Dear Editor,

The authors would like to appreciate Editor for taking your precious time to handle our manuscript. First, I'd like to explain about revisions of datasets in the manuscript:

1. Both Referees #1 and #2 pointed out necessity of providing details of the methodology. Authors agreed the comment and created a new supplement of the manuscript entitled "Supplementary information and data related to methodology of REASv3" (hereafter "the Supplement").
2. For development of the Supplement, we thoroughly checked the data and system of REASv3.1 (a version of ACPD paper) and found several points needed to be revised including trivial errors. Base on the results of the checks, revisions of the data and system were conducted. All values, tables, and figures in the manuscript including supplementary tables and figures were revised using the updated data.
3. Although discussions and conclusions of the manuscript were generally not influenced by the revisions, there were discrepancies in some species, and countries and regions between REASv3.1 and the revised data (hereafter tentatively named as REASv3.2). Therefore, we prepared another new supplemental document showing the differences between REASv3.2 and REASv3.1.
4. For distribution of the revised data, considering the possibility of additional modification during the revision processes, we would like to take the following processes:
   - We did not use the detailed version number (REASv3.1), but used REAS version 3 (REASv3) in the revised main manuscript including the title. The detailed version number were described only in the "Data availability" section.
   - The tentative data during the revision processes will not be opened in the download site of REAS.
   - When the revision process has been completed, the final version will be opened at the REAS download site as REASv3.2.

Above points were also described in the author comments for Referees #1 and #2.

The structure of this document is as follows:

(1) Comments, author's response, and author's changes in manuscript related to Referee #1
(2) Comments, author's response, and author's changes in manuscript related to Referee #2
(3) The revised main manuscript where changed parts were yellow highlighted
(4) The revised main manuscript with track changes
(5) Revision of the supplementary materials

Sincerely Yours,

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(1) Comments, author's response, and author's changes in manuscript related to Referee #1

No. 1

Referee comments

I've reviewed the paper "Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3.1" by Kurokawa and Ohara. This is an important contribution to the literature, coverring a region where emissions are changing rapidly, as mentioned by the authors. The authors should also be commended for providing summary data on-line. As detailed below, however, significantly more detail needs to be provided in terms of definitions and methodology. The present paper gives a general overview of the methodology, but the resulting emissions are still too much of a "black box" otherwise. I would like to re-iterate that this is important and useful work, but I believe additional documentation is required as discussed below.

Author's response to the Referee comments

One major point which was pointed out by both Referees #1 and #2 are necessity of providing details of the methodology in the manuscript. We totally agree the indications and created a new supplement of the manuscript entitled "Supplementary information and data related to methodology of REASv3" which provides detailed descriptions for the framework, activity data, emission factors, emission controls and other settings adopted in REASv3 including definition of sectors, data sources, treatment of the data, related assumptions, etc. (Hereafter, referred as "the Supplement")

For development of the Supplement, we thoroughly checked the data and system of REASv3.1 (a version of the ACPD paper) and found several points which should be revised including trivial errors in the data and system. Based on the results of the checks, revisions of the data and system were conducted including correction of the errors. In general, discussion and conclusions of the manuscript were not influenced by the revision. However, for some species, countries and regions, there were discrepancies between REASv3.1 and the revised one which is tentatively named as REASv3.2. Therefore, we prepared another supplemental document showing the differences between REASv3.2 and REASv3.1 and causes of the discrepancies entitled "Differences between REASv3.2 and REASv3.1". For distribution of the revised data, considering the possibility of additional modification during the revision processes, we would like to take the following processes:

- We did not use the detailed version number (REASv3.1), but used REAS version 3 (REASv3) in the revised main manuscript including the title. The detailed version number were described only in the "Data availability" section.
- The tentative data during the revision processes will not be opened in the download site of REAS.
- When the revision process has been completed, the final version will be opened at the REAS

download site as REASv3.2.

Author’s changes in manuscript

- As described in the author’s response, following two new supplemental documents were created
  - Supplementary information and data related to methodology of REASv3
  - Differences between REASv3.2 and REASv3.1
- All figures and tables in the main manuscript and supplement were recreated by the updated datasets which were also explained in the author’s response.
- “version 3” and “REASv3” were used instead of “version 3.1” and “REASv3.1”, respectively. The differences of versions were described in “Data availability” and the new supplement was introduced there.
- Numbers such as emission amounts and growth rates in abstract, Sects 3.1, and 4 were revised as seen in yellow highlighted parts in abstract, Sects 3.1, and 4 in (3) and corresponding track changes in (4). Line numbers (Page numbers) including corresponding revisions in the revised main manuscript are as follows: L11-14 (P1); L437-439 (P14); L460, L463-465, L471-472 (P15); L499, L511-512 (P16); L535-536 (P17); L549-551, L554-555, L565, L569 (P18); L588 (P19); and L927-930 (P29).

No. 2

Referee comments

Section 2 "Methodology and data" provides a reasonable overview of the methodology, but this needs to be supplemented with much more detail in the supplementary information. There are numerous places throughout this section where a general procedure is described, but then no details are provided. This additional detail is needed is both to satisfy the general scientific publishing principle that work must be, in principle, reproducible, but also so that readers and users of the data can better interpret these results. This information should largely be in the supplementary material.

Author’s response to the Referee comments

As describe above, we developed the Supplement providing detailed information and explanations related to Sect. 2 “Methodology and data”. In order to avoid making the Sect. 2 long, detailed descriptions were not added to the main manuscript, but appropriate parts in the Supplement were indicated in Sect. 2. Following revisions were conducted for the main manuscript related to the Supplement:

- Sect. 2.1 (General description) was fully revised also referring comments from Referee #2, including addition of a new table (Table 2 entitled “Emission inventories from other research works and officially opened data utilized in REASv3."). In Sect. 2.1 of the revised main manuscript, the Supplement was introduced.

- In Sect. 2.2.1, Sects. S2.4.1 and S2.4.2 of the Supplement were cited for descriptions for combustion and non-combustion sources.
- In Sect. 2.2.2, Sects. S3.1.1-6, and S4.1 of the Supplement were cited for definition of fuel types and details of activity data for stationary sources, including fuel consumption, industrial production, and other transformation.
- In Sect. 2.2.3, Sects. S3.2, S4.2, S5.1.5, S5.2.5, and S8.3 of the Supplement were cited for emission factors and emission controls for stationary combustion, industrial production, other transformation sector.
- In Sect. 2.3.1, Sects. S6.2.1, S6.2.3, and S6.3 of the Supplement were cited for additional information about methodology of road transport sector.
- In Sect. 2.3.2, Sect. S6.1.1 of the Supplement was cited for number of vehicles and annual vehicles kilometer traveled. In addition, wrong citations of references in the previous main manuscript were corrected as follows:
  - L246 of the previous manuscript: Road Transport Yearbook (Morth, 2003-2017) was changed to TERI Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018).
  - L249 of the previous manuscript: Pandey and Venkataraman (2014) was deleted.
  - L252-253 of the previous manuscript: "In this study, settings of REASv2.1 were used as default and were updated if new information was available, such as Pandey and Venkataraman (2014), Sahu et al. (2014) and Mishra and Goyal (2014).“ was revised as "In this study, settings of Streets et al. (2003a) and REASv2.1 were used as default and were updated if national information was available, such as He et al. (2005), Yan and Crookes (2009), Sahu et al. (2014), and Malla (2014).".
- Sect. 2.3.3 for emission factors of road transport was fully revised and Sect. S6.2 of the Supplement was cited.
- In Sect. 2.4.1, Sect. S8.1 of the Supplement was cited for methodologies and data sources for manure management sector for NH_3.
- In Sect. 2.4.2, Sect. S8.2 of the Supplement was cited for methodologies and data sources for fertilizer application sector for NH_3.
- In Sect. 2.5, Sects. S5, S7, S8.4, and S8.5 of the Supplement were cited for activity data and emission factors for non-combustion sources of NMVOC, NH_3, and other transport sector.
- In Sect. 2.6, Sects. S9.1 and S9.2 of the Supplement were cited for methodologies and data sources for grid allocation and monthly variation factors.
- In Sect. 3.4, Sect. S10 of the Supplement was cited for methodologies and settings of uncertainties of each component.

Author's changes in manuscript

- Changes of Sect 2.1 are described in (2).
- Sect. 2.3.3 was fully revised.
- In Sects. 2.2-2.5, descriptions related to citing the Supplement were added as seen in yellow highlighted parts in (3). Line numbers (Page numbers) including corresponding parts in the revised main manuscript are as follows: L149-150, L157-L158 (P5); L167-171, L171-172, L174-175 (P6); L192-193, L202-203, L214-215 (P7); L247-L250 (P8); L261, L266, L268-269, L276-277 (P9); L303 (P10); L353-354, L369, L380-381 (P12); L386-387, L388-391, L410 (P13); and L422 (P14); L872-873 (P27); and L924-926 (P29).
- In Sects. 2.2-2.5, incorrect citations including by typos were corrected as seen in yellow highlighted parts in (3) and corresponding track changes in (4). Line numbers (Page numbers) including corresponding revisions in the revised main manuscript are as follows: L176, L186 (P6); L231, L242, L245 (P8); L283-284, L286 (P9); L289-290 (P10); L346, L347 (P11); L377 (P12); L385, L396, L417 (P13); L420 (P14).

No. 3

Referee comments

Some specific points in this regard:

Definitions of the sectors that are included within REAS needs to be provided in much more detail. The EDGAR inventory, for example, provides a fairly comprehensive list of sectors (although even here not all sources are included - REAS includes human waste sources of NH3, for example, while EDGAR does not. This is an example of why it is important to have comprehensive documentation of the methodology and definitions). It is important for users of this data to have this information available.

What is needed is a detailed table with sub-sector definitions. For example Table S3 in Janssens-Maenhout et al 2019 and Tables A1 and A2 in Hoesly et al. 2018.

It is not clear the extent to which non-combustion emissions are included in REAS (I presume they are), and which sectors are included and how they are estimated. For example see the list of non-combustion sectors from EDGAR noted above. Are all of these included for all species in REAS, or just some? (Industrial process emission sources are mentioned in the text, but that is not very specific. Some of the specific non-combustion sources that should be discussed (whose level of inclusion varies between inventories) include: emissions from coke production, non-ferrous metal smelters, refineries, agricultural NOx, open residential waste burning (and waste burning in dumps), and so on.

For these sectors, clarify if just combustion-related emissions are considered, or also if process *(non-combustion) emissions. This is particularly important for sectors that have both combustion-related and non-combustion emissions such as refining, coke production, etc.*

Author's response to the Referee comments

Tables of sub-sectors included in REASv3 are provided in Sect. S2 of the Supplement. For combustion sources, sub-sector categories are compared with IEA code. For some sectors such as iron, steel, and coke production, as suggested, relationships between combustion and non-combustion emissions are also complicated in REASv3. The details are described in Sects. S3 and S4 of the Supplement. In addition, for non-combustion sources of NMVOC and NH_3, details are described in Sects. S5 and S8 as well as Sect. S2 in the Supplement. Related descriptions were added to Sect. 2.1 of the revised main manuscript as described above.

Author's changes in manuscript

- The framework of REASv3 including tables of sub-sectors were provided in Sect. S2 of the Supplement and related description was added to Sect. 2.1 of the revised main manuscript (L125-136 (P4)).
- For combustion and non-combustion sectors, general descriptions were written in Sect. S2 and details were described in Sects. S3 (Combustion sources) and S4 (Industrial process and other transformation) of the Supplement. Related description was added to Sect. 2.2.1 of the revised main manuscript (L149-150 (P5)).
- Non-combustion sources of NMVOC and NH_3 were also described in Sects. S5 and S8 of the Supplement which were cited in Sects. 2.2.3, 2.4.1, and 2.5 of the revised main manuscript (L249-250 (P8); L353-354, L369, L380-381 (P12)).

No. 4

Referee comments

Similar to sectors, the fuel categories used in the calculation need to be discussed as well. While the table "3.2 Fuel types" in the document "Brief description about table data v3.1.pdf" gives an overview, this is not a complete definition of each fuel type. To give just one example, it is not clear what category LPG is in. Since the authors use the IEA energy statistics, what would be most useful is a correspondence between the fuels detailed in IEA and the aggregate fuels reported by the authors.

Given that multiple data sources are used for fuel consumption, comparability of definitions and data time series between multiple sources used for some countries should be addressed.

The authors mention that fuel use by sector is extrapolated back into time before the point where detailed data is used by constant sector shares. This is an important assumption, so the year at *which this assumption comes into force should be listed in the supplement. (For example, given the greater data availability for Japan, perhaps fuel use by sector is available over all time periods.)*

Author's response to the Referee comments

List of detailed fuel types and definition of aggregated categories used in the main manuscript and supplement are provided in Sect. S3.1.1 and Table 3.1 in the Supplement. Data sources of fuel consumption and assumptions to estimate missing historical data are also provided for each country in Sect. S3.1.2 and Table 3.2 in the Supplement document. Related descriptions were added to Sect. 2.1 of the main manuscript as described above.

Author's changes in manuscript

- Definition of fuel type was provided in Table 3.1 in Sect. S3.1.1 of the Supplement and related description was added to Sect. S2.2.1 of the revised main manuscript (L149-150 (P5)).
- Details of data sources, treatments, and related assumptions for development of historical data were also provided in Sect. S3.1.2 and Table 3.2. Related description was added to Sect. 2.2.2 of the revised main manuscript (L171-172 (P6)).

No. 5

Referee comments

For some countries and time periods fuel consumption in non-power plant transformation sectors, particularly coal coke production, is large. How this and other non-combustion or feedstock use of fuels is dealt with over time should be discussed.

Author's response to the Referee comments

For fuel consumption including input amounts of coal for coke ovens and crude oil for oil refinery, data sources and assumptions for estimating missing historical data were described in Sect. S3 of the Supplement document. The historical data and assumptions for coke production amounts were described in Sect. S4 of the Supplement.

Author's changes in manuscript

- Details of data sources and treatments of fuel consumption were provided in Sect. 3 of the Supplement. Fuel data for coke ovens and oil refinery were described in Table 3.3 and Sect. 4.1.8 of the Supplement.

No. 6

Referee comments

Overall, the data sources for assumptions for both activity data (including associated information such as fuel use by vehicle type) and emission factors are unclear. What would be helpful is a comprehensive table (or, likely set of tables), that list this information by sector and country (and species, where relevant). It appears that there are specific data sources for some countries, while for others more generic assumptions are used (in which cases, those countries could be grouped together).

Author's response to the Referee comments

As described above, details of activity data, emission factors, and emission controls adopted in REASv3 were described in the Supplement document for all sources and species. When country-specific settings were adopted, related data and information were also described in the Supplement.

Author's changes in manuscript

- The situations of available data and information were different among countries and regions. Therefore, considering comments from both Referee #1 and #2, we created the Supplement which provides details of activity data, emission factors, settings of emission controls, etc.

No. 7

Referee comments

For road vehicles, the relationship between the "tentative emission factors" and final emission factors in section 2.3.3 Emission factors is unclear.

Author's response to the Referee comments

Thank you for pointing out the problem. We agree that explanations and expressions in Sect. 2.3.3 were unclear and inappropriate. In addition, we also found some wrong citations. In Sect. S6 of the Supplement, detailed data and information for road transport sector were provided. We revised the Sect. 2.3.3 and cited Sect. S6 of the Supplement, as described above.

Author's changes in manuscript

- Sect. 2.3.3 was fully revised.

No. 8

Referee comments

One of the signifiant contributions of this work is the consideration of emission controls. The data sources and assumptions for these calculations by country should be described in greater detail

(again, largely in the supplement). This will provide the readers with important context in terms of how reliable this information might be (e.g., presumably more reliable for Japan than for some other countries!)
At minimum, figures for countries other than Japan and China (e.g. Figure 3, and 5) should be provided in the supplement (at least for those countries with non-zero levels of control). Ideally numerical values would be provided as well.
The development of emission control assumptions for industry is not described in the text. This is an important sector for air pollutions, so further information should be provided. For example, it is not clear to me what BC or SO2 controls in China represent going back as far as 1990. (Very recently SO2 controls were mandated in industrial boilers, but my understanding is that these controls were only for very recent years.)
Also, are emission controls considered for only industrial combustion emissions? Or also for industrial process emissions. Note that technology changes can have a very significant impact on emissions from industrial process technologies (e.g., metal smelters, coke production plants, refineries, etc.). The assumptions here should be discussed. (As noted above, it is not clear the extent to which emissions from these sources/sectors, other than combustion emissions, are included.)

Author's response to the Referee comments

We appreciate the important comments. In Sects. S3 and S4 of the Supplement document, details of settings and assumptions for emission controls both for power plants and industry sectors adopted in REASv3 were described for all countries and regions. However, except for China and Japan, available data and information were limited. For emission controls in industrial processes, the same settings for combustion emissions were adopted except for China where information on some sectors was available from studies for emission inventories of China. Considering the above status, in this manuscript, detailed discussions on effects of emission controls using the figures (like Figs. 3 and 5) were conducted only for China and Japan. Further surveys of local information of emission controls and related abatement technologies are necessary especially for countries and regions other than China and Japan and detailed discussions are important tasks in future studies. These points were emphasized in Sect. 4 of the revised main manuscript.

Author's changes in manuscript

- Details for settings of emission controls were described in Sects. S3 and S4 of the Supplement. and related description was added to Sect. 2.2.1 of the revised main manuscript (L247-249 (P8)).
- Due to reasons described in the author's response, detailed discussion on effects of emission controls using the figures were focused to China and Japan in this study and those for other

countries and regions are important issues in future studies. Related descriptions were added to Sects. 1 and 4 (L78-79 (P3); L949-951 (P30)).

No. 9

Referee comments

It is unclear what is meant by "In REASv3.1, aviation and ship emissions are not included", except then it is said "emissions from domestic shipping including fishing ships were roughly calculated for comparison with other inventories (see Section 3.3)." Clarify. I assume this means that international shipping emissions are not included at all. Does this mean that domestic shipping and fishing emissions are included? What are the data sources for these? (Presumably largely IEA?) So are these emissions C4 included in the REAS3.1 totals? Or are they just included for comparison with other inventories?

Note the substantial literature on shipping emissions, which notes that shipping fuel use, particularly into the past, is inconsistently and incompletely reported in general.

Author's response to the Referee comments

In the first manuscript, we included roughly estimated emissions of domestic and fishing ships just for comparisons with other inventories in Sect. 3.3. However, we reconsidered that including roughly estimated ship emissions for the comparison with other inventories was not appropriate. In the revised main manuscript, we did not add any shipping emissions to REASv3 for the comparison with other studies. This means that in REASv3, emissions from both international and domestic aviation and navigation including fishing ships are totally out of scope. This was also clarified in the revised main manuscript.

Author's changes in manuscript

- In REASv3, international and domestic aviation and navigation including fishing ships are out of scope. This was clarified in Sects. 2.1 (L108-109 (P4)) and 3.3 (L703-704 (P22)) in the revised main manuscript.

No. 10

Referee comments

I note that with these additional details in the supplement, it will likely be possible to streamline the main text somewhat to make the paper more readable.

Author's response to the Referee comments

We agree the suggestion. The corresponding sections in the Supplement were indicated in the main manuscript to provide details for data sources, their treatment, settings and related assumptions as described above.

Author’s changes in manuscript

- This is a general response to the general comment. Corresponding revisions were described above.

No. 11

Referee comments

Section "3.1 Trends of Asian and national emissions" is a bit tedious with the reporting of numerical results. Given that the numerical results are available on-line, the detailed recitation of numerical values in the text is not necessary and detracts from reading the manuscript. A more general overview of the trends and drivers with fewer numerical values would be more useful.

Author’s response to the Referee comments

We appreciate the comments. For the indicated numerical results, we left data of total Asia in the abstract, the first paragraph of Sects. 3.1.1. and 4 to provide general status in Asia. For China and India, results of growth rates in these 60 years were left because these were key features in Asia. For other countries and regions, the indicated numerical values for all species were deleted. In Sects. 3.1.2-3.1.5, more descriptions for features of trends and their drivers were added. On the other hand, Referee #2 gives a following comment: “Please quantitatively estimate the contributions of the energy consumption growth and of the air pollution control progresses on the emission changes over each region discussed in Sect. 3.”. Considering the comment, some quantitative discussions were added for major points of trends and their drivers. For effects of emission controls, as explained above, detailed discussions were conducted only for China and Japan.

Author’s changes in manuscript

- For the numerical results (reports of emissions of each air pollutant), different revisions were done for different countries and regions as follows:
    - Asia: The numerical results in abstract (L11-14 (P1)), Sects. 3.1.1 (L437-439 (P14)), and 4 (L927-930 (P29)) were not deleted to provide general status in Asia. But values were revised as mentioned first.
    - China and India: The results of growth rates in these 60 years were left (China: L470-472 (P15), India: L534-536 (P17)) considering that they were key features in Asia. Values were also revised.
    - Other countries and regions: Corresponding results were deleted in the revised main

manuscript. (In the ACPD paper: L502-505 (P16) for Japan, L546-553 (P16) for Southeast Asia and South Asia other than India, L571-575 (P16) for East Asia other than China and Japan).

- New discussions on features of trends and their drivers including quantitative evaluations were added to descriptions in Sects. 3.1.2-3.1.5. Line numbers (Page numbers) including corresponding parts in the revised main manuscript are as follows:
  - Sect. 3.1.2: L481, L486-491, L495-499, L499-502, L504-507, L508 (P16); L519-520, L529-532 (P17).
  - Sect. 3.1.3: L542-544, L546-L548 (P17); L552-553, L558-561, L563-564, L566, L568, L571-573 (P18).
  - Sect. 3.1.4: L577-578 (P18); L582-585, L590-596, L602-606, L610, L614-617 (P19).
  - Sect. 3.1.5: L630, L638-640, L642-646 (P20); L648-651, L660, L663-665, L667, L668-670 (P21).

No. 12

Referee comments

Line 595 "As described in Section 2.5". I believe this is Section 2.6?

Author's response to the Referee comments

Thank you for pointing out the typo. It was corrected.

Author's changes in manuscript

- The pointed out part (P22L684 in the revised main manuscript) was corrected.

No. 13

Referee comments

In "3.3 Comparison with other inventories" it should be mentioned if the sectoral coverage of the inventories compared are the same? Different sectoral coverage can lead to artificial differences in such comparisons.

Note that for many emissions and countries in Asia the CEDS data was calibrated to REAS2.1 - hence the similarity in results.

This section is long and difficult to read. The extremely long paragraphs should be broken up and, where possible, streamlined.

Author's response to the Referee comments

First, we agree the comment that the Sect. 3.3 is too long and should be streamlined. In the revised main manuscript, we divided the contents to 4 sub-sections: China, India, Other regions, and Relative ratios of emissions from each country and region in Asia and descriptions in these sections were revised. In addition, comparison of emissions in total Asia among REASv3, CEDS, and EDGARv4.3.2 were added to Figs. 12 and S20. For the relationship between CEDS and REASv2.1, the information was added appropriate parts of the main manuscript. For sector categories, as described above, emissions from aviation and navigation were not included in REASv3. Therefore, if it was possible, corresponding emissions were subtracted from total emissions of other inventories. But, unfortunately, there were many inventories where no independent emission data of domestic navigation were available. In this study, such inventories were also included in the comparisons with the notices in the figure captions. These procedures were also mentioned in the revised main manuscript. In addition, based on a comment from Referee #2, we included following top-down emission data.

- Ding, J., Miyazaki, K., van der A, R. J., Mijling, B., Kurokawa, J.-I., Cho, S., Janssens-Maenhout, G., Zhang, Q., Liu, F., and Levelt, P. F.: Intercomparison of NO_x emission inventories over East Asia, Atmos. Chem. Phys., 17, 10125–10141, https://doi.org/10.5194/acp-17-10125-2017, 2017.
- Itahashi, S., Yumimoto, K., Kurokawa, J., Morino, Y., Nagashima, T., Miyazaki, K., Maki, T., and Ohara, T.: Inverse estimation of NO_x emissions over China and India2005–2016: contrasting recent trends and future perspectives, Environ. Res. Lett., 14, 124020, https://doi.org/10.1088/1748-9326/ab4d7f, 2019.
- Jiang, Z., Worden, J. R., Worden, H., Deeter, M., Jones, D. B. A., Arellano, A. F., and Henze, D. K.: A 15-year record of CO emissions constrained by MOPITT CO observations, Atmos. Chem. Phys., 17, 4565–4583, https://doi.org/10.5194/acp-17-4565-2017, 2017.
- Miyazaki, K., Bowman, K., Sekiya, T., Eskes, H., Boersma, F., Worden, H., Livesey, N., Payne, V. H., Sudo, K., Kanaya, Y., Takigawa, M., and Ogochi, K.: An updated tropospheric chemistry reanalysis and emission estimates, TCR-2, for 2005–2018, Earth Syst. Sci. Data Discuss., https://doi.org/10.5194/essd-2020-30, in review, 2020.
- Qu, Z., Henze, D. K., Li, C., Theys, N., Wang, Y., Wang, J., Wang, W., Han, J., Shim, C., Dickerson, R. R., and Ren, X.: SO_2 emission estimates using OMI SO_2 retrievals for 2005–2017, J. Geophys. Res. Atmos., 124, 8336-8359, https://doi.org/10.1029/2019JD030243, 2019.
- Stavrakou, T., Muller, J. F., Bauwens, M., De Smedt, I.: Sources and long-term trends of ozone precursors to Asian Pollution, Air Pollution in Eastern Asia: an integrated perspective, eds. Bouarar, I., Wang, X., Brasseur, G., Springer international Publishing, 167–189, https://doi.org/10.1007/978-3-319-59489-7-8, 2017.
- Zheng, B., Chevallier, F., Yin, Y., Ciais, P., Fortems-Cheiney, A., Deeter, M. N., Parker, R. J., Wang, Y., Worden, H. M., and Zhao, Y.: Global atmospheric carbon monoxide budget 2000–

2017 inferred from multi-species atmospheric inversions, Earth Syst. Sci. Data, 11, 1411–1436, https://doi.org/10.5194/essd-11-1411-2019, 2019.

Furthermore, we added following two bottom-up historical emission inventories of China:

- Sun, W., Shao, M., Granier, C., Liu, Y., Ye, C. S., and Zheng, J. Y.: Long-term trends of anthropogenic SO_2, NO_x, CO, and NMVOCs emissions in China, Earth's Future, 6, 1112-1133, https://doi.org/10.1029/2018EF000822, 2018.
- Wang, R., Tao, S., Wang, W., Liu, J., Shen, H., Shen, G., Wang, B., Liu, X., Li, W., Huang, Y., Zhang, Y., Lu, Y., Chen, H., Chen, Y., Wang, C., Zhu, D., Wang, X., Li, B., Liu, X., and Ma, J.: Black Carbon Emissions in China from 1949 to 2050, Environ. Sci. Technol., 46, 7595-7603, https://doi.org/10.1021/es3003684, 2012.

Author's changes in manuscript

- Sect. 3.3 were fully revised. The section was divided into 4 independent sub-sections: 3.3.1 China, 3.3.2 India, 3.3.3 Other regions, and 3.3.4 Relative rations of emissions from each country and region in Asia. Furthermore, comparison of total emissions in Asia among REASv3, CEDS, and EDGARv4.3.2 were added to Figs. 12 and S20.
- Relationships between CEDS and REASv2.1 were pointed out in the revised main manuscript (L754 (P24); L786, L800 (P25); L812 (P26))
- Descriptions for problems of sector coverage especially about domestic navigation were added to Sect. 3.3 of the revised main manuscript (L703-708 (P22)) and notes were added to captions of Figs. 10 and 11 of the revised main manuscript and Figs. S14-S19 in the supplementary figures.

No. 14

Referee comments

The uncertainty calculation is a valuable and important portion of the paper. However, the specific uncertainty assumptions used need to be provided (e.g. in supplemental information), It would be useful to provide some equations here.

How do these assumptions vary by sector and fuel? Are these assumptions constant across countries? And across time? How is uncertainty in emission control percentages handled? One important assumption is how uncertainty was combined across fuels and sectors is a critical part of the methodology and needs to be described (e.g. are independent uncertainties assumed, or is some correlation assumed?).

Author's response to the Referee comments

We appreciate valuable comments for uncertainties. First, we realized that in the first manuscript, uncertainties in settings of emission controls such as timing of introduction and penetration rates of abatement equipment were not considered. Therefore, we revisited the settings and assumptions for uncertainties of removal efficiencies. Because it is difficult to assume the corresponding uncertainties in each year of the target period of REASv3, we decided to analyze the uncertainties of emissions in REASv3 focusing in the years 1955, 1985, and 2015. Uncertainties for all target years of REASv3 will be analyzed in future studies. Details of methodology including equations, settings of uncertainties of each component, and related assumptions were described in Sect. S10 of the Supplement. In addition, as described in "General Reply", we thoroughly checked the data and system of REASv3 which include those for estimation of uncertainties and also found several points need to be revised including trivial errors. By the revisions, uncertainties of SO_2 became lager and those of CO_2 became smaller compared to previous results. Corresponding descriptions in Sect. 3.4 of the main manuscript were revised.

For combination of uncertainties across emission sources, in this study, it was assumed that the uncertainties were independent.

Author's changes in manuscript

- Descriptions about the settings of uncertainties including removal efficiencies and combination of different emission sources were added to the revised main manuscript (L860-868, L870-872 (P27)). In addition, details of methodology and assumption of settings were provided in the Supplement and cited in the revise main manuscript (L872-873 (P27)).
- As described in the Author's response to the Referee comments, settings and assumptions for uncertainties of removal efficiencies were reconsidered and several errors were corrected. Furthermore, emissions themselves were updated in the revised main manuscript. Therefore, all uncertainties were recalculated focusing in the years 1955, 1985, and 2015 and related discussions in the following parts were revised: L878-887, L892-893, L894-897, L899-902 (P28).

(2) Comments, author's response, and author's changes in manuscript related to Referee #2

No. 1

Referee comments

The authors developed a new version of the REAS emission inventory, presented the trends in Asian air pollutant emissions, and analyzed the regional and sectoral drivers. The comparison with the up-to-date regional inventories presents broadly consistent emission trends, and the uncertainties in the REAS v3.1 estimates are quantified according to the errors in each parameter. This is quite important work, because the REAS inventory has been widely used in the modeling of climate and of air quality. This new version extended the emission time series to 1950-2015 and made necessary updates in both the methods and the data input. My major concern is that the method part is not well structured and is very difficult to follow, and the comparison with the previous emissions data lacks the top-down inversion estimates, which should

Author's response to the Referee comments

One major point which was pointed out by both Referees #1 and #2 are necessity of providing details of the methodology in the manuscript. We totally agree the indications and created a new supplement of the manuscript entitled "Supplementary information and data related to methodology of REASv3" which provides detailed descriptions for the framework, activity data, emission factors, emission controls and other settings adopted in REASv3 including definition of sectors, data sources, treatment of the data, related assumptions, etc. (Hereafter, referred as "the Supplement")

For development of the Supplement, we thoroughly checked the data and system of REASv3.1 (a version of the ACPD paper) and found several points which should be revised including trivial errors in the data and system. Based on the results of the checks, revisions of the data and system were conducted including correction of the errors. In general, discussion and conclusions of the manuscript were not influenced by the revision. However, for some species, countries and regions, there were discrepancies between REASv3.1 and the revised one which is tentatively named as REASv3.2. Therefore, we prepared another supplemental document showing the differences between REASv3.2 and REASv3.1 and causes of the discrepancies entitled "Differences between REASv3.2 and REASv3.1". For distribution of the revised data, considering the possibility of additional modification during the revision processes, we would like to take the following processes:

- We did not use the detailed version number (REASv3.1), but used REAS version 3 (REASv3) in the revised main manuscript including the title. The detailed version number were described only in the "Data availability" section.
- The tentative data during the revision processes will not be opened in the download site of REAS.

- When the revision process has been completed, the final version will be opened at the REAS download site as REASv3.2.

Author's changes in manuscript

- The same changes described in "Author's change in manuscript" for Referee Comments No.1 of Referee #1.

No. 2

Referee comments

1) Method. The method section should put more focus on the new features of the new REAS version 3.1 compared to the last version 2.1. Please summarize the new data development process and give a detailed table to show the new methods developed and the new data sources used in the REAS v3.1. Part of the REAS v2.1 emissions data are directly adopted by the REAS v3.1, such as the agricultural sources in Japan, which should be described clearly in this table. The REAS inventory relies on plenty of other emission inventories to provide the emissions data or the spatial proxies used in the emission distribution. The data dependencies across different inventories would better be clarified specifically in a new table, which would benefit the users of different inventories.

Author's response to the Referee comments

The major new feature of REASv3 is a development of the long-term period emission inventory in Asia. A lot of database, statistics, literatures, and information were corrected, surveyed and processed for the development. On the other hand, for estimation of emissions, more detailed information such as for abatement technologies and regulations for road vehicles were taken into considered compare to previous versions of REAS, but basic methodologies themselves were based on traditional ones. Therefore, providing a new table seems to be too much for the new features, but instead, important points and updates from previous versions of REAS were summarized in bullet point format to emphasize them in Sect. 2.1 of the revised main manuscript. In addition, as described above, we developed the Supplement describing details of REASv3 including processing of historical data and appropriate parts of the Supplement were cited in Sect. 2 of the revised main manuscript as described in the Reply for 2). For data dependencies across different inventories, a new table was created (as Table 2) providing other inventories utilized in REASv3 with descriptions how the datasets were utilized.

Author's changes in manuscript

- Sect. 2.1 was fully revised and Table 2 was newly added. (Therefore, the numbers of Tables 2 and 3 in ACPD papers were shifted to 3 and 4, respectively.)

- Table S1 of the ACPD paper for countries and sub-regions included in REASv3.1 was moved to Table 2.3 in Sect. S2 of the Supplement which provides Framework of REASv3. The corresponding description was added to Sect. 2.1 of the revised main manuscript (L125-126 (P4)). For numbers of supplementary tables, Tables S2-S4 in the supplement of ACPD paper were shifted to Tables S1-S3 in the revised supplementary tables.

No. 3

Referee comments

2) Data sources. The manuscript briefly describes the sources of the input data, but the values of parameters are not given. I understand that it is difficult to present all the detailed input data of a large-scale emission inventory. However, knowing the exact values of some key parameters can help the audience understand the drivers of emissions changes. I suggest the authors present some key parameters that determine the curve of emission changes, show their values, and discuss why such values are adopted (e.g., due to more stringent emission legislations). I noticed that the authors used many proxy data to calculate the "trend factors" when the activity data of the past years are not available. This method needs to be justified. Please show the relationship between the proxy data and the associated activity data using the historical values when they are both available.

Author's response to the Referee comments

As described above, we developed the Supplement providing details of REASv3 including values of emission factors and removal efficiencies for major sources, data sources and treatment of activity data with assumptions for estimating missing historical data. Appropriate parts of the Supplement were indicated in Sect. 2 of the revised main manuscript. The revisions conducted in the main manuscript related to the Supplement were as follows:

- Sect. 2.1 (General description) was fully revised also referring comments from Referee #2, including addition of a new table (Table 2 entitled "Emission inventories from other research works and officially opened data utilized in REASv3."). In Sect. 2.1 of the revised main manuscript, the Supplement was introduced.
- In Sect. 2.2.1, Sects. S2.4.1 and S2.4.2 of the Supplement were cited for descriptions for combustion and non-combustion sources.
- In Sect. 2.2.2, Sects. S3.1.1-6, and S4.1 of the Supplement were cited for definition of fuel types and details of activity data for stationary sources, including fuel consumption, industrial production, and other transformation.
- In Sect. 2.2.3, Sects. S3.2, S4.2, S5.1.5, S5.2.5, and S8.3 of the Supplement were cited for emission factors and emission controls for stationary combustion, industrial production, other transformation sector.

- In Sect. 2.3.1, Sects. S6.2.1, S6.2.3, and S6.3 of the Supplement were cited for additional information about methodology of road transport sector.
- In Sect. 2.3.2, Sect. S6.1.1 of the Supplement was cited for number of vehicles and annual vehicles kilometer traveled. In addition, wrong citations of references in the previous main manuscript were corrected as follows:
  - L246 of the previous manuscript: Road Transport Yearbook (Morth, 2003-2017) was changed to TERI Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018).
  - L249 of the previous manuscript: Pandey and Venkataraman (2014) was deleted.
  - L252-253 of the previous manuscript: "In this study, settings of REASv2.1 were used as default and were updated if new information was available, such as Pandey and Venkataraman (2014), Sahu et al. (2014) and Mishra and Goyal (2014).“ was revised as "In this study, settings of Streets et al. (2003a) and REASv2.1 were used as default and were updated if national information was available, such as He et al. (2005), Yan and Crookes (2009), Sahu et al. (2014), and Malla (2014).".
- Sect. 2.3.3 for emission factors of road transport was fully revised and Sect. S6.2 of the Supplement was cited.
- In Sect. 2.4.1, Sect. S8.1 of the Supplement was cited for methodologies and data sources for manure management sector for NH_3.
- In Sect. 2.4.2, Sect. S8.2 of the Supplement was cited for methodologies and data sources for fertilizer application sector for NH_3.
- In Sect. 2.5, Sects. S5, S7, S8.4, and S8.5 of the Supplement were cited for activity data and emission factors for non-combustion sources of NMVOC, NH_3, and other transport sector.
- In Sect. 2.6, Sects. S9.1 and S9.2 of the Supplement were cited for methodologies and data sources for grid allocation and monthly variation factors.
- In Sect. 3.4, Sect. S10 of the Supplement was cited for methodologies and settings of uncertainties of each component.

Author's changes in manuscript

- The same changes described in "Author's change in manuscript" for Referee Comments No.2 of Referee #1.

No. 4

Referee comments

3) Results. The results section mainly focuses on the emissions of SO2, NOx, and BC. Please add CO2 in each plot of the results to reflect the energy consumption trends. It is difficult to understand the drivers of emission changes from the text now. Please quantitatively estimate the contributions of *the energy consumption growth and of the air pollution control progresses on the emission changes over each region discussed in Sect. 3.*

Author's response to the Referee comments

First, the curves of CO_2 emissions were added to each panel of SO_2, NO_x, and BC emissions. Then, we added some quantitative discussion on drivers of emission changes for major points of trends in Sects. 3.1.2-3.1.5. However, for emission controls, as seen in Sects. S3 and S4 of the Supplement, available data and information were limited except for China and Japan. Therefore, in this manuscript, detailed discussions on effects of emission controls were conducted focusing on China and Japan. Further surveys of local information of emission controls and related abatement technologies are necessary especially for countries and regions other than China and Japan and detailed discussion are important tasks in future studies. These points were emphasized in Sect. 4 of the revised main manuscript.

Author's changes in manuscript

- The curves of total CO_2 emissions were added to Figs. 3-7. Descriptions related to the CO_2 emissions were added to Sects. 3.1.2-3.1.5 as follows: L479 (P15); L489-L491, L504-505 (P16); L530-532, L540 (P17); L558-561 (P18); L582-585, L592-596 (P19); L631-634 (P20); L649-651 (P21).
- Descriptions related to quantitative discussions on drivers of emission changes for major points of trends were also added to Sects. 3.1.2-3.1.5 as follows: L486-488, L496-499, L504-506 (P16); L519-520, L542-544, L547-548 (P17); L552-553, L563-564, L571-573 (P18); L590-592, L602-606, L614-616 (P19); L638-640, L642-646 (P20)

No. 5

Referee comments

For the comparison with other inventories, the authors only compared their emission results with other bottom-up emission inventories, while did not consider topdown emissions data constrained by satellite observations that have developed very fast in recent years. In my opinion, different bottom-up emission inventories commonly share the same sources of input data, which are not completely independent of each other. It would be better to evaluate the long-term emission trends with top-down information from previous literature.

Author's response to the Referee comments

We agree with importance of comparison of bottom-up emission inventories with top-down emissions data. The following data were plotted to the figures for comparisons of inventories and discussed:

- Ding, J., Miyazaki, K., van der A, R. J., Mijling, B., Kurokawa, J.-I., Cho, S., Janssens-Maenhout, G., Zhang, Q., Liu, F., and Levelt, P. F.: Intercomparison of NO_x emission inventories over East Asia, Atmos. Chem. Phys., 17, 10125–10141, https://doi.org/10.5194/acp-17-10125-2017, 2017.
- Itahashi, S., Yumimoto, K., Kurokawa, J., Morino, Y., Nagashima, T., Miyazaki, K., Maki, T., and Ohara, T.: Inverse estimation of NO_x emissions over China and India2005–2016: contrasting recent trends and future perspectives, Environ. Res. Lett., 14, 124020, https://doi.org/10.1088/1748-9326/ab4d7f, 2019.
- Jiang, Z., Worden, J. R., Worden, H., Deeter, M., Jones, D. B. A., Arellano, A. F., and Henze, D. K.: A 15-year record of CO emissions constrained by MOPITT CO observations, Atmos. Chem. Phys., 17, 4565–4583, https://doi.org/10.5194/acp-17-4565-2017, 2017.
- Miyazaki, K., Bowman, K., Sekiya, T., Eskes, H., Boersma, F., Worden, H., Livesey, N., Payne, V. H., Sudo, K., Kanaya, Y., Takigawa, M., and Ogochi, K.: An updated tropospheric chemistry reanalysis and emission estimates, TCR-2, for 2005–2018, Earth Syst. Sci. Data Discuss., https://doi.org/10.5194/essd-2020-30, in review, 2020.
- Qu, Z., Henze, D. K., Li, C., Theys, N., Wang, Y., Wang, J., Wang, W., Han, J., Shim, C., Dickerson, R. R., and Ren, X.: SO_2 emission estimates using OMI SO_2 retrievals for 2005–2017, J. Geophys. Res. Atmos., 124, 8336-8359, https://doi.org/10.1029/2019JD030243, 2019.
- Stavrakou, T., Muller, J. F., Bauwens, M., De Smedt, I.: Sources and long-term trends of ozone precursors to Asian Pollution, Air Pollution in Eastern Asia: an integrated perspective, eds. Bouarar, I., Wang, X., Brasseur, G., Springer international Publishing, 167–189, https://doi.org/10.1007/978-3-319-59489-7-8, 2017.
- Zheng, B., Chevallier, F., Yin, Y., Ciais, P., Fortems-Cheiney, A., Deeter, M. N., Parker, R. J., Wang, Y., Worden, H. M., and Zhao, Y.: Global atmospheric carbon monoxide budget 2000–2017 inferred from multi-species atmospheric inversions, Earth Syst. Sci. Data, 11, 1411–1436, https://doi.org/10.5194/essd-11-1411-2019, 2019.

Furthermore, we added following two bottom-up historical emission inventories of China:

- Sun, W., Shao, M., Granier, C., Liu, Y., Ye, C. S., and Zheng, J. Y.: Long-term trends of anthropogenic SO_2, NO_x, CO, and NMVOCs emissions in China, Earth's Future, 6, 1112-1133, https://doi.org/10.1029/2018EF000822, 2018.
- Wang, R., Tao, S., Wang, W., Liu, J., Shen, H., Shen, G., Wang, B., Liu, X., Li, W., Huang, Y., Zhang, Y., Lu, Y., Chen, H., Chen, Y., Wang, C., Zhu, D., Wang, X., Li, B., Liu, X., and Ma, J.: Black Carbon Emissions in China from 1949 to 2050, Environ. Sci. Technol., 46, 7595-7603, https://doi.org/10.1021/es3003684, 2012.

Author’s changes in manuscript

- For comparison with other inventories, results of inverse modeling were added to Figs. 10 (SO_2 and NO_x for China), 11 (SO_2 and NO_x for India), S14 (CO and NMVOC for China) and S15 (CO for India). Discussions with inverse modeling results were added to Sect. S3.3 as follows: L716-719, L723-726, L740-743, L744-754 (P23); L756-759, L763-766, L773-776 (P24)

- Furthermore, two bottom-up emission inventories were added for China. (Sun et al. (2018) for SO_2 and NO_x in Fig. 10 and for CO and NMVOC in Fig. S14; Wang et al. (2012) for BC in Fig. 10)

No. 6

Referee comments

For the uncertainty assessment, I cannot understand why the uncertainties of CO2 emissions are so large, particularly ±28% for China and ±23% for Japan, which are much higher than the typical uncertainty range (±10%) of country CO2 emissions.

Author’s response to the Referee comments

Thank you for pointing out the issue. First, from comments of Referee #1, we realized that in the first manuscript, uncertainties in settings of emission controls such as timing of introduction and penetration rates of abatement equipment were not considered. Therefore, we revisited the settings and assumptions for uncertainties of removal efficiencies. Details of methodology including equations, settings of uncertainties of each component, and related assumptions were described in Sect. S10 of the Supplement. In addition, as described in “General Reply”, we thoroughly checked the data and system of REASv3 which include those for estimation of uncertainties and found several points need to be revised including trivial errors. By the revisions, uncertainties of SO_2 became lager and those of CO_2 became smaller compared to previous results. Corresponding descriptions in Sect. 3.4 were revised.

For CO_2, in the revisiting process, we found errors in settings of uncertainties of CO_2 emission factors for fossil fuel combustion. After the correction of errors, as described above, uncertainties of CO_2 emissions became lower than those in first manuscript. For Japan, the updated uncertainties are ±13% from ±23% in the first manuscript. However, for China, even after the correction of errors, the updated uncertainties (±19%) were still higher than ±10%. One reason is that high uncertainties were assumed for emission factors of biofuel combustion (50%). Another considerable reason is that in REASv3, uncertainties in fossil fuel consumption data were assumed to be higher than those of OECD countries except for Japan, Republic of Korea, and Taiwan. For example, uncertainties in coal consumption in power plants, small industries, and residential sectors in China were assumed to be 10%, 15%, and 20%, respectively.

Author’s changes in manuscript

- The changes related to uncertainties are the same as for “Author’s change in manuscript” for Referee Comments No.14 of Referee #1.
- Discussions on problems in uncertainties for CO_2 emissions described in the authors response were not included in the revised main manuscript because corresponding discussions are considered to be a bit too specific for this manuscript.

No. 7

Referee comments

1) Line 47 on Page 2. The GAINS model not the GANS model.

Author’s response to the Referee comments

Thank you for pointing out the typo. It was corrected.

Author’s changes in manuscript

- The corresponding type was corrected (P2L47).

No. 8

Referee comments

2) Line 318 on Page 10. For spatial distribution not the special distribution.

Author’s response to the Referee comments

Thank you for pointing out the typo. It was corrected.

Author’s changes in manuscript

- The corresponding type was corrected (P12L362).

No. 9

Referee comments

3) Lines 354 and 355 on Page 12. Please clarify how the information of large plants is used for developing allocation factors for corresponding emission source sectors.

Author’s response to the Referee comments

In Sect. S9 of the Supplement, how to utilize the information of large plants is explained. It was referred in the main manuscript.

Author's changes in manuscript

- Details were described in Sect. S9.1 of the Supplement and the description related to citing the Supplement was added to Sect. 2.6 (L410 (P13)).

No. 10

Referee comments

4) The caption of Figure 3. During 1990-2015 not 1950-2015.

Author's response to the Referee comments

Thank you for pointing out the typo. It was corrected.

Author's changes in manuscript

- The corresponding type was corrected (the caption of Fig.3).

No. 10

Referee comments

5) Figures 10 and 11. The colors of some curves are close to each other and are difficult to distinguish. And please also add the uncertainty range of REAS v3.1 in the plots.

Author's response to the Referee comments

Thank you for pointing out the issue. For curves which were difficult to be identified, colors or line thicknesses were changed. For uncertainties, as described above, we revisited the settings and assumptions for uncertainties of removal efficiencies. Because it is difficult to assume the corresponding uncertainties in each year of the target period of REASv3, we decided to analyze the uncertainties of emissions in REASv3 focusing in the years 1955, 1985, and 2015. For the uncertainty ranges, error bars in 1955, 1985, and 2015 were added to the plots of comparisons. Uncertainties for all target years of REASv3 will be analyzed in future studies.

Author's changes in manuscript

- Colors, shapes, thickness, etc. of plotted data in Figs. 10, 11, and S14-19 were reconsidered and revised.
- Error bars based on uncertainties ranges of REASv3 were added to data in 1955, 1985, and 2015 for all panels in Figs. 10, 11, and S14-S19. Related descriptions were added to Sect. S3.3 (L701-703 (P22)) and captions of Figs. 10, 11, and S14-S19.

(3) The revised main manuscript where changed parts were yellow highlighted

From next pages, the revised main manuscript where changed parts were yellow highlighted is provided. In addition to the revisions based on comments from Referees #1 and #2 described in (1) and (2), some additional revisions were done mainly for correction of typos and English problems as follows:

- “control measures” were used instead of “regulations and laws” in abstract (L21 (P1)) and Sect. 4 (L945 (P30)).
- “the Republic of Korea” was changed to “Republic of Korea” in several points: L22 (P1); L658 (P21); L875 (P27); and L945 (P30)).
- “the REAS series” was changed to “the Regional Emission inventory in ASia (REAS) series” in Sect.1 (L70 (P3)).
- “PM” was changed to “PM species” in several points: L240 (P8); L621 (P20); and L666, L668 (P21).
- Acronyms SEA (Southeast Asia), OEA (East Asia other than China and Japan), and OSA (South Asia other than India) were introduced in Sect. 3.1.1 (P14L435-436) and corresponding changes were done in the following points: L466-467 (P15); L630, L631, L633, L637, L638, L642 (P20); L652, L680 (P21); L683, L686, L691, L693 (P22); and L876-877 (P28). In addition, for readers’ conveniences, colors of country names in Fig.1 were changed to indicate SEA, OEA, and OSA. Furthermore, for captions of figures and tables in the revised main manuscript and supplementary materials, SEA, OEA, and OSA were used and the description “See Fig. 1 for countries included in SEA, OEA, and OSA” was added to them.
- Other small revisions such as correction of grammatical problems and English expressions were done in the following points: L19 (P1); L266, L267 (P9); L412 (P13); L430 (P14); L482, L483, L513 (P16); L523 (P17); L549, L551, L569 (P18); L588, L608, L609 (P19); L622, L624, L625, L627, L631, L633, L636, L637 (P20); L657, L675, L676, L680 (P21); L682, L685, L689, L690,. L691, L692, L693, L694 (P22); L859 (P27); L877 (P28); L918-919, L941 (P29); L956 (P30)

Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3

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Abstract. A long-term historical emission inventory of air and climate pollutants in East, Southeast, and South Asia from 1950-2015 was developed as the Regional Emission inventory in ASia version 3 (REASv3). REASv3 provides details of emissions from major anthropogenic sources for each country and its sub-regions and also provides monthly gridded data with 0.25° × 0.25° resolution. The average total emissions in Asia during 1950-1955 and from 2010-2015 (growth rates in these 60 years) are as follows: SO_2: 3.2 Tg, 42.4 Tg (13.1); NO_x: 1.6 Tg, 47.3 Tg (29.1); CO: 56.1 Tg, 303 Tg (5.4); non-methane volatile organic compounds: 7.0 Tg, 57.8 Tg (8.3); NH_3: 8.0 Tg, 31.3 Tg (3.9); CO_2: 1.1 Pg, 18.6 Pg (16.5); PM_{10}: 5.9 Tg, 30.2 Tg (5.1); $PM_{2.5}$: 4.6 Tg, 21.3 Tg (4.6); black carbon: 0.69 Tg, 3.2 Tg (4.7); and organic carbon: 2.5 Tg, 6.6 Tg (2.7). Clearly, all the air pollutant emissions in Asia increased significantly during these six decades, but situations were different among countries and regions. Due to China's rapid economic growth in recent years, its relative contribution to emissions in Asia has been the largest. However, most pollutant species reached their peaks by 2015 and the growth rates of other species was found to be reduced or almost zero. On the other hand, air pollutant emissions from India showed an almost continuous increasing trend. As a result, the relative ratio of emissions of India to that of Asia have increased recently. The trend observed in Japan was different from the rest of Asia. In Japan, emissions increased rapidly during the 1950s-1970s, which reflected the economic situation of the period; however, most emissions decreased from their peak values, which were approximately 40 years ago, due to the introduction of control measures for air pollution. Similar features were found in Republic of Korea and Taiwan. In the case of other Asian countries, air pollutant emissions generally showed an increase along with economic growth and motorization. Trends and spatial distribution of air pollutants in Asia are becoming complicated. Datasets of REASv3, including table of emissions by countries and sub-regions for major sectors and fuel types, and monthly gridded data with 0.25° × 0.25° resolution for major source categories are available through the following URL: http;//www.nies.go.jp/REAS/.

1 Introduction

With an increase in demand for energy, motorization, and industrial and agricultural products, air pollution from anthropogenic emissions is becoming a serious problem in Asia, especially due to its impact on human health. In addition, a significant increase in anthropogenic emissions in Asia is considered to affect not only the local air quality, but also regional, inter-continental, and global air quality and climate change. Therefore, reduction in air and climate pollutants emissions are urgent issues in Asia (UNEP, 2019). Short-Lived Climate Pollutants (SLCPs), which are gases and particles that contribute to warming and have short lifetimes, have been recently considered to play important roles in the mitigation both air pollution and climate change (UNEP, 2019). SLCPs such as black carbon (BC) and ozone are warming agents, which cause harm to people and ecosystems. A decrease in the emissions of BC and ozone precursors from fuel combustion led to the decrease of other particulate matter (PM) species, such as sulfate and nitrate aerosols. Even though this is a positive step for human health, it has a negative effect on global warming as sulfate and nitrate aerosols act as cooling agents in the troposphere. Therefore, to find effective ways to mitigate both air pollution and climate change, accurate understanding of the current status and historical trends of air and climate pollutants are fundamentally important.

Recently, Hoesly et al. (2018) developed a long-term historical global emission inventory from 1750 to 2014 using the Community Emission Data System (CEDS). This data set is used as input data for the Coupled Model Intercomparison Project phase 6 (CMIP6). The Emission Database for Global Atmospheric Research (EDGAR) also provides global emissions data of both air pollutants and greenhouse gases, with the current version 4.3.2 ranging from the period between 1970-2012 (Crippa et al., 2016). The EDGAR is used as the default data of input emissions for the Task Force on Hemispheric Transport of Air Pollution phase 2 (HTAPv2) (Janssens-Maenhout et al., 2015). For SLCPs, the European Union's Seventh Framework Programme project ECLIPSE (Evaluating the Climate and Air Quality Impact of Short-Lived Pollutants) developed a global emission inventory based on the GAINS model. Current version 5 provides gridded emissions for every five years from 1990 to 2030 and also from 2040 to 2050 (Stohl et al., 2015). However, data from Asia in global emission inventories are generally based on limited country specific information. For the Asian region, several project-based emission inventories are developed, such as Transport and Chemical Evolution over the Pacific (TRACE-P) field campaigns (Streets et al., 2003a, b) and its successor mission, that is Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) (Zhang et al., 2009). Recently, the MIX inventory (mosaic Asian anthropogenic emission inventory) was developed as input emission data sets for the Model Intercomparison Study for Asia (MICS-Asia) Phase 3 by a mosaic of up-to-date regional emission inventories. The MIX inventory is also a component of the HTAPv2 inventory (Li et al., 2017a). For national emission inventories, numerous studies, research papers, and reports have been published. MEIC (Multi-resolution Emission Inventory for China) developed by Tsinghua University is a widely used emission inventory database for China (Zhang et al., 2009; Li et al., 2014; Zheng et al., 2014, Liu et al., 2015) and is included in the MIX inventory. Zhao et al. (2011, 2012, 2013, and 2014) developed recent and projected emission inventories of air pollutants in China. In addition, research papers for regional emission inventories of China were also published recently (Zhu et al., 2018; Zheng et al., 2019a). In the case of India, Garg et al., (2006) developed a historical emission inventory of air pollutants and greenhouse gases from 1985 to 2005. For recent years, Sadavarte and Venkataraman (2014) developed multi-pollutant emission inventories for industry and transport sectors and Pandey et al. (2014) developed the same for domestic and small industry sectors for the same time period, that is 1996-2015. For Japan, several project-based emission data sets were developed, such as the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC 2012a, b, c; 2014), East Asian Air Pollutant Emission Grid Database (EAGrid) (Fukui et al., 2014), and emission data sets for Japan's Study for Reference Air Quality Modeling (J-STREAM) (Chatani et al., 2018). In addition, there are studies for other countries and regions, such as the Clean Air Policy Support System (CAPSS) for Republic of Korea (Lee et al., 2011), Thailand (Thao Pham et al., 2008), Indonesia (Permadi et al., 2017), and Nepal (Jayarathne et al., 2018; Sadavarte et al., 2019). However, these regional and national emission inventories in Asia are available for a limited period, with data of the past missing.

The authors of this study have been devoted in developing the Regional Emission inventory in ASia (REAS) series. First version of REAS (REASv1.1) were developed by Ohara et al. (2007), which accounted for actual emissions during 1980-2003 and projected ones in 2010 and 2020. Kurokawa et al. (2013) updated the inventory in REASv2.1, which focused on the period between 2000-2008 when emissions in China drastically increased. REASv2.1 is used as the default data of the MIX inventory. In this study, a long historical emission inventory in the Asian region from 1950-2015 has been newly developed as REAS version 3 (REASv3). This study provides methodology, results and discussion of REASv3.1. Section 2 gives the basic methodology, including collecting activity data, settings of emission factors and removal efficiencies, and spatial and temporal allocation of emissions to create monthly gridded data sets of REASv3. In Section 3.1, trends in air pollutants emissions in Asia are described in detail and effects of emission controls on emissions in China and Japan are discussed. Spatial and temporal distributions are overviewed in Section 3.2. Section 3.3 compares the results of REASv3.1 with other emission inventories. Uncertainties of REASv3.1 are discussed in Section 3.4. Finally, summary and remarks are presented in Section 4.

2 Methodology and data

2.1 General description

Table 1 summarizes the general information of REASv3. Major updates from previous versions are as follows:

- Target years are from 1950 to 2015 covering much longer periods than REASv1.1 (1980-2003) and REASv2.1 (2000-2008).
- The long historical data sets of activity data were developed by collecting international and national statistics and related proxy data.
- Emission factors and information of emission controls especially for China and Japan were surveyed from research papers of emission inventories in Asia and related literatures.
- Large power plants constructed after 2008 were added as new point sources.
- Allocation factors for spatial and temporal distribution were updated although several emission inventories developed by other research works were utilized (see Table 2).
- Emissions from Japan, Republic of Korea, and Taiwan were originally estimated except for NMVOC evaporative sources (see Table 2).

REASv3 focuses on the long historical trends of air pollutants emissions in Asia. The start year was chosen to be 1950 as severe air pollution in Japan started from the mid-1950s. For the emission inventory framework, there are two major changes from REASv2.1. One is the target species. REASv3 includes the following major air and climate pollutants: SO_2, NO_x, CO, non-methane volatile organic compounds (NMVOC), NH_3, PM_{10}, $PM_{2.5}$, BC, organic carbon (OC), and CO_2. However, CH_4, and N_2O that were included in REASv2.1 are not in the scope of this version. CH_4 is one of important components of SLCP and will be considered in the next version. Another is the target areas. Figure 1 shows the inventory domain of REASv3 which includes East, Southeast, and South Asia. China, India, and Japan have been divided into 33, 17, and 6 regions, respectively to reduce the uncertainties in the spatial distribution. Definition of the sub-regions are the same as for REASv2.1. In REASv3, Central Asia and the Asian part of Russia, which were target areas of REASv2.1 are not included because of the difficulty in collecting necessary data for estimating long historical emissions in these areas. The source categories considered in REASv3 are the same as those in REASv2.1. Major sources include fuel combustion in power plants, industry, transport, and domestic sectors. Non-combustion sources include industrial process, evaporation (NMVOC), and agricultural activities (NH_3). However, NO_x emissions from soil as well as from international and domestic aviation and navigation, including fishing ships are exceptions. They were not included in REASv3. The spatial and temporal resolution are the same as those of REASv2.1. Spatial resolution is 0.25° × 0.25°, except in the case of large power plants, which are treated as point sources. Temporal resolution is monthly.

In REASv3, most emissions were originally estimated. However, several emission inventories from other research works and officially opened data were utilized as summarized in Table 2. NMVOC emissions in Japan and Republic of Korea from evaporative sources were obtained from the Ministry of the Environment of Japan (MOEJ, 2017) and the National Air Pollutants Emission Service of the National Institute of Environmental Research (available at http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do), respectively. For NH_3 emissions from agricultural activities, data of base year (2000 and 2005 for Japan and 2000 for others) were obtained from other research works as follows (see Sect. 2.4): REASv2.1 (Kurokawa et al., 2013: JPEC 2012a, b, c; 2014) for Japan and REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) for other counties and regions. In addition, EDGARv4.3.2 were utilized to create grid allocation factors for road transport sector for all species and manure management for NH_3 (see Sects. 2.4.1 and 2.6, respectively).

In the following sub-sections, general methodologies and data used in REASv3 are overviewed for stational sources, road transport, agricultural sources, other sources, and spatial and temporal distribution. Details of the methodologies such as data sources and treatments, settings of emission factors and emission controls, and related assumptions are provided in the supplement document entitled "Supplementary information and data to methodology of REASv3" (hereafter, this document is expressed as "the Supplement"). In Sect. S2 of the Supplement, details of frame work of REASv3 including definitions of sub-categories of emission sources, and target countries and sub-regions of China, India, and Japan was provided.

2.2 Stationary sources

2.2.1 Basic methodology

Emissions from stationary fuel combustion and industrial processes are traditionally calculated using activity data and emission factors, including the effect of control technologies. In order to increase the accuracy of estimation and to analyze the effects of abatement measures, emissions should be calculated using information on technologies related to emission sources as much as possible. In REASv3, emissions from stationary combustion and industrial processes are estimated based on the following equation:

$$E = \sum_i \sum_j \sum_{k,l} \{A_{i,j} \times F_{i,j,k,l} \times EF_{i,j,k} \times (1 - R_{i,j,l})\} \quad (1)$$

where, *E* represents emission, *i* is the type of activity data, *j* is the type of sector category, *k* is the type of technology related to emission factor, *l* stands for the control technology after emission, *A* is amount of activity data, *EF* is the emission factor of each technology, *R* is the removal efficiency of each technology, and *F* is the fraction rate of activity data for combination of *i*, *j*, *k*, and *l*. When SO_2 emissions from combustion sources are estimated using sulfur contents of fuels, $EF_{i,j,k}$ in eq. (1) is calculated, as follows:

$$EF_{i,j,k} = NCV_{i,j} \times S_{i,j} \times (1 - SR_{i,j,k}) \times 2 \quad (2)$$

where, *NCV* is the net calorific value of fuel, *S* is the sulfur content of fuel, and *SR* is the sulfur retention in ash for combination of *i*, *j*, and *k*. *2* is a factor to convert the value of S to SO_2.

Unfortunately, in the case of Asia, information available on emission factors and removal efficiencies is limited. Even though there is information on the introduction rates of technologies both for emission factors and removal efficiencies, they are available independently. Therefore, for most cases, an average of the removal efficiencies is calculated using the values of each abatement equipment and its penetration rate. Then, the average removal efficiencies are commonly used to calculate the emission factors of each technology.

Note that several sub-sectors in stationary sources such as coke production and cement industry include both combustion and non-combustion emission sources. See Sects. S2.4.1 and S2.4.2 of the Supplement for details.

2.2.2 Activity data

Fuel consumption is the core activity data of the emission inventory of air pollutants and greenhouse gases. For most countries, the amount of energy consumption for each fuel type and sector was primarily obtained from the International Energy Agency (IEA) World Energy Balances (IEA, 2017). For China, province-level tables in the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017) were used. For countries and regions whose energy data are not included in IEA (2017), fuel consumption data were taken from the United Nations (UN) Energy Statistics Database (UN, 2016) and the UN data, which is a web-based data service of the UN (http://data.un.org/). See Sect. S.3.1.1 of the Supplement for definition of fuel types.

One major obstacle in this study was collecting activity data for the entire target period of REASv3, that is from 1950-2015. IEA (2017) includes data from Japan during 1960-2015 and those from other countries during 1971-2015; however, for many countries, fuel types and sector categories, the oldest years when data exist are more later than 1971. Furthermore, past data for sectors do not contain as many categories. For example, coal consumption data in detailed sub-categories of the industrial sector existed in Indonesia only after 2000, but corresponding data are only available for industry total before 1999. In this case, relative ratios of fuel consumption in detailed sub-categories to total industry in 2000 were used to distribute the total industry data to each sub-category for the years before 1999. This procedure is performed for similar cases for all sectors and sometimes for total final consumption. In cases where data did not exist beyond a certain year, fuel consumption data were extrapolated using trends of related data for each sub-category. For example, power generation and amount of industrial products were used to observe trends of fuel consumption in power plant and each industry's sub-category, respectively. Data for long historical trends were obtained from a variety of sources. For example, power generation data and amounts of major industrial products were obtained from Mitchell (1998) and national and international statistics as well as related literatures were surveyed. See Sect. S3.1.2 of the Supplement for details of data sources of fuel consumption and assumptions to estimate missing historical data. For China, data of CESY for each province were available from 1985 to 2015. During 1950-1984, first, total energy data in China were developed based on IEA (2017) and then, fuel consumption in each province was extrapolated using the total data of China in each fuel type and sector category. See Sect. S3.1.3 of the Supplement for details of regional fuel consumption data in China. For countries which used Energy Statistics Database, fuel consumption of each fuel and sector was taken from the UN data (available at http://data.un.org/) for the period between 1990-2015 and was extrapolated using the trend of total consumption of each fuel type obtained from the UN Energy Statistics Database.

As described in Section 2.1, India and Japan have 17 and 6 sub-regions, respectively. Therefore, for them, country total data of IEA (2017) need to be divided for each sub-region. For Japan, energy consumption statistics of each prefecture that were obtained from the Agency for Natural Resources and Energy (available at https://www.enecho.meti.go.jp/statistics/energy_consumption/ec002/results.html) were used as default weighting factors to allocate country total data to the six regions. Similarly, for India, default weighting factors for regional allocation were estimated from TERI (The Energy and Resources Institute) Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018), Annual Survey of Industries (Ministry of Statistics & Programme Implementation, available at http://www.csoisw.gov.in/cms/en/1023-annual-survey-of-industries.aspx), and Census of India (Chandramouli, 2011), among others. In general, details of these weighting factors are less than those of the country's total fuel consumption. In addition, these data are not available for all the years during 1950-2015. Therefore, regional allocation factors for some sectors were developed independently if corresponding proxy data were available. For the power plant sector, generation capacities of each region and year were calculated as proxy data using the World Electric Power Plants Database (WEPP) (Platts, 2018). For India, traffic volumes (see Section 2.3.1) and amount of industrial production in each region (see the last paragraph of this section) were used as proxy data. Details of regional fuel consumption data in India and Japan were provided in Sects. S3.1.4. and S3.1.5, respectively.

Similar to REASv2.1, large power plants are treated as point sources in REASv3 and are updated based on REASv2.1 database. Before 2007, power plants that were classified as point sources were the same as those in REASv2.1 and their information, such as generating capacities, and start and retire years were updated using WEPP. During 2000 to 2007, fuel consumption data were the same as that in REASv2.1. In REASv3, power plants whose start years were after 2007 and generation capacities were larger than 300 MW were added as new point sources. Fuel consumption of new power plants were estimated based on relations between fuel consumption amounts and generation capacities of the point data in REASv2.1. If the (A) total fuel consumption of each power plant in a country is larger than (B) the corresponding data in power plant sector, values of each power plant were adjusted by ratios of (B) per (A). If (B) was larger than (A), differences between (B) and (A) were treated as data of area sources. See also Sect. S3.1.6 of the Supplement for fuel consumption data in power plants.

For emissions from industrial processes, activity data included amount of industrial products. Corresponding data were mainly obtained from related international statistics and national statistics. For example, iron and steel production data were taken from Steel Statistical Yearbook (World Steel Association, 1978-2016) and data for non-ferrous metals and non-metallic minerals were obtained from the United States Geological Survey (USGS) Minerals Yearbook (USGS, 1994-2015). Brick production data were obtained from a variety of sources, such as Zhang (1997), Maithel (2013), Klimont et al. (2017), and the UN data. For China and India, the authors also used internet database services, namely China Data Online (https://www.china-data-online.com/) and Indiastat (https://www.indiastat.com/), respectively, which provided both national and regional statistics. The USGS Minerals Yearbook (USGS, 1994-2015) also provided information on plants in each sub-region of China, India, and Japan. Data in the aforementioned statistics were not available for the early years of the target period of REASv3.1. In such cases, data of Mitchell (1998) were used as factors to extrapolate the activity data until 1950. Details of activity data related to industrial production and other transformation were described in Sect. S4.1 of the Supplement.

2.2.3 Emission factors

Setting up of emission factors and removal efficiencies for stationary combustion and industrial processes are difficult procedures, especially for a long historical emission inventory. In this study, emission factors without effects of abatement measures were set, which were used for the entire target period of REASv3. Then, effects of control measures were set considering their temporal variations, both for abatement measures before emissions such as using low sulfur fuels and low NO_x burners and those after emissions such as flue gas desulfurization (FGD) and electrostatic precipitator (ESP). These settings were done for each country and region based on country and region-specific information. However, such information is still limited, especially in the Asian region. Therefore, default values of unabated emission factors were selected and default removal efficiencies were set to zero. Then, these default values were updated in case information and literature on each country and region were available. For default emission factors, a majority of settings was continuously used from REASv2.1, but some of them, including effects to control measures (net emission factors) were changed to unabated emission factors. Default emission factors were mainly obtained from Kato and Akimoto (1992) for SO_2 and NO_x; Bond et al. (2004), Kupiainen and Klimont (2004), and Klimont et al. (2002, 2017) for PM species; the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006) for CO_2; and the AP-42 (US EPA, 1995), the Global Atmospheric Pollution Forum Air Pollutant Emission Inventory Manual (SEI, 2012), Shrestha et al. (2013), the EMEP/EEA emission inventory guidebook 2016 (EEA, 2016), and other literatures for others.

For country and region-specific settings, in addition to literatures used in REASv2.1 (see Kurokawa et al., 2013), new information, especially for technologies related to settings of emission factors and removal efficiencies was surveyed. Although such information is still limited in Asia, the volume of accessible information on China is relatively large. General information on China in recent years was mainly obtained from Li et al. (2017b) and Zheng (2018). Introduction rates of technologies were obtained from Hua et al. (2016) for cement, Wu et al. (2017) for iron and steel, Huo et al. (2012a) for coke ovens, and Zhao et al. (2013, 2014, and 2015) for a variety of sources. For India, information for technology settings was mainly taken from Sadavarte and Venkataraman (2014), Pandey et al. (2014), Guttikunda and Jawahar (2014), and Reddy and Venkataraman (2002a). For power plants, WEPP database has elements for installed equipment to control SO_2, NO_x, and PM species which were used for settings of emission factors and removal efficiencies of power plants treated as point sources. However, these data are not available for most power plants, especially in Asia. Therefore, in the case of South and Southeast Asia, a variety of literatures, such as Sloss (2012) and UN Environment (2018) were referred to, to set emission factors and removal efficiencies. For Japan, introduction of control technologies for air pollutants were initiated earlier than other countries in Asia. A lot of domestic reports for air pollution and control technologies in power and industry plants published in Japanese, such as MRI (2015), Shimoda (2016), Suzuki (1990), and Goto (1981) were referred to, to determine emission factors, removal efficiencies, and their temporal variations.

Details of emission factors and settings of emission controls for stationary combustion sources were provided in Sect. S3.2 of the Supplement. Those for stationary non-combustion emissions from industrial production and other transformation sectors were described in Sect. S4.2. Activity data and emission factors of NMVOC from chemical industry were obtained from Sects. S5.1.5 and S5.2.5, respectively. Those for NH_3 emissions from industrial production were provided in Sect S8.3.

2.3 Road transport

2.3.1 Basic methodology

Methodology for road transport sector is the same as that of REASv2.1. Equations to estimate hot and cold start emissions (except for SO_2 and CO_2) are, as follows:

$$E_{HOT} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi}\} \quad (3)$$

where, E_{HOT} is the hot emission, i is the vehicle type, NV is the number of vehicles in operation, ADT is the annual distance traveled, and EF_{HOT} is the emission factor. SO_2 emissions are calculated using sulfur contents in gasoline and diesel consumed in road transport sector, assuming sulfur retention in ash is zero. CO_2 emissions are estimated by calculating the consumption amounts of fuels (gasoline, diesel, liquefied petroleum gas, and natural gas) and the corresponding emission factors (IPCC, 2006). Details for SO_2 and CO_2 from road transport were described in S6.2.3 of the Supplement.

Cold start emissions (E_{COLD}) are estimated for NO_x, CO, PM_{10}, $PM_{2.5}$, BC, OC, and NMVOC using the following equation:

$$E_{COLD} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi} \times \beta_i(T) \times F_i(T)\} \tag{4}$$

where, β is the fraction of distance traveled driven with a cold engine or with the catalyst operating below the light-off temperature, and F is the correction factor of EF_{HOT} for cold start emission. β and F are functions of temperature T and are taken from EEA (2016) (See Sect. S6.2.1 of the Supplement for additional information of the settings). For Japan, the ratios of cold start and hot emissions for each vehicle type were estimated from the JEI-DB. Then, cold start emissions were calculated by hot emissions and the ratios for each vehicle type. In REASv3, effects of regulations on cold start emissions were ignored and need to be considered in the next version.

For evaporation from gasoline vehicles, emissions (E_{EVP}) were estimated using the following equation of Tier 1 of EEA (2016):

$$E_{EVP} = \sum_i \{NV_i \times EF_{EVPi}(T)\} \tag{5}$$

where, EF_{EVP} is the emission factor as a function of temperature. For Japan, evaporative emissions in 2000, 2005, and 2010 were obtained from the JEI-DB and those between 2000 (2005) and 2005 (2010) were interpolated. For emissions before 2000 and after 2010, emissions from running loss were extrapolated using trends of traffic volume and those from hot soak loss and diurnal breaking loss were extrapolated by trends of vehicle numbers. See Sect. S6.3 of the Supplement for the NMVOC evaporative emissions.

2.3.2 Activity data

Basic activity data of road transport sector include number of vehicles in operation for each type. Data on the registered number of vehicles are available in the national statistics of each country and the World Road Statistics (IRF, 1990-2018). If these statistics did not contain data until 1950, the numbers were extrapolated using trends of data for aggregated vehicle categories in Mitchell (1998). For China, data for each sub-region were obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986-2016) and the China Data Online. Those for India were taken from TERI Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018) and the Indiastat. A problem that was encountered was that registered vehicles were not always in operation. For India, the number of vehicles obtained as registered vehicles were corrected based on Baidya and Borken-Kleefeld (2009) and Prakash and Habib (2018). For other countries, the number of registered vehicles were considered as those in operation due to lack of information. In addition, to estimate emissions, these numbers must be further divided into vehicles based on each fuel type. However, such information is not easily available in national statistics. In this study, settings of Streets et al. (2003a) and REASv2.1 were used as default and were updated if national information was available, such as He et al. (2005), Yan and Crookes (2009), Sahu et al. (2014), and Malla (2014). If the number of LPG and CNG vehicles were available only for recent years, data were extrapolated using amounts of fuel consumption in road transport sector in IEA (2017).

Emission factors of road transport sector used in this study were given as emission amounts per traffic volumes. Therefore, annual vehicles kilometer traveled (VKT) per each vehicle type need to be set for each country. We used data of Clean Air Asia (2012) for many countries. Clean Air Asia (2012) includes data for China and India, but data of China were estimated based on Huo et al. (2012b) and those of India were set after Prakash and Habib (2018) and Pandey and Venkataraman (2014). For Japan, the total annual VKT for detailed vehicle types were obtained from reports of Pollutants Release and Transfer Register published by the Ministry of Economy Trade and Industry until 2001 (METI, 2003-2017), which was originally estimated from Road Transport Census of Japan developed by the Ministry of Land, Infrastructure, Transport and Tourism. Before 2001, the total annual VKT was extrapolated using data of more aggregated vehicle categories in the Annual Report of Road Statistics (MLIT, 1961-2016) until 1960 and from the Historical Statistics of Japan (Japan Statistical Association, 2006) until 1950.

Details of number of vehicles and annual vehicles kilometer traveled were described in Sect S6.1.1 of the Supplement.

2.3.3 Emission factors

For most countries, road transport is one of major causes of air pollution. In many Asian countries, vehicle emission standards were introduced after the late 1990s and were strengthened in phases (Clean Air Asia, 2014). Therefore, for road vehicles, year-to-year variation of emission factors must be taken into considered for a long historical emission inventory. In REASv3, emission factors of NO_x, CO, NMVOC, and PM species for exhaust emissions from road vehicles were estimated by following procedures:

1. Emission factors of each vehicle type in a base year were estimated.
2. Trends of the emission factors for each vehicle type were estimated considering the timing of road vehicle regulations in each country and the regions and the ratios of vehicles production years.
3. Emission factors of each vehicle type during the target period of REASv3 were calculated using those of base years and the corresponding trends.

The information of road vehicle regulations in each country and regions were taken from Clean Air Asia (2014). For the ratios of vehicle production years, due to lack of information, data for Macau derived from Zhang et al. (2016) were used for Hong Kong, Republic of Korea, and Taiwan and those from Japan Environmental Sanitation Center and Suuri Keikaku (2011) for Vietnam were used for other countries and regions. Then, trends of emission factors were estimated using the above data and information with values of Europe and United States standards. Finally, emission factors used to estimate emissions were calculated for each vehicle type. For most countries, the years just before the regulations for road vehicles began were set as base years and no-controlled emission factors that were used in REASv1.1 and REASv2.1 were adopted for emission factors of the base years. Countries for which information on regulations were not obtained, the no-controlled emission factors were used for the entire target period of REASv3. For China and India, emission factors in 2010 were estimated as base year's data using recently published papers, such as Huo et al. (2012b), Xia et al. (2016), Mishra and Goyal (2014), and Sahu et al. (2014). For Republic of Korea and Taiwan whose emissions were not originally estimated in REASv2.1, emission factors were estimated with high uncertainties based on values of Europe and United States standards, respectively. For Japan, emission factors for each emission standard are available for several vehicle speeds (JPEC, 2012a). Combining these data with information for annual VKT of each vehicle speed, ratios of vehicle ages, and time series of regulation standards, emission of road transport in Japan were calculated. Details of emission factors of exhaust emissions were provided in Sect. S6.2 of the Supplement.

2.4 Agricultural sources

REASv3 includes NH_3 emissions from manure management and fertilizer application in agricultural sources. Approaches similar to REASv2.1 were adopted to estimate historical emissions and develop monthly gridded data. First, annual emissions of each country and sub-region except for Japan and their gridded data for the year 2000 were selected from REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) as base data. For Japan, corresponding base data were obtained from REASv2.1 (Kurokawa et al., 2013: JPEC 2012a, b, c; 2014) for the year 2000 and 2005. Second, trends of emissions during 1950-2015 were estimated for each country and sub-region. Third, annual emissions for the period were calculated using the trends and base data. Fourth, changes in spatial distribution from base years to target years and monthly variations in each country and sub-region were estimated. Finally, monthly gridded data of emissions were developed for 1950-2015. For Japan, emission data during 2001-2004 were interpolated between those in 2000 and 2005. Details for manure management and fertilizer application are described in Sections 2.4.1 and 2.4.2, respectively.

2.4.1 Manure management

Trends in NH_3 emissions from manure management of livestock, except for its application as fertilizer, were estimated based on the Tier 1 method of EEA (2016). In this method, emissions are calculated based on the numbers of livestock and the corresponding emission factors. Statistics on the number of animals, such as broilers, dairy cow, and swine are mainly obtained from FAOSTAT (available at http://www.fao.org/faostat/en/) of the Food and Agriculture Organization (FAO) of the UN from the period between 1961 to 2015. For the years before 1960, data were obtained from Mitchell (1998). National statistics were surveyed for data on provinces, states, and prefectures in China, India, and Japan, respectively to develop activity data for each sub-region. Emission factors are obtained from EEA (2016). For spatial distribution, changes in grid allocation for each country and sub-region from the year 2000 were estimated using EDGARv4.3.2 from 1970 to 2012. Grid allocation factors in 1970 and 2012 were used for the period before and after 1970 and 2012, respectively. For temporal variations, monthly allocation factors are estimated as a function of temperature by referring to the monthly variations of emissions in Japan based on the JEI-DB. Detailed methodologies and data sources for manure management were provided in Sect. S8.1 in the Supplement.

2.4.2 Fertilizer

In most countries, fertilizer application is the largest source of NH_3 emissions. Emission trends after the application of manure and synthetic N fertilizer were estimated using EEA (2016). Manure application is one of the processes of manure management whose emissions trend was calculated based on the number of animals and the corresponding emission factor. For synthetic N fertilizer, trends of total consumption of fertilizer were used in REASv2.1. However, this simple approach causes uncertainties because emission factors are different among types of fertilizer (EEA, 2016). Therefore, in REASv3, emissions from each N fertilizer, such as ammonium phosphate and urea were estimated separately and trends in total emissions were calculated. For spatial distribution, changes in grid allocation factors for each country and sub-region from the year 2000 were estimated using a historical global N fertilizer application map during 1961-2010, developed by Nishina et al. (2017). Data for 1961 and 2010 were used for the period before 1961 and after 2010, respectively. For seasonal variations, monthly factors of China and Japan were determined based on Kang et al. (2016) and the JEI-DB, respectively. For other countries, data from Nishina et al. (2017) have monthly application amounts in each grid. However, there are cases that some months have high factors, whereas the others have almost zero. Referring to Janssens-Maenhout et al. (2015), we adopted the conservative way, such that the highest monthly factor was set at 0.2 and the factors of all months were adjusted accordingly. See Sect. S8.2 for details of methodologies and data sources for emissions from fertilizer application.

2.5 Other sources

NMVOC emissions from evaporative sources are increasing significantly in Asia along with economic growth. Major sources of NMVOC emissions include usage of solvents for dry cleaning, degreasing operations, and adhesive application as well as for paint use. Fugitive emissions related to fossil fuels, such as extraction and handling of oil and gas, oil refinery, and gasoline stations are also important. However, statistics on activity data and information of emission factors for these sources are often less available than those for fuel combustion and industrial processes. In this study, default activity data and emission factors were obtained from REASv2.1 and were updated if information was available in recently published papers (such as Wei et al. (2011) for China and Sharma et al. (2015) for India). In general, activity data of the past years are not available, and, in such cases, proxy data are prepared for trend factors. For example, population numbers were used for dry cleaning and production numbers of vehicles were used for paint application for automobile manufacturing. GDP was used for default trend factors. For emission calculation, the same equation for stationary combustion was adopted. Details of activity data and emission factors for non-combustion sources of NMVOC were provided in Sect. S5 of the Supplement.

In addition to agricultural activities, latrines are an important source of NH_3, especially in rural areas. Activity data are population numbers in no sewage service areas estimated referring settings of REASv2.1 and emission factor were based on EEA (2016) and SEI (2012). Also, humans themselves are sources of NH_3 emissions through perspiration and respiration.

For these sources, population numbers are activity data mainly taken from UN (2018) and emission factors are obtained from EEA (2016). Equation to estimate emission is also the same as that of stationary combustion. Additional data and information for emissions from human and latrines were described in Sects. S8.4 and S8.5, respectively.

In REASv3, aviation and ship emissions including fishing ships are not included, but emissions of fuel combustion in other transport sector (namely, except for aviation, navigation, and road), such as railway and pipeline transport were estimated. Equation (1) is also used for estimating emissions of these sources. See Sect. S7 of the Supplement for additional data and information for other transport sector.

2.6 Spatial and temporal distribution

Procedures for developing gridded emission data were the same as those of REASv2.1. Large power plants were treated as point sources, and longitude and latitude of each power plant were provided. Positions of power plants were surveyed based on detailed information, such as names of units, plants, and companies from WEPP (Platts, 2018). These were searched on internet sites, such as Industry About (https://www.industryabout.com/) and Global Energy Observatory (http://globalenergyobservatory.org/). Positions for newly added power plants in REASv3 as well as those in REASv2.1 were surveyed because some of these services were not available when REASv2.1 was developed. For cement, iron, and steel plants (and non-ferrous metal plants in Japan), REASv3 still did not treat them as point sources due to lack of activity data. However, positions, production capacities, start and retire years for large plants were surveyed similar to power plants and used for developing allocation factors for corresponding sub-sectors. For road transport sector, REASv2.1 used coarse grid allocation data of REASv1.1 with 0.5° × 0.5° resolution. Therefore, in REASv3, grid allocation factors for each country and sub-region, except Japan, were updated using gridded emission data of road transport sector of EDGARv4.3.2 during 1970-2012. Before 1970 (after 2012), data for 1970 (2012) were used. For Japan, gridded emission data of the JEI-DB in 2000, 2005, and 2010 were used to develop grid allocation factors. For the year between 2000 (2005) and 2005 (2010), the JEI-DB data were interpolated. For years before 2000 (after 2010), the JEI-DB data for 2000 (2010) were used. For residential sectors, rural, urban, and total population of HYDE 3.2.1 (Klein Goldewijk et al., 2017) with 5′ × 5′ were used to create allocation factors. Data of HYDE 3.2.1 were available for 1950, 1960, 1970, 1980, 1990, 2000, 2005, 2010, and 2015 and the years between them were interpolated. Spatial distributions of total population were used for grid allocation of all other sources. Detailed methodologies and data sources for grid allocation were provided in Sect. S9.1 in the Supplement.

Methodology to estimate monthly emission data in REASv3 was the same as that of REASv2.1. In general, monthly emissions were estimated by allocating annual emissions to each month using monthly proxy data. Monthly generated power and production amounts of industrial products were used as the monthly allocation factors for power plant sector and corresponding industry sub-sectors, respectively. Basically, monthly factors of REASv2.1 during 2000-2008 were also used in REASv3 and were extended if data existed before (after) 2000 (2008). For the years where surrogate data were unavailable, the data of oldest (newest) year were used before (after) the year. For brick production, monthly allocation factors for Southeast and South Asian countries were estimated referring Maithel et al. (2012) and Maithel (2013). For the residential sector, monthly variations of emissions were estimated using surface temperature in each grid cell, similar to REASv2.1. Surface temperatures during 1950-2015 were taken from NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html). For Thailand and Japan, most monthly factors were set based on country specific information from Thao Pham et al. (2008) and JPEC (2014), respectively. See Sect. S9.2 of the Supplement for details of monthly variation factors.

3 Results and discussion

3.1 Trends of Asian and national emissions

Trends in air pollutants emissions from Asia, China, India, Japan, and other countries are described in this section, mainly focusing on SO_2, NO_x, and BC emissions as they have important roles in both air pollution and climate change. SO_2 and NO_x are precursors of sulfate and nitrate aerosols, respectively, which are the major components of secondary $PM_{2.5}$. NO_x is also a precursor of ozone. Furthermore, BC is a major component of primary $PM_{2.5}$. $PM_{2.5}$ and ozone not only harm human health and ecosystems, but influence climate change. BC and ozone have a warming effect on climate change, whereas sulfate and nitrate aerosols have a cooling effect. Note that all the air pollutant emissions from major countries and regions between 1950 to 2015 categorized based on major sectors and fuel types, are provided in the Supplement material (Figs. S1-S12).

3.1.1 Asia

Table 3 summarizes the national emissions of each species in 2015 and the total emissions from Asia in 1950, 1960, 1970, 1980, 1990, 2000, and from 2010-2015. Figure 2 shows emissions of SO_2, NO_x, CO, NMVOC, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) from 1950 to 2015. Average total emissions in Asia during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO_2: 3.2 Tg, 42.4 Tg (13.1); NO_x: 1.6 Tg, 47.3 Tg (29.1); CO: 56.1 Tg, 303 Tg (5.4); NMVOC: 7.0 Tg, 57.8 Tg (8.3); NH_3: 8.0 Tg, 31.3 Tg (3.9); CO_2: 1.1 Pg, 18.6 Pg (16.5); PM_{10}: 5.9 Tg, 30.2 Tg (5.1); $PM_{2.5}$: 4.6 Tg, 21.3 Tg (4.6); BC: 0.69 Tg, 3.2 Tg (4.7); and OC: 2.5 Tg, 6.6 Tg (2.7). Clearly, all the air pollutant emissions in Asia increased significantly during these six decades. However, this increase was different among the aforementioned species. Growth rates of emissions were relatively large for SO_2, NO_x, and CO_2 because the major sources of these species are power plants, industries, and road transport, for which fuel consumption increased significantly along with economic development in Asia. SO_2 increased before the other species because a majority of the emissions were obtained from the combustion of coal, which is easier to obtain than oil and gas. SO_2, NO_x, and CO_2 emissions increased keenly in the early 2000s, along with rapid growth of emissions of these species in China. For NO_x, combustion of oil fuels, especially by road vehicles, contributed to a large growth of emissions in the latter half of 1950-2015. Growth rates of NMVOC have also increased recently due to an increase in the emissions from road vehicles and evaporative sources, such as paint and solvent usage in accordance with economic growth of Asian countries. On the other hand, rates of growth of CO, PM_{10}, $PM_{2.5}$, BC, and OC are relatively small. One reason is that emissions of these species are mainly from incomplete combustion in low temperature and thus, emissions from power plants and large industry plants are relatively small. Another reason is that a major source of these species is the combustion of coal and biofuels in residential sector, which were relatively large even in past years in Asia. Recently, emissions of these species from industries, including combustion and non-combustion processes are increasing. In addition, gasoline and diesel vehicles have contributed recently to the growth of CO and BC emissions, respectively. Agricultural activities, such as manure management of livestock and fertilizer application, which are major sources of NH_3 are rising to support a growing population in Asia. Although the growth rate of NH_3 emissions is smaller than other species, it still shows an increasing trend.

Differences in the trends of emissions were also observed on the basis of countries and regions. SO_2 and NO_x, emissions from Japan were relatively large in Asia during the 1950s-1970s. Emissions from Japan in 1965 are comparable with and are larger than those of China for SO_2 and NO_x, respectively. In 2015, emissions of SO_2 and NO_x in Japan decreased largely and contribute only about 1.5 and 3.8% of Asia's total emissions, respectively. Similar tendencies were also observed in the case of other species. In 2015, China was the largest contributor of emissions for all the species. Recently, emissions of most species in China have shown decreasing or stable trends. In the case of SO_2, China contributed about 72% of emissions in 2005, but about 49% in 2015. On the other hand, emissions and their relative ratios are increasing in the case of India. Actually, contribution rates of SO_2, NO_x, and BC emissions in India increased from 14%, 16%, and 23% in 2005 to 30%, 22%, and 27% in 2015, respectively. Li et al. (2017c) suggested that, in 2016, SO_2 emissions in India exceeded those in China. Recent increase in air pollutants emissions have also been observed in SEA and OSA. On the other hand, emissions from OEA started to increase slightly later than Japan and then, recently show decreasing trends mainly reflecting trends of emissions from Republic of Korea and Taiwan.

3.1.2 China

Growth rates of all pollutants emissions in China in these 60 years estimated from average during 1950-1955 and 2010-2015 are as follows: 21% for SO_2, 54% for NO_x, 7.0% for CO, 13% for NMVOC, 4.7% for NH_3, 28% for CO_2, 6.8% for PM_{10}, 6.1% for $PM_{2.5}$, 5.5% for BC, and 2.7% for OC. It was observed that emissions of all pollutants increased largely during these six decades, but most species reached their peaks up to 2015 as shown in Fig. 2. Exceptions to this were NMVOC, NH_3, and CO_2; however, their growth rates are at least small or almost zero. Emission trends in China for all the pollutants in each sector and for each fuel type during 1950-2015 were presented in Figs. S1 and S2, respectively. Figure 3 shows recent trends in actual emissions (solid colored areas) and reduced emissions by control measures (hatched areas) from each sector for SO_2, NO_x, and BC during 1990-2015 in China. The reduced emission by control measures was the difference between emissions calculated without effects of all control measures (such as FGD, ESP, using low sulfur fuels, regulated vehicles, etc.) and actual emissions. Total CO_2 emissions were also plotted for each panel of Fig. 3 as an indicator of energy consumption. Note that reduced emissions here do not include effects of substitution of fuel types, such as from coal to natural gas.

For SO_2, most emissions in China were from coal combustion which controlled trends of total emissions. SO_2 emissions in China increased rapidly in the early 2000s, but decreased after 2006 and showed a continuous decline until 2015. Drastic changes in the 2000s were mainly caused by emissions from coal-fired power plants, which increased rapidly along with large economic growth and later decreased due to the introduction of FGD based on the 11th Five Year Plan of China. After 2011, control measures for large industry plants started to become effective and as a result, total emissions in 2015 became comparable with those in 1990. Without effects of emission controls, emissions from power plants and industry in 2015 would be 3.7 and 2.6 times higher than those in 2000, respectively. In this study, the emissions in 2015 were estimated to be reduced by about 90% for power plants and 76 % for industry. On the other hand, even without emission controls, SO_2 emissions from power plants were almost stable after 2010. The same tendencies were also found in CO_2. One considerable reason is an increasing energy supply from nuclear power plants. According to IEA (2017), the total primary energy supply from nuclear power plants increased rapidly recently and those in 2015 were about 2.3 time higher than in 2010.

Similar to SO_2, NO_x emissions increased rapidly from the early 2000s, but continued to increase until 2011 and then, started to decline. In the 2000s, low NO_x burner to power plants and regulation of road vehicles were introduced, but their effects were limited. From 2011, introduction of denitrification technologies, such as selective catalytic reduction (SCR) to large power plants and regulations for road vehicles were strengthened based on the 12th Five Year Plan of China. Three major drivers of NO_x emissions in China are power plants, industry sector, and road transport. If no emission mitigation was considered, their emissions would be increased by 3.6, 3.0, and 4.7 times from 2000 to 2010, respectively. In 2015, reduction rates of emissions due to emission controls were about 61%, 19%, and 62% for power plants, industry, and road transport respectively. As a result, in 2015, NO_x emissions were about 81% of their peak values in 2011. In 2015, actual NO_x emissions from industry sector were larger than those from power plants and road transport which were comparable each other. Major industries such as iron and steel, chemical and petrochemical, and cement industries were large contributor of NO_x emissions in China.

For BC, emissions also increased from early 2000s, but growth rates were smaller than SO_2 and NO_x due to the effects of control equipment in the industrial sector. Actually, trends of BC emissions assuming no emission controls were close to those of CO_2 and the BC emissions in 2015 were increased by 2.2 times from 2000. The emissions in 2015 were reduced by about 41% by abatement measures in industry plants and 9% by regulations especially for diesel vehicles. In 2015, large contributors in industry sectors were brick production, coke ovens, and coal combustion in other industry plants. Another reason of relatively small growth rates could be that BC emission factors for coal-fired power plants are originally low. Recently, BC emissions from residential sector as well as industrial sector show decreasing trends. In this study, the reductions in BC emissions in residential sector were mainly caused by a decrease in emissions from biofuel combustion. During 2010 to 2015, consumptions of primary solid biofuels were reduced about 28%, whereas consumption of natural gas and liquefied petroleum gas increased about 62% in the residential sector.

For CO, most emissions in the 1950s were from residential sectors and gradually increased with increasing coal consumption in the industrial sector. CO emissions increased largely in 2000s due to coal combustion and iron and steel production processes. Recently, CO emissions have seen a decline. A major reason for this declining trend is the decrease in biofuel consumption in residential sector and the phasing out of shaft kiln with high CO emission factor in the cement industry. NMVOC emissions increased significantly from the early 2000s, similar to other species. However, their major sources were different from others. Recent increasing trends are not caused by stationary combustion sources, but by road transport and evaporative sources, such as paint and solvent use. In particular, emissions from non-combustion sources increased largely from 2000 to 2015 (about 3.7 time) and as a result, their contribution rate in 2015 was about 65%. Growth rates of NMVOC emissions tended to slow down around 2015, but emissions increased almost monotonically after the 2000s. NH_3 emissions were mostly from agricultural activities. In China, emissions from fertilizer application showed a significant increase from the early 1970s to the early 2000s. In recent years, NH_3 emissions are almost stable. For PM_{10} and $PM_{2.5}$, majority of the emissions are from the industrial sector, followed by residential sector and power plants. Emissions increased largely from the early 1990s mainly due to coal combustion and industrial processes, especially in cement plants. Compared to SO_2 and NO_x, growth rates of PM_{10} and $PM_{2.5}$ emissions during the early 2000s were small, and later decreased due to the effects of control equipment in industrial plants. OC emissions were mostly from biofuel combustion in the residential sector. Contributions from the industrial sector has been increasing recently, but total OC emissions have decreased due to reduced usage of biofuels. CO_2 emissions were mainly controlled by coal combustion and their trend were similar to those of SO_2, NO_x, and BC without emission controls as shown in Fig.3. After 2011, CO_2 emissions in China were found to be almost stable. As described above, one reason is a trend of emissions from power plants. In addition, emissions from coal combustion in industry sectors were slightly decreased from 2014 to 2015.

3.1.3 India

Growth rates of air pollutants emissions in India based on averaged values during 1950-1955 and 2010-2015 are as follows: 19% for SO_2, 23% for NO_x, 4.2% for CO, 5.3% for NMVOC, 3.1% for NH_3, 8.9% for CO_2, 4.8% for PM_{10}, 4.0% for $PM_{2.5}$, 4.8% for BC, and 2.8% for OC. Figures S3 and S4 provide trends of emissions in India from each sector and fuel type for all the pollutants, respectively, from 1950 to 2015. In general, all the air pollutants show monotonous increase from 1950 to 2015 and growth rates (especially of recent years) are larger for SO_2, NO_x, NMVOC, and CO_2, which is similar to the case of Asia.

Figure 4 shows trends in emission of SO_2, NO_x, and BC from each fuel type as well as sector with total CO_2 emissions during 1950-2015 in India. Clear differences were seen in the structure of emissions in these species. For SO_2, large parts of emissions were from coal combustion in power plants and industry sector. SO_2 emissions in 2015 were about 3.3 times larger than those in 1990 and contribution rates of the increases from power plants and industry sectors were about 66% and 33%, respectively. Trend so total NO_x, emissions were close to those of SO_2 and contributions from coal-fired power plants were also large. In addition, for NO_x, contribution from road transport especially diesel vehicles were comparable with those of power plants. Around the year 2005, the contributions from road transport were almost the same or slightly larger than power plants. However, from 2005 to 2015, growth rates of NO_x emissions from power plants were about twice higher than those of road transport emissions. For BC, contributions from the residential sector and biofuel combustion were large, especially in the 1950s-1960s. Contribution rates of residential sector were 73% in 1950 and 38% in 2015, and those of biofuel combustion, which were mainly used in residential sector and some parts are used in industry sector were 86% in 1950 and 45% in 2015. On the other hand, recent increasing trends of BC emissions were also caused by growth of emissions from diesel vehicles and industry sector. From 1990 to 2015, contribution rates of increased emissions from industry, road transport, and residential sectors were 27%, 43%, and 23%, respectively. For recent trends, relative ratios of SO_2 emissions from power plants were increased from 43% to 59% during 1990-2015. For NO_x, contribution rates from both power plants and road transport were increased and accounted for about 75% of the total emissions in 2015. Even in 2015, about half of the BC emissions were from the residential sector. However, as previously described, recent emission growths were mainly caused by the industrial sector and road transport. These tendencies were similar to Japan and China during their rapid emission growth periods. These features were consistent with trends of CO_2 emissions. Before the mid-1980s, majority of CO_2 emissions were from biofuel combustion and the trends were close to those of BC. Then, recently, contributions from fossil fuel combustion increased largely and trends of CO_2 became close to those of SO_2 and NO_x, especially after the early 2000s.

Trends and structure of CO emissions were similar to those of BC but contribution rates of the residential sector were larger and those from road transport (mainly from gasoline vehicle) were smaller, as compared to BC. On the other hand, for recent trends, half (51%) of increased emissions during 2005 and 2015 were from industry sector. Similar tendency was also found in OC; however, relative ratios of emissions from residential sector were much larger (about 71% in 2015) and those of industry and road transport sectors were much smaller. For PM_{10} and $PM_{2.5}$, a majority of the emissions was from residential and industrial sectors. Both amounts were almost comparable in PM_{10} and those from residential sectors were larger in $PM_{2.5}$. Different from BC and OC, contributions from coal-fired power plants exist in PM_{10} and $PM_{2.5}$ whose contribution rates in 2015 are about 20% and 13%, respectively. For NMVOC, most emissions were from biofuel combustion before the 1980s. Later, emissions from variety of sources, such as road transport, extraction and handling of fossil fuels, usage of paint and solvents are increasing and are controlling recent trends. For increases of emissions from 1990 to 2015, about 52% were from stationary combustion and road transport and the rest were from stationary non-combustion sectors such as paint and solvent use. Most NH_3 emissions are from agricultural activities. Contributions from manure management and fertilizer use were comparable before 1980s. However, emissions from fertilizer application have increased largely which are now determining recent trends.

3.1.4 Japan

As described in Sect. 3.1.1, trends of air pollutants emissions in Japan were different from other countries and regions in Asia. The trends from each sector and fuel type during 1950-2015 in Japan were shown in Figs. S5 and S6. Compared to the rest of Asia, emissions of all species in Japan except CO_2 were reduced significantly after reaching peak values. In addition, peak years were mostly 40 years ago (about 1960 for PM_{10}, $PM_{2.5}$, and OC, 1970 for SO_2 and CO, 1980 for NO_x and NH_3, 1990 for BC, and 2000 for NMVOC). Figure 5, similar to Fig. 3, shows trends of actual emissions (solid colored areas) and reduced emissions by control measures (hatched areas) from each sector for SO_2, NO_x, and BC during 1950-2015. Total CO_2 emissions were also plotted to each panel of Fig. 5. CO_2 emissions increased rapidly in the 1960s and have generally continued to increase, but growth rates are much smaller than those in the 1960s reflecting trends of economic status of Japan.

SO_2 emissions, especially from power plants and industry sector increased significantly in the 1960s (reflecting the rapid economic growth) and caused severe air pollutions in Japan. In the 1950s, more than half the emissions were from coal combustion and then, contributions from heavy fuel oil increased rapidly in the 1960s (more than 50% around the peak year). In order to mitigate air pollution, first, regulation of sulfur contents, especially in heavy fuel oil, were strengthened. Then, desulfurization equipment was mainly introduced from the mid-1970s. As a result, about 68%, 84%, and 93% of the SO_2 emissions were reduced by regulatory measures in 1975, 1990, and 2015, respectively. Furthermore, although coal consumption in power plants increased in the 1990s, SO_2 emissions almost did not change due to these measures. For trends of SO_2 emissions assuming without emission controls and those of CO_2, there are clear differences in the 1970s and after the 1980s. The causes of the differences in the 1970s were decrease of heavy fuel oil consumption whose contribution rates on SO_2 were much higher than CO_2. On the contrary, causes of the differences in the 1980s were increasing consumption of gas and light fuel oil whose sulfur contents were small.

NO_x emissions also increased rapidly from the 1960s mainly by steep increase of traffic volumes and fossil fuel combustion in power and large industry plants. The largest contribution to NO_x emissions during the peak periods was from road transport sector, that is greater than 50% of total emissions. Regulations for road vehicles became effective from the late 1970s but an increase in the number of vehicles partially cancelled the effects. For stationary sources, the number of introduced denitrification equipment increased largely in the 1990s. As a result, NO_x emissions peaked later; furthermore, reduction rates after the peak were smaller compared to that of SO_2. From 1975 to 2015, emissions assuming without emission mitigations would be increased by about 2.0 times for power plants and 2.4 times for road transport. In 2015, by emission abatement equipment for power plants and control measures for road vehicles, the emissions were reduced by 77% and 90%, respectively. As a result, the reduction rate of total NO_x emissions in 2015 was 78%, but it was smaller than SO_2 as described above.

For BC, contributing sectors changed during 1950-2015. In the 1950s, most emissions were from industries and residential sectors and their amounts were almost comparable. After the 1960s, both types of emissions declined, but reasons for declines were different. In the 1950s, coal and biofuels, which have large BC emission factors were mainly used in residential sectors. However, these fuels were substituted for cleaner ones, such as natural gas and liquefied petroleum gas which reduced BC emissions significantly. Emissions in industrial sectors decreased gradually after the 1960s due to the introduction of abatement equipment for PM. Instead, emissions from road transport sector from diesel vehicles increased from the late 1960s to around 1990. Then, regulations for road vehicles were strengthened and BC emissions were reduced largely from peak values. Before 1986, emission controls for BC were only considered for stationary sources. In 1985, by effects of abatement equipment to power and industrial plants, emissions were reduced by about 58% from those assuming no emission controls. Then, by introducing regulations for diesel vehicles, the reduction rates became about 91% in 2015. For CO and OC, most emissions in 1950s were from biofuel combustion in the residential sector. CO and NMVOC emissions in road transport increased largely in the 1960s and then decreased gradually, similar to the case of NO_x. Recently, a majority of NMVOC emissions were from evaporative sources, such as paint and solvent use. These started to increase from the 1980s and then decreased after 2000. Emissions of CO and OC from the industrial sector showed a similar increase before 1970, whereas OC emissions started to decrease due to control equipment for PM species and CO emissions were almost stable after 1970. The majority of NH_3 emissions in Japan was from agricultural activities, especially manure management; however, contributions from latrines were also large in the past years. Overall, NH_3 emissions increased from 1950 to the 1970s but, showed slightly decreasing trends after the 1990s. PM_{10} and $PM_{2.5}$ emission trends were almost the same. The majority of emissions was from the industrial sector, which grew during the 1950s but decreased largely in the 1970s due to the effects of abatement equipment for PM. Contributions from the residential sector were relatively large from the 1950s to the 1960s. Furthermore, contributions from road transport increased from the 1970s and started to decrease after 1990, similar to BC.

3.1.5 Other regions

Similar to India, air pollutant emissions in SEA and OSA tended to increase during these six decades. Figures S7 and S8 (S11 and S12) provide trends for all the air pollutant emissions in SEA (in OSA) for each sector and fuel type, respectively, from 1950 to 2015. Figures 6 and 7 show emission trends of SO_2, NO_x, and BC for each sector category and contribution rates of each country from 1950-2015 in SEA and OSA, respectively. Total CO_2 emissions were also plotted to upper panels of Figs. 6 and 7.

Contributing sources and their relative ratios in SO_2, NO_x and BC emissions are generally close between these regions. For both the regions, major sources of SO_2 emissions are power plants and industry sector. For fuel types, contributions from heavy fuel oil were large in the case of SO_2 emissions in OSA and were almost comparable to those of coal in SEA during the 1990s. After 2010, emissions from coal-fired power plants in SEA increased rapidly which were doubled during 2010-2015. On the other hand, in OSA, heavy fuel consumption in power plants increased by 1.8 times from 2005 to 2015 which mainly caused the large increase of SO_2 emission. For NO_x, majority of the emissions were from road transport, mainly diesel vehicles. This controlled the recent trends in both regions. Contributions from gasoline vehicles were small in OSA, but relatively large in SEA (about 16% in 2015). On the other hand, NO_x emissions from natural gas vehicles increased from the 2000s in OSA and contribution rates in road transport sector were more than 15% after the late 2000s. Recently, similar to SO_2, NO_x emissions from power plants have been increasing by coal and heavy fuel oil combustion in SEA and OSA, respectively. From 2010 to 2015, increases of emissions were mainly caused by power plants in both regions (about 67% for SEA and 82% for OSA). Although trends are almost stable, emissions from biofuel combustion in the residential sector are relatively large in OSA. BC emissions are mostly from biofuel combustion in the residential sector, especially in OSA. and increased constantly during the period of REASv3. After the late 2000s, BC emissions from road transport show decreasing trends due to effect of emission regulations especially in SEA. Relations between trends of SO_2, NO_x, BC, and CO_2 emissions were similar to the case of India that trends of CO_2 were close to those of BC before the 1980s and then those of SO_2 and NO_x after the 1990s. In the case of country-wise emissions, currently, the largest contributing countries are Indonesia and Pakistan in SEA and OSA, respectively. In 2015, the second and third highest contributing countries in SEA were Philippines and Vietnam for SO_2, Thailand and Philippines for NO_x, and Vietnam and Thailand for BC. Relative ratios of SO_2 emissions in Thailand were large in the early 1990s but decreased significantly due to the introduction of FGD in large coal-fired power plants. For OSA, the second highest contributing country is Bangladesh; Sri Lanka is ranked third for SO_2 and NO_x and Nepal for BC.

Emission trends in OEA from each sector during 1950-2015 were presented for all the air pollutants in Figs. S9 and S10. Emission trends in Republic of Korea and Taiwan were similar to those of Japan. SO_2 emissions increased rapidly in the 1970s and reduced largely from their peak values due to the introduction of low sulfur fuels and FGD. NO_x emissions started to increase steeply from the 1980s due to emissions from road vehicles, in addition to those from power and industry plants. Then, NO_x emissions decrease after 2000 due to regulations related to road vehicles and the introduction of control equipment to power plants. However, their rate of decrease was lower than that of SO_2. BC emission trends were similar to those of NO_x until around the year 2000, but the ratio of decrease after 2000 is much larger than that of NO_x. The differences of reduction rates of emissions between NO_x and BC were caused by effects of emission controls in road transport sector. These features and drivers of trends were generally similar to the case of Japan. For Democratic People's Republic of Korea, emissions of SO_2, NO_x, CO_2, and PM species decreased and those of CO, NMVOC, and NH_3 were almost stable recently. The recent decreasing trends were mainly caused by coal consumption amounts in industry sector. For Mongolia, emissions of all the air pollutants, except PM species, show increasing trends recently. The increasing trends were mainly caused by coal-fired power plants for SO_2 and CO_2, road transport for NO_x, CO, NMVOC, and BC, and domestic sector for OC. For PM_{10} and $PM_{2.5}$, due to effects of abatement equipment in power plants, emissions were almost stabilized after 2000. Note that information of these two countries are limited and therefore uncertainties are large.

3.2 Spatial distribution and monthly variation

Figure 8 presents the emission map of SO_2, NO_x, CO, NMVOC, NH_3, $PM_{2.5}$, BC, and OC in 1965 and 2015 at 0.25° × 0.25° resolution. Emission maps of CO_2 and PM_{10} are presented in Fig. S13. In 1965, high emission grids appeared in industrial areas of Japan, especially for NO_x, SO_2, and CO_2. On the other hand, high emission grids were seen in wide areas in China and India, for CO and PM species, especially OC. This is because emissions of these species were mainly from the residential sector and small industrial plants. In 2015, high emission areas for all species clearly appeared in China and India, especially in the northeastern area, around the Sichuan province, and Pearl River Delta for China and Indo-Gangetic Plain, around Gujarat, and southern area for India. High emission areas of SO_2 and PM species in Japan disappeared or shrinked in 2015 compared to 1965, but still remained in the NO_x, CO, NMVOC, and CO_2 maps. In SEA, high emission areas were seen in the Java island of Indonesia and around large cities, such as Bangkok (Thailand) and Hanoi (Vietnam). NH_3 and OC emissions, whose major sources were agriculture and residential sector, respectively were found in relatively large areas of China, India, and SEA.

As described in Section 2.6, seasonality of emissions is taken into considered for sectors where proxy data for monthly profiles were available or could be estimated. Monthly variations of total emissions of SO_2, NO_x, BC, and NH_3 are shown for China, India, Japan, SEA, OEA, and OSA for the year 2015 in Fig. 9. For SO_2 and NO_x, monthly variations were generally small. In China, emissions were slightly larger in the second half of the year. Monthly factors of SO_2 emissions in OSA were high from December to May and low during July and September due to the timings of brick production. For BC, emissions in winter season were relatively large, especially in China and OEA. This seasonality was mainly determined by fuel consumption in residential sector for the purposes of heating. Therefore, monthly variations of BC emissions were smaller in SEA. For NH_3, seasonality of emissions was controlled by the seasonality of emissions from fertilizer application and manure management. In China, Japan, and OEA, peaks of emissions appeared during summertime. Monthly variations of emissions in the whole of SEA were small, but seasonality was different from each country. Finally, it must be noted that monthly variations of emissions in each grid were different to each other because they were determined by monthly profiles of major emission sources in each grid cell.

3.3 Comparison with other inventories

In this section, estimated emissions of REASv3 were compared with other global, regional, and national bottom-up inventories and several top-down estimates. Figures 10 and 11 compare the results of REASv3 with other studies for SO_2, NO_x, and BC emissions in China and India, respectively. For other species, results based on comparison with China are presented in Fig. S14 and those with India are shown in Fig. S15. Furthermore, Figures S16-S19 provide the comparisons of emissions from Japan, SEA, OEA, and OSA, respectively. In Figs. 10, 11, and S14-S19, error bars were plotted in 2015, 1985, and 1955 of emissions in REASv3. These error bars were based on uncertainties estimated in this study for corresponding emissions. See Sect. 3.4 for details about the uncertainties of emissions in REASv3. Note that as described in Sect. 2.1, emissions from domestic and fishing ships are not included in REASv3. Therefore, corresponding data need to be excluded from values of other inventories in the comparisons. This procedure was done for REAS series, EDGARv4.3.2, CEDS, and several research works. For other inventories where emissions from domestic ship were not available independently, total emissions were plotted in the figures. It was confirmed that other sources out of scope of REASv3 such as open biomass burning were not included in the other inventories.

3.3.1 China

For long historical trends of SO_2 emissions in China, most studies generally agreed with the trends of REASv3 although values of REASv3 during 1995 and 2005 were slightly larger than other inventories. Emissions increased almost monotonically until around 1995 and became stable during the late 1990s. Then, emission increased rapidly from the early

2000s and started to decrease from the late 2000s. However, the decreasing rates were different especially after 2010. Recent rapid decreasing tendency in REASv3 was similar to that of Zheng et al. (2018), but decreasing rates of other studies such as Xia et al. (2016) and Sun et al. (2018) were smaller than REASv3. Values of REASv3 were slightly larger than REASv2.1 during 2000-2005, but the discrepancies were reduced due to a larger decreasing rate of REASv3. For top-down estimates (Qu et al., 2019 [based on retrieval products by National Aeronautics and Space Administration (NASA) standard (SP) and Belgian Institute for Space Aeronomy (BIRA)]; Miyazaki et al., 2020), emission amounts were smaller than most bottom-up inventories, but all top-down results showed large decreasing trends after the late 2000s.

Variability of NO_x emissions among estimations plotted in Fig. 10 was smaller than that of SO_2. NO_x emissions in most results increased largely in the 2000s and then decreased or stabilized. Growth rates of Sun et al. (2018) were smaller than others after 2005, but showed similar decreasing trends after 2010. Values of CEDS were slightly larger than other studies. Similar to SO_2, values of top-down estimates (Ding et al., 2017 [based on OMI and GOME-2]; Itahashi et al., 2019; Miyazaki et al., 2020) were generally smaller than those of bottom-up results. But, top-down emissions showed similar tendencies that emission increased until the early 2010s and turned to decrease. Trends of Itahashi et al. (2019) where emissions in 2008 of REASv2.1 were used as a priori data were close to those of REASv3.

Compared to SO_2 and NO_x, relatively large discrepancies were observed in BC emissions among plotted results in Fig. 10. Emissions of REASv3 increased until 1995, slightly decreased during the late 1990s, increased from the early 2000s and then, turned to decrease from the early 2010s. The decreasing rate in the late 1990s of Wang et al. (2012) was much larger than that of REASv3. On the other hand, emissions of Klimont et al. (2017) increased from 1995 to 2000. The majority of results showed increasing trends during the early 2000s, but the following trends were different. Emissions of CEDS increased constantly after 2005, but those of Wang et al. (2012) decreased after 2005 and then started to increase slightly after 2010. BC emissions of both REASv3 and Zheng et al. (2018) decreased from the early 2010s, but the ratio of decrease was larger in Zheng et al. (2018). Values of BC emissions of REASv3 were larger than those of REASv2.1, especially in the early 2000s, but the difference in 2008 was small. For trends and emission amounts of PM_{10} and $PM_{2.5}$, tendencies of relationships among each result were similar. The majority of results showed clear decreasing trends after 2005 except for REASv2.1, EDGARv4.3.1 and Klimont et al. (2017). For OC, most results decreased from 1995 to 2000 and then increase from the early 2000s. After 2005, trends of OC emissions were different among studies.

CO emissions trends were relatively similar among most studies. Increasing rates after the early 2000s are close except for EDGARv4.3.2, but emission amounts of REASv3 were smaller than other studies before 2010. After 2010, the majority of results showed decreasing trends which agreed with top-down estimates (Jiang et al., 2017 [A: MOPITT Column, B: MOPITT Profile, and C: MOPITT Lower Profile]; Zheng et al., 2019b; Miyazaki et al, 2020). However, before the late 2000s, the trends of CO emissions were much different between bottom-up inventories and top-down results. For NMVOC, most studies showed significant increasing trends after the early 2000s. Compared to bottom-up inventories, top-down estimates of Stavrakou et al. (2017) were almost stable between 2007 and 2012, but increased rapidly after that. Values of REASv3 were generally smaller than others before 2010. Differences among studies of NH_3 emissions were large not only in emission amounts, but also in temporal variations. REAS inventories, CEDS, and EDGARv4.3.2 generally showed increasing trends. On the other hand, trends of MEICv1.2 and Zheng et al. (2018) were almost stable after 2000 and the results of Kang et al. (2016) showed decreasing trends after the mid-2000s. Emissions of REASv3 were also almost stable after 2010.

3.3.2 India

For SO_2, emissions of most bottom-up inventories showed monotonically increasing trends. However, after the 1990s, two different emission pathways were shown among studies. The growth rates of REASv3 were close to those of Klimont et al. (2013), CEDS (scaled to REASv2.1 for India; Hoesly et al., 2018), Streets et al. (2000), and REASv2.1. On the other hand, the increasing rates of national studies by Garg et al. (2006), Sadavarte and Venkataraman (2014) and Pandey et al. (2014) were smaller than those of REASv3. In 2005, top-down estimates of Qu et al. (2019) were close to results of Sadavarte and Venkataraman (2014) and Pandey et al. (2014). Another top-down emissions of Miyazaki et al. (2020) were smaller than other inventories. Both bottom-up and top-down emissions after 2005 show increasing trends, but growth rates of bottom-up inventories were higher than those of top-down estimates.

NO_x emissions of REASv3 also increased monotonically during 1950-2015 and the majority of other bottom-up inventories generally agreed with the trends including national studies of Sahu et al. (2012). However, similar to SO_2, growth rates of Venkataraman (2014) and Pandey et al. (2014) were smaller than REASv3 although emission amounts in 2000 and 2005 were almost comparable each other. For the increasing rates, those of top-down estimates of Itahashi et al (2019) using REASv2.1 as a priori emissions were close to those of REASv3. On the other hand, growth rates of another top-down results of Qu et al. (2019) were similar with those of Sadavarte and Venkataraman (2014) and Pandey et al. (2014). Emission amounts of the top-down estimates were much higher than REASv3.

For BC, as in the case of China, discrepancies among studies plotted in Fig. 11 were large. These tendencies were also found in the comparisons of PM_{10}, $PM_{2.5}$, and OC emissions provided in Fig. S15. Generally, the majority of bottom-up emission inventories of PM species showed slightly continuous increasing trends and growth rates were smaller than those of SO_2 and NO_x. On the contrary to the case of SO_2 and NO_x, emissions of BC and $PM_{2.5}$ of REASv3 were slightly smaller than those of Sadavarte and Venkataraman (2014) and Pandey et al. (2014), but their growth rates were almost comparable.

Amounts and trends of CO emissions compared in Fig. S15 generally agreed well except for REASv1.1 which were much higher than others. Emission increased almost constantly until around 2005 ant then growth rates increased slightly. Values of REASv3 were much smaller than top-down results of Jiang et al., 2017 [A: MOPITT Column, B: MOPITT Profile, and C: MOPITT Lower Profile] and Miyazaki et al. (2020). However, recent growth rates of REASv3 were close to those of top-down estimates except for Jiang et al. (2017) [C]. For NMVOC, plotted results were generally comparable except for REASv2.1 and CEDS and indicated increasing trends of emissions. Similar to the case of SO_2 and NO_x, growth rates of REASv3 were smaller than those of Sadavarte and Venkataraman (2014) and Pandey et al. (2014). For NH_3, a comparison of the emissions in Fig. S15 show similar increasing trends. Differences in emission amounts are also relatively small, except for EDGARv4.3.2.

3.3.3 Other regions

Comparisons of emissions in Japan between REASv3 and other studies were provided in Fig. S16. For tends of SO_2 emissions in Japan, the majority of studies agreed with results of REASv3 that rapid increases in the 1960s, keen decreases in the 1970s, and gradually decreasing trends except for EDGARv4.3.2 and Streets et al. (2000), whose values were lager and smaller, respectively. For NO_x, emissions amounts of REASv3 were larger than those of most studies especially before 2000, except for CEDS (scaled to preliminary historical data of REAS for Japan; Hoesly et al., 2018), Kannari et al. (2007), Zhang et al. (2009) based on Kannari et al (2007) and Fukui et al. (2014). For PM species, the majority of results in Fig. S16 agreed with decreasing trends of REASv3 after 1990. On the other hand, emissions of BC and OC of CEDS increased almost monotonically until their peak around 1990. These tendencies were much different from REASv3. For CO, emission amounts of REASv3 were larger than other results of especially REASv1, EDGARv4.3.2, and CEDS. However, after 2000, emissions and their decreasing trends of other studies were generally comparable to those of REASv3. For NMVOC, results of REASv3 after 2000 generally agreed well with other studies which showed large decreasing trends except for EDGARv4.3.2 and Zhang et al. (2009) based on Kannari et al. (2007). Trends of NH_3 emissions shown in Fig. S16 were similar except for EDGARv4.3.2 before the mid-1990s which showed larger growth rates. Emission amounts of REASv3 were smaller than national inventories by Kannari et al. (2001) and Fukui et al. (2014).

For SEA (see Fig. S17), increasing trends and amounts of SO_2 emissions of REASv3 agreed with other results except for CEDS in the 1990s, Zhang et al. (2009), and Klimont et al. (2013). In CEDS, emissions decreased keenly during the late 1990s. A similar feature was also seen in REASv3 but its rate of decrease was much smaller. For NO_x, all results plotted in Fig. S17 indicated monotonically increasing trends of emissions and agreed well until the early 2000s. After that, growth rates of REASv3 became larger than EDGARv4.3.2 and smaller than CEDS (scaled to REASv2.1 for SEA; Hoesly et al., 2018). For BC, REAS series and CEDS showed similar growth rates until around 2005. On the other hand, increasing rates of Klimont et al. (2017) and EDGARv4.3.2 after 1990 were much smaller and close to those of REASv3 after 2005.

Most results of SO_2 emissions in OEA in Fig. S18 show increasing and decreasing trends from the late 1960s and the early 1990s, respectively, although amounts in CEDS from 1970 and 2000 were much smaller. For NO_x, all results agreed well until the late 1980s and REASv3, REASv1.1 and EDGARv4.3.2 showed similar increasing trends until around 2000. Emissions of CEDS became almost stable after the late 1980s and started to decrease after 2005. The decreasing rates of REASv3 and CEDS are close after 2005. On the other hand, emissions of EDGARv4.3.2 were not changed largely after around 2000. The similar tendencies were shown in the case of SO_2. BC emissions of REASv3 and CEDS showed similar trends until 2000. Then, emissions of REASv3 decreased almost monotonically, while those of CEDS were almost stable. Similarly, decreasing rates of EDGARv4.3.2 after 2000 were much smaller than those of REASv3.

For OSA, increasing trends and amounts of SO_2 and NO_x emissions were generally similar among studies plotted in Fig. S19. SO_2 emission of Streets et al. (2003a) and Zhang et al. (2009) and NO_x emissions of CEDS (scaled to REASv2.1; Hoesly et al., 2018) were higher than other results. For BC, discrepancies among studies were larger than those of SO_2 and NO_x, but similar small monotonically growth rates were shown in all results.

3.3.4 Relative rations of emissions from each country and region in Asia

Figure 12 compares trends of total emissions in Asia and relative ratios of emissions from China, India, Japan, SEA, OEA, and OSA among REASv3, CEDS, and EDGARv4.3.2 for SO_2, NO_x, and BC. Comparisons of other species are presented in Fig. S20. From 1950 to the early 2000s, total SO_2 emissions in Asia of all inventories showed similar results. For relative ratios, REASv3 and CEDS values were similar until the mid-2000s. Contributions from Japan were relatively large from 1950 until around 1970 and then, decreased keenly. This was also found in EDGARv4.3.2, but the rate of decrease was smaller than that of REASv3 and CEDS. Then, while emissions of REASv3 decreased largely after the mid-2000s, those of CDES and EDGARv4.3.2 continued to increase. These discrepancies were mainly due to different trends of emissions from China. Actually, after the mid-2000s, relative ratios of SO_2 emissions in China were stable in CEDS and EDGARv4.3.2, but those in REASv3 decreased significantly. Recently, increasing trends of relative ratios of SO_2 emissions in India are a common feature in REASv3, EDGARv4.3.2, and CEDS.

For NO_x, Asia total emissions of REASv3 and EDGARv4.3.2 were close. Although emissions of CEDS were larger than REASv3 and EDGARv4.3.2, trends were similar until early 2010. The different trends after 2010 between REASv3 and CEDS were caused by those of emissions in China. For the contributing rates, REASv3 and CEDS generally showed similar temporal variations, although relative ratios of OSA were larger in CEDS. Contribution rates of Japan were large around 1970 and then gradually decreased. Instead, those from China increased almost monotonically until 2010. Similar to the case of SO_2, relative ratios of China decreased recently in REASv3, but they were almost stable in CEDS and EDGARv4.3.2. In addition, contribution rates from India showed gradual increasing trends in all the results

For total Asia emissions of BC, trends of REASv3 and CEDS were similar until the late 1990s, but after 2000 while growth rats of CEDS became larger, emissions of REAS were not changed largely and turned to decrease after 2010. Emission amounts and growth rates of EDGARv4.3.2 were smaller than others until the mid-1990s, but after that the trends were similar to those of REASv3. Compared to SO_2 and NO_x, temporal variations of relative ratios of BC emissions from each country and regions were small in all the results. In REASv3, contribution rates of Japan were large before 1970 and then decreased afterwards. On the other hand, in CEDS, contribution rates of Japan after 1970 were larger than those before 1970. After 2000, relative ratios of China in REASv3 were almost stable and showed a marginal decrease after 2011. In CEDS and EDGARv4.3.2, contribution rates of China increased during the first half of 2000s and then became almost stable. Similar tendencies were seen in OC. Compared to BC, relative ratios of China started to decrease earlier only in REASv3.

For Asia total emissions of CO, although amounts of CEDS were larger than others, trends of all results were close until the early 2000s. After that, REASv3 showed large increases until 2010 and then started to decreased slightly. These tendencies were mainly controlled by emissions in China. Trends of the relative ratios were similar to those of BC. But contribution rates of China in REASv3 increased gradually until the mid-2000s and then decreased, while those in CEDS and EDGARv4.3.2 were almost stable. For NMVOC, total emissions in Asia of REASv3 were smaller than others, but large increases of emissions were found from the early 1990s. The corresponding feature was shown in contribution rates. Relative ratios of emissions from China in REASv3 increased largely during the 1990s and 2000s. Similar increasing trends were seen in EDGARv4.3.2 but growth rates of REASv3 were much larger. On the other hand, both temporal variations and values of contribution rates of China were relatively small in CEDS.

For NH_3, trends of total emissions in Asia of REASv3 were close to EDGARv4.3.2 and slightly larger than CEDS until around 2000. After that, growth rates of REASv3 were close to CEDS and those of EDGARv4.3.2 became larger. As a result, amounts of total Asia emissions of all inventories became almost the same after 2010. For relative rations of regions, contribution rates of China in REASv3 increased gradually until the mid-2000s and then became almost stable, whereas those in CEDS and EDGARv4.3.2 show slightly decreasing and increasing trends, respectively. In 2015, relative ratios of NH_3 emissions from China in REASv3 were between those of EDGARv4.3.2 and CEDS. Compared to EDGARv4.3.2 and CEDS, contribution rates of NH_3 emissions from SEA region were relatively small in REASv3.

3.4 Uncertainty

In REASv3, uncertainties in emissions were estimated for each country and region in 1955, 1985, and 2015 using basically the same methodology as that of REASv2.1 (Kurokawa et al., 2013). First, uncertainties in all parameters used to calculate emissions, such as activity data, emission factors, removal efficiencies, and sulfur contents of fuels were estimated in the range of 2-150%. In estimation of the uncertainties except for activity data, following three causes need to be considered: uncertainties in the data themselves, those caused by selections of the data, and those in settings related to emission controls such as timing of introduction and penetration rates of abatement equipment. In this study, uncertainties in settings of emission controls were explicitly considered only for removal efficiencies. The uncertainties of removal efficiencies were assumed to be zero for emission sources where no emission controls were considered which means that uncertainties caused by neglecting emission controls were not considered. Furthermore, for emission sources where introduction rates of abatement equipment were small, uncertainties caused by settings of emission controls were assumed to be small. Then, uncertainties in emissions from power plants, industries, road transport, other transport, domestic and other sectors, as well as uncertainties in total emissions were calculated for all the species. The uncertainties of different sub-sectors and activities were combined in quadrature assuming they were independent. On the other hand, for uncertainties of national emissions in China, India, and Japan, those in their sub-regions were added linearly. Details of the methodology and settings of uncertainties of each component were described in Sect. S10 of the Supplement. Similar to REASv2.1, uncertainties of emissions that were not originally developed in REASv3 (NH_3 emissions from manure management and fertilizer application, and NMVOC evaporative emissions from Japan and Republic of Korea) were not evaluated in this study.

Table 4 summarizes the estimated uncertainties in total emissions of each species for China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. Uncertainties in emissions from each sector were provided in the Supplement tables (Table S1 for SO_2, NO_x, CO, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC, in Table S2 for NMVOC, and in Table S3 for NH_3). For most regions and years, uncertainties for SO_2, NO_x and CO_2 are smaller than other species. Major emission sources of these species are power plants and large industry sectors. Uncertainties of activity data of these species were assumed to be small because power plants and large industries are critically important for each country and related statistics are expected to be accurate. In addition, uncertainties of emission factors of combustion at high temperature in power plants and large industries are considered to be small. For SO_2 emissions in China, uncertainties in 2015 were estimated to be slightly larger than those in 1985 due to uncertainties for removal efficiencies which were not considered in 1985. For South and Southeast Asia, uncertainties of SO_2 emissions in 1985 were slightly smaller than those in 2015. This is because settings of sulfur contents in fuels were based on surveys conducted in 1990 (Kato and Akimoto, 1992) and thus, the uncertainties in 1985 were assumed to be smaller than those in 2015.

On the other hand, uncertainties of PM species are large compared to other species for most regions and years. For most countries in Asia, a majority of their emissions was from combustion at relatively low temperatures in small industries and residential sectors. Accuracies of activity data and emission factors for these sources are assumed to be low, especially for biofuel combustion. Therefore, uncertainties of OC emissions mainly from biofuel combustion in Asia are the largest for most regions and years. Uncertainties of PM_{10} are generally smaller than other PM species. This is because for PM_{10} emissions, contribution rates of power plants and industry sectors are generally larger than those of other PM species. For CO and NMVOC, in general, uncertainties of emission factors are assumed to be greater than SO_2, NO_x, and CO_2, but smaller than $PM_{2.5}$, BC and OC. Therefore, uncertainties of total CO and NMVOC emissions are generally between those of other species. For Southeast and South Asia, uncertainties of CO and NMVOC are comparable to PM_{10} as their relative contribution from biofuel combustion is large.

Uncertainties in emissions from Japan are lesser than those of other countries and regions. This is mainly due to the accuracy of activity data. Accessibility to detailed information in Japan is relatively high compared to other countries in REASv3. In Japan, uncertainties of emission in 1985 were comparable to or slightly smaller than those in 2015. This is because relative ratios of emissions from road transport whose uncertainties were the smallest in Japan were reduced largely from 1985 to 2015. For China and India, accuracies of emissions are generally improved for most species compared to REASv2.1 using information from recently published literatures of emission inventory of these countries. However, the improvement is not significant due to the lack of country specific information. This situation is almost the same for other countries and regions. Although studies of national emission inventories in Asia are being published, as described in Section 1, information on technologies related to emissions and their introduction rates is not as easily available. Therefore, continuous efforts to update emission inventories by collecting information of each country and region are essential. For all countries, uncertainties of emissions in 1955 were much larger than those in 2015. This is because most activity data were not obtained directly from statistics, especially in the early half of the target period of REASv3.1. In this study, activity data, which were not available in statistics were extrapolated or assumed using proxy data as described in Section 2. In order to reduce uncertainties of emissions in long past years, these procedures need to be considered based on detailed information of each country and region during the period.

4 Summary and remarks

A long historical emission inventory of major air and climate pollutants in Asia during 1950-2015 was developed as Regional Emission inventory in ASia version 3 (REASv3). Target species were SO_2, NO_x, CO, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, BC, OC, and CO_2 and the domain areas included East, Southeast, and South Asia. Emissions from fuel combustion in power plants, industries, transport, and domestic sectors and those from industrial processes were estimated for all the species. In addition, emissions from evaporative sources were included in NMVOC and those from agricultural activities and human physiological phenomenon were considered for NH. REASv3 provides gridded data as well as emissions from each country and sub-region. Spatial resolution is mainly 0.25° × 0.25° and large power plants are treated as point sources. Temporal resolution is monthly. Emissions were estimated based on information of technologies related to emission factors and removal efficiencies, although available data and literatures are limited in the case of Asia. Activity data for recent years were collected from international and national statistics and those of past years, when detailed information was not available, were extrapolated using proxy data for the target period of REASv3. Details of methodologies such as data sources and treatments, settings of emission factors and emission controls, and related assumptions were provided in the supplement document entitled “Supplementary information and data to methodology of REASv3”.

Total emissions in Asia averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are: SO_2: 3.2 Tg, 42.4 Tg (13.1); NO_x: 1.6 Tg, 47.3 Tg (29.1); CO: 56.1 Tg, 303 Tg (5.4); NMVOC: 7.0 Tg, 57.8 Tg (8.3); NH_3: 8.0 Tg, 31.3 Tg (3.9); CO_2: 1.1 Pg, 18.6 Pg (16.5); PM_{10}: 5.9 Tg, 30.2 Tg (5.1); $PM_{2.5}$: 4.6 Tg, 21.3 Tg (4.6); BC: 0.69 Tg, 3.2 Tg (4.7); and OC: 2.5 Tg, 6.6 Tg (2.7). Clearly, all the air pollutant emissions in Asia increased significantly during these six decades. However, situations were different among countries and regions. In recent years, the relative contribution of air pollutant emissions from China was the largest along with rapid increase in economic growth, but most species have reached their peaks and the growth rates of other species have become at least small or almost zero. For SO_2 and NO_x, introduction of abatement equipment, especially for coal-fired power plants, such as FGD and SCR were considered to be effective in reducing emissions. For PM species, in addition to control equipment in industrial plants, emissions decreased recently due to reduced usage of biofuels. On the other hand, air pollutant emissions from India showed an almost continuous increase. Growth rates were larger for SO_2 and NO_x, but their structures of emissions were different. Large parts of SO_2 emissions were obtained from coal combustion in power plants and industrial sector, and the recent rapid increase of SO_2 emission was mainly from coal-fired power plants. For NO_x, contribution from road transport especially diesel vehicles were almost comparable with those of power plants. For PM species, a majority of emissions was from the residential sector in the 1950s-1960s and its contribution is still considered to be large. Recent increasing trends were mainly caused by emissions from power and industrial plants and road vehicles. Trends in Japan were much different than those of the whole of Asia. Emissions increased rapidly along with economic growth during the 1950s-1970s, but those of most species were reduced largely from peak values. In addition, peak years were mostly 40 years ago, reflecting the time series of introduction of control measures to mitigate air pollution. Similar features were found in Republic of Korea and Taiwan. For other countries in Asia, emissions of air pollutants generally showed increasing trends along with economic situation and motorization. As described above, trends and spatial distribution of air pollutants in Asia are not simple and are becoming complicated.

Mitigation of air and climate pollutant emissions is an urgent issue in most Asian countries, but the situation is different country-wise. In this study, detailed discussion on effects of emission controls were conducted only for China and Japan due to limitation of information. Therefore, continuous efforts to develop and update emission inventories in Asia based on country specific information are essential especially for countries and regions other than China and Japan. On the other hand, there are inevitable uncertainties in parameters required to develop emission inventories, such as activity data and emission factors. In addition, it is fundamentally impossible to develop a real-time emission inventory because there is a time lag in the publication of basic statistics essential to estimate emissions. Recently, satellite observation data of air pollutants are becoming available at a finer scale for many species, such as NO_x, SO_2 and NH_3. Evaluations and improvements of REASv3 based on these data as well as results of modeling studies, such as inverse modeling are more important next steps. Also, addition of target species, especially CH_4, which is one of the key species to mitigate both air pollution and global warming is another important task for future studies.

Data availability:

Monthly gridded emission data sets at 0.25° × 0.25° resolution for major sectors from 1950 to 2015 are available from a data download site of REAS. The URL of the site is http://www.nies.go.jp/REAS/. Country and regional emission table data for major sectors during 1950-2015 and those for major fuel types are also provided at the site. Note that datasets of REASv3.1 were released after a publication of Kurokawa et al. (2019) from December 2019. The datasets were revised and the updated data are available as REASv3.2 together with a publication of this paper. Differences between REASv3.2 and REASv3.1 were presented and discussed in the Supplement document entitled “Differences between REASv3.2 and REASv3.1”.

Author contribution:

JK and TO conducted the study design. JK contributed to actual works for development of REASv3 such as collecting data and information, settings of parameters, calculating emissions and creating final data sets. JK and TO analyzed and discussed the estimated emissions in REASv3. JK prepared the manuscript with contributions from TO.

Competing interest:

The authors declare that they have no conflict of interest.

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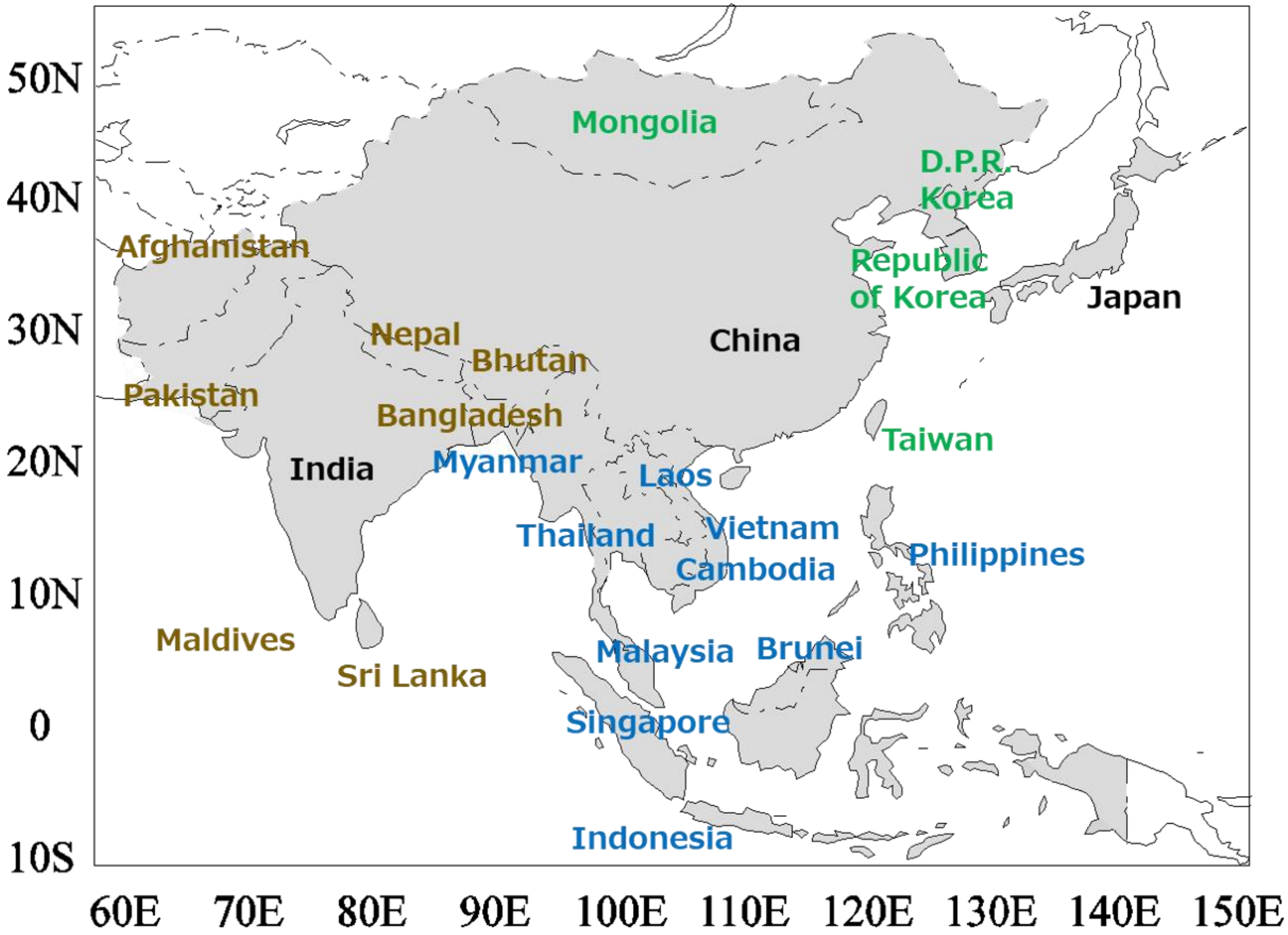

Figure 1. Domain and target countries of REASv3. In this paper, countries written in blue, green, and brown characters were defined as SEA (Southeast Asia), OEA (East Asia other than China and Japan), and OSA (South Asia other than India), respectively.

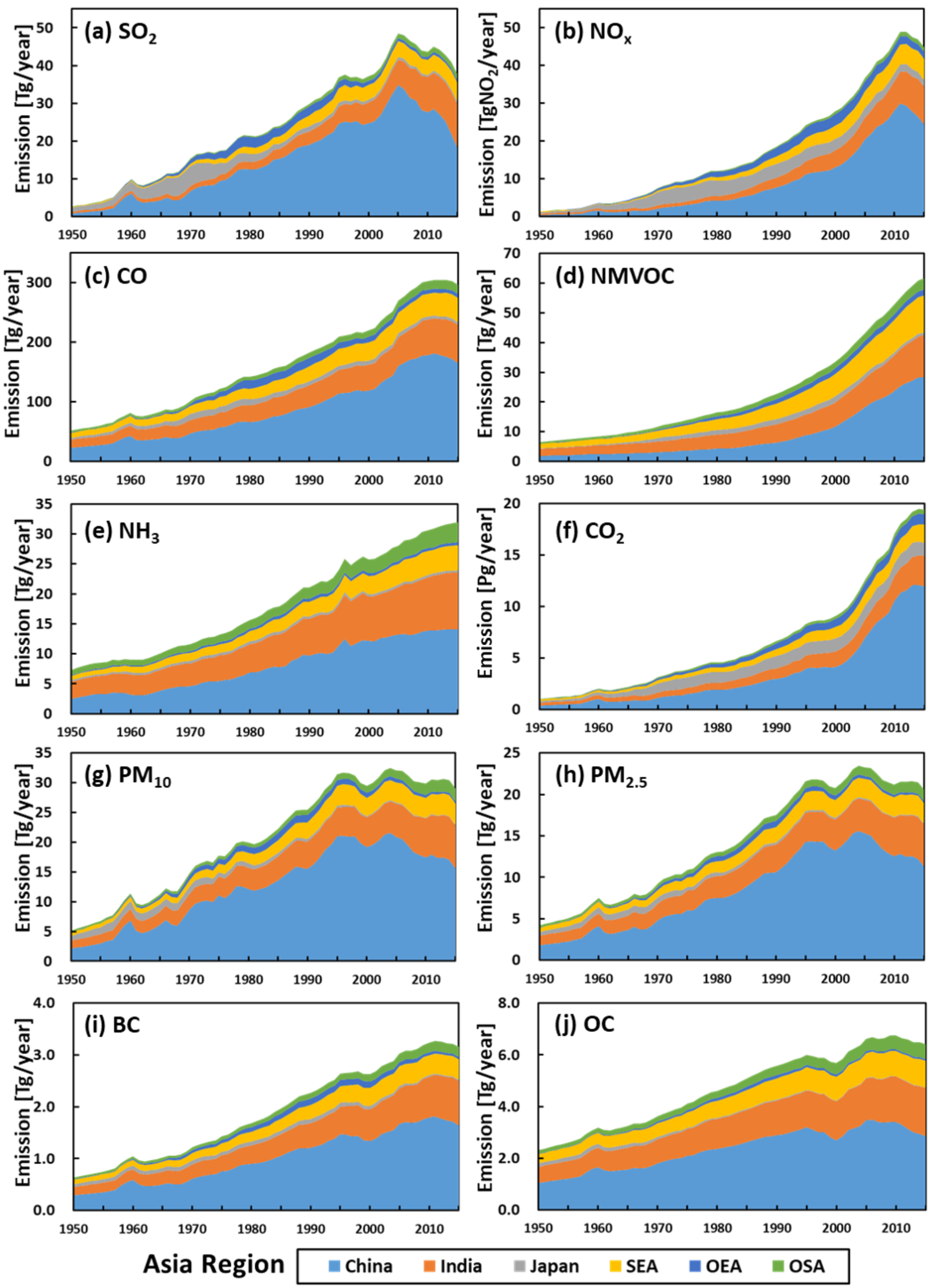

Figure 2. Trends of (a) SO_2, (b) NO_x, (c) CO, (d) NMVOC, (e) NH_3, (f) CO_2, (g) PM_{10}, (h) $PM_{2.5}$, (i) BC, and (j) OC emissions in Asia during 1950-2015 for each region. See Fig. 1 for countries included in SEA, OEA, and OSA.

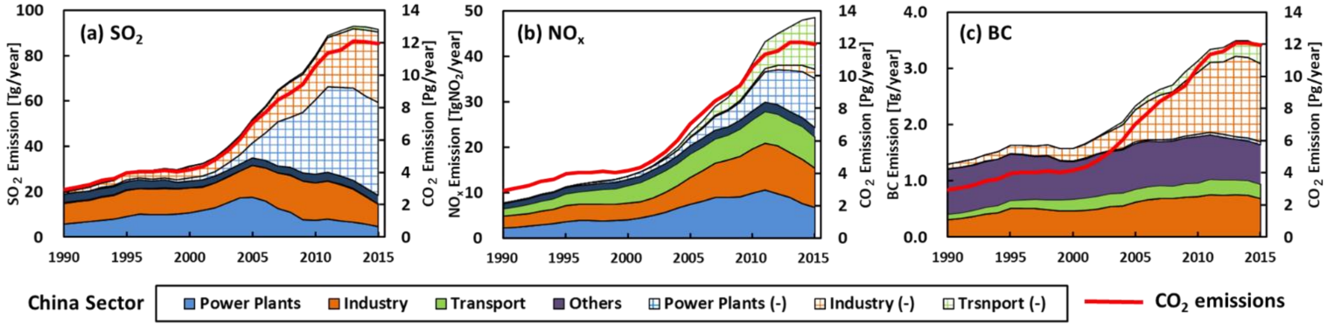

Figure 3. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in China during 1990-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures. Red lines in the panels are total CO_2 emissions.

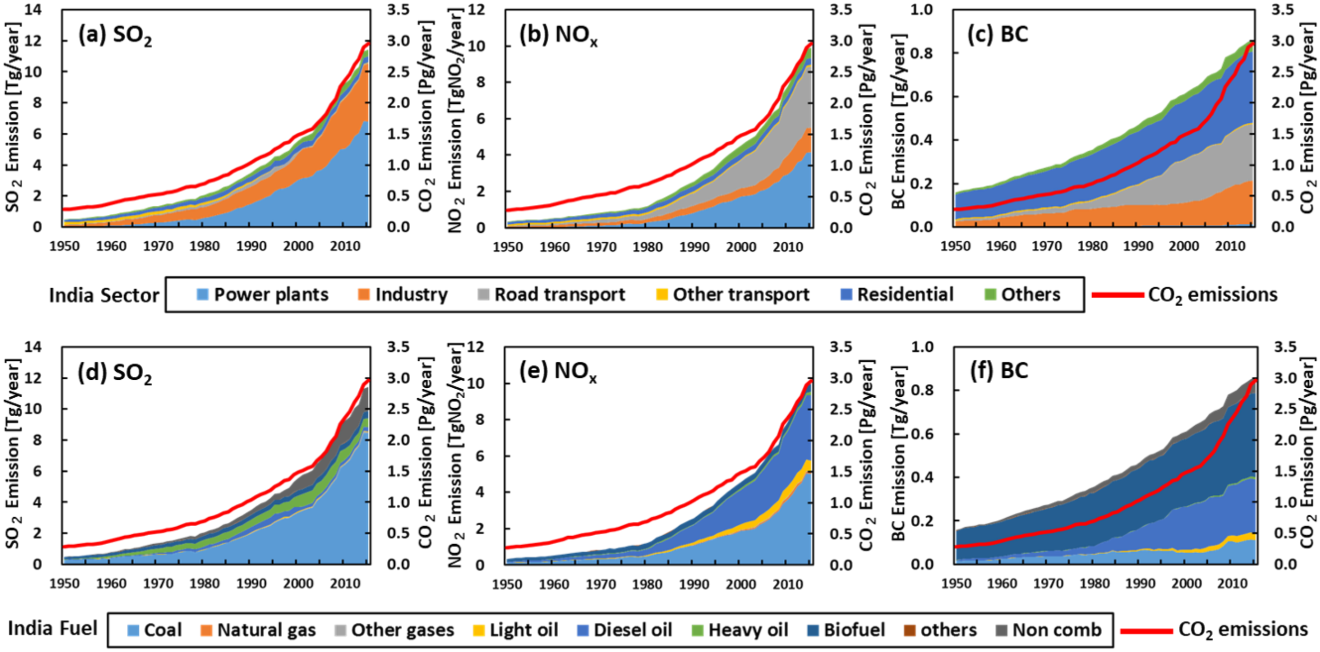

Figure 4. Emissions of (a, d) SO_2, (b, e) NO_x, and (c, f) BC from each major sector category (upper panels) and fuel type (lower panels) in India from 1950 to 2015 (Non comb = Non combustion sources). Red lines in the panels are total CO_2 emissions.

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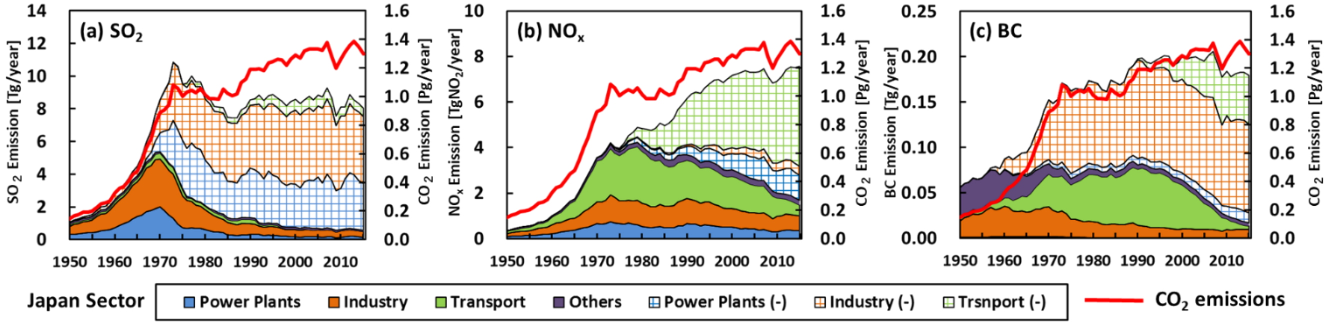

Figure 5. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in Japan during 1950-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures. Red lines in the panels are total CO_2 emissions.

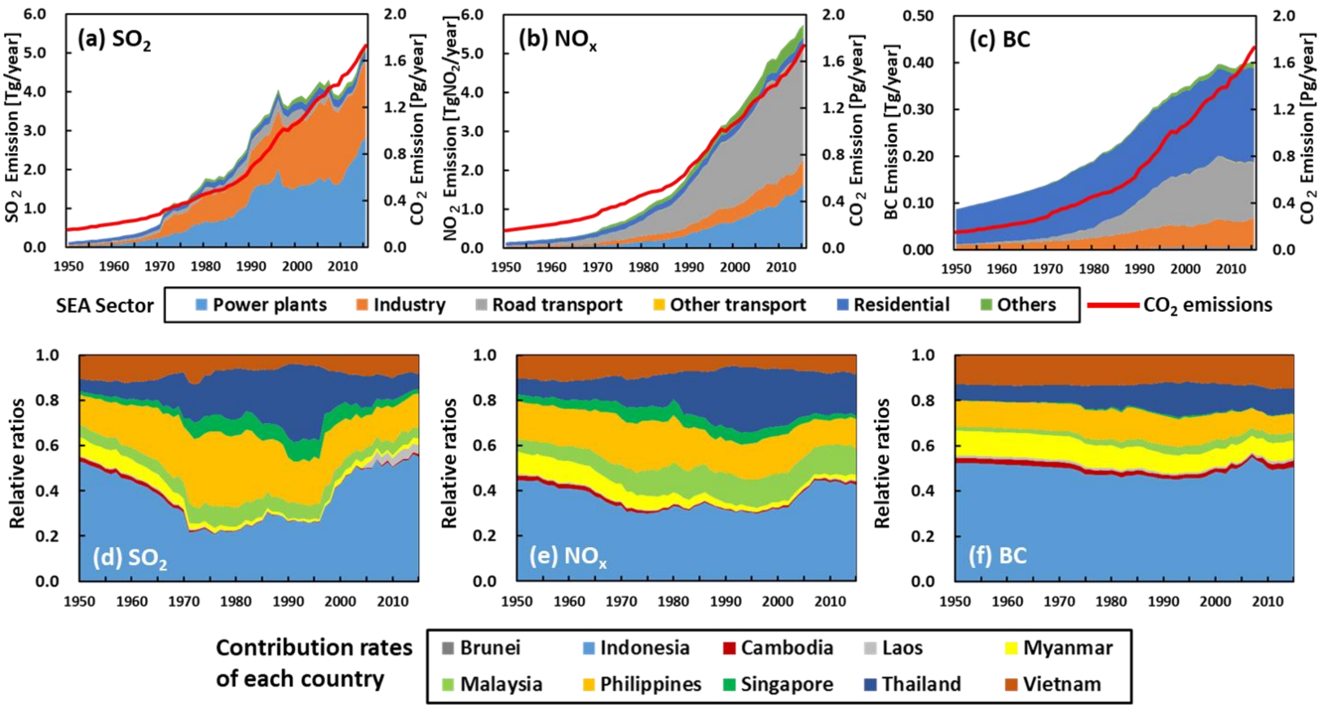

Figure 6. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in SEA (upper panels) and (d, e, f) relative ratios of emissions from each country in SEA (lower panels) during 1950-2015. Red lines in the upper panels are total CO_2 emissions.

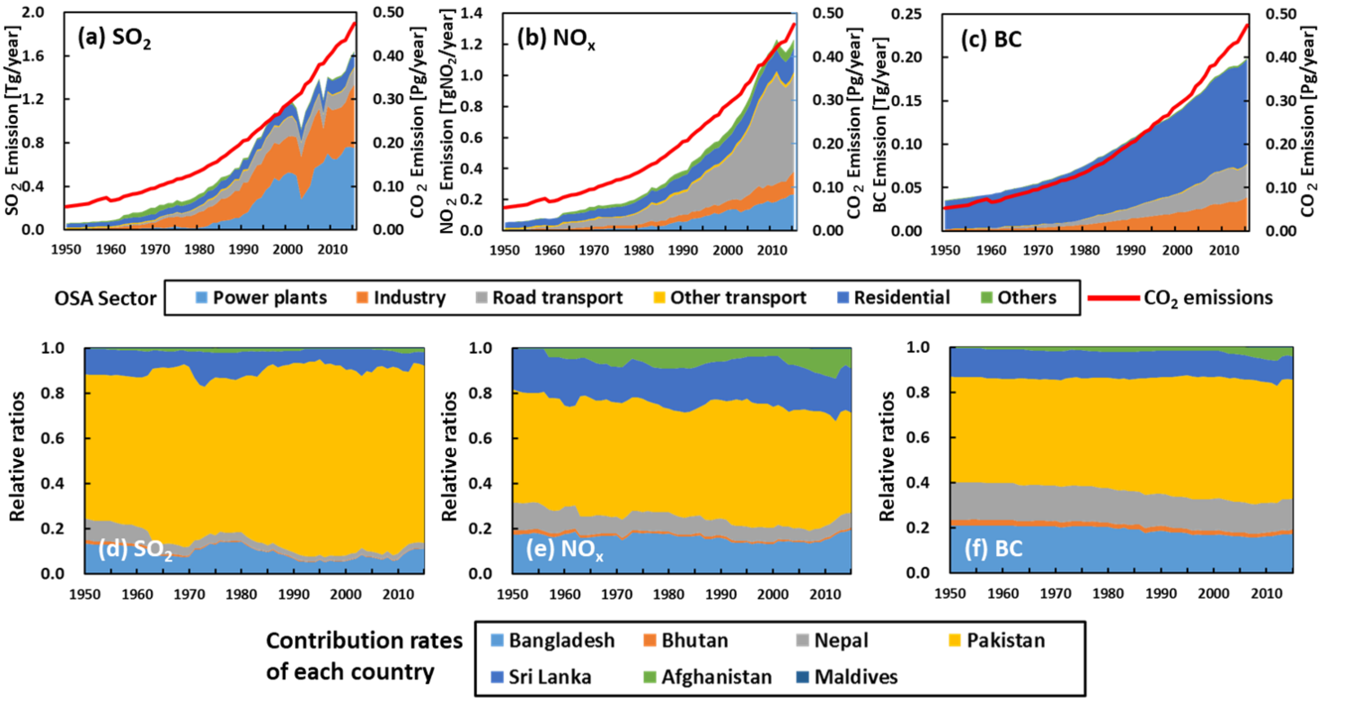

Figure 7. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in OSA (upper panels) and (d, e, f) relative ratios of emissions from each country in OSA (lower panels) from 1950 to 2015.

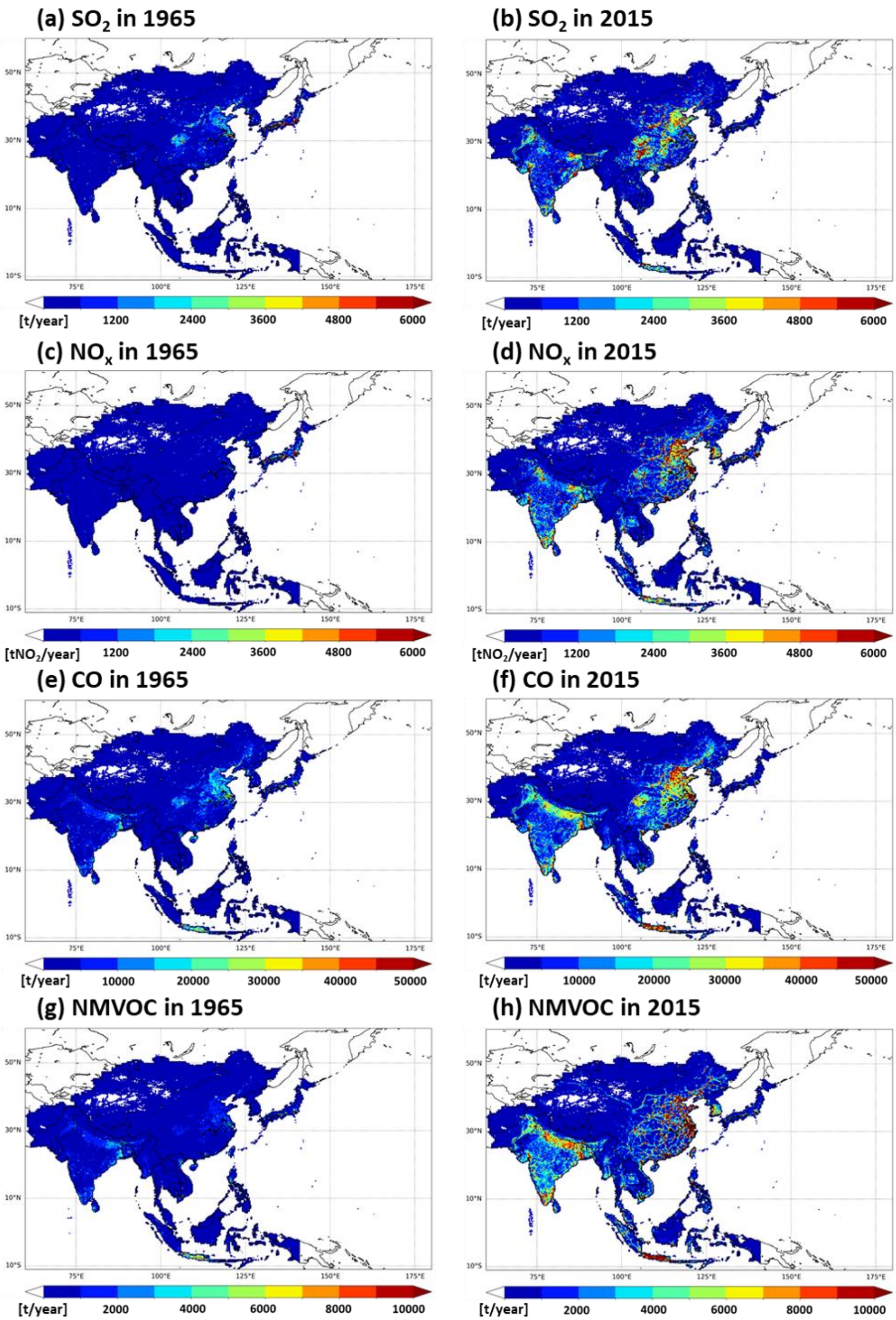

Figure 8. Grid maps of annual emissions of (a, b) SO_2, (c, d) NO_x, (e, f) CO, (g, h) NMVOC, (i, j) NH_3, (k, l) $PM_{2.5}$, (m, n) BC, and (o, p) OC in 1965 (left panels) and 2015 (right panels).

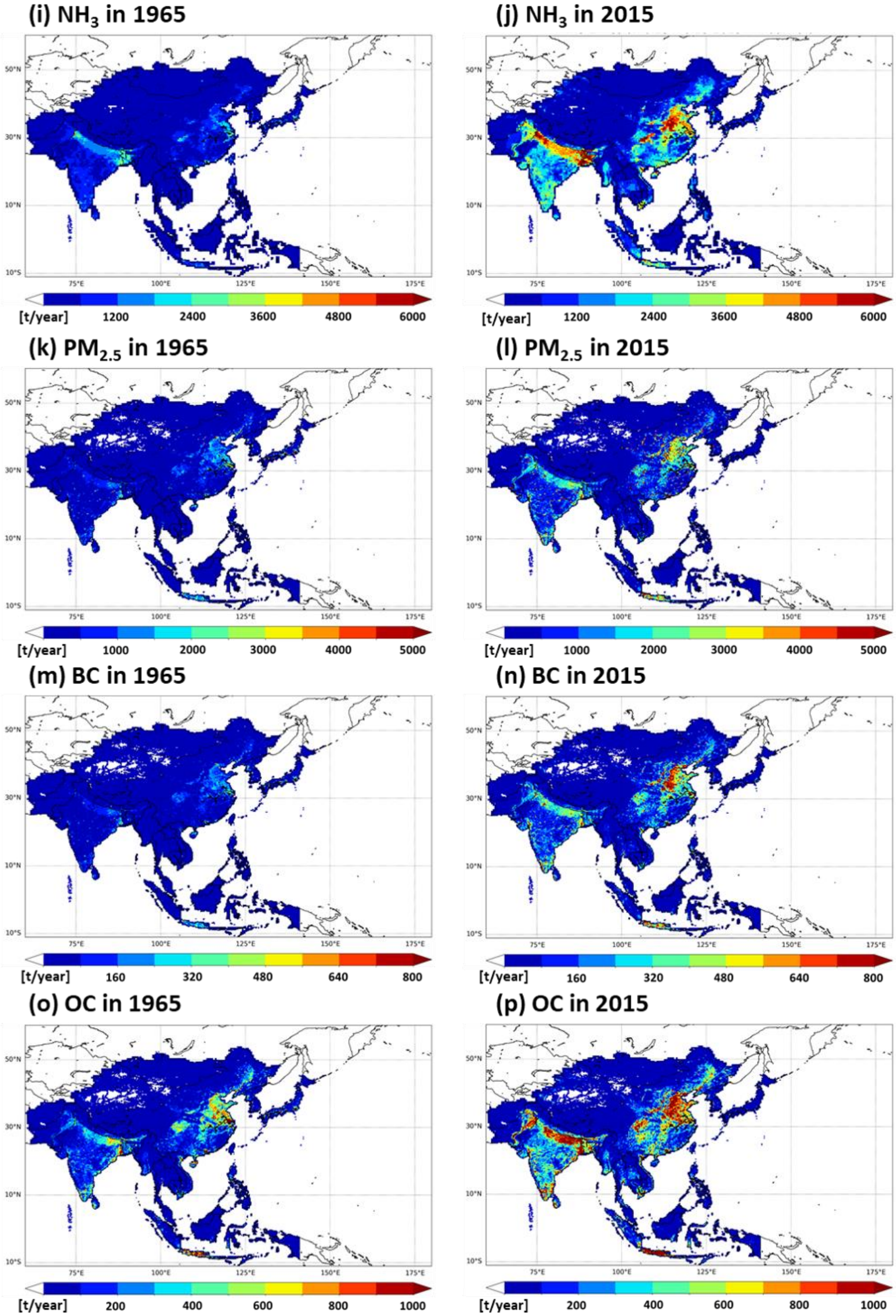

Figure 8. Continued.

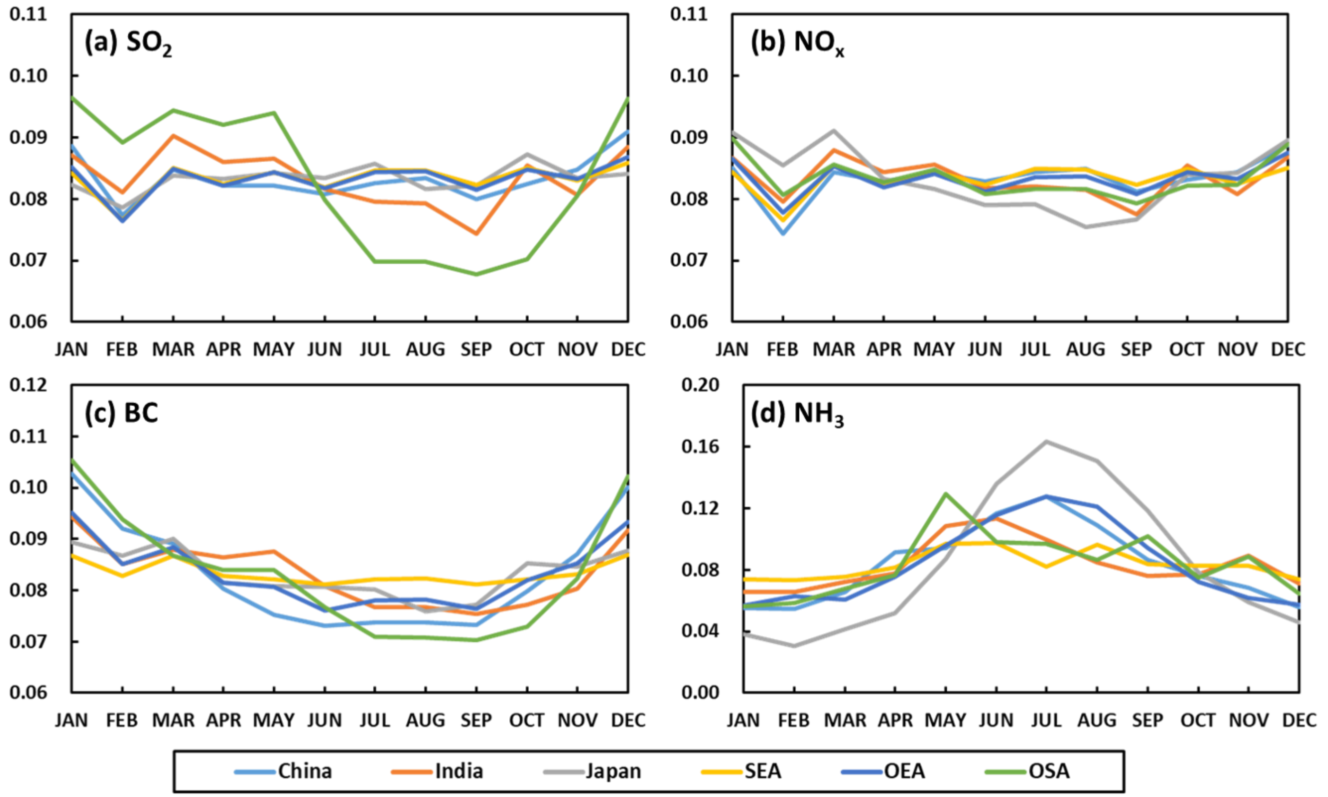

Figure 9. Monthly variations of (a) SO_2, (b) NO_x, (c) BC, and (d) NH_3 emissions for each region of Asia in 2015. See Fig. 1 for definitions of SEA OEA, and OSA.

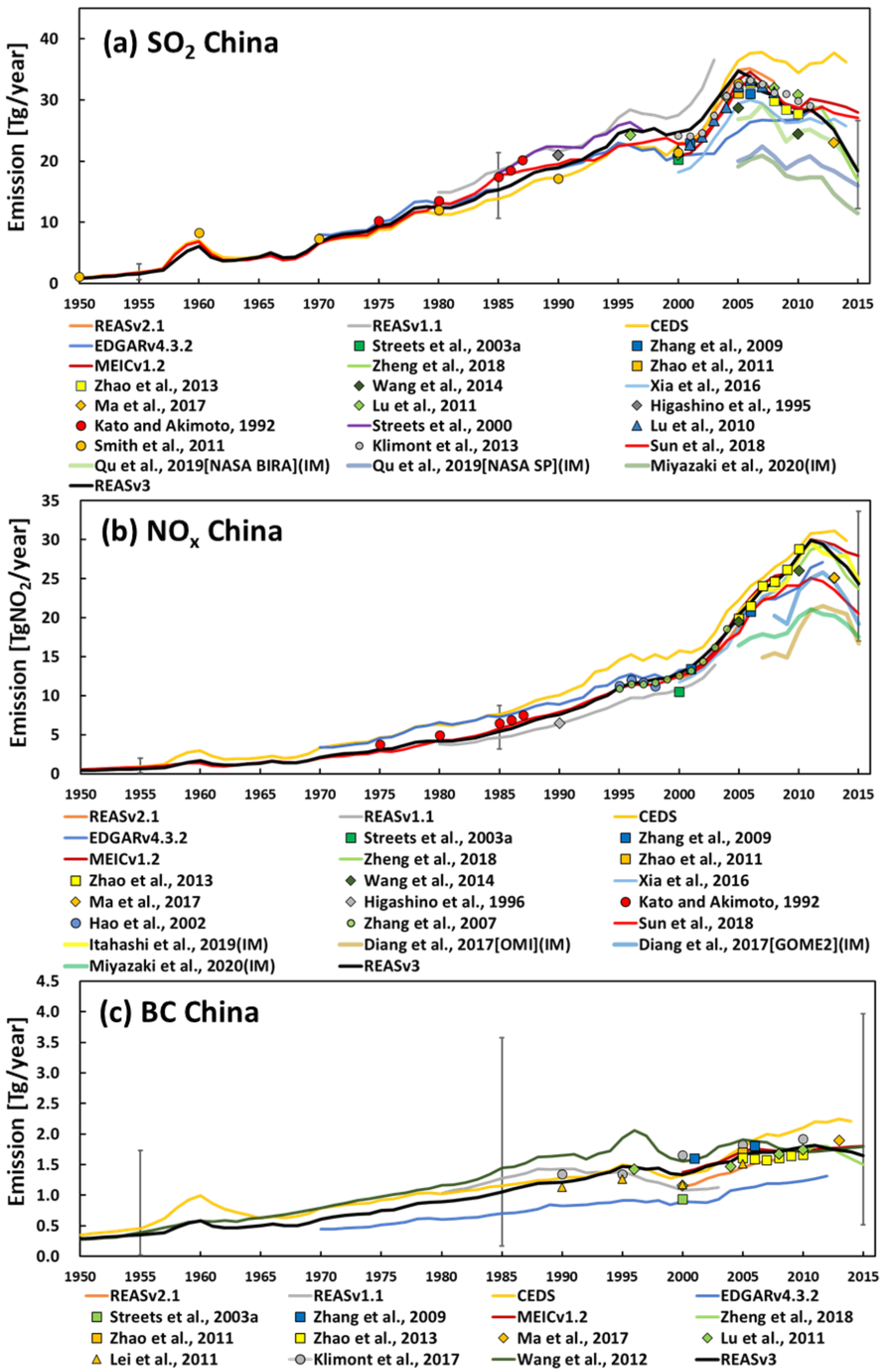

Figure 10. Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in China between REASv3 and other studies. Note that emissions from domestic and fishing ships were excluded from REAS series, CEDS, EDGARv4.3.2, and Higashino et al. (1996). IM means estimates by inverse modeling. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.

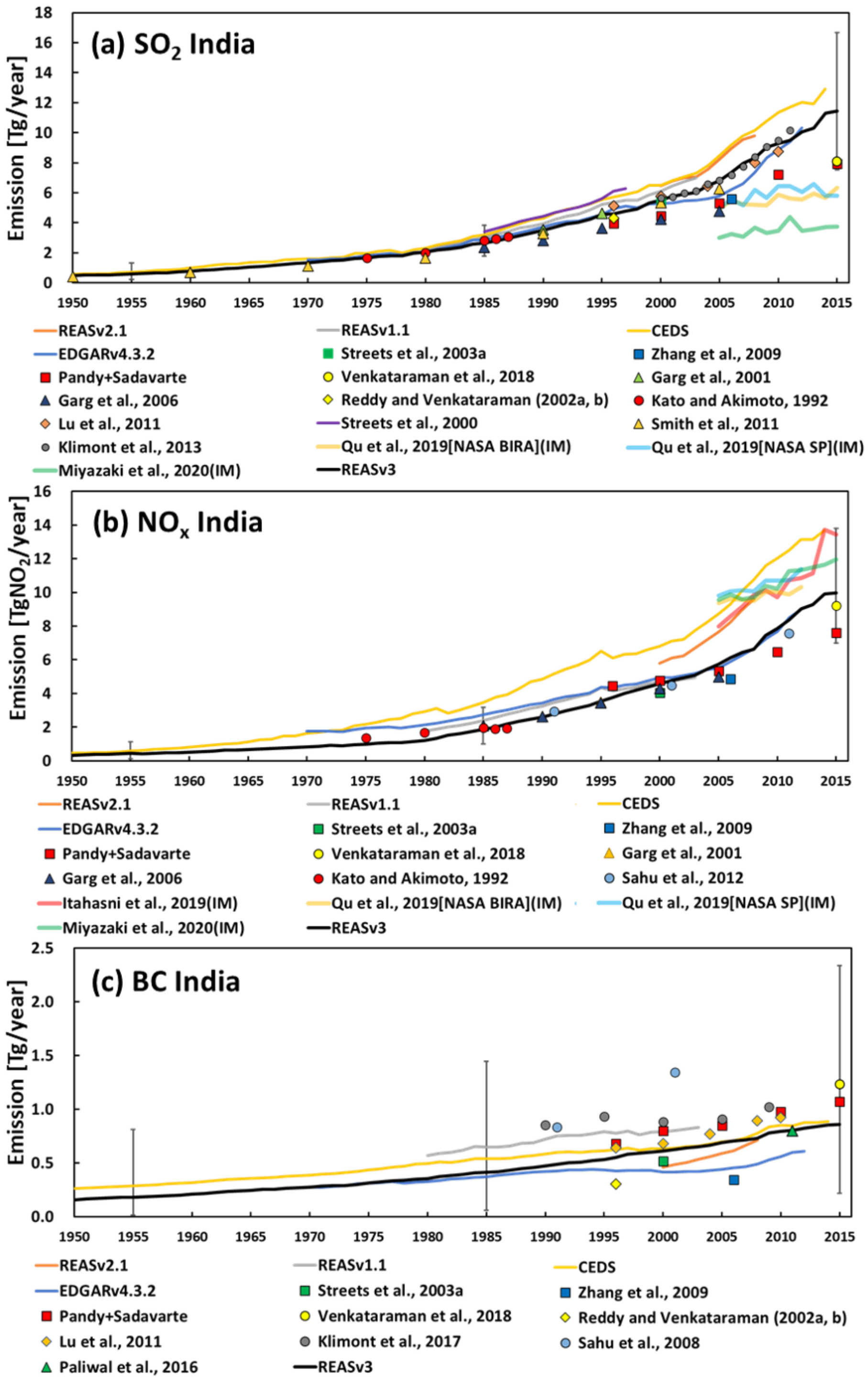

Figure 11. Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in India between REASv3 and other studies. Note that values of "Pandy+Sadavarte" are calculated from Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014). Emissions from domestic and fishing ships were excluded from REAS series, CEDS, EDGARv4.3.2, Garg et al. (2006) and Paliwai et al. (2016). IM means estimates by inverse modeling. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.

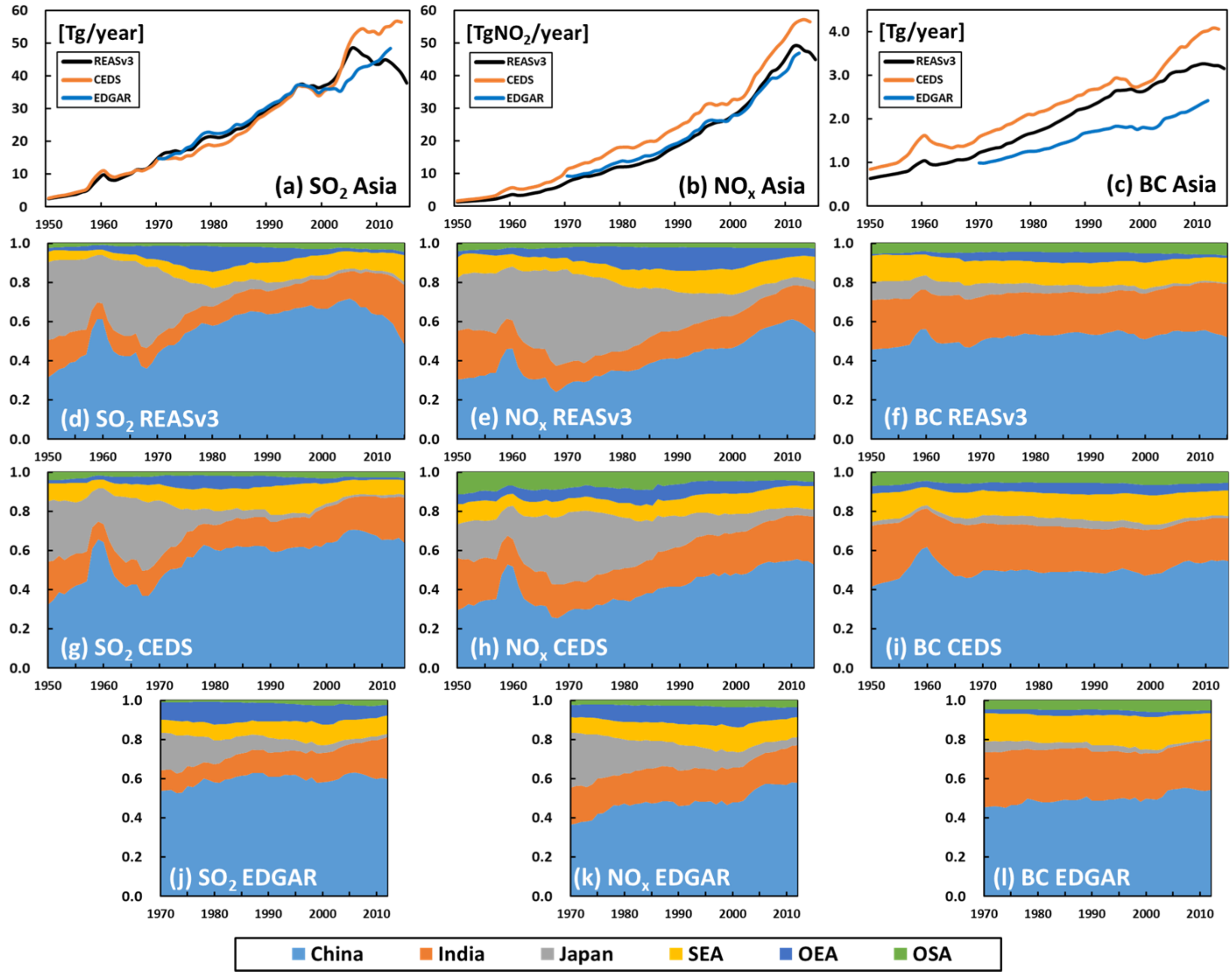

Figure 12. Comparison of trends of (a) SO_2, (b) NO_x and (c) BC emissions in Asia and relative ratios of emissions from China, India, Japan, SEA, OEA, and OSA for (d, g, j) SO_2, (e, h, k) NO_x, and (f, i, l) BC among (d, e, f) REASv3, (g, h, i) CEDS, and (j, k. l) EDGARv4.3.2. Note that periods of CEDS and EDGARv4.3.2 shown here are during 1950-2014 and 1970-2012, respectively. See Fig. 1 for definitions of SEA, OEA, and OSA.

Table 1. General information on REASv3.

Item	Description
Species	SO_2, NO_x, CO, NMVOC, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC
Years	1950–2015
Areas	East, Southeast, and South Asia
Emission sources	Fuel combustion in power plans, industry, transport, and domestic sectors; Industrial processes; Agricultural activities (fertilizer application and livestock); and Others (fugitive emissions, solvent use, human, etc.)
Spatial resolution	0.25 degree by 0.25 degree
Temporal resolution	Monthly
Data distribution	http://www.nies.go.jp/REAS/

Table 2. Emission inventories from other research works and officially opened data utilized in REASv3.

Other emission inventories and data sources	How utilized in REASv3
VOC Emission Inventory in Japan (MOEJ, 2017)	Evaporative emissions of NMVOC in Japan[a]
The National Air Pollutants Emission Service of the National Institute of Environmental Research (http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do)	Evaporative emissions of NMVOC in Republic of Korea[a]
REASv2.1 (Kurokawa et al., 2013; JPEC 2012a, b, c; 2014)	NH_3 emissions from agricultural sources in Japan[b]
REASv1.1 (Yamaji et al., 2004; Yan et al., 2003)	NH_3 emissions from agricultural sources in countries and regions other than Japan[b]
REASv2.1 (Kurokawa et al., 2013; JPEC 2012a, b, c; 2014)	Grid allocation factors for manure management[c] and road transport sectors for Japan[d]
EDGARv4.3.1 (Crippa et al., 2016)	Grid allocation factors for manure management[c] and road transport[d] sectors for countries and regions other than Japan

[a]See Sect. S5.3 of the Supplement. [b]See Sect. 2.4. [c]See Sect. S8.1 of the Supplement. [d]See Sect. 2.6.

Table 3. Summary of national emissions in 2015 for each species and total annual emissions in Asia in 1950, 1960, 1970, 1980, 1990, 2000, and 2010-2015 (Gg yr^{-1}).

Country	SO_2	NO_x[a]	CO	NMVOC	NH_3	CO_2[b]	PM_{10}	$PM_{2.5}$	BC	OC
China	18404	24318	165133	28189	14063	11941	15501	11342	1643	2860
India	11438	9969	64366	14286	9505	2959	7213	5052	858	1868
Japan	565	1687	3877	895	349	1300	129	89	17	13
Korea, D.P.R.	116	200	2663	134	92	29	106	56	11	18
Korea, Rep of	336	1120	1931	960	170	689	139	114	19	34
Mongolia	99	127	986	50	139	18	44	20	2.9	3.2
Taiwan	124	371	1027	770	85	281	45	37	6.9	7.3
Brunei	4.0	13	29	43	3.8	6.1	7.5	2.9	0.2	0.1
Cambodia	55	61	1087	212	78	22	115	69	9.0	32
Indonesia	2852	2463	20517	6130	1591	655	1606	1160	196	556
Laos	201	35	325	66	67	12	46	25	3.6	10
Malaysia	233	613	1288	936	163	230	206	119	14	12
Myanmar	154	121	2925	867	621	59	184	165	29	98
Philippines	786	767	3292	898	388	134	284	183	38	61
Singapore	87	89	76	302	6.4	46	81	62	1.2	0.5
Thailand	341	1137	5436	1543	542	320	522	363	49	125
Vietnam	436	507	6078	1552	747	250	587	362	59	146
Afghanistan	24	97	404	93	251	9.4	18	14	6.9	4.4
Bangladesh	171	305	2755	704	883	110	519	287	40	102
Bhutan	3.3	6.8	269	55	9.5	4.7	29	19	3.0	10
Maldives	3.1	4.1	9.4	3.7	0.4	0.8	0.2	0.2	0.1	0.0
Nepal	42	64	2381	533	321	40	207	161	26	89
Pakistan	1310	573	8576	2031	1772	273	1310	841	105	324
Sri Lanka	92	187	1382	374	103	37	135	98	19	49
Asia[c] 1950	2540	1339	51804	6551	7310	1005	5089	4162	630	2308
Asia[c] 1960	9880	3639	81220	8461	8968	2016	11405	7487	1040	3185
Asia[c] 1970	15287	7470	100368	11599	11579	3117	14770	9217	1221	3629
Asia[c] 1980	21425	12080	142102	16432	15632	4550	19900	13060	1680	4602
Asia[c] 1990	29721	18481	182418	22670	21035	6595	25427	17542	2264	5574
Asia[c] 2000	37074	27782	219516	33498	25775	9083	29461	20758	2626	5682
Asia[c] 2010	43635	46368	302562	52711	30621	17055	29880	21220	3233	6757
Asia[c] 2011	45003	48868	304900	55136	30878	18047	30540	21559	3266	6652
Asia[c] 2012	44227	48962	304396	57285	31283	18496	30414	21526	3254	6587
Asia[c] 2013	42725	47561	304484	58971	31559	19200	30649	21627	3227	6485
Asia[c] 2014	40864	46970	302718	60801	31770	19447	30469	21475	3219	6478
Asia[c] 2015	37876	44835	296809	61627	31950	19423	29034	20644	3155	6422

[a]Gg-NO_2 yr^{-1}.
[b]Tg yr^{-1}.
[c]Asia in this table include all target countries and sub-regions in REASv3.

Table 4. Uncertainties [%] of emissions in China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. See Fig. 1 for definitions of SEA OEA, and OSA.

	SO_2	NO_x	CO	NMVOC	NH_3	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
1955										
China	±85	±167	±291	±277	±174	±133	±253	±315	±334	±365
India	±96	±122	±265	±295	±161	±116	±257	±294	±277	±314
Japan	±59	±62	±157	±135	±141	±49	±94	±117	±170	±270
SEA	±134	±153	±260	±272	±169	±126	±291	±307	±323	±317
OEA	±73	±88	±146	±184	±148	±59	±120	±157	±157	±262
OSA	±70	±112	±272	±270	±168	±110	±219	±281	±310	±345
1985										
China	±36	±53	±157	±150	±139	±39	±101	±129	±182	±250
India	±40	±60	±196	±212	±135	±58	±160	±201	±191	±259
Japan	±30	±31	±44	±50	±93	±14	±72	±71	±53	±67
SEA	±40	±56	±185	±162	±141	±56	±157	±191	±218	±259
OEA	±48	±70	±72	±78	±113	±27	±80	±82	±88	±102
OSA	±36	±44	±144	±137	±134	±33	±108	±137	±176	±248
2015										
China	±40	±35	±73	±76	±82	±19	±83	±94	±111	±193
India	±41	±35	±136	±115	±111	±27	±120	±151	±133	±233
Japan	±34	±32	±45	±63	±103	±13	±68	±74	±58	±100
SEA	±46	±38	±124	±86	±115	±25	±125	±155	±161	±232
OEA	±38	±60	±67	±63	±94	±19	±69	±85	±82	±168
OSA	±40	±34	±87	±73	±93	±19	±96	±112	±124	±211

(4) The revised main manuscript with track changes

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Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3~~.1~~

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Abstract. A long-term historical emission inventory of air and climate pollutants in East, Southeast, and South Asia from 1950-2015 was developed as the Regional Emission inventory in ASia version 3~~.1~~ (REASv3~~.1~~). REASv3~~.1~~ provides details of emissions from major anthropogenic sources for each country and its sub-regions and also provides monthly gridded data with 0.25° × 0.25° resolution. The average total emissions in Asia during 1950-1955 and from 2010-2015 (growth rates in these 60 years) are as follows: SO_2: 3.2 Tg, 42.4 Tg (13.1); NO_x: 1.6 Tg, 47.3 Tg (29.1); CO: 56.1 Tg, 303 Tg (5.4); non-methane volatile organic compounds: 7.0 Tg, 57.8 Tg (8.3); NH_3: 8.0 Tg, 31.3 Tg (3.9); CO_2: 1.1 Pg, 18.6 Pg (16.5); PM_{10}: 5.9 Tg, 30.2 Tg (5.1); $PM_{2.5}$: 4.6 Tg, 21.3 Tg (4.6); black carbon: 0.69 Tg, 3.2 Tg (4.7); and organic carbon: 2.5 Tg, 6.6 Tg (2.7)~~SO_2: 3.15 Tg, 42.4 Tg (13.5); NO_x: 1.83 Tg, 47.6 Tg (26.0); CO: 62.2 Tg, 319 Tg (5.13); non-methane volatile organic compounds: 9.14 Tg, 61.8 Tg (6.77); NH_3: 7.99 Tg, 31.3 Tg (3.92); CO_2: 1.12 Pg, 18.3 Pg (16.3); PM_{10}: 5.76 Tg, 28.4 Tg (4.92); $PM_{2.5}$: 4.52 Tg, 20.3 Tg (4.50); black carbon: 0.751 Tg, 3.38 Tg (4.51); and organic carbon: 2.62 Tg, 6.92 Tg (2.64)~~. Clearly, all the air pollutant emissions in Asia increased significantly during these six decades, but situations were different among countries and regions. Due to China's rapid economic growth in recent years, its relative contribution to emissions in Asia has been the largest. However, most pollutant species reached their peaks by 2015 and the growth rates of other species was found to be reduced or almost zero. On the other hand, air pollutant emissions from India showed an almost continuous increasing trend. As a result, the relative ratio of emissions of India to that of Asia have increased recently. The trend observed in Japan was different from the rest of Asia. In Japan, emissions increased rapidly during the 1950s-1970s, which reflected the economic situation of the period; however, most emissions decreased from their peak values, which were approximately 40 years ago, due to the introduction of ~~regulations and laws~~control measures for air pollution. Similar features were found in ~~the~~ Republic of Korea and Taiwan. In the case of other Asian countries, air pollutant emissions generally showed an increase along with economic growth and motorization. Trends and spatial distribution of air pollutants in Asia are becoming complicated. Datasets of REASv3~~.1~~, including table of emissions by countries and sub-regions for major sectors and fuel types, and monthly gridded data with 0.25° × 0.25° resolution for major source categories are available through the following URL: http;//www.nies.go.jp/REAS/.

1 Introduction

With an increase in demand for energy, motorization, and industrial and agricultural products, air pollution from anthropogenic emissions is becoming a serious problem in Asia, especially due to its impact on human health. In addition, a significant increase in anthropogenic emissions in Asia is considered to affect not only the local air quality, but also regional, inter-continental, and global air quality and climate change. Therefore, reduction in air and climate pollutants emissions are urgent issues in Asia (UNEP, 2019). Short-Lived Climate Pollutants (SLCPs), which are gases and particles that contribute to warming and have short lifetimes, have been recently considered to play important roles in the mitigation both air pollution and climate change (UNEP, 2019). SLCPs such as black carbon (BC) and ozone are warming agents, which cause harm to people and ecosystems. A decrease in the emissions of BC and ozone precursors from fuel combustion led to the decrease of other particulate matter (PM) species, such as sulfate and nitrate aerosols. Even though this is a positive step for human health, it has a negative effect on global warming as sulfate and nitrate aerosols act as cooling agents in the troposphere. Therefore, to find effective ways to mitigate both air pollution and climate change, accurate understanding of the current status and historical trends of air and climate pollutants are fundamentally important.

Recently, Hoesly et al. (2018) developed a long-term historical global emission inventory from 1750 to 2014 using the Community Emission Data System (CEDS). This data set is used as input data for the Coupled Model Intercomparison Project phase 6 (CMIP6). The Emission Database for Global Atmospheric Research (EDGAR) also provides global emissions data of both air pollutants and greenhouse gases, with the current version 4.3.2 ranging from the period between 1970-2012 (Crippa et al., 2016). The EDGAR is used as the default data of input emissions for the Task Force on Hemispheric Transport of Air Pollution phase 2 (HTAPv2) (Janssens-Maenhout et al., 2015). For SLCPs, the European Union's Seventh Framework Programme project ECLIPSE (Evaluating the Climate and Air Quality Impact of Short-Lived Pollutants) developed a global emission inventory based on the GAINS model. Current version 5 provides gridded emissions for every five years from 1990 to 2030 and also from 2040 to 2050 (Stohl et al., 2015). However, data from Asia in global emission inventories are generally based on limited country specific information. For the Asian region, several project-based emission inventories are developed, such as Transport and Chemical Evolution over the Pacific (TRACE-P) field campaigns (Streets et al., 2003a, b) and its successor mission, that is Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) (Zhang et al., 2009). Recently, the MIX inventory (mosaic Asian anthropogenic emission inventory) was developed as input emission data sets for the Model Intercomparison Study for Asia (MICS-Asia) Phase 3 by a mosaic of up-to-date regional emission inventories. The MIX inventory is also a component of the HTAPv2 inventory (Li et al., 2017a). For national emission inventories, numerous studies, research papers, and reports have been published. MEIC (Multi-resolution Emission Inventory for China) developed by Tsinghua University is a widely used emission inventory database for China (Zhang et al., 2009; Li et al., 2014; Zheng et al., 2014, Liu et al., 2015) and is included in the MIX inventory. Zhao et al. (2011, 2012, 2013, and 2014) developed recent and projected emission inventories of air pollutants in China. In addition, research papers for regional emission inventories of China were also published recently (Zhu et al., 2018; Zheng et al., 2019a). In the case of India, Garg et al., (2006) developed a historical emission inventory of air pollutants and greenhouse gases from 1985 to 2005. For recent years, Sadavarte and Venkataraman (2014) developed multi-pollutant emission inventories for industry and transport sectors and Pandey et al. (2014) developed the same for domestic and small industry sectors for the same time period, that is 1996-2015. For Japan, several project-based emission data sets were developed, such as the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC 2012a, b, c; 2014), East Asian Air Pollutant Emission Grid Database (EAGrid) (Fukui et al., 2014), and emission data sets for Japan's Study for Reference Air Quality Modeling (J-STREAM) (Chatani et al., 2018). In addition, there are studies for other countries and regions, such as the Clean Air Policy Support System (CAPSS) for Republic of Korea (Lee et al., 2011), Thailand (Thao Pham et al., 2008), Indonesia (Permadi et al., 2017), and Nepal (Jayarathne et al., 2018; Sadavarte et al., 2019). However, these regional and national emission inventories in Asia are available for a limited period, with data of the past missing.

The authors of this study have been devoted in developing the Regional Emission inventory in ASia (REAS) series. First version of REAS (REASv1.1) were developed by Ohara et al. (2007), which accounted for actual emissions during 1980-2003 and projected ones in 2010 and 2020. Kurokawa et al. (2013) updated the inventory in REASv2.1, which focused on the period between 2000-2008 when emissions in China drastically increased. REASv2.1 is used as the default data of the MIX inventory. In this study, a long historical emission inventory in the Asian region from 1950-2015 has been newly developed as REAS version 3~~.1~~ (REASv3~~.1~~). This study provides methodology, results and discussion of REASv3.1. Section 2 gives the basic methodology, including collecting activity data, settings of emission factors and removal efficiencies, and spatial and temporal allocation of emissions to create monthly gridded data sets of REASv3~~.1~~. In Section 3.1, trends in air pollutants emissions in Asia are described in detail and effects of emission controls on emissions in China and Japan are discussed. Spatial and temporal distributions are overviewed in Section 3.2. Section 3.3 compares the results of REASv3.1 with other emission inventories. Uncertainties of REASv3.1 are discussed in Section 3.4. Finally, summary and remarks are presented in Section 4.

2 Methodology and data

2.1 General description

Table 1 summarizes the general information of REASv3. Major updates from previous versions are as follows:

- Target years are from 1950 to 2015 covering much longer periods than REASv1.1 (1980-2003) and REASv2.1 (2000-2008).
- The long historical data sets of activity data were developed by collecting international and national statistics and related proxy data.
- Emission factors and information of emission controls especially for China and Japan were surveyed from research papers of emission inventories in Asia and related literatures.
- Large power plants constructed after 2008 were added as new point sources.

- Allocation factors for spatial and temporal distribution were updated although several emission inventories developed by other research works were utilized (see Table 2).
- Emissions from Japan, Republic of Korea, and Taiwan were originally estimated except for NMVOC evaporative sources (see Table 2).

REASv3 focuses on the long historical trends of air pollutants emissions in Asia. The start year was chosen to be 1950 as severe air pollution in Japan started from the mid-1950s. For the emission inventory framework, there are two major changes from REASv2.1. One is the target species. REASv3 includes the following major air and climate pollutants: SO_2, NO_x, CO, non-methane volatile organic compounds (NMVOC), NH_3, PM_{10}, $PM_{2.5}$, BC, organic carbon (OC), and CO_2. However, CH_4, and N_2O that were included in REASv2.1 are not in the scope of this version. CH_4 is one of important components of SLCP and will be considered in the next version. Another is the target areas. Figure 1 shows the inventory domain of REASv3 which includes East, Southeast, and South Asia. China, India, and Japan have been divided into 33, 17, and 6 regions, respectively to reduce the uncertainties in the spatial distribution. Definition of the sub-regions are the same as for REASv2.1. In REASv3, Central Asia and the Asian part of Russia, which were target areas of REASv2.1 are not included because of the difficulty in collecting necessary data for estimating long historical emissions in these areas. The source categories considered in REASv3 are the same as those in REASv2.1. Major sources include fuel combustion in power plants, industry, transport, and domestic sectors. Non-combustion sources include industrial process, evaporation (NMVOC), and agricultural activities (NH_3). However, NO_x emissions from soil as well as from international and domestic aviation and navigation, including fishing ships are exceptions. They were not included in REASv3. The spatial and temporal resolution are the same as those of REASv2.1. Spatial resolution is 0.25° × 0.25°, except in the case of large power plants, which are treated as point sources. Temporal resolution is monthly.

In REASv3, most emissions were originally estimated. However, several emission inventories from other research works and officially opened data were utilized as summarized in Table 2. NMVOC emissions in Japan and Republic of Korea from evaporative sources were obtained from the Ministry of the Environment of Japan (MOEJ, 2017) and the National Air Pollutants Emission Service of the National Institute of Environmental Research (available at http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do), respectively. For NH_3 emissions from agricultural activities, data of base year (2000 and 2005 for Japan and 2000 for others) were obtained from other research works as follows (see Sect. 2.4): REASv2.1 (Kurokawa et al., 2013: JPEC 2012a, b, c; 2014) for Japan and REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) for other counties and regions. In addition, EDGARv4.3.2 were utilized to create grid allocation factors for road transport sector for all species and manure management for NH_3 (see Sects. 2.4.1 and 2.6, respectively).

In the following sub-sections, general methodologies and data used in REASv3 are overviewed for stational sources, road transport, agricultural sources, other sources, and spatial and temporal distribution. Details of the methodologies such as data sources and treatments, settings of emission factors and emission controls, and related assumptions are provided in the supplement document entitled “Supplementary information and data to methodology of REASv3” (hereafter, this document is expressed as “the Supplement”). In Sect. S2 of the Supplement, details of frame work of REASv3 including definitions of sub-categories of emission sources, and target countries and sub-regions of China, India, and Japan was provided.~~Table 1 summarizes the general information of REASv3.1. REASv3.1 focuses on the long historical trends of air pollutants emissions in Asia. Target species include the following major air and climate pollutants: SO_2, NO_x, CO, non-methane volatile organic compounds (NMVOC), NH_3, PM_{10}, $PM_{2.5}$, BC, organic carbon (OC), and CO_2. CH_4, and N_2O that were included in REASv2.1 are not in the scope of this version. CH_4 is one of important components of SLCP and will be considered in the next version. The target years are from 1950 to 2015. The start year was chosen to be 1950 as severe air pollution in Japan started from mid-1950s. Figure 1 shows the inventory domain of REASv3.1 which includes East, Southeast, and South Asia. Central Asia and the Asian part of Russia, which were target areas of REASv2.1 are not included in REASv3 because of the difficulty in collecting necessary data for estimating long historical emissions in these areas. China, India, and Japan have been divided into 33, 17, and 6 regions, respectively to reduce the uncertainties in the spatial distribution as that of the previous versions. All target countries and sub-regions are listed in Table S1 in the Supplementary material. Spatial resolution is 0.25° × 0.25°, except in the case of large power plants, which are treated as point sources. Temporal resolution is monthly. These spatial and temporal resolution settings are the same as those of REASv2.1, except that more information for grid allocation and temporal distribution have been collected in this version (see Section 2.5).~~

~~The source categories considered in REASv3.1 are the same as those in REASv2.1. Major sources include fuel combustion in power plants, industry, transport, and domestic sectors. Non-combustion sources include industrial process, evaporation (NMVOC), and agricultural activities (NH_3). NO_x emissions from soil as well as from international and domestic aviation and navigation, including fishing ships are exceptions and were not included in REASv3.1. For domestic and fishing ships, emissions were roughly estimated but their results were only used for comparison with other inventories (see Section 3.3).~~

~~In the case of Japan, the Republic of Korea, and Taiwan, REASv2.1 relied on results from other projects or officially opened data. However, in order to develop long historical inventories in a consistent way, their emissions were originally estimated in REASv3.1. One exception to this is NMVOC emission from evaporative sources, which was obtained from the Ministry of the Environment of Japan (MOEJ, 2017) and the National Air Pollutants Emission Service of the National Institute of Environmental Research (http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do) for Japan and the Republic of Korea, respectively. Another is NH_3 emissions from agricultural activities. Data of base year (2000 and 2005 for Japan and 2000 for others) were obtained from other research works (see Sect. 2.4).~~

2.2 Stationary sources

2.2.1 Basic methodology

Emissions from stationary fuel combustion and industrial processes are traditionally calculated using activity data and emission factors, including the effect of control technologies. In order to increase the accuracy of estimation and to analyze the effects of abatement measures, emissions should be calculated using information on technologies related to emission sources as much as possible. In REASv3~~.1~~, emissions from stationary combustion and industrial processes are estimated based on the following equation:

$$E = \sum_i \sum_j \sum_{k,l} \{A_{i,j} \times F_{i,j,k,l} \times EF_{i,j,k} \times (1 - R_{i,j,l})\} \quad (1)$$

where, E represents emission, i is the type of activity data, j is the type of sector category, k is the type of technology related to emission factor, l stands for the control technology after emission, A is amount of activity data, EF is the emission factor of each technology, R is the removal efficiency of each technology, and F is the fraction rate of activity data for combination of i, j, k, and l. When SO_2 emissions from combustion sources are estimated using sulfur contents of fuels, $EF_{i,j,k}$ in eq. (1) is calculated, as follows:

$$EF_{i,j,k} = NCV_{i,j} \times S_{i,j} \times (1 - SR_{i,j,k}) \times 2 \quad (2)$$

where, NCV is the net calorific value of fuel, S is the sulfur content of fuel, and SR is the sulfur retention in ash for combination of i, j, and k. 2 is a factor to convert the value of S to SO_2.

Unfortunately, in the case of Asia, information available on emission factors and removal efficiencies is limited. Even though there is information on the introduction rates of technologies both for emission factors and removal efficiencies, they are available independently. Therefore, for most cases, an average of the removal efficiencies is calculated using the values of each abatement equipment and its penetration rate. Then, the average removal efficiencies are commonly used to calculate the emission factors of each technology.

Note that several sub-sectors in stationary sources such as coke production and cement industry include both combustion and non-combustion emission sources. See Sects. S2.4.1 and S2.4.2 of the Supplement for details.

2.2.2 Activity data

Fuel consumption is the core activity data of the emission inventory of air pollutants and greenhouse gases. For most countries, the amount of energy consumption for each fuel type and sector was primarily obtained from the International Energy Agency (IEA) World Energy Balances (IEA, 2017). For China, province-level tables in the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017) were used. For countries and regions whose energy data are not included in IEA (2017), fuel consumption data were taken from the United Nations (UN) Energy Statistics Database (UN, 2016) and the UN data, which is a web-based data service of the UN (http://data.un.org/). See Sect. S.3.1.1 of the Supplement for definition of fuel types.

One major obstacle in this study was collecting activity data for the entire target period of REASv3~~.1~~, that is from 1950-2015. IEA (2017) includes data from Japan during 1960-2015 and those from other countries during 1971-2015; however, for many countries, fuel types and sector categories, the oldest years when data exist are more later than 1971. Furthermore, past data for sectors do not contain as many categories. For example, coal consumption data in detailed sub-categories of the industrial sector existed in Indonesia only after 2000, but corresponding data are only available for industry total before 1999. In this case, relative ratios of fuel consumption in detailed sub-categories to total industry in 2000 were used to distribute the total industry data to each sub-category for the years before 1999. This procedure is performed for similar cases for all sectors and sometimes for total final consumption. In cases where data did not exist beyond a certain year, fuel consumption data were extrapolated using trends of related data for each sub-category. For example, power generation and amount of industrial products were used to observe trends of fuel consumption in power plant and each industry's sub-category, respectively. Data for long historical trends were obtained from a variety of sources. For example, power generation data and amounts of major industrial products were obtained from Mitchell (1998) and national and international statistics as well as related literatures were surveyed. See Sect. S3.1.2 of the Supplement for details of data sources of fuel consumption and assumptions to estimate missing historical data.~~For example, power generation, amount of industrial products, and population were used to observe trends of fuel consumption in power plant, each industry's sub-category, and residential sectors, respectively. Data for long historical trends were obtained from a variety of sources. For example, power generation data were obtained from Mitchell (1998) and population data were taken from UN (2018). Sources for industrial production are described toward the end of this section.~~ For China, data of CESY for each province were available from 1985 to 2015. During 1950-1984, first, total energy data in China were developed based on IEA (2017) and then, fuel consumption in each province was extrapolated using the total data of China in each fuel type and sector category. See Sect. S3.1.3 of the Supplement for details of regional fuel consumption data in China. For countries which used Energy Statistics Database, fuel consumption of each fuel and sector was taken from the UN data (available at http://data.un.org/~~Default.aspx~~) for the period between 1990-2015 and was extrapolated using the trend of total consumption of each fuel type obtained from the UN Energy Statistics Database.

As described in Section 2.1, India and Japan have 17 and 6 sub-regions, respectively. Therefore, for them, country total data of IEA (2017) need to be divided for each sub-region. For Japan, energy consumption statistics of each prefecture that were obtained from the Agency for Natural Resources and Energy (available at https://www.enecho.meti.go.jp/statistics/energy_consumption/ec002/results.html) were used as default weighting factors to allocate country total data to the six regions. Similarly, for India, default weighting factors for regional allocation were estimated from TERI (The Energy and Resources Institute) Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018), Annual Survey of Industries (Ministry of Statistics & Programme Implementation, available at http://www.csoisw.gov.in/cms/en/1023-annual-survey-of-industries.aspx), and Census of India (Chandramouli, 2011)~~Yevich and Logan (2003)~~, among others. In general, details of these weighting factors are less than those of the country's total fuel consumption. In addition, these data are not available for all the years during 1950-2015. Therefore, regional allocation factors for some sectors were developed independently if corresponding proxy data were available. For the power plant sector, generation capacities of each region and year were calculated as proxy data using the World Electric Power Plants Database (WEPP) (Platts, 2018). For India, traffic volumes (see Section 2.3.1) and amount of industrial production in each region (see the last paragraph of this section) were used as proxy data. Details of regional fuel consumption data in India and Japan were provided in Sects. S3.1.4. and S3.1.5, respectively.

Similar to REASv2.1, large power plants are treated as point sources in REASv3~~.1~~ and are updated based on REASv2.1 database. Before 2007, power plants that were classified as point sources were the same as those in REASv2.1 and their information, such as generating capacities, and start and retire years were updated using WEPP. During 2000 to 2007, fuel consumption data were the same as that in REASv2.1. In REASv3~~.1~~, power plants whose start years were after 2007 and generation capacities were larger than 300 MW were added as new point sources. Fuel consumption of new power plants were estimated based on relations between fuel consumption amounts and generation capacities of the point data in REASv2.1. If the (A) total fuel consumption of each power plant in a country is larger than (B) the corresponding data in power plant sector, values of each power plant were adjusted by ratios of (B) per (A). If (B) was larger than (A), differences between (B) and (A) were treated as data of area sources. See also Sect. S3.1.6 of the Supplement for fuel consumption data in power plants.

For emissions from industrial processes, activity data included amount of industrial products. Corresponding data were mainly obtained from related international statistics and national statistics. For example, iron and steel production data were taken from Steel Statistical Yearbook (World Steel Association, 1978-2016) and data for non-ferrous metals and non-metallic minerals were obtained from the United States Geological Survey (USGS) Minerals Yearbook (USGS, 1994-2015). Brick production data were obtained from a variety of sources, such as Zhang (1997), Maithel (2013), Klimont et al. (2017), and the UN data. For China and India, the authors also used internet database services, namely China Data Online (https://www.china-data-online.com/) and Indiastat (https://www.indiastat.com/), respectively, which provided both national and regional statistics. The USGS Minerals Yearbook (USGS, 1994-2015) also provided information on plants in each sub-region of China, India, and Japan. Data in the aforementioned statistics were not available for the early years of the target period of REASv3.1. In such cases, data of Mitchell (1998) were used as factors to extrapolate the activity data until 1950. Details of activity data related to industrial production and other transformation were described in Sect. S4.1 of the Supplement.

2.2.3 Emission factors

Setting up of emission factors and removal efficiencies for stationary combustion and industrial processes are difficult procedures, especially for a long historical emission inventory. In this study, emission factors without effects of abatement measures were set, which were used for the entire target period of REASv3~~.1~~. Then, effects of control measures were set considering their temporal variations, both for abatement measures before emissions such as using low sulfur fuels and low NO_x burners and those after emissions such as flue gas desulfurization (FGD) and electrostatic precipitator (ESP). These settings were done for each country and region based on country and region-specific information. However, such information is still limited, especially in the Asian region. Therefore, default values of unabated emission factors were selected and default removal efficiencies were set to zero. Then, these default values were updated in case information and literature on each country and region were available. For default emission factors, a majority of settings was continuously used from REASv2.1, but some of them, including effects to control measures (net emission factors) were changed to unabated emission factors. Default emission factors were mainly obtained from Kato and Akimoto (1992) for SO_2 and NO_x; Bond et al. (2004), Kupiainen and Klimont (2004), and Klimont et al. (2002, 2017) for PM species; the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006) for CO_2; and the AP-42 (US EPA, 1995), the Global Atmospheric Pollution Forum Air Pollutant Emission Inventory Manual (SEI, 2012), Shrestha et al. (2013), the EMEP/EEA emission inventory guidebook 2016~~09~~ (EEA, 2016), and other literatures for others.

For country and region-specific settings, in addition to literatures used in REASv2.1 (see Kurokawa et al., 2013), new information, especially for technologies related to settings of emission factors and removal efficiencies was surveyed. Although such information is still limited in Asia, the volume of accessible information on China is relatively large. General information on China in recent years was mainly obtained from Li et al. (2017b) and Zheng (2018). Introduction rates of technologies were obtained from Hua et al. (2016) for cement, Wu et al. (2017) for iron and steel, Huo et al. (2012a) for coke ovens, and Zhao et al. (2013, 2014, and 2015) for a variety of sources. For India, information for technology settings was mainly taken from Sadavarte and Venkataraman (2014), Pandey et al. (2014), Guttikunda and Jawahar (2014), and Reddy and Venkataraman (2002a). For power plants, WEPP database has elements for installed equipment to control SO_2, NO_x, and PM species which were used for settings of emission factors and removal efficiencies of power plants treated as point sources. However, these data are not available for most power plants, especially in Asia. Therefore, in the case of South and Southeast Asia, a variety of literatures, such as Sloss (2012) and UN Environment (~~2017, ~~2018) were referred to, to set emission factors and removal efficiencies. For Japan, introduction of control technologies for air pollutants were initiated earlier than other countries in Asia. A lot of domestic reports for air pollution and control technologies in power and industry plants published in Japanese, such as MRI (2015), Shimoda (2016), Suzuki (1990), and Goto (1981) were referred to, to determine emission factors, removal efficiencies, and their temporal variations.

Details of emission factors and settings of emission controls for stationary combustion sources were provided in Sect. S3.2 of the Supplement. Those for stationary non-combustion emissions from industrial production and other transformation sectors were described in Sect. S4.2. Activity data and emission factors of NMVOC from chemical industry were obtained from Sects. S5.1.5 and S5.2.5, respectively. Those for NH_3 emissions from industrial production were provided in Sect S8.3.

2.3 Road transport

2.3.1 Basic methodology

Methodology for road transport sector is the same as that of REASv2.1. Equations to estimate hot and cold start emissions (except for SO_2 and CO_2) are, as follows:

$$E_{HOT} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi}\} \quad (3)$$

where, E_{HOT} is the hot emission, i is the vehicle type, NV is the number of vehicles in operation, ADT is the annual distance traveled, and EF_{HOT} is the emission factor. SO_2 emissions are calculated using sulfur contents in gasoline and diesel consumed in road transport sector, assuming sulfur retention in ash is zero. CO_2 emissions are estimated by calculating the consumption amounts of fuels (gasoline, diesel, liquefied petroleum gas, and natural gas) and the corresponding emission factors (IPCC, 2006). Details for SO_2 and CO_2 from road transport were described in S6.2.3 of the Supplement.

Cold start emissions (E_{COLD}) are estimated for NO_x, CO, PM_{10}, $PM_{2.5}$, BC, OC, and NMVOC using the following equation:

$$E_{COLD} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi} \times \beta_i(T) \times F_i(T)\} \quad (4)$$

where, β is the fraction of distance traveled driven with a cold engine or with the catalyst operating below the light-off temperature, and F is the correction factor of EF_{HOT} for cold start emission. β and F are functions of temperature T and are ~~estimated based on~~taken from EEA (2016) (See Sect. S6.2.1 of the Supplement for additional information of the settings). For Japan, the ratios of cold start and hot emissions for each vehicle type ~~was~~ were estimated from the JEI-DB. ~~Furthermore~~Then, cold start emissions were calculated by hot emissions and the ratios for each vehicle type. In REASv3, effects of regulations on cold start emissions were ignored and need to be considered in the next version.

For evaporation from gasoline vehicles, emissions (E_{EVP}) were estimated using the following equation of Tier 1 of EEA (2016):

$$E_{EVP} = \sum_i \{NV_i \times EF_{EVPi}(T)\} \quad (5)$$

where, EF_{EVP} is the emission factor as a function of temperature. For Japan, evaporative emissions in 2000, 2005, and 2010 were obtained from the JEI-DB and those between 2000 (2005) and 2005 (2010) were interpolated. For emissions before 2000 and after 2010, emissions from running loss were extrapolated using trends of traffic volume and those from hot soak loss and diurnal breaking loss were extrapolated by trends of vehicle numbers. See Sect. S6.3 of the Supplement for the NMVOC evaporative emissions.

2.3.2 Activity data

Basic activity data of road transport sector include number of vehicles in operation for each type. Data on the registered number of vehicles are available in the national statistics of each country and the World Road Statistics (IRF, 1990-2018). If these statistics did not contain data until 1950, the numbers were extrapolated using trends of data for aggregated vehicle categories in Mitchell (1998). For China, data for each sub-region were obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986-2016) and the China Data Online. Those for India were taken from TERI Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018)~~Road Transport Yearbook (Morth, 2003-2017)~~ and the Indiastat. A problem that was encountered was that registered vehicles were not always in operation. For India, the number of vehicles obtained as registered vehicles were corrected based on Baidya and Borken-Kleefeld (2009)~~, Pandey and Venkataraman (2014),~~ and Prakash and Habib (2018). For other countries, the number of registered vehicles were considered as those in operation due to lack of information. In addition, to estimate emissions, these numbers must be further divided into vehicles based on each fuel type. However, such information is not easily available in national statistics. In this study, settings of Streets et al. (2003a) and REASv2.1 were used as default and were updated if national information was available, such as He et al. (2005), Yan and Crookes (2009), Sahu et al. (2014), and Malla (2014). ~~In this study, settings of REASv2.1 were used as default and were updated if new information was available, such as Pandey and Venkataraman (2014), Sahu et al. (2014) and Mishra and Goyal (2014).~~ If the number of LPG and CNG vehicles were available only for recent years, data were extrapolated using amounts of fuel consumption in road transport sector in IEA (2017).

Emission factors of road transport sector used in this study were given as emission amounts per traffic volumes. Therefore, annual vehicles kilometer traveled (VKT) per each vehicle type need to be set for each country. We used data of Clean Air Asia (2012) for many countries. Clean Air Asia (2012) includes data for China and India, but data of China were estimated based on Huo et al. (2012b) and those of India were set after Prakash and Habib (2018) and Pandey and Venkataraman (2014). For Japan, the total annual VKT for detailed vehicle types were obtained from reports of Pollutants Release and Transfer Register published by the Ministry of Economy Trade and Industry until 2001 (METI, 2003-2017), which was originally estimated from Road Transport Census of Japan developed by the Ministry of Land, Infrastructure, Transport and Tourism. Before 2001, the total annual VKT was extrapolated using data of more aggregated vehicle categories in the Annual Report of Road Statistics (MLIT, 1961-2016) until 1960 and from the Historical Statistics of Japan (Japan Statistical Association, 2006) until 1950.

Details of number of vehicles and annual vehicles kilometer traveled were described in Sect S6.1.1 of the Supplement.

2.3.3 Emission factors

For most countries, road transport is one of major causes of air pollution. In many Asian countries, vehicle emission standards were introduced after the late 1990s and were strengthened in phases (Clean Air Asia, 2014). Therefore, for road vehicles, year-to-year variation of emission factors must be taken into considered for a long historical emission inventory. In REASv3, emission factors of NO_x, CO, NMVOC, and PM species for exhaust emissions from road vehicles were estimated by following procedures:

1. Emission factors of each vehicle type in a base year were estimated.
2. Trends of the emission factors for each vehicle type were estimated considering the timing of road vehicle regulations in each country and the regions and the ratios of vehicles production years.
3. Emission factors of each vehicle type during the target period of REASv3 were calculated using those of base years and the corresponding trends.

The information of road vehicle regulations in each country and regions were taken from Clean Air Asia (2014). For the ratios of vehicle production years, due to lack of information, data for Macau derived from Zhang et al. (2016) were used for Hong Kong, Republic of Korea, and Taiwan and those from Japan Environmental Sanitation Center and Suuri Keikaku (2011) for Vietnam were used for other countries and regions. Then, trends of emission factors were estimated using the above data and information with values of Europe and United States standards. Finally, emission factors used to estimate emissions were calculated for each vehicle type. For most countries, the years just before the regulations for road vehicles began were set as base years and no-controlled emission factors that were used in REASv1.1 and REASv2.1 were adopted for emission factors of the base years. Countries for which information on regulations were not obtained, the no-controlled emission factors were used for the entire target period of REASv3. For China and India, emission factors in 2010 were estimated as base year's data using recently published papers, such as Huo et al. (2012b), Xia et al. (2016), Mishra and Goyal (2014), and Sahu et al. (2014). For Republic of Korea and Taiwan whose emissions were not originally estimated in REASv2.1, emission factors were estimated with high uncertainties based on values of Europe and United States standards, respectively. For Japan, emission factors for each emission standard are available for several vehicle speeds (JPEC, 2012a). Combining these data with information for annual VKT of each vehicle speed, ratios of vehicle ages, and time series of regulation standards, emission of road transport in Japan were calculated. Details of emission factors of exhaust emissions were provided in Sect. S6.2 of the Supplement. ~~For most countries, road transport is one of major causes of air pollution. In Asian countries, vehicle emission standards were introduced after the late 1990s and were strengthened in phases (Clean Air Asia, 2014). Therefore, for road vehicles, year-to-year variation of emission factors must be taken into considered for a long historical emission inventory. In this study, trends in emission factors were estimated considering the timings of road vehicle regulations in each country and the ratios of vehicle production years. Moreover, emission factors of each vehicle type in selected base years were estimated. Finally, emission factors for the entire target period of REASv3.1 were calculated. For year-to-year variations of emission factors, ratios of vehicles based on annual production for each country were estimated using data on the age of vehicles from literatures, such as Zhang et al. (2016), Pandey and Venkataraman (2014), and Huo et al. (2012b). Moreover, emission factors for each production year were selected from the values of Europe and the United States standards and were based on regulation schedules of the target country. Then, tentative emission factors of the target country for each year were estimated during the target period of REASv3.1 using ratios of vehicles' production years as weighting factors. Finally, trends in emission factors from base years were calculated using the calculated tentative emission factors. For most countries, the years just before the regulations for road vehicles began were set as base years and no-controlled emission factors that were used in REASv2.1 were adopted for emission factors of the base years. Countries for which information on regulations were not obtained, the no-controlled emission factors were used for the entire target period of REASv3.1. For China and India, emission factors in 2010 were estimated to be base year's data using setting of REASv2.1 and recently published papers, such as Huo et al. (2012b), Xia et al. (2016), Mishra and Goyal (2014), and Sahu et al. (2014). For the Republic of Korea and Taiwan whose emissions were not originally estimated in REASv2.1, tentative emission factors calculated by the above procedures were used. For Japan, emission factors for each emission standard are available for several vehicle speeds (JPEC, 2012a). Combining these data with information for annual VKT of each vehicle speed, ratios of vehicle ages, and time series of regulation standards, emission of road transport in Japan were calculated.~~

2.4 Agricultural sources

REASv3 includes NH_3 emissions from manure management and fertilizer application in agricultural sources. Approaches similar to REASv2.1 were adopted to estimate historical emissions and develop monthly gridded data. First, annual emissions of each country and sub-region except for Japan and their gridded data for the year 2000 were selected from

REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) as base data. For Japan, corresponding base data were obtained from REASv2.1 (Kurokawa et al., 2013: JPEC 2012a, b, c; 2014) for the year 2000 and 2005. Second, trends of emissions during 1950-2015 were estimated for each country and sub-region. Third, annual emissions for the period were calculated using the trends and base data. Fourth, changes in spatial distribution from base years to target years and monthly variations in each country and sub-region were estimated. Finally, monthly gridded data of emissions were developed for 1950-2015. For Japan, emission data during 2001-2004 were interpolated between those in 2000 and 2005. Details for manure management and fertilizer application are described in Sections 2.4.1 and 2.4.2, respectively.

2.4.1 Manure management

Trends in NH_3 emissions from manure management of livestock, except for its application as fertilizer, were estimated based on the Tier 1 method of EEA (2016). In this method, emissions are calculated based on the numbers of livestock and the corresponding emission factors. Statistics on the number of animals, such as broilers, dairy cow, and swine are mainly obtained from FAOSTAT (available at http://www.fao.org/faostat/en/) of the Food and Agriculture Organization (FAO) of the UN~~nited Nations~~ from the period between 1961 to 2015. For the years before 1960, data were obtained from Mitchell (1998). National statistics were surveyed for data on provinces, states, and prefectures in China, India, and Japan, respectively to develop activity data for each sub-region. Emission factors are obtained from EEA (2016). For spatial distribution, changes in grid allocation for each country and sub-region from the year 2000 were estimated using EDGARv4.3.2 from 1970 to 2012. Grid allocation factors in 1970 and 2012 were used for the period before and after 1970 and 2012, respectively. For temporal variations, monthly allocation factors are estimated as a function of temperature by referring to the monthly variations of emissions in Japan based on the JEI-DB. Detailed methodologies and data sources for manure management were provided in Sect. S8.1 in the Supplement.

2.4.2 Fertilizer

In most countries, fertilizer application is the largest source of NH_3 emissions. Emission trends after the application of manure and synthetic N fertilizer were estimated using EEA (2016). Manure application is one of the processes of manure management whose emissions trend was calculated based on the number of animals and the corresponding emission factor. For synthetic N fertilizer, trends of total consumption of fertilizer were used in REASv2.1. However, this simple approach causes uncertainties because emission factors are different among types of fertilizer (EEA, 2016). Therefore, in REASv3~~.1~~, emissions from each N fertilizer, such as ammonium phosphate and urea were estimated separately and trends in total emissions were calculated. For ~~special~~ spatial distribution, changes in grid allocation factors for each country and sub-region from the year 2000 were estimated using a historical global N fertilizer application map during 1961-2010, developed by Nishina et al. (2017). Data for 1961 and 2010 were used for the period before 1961 and after 2010, respectively. For seasonal variations, monthly factors of China and Japan were determined based on Kang et al. (2016) and the JEI-DB, respectively. For other countries, data from Nishina et al. (2017) have monthly application amounts in each grid. However, there are cases that some months have high factors, whereas the others have almost zero. Referring to Janssens-Maenhout et al. (2015), we adopted the conservative way, such that the highest monthly factor was set at 0.2 and the factors of all months were adjusted accordingly. See Sect. S8.2 for details of methodologies and data sources for emissions from fertilizer application.

2.5 Other sources

NMVOC emissions from evaporative sources are increasing significantly in Asia along with economic growth. Major sources of NMVOC emissions include usage of solvents for dry cleaning, degreasing operations, and adhesive application as well as for paint use. Fugitive emissions related to fossil fuels, such as extraction and handling of oil and gas, oil refinery, and gasoline stations are also important. However, statistics on activity data and information of emission factors for these sources are often less available than those for fuel combustion and industrial processes. In this study, default activity data and emission factors were obtained from REASv2.1 and were updated if information was available in recently published papers (such as Wei et al. (2011) for China and Sharma et al. (2015) for India). In general, activity data of the past years are not available, and, in such cases, proxy data are prepared for trend factors. For example, population numbers were used for dry cleaning and production numbers of vehicles were used for paint application for automobile manufacturing. GDP was used for default trend factors. For emission calculation, the same equation for stationary combustion was adopted. Details of activity data and emission factors for non-combustion sources of NMVOC were provided in Sect. S5 of the Supplement.

In addition to agricultural activities, latrines are an important source of NH_3, especially in rural areas. Activity data are population numbers in no sewage service areas estimated referring settings of REASv2.1 and emission factor were based on EEA (2016) and SEI (2012). Also, humans themselves are sources of NH_3 emissions through perspiration and respiration. For these sources, population numbers are activity data mainly taken from UN (2018) and emission factors are obtained from EEA (2016). Equation to estimate emission is also the same as that of stationary combustion. Additional data and information for emissions from human and latrines were described in Sects. S8.4 and S8.5, respectively.

In REASv3, aviation and ship emissions including fishing ships are not included, but emissions of fuel combustion in other transport sector (namely, except for aviation, navigation, and road), such as railway and pipeline transport were estimated. Equation (1) is also used for estimating emissions of these sources. See Sect. S7 of the Supplement for additional data and information for other transport sector.~~In REASv3.1, aviation and ship emissions are not included, but emissions of fuel combustion in other transport sector (except for aviation, navigation, and road), such as railway and pipeline transport were estimated. Also, emissions from domestic shipping including fishing ships were roughly calculated for comparison with other inventories (see Section 3.3). Equation (1) is also used for estimating emissions of these sources.~~

2.6 Spatial and temporal distribution

Procedures for developing gridded emission data were the same as those of REASv2.1. Large power plants were treated as point sources, and longitude and latitude of each power plant were provided. Positions of power plants were surveyed based on detailed information, such as names of units, plants, and companies from WEPP (Platts, 2018). These were searched on internet sites, such as Industry About (~~available at~~ https://www.industryabout.com/) and Global Energy Observatory (http://globalenergyobservatory.org/). Positions for newly added power plants in REASv3~~.1~~ as well as those in REASv2.1 were surveyed because some of these services were not available when REASv2.1 was developed. For cement, iron, and steel plants (and non-ferrous metal plants in Japan), REASv3~~.1~~ still did not treat them as point sources due to lack of activity data. However, positions, production capacities, start and retire years for large plants were surveyed similar to power plants and used for developing allocation factors for corresponding sub-sectors. For road transport sector, REASv2.1 used coarse grid allocation data of REASv1.1 with 0.5° × 0.5° resolution. Therefore, in REASv3~~.1~~, grid allocation factors for each country and sub-region, except Japan, were updated using gridded emission data of road transport sector of EDGARv4.3.2 during 1970-2012. Before 1970 (after 2012), data for 1970 (2012) were used. For Japan, gridded emission data of the JEI-DB in 2000, 2005, and 2010 were used to develop grid allocation factors. For the year between 2000 (2005) and 2005 (2010), the JEI-DB data were interpolated. For years before 2000 (after 2010), the JEI-DB data for 2000 (2010) were used. For residential sectors, rural, urban, and total population of HYDE 3.2.1 (Klein Goldewijk et al., 2017) with 5′ × 5′ were used to create allocation factors. Data of HYDE 3.2.1 were available for 1950, 1960, 1970, 1980, 1990, 2000, 2005, 2010, and 2015 and the years between them were interpolated. Spatial distributions of total population were used for grid allocation of all other sources. Detailed methodologies and data sources for grid allocation were provided in Sect. S9.1 in the Supplement.

Methodology to estimate monthly emission data in REASv3~~.1~~ was the same as that of REASv2.1. In general, monthly emissions were estimated by allocating annual emissions to each month using monthly proxy ~~index~~data. Monthly generated power and production amounts of industrial products were used as the monthly allocation factors for power plant sector and corresponding industry sub-sectors, respectively. Basically, monthly factors of REASv2.1 during 2000-2008 were also used in REASv3~~.1~~ and were extended if data existed before (after) 2000 (2008). For the years where surrogate data were unavailable, the data of oldest (newest) year were used before (after) the year. For brick production, monthly allocation factors for Southeast and South Asian countries were estimated ~~using~~ referring Maithel et al. (2012) and Maithel (2013). For the residential sector, monthly variations of emissions were estimated using surface temperature in each grid cell, similar to REASv2.1. Surface temperatures during 1950-2015 were taken from NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html~~available at https://www.esrl.noaa.gov/psd/~~). For Thailand and Japan, most monthly factors were set based on country specific information from Thao Pham et al. (2008) and JPEC (2014), respectively. See Sect. S9.2 of the Supplement for details of monthly variation factors.

3 Results and discussion

3.1 Trends of Asian and national emissions

Trends in air pollutants emissions from Asia, China, India, Japan, and other countries are described in this section, mainly focusing on SO_2, NO_x, and BC emissions as they have important roles in both air pollution and climate change. SO_2 and NO_x are precursors of sulfate and nitrate aerosols, respectively, which are the major components of secondary $PM_{2.5}$. NO_x is also a precursor of ozone. Furthermore, BC is a major component of primary $PM_{2.5}$. $PM_{2.5}$ and ozone not only harm human health and ecosystems, but influence climate change. BC and ozone have a warming effect on climate change, whereas sulfate and nitrate aerosols have a cooling effect. Note that all the air pollutant emissions from major countries and regions ~~each country~~ between 1950 to 2015 categorized based on major sectors and fuel types, are provided in the Supplement material (Figs. S1-S12).

3.1.1 Asia

Table 3~~2~~ summarizes the national emissions of each species in 2015 and the total emissions from Asia in 1950, 1960, 1970, 1980, 1990, 2000, and from 2010-2015. Figure 2 shows emissions of SO_2, NO_x, CO, NMVOC, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) from 1950 to 2015. Average total emissions in Asia during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO_2: 3.2 Tg, 42.4 Tg (13.1); NO_x: 1.6 Tg, 47.3 Tg (29.1); CO: 56.1 Tg, 303 Tg (5.4); NMVOC: 7.0 Tg, 57.8 Tg (8.3); NH_3: 8.0 Tg, 31.3 Tg (3.9); CO_2: 1.1 Pg, 18.6 Pg (16.5); PM_{10}: 5.9 Tg, 30.2 Tg (5.1); $PM_{2.5}$: 4.6 Tg, 21.3 Tg (4.6); BC: 0.69 Tg, 3.2 Tg (4.7); and OC: 2.5 Tg, 6.6 Tg (2.7)~~SO₂: 3.15 Tg, 42.4 Tg (13.5); NOₓ: 1.83 Tg, 47.6 Tg (26.0); CO: 62.2 Tg, 319 Tg (5.13); NMVOC: 9.14 Tg, 61.8 Tg (6.77); NH₃: 7.99 Tg, 31.3 Tg (3.92); CO₂: 1.12 Pg, 18.3 Pg (16.3); PM₁₀: 5.76 Tg, 28.4 Tg (4.92); PM₂.₅: 4.52 Tg, 20.3 Tg (4.50); BC 0.751 Tg, 3.38 Tg (4.51); and OC 2.62 Tg, 6.92 Tg (2.64)~~. Clearly, all the air pollutant emissions in Asia increased significantly during these six decades. However, this increase was different among the aforementioned species. Growth rates of emissions were relatively large for SO_2, NO_x, and CO_2 because the major sources of these species are power plants, industries, and road transport, for which fuel consumption increased significantly along with economic development in Asia. SO_2 increased before the other species because a majority of the emissions were obtained from the combustion of coal, which is easier to obtain than oil and gas. SO_2, NO_x, and CO_2 emissions increased keenly in the early 2000s, along with rapid growth of emissions of these species in China. For NO_x, combustion of oil fuels, especially by road vehicles, contributed to a large growth of emissions in the latter half of 1950-2015. Growth rates of NMVOC have also increased recently due to an increase in the emissions from road vehicles and evaporative sources, such as paint and solvent usage in accordance with economic growth of Asian countries. On the other hand, rates of growth of CO, PM_{10}, $PM_{2.5}$, BC, and OC are relatively small. One reason is that emissions of these species are mainly from incomplete combustion in low temperature and thus, emissions from power plants and large industry plants are relatively small. Another reason is that a major source of these species is the combustion of coal and biofuels in residential sector, which were relatively large even in past years in Asia. Recently, emissions of these species from industries, including combustion and non-combustion processes are increasing. In addition, gasoline and diesel vehicles have contributed recently to the growth of CO and BC emissions, respectively. Agricultural activities, such as manure management of livestock and fertilizer application, which are major sources of NH_3 are rising to support a growing population in Asia. Although the growth rate of NH_3 emissions is smaller than other species, it still shows an increasing trend.

Differences in the trends of emissions were also observed on the basis of countries and regions. SO_2 and NO_x, emissions from Japan were relatively large in Asia during the 1950s-1970s. Emissions from Japan in 1965 are comparable with and are larger than those of China for SO_2 and NO_x, respectively. In 2015, emissions of SO_2 and NO_x in Japan decreased largely and contribute only about 1.5 and 3.8~~4~~% of Asia's total emissions, respectively. Similar tendencies were also observed in the case of other species. In 2015, China was the largest contributor of emissions for all the species. Recently, emissions of most species in China have shown decreasing or stable trends. In the case of SO_2, China contributed about 72% of emissions in 2005, but about 49~~8~~% in 2015. On the other hand, emissions and their relative ratios are increasing in the case of India. Actually, contribution rates of SO_2, NO_x, and BC emissions in India increased from 14%, 16~~7~~%, and 23~~4~~% in 2005 to 30~~1~~%, ~~23~~22%, and ~~28~~27% in 2015, respectively. Li et al. (2017c) suggested that, in 2016, SO_2 emissions in India exceeded those in China. Recent increase in air pollutants emissions have also been observed in ~~Southeast Asia~~SEA and ~~South Asia other than India~~OSA. ~~Furthermore~~On the other hand, emissions from ~~East Asia other than China and Japan~~OEA started to increase slightly later than Japan and then, recently show decreasing trends mainly reflecting trends of emissions from Republic of Korea and Taiwan.

3.1.2 China

Growth rates of all pollutants emissions in China in these 60 years estimated from average during 1950-1955 and 2010-2015 are as follows: 21% for SO_2, 54% for NO_x, 7.0% for CO, 13% for NMVOC, 4.7% for NH_3, 28% for CO_2, 6.8% for PM_{10}, 6.1% for $PM_{2.5}$, 5.5% for BC, and 2.7% for OC.~~Air pollutants emissions in China averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are, as follows: SO_2 1.10 Tg, 24.6 Tg (22.4); NO_x 0.544 Tg, 26.6 Tg (48.8); CO 27.2 Tg, 188 Tg (6.90); NMVOC 2.73 Tg, 27.8 Tg (10.2); NH_3 2.94 Tg, 14.0 Tg (4.74); CO_2 0.414 Pg, 11.4 Pg (27.5); PM_{10} 2.46 Tg, 16.8 Tg (6.81); $PM_{2.5}$ 1.96 Tg, 12.0 Tg (6.10); BC 0.316 Tg, 1.76 Tg (5.55); and OC 1.10 Tg, 2.93 Tg (2.65).~~ ~~Emission trends in China for all the pollutants in each sector and for each fuel type during 1950-2015 were presented in Figs. S1 and S2, respectively.~~ It was observed that emissions of all pollutants increased largely during these six decades, but most species reached their peaks up to 2015 as shown in Fig. 2. Exceptions to this were NMVOC, NH_3, and CO_2; however, their growth rates are at least small or almost zero. Emission trends in China for all the pollutants in each sector and for each fuel type during 1950-2015 were presented in Figs. S1 and S2, respectively.

Figure 3 shows recent trends in actual emissions (solid colored areas) and reduced emissions by control measures (hatched areas) from each sector for SO_2, NO_x, and BC during 1990-2015 in China. The reduced emission by control measures was the difference between emissions calculated without effects of all control measures (such as FGD, ESP, using low sulfur fuels, regulated vehicles, etc.) and actual emissions. Total CO_2 emissions were also plotted for each panel of Fig. 3 as an indicator of energy consumption. Note that reduced emissions here do not include effects of substitution of fuel types, such as from coal to natural gas.

For SO_2, most emissions in China were from coal combustion which controlled trends of total emissions. SO_2 emissions in China increased rapidly in the early 2000s, but decreased after 2006 and showed a continuous decline until 2015. Drastic changes in the 2000s were mainly caused by emissions from coal-fired power plants, which increased rapidly along with large economic growth and later decreased due to the introduction of FGD based on the 11th Five Year Plan of China. After 2011, control measures for large industry plants started to become effective and as a result, total emissions in 2015 became comparable with those in 1990. Without effects of emission controls, emissions from power plants and industry in 2015 would be 3.7 and 2.6 times higher than those in 2000, respectively. In this study, the emissions in 2015 were estimated to be reduced by about 90% for power plants and 76 % for industry. On the other hand, even without emission controls, SO_2 emissions from power plants were almost stable after 2010. The same tendencies were also found in CO_2. One considerable reason is an increasing energy supply from nuclear power plants. According to IEA (2017), the total primary energy supply from nuclear power plants increased rapidly recently and those in 2015 were about 2.3 time higher than in 2010.

Similar to SO_2, NO_x emissions increased rapidly from the early 2000s, but continued to increase until 2011 and then, started to decline. In the 2000s, low NO_x burner to power plants and regulation of road vehicles were introduced, but their effects were limited. From 2011, introduction of denitrification technologies, such as selective catalytic reduction (SCR) to large power plants and regulations for road vehicles were strengthened based on the 12th Five Year Plan of China. Three major drivers of NO_x emissions in China are power plants, industry sector, and road transport. If no emission mitigation was considered, their emissions would be increased by 3.6, 3.0, and 4.7 times from 2000 to 2010, respectively. In 2015, reduction rates of emissions due to emission controls were about 61%, 19%, and 62% for power plants, industry, and road transport respectively. As a result, in 2015, NO_x emissions were about 81~~3~~% of their peak values in 2011. In 2015, actual NO_x emissions from industry sector were larger than those from power plants and road transport which were comparable each other. Major industries such as iron and steel, chemical and petrochemical, and cement industries were large contributor of NO_x emissions in China.

For BC, emissions also increased from early 2000s, but growth rates were smaller than SO_2 and NO_x due to the effects of control equipment in the industrial sector. Actually, trends of BC emissions assuming no emission controls were close to those of CO_2 and the BC emissions in 2015 were increased by 2.2 times from 2000. The emissions in 2015 were reduced by about 41% by abatement measures in industry plants and 9% by regulations especially for diesel vehicles. In 2015, large contributors in industry sectors were brick production, coke ovens, and coal combustion in other industry plants. Another reason of relatively small growth rates could be that BC emission factors for coal-fired power plants are originally low. Recently, BC emissions from residential sector as well as industrial sector show decreasing trends. In this study, the reductions in BC emissions in residential sector were mainly caused by a decrease in emissions from biofuel combustion.

During 2010 to 2015, consumptions of primary solid biofuels were reduced about ~~30~~28%, whereas consumption of natural gas and liquefied petroleum gas increased about 62~~0~~% in the residential sector.

For CO, most emissions in the 1950s were from residential sectors and~~, which~~ gradually increased with increasing coal consumption in the industrial sector. CO emissions increased largely in 2000s due to coal combustion and iron and steel production processes. Recently, CO emissions have seen a decline. A major reason for this declining trend is the decrease in biofuel consumption in residential sector and the phasing out of shaft kiln with high CO emission factor in the cement industry. NMVOC emissions increased significantly from the early 2000s, similar to other species. However, their major sources were different from others. Recent increasing trends are not caused by stationary combustion sources, but by road transport and evaporative sources, such as paint and solvent use. In particular, emissions from non-combustion sources increased largely from 2000 to 2015 (about 3.7 time) and as a result, their contribution rate in 2015 was about 65%. Growth rates of NMVOC emissions tended to slow down around 2015, but emissions increased almost monotonically after the 2000s. NH_3 emissions were mostly from agricultural activities. In China, emissions from fertilizer application showed a significant increase from the early 1970s to the early 2000s. In recent years, NH_3 emissions are almost stable. For PM_{10} and $PM_{2.5}$, majority of the emissions are from the industrial sector, followed by residential sector and power plants. Emissions increased largely from the early 1990s mainly due to coal combustion and industrial processes, especially in cement plants. Compared to SO_2 and NO_x, growth rates of PM_{10} and $PM_{2.5}$ emissions during the early 2000s were small, and later decreased due to the effects of control equipment in industrial plants. OC emissions were mostly from biofuel combustion in the residential sector. Contributions from the industrial sector has been increasing recently, but total OC emissions have decreased due to reduced usage of biofuels. CO_2 emissions were mainly controlled by coal combustion and their trend were similar to those of SO_2, NO_x, and BC without emission controls as shown in Fig.3. After 2011, CO_2 emissions in China were found to be almost stable. As described above, one reason is a trend of emissions from power plants. In addition, emissions from coal combustion in industry sectors were slightly decreased from 2014 to 2015.~~NOx until 2011. After 2011, CO2 emissions in China were found to be almost stable.~~

3.1.3 India

Growth rates of air pollutants emissions in India based on averaged values during 1950-1955 and 2010-2015 are as follows: 19% for SO_2, 23% for NO_x, 4.2% for CO, 5.3% for NMVOC, 3.1% for NH_3, 8.9% for CO_2, 4.8% for PM_{10}, 4.0% for $PM_{2.5}$, 4.8% for BC, and 2.8% for OC.~~Emissions of air pollutants in India averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO2 0.541 Tg, 10.5 Tg (19.5); NOx 0.484 Tg, 9.62 Tg (19.9); CO 16.1 Tg, 62.9 Tg (3.91); NMVOC 2.86 Tg, 14.3 Tg (4.99); NH3 2.95 Tg, 9.26 Tg (3.14); CO2 0.299 Pg, 2.64 Pg (8.82); PM10 1.40 Tg, 6.32 Tg (4.50); PM2.5 1.17 Tg, 4.57 Tg (3.91); BC 0.203 Tg, 0.903 Tg (4.45); and OC 0.701 Tg, 1.98 Tg (2.83).~~ Figures S3 and S4 provide trends of emissions in India from each sector and fuel type for all the pollutants, respectively, from 1950 to 2015. In general, all the air pollutants show monotonous increase from 1950 to 2015 and growth rates (especially of recent years) are larger for SO_2, NO_x, NMVOC, and CO_2, which is similar to the case of Asia.

Figure 4 shows trends in emission of SO_2, NO_x, and BC from each fuel type as well as sector with total CO_2 emissions during 1950-2015 in India. Clear differences were seen in the structure of emissions in these species. For SO_2, large parts of emissions were from coal combustion in power plants and industry sector. SO_2 emissions in 2015 were about 3.3 times larger than those in 1990 and contribution rates of the increases from power plants and industry sectors were about 66% and 33%, respectively. Trend so total ~~Recently, a rapid increase in SO_2 emission was mainly from coal-fired power plants. For~~ NO_x, emissions ~~trends~~ were close to those of SO_2 and contributions from coal-fired power plants were also large. ~~However~~In addition, for NO_x, contribution ~~and growth rates~~ from road transport especially diesel vehicles were ~~almost~~ comparable with those of power plants ~~for NO_x~~. Around the year 2005, the contributions from road transport were almost the same or slightly larger than power plants. However, from 2005 to 2015, growth rates of NO_x emissions from power plants were about twice higher than those of road transport emissions. For BC, contributions from the residential sector and biofuel combustion were large, especially in the 1950s-1960s. Contribution rates of residential sector were ~~76~~73% in 1950 and ~~47~~38% in 2015, and those of biofuel combustion, which were mainly used in residential sector and some parts are used in industry sector were ~~87~~86% in 1950 and ~~53~~45% in 2015. ~~However~~On the other hand, recent increasing trends of BC emissions were also caused by growth of emissions from diesel vehicles and industry sector. From 1990 to 2015, contribution rates of increased emissions from industry, road transport, and residential sectors were 27%, 43%, and 23%, respectively. For recent trends, relative ratios of SO_2 emissions from power plants were increased from 43% to ~~60~~59% during 1990-2015. For NO_x, contribution rates from both power plants and road transport were increased and accounted for about ~~80~~75% of the total emissions in 2015. Even in 2015, about half of the BC emissions were from the residential sector. However, as previously described, recent emission growths were mainly caused by the industrial sector and road transport. These tendencies were similar to Japan and China during their rapid emission growth periods. These features were consistent with trends of CO_2 emissions. Before the mid-1980s, majority of CO_2 emissions were from biofuel combustion and the trends were close to those of BC. Then, recently, contributions from fossil fuel combustion increased largely and trends of CO_2 became close to those of SO_2 and NO_x, especially after the early 2000s.

Trends and structure of CO emissions were similar to those of BC but contribution rates of the residential sector were larger and those from road transport (mainly from gasoline vehicle) were smaller, as compared to BC. On the other hand, for recent trends, half (51%) of increased emissions during 2005 and 2015 were from industry sector. Similar ~~This~~ tendency was also found in OC; however, relative ratios of emissions from residential sector were much larger (about 71~~5~~% in 2015) and those of industry and road transport sectors were much smaller. For PM_{10} and $PM_{2.5}$, a majority of the emissions was from residential and industrial sectors. Both amounts were almost comparable in PM_{10} and those from residential sectors were larger in $PM_{2.5}$. Different from BC and OC, contributions from coal-fired power plants exist in PM_{10} and $PM_{2.5}$ whose contribution rates in 2015 are about 20~~2~~% and 13~~7~~%, respectively. For NMVOC, most emissions were from biofuel combustion before the 1980s. Later, emissions from variety of sources, such as road transport, extraction and handling of fossil fuels, usage of paint and solvents are increasing and are controlling recent trends. For increases of emissions from 1990 to 2015, about 52% were from stationary combustion and road transport and the rest were from stationary non-combustion sectors such as paint and solvent use. Most NH_3 emissions are from agricultural activities. Contributions from manure management and fertilizer use were comparable before 1980s. However, emissions from fertilizer application have increased largely which are now determining recent trends. ~~For CO_2, trends during 1950-2015 were similar to SO_2 and NO_x, which show rapid growth after the 2000s mainly due to increasing consumption of fossil fuels.~~

3.1.4 Japan

As described in Sect. 3.1.1, trends of air pollutants emissions in Japan were different from other countries and regions in Asia. The t~~Air pollutants emissions in Japan averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are as follows: SO_2 1.23 Tg, 0.601 Tg (0.491); NO_x 0.483 Tg, 2.03 Tg (4.21); CO 4.27 Tg, 4.54 Tg (1.06); NMVOC 0.326 Tg, 0.977 Tg (3.00); NH_3 0.231 Tg, 0.365 Tg (1.58); CO_2 0.178 Pg, 1.30 Pg (7.31); PM_{10} 0.922 Tg, 0.142 Tg (0.154); $PM_{2.5}$ 0.489 Tg, 0.113 Tg (0.232); BC 0.0625 Tg, 0.0196 Tg (0.314); and OC 0.141 Tg, 0.0161 Tg (0.114). T~~rends ~~in air pollutants emissions~~ from each sector and fuel type during 1950-2015 in Japan were shown in Figs. S5 and S6. Compared to the rest of Asia, emissions of all species in Japan except CO_2 were reduced significantly after reaching peak values. In addition, peak years were mostly 40 years ago (about 1960 for PM_{10}, $PM_{2.5}$, and OC, 1970 for SO_2 and CO, 1980 for NO_x and NH_3, 1990 for BC, and 2000 for NMVOC).

Figure 5, similar to Fig. 3, shows trends of actual emissions (solid colored areas) and reduced emissions by control measures (hatched areas) from each sector for SO_2, NO_x, and BC during 1950-2015. Total CO_2 emissions were also plotted to each panel of Fig. 5. CO_2 emissions increased rapidly in the 1960s and have generally continued to increase, but growth rates are much smaller than those in the 1960s reflecting trends of economic status of Japan.

SO_2 emissions, especially from power plants and industry sector increased significantly in the 1960s (reflecting the rapid economic growth) and caused severe air pollutions in Japan. In the 1950s, more than half the emissions were from coal combustion and then~~; moreover~~, contributions from heavy fuel oil increased rapidly in the 1960s (~~about~~ more than 50% around the peak year). In order to mitigate air pollution, first, regulation of sulfur contents, especially in heavy fuel oil, were strengthened. Then, desulfurization equipment was mainly introduced from the mid-1970s. As a result, about 68%, 84%, and 93% of the SO_2 emissions were reduced by regulatory measures in 1975, 1990, and 2015, respectively. Furthermore, although coal consumption in power plants increased in the 1990s, SO_2 emissions almost did not change due to these measures. For trends of SO_2 emissions assuming without emission controls and those of CO_2, there are clear differences in the 1970s and after the 1980s. The causes of the differences in the 1970s were decrease of heavy fuel oil consumption whose contribution rates on SO_2 were much higher than CO_2. On the contrary, causes of the differences in the 1980s were increasing consumption of gas and light fuel oil whose sulfur contents were small.

~~As a result, SO_2 emissions decreased keenly in the 1970s. Although coal consumption in power plants increased in 1990s, SO_2 emissions almost did not change due to these measures. In 2015, about 90% of the SO_2 emissions were reduced by regulatory measures.~~ NO_x emissions also increased rapidly from the 1960s mainly by steep increase of traffic volumes and fossil fuel combustion in power and large industry plants. The largest contribution to NO_x emissions during the peak periods was from road transport sector, that is greater than 50% of total emissions. Regulations for road vehicles became effective from the late 1970s but an increase in the number of vehicles partially cancelled the effects. For stationary sources, the number of introduced denitrification equipment increased largely in the 1990s. As a result, NO_x emissions peaked later; furthermore, reduction rates after the peak were smaller compared to that of SO_2. From 1975 to 2015, emissions assuming without emission mitigations would be increased by about 2.0 times for power plants and 2.4 times for road transport. In 2015, by emission abatement equipment for power plants and control measures for road vehicles, the emissions were reduced by 77% and 90%, respectively. As a result, the reduction rate of total NO_x emissions in 2015 was 78%, but it was smaller than SO_2 as described above.

For BC, contributing sectors changed during 1950-2015. In the 1950s, most emissions were from industries and residential sectors and their amounts were almost comparable. After the 1960s, both types of emissions declined, but ~~their~~ reasons for declines were different. In the 1950s, coal and biofuels, which have large BC emission factors were mainly used in residential sectors. However, these fuels were substituted for cleaner ones, such as natural gas and liquefied petroleum gas which reduced BC emissions significantly. Emissions in industrial sectors decreased gradually after the 1960s due to the introduction of abatement equipment for PM. Instead, emissions from road transport sector from diesel vehicles increased from the late 1960s to around 1990. Then, regulations for road vehicles were strengthened and BC emissions were reduced largely from peak values. Before 1986, emission controls for BC were only considered for stationary sources. In 1985, by effects of abatement equipment to power and industrial plants, emissions were reduced by about 58% from those assuming no emission controls. Then, by introducing regulations for diesel vehicles, the reduction rates became about 91% in 2015.

For CO~~, NMVOC,~~ and OC, most emissions in 1950s were from biofuel combustion in the residential sector. CO and NMVOC emissions in road transport increased largely in the 1960s and then decreased gradually, similar to the case of NO_x. Recently, a majority of NMVOC emissions were from evaporative sources, such as paint and solvent use. These started to increase from the 1980s and then decreased after 2000. Emissions of CO and OC from the industrial sector showed a similar increase before 1970, whereas OC emissions started to decrease due to control equipment for PM species and CO emissions were almost stable after 1970. The m~~M~~ajority of NH_3 emissions in Japan ~~were~~ was from agricultural activities, especially manure management; however, contributions from latrines were also large in the past years. Overall, NH_3 emissions increased from 1950 to the 1970s but, showed slightly decreasing trends after the 1990s. PM_{10} and $PM_{2.5}$ emission trends were almost the same. The m~~M~~ajority of ~~the~~ emissions ~~were~~ was from the industrial sector, which grew during the 1950s but decreased largely in the 1970s due to the effects of abatement equipment for PM. Contributions from the residential sector were relatively large from the 1950s to the 1960s. Furthermore, contributions from road transport increased from the 1970s and started to decrease after 1990, similar to BC. ~~For CO_2, emission increased rapidly in 1960s, similar to that of SO_2 and NO_x. After that, CO_2 emissions have generally continued to increase, but growth rates are much smaller than those in the 1960s, thereby reflecting the economic status of Japan.~~

3.1.5 Other regions

Similar to India, air pollutant emissions in ~~Southeast Asia~~SEA and ~~South Asia other than India (~~OSA~~)~~ tended to increase during these six decades. ~~Emissions in Southeast Asia averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are, as follows: SO_2 0.161 Tg, 4.52 Tg (28.1); NO_x 0.215 Tg, 6.10 Tg (28.3); CO 10.8 Tg, 43.2 Tg (4.00); NMVOC 2.36 Tg, 13.3 Tg (5.61); NH_3 0.784 Tg, 4.09 Tg (5.22); CO_2 0.161 Pg, 1.56 Pg (9.68); PM_{10} 0.680 Tg, 3.07 Tg (4.51); $PM_{2.5}$ 0.636 Tg, 2.28 Tg (3.59); BC 0.119 Tg, 0.422 Tg (3.55); and OC 0.486 Tg, 1.28 Tg (2.63); corresponding data in OSA are, as follows: SO_2 0.0602 Tg, 1.41 Tg (23.4); NO_x 0.0713 Tg, 1.26 Tg (17.7); CO 3.52 Tg, 14.5 Tg (4.11); NMVOC 0.783 Tg, 3.66 Tg (4.67); NH_3 0.907 Tg, 3.13 Tg (3.44); CO_2 0.0566 Pg, 0.435 Pg (7.69); PM_{10} 0.256 Tg, 1.69 Tg (6.62); $PM_{2.5}$ 0.240 Tg, 1.14 Tg (4.73); BC 0.0451 Tg, 0.222 Tg (4.93); and OC 0.180 Tg, 0.661 Tg (3.67).~~ Figures S7 and S8 (~~S9~~ S11 and ~~S10~~S12) provide trends for all the air pollutant emissions in ~~Southeast Asia~~SEA (in OSA) for each sector and fuel type, respectively, from 1950 to 2015.

Figures 6 and 7 show emission trends of SO_2, NO_x, and BC for each sector category and contribution rates of each country from 1950-2015 in ~~Southeast Asia~~SEA and OSA, respectively. Total CO_2 emissions were also plotted to upper panels of Figs. 6 and 7.

Contributing sources and their relative ratios in SO_2, NO_x and BC emissions are generally close between these regions. For both the regions, major sources of SO_2 emissions are power plants and industry sector. For fuel types, contributions from heavy fuel oil ~~was~~ were large in the case of SO_2 emissions ~~from~~ in OSA and ~~was~~ were almost comparable to those of coal ~~from~~ in ~~Southeast Asia~~SEA ~~in~~ during the 1990s. After 2010, emissions from coal-fired power plants in ~~Southeast Asia~~SEA increased rapidly which were doubled during 2010-2015. On the other hand, in OSA, heavy fuel consumption in power plants increased by 1.8 times from 2005 to 2015 which mainly caused the large increase of SO_2 emission.~~.~~ For NO_x, majority of the emissions were from road transport, mainly diesel vehicles. This controlled the recent trends in both regions. Contributions from gasoline vehicles were small in OSA, but relatively large in SEA (about 16% in 2015). On the other hand, NO_x emissions from natural gas vehicles increased from the 2000s in OSA and contribution rates in road transport sector were more than 15% after the late 2000s. Recently, similar to SO_2, NO_x emissions from power plants have been increasing by coal and heavy fuel oil combustion in SEA and OSA, respectively. From 2010 to 2015, increases of emissions were mainly caused by power plants in both regions (about 67% for SEA and 82% for OSA). ~~Southeast Asia. Recently, NO_x emissions from coal-fired power plants have been increasing, especially in Southeast Asia.~~ Although trends are almost stable, emissions from biofuel combustion in the residential sector are relatively large in OSA. BC emissions are mostly from biofuel combustion in the residential sector, especially in OSA. and increased constantly during the period of REASv3. After the late 2000s, BC emissions from road transport show decreasing trends due to effect of emission regulations especially in SEA. Relations between trends of SO_2, NO_x, BC, and CO_2 emissions were similar to the case of India that trends of CO_2 were close to those of BC before the 1980s and then those of SO_2 and NO_x after the 1990s. ~~Recent increasing trends are caused by emissions from industry and road transport sectors.~~ In the case of country-wise emissions, currently, the largest contributing countries are Indonesia and Pakistan in ~~Southeast Asia~~SEA and OSA, respectively. In 2015, the second and third highest contributing countries in ~~Southeast Asia~~SEA were Philippines and Vietnam for SO_2, Thailand and Philippines for NO_x, and Vietnam and Thailand for BC. Relative ratios of SO_2 emissions in Thailand were large in the early 1990s but decreased significantly due to the introduction of FGD in large coal-fired power plants. For OSA, the second highest contributing country is Bangladesh; Sri Lanka is ranked third for SO_2 and NO_x and Nepal for BC.

E~~For East Asia other than China and Japan (OEA), all the air pollutant emissions averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are, as follows: SO_2 0.0589 Tg, 0.753 Tg (12.8); NO_x 0.0339 Tg, 2.00 Tg (58.9); CO 0.349 Tg, 6.41 Tg (18.3); NMVOC 0.0828 Tg, 1.88 Tg (22.7); NH_3 0.169 Tg, 0.467 Tg (2.76); CO_2 0.0123 Pg, 0.981 Pg (79.7); PM_{10} 0.0412 Tg, 0.375 Tg (9.11); $PM_{2.5}$ 0.0244 Tg, 0.263 Tg (10.8); BC 0.00529 Tg, 0.0604 Tg (11.4); and OC 0.0109 Tg, 0.0592 Tg (5.46). In Figs. S11 and S12, e~~mission trends in OEA from each sector during 1950-2015 were presented for all the air pollutants in Figs. S9 and S10. ~~.~~Emission trends in ~~the~~ Republic of Korea and Taiwan were similar to those of Japan. SO_2 emissions increased rapidly in the 1970s and reduced largely from their peak values due to the introduction of low sulfur fuels and FGD. NO_x emissions started to increase steeply from the 1980s due to ~~the~~ emissions from road ~~transport sector~~vehicles, in addition to those from power and industry plants. Then, NO_x emissions decrease after 2000 due to regulations related to road vehicles and the introduction of control equipment to power plants. However, their rate of decrease was lower than that of SO_2. BC emission trends were similar to those of NO_x until around the year 2000, but the ratio of decrease after 2000 is much larger than that of NO_x. The differences of reduction rates of emissions between NO_x and BC were caused by effects of emission controls in road transport sector. These features and drivers of trends were generally similar to the case of Japan. ~~These features are mainly determined by emissions from road vehicles.~~ For Democratic People's Republic of Korea, emissions of SO_2, NO_x, CO_2, and PM species decreased and those of CO, NMVOC, and NH_3 were almost stable recently. The recent decreasing trends were mainly caused by coal consumption amounts in industry sector. For Mongolia, emissions of all the air pollutants, except PM species, show increasing trends recently. The increasing trends were mainly caused by coal-fired power plants for SO_2 and CO_2, road transport for NO_x, CO, NMVOC, and BC, and domestic sector for OC. For PM_{10} and $PM_{2.5}$, due to effects of abatement equipment in power plants, emissions were almost stabilized after 2000. ~~BC and OC emissions also increased after the 2000s but decreased after 2013. Emissions of PM_{10} and $PM_{2.5}$ almost stabilized after 2000.~~ Note that information of these two countries are limited and therefore uncertainties are large.

3.2 Spatial distribution and monthly variation

Figure 8 presents the emission map of SO_2, NO_x, CO, NMVOC, NH_3, $PM_{2.5}$, BC, and OC in 1965 and 2015 at 0.25° × 0.25° resolution. Emission maps of CO_2 and PM_{10} are presented in Fig. S13. In 1965, high emission grids appeared in industrial areas of Japan, especially for NO_x, SO_2, and CO_2. On the other hand, high emission grids ~~are~~ were seen in wide areas in China and India, ~~in addition to Japan~~ for CO and PM species, especially OC. This is because emissions of these species ~~are~~ were mainly from the residential sector and small industrial plants. In 2015, high emission areas for all species clearly appeared in China and India, especially in the northeastern area, around the Sichuan province, and Pearl River Delta for China and Indo-Gangetic Plain, around Gujarat, and southern area for India. High emission areas of SO_2 and PM species in Japan disappeared or shrinked in 2015 compared to 1965, but still remained in the ~~case of~~ NO_x, CO, NMVOC, and CO_2 maps. In ~~Southeast Asia~~SEA, high emission areas ~~are~~ were seen in the Java island of Indonesia and around large cities, such as Bangkok (Thailand) and Hanoi (Vietnam). NH_3 and OC emissions, whose major sources ~~are~~ were agriculture and residential sector, respectively ~~are~~ were found in relatively large areas of China, India, and SEA~~Southeast Asia~~.

As described in Section 2.6~~5~~, seasonality of emissions is taken into considered for sectors where proxy data for monthly profiles were~~are~~ available or could~~can~~ be estimated. Monthly variations of total emissions of SO_2, NO_x, BC, and NH_3 are shown for China, India, Japan, ~~Southeast Asia~~SEA, OEA, and OSA for the year 2015 in Fig. 9. For SO_2 and NO_x, monthly variations were generally small. In China, emissions were slightly larger in the second half of the year. Monthly factors of SO_2 emissions in OSA were high from December to May and low during July and September due to the timings of brick production. For BC, emissions in winter season were relatively large, especially in China and OEA. This seasonality ~~is~~ was mainly determined by fuel consumption in residential sector for the purposes of heating. Therefore, monthly variations of BC emissions ~~are~~ were smaller in ~~Southeast Asia~~SEA. For NH_3, seasonality of emissions ~~is~~ was controlled by the seasonality of emissions from fertilizer application and manure management. In China, Japan, and OEA, peaks of emissions appeared during summertime. Monthly variations of emissions in the whole of ~~Southeast Asia~~SEA ~~are~~ were small, but seasonality ~~is~~ was different from each country. Finally, it must be noted that monthly variations of emissions in each grid ~~are~~ were different to each other because they ~~are~~ were determined by monthly profiles of major emission sources in each grid cell.

3.3 Comparison with other inventories

In this section, estimated emissions of REASv3 were compared with other global, regional, and national bottom-up inventories and several top-down estimates. Figures 10 and 11 compare the results of REASv3 with other studies for SO_2, NO_x, and BC emissions in China and India, respectively. For other species, results based on comparison with China are presented in Fig. S14 and those with India are shown in Fig. S15. Furthermore, Figures S16-S19 provide the comparisons of emissions from Japan, SEA, OEA, and OSA, respectively. In Figs. 10, 11, and S14-S19, error bars were plotted in 2015, 1985, and 1955 of emissions in REASv3. These error bars were based on uncertainties estimated in this study for corresponding emissions. See Sect. 3.4 for details about the uncertainties of emissions in REASv3. Note that as described in Sect. 2.1, emissions from domestic and fishing ships are not included in REASv3. Therefore, corresponding data need to be excluded from values of other inventories in the comparisons. This procedure was done for REAS series, EDGARv4.3.2, CEDS, and several research works. For other inventories where emissions from domestic ship were not available independently, total emissions were plotted in the figures. It was confirmed that other sources out of scope of REASv3 such as open biomass burning were not included in the other inventories.

3.3.1 China

For long historical trends of SO_2 emissions in China, most studies generally agreed with the trends of REASv3 although values of REASv3 during 1995 and 2005 were slightly larger than other inventories. Emissions increased almost monotonically until around 1995 and became stable during the late 1990s. Then, emission increased rapidly from the early 2000s and started to decrease from the late 2000s. However, the decreasing rates were different especially after 2010. Recent rapid decreasing tendency in REASv3 was similar to that of Zheng et al. (2018), but decreasing rates of other studies such as Xia et al. (2016) and Sun et al. (2018) were smaller than REASv3. Values of REASv3 were slightly larger than REASv2.1 during 2000-2005, but the discrepancies were reduced due to a larger decreasing rate of REASv3. For top-down estimates (Qu et al., 2019 [based on retrieval products by National Aeronautics and Space Administration (NASA) standard (SP) and Belgian Institute for Space Aeronomy (BIRA)]; Miyazaki et al., 2020), emission amounts were smaller than most bottom-up inventories, but all top-down results showed large decreasing trends after the late 2000s.

Variability of NO_x emissions among estimations plotted in Fig. 10 was smaller than that of SO_2. NO_x emissions in most results increased largely in the 2000s and then decreased or stabilized. Growth rates of Sun et al. (2018) were smaller than others after 2005, but showed similar decreasing trends after 2010. Values of CEDS were slightly larger than other studies. Similar to SO_2, values of top-down estimates (Ding et al., 2017 [based on OMI and GOME-2]; Itahashi et al., 2019; Miyazaki et al., 2020) were generally smaller than those of bottom-up results. But, top-down emissions showed similar tendencies that emission increased until the early 2010s and turned to decrease. Trends of Itahashi et al. (2019) where emissions in 2008 of REASv2.1 were used as a priori data were close to those of REASv3.

Compared to SO_2 and NO_x, relatively large discrepancies were observed in BC emissions among plotted results in Fig. 10. Emissions of REASv3 increased until 1995, slightly decreased during the late 1990s, increased from the early 2000s and then, turned to decrease from the early 2010s. The decreasing rate in the late 1990s of Wang et al. (2012) was much larger than that of REASv3. On the other hand, emissions of Klimont et al. (2017) increased from 1995 to 2000. The majority of results showed increasing trends during the early 2000s, but the following trends were different. Emissions of CEDS increased constantly after 2005, but those of Wang et al. (2012) decreased after 2005 and then started to increase slightly after 2010. BC emissions of both REASv3 and Zheng et al. (2018) decreased from the early 2010s, but the ratio of decrease was larger in Zheng et al. (2018). Values of BC emissions of REASv3 were larger than those of REASv2.1, especially in the early 2000s, but the difference in 2008 was small. For trends and emission amounts of PM_{10} and $PM_{2.5}$, tendencies of relationships among each result were similar. The majority of results showed clear decreasing trends after 2005 except for REASv2.1, EDGARv4.3.1 and Klimont et al. (2017). For OC, most results decreased from 1995 to 2000 and then increase from the early 2000s. After 2005, trends of OC emissions were different among studies.

CO emissions trends were relatively similar among most studies. Increasing rates after the early 2000s are close except for EDGARv4.3.2, but emission amounts of REASv3 were smaller than other studies before 2010. After 2010, the majority of results showed decreasing trends which agreed with top-down estimates (Jiang et al., 2017 [A: MOPITT Column, B:

MOPITT Profile, and C: MOPITT Lower Profile]; Zheng et al., 2019b; Miyazaki et al, 2020). However, before the late 2000s, the trends of CO emissions were much different between bottom-up inventories and top-down results. For NMVOC, most studies showed significant increasing trends after the early 2000s. Compared to bottom-up inventories, top-down estimates of Stavrakou et al. (2017) were almost stable between 2007 and 2012, but increased rapidly after that. Values of REASv3 were generally smaller than others before 2010. Differences among studies of NH_3 emissions were large not only in emission amounts, but also in temporal variations. REAS inventories, CEDS, and EDGARv4.3.2 generally showed increasing trends. On the other hand, trends of MEICv1.2 and Zheng et al. (2018) were almost stable after 2000 and the results of Kang et al. (2016) showed decreasing trends after the mid-2000s. Emissions of REASv3 were also almost stable after 2010.

3.3.2 India

For SO_2, emissions of most bottom-up inventories showed monotonically increasing trends. However, after the 1990s, two different emission pathways were shown among studies. The growth rates of REASv3 were close to those of Klimont et al. (2013), CEDS (scaled to REASv2.1 for India; Hoesly et al., 2018), Streets et al. (2000), and REASv2.1. On the other hand, the increasing rates of national studies by Garg et al. (2006), Sadavarte and Venkataraman (2014) and Pandey et al. (2014) were smaller than those of REASv3. In 2005, top-down estimates of Qu et al. (2019) were close to results of Sadavarte and Venkataraman (2014) and Pandey et al. (2014). Another top-down emissions of Miyazaki et al. (2020) were smaller than other inventories. Both bottom-up and top-down emissions after 2005 show increasing trends, but growth rates of bottom-up inventories were higher than those of top-down estimates.

NO_x emissions of REASv3 also increased monotonically during 1950-2015 and the majority of other bottom-up inventories generally agreed with the trends including national studies of Sahu et al. (2012). However, similar to SO_2, growth rates of Venkataraman (2014) and Pandey et al. (2014) were smaller than REASv3 although emission amounts in 2000 and 2005 were almost comparable each other. For the increasing rates, those of top-down estimates of Itahashi et al (2019) using REASv2.1 as a priori emissions were close to those of REASv3. On the other hand, growth rates of another top-down results of Qu et al. (2019) were similar with those of Sadavarte and Venkataraman (2014) and Pandey et al. (2014). Emission amounts of the top-down estimates were much higher than REASv3.

For BC, as in the case of China, discrepancies among studies plotted in Fig. 11 were large. These tendencies were also found in the comparisons of PM_{10}, $PM_{2.5}$, and OC emissions provided in Fig. S15. Generally, the majority of bottom-up emission inventories of PM species showed slightly continuous increasing trends and growth rates were smaller than those of SO_2 and NO_x. On the contrary to the case of SO_2 and NO_x, emissions of BC and $PM_{2.5}$ of REASv3 were slightly smaller than those of Sadavarte and Venkataraman (2014) and Pandey et al. (2014), but their growth rates were almost comparable.

Amounts and trends of CO emissions compared in Fig. S15 generally agreed well except for REASv1.1 which were much higher than others. Emission increased almost constantly until around 2005 ant then growth rates increased slightly. Values of REASv3 were much smaller than top-down results of Jiang et al., 2017 [A: MOPITT Column, B: MOPITT Profile, and C:

MOPITT Lower Profile] and Miyazaki et al. (2020). However, recent growth rates of REASv3 were close to those of top-down estimates except for Jiang et al. (2017) [C]. For NMVOC, plotted results were generally comparable except for REASv2.1 and CEDS and indicated increasing trends of emissions. Similar to the case of SO_2 and NO_x, growth rates of REASv3 were smaller than those of Sadavarte and Venkataraman (2014) and Pandey et al. (2014). For NH_3, a comparison of the emissions in Fig. S15 show similar increasing trends. Differences in emission amounts are also relatively small, except for EDGARv4.3.2.

3.3.3 Other regions

Comparisons of emissions in Japan between REASv3 and other studies were provided in Fig. S16. For tends of SO_2 emissions in Japan, the majority of studies agreed with results of REASv3 that rapid increases in the 1960s, keen decreases in the 1970s, and gradually decreasing trends except for EDGARv4.3.2 and Streets et al. (2000), whose values were lager and smaller, respectively. For NO_x, emissions amounts of REASv3 were larger than those of most studies especially before 2000, except for CEDS (scaled to preliminary historical data of REAS for Japan; Hoesly et al., 2018), Kannari et al. (2007), Zhang et al. (2009) based on Kannari et al (2007) and Fukui et al. (2014). For PM species, the majority of results in Fig. S16 agreed with decreasing trends of REASv3 after 1990. On the other hand, emissions of BC and OC of CEDS increased almost monotonically until their peak around 1990. These tendencies were much different from REASv3. For CO, emission amounts of REASv3 were larger than other results of especially REASv1, EDGARv4.3.2, and CEDS. However, after 2000, emissions and their decreasing trends of other studies were generally comparable to those of REASv3. For NMVOC, results of REASv3 after 2000 generally agreed well with other studies which showed large decreasing trends except for EDGARv4.3.2 and Zhang et al. (2009) based on Kannari et al. (2007). Trends of NH_3 emissions shown in Fig. S16 were similar except for EDGARv4.3.2 before the mid-1990s which showed larger growth rates. Emission amounts of REASv3 were smaller than national inventories by Kannari et al. (2001) and Fukui et al. (2014).

For SEA (see Fig. S17), increasing trends and amounts of SO_2 emissions of REASv3 agreed with other results except for CEDS in the 1990s, Zhang et al. (2009), and Klimont et al. (2013). In CEDS, emissions decreased keenly during the late 1990s. A similar feature was also seen in REASv3 but its rate of decrease was much smaller. For NO_x, all results plotted in Fig. S17 indicated monotonically increasing trends of emissions and agreed well until the early 2000s. After that, growth rates of REASv3 became larger than EDGARv4.3.2 and smaller than CEDS (scaled to REASv2.1 for SEA; Hoesly et al., 2018). For BC, REAS series and CEDS showed similar growth rates until around 2005. On the other hand, increasing rates of Klimont et al. (2017) and EDGARv4.3.2 after 1990 were much smaller and close to those of REASv3 after 2005.

Most results of SO_2 emissions in OEA in Fig. S18 show increasing and decreasing trends from the late 1960s and the early 1990s, respectively, although amounts in CEDS from 1970 and 2000 were much smaller. For NO_x, all results agreed well until the late 1980s and REASv3, REASv1.1 and EDGARv4.3.2 showed similar increasing trends until around 2000. Emissions of CEDS became almost stable after the late 1980s and started to decrease after 2005. The decreasing rates of REASv3 and CEDS are close after 2005. On the other hand, emissions of EDGARv4.3.2 were not changed largely after around 2000. The similar tendencies were shown in the case of SO_2. BC emissions of REASv3 and CEDS showed similar trends until 2000. Then, emissions of REASv3 decreased almost monotonically, while those of CEDS were almost stable. Similarly, decreasing rates of EDGARv4.3.2 after 2000 were much smaller than those of REASv3.

For OSA, increasing trends and amounts of SO_2 and NO_x emissions were generally similar among studies plotted in Fig. S19. SO_2 emission of Streets et al. (2003a) and Zhang et al. (2009) and NO_x emissions of CEDS (scaled to REASv2.1; Hoesly et al., 2018) were higher than other results. For BC, discrepancies among studies were larger than those of SO_2 and NO_x, but similar small monotonically growth rates were shown in all results.

3.3.4 Relative rations of emissions from each country and region in Asia

Figure 12 compares trends of total emissions in Asia and relative ratios of emissions from China, India, Japan, SEA, OEA, and OSA among REASv3, CEDS, and EDGARv4.3.2 for SO_2, NO_x, and BC. Comparisons of other species are presented in Fig. S20. From 1950 to the early 2000s, total SO_2 emissions in Asia of all inventories showed similar results. For relative ratios, REASv3 and CEDS values were similar until the mid-2000s. Contributions from Japan were relatively large from 1950 until around 1970 and then, decreased keenly. This was also found in EDGARv4.3.2, but the rate of decrease was smaller than that of REASv3 and CEDS. Then, while emissions of REASv3 decreased largely after the mid-2000s, those of CDES and EDGARv4.3.2 continued to increase. These discrepancies were mainly due to different trends of emissions from China. Actually, after the mid-2000s, relative ratios of SO_2 emissions in China were stable in CEDS and EDGARv4.3.2, but those in REASv3 decreased significantly. Recently, increasing trends of relative ratios of SO_2 emissions in India are a common feature in REASv3, EDGARv4.3.2, and CEDS.

For NO_x, Asia total emissions of REASv3 and EDGARv4.3.2 were close. Although emissions of CEDS were larger than REASv3 and EDGARv4.3.2, trends were similar until early 2010. The different trends after 2010 between REASv3 and CEDS were caused by those of emissions in China. For the contributing rates, REASv3 and CEDS generally showed similar temporal variations, although relative ratios of OSA were larger in CEDS. Contribution rates of Japan were large around 1970 and then gradually decreased. Instead, those from China increased almost monotonically until 2010. Similar to the case of SO_2, relative ratios of China decreased recently in REASv3, but they were almost stable in CEDS and EDGARv4.3.2. In addition, contribution rates from India showed gradual increasing trends in all the results

For total Asia emissions of BC, trends of REASv3 and CEDS were similar until the late 1990s, but after 2000 while growth rats of CEDS became larger, emissions of REAS were not changed largely and turned to decrease after 2010. Emission amounts and growth rates of EDGARv4.3.2 were smaller than others until the mid-1990s, but after that the trends were similar to those of REASv3. Compared to SO_2 and NO_x, temporal variations of relative ratios of BC emissions from each country and regions were small in all the results. In REASv3, contribution rates of Japan were large before 1970 and then decreased afterwards. On the other hand, in CEDS, contribution rates of Japan after 1970 were larger than those before 1970. After 2000, relative ratios of China in REASv3 were almost stable and showed a marginal decrease after 2011. In CEDS and

EDGARv4.3.2, contribution rates of China increased during the first half of 2000s and then became almost stable. Similar tendencies were seen in OC. Compared to BC, relative ratios of China started to decrease earlier only in REASv3.

For Asia total emissions of CO, although amounts of CEDS were larger than others, trends of all results were close until the early 2000s. After that, REASv3 showed large increases until 2010 and then started to decreased slightly. These tendencies were mainly controlled by emissions in China. Trends of the relative ratios were similar to those of BC. But contribution rates of China in REASv3 increased gradually until the mid-2000s and then decreased, while those in CEDS and EDGARv4.3.2 were almost stable. For NMVOC, total emissions in Asia of REASv3 were smaller than others, but large increases of emissions were found from the early 1990s. The corresponding feature was shown in contribution rates. Relative ratios of emissions from China in REASv3 increased largely during the 1990s and 2000s. Similar increasing trends were seen in EDGARv4.3.2 but growth rates of REASv3 were much larger. On the other hand, both temporal variations and values of contribution rates of China were relatively small in CEDS.

For NH_3, trends of total emissions in Asia of REASv3 were close to EDGARv4.3.2 and slightly larger than CEDS until around 2000. After that, growth rates of REASv3 were close to CEDS and those of EDGARv4.3.2 became larger. As a result, amounts of total Asia emissions of all inventories became almost the same after 2010. For relative rations of regions, contribution rates of China in REASv3 increased gradually until the mid-2000s and then became almost stable, whereas those in CEDS and EDGARv4.3.2 show slightly decreasing and increasing trends, respectively. In 2015, relative ratios of NH_3 emissions from China in REASv3 were between those of EDGARv4.3.2 and CEDS. Compared to EDGARv4.3.2 and CEDS, contribution rates of NH_3 emissions from SEA region were relatively small in REASv3.~~In this section, estimated emissions of REASv~~3~~.1 were compared with those of other global, regional, and national inventories. As mentioned in Section 2.1, values of REASv3.1 include emissions from domestic and fishing ships, which are roughly estimated using fuel consumption and default emission factors. This procedure is done because most other inventories include ship emissions and it is difficult to remove their contribution for comparison with REASv3.1. Note that domestic and fishing ships are not officially included in target sources of REASv3.1, as described in Sect. 2.1. Figures 10 and 11 compare the results of REASv3.1 with other studies for SO_2, NO_x, and BC emissions in China and India, respectively. For other species, results based on comparison with China are presented in Fig. S14 and those with India are shown in Fig. S15. Furthermore, Figures S16-S19 provide the comparisons of emissions from Japan, Southeast Asia, East Asia other than China and Japan, and South Asia other than India, respectively.~~

~~For long historical trends of SO_2 emissions in China, values of REASv3.1 were similar to those of EDGARv4.3.2 and CEDS, until 1995. After this, EDGARv4.3.2 and CEDS showed a marginal decline, although the values of REASv3.1 were almost constant. SO_2 emissions in most results increased rapidly from the early 2000s and a majority of them decreased from the late 2000s, although the decreasing rates are different. Recent rapid decreasing tendency in REASv3.1 is similar to that of Zheng et al. (2018). Values of REASv3.1 were slightly larger than those of REASv2.1 during 2000-2005, but then reduced due to a larger decreasing rate of REASv3.1. Compared to SO_2, variability of NO_x emissions among estimations plotted in Fig. 10 is small, although values of CEDS are slightly larger than others. NO_x emissions in all the results increased largely in~~

~~the 2000s and then decreased or stabilized. For BC, there were large discrepancies observed among plotted results in Fig. 10. Trends observed in REASv3.1 and CEDS were similar until early 2000s, but then CEDS showed a large and constant increase. Differences in REASv3.1 and REASv2.1 are small. BC emissions of both REASv3.1 and Zheng et al. (2018) decreased from 2012, but the ratio of decrease was much larger in Zheng et al. (2018). Values of BC emissions of REASv3.1 were larger than those of REASv2.1, especially in the early 2000s, but the difference in 2008 was small. For trends and emission amounts of PM_{10} and $PM_{2.5}$, tendencies of relationships among each result were similar. Growth rates of REASv3.1 were larger than EDGARv4.3.1 until mid-1990s, following which, similar temporal variations were shown in both results until the early 2000s. After the early 2000s, REASv3.1, MEICv1.2, Zhao et al. (2013), and Zheng et al. (2018) show decreasing trends, although REASv2.1, EDGARv4.3.2, and Klimont et al. (2017) continued to increase. For OC, most results decreased from 1995 to 2000 and then increase from the early 2000s. Then, peaks of REASv3.1, REASv2.1, and MEICv1.2 appeared around 2005, whereas EDGARv4.3.2 and CEDS show continuous increasing trends. CO emissions trends were relatively similar among most studies, although values of REASv3.1 were smaller (larger) before the mid-1990s (after middle of 2000s), as compared to other inventories. Emissions and their growth rates of EDGARv4.3.2 were smaller than other results. For NMVOC, emission amounts of REASv3.1 were smaller, but growth rates were larger than CEDS and EDGARv4.3.2 until the early 2000s. After this period, most studies showed significant increasing trends, but emission amounts of REASv3.1 in 2015 were larger than MEICv1.2, Zheng et al. (2018), and Li et al. (2019). Differences among studies in terms of NH_3 emissions were large not only in emission amounts, but also in temporal variations. REAS inventories, CEDS, and EDGARv4.3.2 generally showed increasing trends. On the other hand, trends of MEICv1.2 and Zheng et al. (2018) were almost stable after 2000 and the results of Kang et al. (2016) showed decreasing trends after mid-2000s. Emissions of REASv3.1 were also almost stable after 2010.~~

~~For SO_2 emissions in India, REASv3.1, EDGARv4.3.2, and Smith et al. (2011) showed similar long historical trends, until 2000. Then, values of REASv3.1 became slightly larger than these, but were close to those of Klimont et al. (2013). CEDS, Streets et al. (2000), and REASv2.1 values were similar but were larger than REASv3.1. On the other hand, emissions of REASv3.1 were larger than the recent national studies by Sadavarte and Venkataraman (2014) and Pandey et al. (2014). For long historical trends of NO_x, emissions of REASv3.1 are much smaller than CEDS and REASv2.1 as well as smaller than those of EDGARv4.3.2 and REASv1.1 until 2000. However, values of REASv3.1 are close to Garg et al. (2001) and those of EDGARv4.3.2 after 2000. Similar to SO_2, NO_x emissions of REASv3.1 are smaller than Sadavarte and Venkataraman (2014) and Pandey et al. (2014). For BC, trends of REASv3.1 and CEDS agree relatively well for all periods. However, as in the case of China, discrepancies among studies plotted in Fig. S15 are large. Trends of REASv1.1 and REASv3.1 showed similar trends but values of REASv3.1 were smaller. On the other hand, emissions of REASv3.1 were larger than EDGARv4.3.2 and REASv2.1 but growth rates of EDGARv4.3.2 after 2008 and REASv2.1 were larger than those of REASv3.1. OC emissions of REASv3.1 were smaller than CEDS and Lu et al. (2011) but growth rates were similar. On the contrary, growth rates of REASv3.1 were larger than EDGARv4.3.2, Pandey and Venkataraman (2014), Sadavarte and Venkataraman (2014), and Klimont et al. (2017). For PM_{10} and $PM_{2.5}$, temporal variations of REASv3.1 and EDGARv4.3.2~~

~~were similar until the late 2000s. Growth rates of EDGARv4.3.2 after late 2000s and REASv2.1 were larger than REASv3.1. $PM_{2.5}$ emissions of Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014) were larger than REASv3.1 but growth rates were alike. On the other hand, Klimont et al. (2017) showed different temporal variations. Emissions of PM_{10} and $PM_{2.5}$ decreased from 2000 to 2005 and differences in the values between 2005 and 2010 were marginal. Amounts and trends of CO emissions compared in Fig. S15 generally agree well except for REASv1.1. Values of REASv3.1 are between those of CEDS and EDGARv4.3.2. For NMVOC, emission amounts of REASv3.1 and EDGARv4.3.2 were almost the same in the early 1990s but growth rates after that were higher for REASv3.1 than for EDGARv4.3.2. Values of REASv3.1 were close to Streets et al. (2003a), Zhang et al. (2009), and Venkataraman et al. (2018). Moreover, values of EDGARv4.3.2 agree with Pandey and Venkataraman (2014), Sadavarte and Venkataraman (2014) and Sharma et al. (2015). Emissions of CEDS and REASv2.1 were more than that of other studies. For NH_3, a comparison of the emissions in Fig. S15 show similar trends. Differences in emission amounts are also relatively small, except for EDGARv4.3.2.~~

~~Results of SO_2 emissions in Japan compared in Fig. S16 generally agree well except for EDGARv4.3.2 and Streets et al. (2000), whose values were lager and smaller, respectively. For NO_x, emissions, amounts of REASv3.1 were larger than those of most studies, except for CEDS; although temporal variations were found to be similar. Emission amounts and trends of BC were generally alike among the results shown in Fig. S16, except for CEDS. Emissions of BC and OC of CEDS increased almost monotonically until their peak around 1990. These tendencies were much different from REASv3.1. In general, most studies show decreasing trends of emissions of PM species from 1990s. However, decreasing ratios of REASv3.1 before 1990s are larger than other studies. For CO emissions, significant increase was observed from the early 1960s in both REASv3.1 and CEDS, but emissions of REASv3.1 reached its peak in the early 1970s while those of CEDS continued to increase until 1980. Emissions of EDGARv4.3.2 decreased largely during the early 1970s. For NMVOC, results of REASv3.1 and CEDS agree well except from the late 1970s to early 1990s and after 2000s. Emission amounts of REASv3.1 and CEDS are smaller than EDGARv4.3.2 and Zhang et al. (2009). Trends of NH_3 emissions shown in Fig. S16 are similar except for EDGARv4.3.2 before the mid-1990s. Emission amounts of REASv3.1 are smaller than Kannari et al. (2001), REASv2.1, and CEDS.~~

~~For SO_2 emissions in Southeast Asia, values decreased keenly in CEDS from the late 1990s. A similar feature was seen in REASv3.1 but its rate of decrease was much smaller. After 2000, emissions of REASv3.1 showed increasing trends while those of Klimont et al. (2013) decreased almost monotonically. For NO_x, all the results shown in Fig. S17 agreed well until late 1990s, but showed different trends later. Emissions of REASv3.1 increased almost monotonically from the 1990s, but those of CEDS and EDGARv4.3.2 were stable until 2000 and then increased again. Growth rates of CEDS and EDGARv4.3.2 after 2000 were larger and smaller than REASv3.1, respectively. Trends of emissions in REASv3.1 and REASv2.1 were similar but amounts of REASv3.1 were larger. For BC, all the results in Fig. S17 increased gradually with similar increasing rates. However, emissions of EDGAR became almost stable after 2000 and those of REASv3.1 decreased during the late 2000s and then increased again.~~

~~Most results of SO_2 emissions in East Asia other than China and Japan show increasing and decreasing trends from late 1960s and early 1990s, respectively, although amounts in CEDS from 1970 and 2000 were much smaller. On the other hand, emission peaks in EDGAR were reached around 2000 and the amounts were much larger than those observed other studies after the late 1990s. For NO_x emissions, amounts and trends shown in Fig. S18 generally agreed until the late 1990s, but the results differed after the 2000s. Decreasing rates of REASv3.1 were similar to those of EDGARv4.3.2, but smaller than those of CEDS and REASv2.1. BC emissions of REASv3.1 and CEDS show similar increasing trends until the late 1980s. Then, emissions of REASv3.1 decreased almost monotonically, but those of CEDS were almost stable. Both emission amounts were larger than REASv1.1., REASv2.1, and EDGARv4.3.2. For South Asia other than India, trends and amounts of SO_2 emissions were similar. Emission values of Streets et al. (2003a) and Zhang et al. (2009) were larger than the results plotted in Fig. S18. For NO_x, values of REASv3.1, Kato and Akimoto (1992), Streets et al. (2003a) and Zhang et al. (2009) were almost the same and trends of REASv3.1 were similar to those of REASv1.1, REASv2.1, and EDGARv4.3.2. Emission amounts of CEDS were much higher than other results. For BC emissions, trends of REASv3.1 were generally similar with the results shown in Fig. S18. Emissions of REASv3.1 were close to those obtained by Streets et al. (2003a), Zhang et al. (2009), CEDS, and REASv2.1, but were larger than EDGARv4.3.2, and much smaller than Klimont et al. (2017).~~

~~Figure 12 compares trends of relative ratios of SO_2, NO_x, and BC emissions from each country and region among REASv3.1, CEDS, and EDGARv4.3.2. Comparisons of other species are presented in Fig. S19. For SO_2, REASv3.1 and CEDS values were similar until the mid-2000s. Contribution from Japan were relatively large from 1950 until around 1970 and then, decreased keenly. This was also found in EDGARv4.3.2, but the rate of decrease was smaller than that of REASv3.1 and CEDS. After the mid-2000s, relative ratios of SO_2 emissions in China were stable in CEDS and EDGARv4.3.2, but those in REASv3.1 decreased significantly. Recently, increasing trends of relative ratios of SO_2 emissions in India are a common feature in REASv3.1, EDGARv4.3.2, and CEDS. For NO_x, REASv3.1 and CEDS generally showed similar temporal variations of the contributing rates, although relative ratios of South Asia other than India were larger in CEDS. Relative ratios of Japan were large around 1970 and then gradually decreased. Instead, contribution rates from China increased almost monotonically until 2010. Similar to the case of SO_2, relative ratios of China decreased recently in REASv3.1, but they were almost stable in CEDS and EDGARv4.3.2. In all the results, contribution rates from India showed gradual increasing trends. Compared to SO_2 and NO_x, temporal variations of relative ratios of BC emissions from each country and regions were small for REASv3.1, CEDS, and EDGARv4.3.2. In REASv3.1, contribution rates of Japan were large before 1970 and then decreased afterwards. On the other hand, in CEDS, contribution rates of Japan after 1970 were larger than those before 1970. After 2000, contribution rates of China in REASv3.1 were almost stable and showed a marginal decrease after 2011. In CEDS and EDGARv4.3.2, contribution rates of China increased during the first half of 2000s and then became almost stable. Similar tendencies were seen in OC. Compared to BC, relative ratios of China started to decrease earlier only in REASv3.1. CO trends were similar to those of BC. But contribution rates of China in REASv3.1 increased gradually until the mid-2000s and then decreased, while those in CEDS and EDGARv4.3.2 were almost stable. For NMVOC, contribution rates of China in REASv3.1 increased largely after the early 2000s. Similar increasing trends were seen in EDGARv4.3.2 but growth rates~~

~~of REASv3.1 were much larger. On the other hand, both temporal variations and values of contribution rates of China were relatively small in CEDS. For NH_3, contribution rates of China in REASv3.1 increased gradually until the mid-2000s and then became almost stable, whereas contribution rates from China in CEDS and EDGARv4.3.2 show slightly decreasing and increasing trends, respectively. In 2015, contribution rates of NH_3 emissions from China in REASv3.1 were between those of EDGARv4.3.2 and CEDS. Compared to EDGARv4.3.2 and CEDS, contribution rates of NH_3 emissions from Southeast Asian region were relatively small in REASv3.1.~~

3.4 Uncertainty

In REASv3~~.1~~, uncertainties in emissions were estimated for each country and region in 1955, 1985, and 2015~~as well as each year~~ using basically the same methodology as that of REASv2.1 (Kurokawa et al., 2013). First, uncertainties in all ~~the~~ parameters used to calculate emissions, such as activity data, emission factors, removal efficiencies, and sulfur contents of fuels were estimated in the range of ~~5~~2-~~200~~150%. In estimation of the uncertainties except for activity data, following three causes need to be considered: uncertainties in the data themselves, those caused by selections of the data, and those in settings related to emission controls such as timing of introduction and penetration rates of abatement equipment. In this study, uncertainties in settings of emission controls were explicitly considered only for removal efficiencies. The uncertainties of removal efficiencies were assumed to be zero for emission sources where no emission controls were considered which means that uncertainties caused by neglecting emission controls were not considered. Furthermore, for emission sources where introduction rates of abatement equipment were small, uncertainties caused by settings of emission controls were assumed to be small. Then, uncertainties in emissions from power plants, industries, road transport, other transport, domestic and other sectors, as well as uncertainties in total emissions were calculated for all the species. The uncertainties of different sub-sectors and activities were combined in quadrature assuming they were independent. On the other hand, for uncertainties of national emissions in China, India, and Japan, those in their sub-regions were added linearly. Details of the methodology and settings of uncertainties of each component were described in Sect. S10 of the Supplement. ~~by combining the estimated uncertainties of used parameters.~~ Similar to REASv2.1, uncertainties of emissions that were not originally developed in REASv3~~.1~~ (NH_3 emissions from manure management and fertilizer application, and NMVOC evaporative emissions from Japan and ~~the~~ Republic of Korea) were not evaluated in this study.

Table ~~3~~ 4 summarizes the estimated uncertainties in total emissions of each species for China, India, Japan, ~~Southeast Asia~~SEA, ~~East Asia other than China and Japan~~OEA, and ~~South Asia other than India~~OSA in ~~2015~~1955, 1985, and ~~1955~~2015. Uncertainties in emissions from each sector were provided in the Supplement tables (Table S1 for SO_2, NO_x, CO, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC, in Table S2 for NMVOC, and in Table S3 for NH_3)~~in Table S2 for SO_2, NO_x, CO, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC, in Table S3 for NMVOC, and in Table S4 for NH_3~~. For most regions and years, uncertainties for SO_2, NO_x and CO_2 are smaller than other species. Major emission sources of these species are power plants and large industry sectors. Uncertainties of activity data of these species were assumed to be small because power plants and large industries are critically important for each country and related statistics are expected to be accurate. In addition, uncertainties of emission factors of combustion at high temperature in power plants and large industries are considered to be small. For SO_2 emissions in China, uncertainties in 2015 were estimated to be slightly larger than those in 1985 due to uncertainties for removal efficiencies which were not considered in 1985. For South and Southeast Asia, uncertainties of SO_2 emissions in 1985 were slightly smaller than those in 2015. This is because settings of sulfur contents in fuels were based on surveys conducted in 1990 (Kato and Akimoto, 1992) and thus, the uncertainties in 1985 were assumed to be smaller than those in 2015.~~For most regions and years, the smallest uncertainties are found for SO_2 emissions; uncertainties for NO_x and CO_2 are smaller than other species. Emissions of SO_2 are mainly determined by the sulfur contents of fuels, with contributions from power plants and industries sector being large. As compared to other sources, the accuracy of data of sulfur contents of fuels are assumed to be high because the information available on sulfur contents is relatively large, even for earlier years in the period of REASv3.1. Another reason for a relatively high accuracy of SO_2 emission data is that uncertainties of emission factors of combustion at high temperature, such as for large power and industry plants are considered to be small. This is also a major reason that uncertainties of NO_x and CO_2 emissions are lesser than others.~~

On the other hand, uncertainties of PM species are large compared to other species for most regions and years. For most countries in Asia, a majority of their emissions was from combustion at relatively low temperatures in small industries and residential sectors. Accuracies of activity data and emission factors for these sources are assumed to be low, especially for biofuel combustion. Therefore, uncertainties of OC emissions mainly from biofuel combustion in Asia are the largest for most regions and years. Uncertainties of PM_{10} are generally smaller than other PM species. This is because for PM_{10} emissions, contribution rates of power plants and industry sectors are generally larger than those of other PM species. For CO and NMVOC, in general, uncertainties of emission factors are assumed to be greater than SO_2, NO_x, and CO_2, but smaller than $PM_{2.5}$, BC and OC~~ and smaller than PM species~~. Therefore, uncertainties of total CO and NMVOC emissions are generally between those of other species~~middle of all species~~. For Southeast and South Asia, uncertainties of CO and NMVOC are comparable to PM_{10} as their relative contribution from biofuel combustion is large.~~Exceptions to these include uncertainties in CO emissions in India and Southeast Asia, which are comparable to PM species as their relative contribution from biofuel combustion is large.~~

Uncertainties in emissions from Japan are lesser than those of other countries and regions. This is mainly due to the accuracy of activity data. Accessibility to detailed information in Japan is relatively high compared to other countries in REASv3~~.1~~. In Japan, uncertainties of emission in 1985 were comparable to or slightly smaller than those in 2015. This is because relative ratios of emissions from road transport whose uncertainties were the smallest in Japan were reduced largely from 1985 to 2015. For China and India, accuracies of emissions are generally improved for most species compared to REASv2.1 using information from recently published literatures of emission inventory of these countries. However, the improvement is not significant due to the lack of country specific information. This situation is almost the same for other countries and regions. Although studies of national emission inventories in Asia are being published, as described in Section 1, information on technologies related to emissions and their introduction rates is not as easily available. Therefore, continuous efforts to update emission inventories by collecting information of each country and region are essential. For all countries, uncertainties of emissions in 1955 were much larger than those in 2015. This is because most activity data were not obtained directly from statistics, especially in the early half of the target period of REASv3.1. In this study, activity data, which were not available in statistics were extrapolated or assumed using proxy data as described in Section 2. In order to reduce uncertainties of emissions in long past years, these procedures need to be considered based on detailed information of each country and region during the period.

4 Summary and remarks

A long historical emission inventory of major air and climate pollutants in Asia during 1950-2015 was developed as Regional Emission inventory in ASia version 3~~.1~~ (REASv3~~.1~~). Target species were SO_2, NO_x, CO, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, BC, OC, and CO_2 and the domain areas included East, Southeast, and South Asia. Emissions from fuel combustion in power plants, industries, transport, and domestic sectors and those from industrial processes were estimated for all the species. In addition, emissions from evaporative sources were included in NMVOC and those from agricultural activities and human physiological phenomenon were considered for NH~~emissions from agricultural activities and human physiological phenomenon were considered for NH_3 and those from evaporative sources were included in NMVOC~~. REASv3~~.1~~ provides gridded data as well as emissions from each country and sub-region. Spatial resolution is mainly 0.25° × 0.25° and large power plants are treated as point sources. Temporal resolution is monthly. Emissions were estimated based on information of technologies related to emission factors and removal efficiencies, although available data and literatures are limited in the case of Asia. Activity data for recent years were collected from international and national statistics and those of past years, when detailed information was not available, were extrapolated using proxy data for the target period of REASv3~~.1~~. Details of methodologies such as data sources and treatments, settings of emission factors and emission controls, and related assumptions were provided in the supplement document entitled “Supplementary information and data to methodology of REASv3”.

Total emissions in Asia averaged during 1950-1955 and 2010-2015 (growth rates in these 60 years) are: SO_2: 3.2 Tg, 42.4 Tg (13.1); NO_x: 1.6 Tg, 47.3 Tg (29.1); CO: 56.1 Tg, 303 Tg (5.4); NMVOC: 7.0 Tg, 57.8 Tg (8.3); NH_3: 8.0 Tg, 31.3 Tg (3.9); CO_2: 1.1 Pg, 18.6 Pg (16.5); PM_{10}: 5.9 Tg, 30.2 Tg (5.1); $PM_{2.5}$: 4.6 Tg, 21.3 Tg (4.6); BC: 0.69 Tg, 3.2 Tg (4.7); and OC: 2.5 Tg, 6.6 Tg (2.7)~~SO_2: 3.15 Tg, 42.4 Tg (13.5); NO_x: 1.83 Tg, 47.6 Tg (26.0); CO: 62.2 Tg, 319 Tg (5.13); NMVOC: 9.14 Tg, 61.8 Tg (6.77); NH_3: 7.99 Tg, 31.3 Tg (3.92); CO_2: 1.12 Pg, 18.3 Pg (16.3); PM_{10}: 5.76 Tg, 28.4 Tg (4.92); $PM_{2.5}$: 4.52 Tg, 20.3 Tg (4.50); BC 0.751 Tg, 3.38 Tg (4.51); and OC 2.62 Tg, 6.92 Tg (2.64)~~. Clearly, all the air pollutant emissions in Asia increased significantly during these six decades. However, situations were different among countries and regions. In recent years, the relative contribution of air pollutant emissions from China was the largest along with rapid increase in economic growth, but most species have reached their peaks and the growth rates of other species have become at least small or almost zero. For SO_2 and NO_x, introduction of abatement equipment, especially for coal-fired power plants, such as FGD and SCR were considered to be effective in reducing emissions. For PM species, in addition to control equipment in industrial plants, emissions decreased recently due to reduced usage of biofuels. On the other hand, air pollutant emissions from India showed an almost continuous increase. Growth rates were larger for SO_2 and NO_x, but their structures of emissions were different. Large parts of SO_2 emissions were obtained from coal combustion in power plants and industrial sector, and the recent rapid increase of SO_2 emission was mainly from coal-fired power plants. For NO_x, contribution ~~and growth rates~~ from road transport especially diesel vehicles were almost comparable with those of power plants. For PM species, a majority of emissions was from the residential sector in the 1950s-1960s and~~;~~ its contribution is still considered to be large. Recent increasing trends were mainly caused by emissions from power and industrial plants and road vehicles. Trends in Japan were much different than those of the whole of Asia. Emissions increased rapidly along with economic growth during the 1950s-1970s, but those of most species were reduced largely from peak values. In addition, peak years were mostly 40 years ago, reflecting the time series of introduction of ~~regulations and laws~~control measures to mitigate air pollution. Similar features were found in ~~the~~ Republic of Korea and Taiwan. For other countries in Asia, emissions of air pollutants generally showed increasing trends along with economic situation and motorization. As described above, trends and spatial distribution of air pollutants in Asia are not simple and are becoming complicated.

Mitigation of air and climate pollutant emissions is an urgent issue in most Asian countries, but the situation is different country-wise. In this study, detailed discussion on effects of emission controls were conducted only for China and Japan due to limitation of information. Therefore, continuous efforts to develop and update emission inventories in Asia based on country specific information are essential especially for countries and regions other than China and Japan. On the other hand, ~~Therefore, continuous efforts to develop and update emission inventories in Asia that are based on country specific information are essential. However,~~ there are inevitable uncertainties in parameters required to develop emission inventories, such as activity data and emission factors. In addition, it is fundamentally impossible to develop a real-time emission inventory because there is a time lag in the publication of basic statistics essential to estimate emissions. Recently, satellite observation data of air pollutants are becoming available at a finer scale for many species, such as NO_x, SO_2 and NH_3. Evaluations and improvements of REASv3~~.1~~ based on these data as well as results of modeling studies, such as inverse modeling are more important next steps. Also, addition of target species, especially CH_4, which is one of the key species to mitigate both air pollution and global warming is another important task for future studies.

Data availability:

Monthly gridded emission data sets at 0.25° × 0.25° resolution for major sectors from 1950 to 2015 are available from a data download site of REAS. The URL of the site is http://www.nies.go.jp/REAS/. Country and regional emission table data for major sectors during 1950-2015 and those for major fuel types are also provided at the site. Note that datasets of REASv3.1 were released after a publication of Kurokawa et al. (2019) from December 2019. The datasets were revised and the updated data are available as REASv3.2 together with a publication of this paper. Differences between REASv3.2 and REASv3.1 were presented and discussed in the Supplement document entitled “Differences between REASv3.2 and REASv3.1”.

Author contribution:

JK and TO conducted the study design. JK contributed to actual works for development of REASv3~~.1~~ such as collecting data and information, settings of parameters, calculating emissions and creating final data sets. JK and TO analyzed and discussed the estimated emissions in REASv3~~.1~~. JK prepared the manuscript with contributions from TO.

Competing interest:

The authors declare that they have no conflict of interest.

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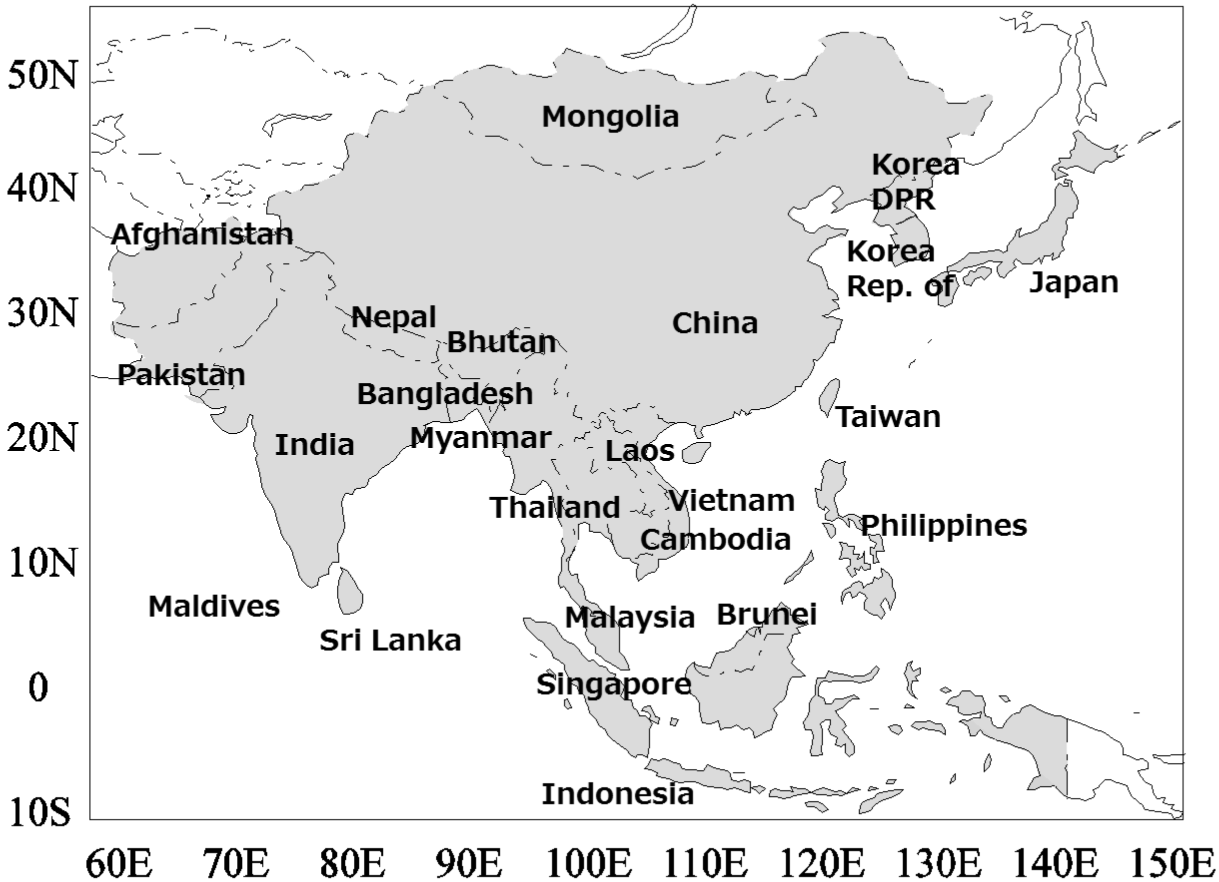
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Korea Rep. of
Japan
Nepal
China
Bhutan
Pakistan
Bangladesh
Taiwan
India
Myanmar
Laos
Vietnam
Thailand
Philippines
Cambodia
Maldives
Malaysia
Brunei
Sri Lanka
Singapore
Indonesia

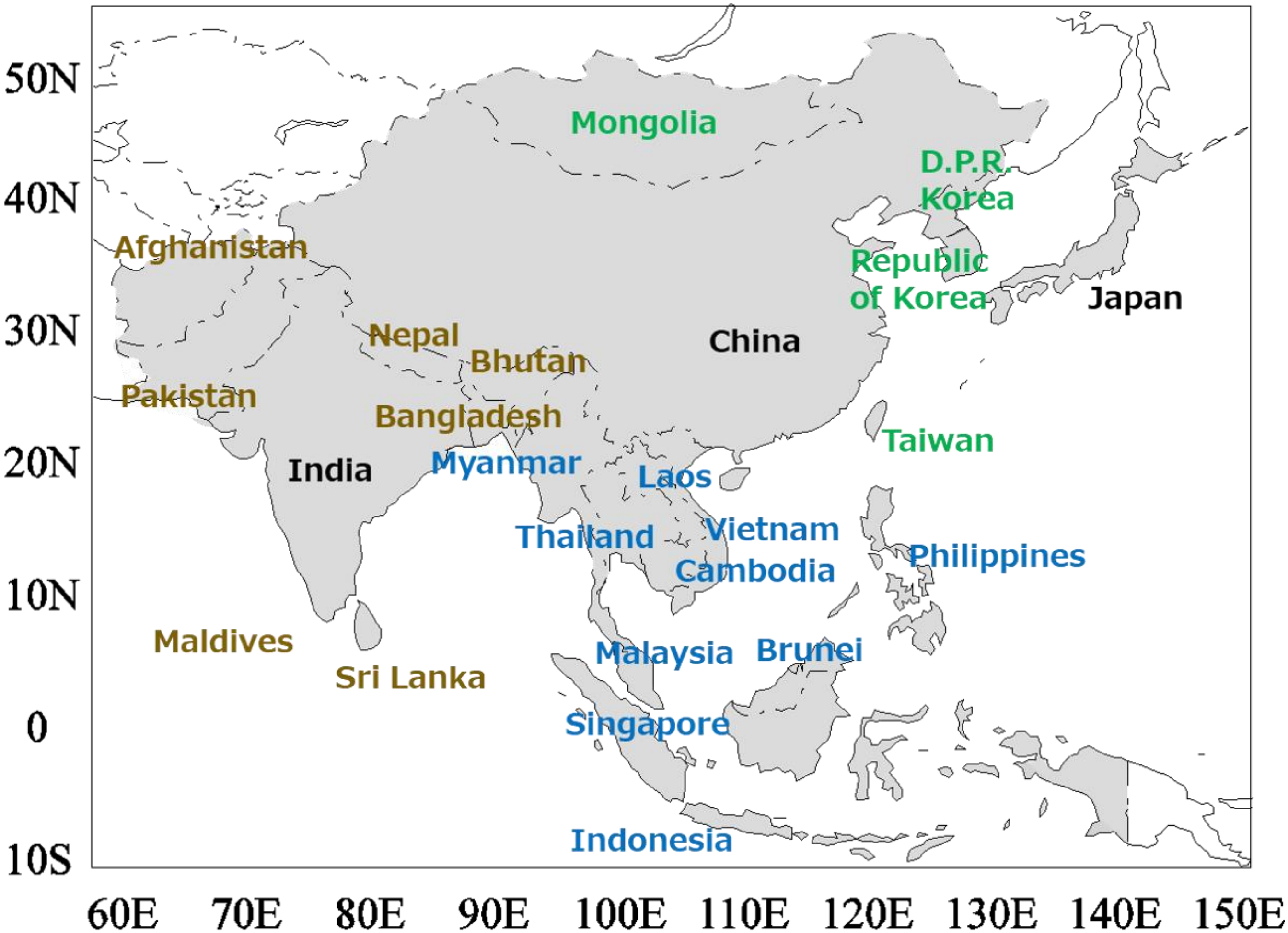
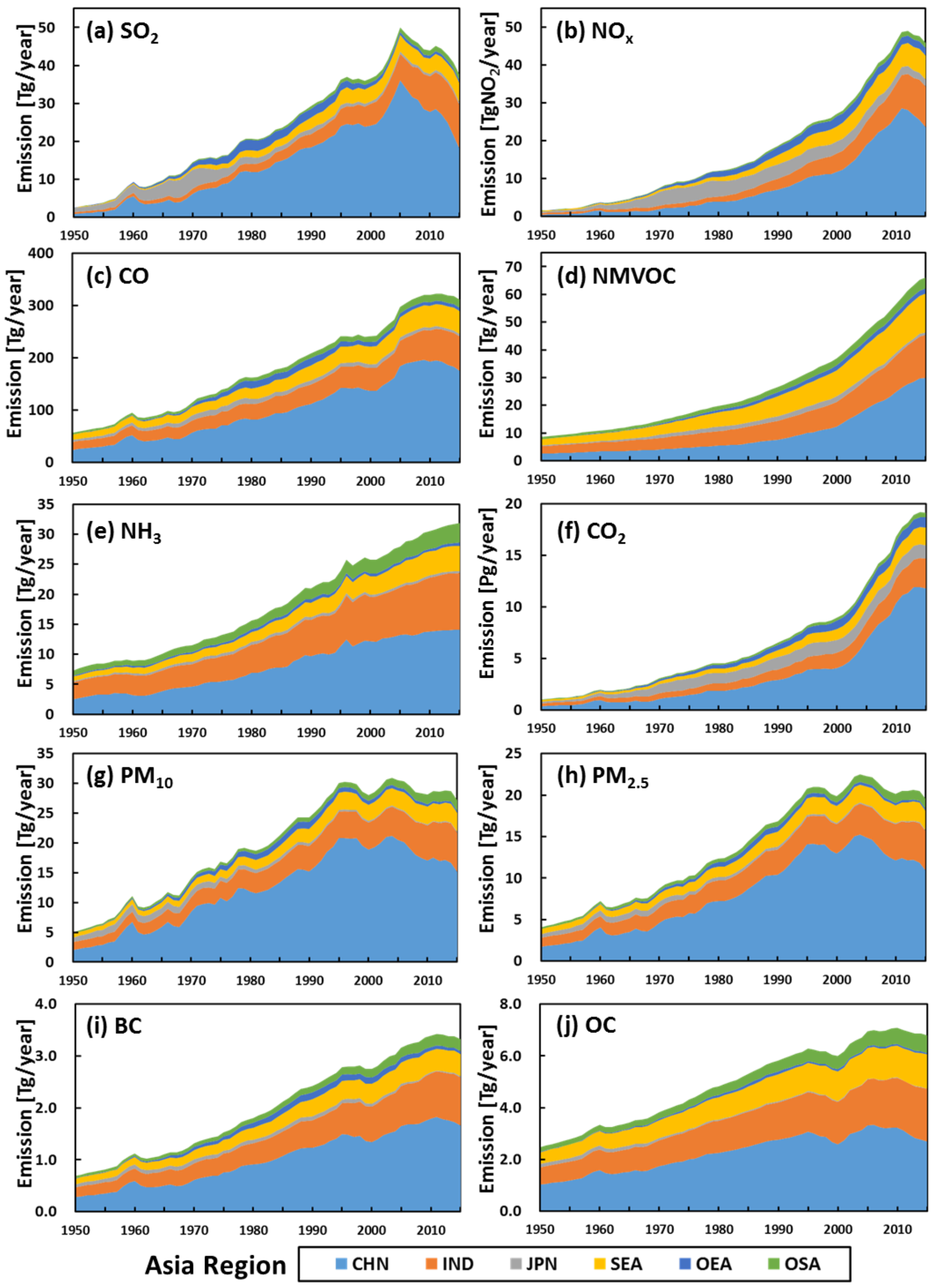


Figure 1. Domain and target countries of REASv3. In this paper, countries written in blue, green, and brown characters were defined as SEA (Southeast Asia), OEA (East Asia other than China and Japan), and OSA (South Asia other than India), respectively.~~**Figure 1: Domain and target countries of REASv3.1.**~~


(a) SO_2
Emission [Tg/year]
(b) NO_x
Emission [$TgNO_2$/year]
(c) CO
Emission [Tg/year]
(d) NMVOC
Emission [Tg/year]
(e) NH_3
Emission [Tg/year]
(f) CO_2
Emission [Pg/year]
(g) PM_{10}
Emission [Tg/year]
(h) $PM_{2.5}$
Emission [Tg/year]
(i) BC
Emission [Tg/year]
(j) OC
Emission [Tg/year]
Asia Region
CHN
IND
JPN
SEA
OEA
OSA

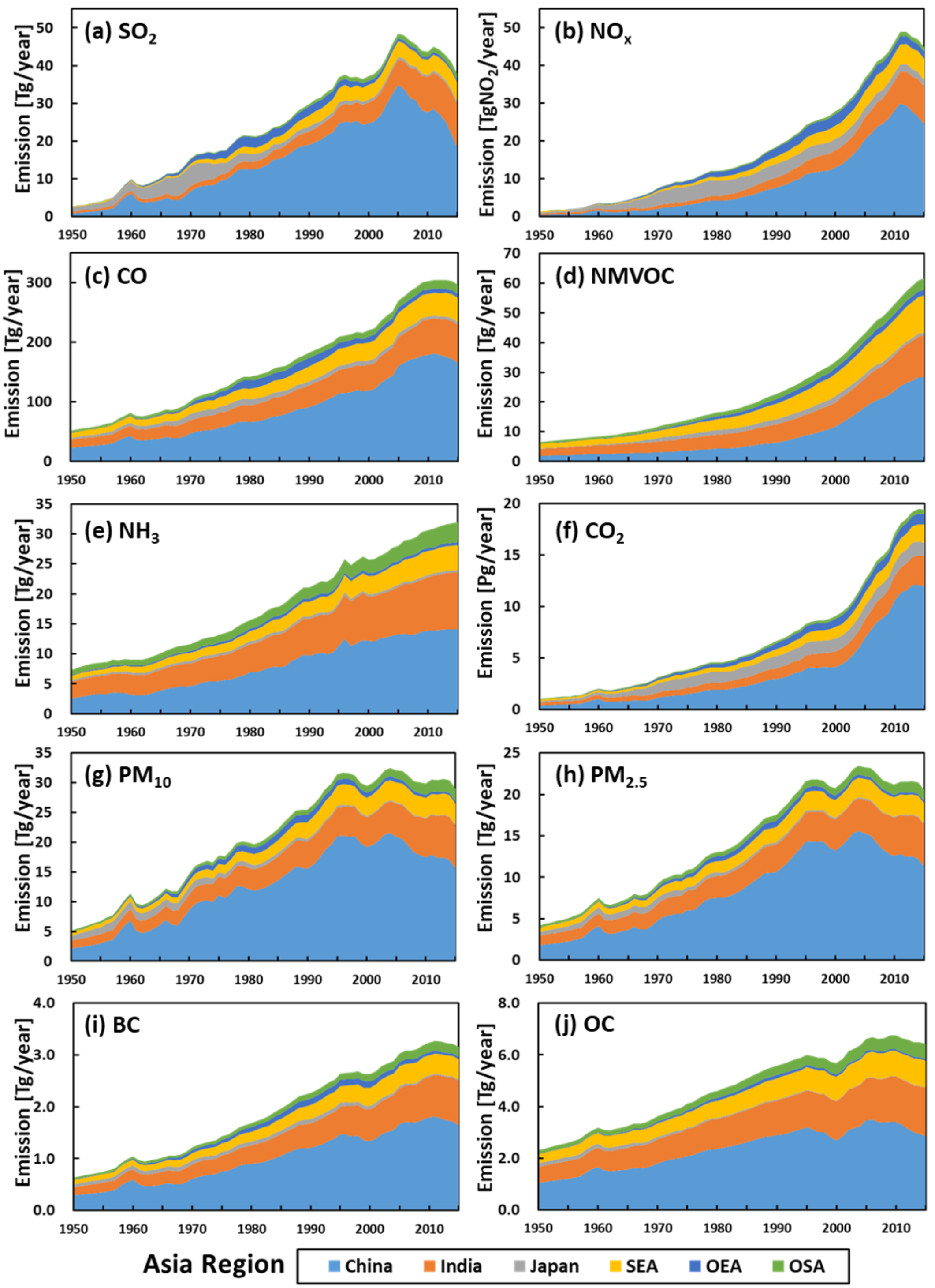
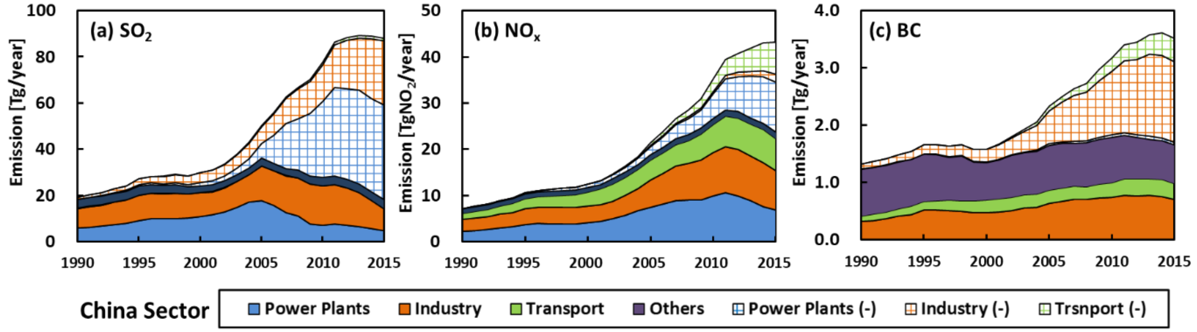

(a) SO_2
Emission [Tg/year]
(b) NO_x
Emission [$TgNO_2$/year]
(c) CO
Emission [Tg/year]
(d) NMVOC
Emission [Tg/year]
(e) NH_3
Emission [Tg/year]
(f) CO_2
Emission [Pg/year]
(g) PM_{10}
Emission [Tg/year]
(h) $PM_{2.5}$
Emission [Tg/year]
(i) BC
Emission [Tg/year]
(j) OC
Emission [Tg/year]
Asia Region
China
India
Japan
SEA
OEA
OSA

Figure 2. Trends of (a) SO_2, (b) NO_x, (c) CO, (d) NMVOC, (e) NH_3, (f) CO_2, (g) PM_{10}, (h) $PM_{2.5}$, (i) BC, and (j) OC emissions in Asia during 1950-2015 for each region. See Fig. 1 for countries included in SEA, OEA, and OSA.~~**Figure 2: Trends of (a) SO_2, (b) NO_x, (c) CO, (d) NMVOC, (e) NH_3, (f) CO_2, (g) PM_{10}, (h) $PM_{2.5}$, (i) BC, and (j) OC emissions in Asia during 1950-2015 for each region. Regions: CHN = China, IND = India, JPN = Japan, SEA = Southeast Asia, OEA = East Asia other than China and Japan, and OSA = South Asia other than India. See Table S1 for countries included in SEA, OEA, and OSA.**~~



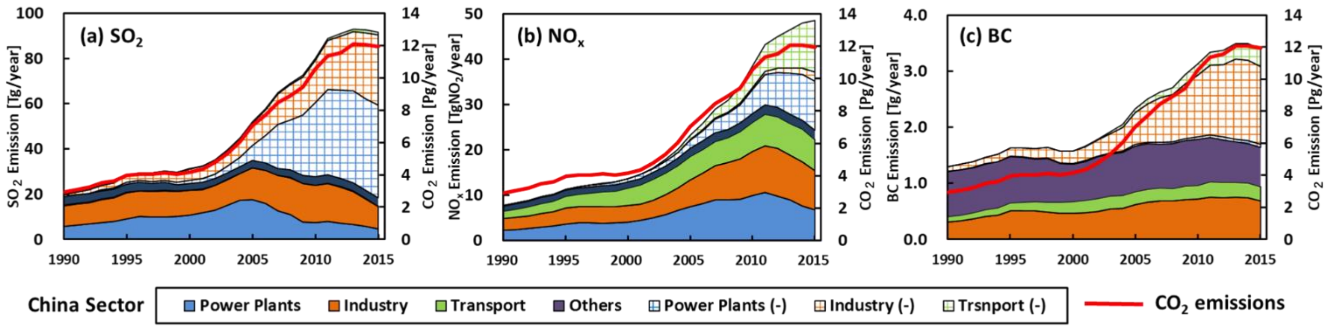
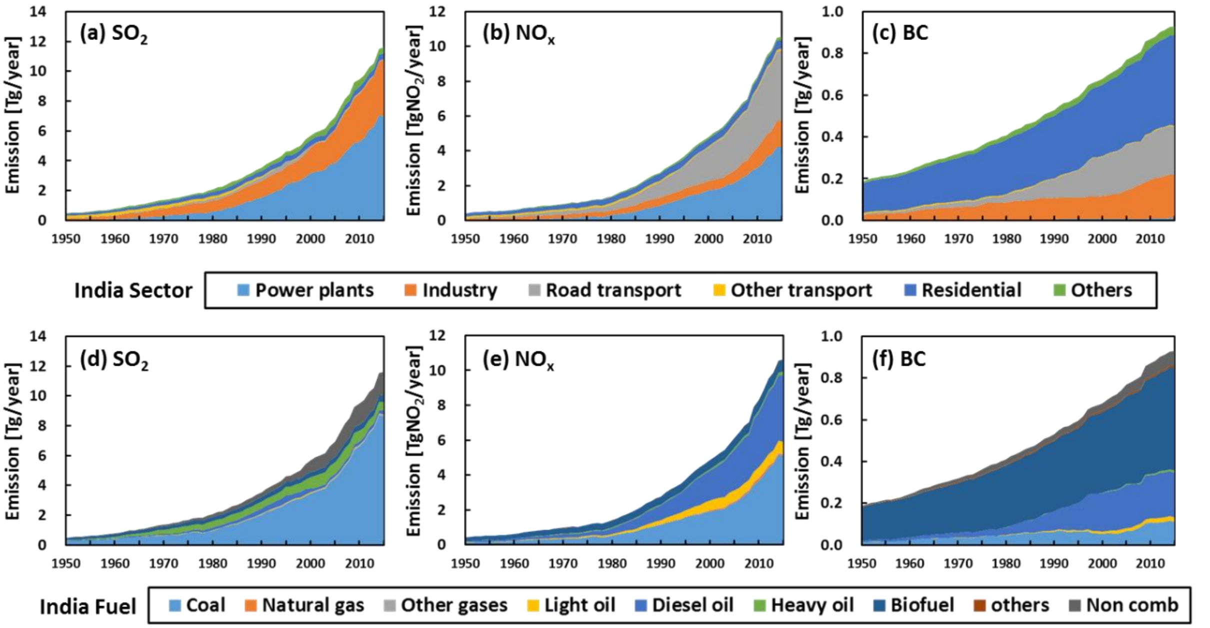


Figure 3. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in China during 1990-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures. Red lines in the panels are total CO_2 emissions.~~**Figure 3: Emissions of (a) SO₂, (b) NOₓ, and (c) BC from each major sector in China during 1950-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures.**~~



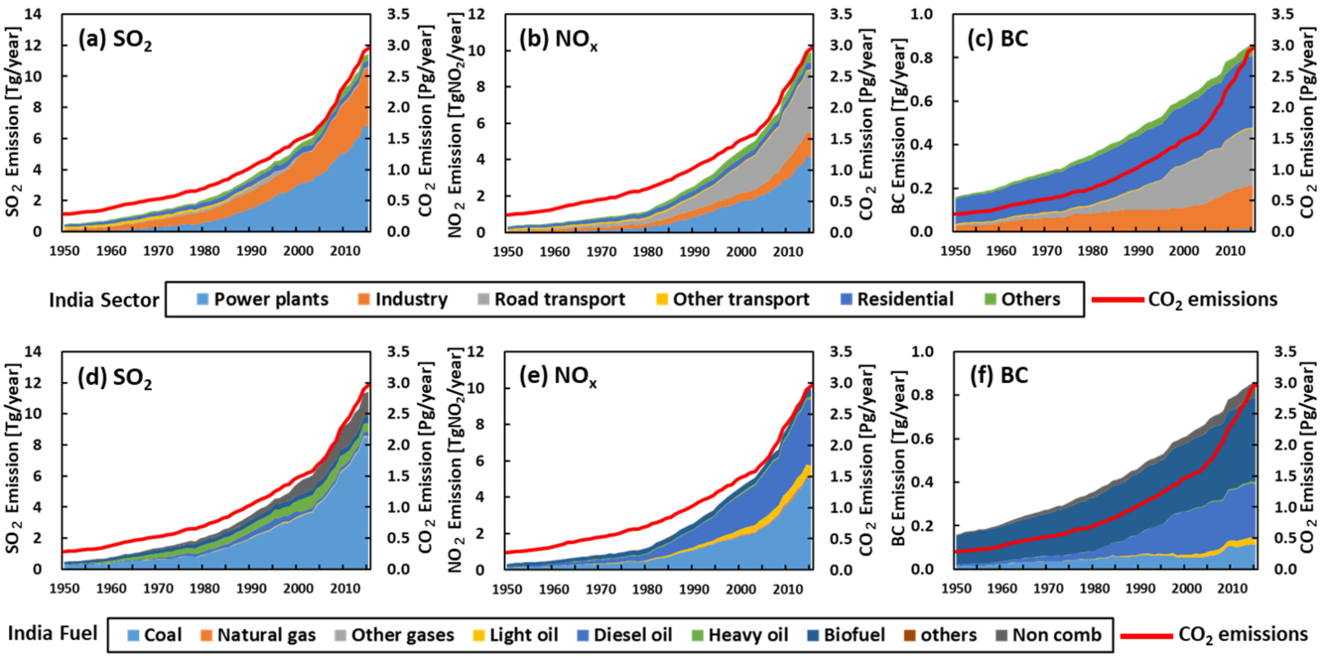
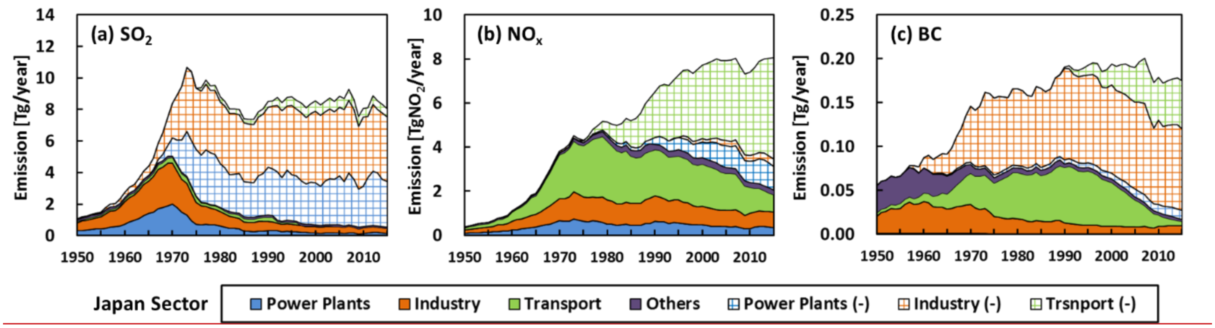


Figure 4. Emissions of (a, d) SO_2, (b, e) NO_x, and (c, f) BC from each major sector category (upper panels) and fuel type (lower panels) in India from 1950 to 2015 (Non comb = Non combustion sources). Red lines in the panels are total CO_2 emissions.~~**Figure 4: Emissions of (a, d) SO_2, (b, e) NO_x, and (c, f) BC from each major sector category (upper panels) and fuel type (lower panels) in India from 1950 to 2015. (Non comb = Non combustion sources)**~~



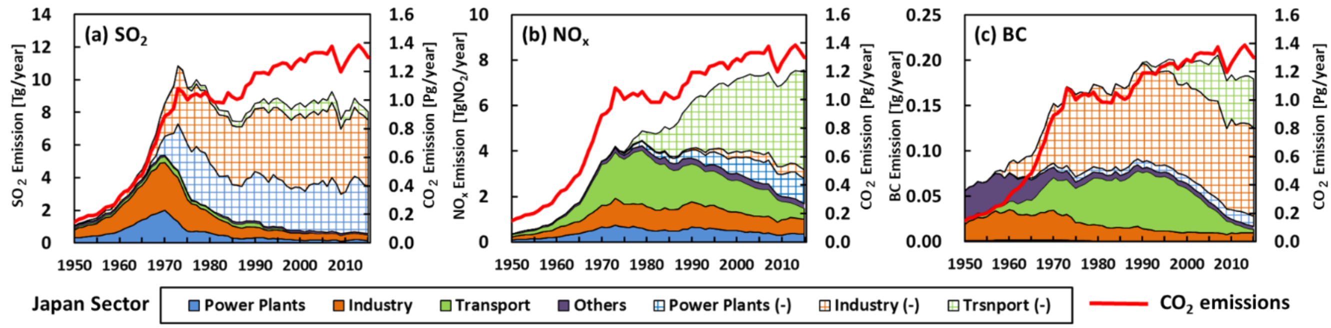
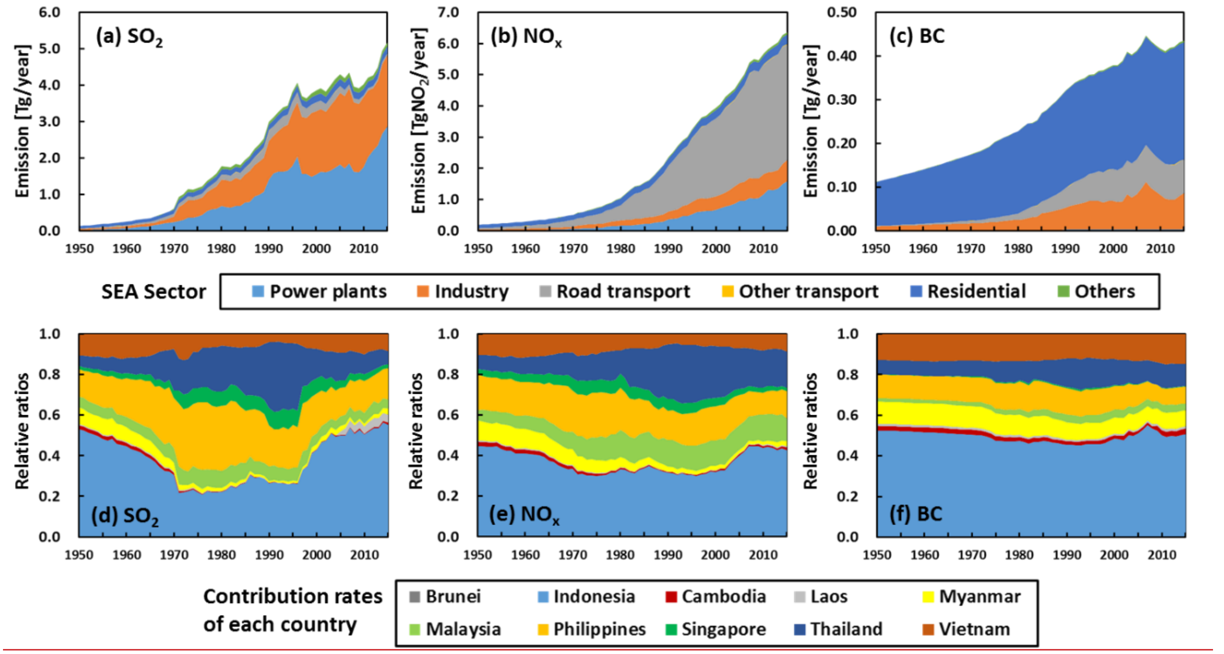


Figure 5. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in Japan during 1950-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures. Red lines in the panels are total CO_2 emissions.~~**Figure 5: Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in Japan during 1950-2015. Solid colored areas are actual emissions and hatched ones (-) are reduced emissions due to control measures.**~~



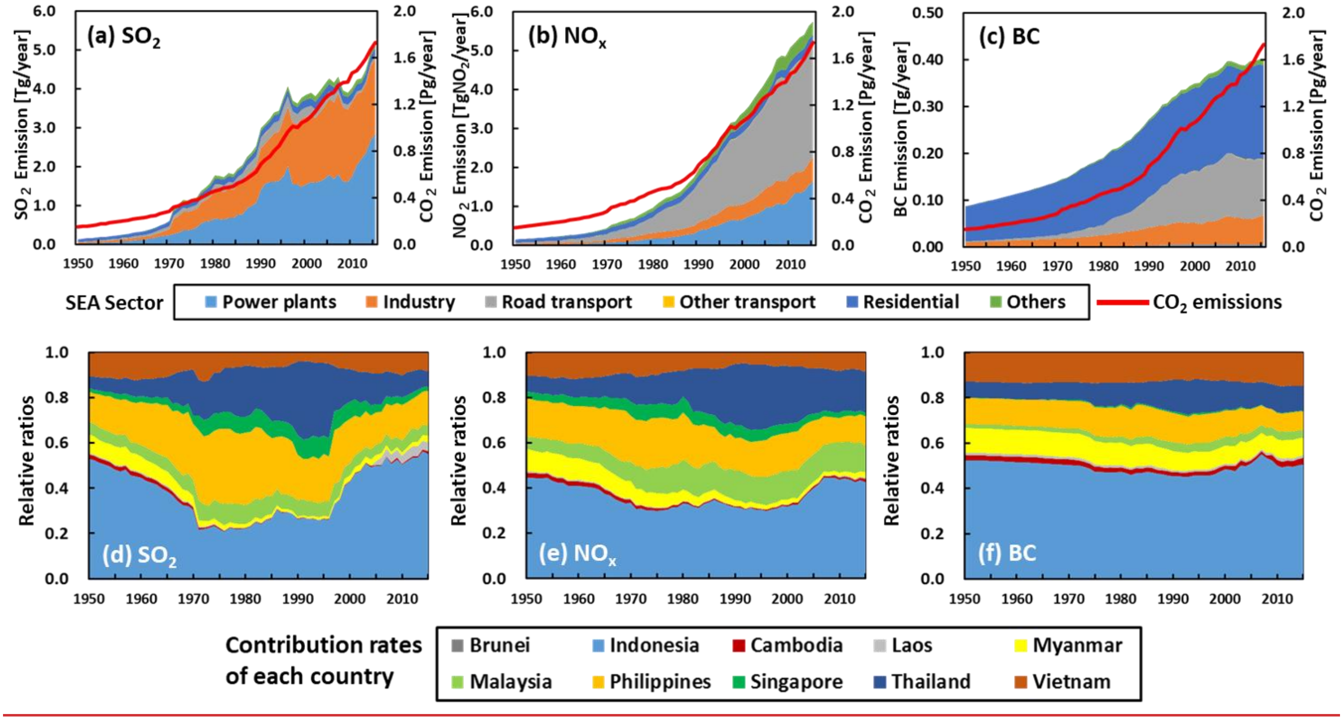
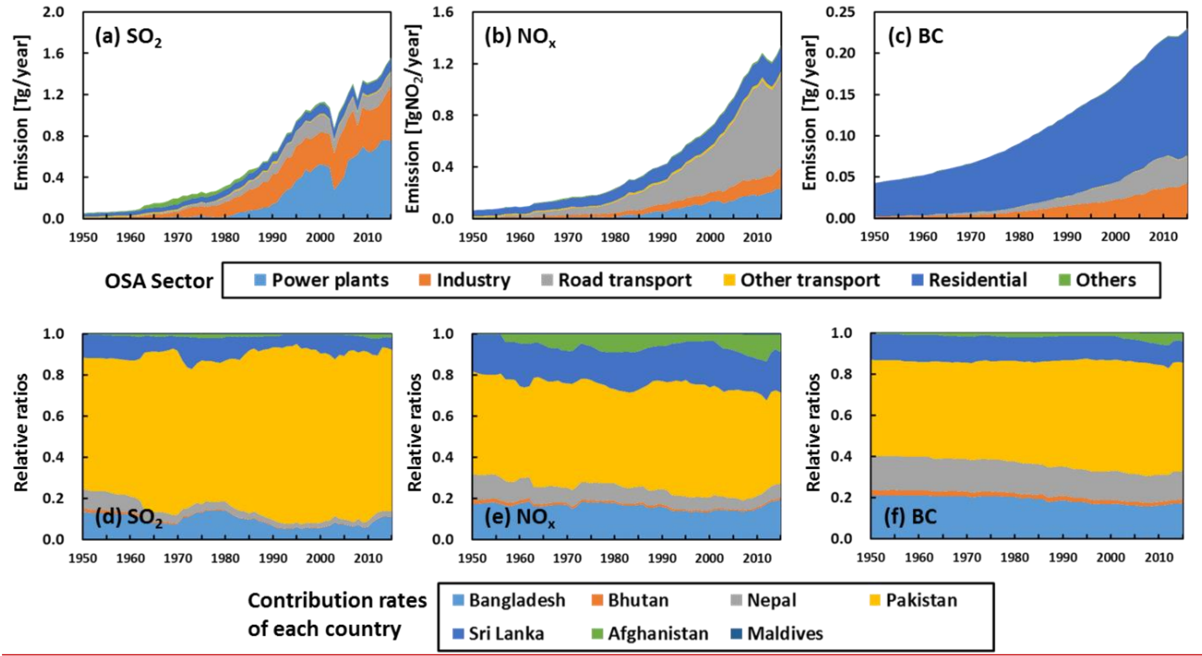


Figure 6. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in SEA (upper panels) and (d, e, f) relative ratios of emissions from each country in SEA (lower panels) during 1950-2015. Red lines in the upper panels are total CO_2 emissions.~~**Figure 6: Emissions of (a) SO2, (b) NOx, and (c) BC from each major sector in total Southeast Asia (SEA) (upper panels) and (d, e, f) relative ratios of emissions from each country in SEA (lower panels) during 1950-2015.**~~



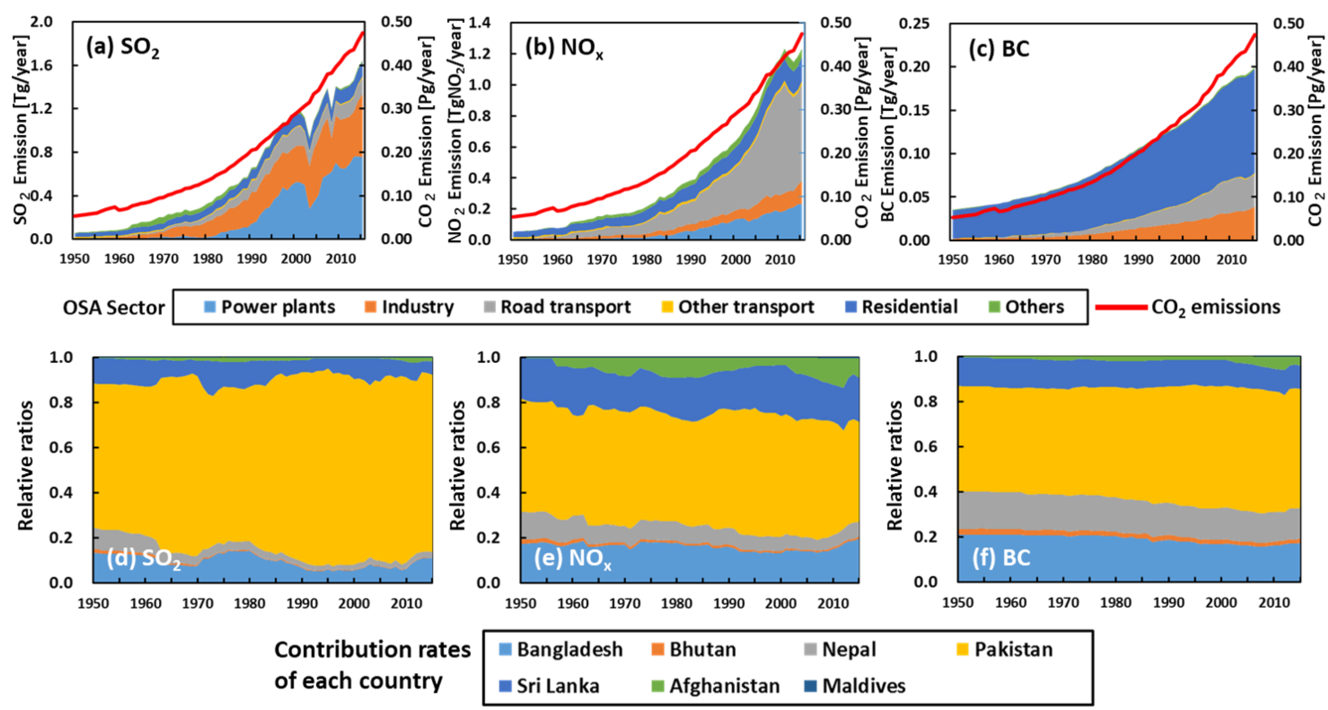
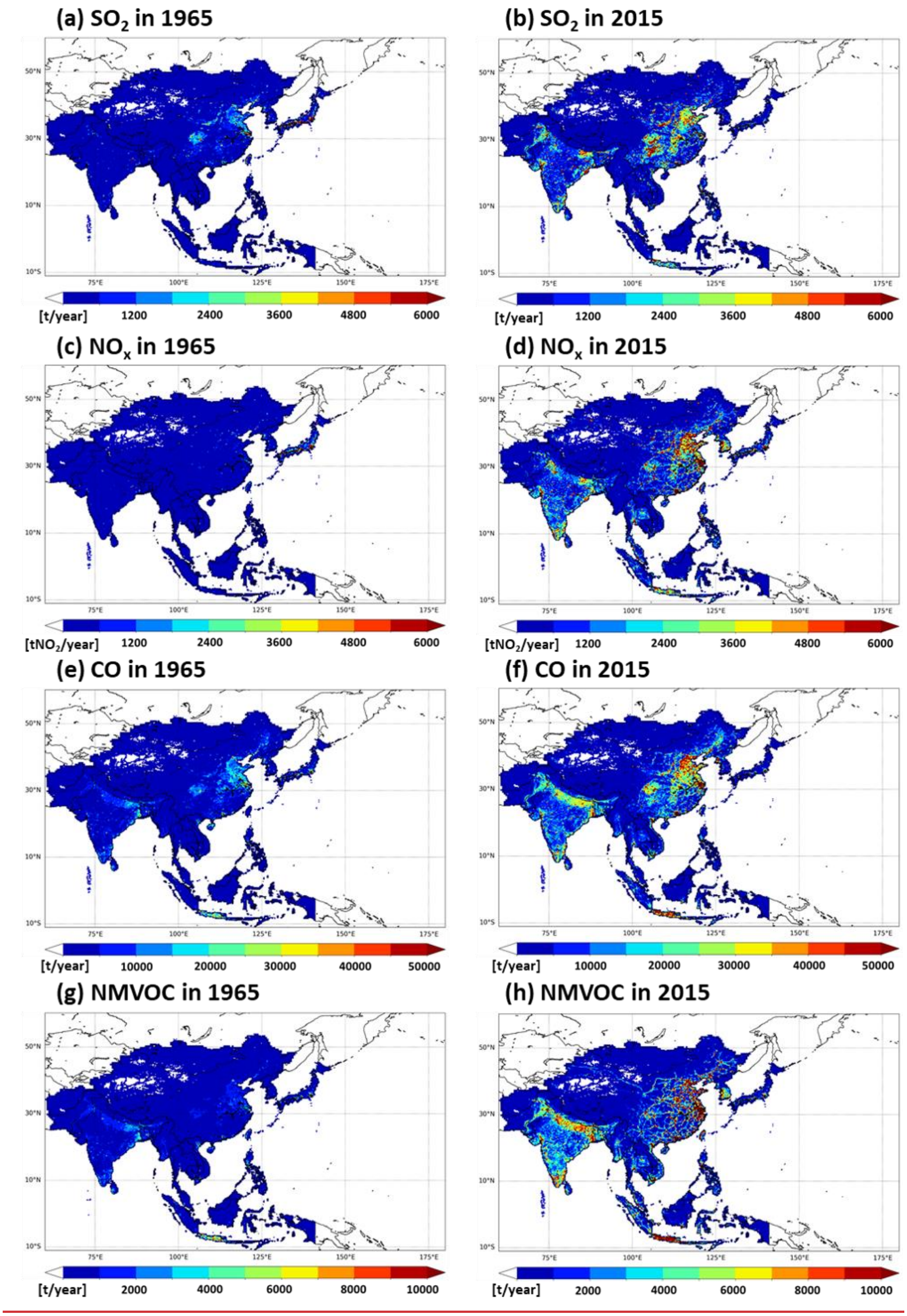


Figure 7. Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in OSA (upper panels) and (d, e, f) relative ratios of emissions from each country in OSA (lower panels) from 1950 to 2015.~~**Figure 7: Emissions of (a) SO_2, (b) NO_x, and (c) BC from each major sector in total South Asia other than India (OSA) (upper panels) and (d, e, f) relative ratios of emissions from each country in OSA (lower panels) from 1950 to 2015.**~~



(a) SO_2 in 1965
(b) SO_2 in 2015
(c) NO_x in 1965
(d) NO_x in 2015
(e) CO in 1965
(f) CO in 2015
(g) NMVOC in 1965
(h) NMVOC in 2015
[t/year] 1200 2400 3600 4800 6000
[tNO_2/year] 1200 2400 3600 4800 6000
[t/year] 10000 20000 30000 40000 50000
[t/year] 2000 4000 6000 8000 10000

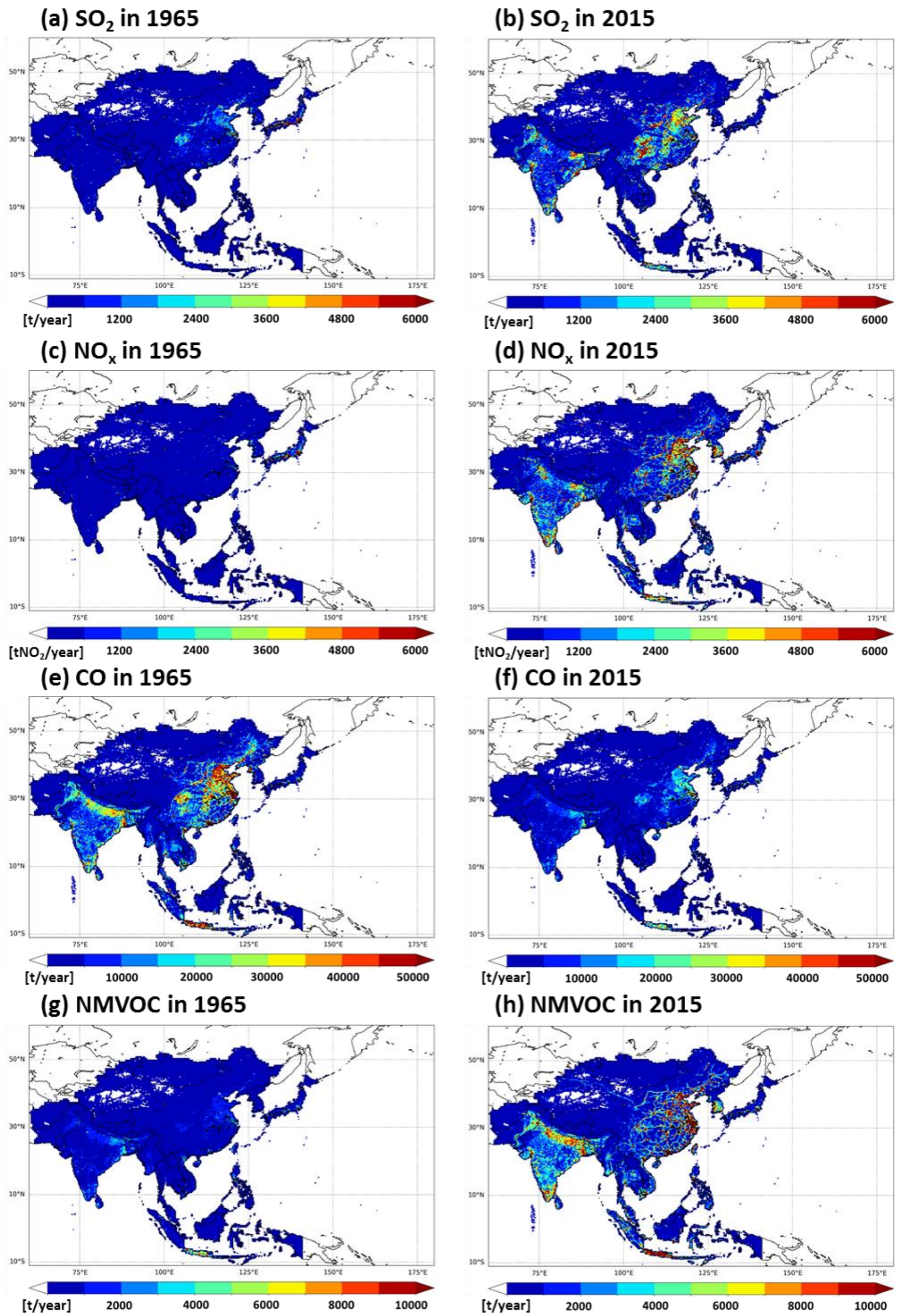
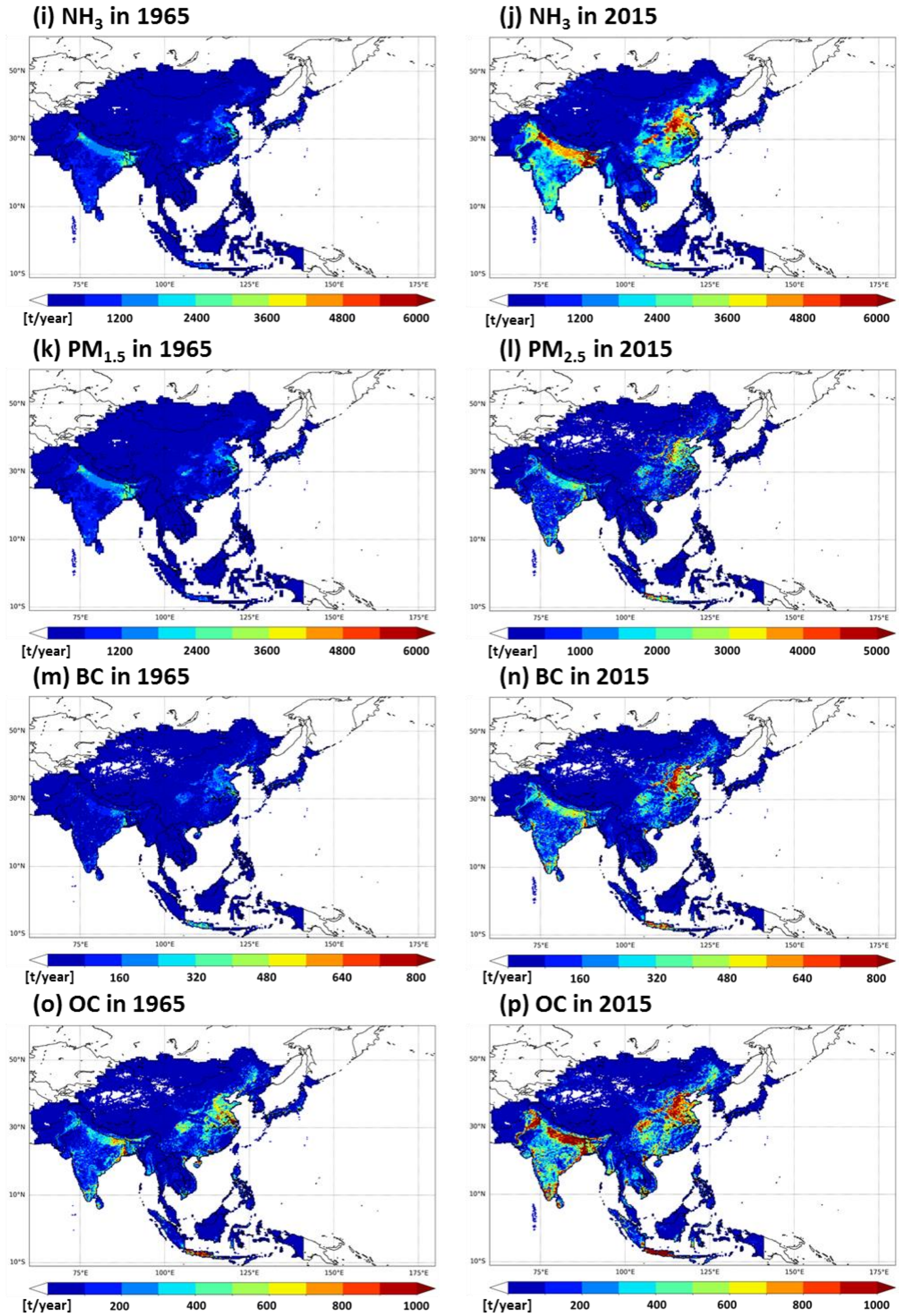

(a) SO_2 in 1965
(b) SO_2 in 2015
[t/year] 1200 2400 3600 4800 6000
(c) NO_x in 1965
(d) NO_x in 2015
[tNO_2/year] 1200 2400 3600 4800 6000
(e) CO in 1965
(f) CO in 2015
[t/year] 10000 20000 30000 40000 50000
(g) NMVOC in 1965
(h) NMVOC in 2015
[t/year] 2000 4000 6000 8000 10000

Figure 8. Grid maps of annual emissions of (a, b) SO_2, (c, d) NO_x, (e, f) CO, (g, h) NMVOC, (i, j) NH_3, (k, l) $PM_{2.5}$, (m, n) BC, and (o, p) OC in 1965 (left panels) and 2015 (right panels).~~**Figure 8: Grid maps of annual emissions of (a, b) SO_2, (c, d) NO_x, (e, f) CO, (g, h) NMVOC, (i, j) NH_3, (k, l) $PM_{2.5}$, (m, n) BC, and (o, p) OC in 1965 (left panels) and 2015 (right panels).**~~


(i) NH3 in 1965
[t/year] 1200 2400 3600 4800 6000
(j) NH3 in 2015
[t/year] 1200 2400 3600 4800 6000
(k) PM1.5 in 1965
[t/year] 1200 2400 3600 4800 6000
(l) PM2.5 in 2015
[t/year] 1000 2000 3000 4000 5000
(m) BC in 1965
[t/year] 160 320 480 640 800
(n) BC in 2015
[t/year] 160 320 480 640 800
(o) OC in 1965
[t/year] 200 400 600 800 1000
(p) OC in 2015
[t/year] 200 400 600 800 1000

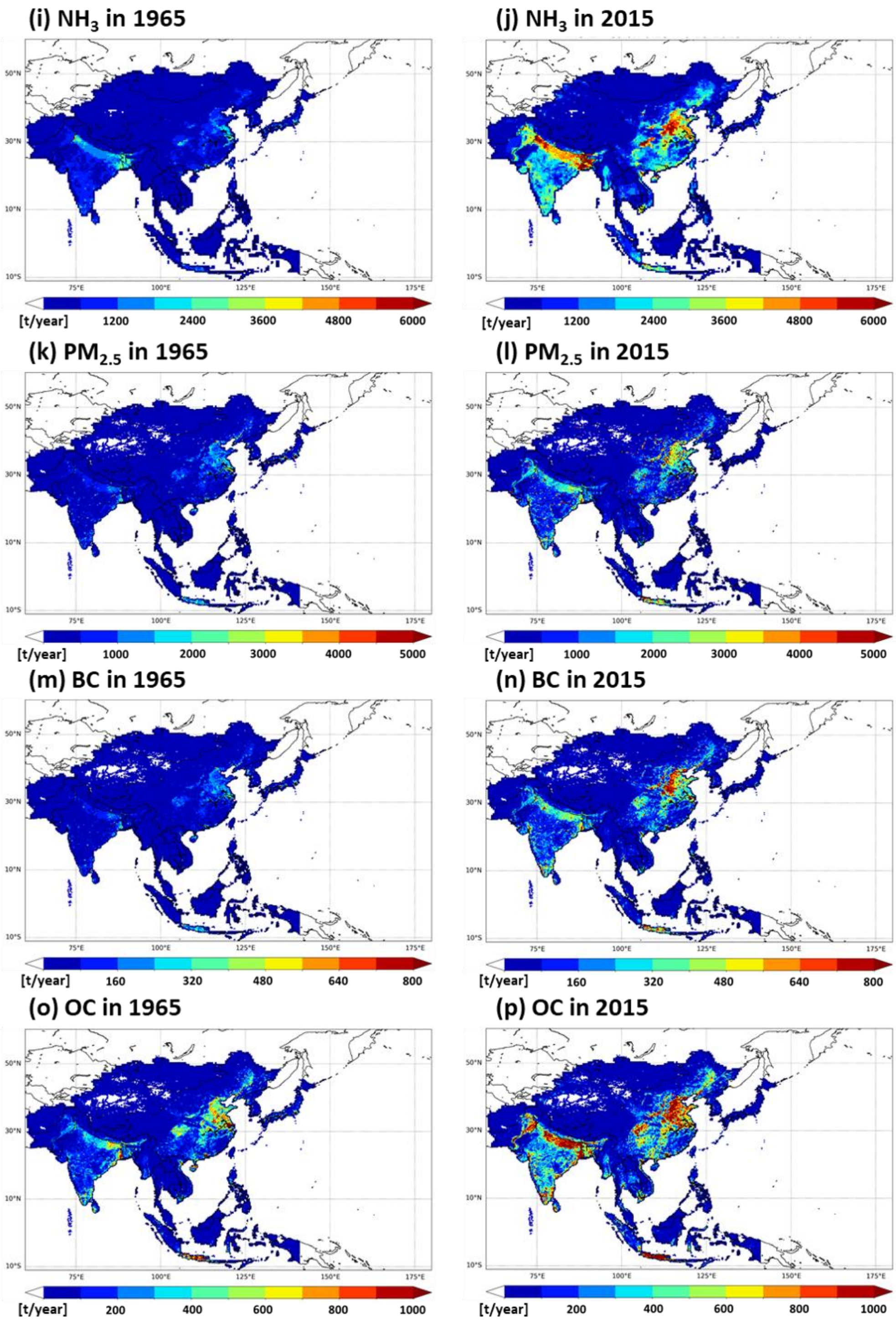
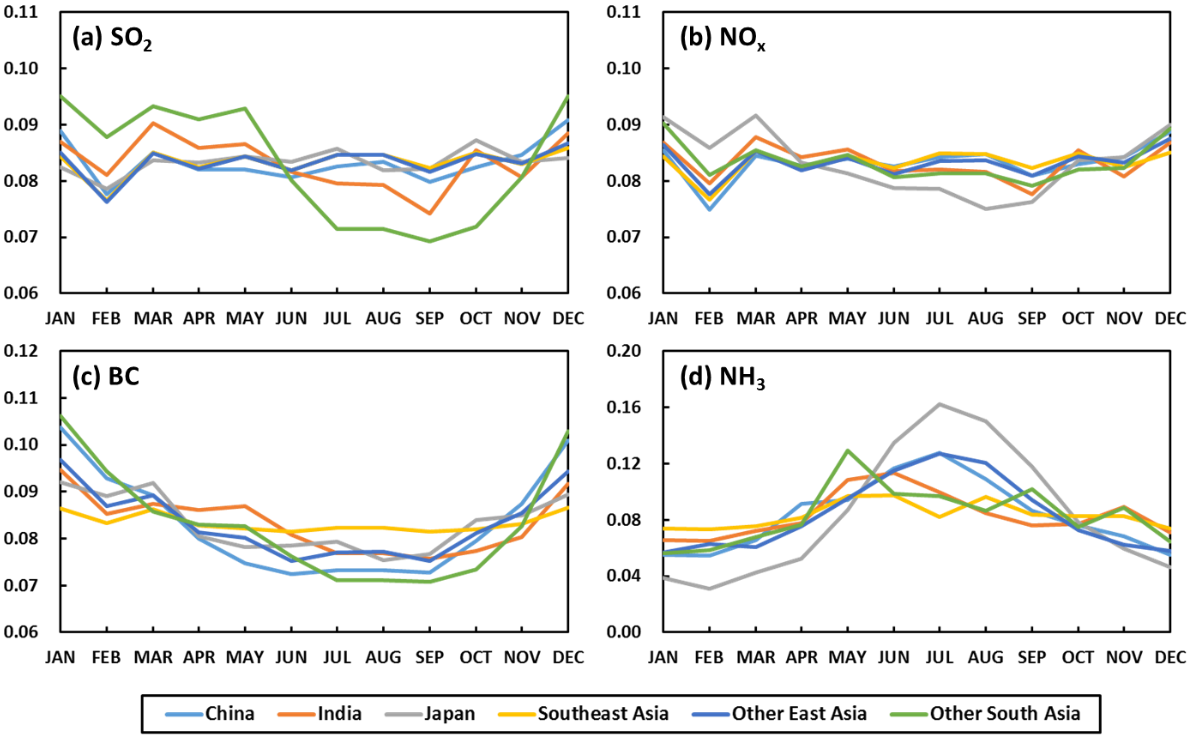


Figure 8. Continued.~~**Figure 8: Continued.**~~


(a) SO2
(b) NOx
(c) BC
(d) NH3
0.11
0.10
0.09
0.08
0.07
0.06
0.12
0.20
0.16
0.12
0.08
0.04
0.00
JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC
China
India
Japan
Southeast Asia
Other East Asia
Other South Asia

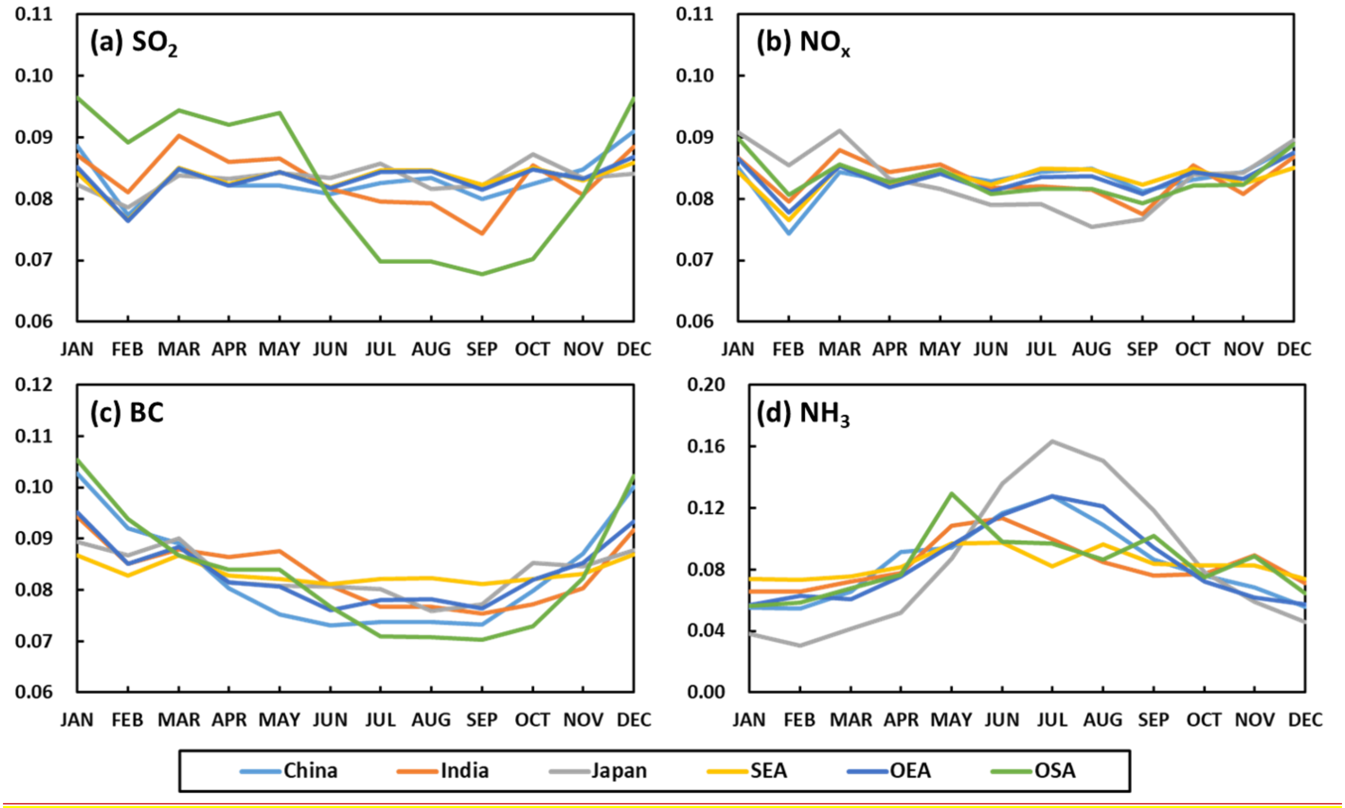
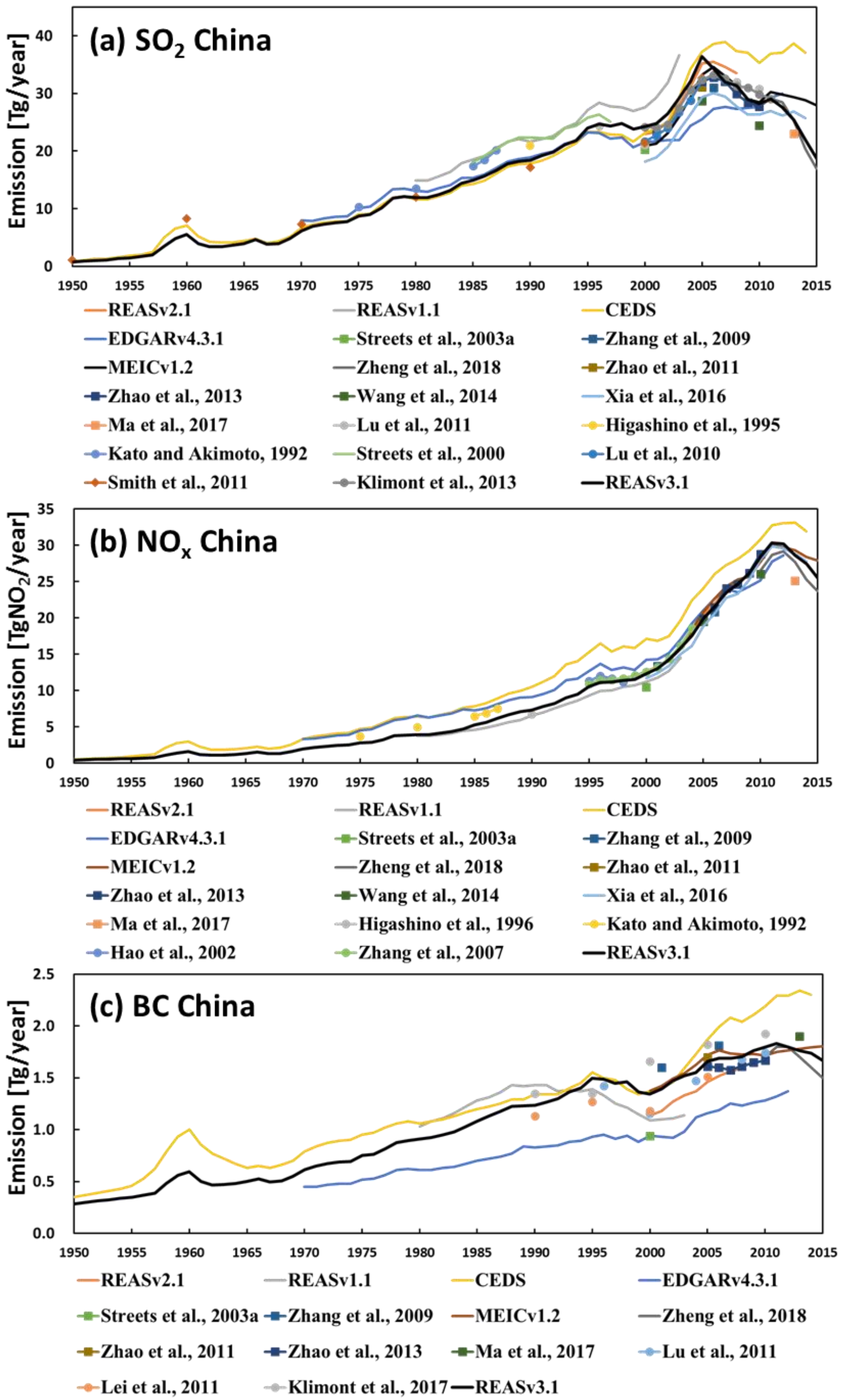


Figure 9. Monthly variations of (a) SO_2, (b) NO_x, (c) BC, and (d) NH_3 emissions for each region of Asia in 2015. See Fig. 1 for definitions of SEA OEA, and OSA.~~**Figure 9: Monthly variations of (a) SO_2, (b) NO_x, (c) BC, and (d) NH_3 emissions for each region of Asia in 2015.**~~


(a) SO_2 China
Emission [Tg/year]
0
10
20
30
40
1950
1955
1960
1965
1970
1975
1980
1985
1990
1995
2000
2005
2010
2015
REASv2.1
REASv1.1
CEDS
EDGARv4.3.1
Streets et al., 2003a
Zhang et al., 2009
MEICv1.2
Zheng et al., 2018
Zhao et al., 2011
Zhao et al., 2013
Wang et al., 2014
Xia et al., 2016
Ma et al., 2017
Lu et al., 2011
Higashino et al., 1995
Kato and Akimoto, 1992
Streets et al., 2000
Lu et al., 2010
Smith et al., 2011
Klimont et al., 2013
REASv3.1
(b) NO_x China
Emission [$TgNO_2$/year]
0
5
10
15
20
25
30
35
1950
1955
1960
1965
1970
1975
1980
1985
1990
1995
2000
2005
2010
2015
REASv2.1
REASv1.1
CEDS
EDGARv4.3.1
Streets et al., 2003a
Zhang et al., 2009
MEICv1.2
Zheng et al., 2018
Zhao et al., 2011
Zhao et al., 2013
Wang et al., 2014
Xia et al., 2016
Ma et al., 2017
Higashino et al., 1996
Kato and Akimoto, 1992
Hao et al., 2002
Zhang et al., 2007
REASv3.1
(c) BC China
Emission [Tg/year]
0.0
0.5
1.0
1.5
2.0
2.5
1950
1955
1960
1965
1970
1975
1980
1985
1990
1995
2000
2005
2010
2015
REASv2.1
REASv1.1
CEDS
EDGARv4.3.1
Streets et al., 2003a
Zhang et al., 2009
MEICv1.2
Zheng et al., 2018
Zhao et al., 2011
Zhao et al., 2013
Ma et al., 2017
Lu et al., 2011
Lei et al., 2011
Klimont et al., 2017
REASv3.1

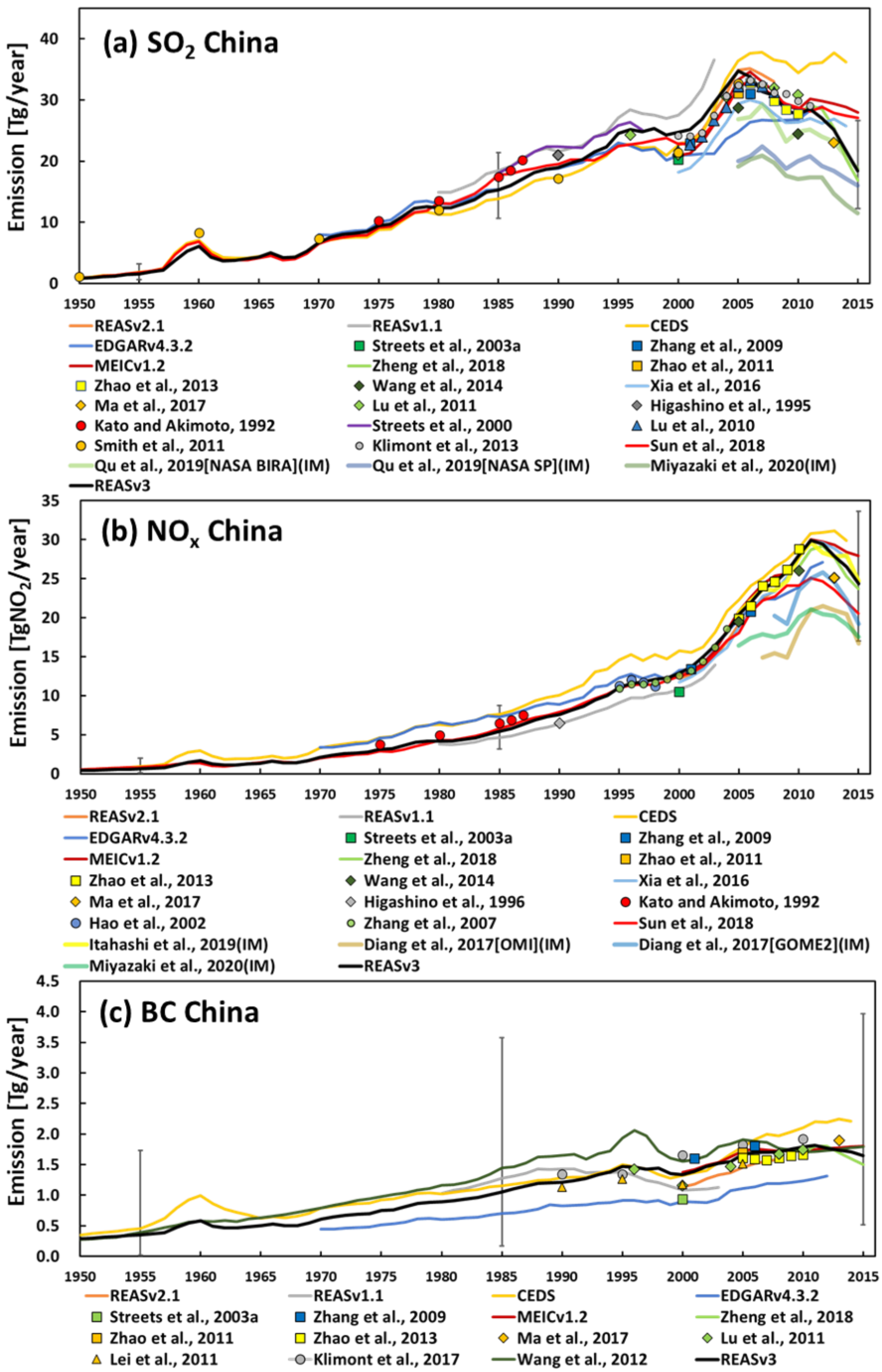
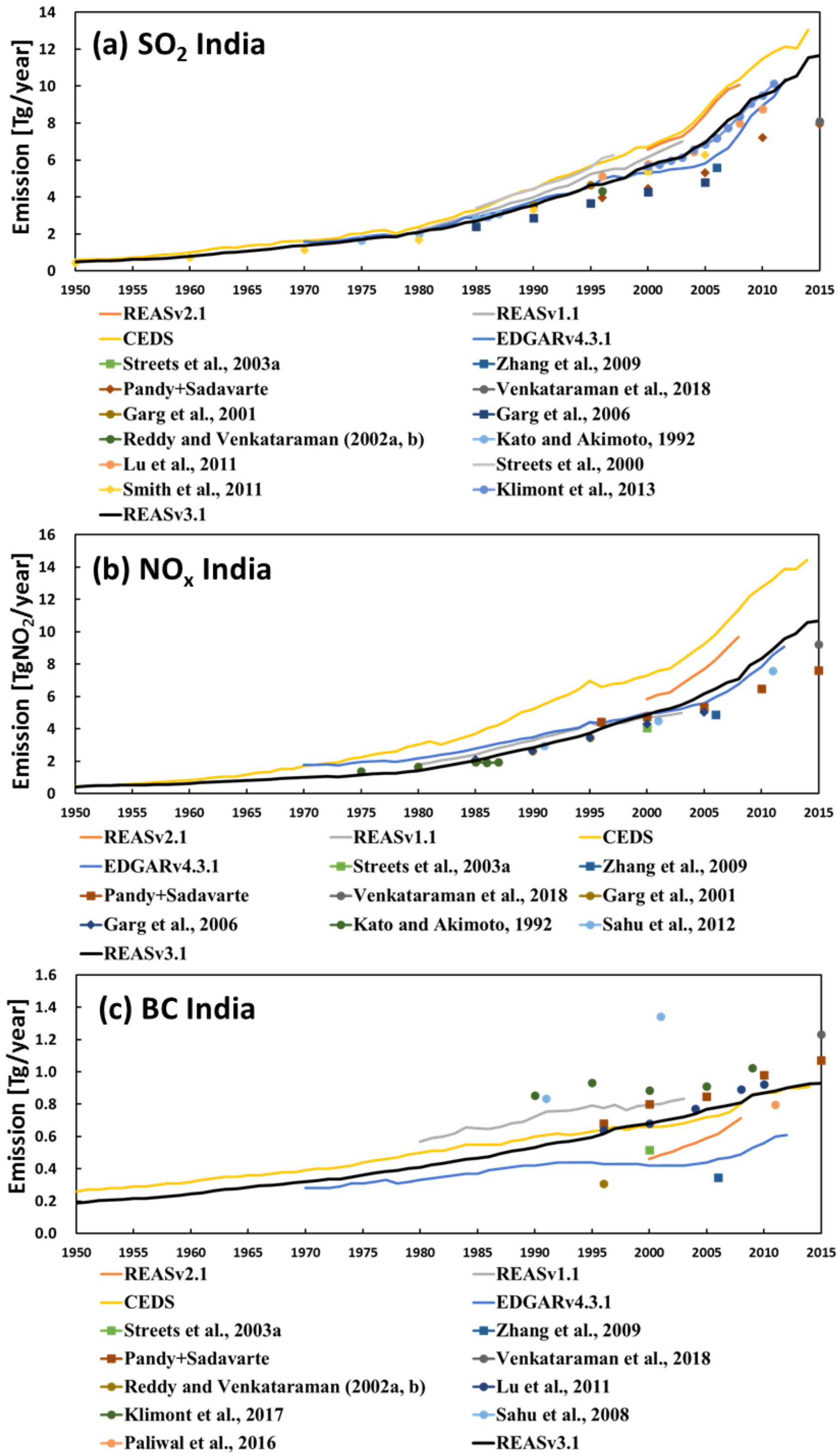


Figure 10. Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in China between REASv3 and other studies. Note that emissions from domestic and fishing ships were excluded from REAS series, CEDS, EDGARv4.3.2, and Higashino et al. (1996). IM means estimates by inverse modeling. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.~~Figure 10: Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in China between REASv3.1 and other studies.~~



(a) SO2 India
Emission [Tg/year]
REASv2.1
REASv1.1
CEDS
EDGARv4.3.1
Streets et al., 2003a
Zhang et al., 2009
Pandy+Sadavarte
Venkataraman et al., 2018
Garg et al., 2001
Garg et al., 2006
Reddy and Venkataraman (2002a, b)
Kato and Akimoto, 1992
Lu et al., 2011
Streets et al., 2000
Smith et al., 2011
Klimont et al., 2013
REASv3.1
(b) NOx India
Emission [TgNO2/year]
REASv2.1
REASv1.1
CEDS
EDGARv4.3.1
Streets et al., 2003a
Zhang et al., 2009
Pandy+Sadavarte
Venkataraman et al., 2018
Garg et al., 2001
Garg et al., 2006
Kato and Akimoto, 1992
Sahu et al., 2012
REASv3.1
(c) BC India
Emission [Tg/year]
REASv2.1
REASv1.1
CEDS
EDGARv4.3.1
Streets et al., 2003a
Zhang et al., 2009
Pandy+Sadavarte
Venkataraman et al., 2018
Reddy and Venkataraman (2002a, b)
Lu et al., 2011
Klimont et al., 2017
Sahu et al., 2008
Paliwal et al., 2016
REASv3.1

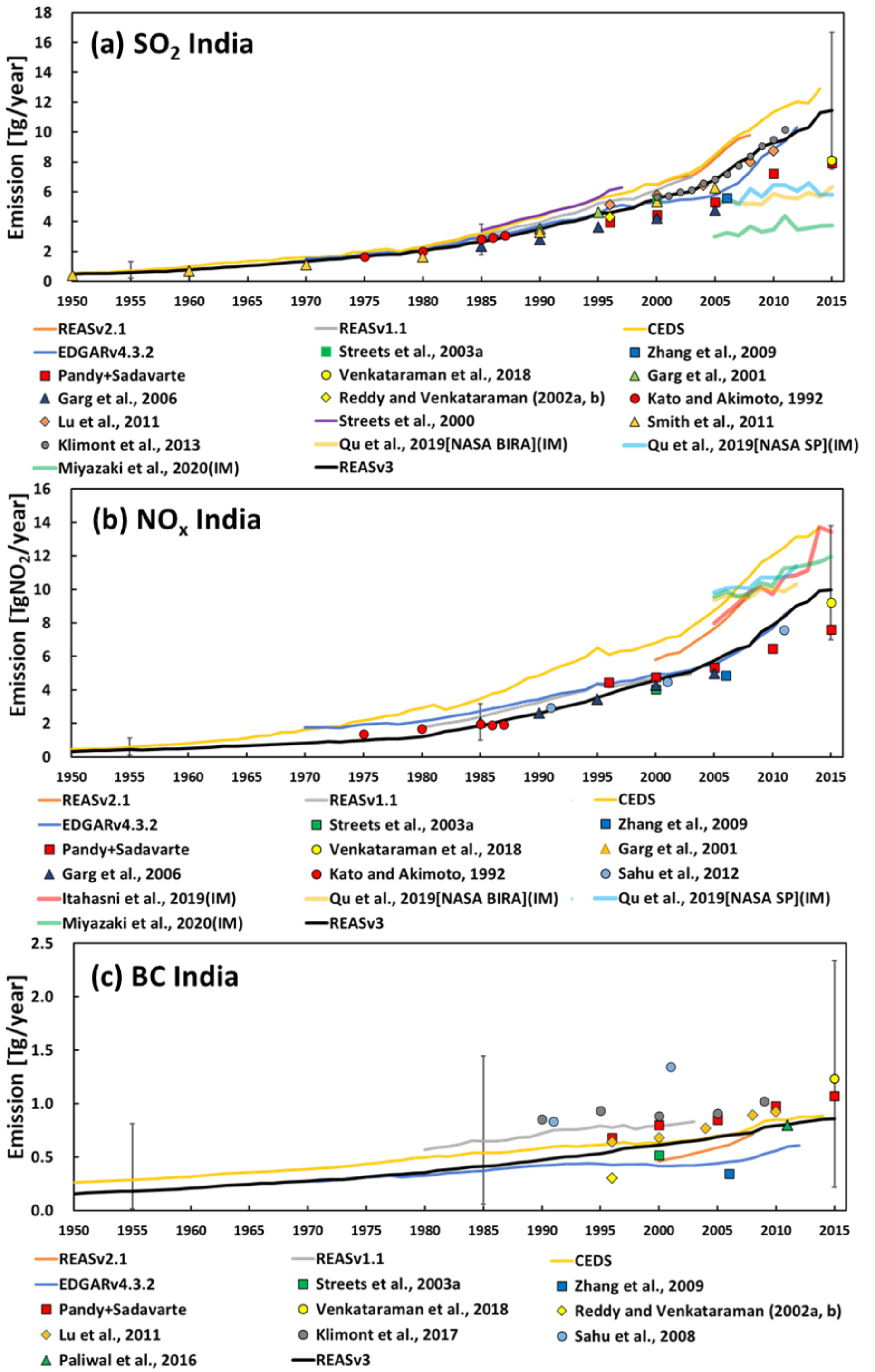
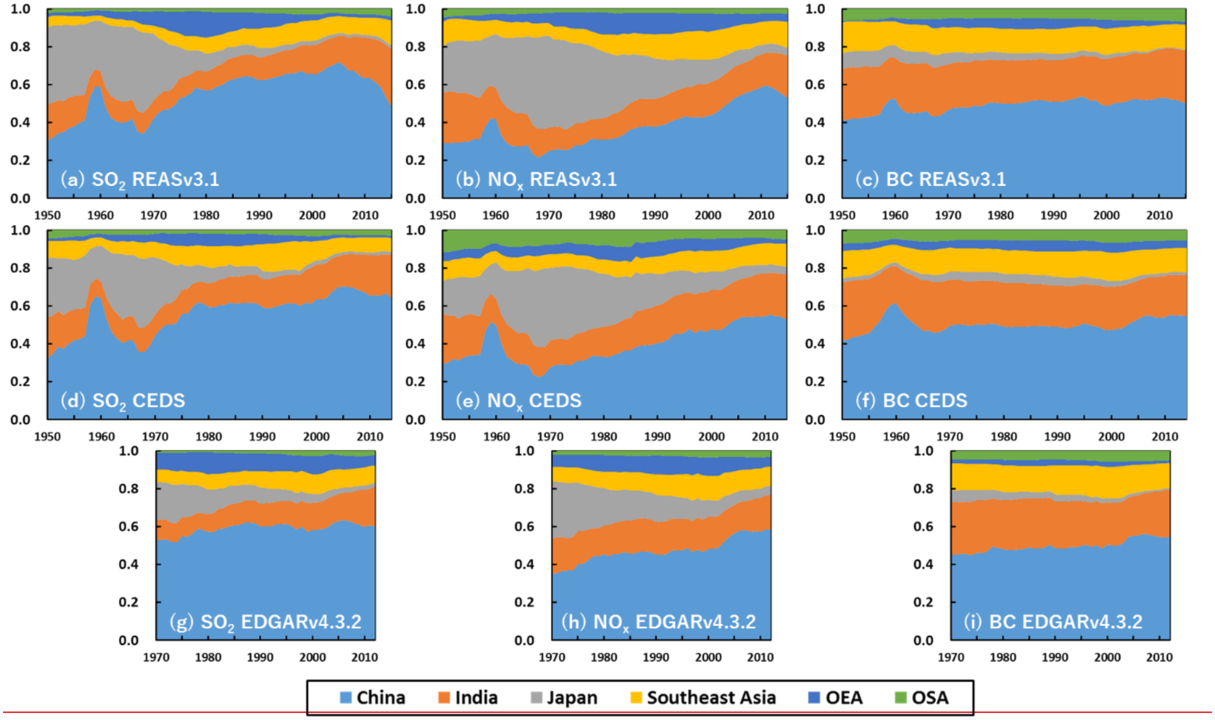


Figure 11. Comparison of (a) SO_2, (b) NO_x, and (c) BC emissions in India between REASv3 and other studies. Note that values of "Pandy+Sadavarte" are calculated from Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014). Emissions from domestic and fishing ships were excluded from REAS series, CEDS, EDGARv4.3.2, Garg et al. (2006) and Paliwal et al. (2016). IM

means estimates by inverse modeling. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.~~**Figure 11: Comparison of (a) SO₂, (b) NOₓ, and (c) BC emissions in India between REASv3.1 and other studies. Note that values of "Pandy+Sadavarte" are calculated from Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014).a あ**~~



(a) SO2 REASv3.1
(b) NOx REASv3.1
(c) BC REASv3.1
(d) SO2 CEDS
(e) NOx CEDS
(f) BC CEDS
(g) SO2 EDGARv4.3.2
(h) NOx EDGARv4.3.2
(i) BC EDGARv4.3.2
China
India
Japan
Southeast Asia
OEA
OSA

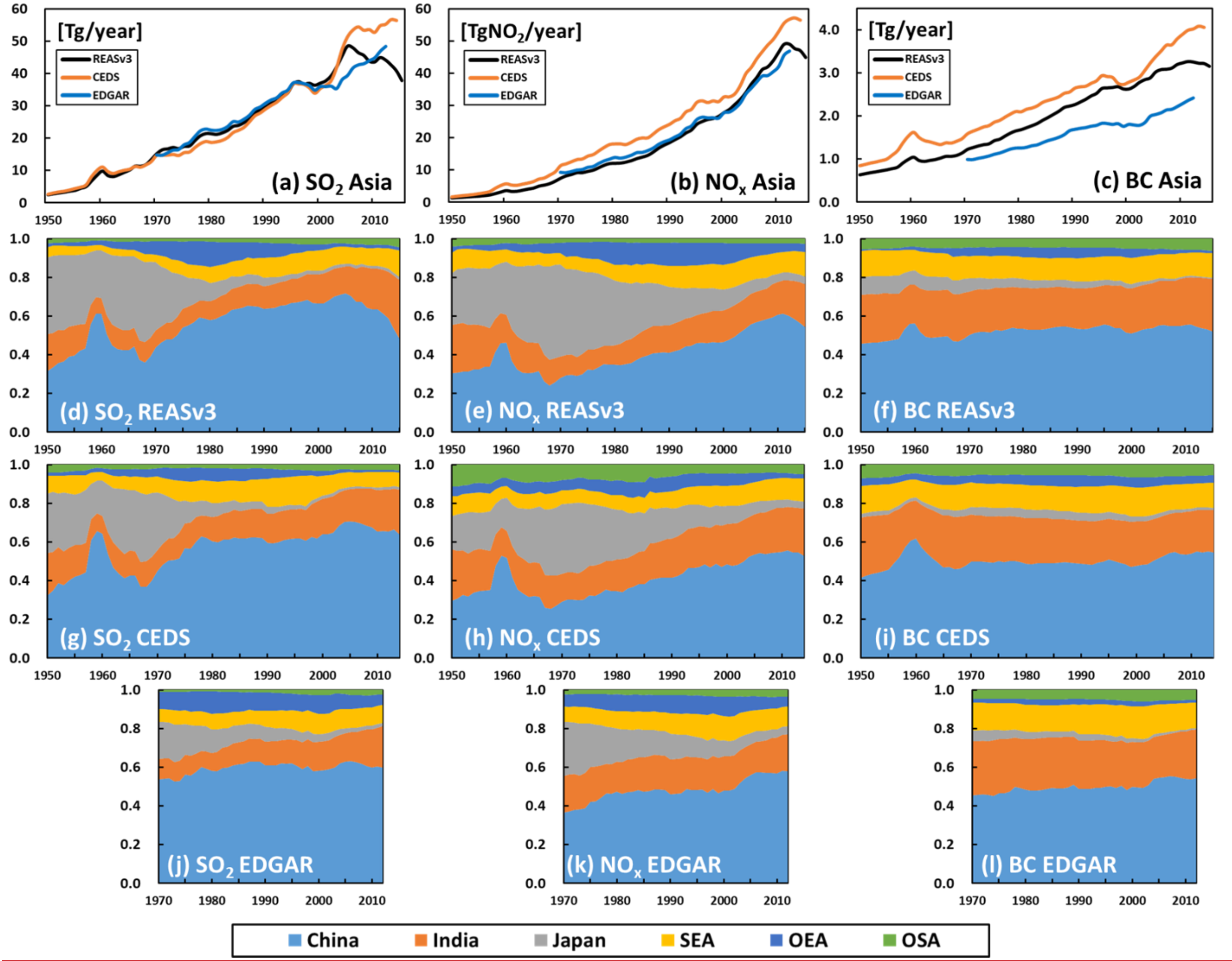


Figure 12. Comparison of trends of (a) SO_2, (b) NO_x and (c) BC emissions in Asia and relative ratios of emissions from China, India, Japan, SEA, OEA, and OSA for (d, g, j) SO_2, (e, h, k) NO_x, and (f, i, l) BC among (d, e, f) REASv3, (g, h, i) CEDS, and (j, k. l) EDGARv4.3.2. Note that periods of CEDS and EDGARv4.3.2 shown here are during 1950-2014 and 1970-2012, respectively. See Fig. 1 for definitions of SEA, OEA, and OSA.~~**Figure 12: Comparison of trends of relative ratios of emissions from China, India, Japan, Southeast Asia, East Asia other than China and Japan (OEA), and South Asia other than India (OSA) for (a, d, g) SO_2, (b, e, h) NO_x, and (c, f, i) BC among REASv3.1 (upper panels), CEDS (middle panels), and EDGARv4.3.2 (lower panels). Note that periods of CEDS and EDGARv4.3.2 shown here are during 1950-2014 and 1970-2012, respectively.**~~

Table 1.~~:~~ General information on REASv~~-~~3.~~1.~~

Item	Description
Species	SO_2, NO_x, CO, NMVOC, NH_3, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC
Years	1950–2015
Areas	East, Southeast, and South Asia
Emission sources	Fuel combustion in power plans, industry, transport, and domestic sectors; Industrial processes; Agricultural activities (fertilizer application and livestock); and Others (fugitive emissions, solvent use, human, etc.)
Spatial resolution	0.25 degree by 0.25 degree
Temporal resolution	Monthly
Data distribution	http://www.nies.go.jp/REAS/

Table 2. Emission inventories from other research works and officially opened data utilized in REASv3.

Other emission inventories and data sources	How utilized in REASv3
VOC Emission Inventory in Japan (MOEJ, 2017)	Evaporative emissions of NMVOC in Japan[a]
The National Air Pollutants Emission Service of the National Institute of Environmental Research (http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do)	Evaporative emissions of NMVOC in Republic of Korea[a]
REASv2.1 (Kurokawa et al., 2013; JPEC 2012a, b, c; 2014)	NH_3 emissions from agricultural sources in Japan[b]
REASv1.1 (Yamaji et al., 2004; Yan et al., 2003)	NH_3 emissions from agricultural sources in countries and regions other than Japan[b]
REASv2.1 (Kurokawa et al., 2013; JPEC 2012a, b, c; 2014)	Grid allocation factors for manure management[c] and road transport sectors for Japan[d]
EDGARv4.3.1 (Crippa et al., 2016)	Grid allocation factors for manure management[c] and road transport[d] sectors for countries and regions other than Japan

[a]See Sect. S5.3 of the Supplement. [b]See Sect. 2.4. [c]See Sect. S8.1 of the Supplement. [d]See Sect. 2.6.

~~Table 2: Summary of national emissions in 2015 for each species and total annual emissions in Asia in 1950, 1960, 1970, 1980, 1990, 2000, and 2010-2015 (Gg yr^{-1}).~~

Country	SO_2	NO_x [a]	CO	NMVOC	NH_3	CO_2 [b]	PM_{10}	$PM_{2.5}$	BC	OC
China	18144	23647	175656	29923	14053	11768	15166	10993	1660	2687
Japan	551	1825	3976	900	354	1277	135	106	16	16
Korea, Rep of	361	1203	1713	985	171	663	142	125	27	35
Korea, DPR	116	205	2809	143	92	29	99	54	15	18
Mongolia	107	126	1059	51	139	18	43	24	2.7	3.2
Taiwan	135	425	660	704	90	278	40	35	4.4	5.9
Brunei	4.0	16	29	45	3.8	6.1	4.8	1.5	0.2	0.0
Cambodia	57	78	1500	290	77	22	117	82	12	44
Indonesia	2856	2741	21006	6814	1589	643	1512	1173	220	690
Laos	201	43	387	94	67	11	43	28	4.6	14
Malaysia	238	715	1480	1012	162	226	149	83	13	15
Myanmar	153	158	2933	1093	620	58	187	171	35	128
Philippines	806	874	3544	1103	387	133	252	174	37	76
Singapore	92	104	74	299	8.4	46	16	11	1.0	1.1
Thailand	332	1140	7150	1598	540	316	409	302	47	158
Vietnam	435	568	6620	1641	746	243	532	349	64	185
Bangladesh	170	263	2465	765	879	108	402	224	40	112
Bhutan	3.3	11	283	64	9.2	4.7	28	21	4.0	14
India	11633	10632	65963	15481	9462	2919	6526	4709	929	2042
Nepal	40	89	2295	573	318	39	194	158	32	111
Pakistan	1230	598	8440	2150	1760	272	1032	670	121	381
Sri Lanka	92	263	1355	325	102	37	132	104	24	59
Afghanistan	24	114	360	71	251	9.4	24	20	10	8.5
Maldives	3.1	6.1	6.9	2.7	0.4	0.8	0.4	0.3	0.2	0.1
Asia[c] 1950	2485	1532	57246	8597	7280	1002	4966	4074	688	2465
Asia[c] 1960	9324	3802	94786	10892	8932	1976	11045	7262	1125	3330

~~Asia[c] 1970~~	~~14384~~	~~7567~~	~~113836~~	~~14398~~	~~11538~~	~~3064~~	~~14088~~	~~8779~~	~~1317~~	~~3824~~
~~Asia[c] 1980~~	~~20660~~	~~12112~~	~~161936~~	~~19861~~	~~15582~~	~~4490~~	~~18925~~	~~12366~~	~~1806~~	~~4823~~
~~Asia[c] 1990~~	~~29186~~	~~18625~~	~~208143~~	~~26603~~	~~20975~~	~~6514~~	~~24249~~	~~16846~~	~~2420~~	~~5831~~
~~Asia[c] 2000~~	~~36592~~	~~27126~~	~~241096~~	~~36916~~	~~25711~~	~~8940~~	~~28057~~	~~19904~~	~~2751~~	~~6004~~
~~Asia[c] 2010~~	~~43884~~	~~46174~~	~~320578~~	~~56423~~	~~30550~~	~~16764~~	~~28110~~	~~20166~~	~~3392~~	~~7095~~
~~Asia[c] 2011~~	~~45111~~	~~48741~~	~~323203~~	~~58893~~	~~30809~~	~~17742~~	~~28750~~	~~20517~~	~~3421~~	~~6998~~
~~Asia[c] 2012~~	~~44407~~	~~49059~~	~~322774~~	~~61230~~	~~31214~~	~~18193~~	~~28591~~	~~20502~~	~~3408~~	~~6939~~
~~Asia[c] 2013~~	~~42669~~	~~47935~~	~~319557~~	~~63100~~	~~31491~~	~~18894~~	~~28818~~	~~20614~~	~~3383~~	~~6858~~
~~Asia[c] 2014~~	~~40797~~	~~47653~~	~~318009~~	~~65179~~	~~31700~~	~~19154~~	~~28683~~	~~20513~~	~~3383~~	~~6857~~
~~Asia[c] 2015~~	~~37785~~	~~45845~~	~~311766~~	~~66126~~	~~31882~~	~~19125~~	~~27185~~	~~19617~~	~~3319~~	~~6801~~

Table 3. Summary of national emissions in 2015 for each species and total annual emissions in Asia in 1950, 1960, 1970, 1980, 1990, 2000, and 2010-2015 (Gg yr^{-1}).

Country	SO_2	NO_x[a]	CO	NMVOC	NH_3	CO_2[b]	PM_{10}	$PM_{2.5}$	BC	OC
China	18404	24318	165133	28189	14063	11941	15501	11342	1643	2860
India	11438	9969	64366	14286	9505	2959	7213	5052	858	1868
Japan	565	1687	3877	895	349	1300	129	89	17	13
Korea, D.P.R.	116	200	2663	134	92	29	106	56	11	18
Korea, Rep of	336	1120	1931	960	170	689	139	114	19	34
Mongolia	99	127	986	50	139	18	44	20	2.9	3.2
Taiwan	124	371	1027	770	85	281	45	37	6.9	7.3
Brunei	4.0	13	29	43	3.8	6.1	7.5	2.9	0.2	0.1
Cambodia	55	61	1087	212	78	22	115	69	9.0	32
Indonesia	2852	2463	20517	6130	1591	655	1606	1160	196	556
Laos	201	35	325	66	67	12	46	25	3.6	10
Malaysia	233	613	1288	936	163	230	206	119	14	12
Myanmar	154	121	2925	867	621	59	184	165	29	98
Philippines	786	767	3292	898	388	134	284	183	38	61
Singapore	87	89	76	302	6.4	46	81	62	1.2	0.5
Thailand	341	1137	5436	1543	542	320	522	363	49	125
Vietnam	436	507	6078	1552	747	250	587	362	59	146
Afghanistan	24	97	404	93	251	9.4	18	14	6.9	4.4
Bangladesh	171	305	2755	704	883	110	519	287	40	102
Bhutan	3.3	6.8	269	55	9.5	4.7	29	19	3.0	10
Maldives	3.1	4.1	9.4	3.7	0.4	0.8	0.2	0.2	0.1	0.0
Nepal	42	64	2381	533	321	40	207	161	26	89
Pakistan	1310	573	8576	2031	1772	273	1310	841	105	324
Sri Lanka	92	187	1382	374	103	37	135	98	19	49
Asia[c] 1950	2540	1339	51804	6551	7310	1005	5089	4162	630	2308
Asia[c] 1960	9880	3639	81220	8461	8968	2016	11405	7487	1040	3185
Asia[c] 1970	15287	7470	100368	11599	11579	3117	14770	9217	1221	3629
Asia[c] 1980	21425	12080	142102	16432	15632	4550	19900	13060	1680	4602
Asia[c] 1990	29721	18481	182418	22670	21035	6595	25427	17542	2264	5574

Asia[c] 2000	37074	27782	219516	33498	25775	9083	29461	20758	2626	5682
Asia[c] 2010	43635	46368	302562	52711	30621	17055	29880	21220	3233	6757
Asia[c] 2011	45003	48868	304900	55136	30878	18047	30540	21559	3266	6652
Asia[c] 2012	44227	48962	304396	57285	31283	18496	30414	21526	3254	6587
Asia[c] 2013	42725	47561	304484	58971	31559	19200	30649	21627	3227	6485
Asia[c] 2014	40864	46970	302718	60801	31770	19447	30469	21475	3219	6478
Asia[c] 2015	37876	44835	296809	61627	31950	19423	29034	20644	3155	6422

[a]Gg-NO_2 yr^{-1}.
[b]Tg yr^{-1}.
[c]Asia in this table include all target countries and sub-regions in REASv3.

~~[a]Gg NO$_2$ yr^{-1}.~~

~~[b]Tg yr^{-1}.~~

~~[c]Asia in this table include all target countries and sub-regions in REASv3.1.~~

Table 4. Uncertainties [%] of emissions in China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. See Fig. 1 for definitions of SEA OEA, and OSA.

	SO_2	NO_x	CO	NMVOC	NH_3	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
1955										
China	±85	±167	±291	±277	±174	±133	±253	±315	±334	±365
India	±96	±122	±265	±295	±161	±116	±257	±294	±277	±314
Japan	±59	±62	±157	±135	±141	±49	±94	±117	±170	±270
SEA	±134	±153	±260	±272	±169	±126	±291	±307	±323	±317
OEA	±73	±88	±146	±184	±148	±59	±120	±157	±157	±262
OSA	±70	±112	±272	±270	±168	±110	±219	±281	±310	±345
1985										
China	±36	±53	±157	±150	±139	±39	±101	±129	±182	±250
India	±40	±60	±196	±212	±135	±58	±160	±201	±191	±259
Japan	±30	±31	±44	±50	±93	±14	±72	±71	±53	±67
SEA	±40	±56	±185	±162	±141	±56	±157	±191	±218	±259
OEA	±48	±70	±72	±78	±113	±27	±80	±82	±88	±102
OSA	±36	±44	±144	±137	±134	±33	±108	±137	±176	±248
2015										
China	±40	±35	±73	±76	±82	±19	±83	±94	±111	±193
India	±41	±35	±136	±115	±111	±27	±120	±151	±133	±233
Japan	±34	±32	±45	±63	±103	±13	±68	±74	±58	±100
SEA	±46	±38	±124	±86	±115	±25	±125	±155	±161	±232
OEA	±38	±60	±67	±63	±94	±19	±69	±85	±82	±168
OSA	±40	±34	±87	±73	±93	±19	±96	±112	±124	±211

~~Table 3: Uncertainties [%] of emissions in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in 2015, 1985, and 1955.~~

	SO_2	NO_x	CO	NMVOC	NH_3	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
2015										
China	±30	±35	±81	±75	±80	±28	±94	±102	±125	±220
India	±34	±39	±137	±122	±99	±40	±128	±157	±156	±240
Japan	±29	±25	±35	±31	±92	±23	±61	±61	±54	±74
SEA	±39	±44	±121	±102	±103	±39	±141	±173	±184	±242
OEA	±37	±68	±84	±78	±93	±33	±72	±84	±88	±204
OSA	±31	±34	±91	±79	±87	±28	±105	±119	±140	±233
1985										
China	±35	±61	±146	±184	±123	±59	±110	±141	±206	±295
India	±33	±61	±202	±231	±116	±89	±168	±208	±214	±275
Japan	±25	±30	±44	±35	±80	±23	±65	±60	±55	±58
SEA	±37	±59	±176	±181	±122	±87	±184	±223	±240	±277
OEA	±43	±73	±79	±90	±96	±41	±74	±72	±91	±110
OSA	±34	±47	±138	±164	±119	±51	±117	±150	±196	±285
1955										
China	±78	±161	±282	±300	±150	±180	±272	±341	±379	±420
India	±72	±118	±259	±306	±138	±156	±262	±296	±296	±326
Japan	±63	±60	±141	±203	±325	±58	±85	±105	±153	±251
SEA	±88	±141	±234	±281	±147	±169	±301	±312	±331	±326
OEA	±80	±94	±126	±186	±123	±71	±111	±145	±140	±284
OSA	±66	±110	±258	±287	±149	±147	±233	±297	±338	±378

(5) Revision of the supplementary materials

For the ACPD paper, one supplement including both supplementary figures and tables was provided. In the revised manuscript, as mentioned above, following two new supplements were created:

- Supplementary information and data related to methodology of REASv3
- Differences between REASv3.2 and REASv3.1

Then, we divided the first supplement into figure and table parts and also prepared the title page. Finally, five files are provided as supplements of the revised main manuscript.

For the supplementary figures, revisions from the ACPD version are as follows:

- All figures were recreated using the updated data (REASv3.2).
- Following revisions were done in Figs. S14-S19:
    - Results of inverse modeling and two bottom-up inventories were added and corresponding references were added.
    - Error bars indicating the uncertainty range in 1955, 1985, and 2015 were added.
    - Markers and lines in the figures were reconsidered and revised.
- Comparisons of total emissions in Asia among REASv3, CEDS, and EDGARv4.3.2 were added to Figs. 12 and S20.

For the supplementary tables, revisions from the ACPD version are as follows:

- Table for target countries and regions (Table S1 for the ACPD version) was moved to the new supplement for methodology (Table 2.3 in the Supplement) and thus, Tables S2-S4 in the ACPD version were shifted to Tables S1-S3.
- All data in Tables S1-S3 in the new supplementary tables were recalculated using the updated data (REASv3.2).

From next page, the five supplementary materials are provided as follows:

- Title page
- Supplementary information and data related to methodology of REASv3
- Supplementary figures
- Supplementary tables
- Differences between REASv3.2 and REASv3.1

Supplement of

Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3

Junichi Kurokawa and Toshimasa Ohara

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- Supplementary information and data related to methodology of REASv3
- Supplementary figures
- Supplementary tables
- Differences between REASv3.2 and REASv3.1

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Supplementary information and data related to methodology of REASv3

Contents

S1. Introduction

This document provides detailed information related to methodologies of Regional Emission inventory in ASia (REAS) version 3 (hereafter REASv3 in this document) developed as a supplementary material of the main manuscript entitled “Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3”. In this document, first and second versions of REAS are often cited and expressed as REASv1 (Ohara et al., 2007) and REASv2 (Kurokawa et al., 2013), respectively. The framework of REASv3 such as target species, countries and regions, and emission sources was summarized in Sect. 2. Sects. 3, 4, 5, 6, and 7 provide details of activity data and emission factors including settings of emission controls for stationary combustion, industrial production, non-combustion sources of NMVOC, road transport, and other transport, respectively. The details related to methodology for non-combustion sources of NH_3 were given in Sect. 8. Grid allocation and monthly variation factors for spatial and temporal distribution were described in Sect. 9. In Sect. 10, details of methodology and settings for estimation of uncertainties were provided.

Note that this document is for REASv3.2 which is an updated version of REASv3.1 (Kurokawa et al., 2019). The differences between REASv3.2 and REASv3.1 and causes of the discrepancies were provided in another document entitled “Differences between REASv3.2 and REASv3.1” developed as an additional supplement of the main manuscript.

S2. Framework of REASv3

S2.1 Target species

Target species of REASv3 are summarized in Table 2.1. In REASv3, NMVOC species were divided into 19 chemical species categories as presented in Table 2.2. Codes of each species used in emission tables and gridded data of REASv3 are also provided in the tables.

Table 2.1. Target species of REASv3.

Species code	Species
SO2	Sulfur dioxide
NOX	Nitrogen oxides (as NO_2)
CO_	Carbon monoxide
NMV	Non-methane volatile organic compounds
NH3	Ammonia
CO2	Carbon dioxide
PM10_	Primary PM_{10}
PM2.5	Primary $PM_{2.5}$
BC_	Black carbon
OC_	Primary Organic carbon

Table 2.2. NMVOC species categories defined in REASv3.

Species number code	NMVOC species
01	Ethane
02	Propane
03	Butanes
04	Pentanes
05	Other Alkanes
06	Ethylene
07	Propene
08	Terminal Alkenes
09	Internal Alkenes
10	Acetylene
11	Benzene
12	Toluene
13	Xylenes

14	Other Aromatics
15	Formaldehyde
16	Other Aromatics
17	Ketones
18	Halocarbons
19	Others
20	Total

S2.2 Target years

Target years of REASv3 are 1950-2015 (each year). In future updated versions, the oldest target year is basically fixed, but data in later years (after 2016) are planned to be added.

S2.3 Target countries and regions

Table 2.3 provides list of countries and sub-regions included in the inventory domain of REASv3. Codes of region, countries, and sub-regions used in the main manuscript, emission tables and gridded data of REASv3 are also provided in the table.

Table 2.3. Region, country, and sub-region included in the inventory domain of REASv3 with codes used in the main manuscript and files of emission tables and gridded data provided from the REAS website (https://www.nies.go.jp/REAS/).

Region name/ Region code	Country name: Sub-region name	Country and sub-region code CCCRR CCC: Country code RR: Sub-region code
China/ CHN	China: Whole Country	CHNWC
	China: Beijing	CHNBJ
	China: Tianjin	CHNTJ
	China: Hebei	CHNHE
	China: Shanxi	CHNSX
	China: Inner Mongolia	CHNNM
	China: Liaoning	CHNLN
	China: Jilin	CHNJL
	China: Heilongjiang	CHNHL

	China: Shanghai	CHNSH
	China: Jiangsu	CHNJS
	China: Zhejiang	CHNZJ
	China: Anhui	CHNAH
	China: Fujian	CHNFJ
	China: Jiangxi	CHNJX
	China: Shandong	CHNSD
	China: Henan	CHNHA
	China: Hubei	CHNHB
	China: Hunan	CHNHN
	China: Guangdong	CHNGD
	China: Guangxi	CHNGX
	China: Hainan	CHNHI
	China: Chongqing	CHNCQ
	China: Sichuan	CHNSC
	China: Guizhou	CHNGZ
	China: Yunnan	CHNYN
	China: Tibet	CHNXZ
	China: Shaanxi	CHNSN
	China: Gansu	CHNGS
	China: Qinghai	CHNQH
	China: Ningxia	CHNNX
	China: Xinjiang	CHNXJ
	China: Hong Kong	CHNHK
	China: Macau	CHNMC
India/ IND	India: Whole Country	INDWC
	India: Andhra Pradesh	INDAP
	India: Bihar, Jharkhand	INDBJ
	India: North East (Arunachal Pradesh/Assam/Manipur/ Meghalaya/Mizoram/Nagaland/Sikkim/Tripura)	INDAN
	India: Gujarat	INDGU
	India: Haryana	INDHA
	India: Karnataka/Goa	INDKG
	India: Kerala	INDKE
	India: Madhya Pradesh/Chhattisgarh	INDMC

	India: Maharashtra	INDMA
	India: Orissa	INDOR
	India: Punjab/Chandigarh	INDPU
	India: Rajasthan	INDRA
	India: Tamil Nadu	INDTN
	India: Utter Pradesh/Uttaranchal	INDUU
	India: West Bengal	INDWB
	India: Himachal Pradesh/Jammu and Kashmir	INDHJ
	India: Delhi	INDDE
Japan/ JPN	Japan: Whole Country	JPNWC
	Japan: Hokkaido-Tohoku (Hokkaido/Aomori/Iwate/ Miyagi/Akita/Yamagata/Fukukshima)	JPNHT
	Japan: Kanto (Ibaraki/Tochigi/Gunma/Saitama/Chiba/ Tokyo/Kanagawa)	JPNKN
	Japan: Chubu (Niigata/Toyama/Ishikawa/Fukui/ Yamanashi/Nagano/Gifu/Shizuoka/Aichi)	JPNCB
	Japan: Kinki (Mie/Shiga/Kyoto/Osaka/Hyogo/Nara/ Wakayama)	JPNKK
	Japan: Chugoku-Shikoku (Tottori/Shimane/Okayama/ Hiroshima/Yamaguchi/Tokushima/Kagawa/Ehime/Kochi)	JPNCS
	Japan: Kyushu-Okinawa (Fukuoka/Saga/Nagasaki/ Kumamoto/Oita/Miyazaki/Kagoshima/Okinawa)	JPNKO
Other East Asia / OEA	Democratic People's Republic of Korea, Whole Country	PRKWC
	Republic of Korea, Whole Country	KORWC
	Mongolia: Whole Country	MNGWC
	Taiwan: Whole Country	TWNWC
Southeast Asia/ SEA	Brunei: Whole Country	BRNWC
	Cambodia: Whole Country	KHMWC
	Indonesia: Whole Country	IDNWC
	Laos: Whole Country	LAOWC
	Malaysia: Whole Country	MYSWC
	Myanmar: Whole Country	MMRWC
	Philippines: Whole Country	PHLWC
	Singapore: Whole Country re	SGPWC
	Thailand: Whole Country	THAWC

	Vietnam: Whole Country	VNMWC
Other South Asia/ OSA	Afghanistan: Whole Country	AFGWC
	Bangladesh: Whole Country	BGDWC
	Bhutan: Whole Country	BTNWC
	Maldives: Whole Country	MDVWC
	Nepal: Whole Country	NPLWC
	Pakistan: Whole Country	PAKWC
	Sri Lanka: Whole Country	LKAWC

S2.4 Target emission sources

S2.4.1 Combustion sources

Table 2.4 provides list of sub-sector categories of combustion sources defined in REASv3. Aggregated sector categories used in the main manuscript and emission tables of REASv3 are presented as "Sector code". IEA codes show relationships between sub-sector categories of REASv3 and the International Energy Agency (IEA) World Energy Balances (IEAWEB) (IEA, 2017). Fuel types defined in REASv3 are provided in Sect S3.1.1. See Sects. S3, S6, and S7 for details of stationary combustion, road transport, and other transport sectors, respectively.

Several emission sources related to transformation sectors except for power plants were included in Table 2.4. Sources categorized as energy sectors in IEAWEB are only considered as combustion sources. For coke ovens (not as the energy sector), emissions were estimated based on coal input for SO_2 and NO_x and coke production for CO, NMVOC, CO_2, and PM species. In REASv3, for coke ovens as energy transformation sectors, contributions from both combustion and non-combustion processes were included in the emissions. In other words, their emissions were not estimated separately. Similarly, the following sources include both combustion and non-combustion emissions which were not estimated separately:

- Charcoal production plants
- Manufacture of other solid fuels
- Gas works

In addition, CO emissions from pig iron, crude steel, and sinter production for all countries, those from brick production except for China, Japan, Republic of Korea, and Taiwan, emissions of PM species from sinter and pig iron production for China, and those from brick production for all countries estimated based on their production amounts include contributions from both combustion and non-combustion sources (not estimated separately).

Table 2.4. Sub-sector categories of combustion sources considered in REASv3 with sector codes used in the main manuscript and emission tables of REASv3 and IEA codes showing relationships between sub-sector categories of REASv3 and the IEAWEB.

Sector code	Sub-sector category	IEA code
Power Plants/ PP	Power plants (point sources/area sources)	MAINELEC/AUTOELEC/ MAINCHP/AUTOCHP/ MAINHEAT/AUTOHEAT/ THEA/TBOILER/TELE
	Power plants (energy)	EPOWERPLT
Industry/ IND	Coke ovens	TCOKEOVS
	Charcoal production plants	TCHARCOAL
	Manufacture of other solid fuels	TPATFUEL/TBKB/TNONSPEC
	Coke ovens (energy)	ECOKEOVS
	Charcoal production plants (energy)	ECHARCOAL
	Manufacture of other solid fuels (energy)	EMINES/EPATFUEL/EBKB/ ENONSPEC
	Petroleum refineries (energy)	EREFINER
	Manufacture of other liquid fuels (energy)	EOILGASEX/ECOALLIQ/EGTL
	Gas works	TGASWKS
	Gas works (energy)	EGASWKS
	Manufacture of other gaseous fuels (energy)	ELNG/EGTL
	Chemical and petrochemical industry	CHEMICAL
	Iron and steel industry	IRONSTL
	Blast furnace	TBLASTFUR
	Blast furnace (energy)	EBLASTFUR
	Non-ferrous metal industry	NONFERR
	Cement industry	NONMET
	Lime industry	
	Brick industry	
	Other non-metallic minerals industries	
	Construction industry	CONSTRUC
	Transport equipment industry	TRANSEQ
	Machinery industry	MACHINE
	Mining and quarrying industry	MINING
	Food and tobacco industry	FOODPRO

	Paper, pulp and printing industry	PAPERPRO
	Wood and wood products industry	WOODPRO
	Textile and leather industry	TEXTILES
	Other industries	INONSPEC
Road transport/ ROAD	Road transport	ROAD
Other transport/ OTRA	Rail	RAIL
	Pipeline transport	PIPLINE
	Other transport*[1]	TRNONSPE
Residential/ RESI	Residential	RESIDENT
Other domestic/ ODOM	Commercial and public services	COMMPUB
	Agriculture*[2]	AGRICULT
	Others	ONONSPEC

*[1]Aviation and navigation (both for domestic and international) are not included.

*[2]Forestry is included, but fishing is not included.

S2.4.2 Non-combustion sources: Industrial production and other transformation

Table 2.5 provides list of sub-sector categories of non-combustion sources defined in REASv3 with target species and notes for each sub-sector category. See Sect. S4 for details of industrial processes and other transformation. See Sects. S5 and S8 for industrial processes related to NMVOC and NH_3, respectively. Note that, as described in Sect S2.4.1, non-combustion emissions from coke production, those of CO from pig iron, crude steel, and sinter productions (for all countries and regions) and from brick production (except for China, Japan, Republic of Korea, and Taiwan), and those of PM species from sinter and pig iron production (for China) and from brick production (for all countries) were not estimated separately. For these sources, estimated emission in REASv3 include contributions from both combustion and non-combustion processes.

Table 2.5. Sub-sector categories of non-combustion sources from industrial production and other transformation considered in REASv3.

Sub-sector category	Target species	Notes
Pig iron production	CO, PM species	Iron and steel industry
Crude steel production	CO, NMVOC, PM species	

Sinter production	CO, PM species	
Rolled steel production	NMVOC	
Copper production	SO_2, PM species	Non-ferrous metal industry
Zinc production	SO_2, PM species	
Lead production	SO_2, PM species	
Almina production	SO_2, PM species	
Aluminium production	SO_2, PM species	
Cement production	CO_2, PM species	Non-metallic minerals industry
Lime production	CO_2, PM species	
Brick production	PM species	
Sulphuric acid production	SO_2	Inorganic chemicals industry
Carbon black production	NMVOC, PM species	
Ethylene production	NMVOC	Organic chemicals industry
Polyethylene production	NMVOC	
Styrene production	NMVOC	
Polystyrene production	NMVOC	
Polyvinylchloride production	NMVOC	
Propylene production	NMVOC	
Polypropylene production	NMVOC	
Polyvinylchloride processing	NMVOC	
Polystyrene processing	NMVOC	
Bread production	NMVOC	Other industries considered for NMVOC
Beer production	NMVOC	
Asphalt production	NMVOC	
Pulp and paper production	NMVOC	
Ammonia	NH_3	Synthetic fertilizer industry considered for NH_3
Ammonium nitrate	NH_3	
Urea	NH_3	
Coke production	CO, NMVOC, CO_2, PM species	Manufacture of solid fuels
Petroleum refineries	SO_2, NMVOC, PM species	Manufacture of liquid fuels For NMVOC, contributions were included in extraction processes. See Sect. S2.4.3.

S2.4.3 Non-combustion sources of NMVOC

Non-combustion sources for NMVOC emissions considered in REASv3 are extraction processes, solvent use, industrial processes, waste disposal and evaporative emissions from road vehicles. Sub-categories of extraction processes and solvent use are summarized in Tables 2.6 and 2.7. Definitions of the sub-sectors are the same as with those of Klimont et al. (2002a). See Table 2.5, Sect. 5.1.7 and Sect. S6.3 for industrial processes, waste disposal, and evaporative emissions from road vehicles, respectively. See Sect. S5 for details of non-combustion sources of NMVOC.

Table 2.6. Sub-sector categories of extraction processes considered in REASv3.

Sub-category
Gas production
Gas distribution
Crude oil production
Crude oil handling
Petroleum refineries[a]
Service station
Transport and depots

a. Except for NMVOC, contributions were included in industrial processes. See Sect. S2.4.2.

Table 2.7. Sub-sector categories of solvent use considered in REASv3.

Sub-category
Dry cleaning
Decreasing operation
Vehicle treatment
Domestic use of solvents
Asphalt blowing
Paint production
Ink production
Tire production
Synthetic rubber production
Textile industry
Preservation of wood
Adhesive application
Printing[a]
Paint application[b]

a. Contributions from following activities were included: packing offset printing, publication, and screen printing were included. b. Contributions from following purposes were included: architecture, domestic usage, automobile manufacture, vehicle refinishing, and other industrial application.

S2.4.4 Non-combustion sources of NH_3

Non-combustion sources for NH_3 emissions considered in REASv3 are manure management of livestock, fertilizer application, industrial processes, human, and latrines as summarized in Table 2.8. See Sect. S8 for details of non-combustion sources of NH_3.

Table 2.8. Sub-sector categories of non-combustion sources of NH_3 considered in REASv3.

Sub-category
Manure management[a]
Fertilizer application[b]
Industrial processes[c]
Human[d]
Latrines

a. Contributions from manure management including housing, storage and yards were included. Those from manure applied to soils were included in fertilizer application. b. Contributions from both synthetic fertilizer and animal manure used as fertilizer were included. c. See Sect. S2.4.2. d. Contributions from perspiration and respiration were included.

S2.5 Spatial and temporal resolution

In REASv3, only large power plants are treated as point sources and gridded data of other emission sources are provided with a horizontal resolution of 0.25° × 0.25°. For temporal resolution, monthly emissions are estimated in REASv3 by allocating annual emissions to each month using monthly proxy data. Details of methodologies and data used for spatial and temporal allocation are described in Sect. S9.

Table 2.9 provides sub-sector categories included in aggregated sector codes for gridded data in REASv3.

Table 2.9. Sector codes for gridded data in REASv3 and sub-sector categories included in each code.

Sector categories code	Sub-sector categories included in each sector code
POWER_PLANTS_POINT	Power plants (points) in Table 2.4
POWER_PLANTS_NON-POINT	Power plants (area sources and energy) in Table 2.4
INDUSTRY	Combustion sources of industry sector in Table 2.4
	Non-combustion sources of industrial production and other transformation sector in Table 2.5
ROAD_TRANSPORT	Road transport sector in Table 2.4
	Evaporative NMVOC emissions from road vehicles described in Sect. S6.3
OTHER_TRANSPORT	Other transport sector in Table 2.4
DOMESTIC	Residential and other domestic sectors in Table 2.4
EXTRACTION	NMVOC emissions from extraction processes in Table 2.6
SOLVENTGS	NMVOC emissions from solvent use in Table 2.7
WASTE	NMVOC emissions from waste disposal described in Sect. S5.1.7
MANURE_MANAGEMENT	NH_3 emissions from manure management described in Sect. S8.1
FERTILIZER	NH_3 emissions from fertilizer application described in Sect. S8.2
MISC	NH_3 emissions from human and latrines described in Sects. S8.4 and S8.5.

S3. Stationary combustion

S3.1 Activity data

S3.1.1 Definition of fuel types

Table 3.1 describes fuel types considered in stationary combustion sources of REASv3. Emissions of air pollutants were estimated individually for each fuel type. In Fig. 4 of the main manuscript and Figs. S2, S4, S6, S8, S10, and S12 of the supplement, fuel types are aggregated to several categories. Definition of the categories are also provided in Table 3.1. For each fuel type, definitions are mostly the same as those of the International Energy Agency (IEA) World Energy Balances (IEAWEB) (IEA, 2017). Exceptions are "Raw coal", "Cleaned coal", "Other washed coal", and "Other coking products" which are defined only for China in the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017). Definition of "Bituminous coal", "Kerosene", and “Diesel oil” in Table 3.1. is the same as that of "Other bituminous coal", "Other kerosene", and “Gas/diesel oil excl. biofuels” of IEAWEB, respectively. For hard (brown) coal, if there is no detailed information, corresponding fuel type is considered as "Bituminous coal" ("Lignite"). For other fuel types, emissions from combustion were ignored in REASv3.

Table 3.1. List of detailed fuel types considered in REASv3 and definition of aggregated categories used in the main manuscript and the supplement.

Aggregated categories (code)	Aggregated categories (description)	Detailed fuel types
COAL	Primary coal	Coking coal
		Anthracite
		Bituminous coal
		Raw coal
		Cleaned coal
		Other washed coal
		Sub-bituminous coal
		Lignite
DC	Secondary coal	Coke oven coke
		Gas coke
		Coal tar
		Patent fuel
		Brown coal briquettes (BKB)
		Other coking products
NGAS	Natural gas	Natural gas
OGAS	Other gas fuels	Gas works gas
		Coke oven gas
		Blast furnace gas
		Other recovered gases
LF	Light oil fuels	Refinery gas
		Liquefied petroleum gas (LPG)
		Natural gas liquids
		Motor gasoline
		Naphtha
		Kerosene
MD	Diesel oil	Diesel oil
HF	Heavy oil fuels	Crude oil
		Heavy fuel oil
		Petroleum coke
		Other oil products
BF	Biofuel	Fuelwood
		Crop Residue

		Animal waste Biogas Biogasoline Biodiesels Charcoal
OTH	Other fuels	Municipal waste (renewable) Municipal waste (non-renewable) Industrial waste

S3.1.2 Data sources of fuel consumption and assumptions to estimate missing historical data

In REASv3, fuel consumption data were primarily obtained from IEAWEB, CESY, the United Nations (UN) Energy Statistics Database (UN, 2016), and UN data, which is a web-based data service of the UN (http://data.un.org/). However, all these sources do not include data for the entire target period of REASv3, that is from 1950-2015. Furthermore, past data for sectors do not contain as many categories. In this sub-section, data sources and assumptions for estimating missing historical data used in REASv3 are summarized in Table 3.2 including how to distribute total or sub-total data to detailed sub-sectors and how to extrapolate data until 1950. Note that descriptions for fuel consumption data in transport sector are also included in this sub-section.

Table. 3.2. Data sources and assumptions for estimating missing historical data used in REASv3 for each country and region.

(a) China

Data sources and treatments	● Fuel consumption for each region except for Tibet, Hong Kong and Macau were obtained from CESY during 1985-2015 and those before 1984 were extrapolated to 1950 using data for whole China during 1950-2015. See Sect. S3.1.3 for regional fuel consumption data in China. ● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950. Those of Tibet were taken from REASv2 (based on GAINS ASIA at that time) during 2000-2008 and extrapolated using data of whole country. See (n) and (o) of this sub-section for Hong Kong and Macau, respectively.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Energy industry own use sector: ✧ Data of bituminous coal and natural gas before 1989 were distributed to sub-sectors based on relative ratios of fuel consumption data in 1990. ✧ Fuel consumption data of coke oven gas in 1990 were extrapolated to 1980 using trends of coke oven gas production in IEAWEB during 1980-1990 and then, extrapolated to 1971 based on trends of coke oven coke production in IEAWEB during 1971-1980. ➢ Industry sector: ✧ Data of coking coal, gas works gas, coke oven gas, refinery gas, and LPG/other bituminous coal and crude oil/natural gas, other kerosene, diesel oil, and heavy fuel oil before 1989/1984/1979 were distributed to sub-sectors based on relative ratios in 1990/1985/1980. ✧ Fuel consumption data of coke oven gas in 1980 were extrapolated to 1971 using trends of coke oven gas production in IEAWEB during 1971-1980. ➢ Transport sector: ✧ Data of diesel oil before 1989 were distributed to road transport, domestic navigation, and agriculture/forestry based on relative ratios of corresponding fuel consumption in 1990. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(b) India

<table>
<tr><td>Data sources and treatments</td><td>● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
● See Sect. S3.1.4 for regional fuel consumption data in India.</td></tr>
<tr><td>Assumptions for estimating missing historical data</td><td>● No major modifications were done for IEAWEB during 1971-2015.
● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.</td></tr>
</table>

(c) Japan

<table>
<tr><td>Data sources and treatments</td><td>● Data of whole country were taken from IEAWEB during 1960-2015 and extrapolated to 1950.
● See Sect. S3.1.5 for regional fuel consumption data in Japan.</td></tr>
<tr><td>Assumptions for estimating missing historical data</td><td>● Assumptions for modifying IEAWEB during 1960-2015 are as follows:
➢ Industry sector:
✧ Data of hard coal and coke oven coke/natural gas and LPG/crude oil/heavy fuel oil before 1974/1981/1965/1969 were distributed to sub-sectors based on relative ratios of fuel consumption data in 1975/1982/1966/1970.
➢ Residential and other sectors:
✧ Data of heavy fuel oil before 1969 were distributed to sub-sectors based on relative ratios in 1970.
➢ Other kerosene and diesel oil:
✧ Data of total final consumption before 1969 were distributed to sub-sectors based on relative ratios in 1970.
● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950 except for following procedures:
➢ Consumption of hard coal, brown coal, patent fuel, coke oven coke, gas works gas, natural gas, and primary solid biofuels in residential sector were extrapolated to 1950 using the Historical Statistics of Japan (Japan Statistical Association, 2006).
➢ Consumption of primary solid biofuels in paper, pulp and printing industry before 1981 were extrapolated to 1950 based on trends of production amounts of paper and pulp in Japan (Economy, Trade and Industry Statistics Association, 1998).</td></tr>
</table>

(d) Republic of Korea

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Industry sector: ✧ Data of coke oven coke/other kerosene, diesel oil, and heavy fuel oil/natural gas before 2001/1980/1992 were distributed to sub-sectors based on relative ratios of fuel consumption data in 2002/1981/1993. ➢ Transport and other sectors: ✧ Data of diesel oil and heavy fuel oil before 1980 were distributed to sub-sectors based on relative ratios in 1981. ➢ Residential and other sectors: ✧ Data of primary solid biofuels before 1989 were distributed to sub-sectors based on relative ratios in 1990. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(e) Taiwan

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Residential and other sectors: ✧ Data of diesel oil/heavy fuel oil before 1979/1981 were distributed to sub-sectors based on relative ratios of fuel consumption data in 1980/1982. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(f) Indonesia

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Industry sector: ✧ Data of other bituminous coal and sub-bituminous coal before 1999 were distributed to sub-sectors based on relative ratios of consumption data of sub-bituminous coal in 2000. ✧ Data of natural gas/diesel oil and heavy fuel oil before 1980/1988 were distributed to sub-sectors based on relative ratios of fuel consumption data in 1981/1989. ✧ Fuel consumption data of primary solid biofuels in 1990 were extrapolated to 1971 using trends of primary solid biofuels consumption data in the other sector in IEAWEB during 1971-1990. ➢ Transport, residential and other sectors: ✧ Data of heavy fuel oil after 2000 were distributed to sub-sectors based on relative ratios in 1999. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(g) Myanmar

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Industry sector: ✧ Data of other bituminous coal/diesel oil before 2010/2011 were distributed to sub-sectors based on relative ratios of fuel consumption data in 2011/2012. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(h) Philippines

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Industry sector: ✧ Data of diesel oil and heavy fuel oil before 1979 were distributed to sub-sectors based on relative ratios of fuel consumption data in 1980. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(i) Singapore

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Residential and other sectors: ✧ Data of natural gas before 2005 were distributed to sub-sectors based on relative ratios of fuel consumption data in 2006. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(j) Thailand

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Industry sector: ✧ Data of other bituminous coal/natural gas before 1988/2001 were distributed to sub-sectors based on relative ratios of fuel consumption data in 1989/2002. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(k) Vietnam

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● Assumptions for modifying IEAWEB during 1971-2015 are as follows: ➢ Industry ✧ Data of anthracite, diesel oil and heavy fuel oil during 1980-2009 were distributed to sub-sectors based on relative ratios of corresponding fuel consumption data in 2010. ✧ Data of natural gas before 2009 were distributed to sub-sectors based on relative ratios in 2010. ✧ Data of other bituminous coal and lignite before 2009 were distributed to sub-sectors based on relative ratios of anthracite consumption data in 2010. ✧ Data of other bituminous coal and sub-bituminous coal after 2011 were distributed to sub-sectors based on relative ratios of anthracite consumption data in corresponding years of 2011-2015. ➢ Hard coal, diesel oil, and heavy fuel oil ✧ Data of total final consumption before 1979 were distributed to sub-sectors based on relative ratios in 1980. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(l) Mongolia

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1985-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● No major modifications were done for IEAWEB during 1985-2015. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(m) Cambodia

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1995-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● No major modifications were done for IEAWEB during 1995-2015. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(n) Hong Kong, Democratic People's Republic of Korea, Brunei, Malaysia, Bangladesh, Nepal, Pakistan, and Sri Lanka

Data sources and treatments	● Data of whole country were taken from IEAWEB during 1971-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● No major modifications were done for IEAWEB during 1995-2015. ● See "Assumption for data extrapolation" in this sub-section how to extrapolate the data of IEAWEB to 1950.

(o) Macau, Laos, Afghanistan, Bhutan, and Maldives

Data sources and treatments	● Data of whole country were taken from UN data during 1990-2015 and extrapolated to 1950.
Assumptions for estimating missing historical data	● No major modifications were done for UN data during 1990-2015. ● Data before 1990 were extrapolated to 1950 using trends of fuel consumption estimated using UN Energy Statistics Database as follows: Consumption = Production + Import – Export + Changes in stocks ● Biofuel consumption data before 1970 were extrapolated to 1950 using trends of population numbers.

Assumption for data extrapolation

As described above, fuel consumption data before 1959 and 1970 were not included in IEAWEB for Japan and other countries, respectively. The missing historical fuel consumption data were estimated by extrapolation using trends of related data for each sub-sector. Trend factors used in REASv3 are summarized in Table 3.3.

Table. 3.3. Trend factors for extrapolating fuel consumption data to 1950 in each sub-sector.

Sub-sectors	Trend factors and data sources
Power plants including energy sector	● Trend factors: Amounts of generated power for all fuel types ● Data sources: ➢ Each region of China: China Data Online ➢ Other countries and regions: Mitchell (1998)
Coke ovens and blast furnace including energy sector	● Trend factors: Amounts of pig iron production for all fuel types ● Data sources: See Sect. S4.1.1
Charcoal production plants	● Trend factors: Amounts of charcoal production for all fuel types ● Data sources: Data after 1961 were obtained from FAOSTAT

	(http://www.fao.org/faostat/en) and trends between 1950 and 1960 were assumed based on Fernandes et al. (2007).
Petroleum Refineries including energy sector	● Trend factors: Amounts of total crude oil consumption for all fuel types ● Data sources: Total crude oil consumption was estimated using Mitchell (1998) as follow: Consumption = Production + Import – Export
Iron and steel	● Trend factors: Total amounts of pig iron and crude steel production for all fuel types ● Data sources: See Sect. S4.1.1
Non-ferrous metals	● Trend factors: Total amounts of copper, lead, zinc, and primary aluminum production for all fuel types ● Data sources: See Sect. S4.1.2
Non-metallic minerals industry (cement, lime, and brick)	● Trend factors: Amounts of cement production for all fuel types ● Data sources: See Sect. S4.1.3
Railway	● Trend factors: Length of railway line for all fuel types ● Data sources: Mitchell (1998)
Road transport	● Trend factors: Total annual mileages of vehicles for each fuel type ● Data sources: See Sect. S6.1.1
Others	● Trend factors and data sources: ➢ Coal fuels except for coke fuels: Total coal consumption estimated using Mitchell (1998) as follows: Consumption = Production + Import – Export ➢ Coke fuels and gas fuels except for natural gas: The same trends as those for coke ovens ➢ Natural gas: Total natural gas consumption estimated using Mitchell (1998) ➢ Oil fuels: The same trends as those for petroleum refineries ➢ Biofuels: See Sect. S3.1.8 ➢ Charcoal: The same trends as those for charcoal production plants ➢ Other fuels: Fuel consumption data were not extended to 1950.

S3.1.3 Regional fuel consumption data in China

REASv3 used CESY for fuel consumption data of regions in China defined in Table 2.1 except for Hong Kong and Macau. However, in CESY, only total data are available in industry and transport sectors which need to be distributed to sub-sectors. In REASv3, weighting factors for the distribution were prepared for each region. Basic methodology and data used for the weighting factors are described briefly in this sub-section. Note that all motor gasoline listed in both industry and transport sectors of CESY are assumed to be consumed in road transport sector based on IEAWEB.

Industry sector

For most regions, total consumption data in industry sector were divided into sub-sectors based on weighting factors prepared using energy data in statistical yearbook of each region. Availabilities of detailed data for the weighting factors are different among regions and summarized in Table 3.4 except for Shanghai, Jiangsu, Zhejiang, Shandong, Hainan and Sichuan where no energy data are available in statistical yearbook of each region.

Table. 3.4. Data sources and treatments of weighting factors for each region to distribute total fuel consumption in industry sector to each sub-sector.

Regions	Data sources and treatments
Beijing	● Data of major fuel types were taken from Beijing Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2005/2007/2010/2014 were used before 2000/for 2004/for 2008/for 2011/for 2015.
Tianjin	● Data of major fuel types were taken from Tianjin Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2010/2013 were used before 2000/for 2011/after 2012.
Hebei	● Consumption of main energy sources were taken from Hebei Statistical Yearbook and used for all fuel types. ● For the year when statistics are not available, data in 2005/2010/2013 were used before 2004/for 2011/after 2012.
Shanxi	● Data of coal, coke, and diesel oil were taken from Shanxi Statistical Yearbook. For other fuels, weighting factors were based on data of REASv2 (based on GAINS ASIA at that time).

	● For the year when Shanxi Statistical Yearbook are not available, data in 2000/2010/2013/2014 were used before 1999/for 2011/for 2012/for 2015. For REASv2 (available during 2000-2008), data in 2000/2008 were used before 1999/after 2009.
Inner Mongolia	● Data of major fuel types were taken from Inner Mongolia Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2007/2010/2013 were used before 2000/for 2006/for 2011/after 2012.
Liaoning	● Data of major fuel types were taken from Liaoning Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2010/2013 were used before 2000/for 2011/after 2012.
Jilin	● Data of major fuel types were taken from Jilin Statistical Yearbook. ● For the year when statistics are not available, data in 2000/2002/2005/2010/2013 were used before 1999/for 2001/for 2004/for 2011/after 2012.
Heilongjiang	● Data of major fuel types were taken from Heilongjiang Statistical Yearbook. ● For the year when statistics are not available, data in 2005/2010/2013 were used before 2004/for 2011/after 2012.
Shanghai	See descriptions below this table.
Jiangsu	See descriptions below this table.
Zhejiang	See descriptions below this table.
Anhui	● Data of major fuel types were taken from Anhui Statistical Yearbook. ● For the year when statistics are not available, data in 2000/2002/2010/2013 were used before 1999/for 2001/for 2011/after 2012.
Fujian	● Data of major fuel types were taken from Fujian Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2010/2013 were used before 2000/for 2011/after 2012.
Jiangxi	● Data of major fuel types were taken from Jiangxi Statistical Yearbook. ● For the year when statistics are not available, data in 2000/2010/2013 were used before 1999/for 2011/after 2012.
Shandong	See descriptions below this table.

Henan	● Data of major fuel types were taken from Henan Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2010/2013 were used before 2000/for 2011/after 2012.
Hubei	● Data of coal and diesel oil were taken from Hubei Statistical Yearbook. For other fuels, weighting factors were based on data of REASv2 (based on GAINS ASIA at that time). ● For the year when Hubei Statistical Yearbook are not available, data in 2000/2010/2013 were used before 1999/for 2011/after 2012. For REASv2 (available during 2000-2008), data in 2000/2008 were used before 1999/after 2009.
Hunan	● Data of major fuel types were taken from Hunan Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2005/2010/2013 were used before 2000/for 2004/for 2011/after 2012.
Guangdong	● Data of coal were taken from Guangdong Statistical Yearbook. For other fuels, weighting factors were based on data of REASv2 (based on GAINS ASIA at that time). ● For the year when Guangdong Statistical Yearbook are not available, data in 2000/2010/2013/2014 were used before 1999/for 2011/for 2012/for 2015. For REASv2 (available during 2000-2008), data in 2000/2008 were used before 1999/after 2009.
Guangxi	● Data of total energy consumption were taken from Guangxi Statistical Yearbook for all fuel types. ● For the year when statistics are not available, data in 1995/2000/2014 were used before 1997/for 1998 and 1999/for 2015.
Hainan	See descriptions below this table.
Chongqing	● Data of major fuel types were taken from Chongqing Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2010/2013 were used before 2000/for 2011/after 2012.
Sichuan	See descriptions below this table.
Guizhou	● Data of major fuel types were taken from Guizhou Statistical Yearbook. ● For the year when statistics are not available, data in 2000/2010/2014 were used before 1999/for 2011/for 2015.
Yunnan	● Data of coal, coke, and oil were taken from Yunnan Statistical

	Yearbook. For other fuels, weighting factors were based on data of REASv2 (based on GAINS ASIA at that time). ● For the year when Yunnan Statistical Yearbook are not available, data in 2000/2013 were used before 1999/after 2014. For REASv2 (available during 2000-2008), data in 2000/2008 were used before 1999/after 2009.
Tibet	● Fuel consumption data were not from CESY. (See Sect. S3.1.2)
Shaanxi	● Data of coal, coke, and diesel oil were taken from Shaanxi Statistical Yearbook. For other fuels, weighting factors were based on data of REASv2 (based on GAINS ASIA at that time). ● For the year when Shanxi Statistical Yearbook are not available, data in 2002/2005/2010/2013 were used before 2001/for 2004/for 2009 and 2011/after 2014. For REASv2 (available during 2000-2008), data in 2000/2008 were used before 1999/after 2009.
Gansu	● Data of major fuel types were taken from Gansu Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2010/2013/2014 were used before 2000/for 2011/for 2012/for 2015.
Qinghai	● Data of coal were taken from Qinghai Statistical Yearbook. For other fuels, weighting factors were based on data of REASv2 (based on GAINS ASIA at that time). ● For the year when Qinghai Statistical Yearbook are not available, data in 2001/2010/2013 were used before 2000/for 2011/after 2014. For REASv2 (available during 2000-2008), data in 2000/2008 were used before 1999/after 2009.
Ningxia	● Data of major fuel types were taken from Ningxia Statistical Yearbook. ● For the year when statistics are not available, data in 2000/2010/2013 were used before 1999/for 2011/after 2014.
Xinjiang	● Data of major fuel types were taken from Xinjiang Statistical Yearbook. ● For the year when statistics are not available, data in 2001/2007/2009/2013 were used before 2000/for 2008/for 2010/after 2014.
Hong Kong	Fuel consumption data were not from CESY. (See Sect. S3.1.2)
Macau	Fuel consumption data were not from CESY. (See Sect. S3.1.2)

For Shanghai, Jiangsu, Zhejiang, Shandong, Hainan and Sichuan, weighting factors were assumed based on sub-sector level fuel consumption data developed using the China total data described in Sect. S3.1.2 and related regional data as follows:

- Weighting factors to distribute fuel consumption in whole China to each region were prepared for each sub-sector and commonly used for all fuel types. The weighting factors for each sub-sector used in REASv3 are as follows:
  - Amounts of steel production in each region (see Sect. S4.1.1) were used for iron and steel sub-sector.
  - Total amounts of copper, lead, zinc, and primary aluminum production in each region (see Sect. S4.1.2) were used for non-ferrous metals sub-sector.
  - Amounts of cement production in each region (see Sect. S4.1.3) were used for non-metallic minerals sub-sector in IEAWEB. (Fuel consumption in non-metallic minerals were further distributed to cement, lime, and brick sub-sectors in REASv3. See Sect. S3.1.7.)
  - Amounts of coal production in each region taken from China Data Online were used for coal mines (in energy sector) and mining and quarrying sub-sectors.
  - Amounts of paper and paperboard production in each region taken from China Data Online were used for paper, pulp and prints sub-sector.
  - Amounts of textile production in each region (see Sect. S5.1.2) were used for textile and leather sub-sector.
  - GDP of each region taken from China Data Online were used for other sectors.
- Using the China total data and the weighting factors, the tentative regional fuel consumption data (TRFCD) were developed. Then, the fuel consumption ratio of each sub-sector to industry sector total was calculated for Shanghai, Jiangsu, Zhejiang, Shandong, Hainan and Sichuan using the TRFCD of each region. Finally, fuel consumption in industry sector of each region in CESY was distributed to sub-sectors using the corresponding ratio. When categories of fuel types are different between the TRFCD and CESY, following procedures were adopted:
  - For raw coal, cleaned coal, and other washed coal in CESY, the ratio for total of anthracite, coking coal and other bituminous coal in the TRFCD were used.
  - For other coking products and other petroleum products in CESY, the ratio for coke oven coke and heavy fuel oil in the TRFCD were used, respectively.

Transport sector

For transport sector, no detailed data are available even in statistical yearbook of each region. Therefore, weighting factors for each region were assumed in the similar procedure for industry sector as follows:

- As mentioned in the first paragraph of this sub-section, all motor gasoline consumption (including those in industry sector) is distributed to road transport sector.
- All solid coal fuels are assumed to be used in railway sector.
- Natural gas consumption before and after 1995 was distributed to pipeline transport and road transport sectors, respectively.
- All heavy fuel oil consumption is distributed to domestic navigation sector.
- For diesel oil, using the same methodology for industry sector, diesel oil consumption data in road transport, railway, and domestic navigation sectors in each region were developed and then, weighting factors were assumed. For regional diesel oil consumption data, those in railway and domestic navigation sectors were taken from REASv2 (based on GAINS ASIA at that time) during 2000-2008 and data in 2000 and 2008 were used before 1999 and 2009, respectively. See Sect. S6.1.2 for diesel oil consumption in each region in road transport sector.
- Consumption of all other fuels is distributed to non-specified transport sector.
- Assumptions of motor gasoline, solid coal fuels, natural gas and heavy fuel oil described above were based on IEAWEB.

S3.1.4 Regional fuel consumption data in India

As defined in Table 2.1, REASv3 has 17 sub-regions for India. Therefore, fuel consumption data of country total based on IEAWEB need to be divided for each sub-region. Table 3.5 provides weighting factors used to allocate country total data to the 17 sub-regions.

Table. 3.5. Weighting factors for allocating country total fuel consumption data to the 17 sub-regions in India.

Sectors and fuel types	Weighting factors and data sources
Power plants including energy sector	● Weighting factors: Total generation capacities in each region ● Data sources: World Electric Power Plants Database (Platts, 2018)
Iron and steel	● Weighting factors: Amounts of crude steel production for all fuel types ● Data sources: See Sect. S4.1.1
Non-ferrous metals	● Weighting factors: Total amounts of copper, lead, zinc, and primary

	aluminum production for all fuel types ● Data sources: See Sect. S4.1.2
Non-metallic minerals industry (cement, lime, and brick)	● Weighting factors: Amounts of cement production for all fuel types ● Data sources: See Sect. S4.1.3
Road	● See Sect. S6.1
Rail	● Weighting factors: Length of railway line open for all fuel types ● Data sources: Factors after 2005 were estimated from TERI (2013, 2018) and those in 2005 were used before 2004.
Biofuels	● See Sect. S3.1.8
Industry and energy sectors (default)	● Weighting factors and data sources: ➢ Factors for LPG, motor gasoline, kerosene, diesel oil, heavy fuel oil, and naphtha after 1998 were estimated from TERI (2013, 2018) and those in 1998 were used before 1997. ➢ Factors for other fuels after 1999 were estimated from "Fuel Consumed" in Annual Survey of Industries (Ministry of Statistics & Programme Implementation, http://www.csoisw.gov.in/cms/en/1023-annual-survey-of-industries.aspx) and those in 1999 were used before 1998.
Residential and other domestic sectors	● Weighting factors and data sources: ➢ Factors for kerosene and LPG after 1983 were estimated from TERI (2013, 2018) and those in 1983 were used before 1982. ➢ Data of LPG were also used for natural gas and for other fuels, those of kerosene were used.

S3.1.5 Regional fuel consumption data in Japan

REASv3 has 6 sub-regions for Japan as defined in Table 2.1 and the same as the case of India, fuel consumption data of country total based on IEAWEB need to be divided for each sub-region. Table 3.6 provides weighting factors used to allocate country total data to the 6 sub-regions.

Table 3.6. Weighting factors for allocating country total fuel consumption data to the 6 sub-regions in Japan.

Sectors and fuel types	Weighting factors and data sources
Power plants including energy sector	● Weighting factors: Total generation capacities in each region ● Data sources: World Electric Power Plants Database (Platts, 2018)
Non-ferrous metals	● Weighting factors: Total amounts of copper, lead, zinc, and primary aluminum production for all fuel types ● Data sources: See Sect. S4.1.2
Road	● See Sect. S6.1
Others	● Weighting factors: ➢ Factors for each sector and fuel type during 1990-2015 were estimated using energy consumption statistics of each prefecture in corresponding years of 1990-2015. ➢ Factors in 1990 were used for those before 1989. ● Data sources: ➢ Website of the Agency for National Resources and Energy https://www.enecho.meti.go.jp/statistics/energy_consumption/ec002/results.html (in Japanese)

S3.1.6 Fuel consumption in power plants

General methodology

In REASv3, power plants with following criteria were treated as point sources:

- Power plants which were treated as point sources in REASv2 (see Kurokawa et al., 2013).
- Power plants which entered commercial operation after 2008 and whose total generating capacities of units in each power plant were larger than 300MW.

Then, fuel consumption in power plants sector was estimated as follows:

1) Fuel consumption in each power plant (point source) was estimated. (see “Fuel consumption in each power plant” below)
2) (A) Total of the fuel consumption in each power plant was calculated in each country and region.
3) If (A) was larger than (B) fuel consumption in total power plant sector in a corresponding country and region, data of each power plants prepared in 1) were adjusted by the ratio of (B) to (A). In this case, fuel consumption of power plants as area sources was assumed to be zero.

4) IF (A) was smaller than (B), the value of (B) minus (A) was assumed to be fuel consumption in area sources. In this case, there is no change for the data of each power plant developed in 1).

Fuel consumption in each power plant

In REASv2, power plants whose annual CO_2 emissions in the Carbon Monitoring for Action (CARMA) Database (Wheeler and Ummel, 2008) were more than 1 Mt in 2000 and/or 2007 were treated as point sources. Before 2007, REASv3 used the same power plants as point sources with some revisions for such as generation capacities, fuel types, etc. using the updated World Electric Power Plants Database (Platts, 2018). For fuel consumption, data between 2000 and 2007 were basically the same as those in REASv2. Before 2000, fuel consumption of each power plant in operation was assumed to be the same as that in 2000 which will be adjusted based on total fuel consumption in power plants sector as described in "General methodology" above. (Note that power plants which were constructed and retired before 2000 were not considered in REASv3.) After 2008, REASv3 included power plants which entered commercial operation after 2008 as new point sources based on the WEPP (see also "General methodology" above). Although major information was available including fuel types used in each power plant, there are no data of fuel consumption in the WEPP. Thus, in REASv3, annual fuel consumption per generation capacity for each fuel type was estimated first using data in 2000 and 2007 for each country. The data were estimated for power plants which started operation before 1999 and after 2000, separately. Then, using the generation capacities data obtained from the WEPP, fuel consumption in each power plant was estimated.

S3.1.7 Fuel consumption in non-metallic minerals

REASv3 defined cement, lime, brick, and non-specified sub-sectors in the non-metallic minerals category in stationary combustion sources. However, energy statistics used in REASv3 including IEAWEB and regional statistical yearbook of China provide fuel consumption in total non-metallic minerals industry which needs to be distributed to each sub-sector.

In REASv3, all primary coal fuels were assumed to be used in cement, lime, and brick production. For China, Hua et al. (2016), Wang et al. (2012), and Streets et al. (2006) give coal consumption in cement (1980-2012), brick (1950-2015), and lime (2001) industries, respectively. Using these data and production amounts of cement, lime and brick, coal consumption per unit of production of cement, lime, and brick was estimated, respectively. Then, coal consumption data in non-metallic minerals in each region were distributed to each sub-sector based on production amounts of cement, lime, and brick in each region and corresponding coal consumption per united of production. Similarly, Maithel (2013) provides coal consumption in cement and brick industries in Pakistan during 2001-2010 and with production amounts of cement and brick, fuel consumption in non-metallic minerals industry were distributed to each sub-sector. For other countries, due to lack of information, averaged coal consumption per unit of production of cement, lime, and brick for China was used for other East and Southeast Asian countries. For other countries in South Asia, averaged coal consumption per unit of production of cement and brick for Pakistan and that of lime for China was used. Then, with production data of cement, lime, and brick, fuel consumption in non-metallic minerals were distributed to each sub-sector. See Sects. S4.1.3, S4.1.4, and S4.1.5 for production data of cement, lime, and brick, respectively.

For other fuels, in REASv3, coke oven coke and heavy fuel oil were assumed to be used in cement industry and others including gas fuels and diesel oil were allocated to the non-specified sub-sector.

S3.1.8 Biofuels

China

CESY provides biofuel consumption data of fuelwood, crop residue, and biogas in each region during 1998-2007 which were used in REASv3. Before 1997, data were extended to 1980 using trends of each fuel consumption data in REASv1 and then extended to 1950 based on trends of biofuel consumption in East Asia obtained from Fernandes et al. (2007). After 2007, fuelwood, crop residue, and biogas consumption in total China were extrapolated to 2015 using trends of primary solid biofuels consumption in IEAWEB. Then, consumption of each fuel in each region in 2007 were tentatively extrapolated to 2015 using trends of rural population numbers in each region. Finally, fuelwood, crop residue, and biogas consumption in total China estimated during 2008-2015 were distributed to each region using the tentatively extrapolated data in each region.

India

Primary solid biofuels in IEAWEB were assumed to be total of fuelwood, crop residue and animal waste in India during 1971-2015. Before 1970, the primary solid biofuels consumption was extrapolated to 1950 using trends of biofuel consumption in South Asia obtained from Fernandes et al. (2007). Then, relative ratios of fuelwood, crop residue, and animal waste consumption in 17 sub-regions to consumption of the primary solid biofuels in total India were calculated for 1990 and 2010 using data in Streets and Waldhoff (1998) and Census of India 2011 (Chandramouli, 2011), respectively and interpolated between 1991 and 2009. Before 1989 and after 2011, the ratios of 1990 and 2010 were assumed to be constant, respectively. Finally, fuel consumption of fuelwood, crop residue, and animal waste in each sub-region during 1950-2015 were calculated.

Japan

Primary solid biofuels consumption in IEAWEB were assumed to be fuelwood consumption in Japan during 1982-2015. Before 1981, as described in Sect. S3.1.2, fuel consumption in residential and paper, pulp and printing industry sectors was extrapolated to 1950 using the Historical Statistics of Japan (Japan Statistical Association, 2006) and trends of production amounts of paper and pulp in Japan, respectively.

Macau, Laos, Afghanistan, Bhutan, and Maldives

See Sect. S3.1.2 for methodology and data sources. Only fuelwood and charcoal were included for this group.

Other countries

Primary solid biofuels data in IEAWEB were assumed to be total of fuelwood, crop residue and animal waste consumption in each country and extrapolated to 1950 using trends of biofuel consumption in East or Southeast or South Asia obtained from Fernandes et al. (2007). For distribution to each fuel type, consumption ratios of fuelwood, crop residue, and animal waste in 1990 obtained from Streets and Waldhoff (1998) were used during 1950-2015.

S3.2 Emission factors and settings of emission controls

S3.2.1 SO_2

Sulfur contents in fuels

In REASv3, default settings were taken from those of REASv1 during 1980-2000 generally based on RAINS ASIA at that time, Streets et al. (2000), Kato and Akimoto (1992) and Kato et al. (1991). For countries using default settings, data in 1980 and 2000 were used before 1979 and after 2001, respectively. For China, India, Japan, Republic of Korea, and Taiwan, additional country-specific settings were considered as described in Table 3.7.

Table 3.7. Settings and assumptions of sulfur contents in fuels for China, India, Japan, Republic of Korea, and Taiwan.

Countries	Settings and assumptions
China	● Coal: ➢ During 1985-2000: Data were taken from REASv1 based on Kato and Akimoto (1992) in 1985 and China Coal Industry Yearbook 2002 (State Administration for Coal Safety, 2003) in 1990 and 1995. In 2000, data in 1995 were adjusted so that the national average sulfur contents were 1.08% after Lu et al. (2010). Data in other years were interpolated. ➢ During 2001-2005: Data were taken from REASv2 where settings of power plants in 2005 were based on Zhao et al. (2008) and national average sulfur contents were adjusted to 1.02% after Lu et al. (2010). Data between 2000 and 2005 were interpolated. ➢ Before 1984 and after 2006, settings in 1980 and 2005 were used, respectively. ● Oil ➢ Before 1985, data were obtained from Kato et al. (1991) and those in 1995 were based on information from Tsinghua University (1.5% for heavy fuel oil and 0.58%, 0.35%, and 0.163% for diesel oil in north, northeast, and other areas, respectively) for REASv1. Data between 1986-1994 were interpolated and after 1996, data in 1995 were used.
India	● Data were taken from REASv1 based on Reddy and Venkataraman (2002) for coal, heavy fuel oil, and light fuels and Kato et al. (1991) for others. The same data were used for the entire target period of REASv3.
Japan	● Coal: Data during 1960-1996 were taken from Li and Dai (2000). The value in 1960 was 1.06% and gradually decreased to 0.60% in 1996. It was assumed that the value was reduced by 10% from 1996 to 2010 referring a report of MOEJ (2012). Data between 1996 and 2010 were interpolated and those in 1960 and 2010 were used before 1959 and after 2011, respectively. ● Heavy fuel oil and crude oil: Settings during 1965-2010 for power plants were based on Iwaya (2013). Those for industry were based on Kato et al. (1991), Streets et al. (2000), and Imura et al. (1999). Data

	in 1965 and 2010 were used before 1964 and after 2011, respectively. ➢ Heavy fuel oil for power plants: The values before 1965 were 2.6% and decreased almost constantly to 0.80% in 1975. Then the values were gradually decreased to 0.75% in 1990 and the values was used after 1990. ➢ Heavy fuel oil for industry: The values before 1965 were 2.60% and assumed to be decreased gradually to 1.4% in 1975, 1.1% in 1985, and 1.0% in 2000. The values after 2000 were assumed to be constant. ➢ Crude oil for power plants: The value before 1966 were 2.8% and decreased almost linearly to 0.20% in 1975. After 1975, values were between 0.15% and 0.20%. ● Diesel: Settings were based on regulations of diesel oil in Japan as follows: 1.2% before 1975, 0.50% during 1976-1991, 0.20% during 1992-1996, 0.05% during 1997-2003, and 0.01% after 2004.
Republic of Korea and Taiwan	● Data during 1980-2000 were taken from REASv1 based on Kato et al. (1991), RAINS ASIA, and Streets et a. (2000) and those in 1975 were obtained from Kato et al. (1991). Data between 1976-1981 were interpolated and those in 1975 and 2000 were used before 1974 and after 2001, respectively.

Emission factors

SO_2 emissions from coal and oil fuels were calculated using sulfur contents in fuels and ratios of sulfur emitted as SO_2. Settings of REASv3 were taken from REASv1 and REASv2 based on Kato and Akimoto (1992), Kato et al. (1991) and RAINS ASIA as follows:

- Power plants (point sources): 0.95
- Power plants (area sources)): 0.90 for Japan, Republic of Korea, and Taiwan; 0.775 for other countries and regions.
- Industry sector: 0.775
- Coke ovens: 0.0685
- Iron and steel: 0.1483
- Transport sector: 0.775
- Domestic sector: 0.60
- Coke oven coke for all sectors: 0.885
- Oil fuels for all sectors: 1.0

For coke ovens, activity data are coal input and it is considered that the estimated SO_2 emissions include both combustion and non-combustion sources.

For gas fuels such as coke oven gas and blast furnace gas, light fuels such as LPG, and other fuels except for primary biofuels such as charcoal and municipal wastes, emission factors were derived from Kato and Akimoto (1991). Those for fuelwood and crop residue were taken from Garg et al. (2001) and those for animal waste were from Gadi et al. (2003).

In cement plants, effects of absorption of SO_2 by cements need to be considered. In REASv3, the absorption rates for China were obtained from Li et al. (2017) and those for other countries were based on Kato et al. (1991).

Settings of emission controls

Settings and assumptions for reduction of SO_2 emissions from combustion sources by abatement equipment adopted in REASv3 are summarized in Table 3.8. For other sources not described in Table 3.8, no emission controls were considered.

Table 3.8. Settings and assumptions of emission controls of SO_2

Countries	Settings and assumption
China	● Power plants: Effects of flue-gas desulfurization (FGD) were considered after 2000 as follows: ➢ Settings during 2000-2008 were taken from REASv2 based on national introduction rates of FGD from Lu et al. (2010) and those of each province from Zhao et al. (2008). ➢ After 2008, increases of penetration of FGD were assumed referring Liu et al. (2015) and Li et al. (2017). In 2015, the introduction rates were assumed to be 100% in power plants considered as point sources and 90% for other power plants. ● Industry: Effects of FGD were roughly assumed as follows: ➢ Referring Li et al. (2017), it was assumed that regulations started from (A) Beijing and Shanghai, then (B) Shandong, Hebei, and Guangdong, and finally (C) other provinces. ➢ Regulations of industrial boiler were strengthened after 2014 referring Zheng et al. (2018). ➢ For (A), it was assumed that introduction of FGD started from 2000 and penetration rates in 2010 were 40% which is a setting for China in 2020 in Business-as-usual scenario of Wang et al.

	(2014). For the penetration rates, linear trends were assumed during 2000-2013. ➢ For (B) and (C), it was assumed that penetration of FGD started 2 and 4 years after (A), respectively and reduction effects were assumed to be smaller than (A) by 10% and 15%, respectively. ➢ In 2015, reduction rates of SO_2 emissions were assumed to be 75%, 63%, and 52% for (A), (B), and (C), respectively.
Japan	● Power plants: Referring MRI (2015), Kato et al. (1991), and MOEJ (2000), effects of FGD were considered after 1968 as follows: ➢ In 1990 and after 2000, introduction rates of FGD in power plants as point sources were assumed to be 95% and 100%, respectively. Trends of the introduction rates during 1968 and 1990 were assumed based on MOEJ (2000) and those between 1990 and 2000 were interpolated. ➢ For introduction rates of FGD in power plants as area sources, it was assumed to be 95% after 2000 and the trends before 1990 were estimated based on those of point sources. ● Other sectors: Referring Kato et al. (1991), reduction rates of SO_2 emissions were assumed as follows: ➢ For large industries including sulphuric acid plants, 80% of reduction rates of power plants as area sources were adopted. ➢ For other industries, reduction rates were assumed to be 50% of large industries. ➢ For commercial and public services, 50% of reduction rates of other industries were adopted.
Republic of Korea	● Effects of FGD were roughly assumed as follows: ➢ Power plants: Referring Ebata et al. (1997) and Wang et al. (2014), it is assumed that introduction of FGD was from 1990. The penetration rates in power plants as point sources in 2000, 2005, and 2010 were 90%, 97%, and 98%, respectively. Data between 1990, 2000, 2005, and 2010 were interpolated and data in 2010 were used after 2011. Effects of FGD on power plants as area sources were assumed to be 5% lower than point sources. ➢ Industry: It was assumed that introduction of FGD started from 1990 and penetration rates of FGD were 80% and 85% in 2005 and 2010, respectively based on Wang et al. (2014). Data between

	1990, 2005, and 2010 were interpolated and data in 2010 were used after 2011.
Taiwan	● Effects of FGD were roughly assumed as follows: ➢ Power plants: Due to lack of information, the same reduction rates of Republic of Korea were adopted after 1995. But according to Ebata et al. (1997), introduction of FGD started earlier than Republic of Korea. It was assumed that penetration rates in 10% and 30% in 1980 and 1990, respectively and data between 1980, 1990, and 1995 were interpolated. ➢ Industry: Similar to power plants, the same reduction rates of Republic of Korea were adopted after 2000 and it was assumed that introduction of FGD started from 1985. Data between 1985 and 2000 were interpolated.
Thailand	● Effects of FGD were assumed as follows: ➢ Power plants as point sources: Referring UN Environment (2018), reduction rates were assumed for four power plants as follows: Mae Moh (0.8-0.97 in 1978-2015), BLCP Power (0.84 from 2006), National Power Supply (0.75 from 1999), and GHECO-One (0.952 from 2012).
Other countries	● Effects of FGD were assumed as follows: ➢ Power plants as point sources: Reduction rets (0.7-0.9) were assumed if units have information of installed FGD equipment in World Electric Power Plants Database (Platts, 2018). ➢ Countries which have power plants with FGD and number of such power plants in 2015 (in parentheses) in REASv3 were as follows: India (10), Indonesia (5), Laos (1), Malaysia (4), Vietnam (10), and Sri Lanka (2).

S3.2.2 NO_x

Default emission factors

Table 3.9 summarized default emission factors used in REASv3 for fuel combustion in power plants, industry and residential sectors. Specific settings for coke ovens, iron and steel industry, cement industry, and emission controls were described below the table.

Table 3.9. Default emission factors of NO_x from fuel combustion in power plants, industry and residential sectors. Unit is t/PJ expressed as NO_2.

Fuel type	Power plants	Industry	Residential
Hard coal[h]	345[a]	260[e]	78[g]
Raw coal[i]	See Table 3.10.	203[f]	61.1[g]
Cleaned coal[i]		162[f]	48.5[g]
Other washed coal[i]		509[f]	153[g]
Sub-bituminous coal	524[a]	A	B
Lignite	433[a]	A	B
Coke oven coke[j]	345	260	78
Natural gas	105[b]	53[b]	37[b]
Gas works gas	10.5[b]	7.4[b]	5.25[b]
Coke oven gas	77.8[b]	55[b]	38[b]
Blast furnace gas	10.5[b]	7.4[b]	38[b]
LPG	79[b]	56[b]	33[b]
Kerosene	485[b]	167[b]	25[b]
Diesel oil	632[b]	222[b]	74[b]
Crude oil	249[b]	145[b]	49[b]
Heavy fuel oil	249[b]	145[b]	49[b]
Fuelwood	45[c]		
Crop residue	91.1[c]		
Animal waste	91.1[c]		
Charcoal	100[d]		

a. AP-42 (US EPA, 1995). b. Kato and Akimoto (1992). c. Streets and Waldhoff (1998), d. Revised 1996 IPCC guidelines (IPCC, 1997). e. Estimated based on ratios of emission factors between power plants and industry in Kato and Akimoto (1992). f. Estimated referring Zhang et al. (2007). g. 30% of emission factors of industry were adopted based on Kato and Akimoto (1992). h. Emission factors were commonly used for coking coal, anthracite and bituminous coal. i. Only defined for China. j. Emission factors for hard coal were adopted. A. Estimated based on ratios of emission factors between power plants and industry in Kato and Akimoto (1992) considering differences of net calorific values. B. 30% of emission factors of industry were adopted.

Coke ovens

For coal input to coke ovens, emission factor was 1.0 t/kt taken from Kato and Akimoto (1992). It is considered that NO_x emissions estimated using this emission factor include contributions from both combustion and non-combustion processes.

Iron and steel industry

In iron and steel industry, emission factors for cokes, coke oven gas, and blast furnace gas were taken from Kato and Akimoto (1992) as follows:

- Coke oven coke: 4.0 t/kt for China and 2.5 t/kt for other countries
- Coke oven gas: 141 t/PJ
- Blast furnace gas: 76.4 t/PJ

For other fuel types, default emission factors were used.

Cement industry

For China, emission factors of coal combustion in each cement kiln type were obtained from Lei et al. (2011) as follows: 15.3 t/kt for precalciner kilns, 18.5 t/kt for other rotary kilns, and 1.7 t/kt for shaft kilns. Coal consumption in each cement kiln type were estimated based on Lei et al. (2011) and Hua et al. (2016). For other fuel types, default emission factors in industry were used.

For Japan, NO_x emissions were not estimated based on fuel consumption, but using amount of cement production in each kiln type. Emission factors (t/kt of clinker produced) were taken from AP-42 (US EPA, 1995) as follows: 3.7 for wet process kilns, 3.0 for long dry process kilns, 2.4 for preheater process kilns and 2.1 for preheater/precalciner kilns. Ratio of clinker to cement was assumed to be 0.85 based on Cement handbook (Japan Cement Association, 2019). (See Sect. S4.1.3 for production data by different kiln types.)

For other countries and regions, default emission factors in industry were used for all fuel types.

Settings of emission controls

Settings and assumptions for reduction of NO_x emissions from combustion sources by abatement equipment adopted in REASv3 are summarized in Table 3.10. For other sources not described in Table 3.10, no emission controls were considered.

Table 3.10. Settings and assumptions of emission controls of NO_x

Countries	Settings and assumption
China	● Power plants ➢ Referring Zhang et al. (2007) and Liu et al. (2015), emission factors [t/PJ] for coal fired power plants were assumed considering effects of low-NO_x burner based on capacity and years as follows: ✧ 227: Larger than 300 MW or equal to 300 MW after 1995 ✧ 300: Smaller than 300 MW but equal to or larger than 100 MW after 1997. ✧ 393: Equal to 300 MW before 1995 or Smaller than 300 MW but equal to or larger than 100 MW before 1997. ✧ 360: Less than 100 MW. ✧ 300: Power plants as area sources (no information of capacity) before 2000. The values were assumed to be decreased by 10% until 2010 and by 15% until 2015. ➢ Penetration rates of selective catalytic reduction (SCR: efficiency 73%) and selective non-catalytic reduction (SNCR: efficiency 30%) for each province in 2011 were taken from Chen et al. (2014). Referring Chen et al. (2014), Li et al. (2017), and Zheng et al. (2018), national introduction rates were assumed to be 12%, 18%, and 75% in 2010, 2011, and 2015 and reduction rates for as point sources were estimated. For area sources, 50% of reduction rates of point sources were adopted. ● Industry ➢ Referring Li et al. (2017), effects of De-NO_x system were considered for precalciner kilns in cement plants and penetration rates were roughly assumed to be 0% in 2010, 50% in 2014 and 90% in 2015.0
Japan	● Power plants: Referring MRI (2015), JMF and ICETT (2003), and MOEJ (2000), effects of low-NO_x burner and SCR were considered as follows: ➢ Effects of low-NO_x burner were considered after 1970 and reduction efficiencies were assumed to be 15%, 35%, and 50% in 1975, 1980, and after 2005, respectively. Data between 1970, 1975, 1980, and 2005 were interpolated.

	➢ Effects of SCR were considered after 1974 and introduction rates in coal, oil, and gas power plants as point sources were assumed to be 80%, 40%, and 72% in 2002 and 90%, 45%, and 80% after 2010, respectively. Trends of the introduction rates during 1974 -2002 were assumed based on MOEJ (2000) and reduction rates during 2002-2010 were interpolated. For power plants as area sources, reduction rates were assumed to be 85% of point sources. ● Industry: Effects of low-NO_x burner and SCR were roughly assumed referring MRI (2015) and Kato et al. (1991) as follows: ➢ It was assumed that trends of introduction rates of low-NO_x burner were the same as for those of power plants, but reduction efficiencies were 50% of those for power plants as point sources. ➢ For large industries such as cement, iron and steel, it was assumed that trends of penetration rates of SCR were the same as those of power plants, but reduction efficiencies were 50% of those for power plants as point sources. For other industries, reduction rates were assumed to be 50% of those for large industries.
Republic of Korea/Taiwan	● For power plants, introduction rates of low-NO_x burner were 84% and 86% in 2005 and 2010, respectively and those of SCR (SNCR) were 56% (5%) and 68% (5%) in 2005, and 2010, respectively based on Wang et al. (2014). It was roughly assumed that low-NO_x burner, SCR, and SNCR were installed from 1990 and their penetration rates in 2015 were 90%, 73%, and 5%, respectively. Reduction rates between 1990, 2005, 2010, and 2015 were interpolated. ● Due to lack of information, the same settings for Republic ok Korea were adopted to Taiwan.
Others	● Effects of low-NO_x burner and De-NO_x system were assumed as follows: ➢ Power plants as point sources: Reduction rets (0.7-0.9) were assumed if units have information of installed FGD equipment in World Electric Power Plants Database (Platts, 2018). ● Countries which have power plants with De-NO_x equipment and number of such power plants in 2015 (in parentheses) in REASv3 were as follows: India (11), Indonesia (5), Malaysia (6), Philippines (4), Singapore (4), Thailand (9), Vietnam (4), Pakistan (1), and Sri Lanka (2).

S3.2.3 CO

Default emission factors

Table 3.11 summarized default emission factors used in REASv3 for fuel combustion in power plants, industry and residential sectors. Specific settings for coal combustion and, iron and steel industry, cement and other non-metallic minerals industries were described below the table.

Table 3.11. Default emission factors of CO from fuel combustion in power plants, industry and residential sectors. Unit is t/PJ.

Fuel type	Power plants	Industry	Residential
Hard coal[e]	20[a]	See "Emission factors for coal combustion" below.	
Raw coal[f]	20[a]		
Cleaned coal[f]	20[a]		
Other washed coal[f]	20[a]		
Sub-bituminous coal	20[a]		
Lignite	20[a]		
Coke oven coke	20[a]	150[a]	2000[a]
Natural gas	20[a]	30[a]	50[a]
Gas works gas	20[a]	150[a]	150[a]
Coke oven gas	20[a]	150[a]	150[a]
Blast furnace gas	20[a]	150[a]	150[a]
LPG	15[a]	10[a]	326[a]
Kerosene	15[a]	15[a]	179[a]
Diesel oil	15[a]	15[a]	20[a]
Crude oil	15[a]	15[a]	20[a]
Heavy fuel oil	15[a]	15[a]	20[a]
Fuelwood	255.5[b]	2555[c]	5110[d]
Crop residue	354.5[b]	3545[c]	7090[d]
Animal waste	330[b]	3300[c]	6600[d]
Charcoal	400[b]	4000[a]	7000[a]

a. The global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012). b. Emission factors of power plants were assumed to be 10% of industry sector. c. Emission factors of industry sector were assumed to be 50% of residential sector. d. Streets and Waldhoff (1999). e. Emission factors were commonly used for coking coal, anthracite and bituminous coal. f. Only defined for China.

Emission factors for coal combustion

(a) Industry sector except for cement and other non-metallic minerals industries

Due to lack of information of detailed boiler and furnace types in industry sub-sectors in each country, CO emission factors of industry sector were roughly assumed in REASv3 as follows:

- 5.75 t/kt: average of emission factors for fluidized bed furnace and automatic stoker boiler based on AP-42 (US EPA, 1995).
  - Default emission factors for Japan, Republic of Korea, and Taiwan
  - Emission factors for large industries in China
- 18.6 t/kt: Emission factors for other industries in China estimated referring Streets et al. (2006) and data for fluidized bed furnace, automatic stoker, and hand-feed stoker in AP-42 (US EPA, 1995).
- 8.5 t/kt: Emission factors based on automatic stoker in AP-42 (UE EPA, 1995) were adopted for large industries in other countries.
- 66.25 t/kt: Emission factors based on average of automatic stoker and hand-feed stoker in AP-42 (UE UPA, 1995) for other industries in other countries.
- It was assumed that emission factors in China were decreased by 25% from 2000 to 2015 linearly assuming improvement in combustion efficiency.

(b) Residential sector

Emission factors for China, India, and other countries were assumed as follows:

- 75 t/kt for China obtained from Streets et al. (2006) for stove in residential sector.
- 275 t/kt for India taken from Pandey et al. (2014) for traditional stove in residential sector.
- 2.61 kt/PJ for other countries as default emission factor derived from the global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012)

Coke production and iron and steel industry

In REASv3, CO emissions from coke production and iron and steel industry were also estimated using production amounts of coke oven coke, sinter, pig iron, and crude steel (see Sects. S4.2.1 and S4.2.8). CO emission factors for coal consumption in coke ovens, those for coal and coke fuels in blast furnace, and coke furls and gas fuels in iron and industry sectors were assumed to be zero assuming their contributions were included in the emissions estimated based on production amounts described in Sects S4.2.1 and S4.2.8. These mean that CO emissions from combustion sources in coke production and iron and steel industry were not estimated separately in REASv3.

Cement industries

For China, emission factors of coal combustion in each cement kiln type were obtained from Lei et al. (2011) as follows: 17.8 t/kt for precalciner kilns, 17.8 t/kt for other rotary kilns, and 155.7 t/kt for shaft kilns. Coal consumption in each cement kiln type were estimated based on Lei et al. (2011) and Hua et al. (2016). For other fuel types, default emission factors in industry were used.

For Japan, CO emissions were not estimated based on fuel consumption, but using amount of cement production in each kiln type. Emission factors (t/kt of clinker produced) were taken from AP-42 (US EPA, 1995) as follows: 0.06 for wet process kilns, 0.11 for long dry process kilns, 0.49 for preheater process kilns and 1.8 for preheater/precalciner kilns. Ratio of clinker to cement was assumed to be 0.85 based on Cement handbook (Japan Cement Association, 2019). (See Sect. S4.1.3 for production data by different kiln types.)

For other countries and regions, 63.8 t/kt were used for emission factors for coal consumption in cement industry based on average of emission factors for precalciner kilns, other rotary kilns, and shaft kilns taken from AP-42 (US EPA, 1995). For other fuel types, default emission factors in industry were used.

Other non-metallic minerals industries

For lime industry, 155.7 t/kt were commonly used for coal combustion in all countries and default emission factors were used for other fuel types. For brick industry, 150 t/kt were used for coal combustion in China and default emission factors were adopted for Japan, Republic of Korea, and Taiwan. For other countries, emissions from brick industry were not estimated based on fuel combustion, but using amount of brick production. Emission factor 2.0 t/kt of brick produced was assumed based on Weyant et al. (2014) (See Sect. S4.2.5). For other sources, default emission factors were used.

S3.2.4 PM species

Default emission factors

Tables 3.12-14 summarized default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC used in REASv3 for fuel combustion in power plants, industry and residential sectors (Note that emissions of PM species from gas fuels were neglected in REASv3). Specific settings for biofuels, iron and steel industry, cement and other non-metallic minerals industries were described below the table.

Table 3.12. Default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC from fuel combustion in power plants. Unit is t/kt.

Fuel type	PM_{10}	$PM_{2.5}$	BC	OC
Hard coal[f]	12.0[a]	5.08[c]	0.072[a]	0.0[a]
Raw coal[g]	46.0[b]	12.0[b]	0.024[b]	0.0[b]
Cleaned coal[g]	46.0[b]	12.0[b]	0.024[b]	0.0[b]
Other washed coal[g]	46.0[b]	12.0[b]	0.024[b]	0.0[b]
Sub-bituminous coal	29.0[a]	9.3[c]	0.174[a]	0.0[a]
Lignite	29.0[a]	9.3[c]	0.174[a]	0.0[a]
Coke oven coke[h]	12.0	5.08	0.072	0.0
Diesel oil	0.49[a]	0.186[d]	0.147[a]	0.0441[a]
Crude oil[i]	1.1	0.775	0.088	0.033
Heavy fuel oil	1.1[a]	0.775[d]	0.088[a]	0.033[a]
Fuelwood	2.2[e]	1.79[e]	0.11[e]	0.44[e]
Crop residue[j]	2.2	1.79	0.11	0.44
Animal waste[j]	2.2	1.79	0.11	0.44
Charcoal	4.1[e]	3.32[e]	0.205[e]	0.82[e]

a. Bond et al. (2004). b. Lei et al. (2011). c. $PM_{2.5}/PM_{10}$ ratios were estimated based on AP-42 (US UPA, 1995). d. $PM_{2.5}/PM_{10}$ ratios were estimated based on Klimont et al. (2002b). e. Emission factors of PM_{10}, BC, and OC for fuelwood and charcoal were taken from Bond et al. (2004). $PM_{2.5}/PM_{10}$ ratios were estimated based on the global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012). f. Emission factors were commonly used for coking coal, anthracite and bituminous coal. g. Only defined for China. h. Emission factors for hard coal were adopted. i. Emission factors for heavy fuel oil were adopted. j. Emission factors for fuelwood were adopted.

Table 3.13. Default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC from fuel combustion in industry sector. Unit is t/kt.

Fuel type	PM_{10}	$PM_{2.5}$	BC	OC
Hard coal[f]	4.2[a]	1.79[c]	0.84[a]	0.168[a]
Raw coal[g]	7.21[b]	2.17[b]	0.412[b]	0.0868[b]
Cleaned coal[g]	7.21[b]	2.17[b]	0.412[b]	0.0868[b]
Other washed coal[g]	7.21[b]	2.17[b]	0.412[b]	0.0868[b]
Sub-bituminous coal	17.0[a]	7.23[c]	0.85[a]	1.7[c]
Lignite	17.0[a]	7.23[c]	0.85[a]	1.7[c]
Coke oven coke[h]	4.2	1.79	0.84	0.168
Kerosene	0.9[a]	0.341[d]	0.117[a]	0.09[a]
Diesel oil	0.49[a]	0.186[d]	0.147[a]	0.0441[a]
Crude oil[i]	1.1	0.775	0.088	0.033
Heavy fuel oil	1.1[a]	0.775[d]	0.088[a]	0.033[a]
Fuelwood	6.1[e]	4.95[e]	0.555[e]	3.22[e]
Crop residue[j]	6.1	4.95	0.555	3.22
Animal waste[j]	6.1	4.95	0.555	3.22
Charcoal	4.1[e]	3.32[e]	0.205[e]	0.82[e]

a. Bond et al. (2004). b. Estimated based on Lei et al. (2011) and Streets et al. (2006). c. $PM_{2.5}/PM_{10}$ ratio was estimated based on the global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012). OC/BC ratio was assumed based on ABC Emission Inventory Manual (Shrestha et al., 2013). d. $PM_{2.5}/PM_{10}$ ratios were estimated based on Klimont et al. (2002b). e. Emission factors of PM_{10}, BC, and OC for fuelwood and charcoal were taken from Bond et al. (2004). $PM_{2.5}/PM_{10}$ ratios were estimated based on the global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012). f. Emission factors were commonly used for coking coal, anthracite and bituminous coal. g. Only defined for China. h. Emission factors for hard coal were adopted. i. Emission factors for heavy fuel oil were adopted. j. Emission factors for fuelwood were adopted.

Table 3.14. Default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC from fuel combustion in residential sector. Unit is t/kt.

Fuel type	PM_{10}	$PM_{2.5}$	BC	OC
Hard coal[i]	7.4[a]	4.49[a]	1.02[a]	2.15[a]
Raw coal[j]	8.82[b]	6.86[b]	1.56[b]	3.29[b]
Cleaned coal[j]	8.82[b]	6.86[b]	1.56[b]	3.29[b]

Other washed coal[j]	8.82[b]	6.86[b]	1.56[b]	3.29[b]
Sub-bituminous coal	4.6[c]	2.79[c]	0.636[c]	1.334[c]
Lignite	4.6[c]	2.79[c]	0.636[c]	1.334[c]
Coke oven coke[k]	7.4	4.49	1.02	2.15
LPG	0.52[d]	0.197[d]	0.0676[d]	0.052[d]
Kerosene	0.9[d]	0.341[d]	0.117[d]	0.09[d]
Diesel oil	0.49[d]	0.186[d]	0.147[d]	0.0441[d]
Crude oil[l]	1.1	0.775	0.088	0.033
Heavy fuel oil	1.1[d]	0.775[d]	0.088[d]	0.033[d]
Fuelwood	5.76[e], 4.80[f]	5.58[e], 4.60[f]	1.12[e], 0.85[f]	4.46[e], 3.20[f]
Crop residue	7.21[e], 6.01[f]	6.98[e], 5.75[f]	1.05[e], 0.95[f]	3.98[e], 3.70[f]
Animal waste	9.8[g]	9.8[g]	0.4[g]	3.1[g]
Charcoal	4.1[h]	3.32[h]	0.205[h]	0.82[h]

a. Estimated based on PM_{10} emission factors for residential sectors in Bond et al. (2004) and ratios of $PM_{2.5}$, BC, and OC to PM_{10} in Lei et al. (2011). b. Estimated based on emission factors for stove in Lei et al. (2011). c. Emission factor for PM_{10} derived from Bond et al. (2004) and ratios of $PM_{2.5}$, BC, and OC to PM_{10} were from those for hard coal. d. Bond et al. (2004) for PM_{10}, BC, and OC and $PM_{2.5}/PM_{10}$ ratios were estimated based on Klimont et al. (2002b). e. Estimated based on Lei et al. (2011) and used for East Asian countries. f. Estimated based on Pandy et al. (2014) and used for Southeast and South Asian countries. g. Estimated based on Pandy et al. (2014) and commonly used for all countries. h. Emission factors of PM_{10}, BC, and OC were taken from Bond et al. (2004). $PM_{2.5}/PM_{10}$ ratios were estimated based on the global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012). i. Emission factors were commonly used for coking coal, anthracite and bituminous coal. j. Only defined for China. Values were gradually decreased from 1990 until their two third by 2005 referring Lei et al. (2011). k. Emission factors for hard coal were adopted. l. Emission factors for heavy fuel oil were adopted.

Coke production and iron and steel industry

The same as for CO, in REASv3, emissions of PM species from coke ovens were also estimated base on production amounts of coke oven coke (see Sect. S4.2.8). Emission factors of PM species for coal consumption in coke ovens were assumed to be zero assuming their contribution were included in the emissions estimated based on production amounts of coke described in Sect. S4.2.8. For China, emissions of PM species from iron and steel production were also estimated base on production amounts of sinter, pig iron, and crude steel (see Sect. S4.2.1). It was assumed that emission factors for sinter and pig iron production obtained from Lei et al (2011) include emissions from coal combustion. Therefore, emission factors of PM species for coal combustion in iron and steel industry were assumed to be zero for China.

Cement industry

Emissions of PM species in China and Japan were not estimated based on fuel consumption, but using amount of cement production in each kiln type. For China, emission factors (t/kt of cement produced) of PM_{10}/$PM_{2.5}$/BC/OC were estimated based on Hua et al. (2016) and Lei et al. (2011) as follows: 44.8/19.2/0.115/0.192 for precalciner kilns, 37.3/14.9/0.0894/0.149 for other rotary kilns, and 8.9/3.2/0.0192/0.032 for shaft kilns. For Japan, emission factors of PM_{10}/$PM_{2.5}$/BC/OC (t/kt of clinker produced) were taken from AP-42 (US EPA, 1995) and Kupiainen and Klimont (2004) as follows: 15.6/4.55/0.0273/0.0455 for wet process kilns, 35.9/15.4/0.0924/0.154 for long dry process kilns, 54.6/23.4/0.140/0.234 for preheater process kilns and preheater/precalciner kilns. Ratio of clinker to cement was assumed to be 0.85 based on Cement handbook (Japan Cement Association, 2019). (See Sect. S4.1.3 for production data by different kiln types.). For other countries and regions, default emission factors in industry were used for all fuel types. See Sect. S4.2.3 for non-combustion emissions from cement production.

Brick industry

Emissions of PM species from brick production were not estimated based on fuel combustion, but using amount of brick production. Emission factors of PM_{10}/$PM_{2.5}$/BC/OC were assumed referring Lei et al. (2011), Weyant et al. (2014), and Klimont et al. (2017) as follows:

- China: 0.71/0.27/0.108/0.0945 t/kt of brick produced
- Japan, Republic of Korea, and Taiwan: 0.473/0.18/0.002/0.0035 t/kt of brick produced
- Other countries: 0.5/0.19/0.15/0.007 t/kt of brick produced

Settings of emission controls

Settings and assumptions for reduction of emissions of PM species from combustion sources by abatement equipment adopted in REASv3 are summarized in Table 3.15. For other sources not described in Table 3.15, no emission controls were considered.

Table 3.15. Settings and assumptions of emission controls of PM species

Countries	Settings and assumption
China	● Power plants ➢ Effects of control technologies by cyclones, wet scrubbers, electrostatic precipitators (ESP), and fabric filters during 1990-2015 were estimated based on their penetration rates in Lei et al. (2011) and Zhao et al. (2014). ➢ Reduction rates of $PM_{10}/PM_{2.5}$ were assumed to be 0.84/0.62, 0.92/0.78, and 0.98/0.94, and in 1990, 2000, and 2015, respectively. It was assumed that reduction rates before 1970 were zero and the values between 1970 and 1990 were interpolated. ● Industry ➢ Iron and steel industry: See Sect. S4.2.1 ➢ Coke ovens: See Sect. S4.2.8. ➢ Non-ferrous metals industry: See Sect. S4.2.2 ➢ Cement industry: See Sect. S4.2.3. ➢ Lime industry: See Sect. S4.2.4. ➢ Brick industry: See Sect. S4.2.5. ➢ Other industries: Due to lack of information, reduction rates were roughly assumed as follows: Reduction rates of PM_{10} and $PM_{2.5}$ in 1990 were 0.55 and 0.25 referring settings of cement industry. Those in 2015 were 0.77 and 0.53 referring Wang et al. (2014) for settings of industry in 2010. It was assumed that reduction rates before 1980 were zero and the values between 1980, 1990, and 2015 were interpolated.
India	● Due to lack of information, referring Sadavarte and Venkataraman (2014), Pandey et al. (2014), Guttikunda and Jawahar (2014), and Reddy and Venkataraman (2002), reduction rates of $PM_{10}/PM_{2.5}$ for power plants and industries during 1980-2015 were roughly assumed as follows:

	➢ Power plants: 0.0/0.0, 0.45/0.40, 0.85/0.81, and 0.87/0.85 in 1980, 1985, 2000, and 2015, respectively. Values between 1980, 1985, 2000, and 2015 were interpolated. ➢ Iron and steel and cement industries: 0.0/0.0, 0.47/0.46, and 0.85/0.83 in 1980, 1995, and 2015, respectively. Values between 1980, 1995, and 2015 were interpolated. ➢ Other industries: 0.0/0.0, 0.40/0.30, and 0.45/0.40 in 1980, 1995, and 2015, respectively. Values between 1980, 1995, and 2015 were interpolated.
Japan	● Referring MRI (2015) and other literatures such as Shimoda (2016), Suzuki (1990) and Goto (1981), following assumptions were considered for control equipment of PM species: ➢ Introduction of control equipment for power plants was expanded from 1957. ➢ Introduction of bag filter was expanded from 1960. ➢ From 1968, installation of ESP in power plants became mandatory. ➢ Introduction of high quality ESP was expanded from 1975. ➢ Regulations for PM species were strengthened from 1995. ● Based on above assumption, reduction rates of $PM_{10}/PM_{2.5}$ for power plants were assumed as follows: 0.37/0.27, 0.9/0.88, and 0.995/0.99 in 1960, 1975, and after 2000, respectively. It was assumed that reduction rates before 1956 were zero and the values between 1950, 1960, 1975, and 2000 were interpolated. ● For industry, reduction rates of $PM_{10}/PM_{2.5}$ after 2000 were assumed to be 0.99/0.985 for iron and steel and cement industries and 0.98/0.96 for other industries. Trends between 1950 and 2000 were assumed to be the same as for those of power plants.
Republic of Korea/Taiwan	● Power plants: Based on Wang et al. (2014), reduction rates of PM_{10} and $PM_{2.5}$ after 2005 were assumed to be 0.985 and 0.97, respectively. Referring Ebata et al. (1997), it was assumed that penetration rates of control equipment of PM species in 1990 were already high. Reduction rates in 1990 were assumed to be 0.9 and 0.88 for PM_{10} and $PM_{2.5}$, respective and zero before 1970. Values between 1970, 1990, and 2005 were interpolated. ● Industry: Based on Wang et al. (2014), reduction rates of $PM_{10}/PM_{2.5}$

	in 2005 and in 2010 were assumed to be 0.944/0.905 and 0.948/0.910, respectively. It was roughly assumed that reduction rates of $PM_{10}/PM_{2.5}$ in 2015 were 0.968/0.935, respectively and zero before 1970. Values between 1970, 2005, 2010, and 2015 were interpolated. ● Due to lack of information, the same settings for Republic of Korea were adopted.
Thailand	● Power plants: Referring Thao Pham et al. (2008), reduction rates of PM_{10} and $PM_{2.5}$ in 2000 were assumed to be 0.84 and 0.80, respectively. For trends of reduction rates, it was roughly assumed that reduction rates of PM_{10} and $PM_{2.5}$ were increased to 0.90 and 0.88 in 2015, respectively and zero before 1980. Values between 1980, 2000, and 2015 were interpolated. ● Industry: Referring Thao Pham et al. (2008), for iron and steel and cement industries, reduction rates of PM_{10} and $PM_{2.5}$ in 2005 were assumed to be 0.82 and 0.80, respectively. For trends of reduction rats, it was roughly assumed that reduction rates of PM_{10} and $PM_{2.5}$ in 2015 were 0.85 and 0.83, respectively and zero before 1980. Values between 1980, 2000, and 2015 were interpolated.
Others	● Due to lack of information, settings of Thailand during 1980-2005 were adopted for those of Indonesia, Malaysia, Myanmar, Philippines, Vietnam and Mongolia during 1990-2015 and the same settings of Thailand were used for Singapore. ● For Laos and Sri Lanka, reduction rates of 0.95/0.92 for $PM_{10}/PM_{2.5}$ were used for large power plants equipped with ESP based on information from World Electric Power Plants Database (Platts, 2018),

S3.2.5 Other species and sources

NMVOC

Emission factors for fossil fuel combustion were taken from REASv2 based on Wei et al. (2008) for East Asian countries and the global atmospheric pollution forum air pollutant emission inventory manual (SEI, 2012) for Southeast and South Asian countries. For fuelwood, crop residue, and animal waste, emission factors were estimated as follows:

- Fuelwood
  - 3.13 t/kt based on Wei et al. (2008) for East Asian countries
  - 15.9 t/kt based on Sharma et al. (2015) for Southeast and South Asian countries
- Crop residue
  - 8.36 t/kt based on Wei et al. (2008) for East Asian countries
  - 13.3 t/kt based on Sharma et al. (2015) for Southeast and South Asian countries
- Animal waste
  - 10.4 t/kt based on Sharma et al. (2015) for all countries
- Charcoal
  - 100 t/PJ taken from IPCC (1997) for all countries

Emission factors described above were for total NMVOC. In REASv3, total NMVOC emissions were allocated to 19 NMVOC species categories defined in Sect. S2.1. The speciation was conducted based on speciation profiles for each sub-sector and fuel type provided by D. G. Streets (private communication) generally based on Klimont et al. (2002a) used for REASv1 and REASv2. The speciation profiles were commonly used for all countries and periods.

NH_3

Emission factors for fossil fuel combustion were taken from REASv1 based on EMEP/CORINAIR Emission Inventory Guidebook (EEA, 1996). For biofuel, 1.29 t/kt for fuelwood and 0.97 t/kt for charcoal were obtained from ABC Emission Inventory Manual (Shrestha et al., 2013). Due to lack of information, the emission factor for fuelwood was adopted to crop residue and animal waste.

CO_2

Emission factors for fuel combustion except for fuelwood, crop residue, and animal wastes were obtained from 2006 IPCC Guidelines for National Greenhouse Gas Inventories (IPCC, 2006). Default emission factors were used except for those of coal combustion in China where lower values were adopted referring Guan et al. (2012). Emission factors for fuelwood, crop residue, and animal wastes were 83.1, 87.0, and 76.9 kt/PJ derived from Streets and Waldhoff (1999).

Agriculture

For emissions from fuel combustion in agriculture sub-sector, emission factors of industry sector were used except for following settings for diesel oil referring Bond et al. (2004) and ABC Emission Inventory Manual (Shrestha et al., 2013):

- 50.3 t/kt for NO_x
- 16.0 t/kt for CO
- 2.0 t/kt for PM_{10}
- 1.72 t/kt for $PM_{2.5}$
- 1.14 t/kt for BC
- 0.36 t/kt for OC

Charcoal production

Activity data to estimate emissions from charcoal production as energy transformation sectors is wood input. Fuelwood consumption data developed based on methodologies described in Sect. S3.1 were used. Emission factors of NO_x, CO, and NMVOC were taken from Revised 1996 IPCC guidelines (IPCC, 1997) and those of others were based on Akagi et al. (2011).

S4. Stationary non-combustion: Industrial production and other transformation

Descriptions for evaporative NMVOC emissions and NH_3 emissions from non-combustion sources are provided in Sects. S5 and S8, respectively.

S4.1 Activity data

S4.1.1 Iron and steel production

Activity data to estimate non-combustion emissions from iron and steel production industry in REASv3 are production amounts of pig iron, crude steel, sinter, and hot rolled products. National total production of pig iron, crude steel, and hot rolled products were obtained from Steel Statistical Yearbook (World Steel Association, https://www.worldsteel.org/steel-by-topic/statistics/steel-statistical-yearbook.html) during 1968-2015 and extrapolated to 1950 using trends of pig iron and crude steel production in Mitchell (1998). For crude steel, production data by each process, oxygen-blown converter, electric furnace, and open-hearth furnace were separately obtained. Sinter production data were taken from Steel Statistical Yearbook during 1977-1992. For China, sinter production data were available during 2000-2015 and those between 1992 and 2000 were interpolated. Then, missing data between 1950 and 2015 were estimated based on trends of pig iron production in each country.

For regional distribution in China, production amounts of steel during 1950-2015 and pig iron during 1983-2015 in each region were available in China Data Online and China Statistical Yearbook (National Bureau of Statistics of China, 1986-2016), respectively. Pig iron data before 1982 were extrapolated for each region using the trends of steel production in China Data Online. Then, using the steel data, production amounts of crude steel and hot rolled products in China total were distributed to each region. Similarly using the regional pig iron data, sinter and pig iron production amounts in whole China were distributed to each region. For India, ratios of crude steel production in 17 sub-regions were estimated using Minerals Yearbook (United States Geological Survey (USGS)) and Indiastat during 2000-2015. Using the regional data, production amounts of pig iron, crude steel, singer, and hot rolled products in India total were distributed to each sub-region. For Japan, ratios of steel production amounts in 6 sub-regions during 2003 and 2011 were estimated using statistics of major factories (https://www.japanmetaldaily.com/statistics/crudemateworks/details/index.html) and production data of pig iron, crude steel, singer, and hot rolled products in India total were distributed to each sub-region.

S4.1.2 Non-ferrous metal production

In REASv3, non-combustion emissions from copper, zinc, lead, and aluminum production were considered in non-ferrous metal production processes. Activity data were production amounts of primary copper, zinc, lead, alumina, aluminum, and secondary aluminum obtained from Minerals Yearbook during 1960-2015 (USGS) and extrapolated to 1950 using trends of corresponding production data in Mitchell (1998). For China, India, and Japan, national total data need to be distributed to each sub-region. Weighting factors for the distribution were estimated during 1995-2015 using annual generation capacities of major plants in Minerals Yearbook (USGS). Before 1994, the weighting factors for 1995 were used.

S4.1.3 Cement production

Activity data for non-combustion emissions from cement industry are production amounts of cement. For China, regional data were basically available in China Data Online during 1950-2015. However, not all regions had complete data during the period and sometimes interpolation and extrapolation procedures were necessary. Therefore, in REASv3, regional data were used for weighting factors to distribute national total data of cement production to each sub-region. For Japan, national cement production during 1990-2015 were obtained from Minerals Yearbook (USGS) and extrapolated to 1950 using trends of corresponding data in the Historical Statistics of Japan (Japan Statistical Association, 2006). For the distribution to each sub-region, first, weighting factors in 2004 and 2018 were estimated using production amounts by major cement plants. Then, those during 2005-2015 were interpolated and data in 2004 were used before 2003. In addition to total amounts, production data by different kiln types were available in China (Hua et al., 2016) and Japan (Japan Cement Association, http://www.jcassoc.or.jp/cement/2eng/index.html). For other countries, national total production during 1960-2015 were obtained from Minerals Yearbook (USGS). For extrapolation to 1950, in REASv3, trends of national CO_2 emissions from cement production taken from CDIAC (Carbon Dioxide Information Analysis Center) (Marland et al., 2008). For regional data in India, weighting factors during 1984 and 2009 were estimated using regional production data in TERI Energy & Environment Data Diary and Yearbook (TERI, 2013, 2018). Before 1983 and after 2010, data in 1984 and 2009 were used, respectively.

S4.1.4 Lime production

Activity data for non-combustion emissions from lime industry are production amounts of lime. Data were obtained from Minerals Yearbook during 1960-2015 (USGS) and were extrapolated to 1950 using trends of cement production estimated in REASv3.

S4.1.5 Brick production

Activity data for non-combustion emissions from brick industry are production amounts of brick. However, unlike the other products in non-metallic minerals industry, brick production data were not available in most international and national statistics. For Japan, national production data during 1950-2007 were taken from Hiragushi (2009) and Japan Statistical Yearbook (Statistics Bureau, 2010-2018) and were distributed to 6 sub-regions using total fuel consumption in non-metallic minerals sector. For other countries, first, default data were prepared taken from REASv2 and GAINS ASIA at that time during 1990-2015 and extrapolated to 1950 using trends of cement production in each country. For China, Vietnam, Bangladesh, India, and Pakistan, national production data in 1990, 2000, 2005, and 2010 were obtained from Klimont et al. (2017) and interpolated during 1990-2010 and extrapolated to 2015 using trends of the default data. For China, data between 1980-1990 were extrapolated based on trends of production in Zhang (1997) and those before 1980 were extrapolated using trends of the default data. For regional distribution, fuel consumption data in brick production in each region (see Sects. S3.1.3 and S3.1.7) were used for weighting factors. For India, data between 1983-1990 were extrapolated based on trends of production in Industrial Commodity Statistical Yearbook taken from UN data, which is a web-based data service of the UN (http://data.un.org/) and those before 1983 were extrapolated using trends of the default data. For regional distribution, common weighting factors during 1950-2015 were estimated based on Maithel et al. (2012). For Vietnam, Bangladesh, and Pakistan, data before 1990 were extrapolated using trends of the default data. For Nepal, production data in 2006 were obtained from Maithel (2013) and extrapolated during 1950-2015 using trends of the default data. For Rep. of Korea, Indonesia, Myanmar, the default data were used during 1990-2015 and before 1990, data were extrapolated to 1985 using trends of production in Industrial Commodity Statistical Yearbook and then extended to 1950 using trends of the default data. For other countries, the default data were directly used.

S4.1.6 Sulphuric acid production

Activity data to estimate non-combustion emissions from sulphuric acid plants are amounts of total sulphuric acid production in each country and region. For China, national total production data during 1950-2015 were obtained from China Data Online and distributed to each region using regional data during 1983-2015 in China Statistical Yearbook (National Bureau of Statistics of China, 1986-2016). Before 1983, data in 1983 were used as weighting factors for the regional distribution. For Japan, national production data were taken from statistics provided by the Sulphuric Acid Association of Japan (http://www.ryusan-kyokai.org/) during 1983-2015 and extrapolated to 1950 using trends of sulphuric acid production in Mitchell (1998). Weighting factors for regional distribution were estimated using annual generation capacities of major plants in 2015 in Minerals Yearbook (USGS). For other countries, national total production data were provided by the Sulphuric Acid Association of Japan during 1980-2015 and extrapolated to 1950 using trends of sulphuric acid production in Mitchell (1998). For India, national total data were distributed to 17 sub-regions using data of REASv2 during 2000-2008 based on GAINS ASIA at that time. For the weighting factors, data in 2000 and 2008 were used before 2000 and 2008, respectively.

S4.1.7 Carbon black production

In REASv3, non-combustion emissions from carbon black production were only considered for China, India, Japan, and, Rep. of Korea. Similar to brick production, default data were prepared taken from REASv2 and GAINS ASIA at that time during 1990-2015 and extrapolated to 1950 using GDP in each country and region. For GDP, regional data in China during 1950-2015 were obtained from China Data Online. For other countries, data during 1970-2015 were derived from UN data, which is a web-based data service of the UN (http://data.un.org/) and extrapolated to 1960 using OECD Data (https://data.oecd.org/gdp/gross-domestic-product-gdp.htm) and then extrapolated to 1950 using trends of total population.

For China, national total production in 2010 were obtained from Wei et al. (2011) and were extrapolated during 1950-2015 and distributed to each region using the default data as weighting factors. For India, national production data during 1983-2003 were taken from Industrial Commodity Statistical Yearbook taken from the UN data and similar to China, the data were extrapolated during 1950-2015 and distributed to each region using the default data. For Japan and Rep. of Korea, national production data during 1964-2014 were obtained from Mineral Yearbook (USGS) and extrapolated during 1950-2015 and data in Japan were distributed to 6 sub-regions using the default data.

S4.1.8 Other transformation sectors

Coke ovens

In REASv3, activity data to estimate emissions from coke ovens as energy transformation sectors are coal input for SO_2 and NO_x and coke production for CO, NMVOC, CO_2, and PM species. Coal consumption was taken from data developed based on methodologies described in Sect. S3.1. For coke production, national data were obtained from the International Energy Agency (IEA) World Energy Balances (IEA, 2017) during 1960-2015 for Japan and 1971-2015 for other countries. The data were extrapolated to 1950 based on trends of pig iron production before 1959 and 1970 for Japan and other countries, respectively. For China, regional production data during 1990-2015 were available in the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017) and used to distribute national total production data to each sub-region. Before 1990, data in 1990 were used. For India and Japan, weighting factors for the regional distribution were based on regional pig iron production data in each country.

Petroleum refineries

Activity data to estimate emissions from petroleum refineries as energy transformation sectors is crude oil input. Consumption data of crude oil developed based on methodologies described in Sect. S3.1 were used.

S4.2 Emission factors and settings of emission controls

S4.2.1 Iron and steel production

Emission factors

In REASv3, emissions of CO, NMVOC, CO_2, and PM species were estimated using production amounts of sinter, pig iron, crude steel, and rolled steel. Default emission factors are summarized in Table 4.1 and emission factors of PM species for China are provided in Table 4.2. Note that emission factors of CO for all countries and those of PM species for China include contributions from both combustion and non-combustion emissions. (See also Sects. S3.2.3 and S3.2.4.)

Table 4.1. Default emission factors of CO, NMVOC, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC from production of sinter, pig iron, crude steel, and rolled steel. It was assumed that both combustion and non-combustion emissions are included in emission factors of CO. Unit is t/kt-produced.

	Sinter	Pig iron	Crude steel/ OHF[a]	Crude steel/ BOF[a]	Crude steel/ EF[a]	Rolled steel
CO	22.0[b]	40.5[c]	34.5[d]	69.0[b]	9.0[b]	-
NMVOC[e]	-	-	0.055	0.055	0.055	0.025
CO_2[f]	-	-	-	-	80.0	-
PM_{10}[g]	1.555	0.490	8.760	14.63	10.18	-
$PM_{2.5}$[g]	0.691	0.300	6.330	10.45	7.550	-
BC[h]	0.005	0.018	-	-	-	-
OC[h]	0.026	-	-	2.090	0.180	-

a. OHF: Open-hearth furnace, BOF: Basic oxygen furnace, and EF: Electric furnace. b. AP-42 (US EPA, 1995), c. Streets et al. (2006), d. 50% of BOF was adopted. e. Klimont et al. (2002a). f. IPCC (2006). g. Klimont et al. (2002b). h. Kupiainen and Klimont (2004).

Table 4.2. Emission factors of PM_{10}, $PM_{2.5}$, BC, and OC from production of sinter, pig iron, crude steel, and rolled steel for China. It was assumed that both combustion and non-combustion emissions are included (except for emission factors of PM species for crude steel production). Unit is t/kt-produced.

	Sinter	Pig iron	Crude steel/ OHF[a]	Crude steel/ BOF[a]	Crude steel/ EF[a]	Rolled steel
CO[b]	22.00	40.50	27.10[d]	54.20	9.000	-
PM_{10}[c]	6.050	9.650	19.10	14.63	8.120	-
$PM_{2.5}$[c]	2.620	6.000	13.80	10.45	6.020	-
BC[c]	0.0262	0.600	0.138	-	-	-
OC[c]	0.131	0.120	0.690	2.090	0.120	-

a. OHF: Open-hearth furnace, BOF: Basic oxygen furnace, and EF: Electric furnace. b. Streets et al. (2006). c. Lei et al. (2011). d. 50% of BOF was adopted.

For CO, the gas from blast furnace and basic oxygen furnace is collected and recycled in modern factories (Streets et al., 2006) and in REASv1, corresponding CO emissions in Japan were neglected. In REASv3, following settings were roughly assumed:

- China: Emission factors in Table 4.2 were used during 1950-2000 and 50% of the value was adopted in 2015. Emission factors between 2000 and 2015 were interpolated.
- Japan: Default emission factors were used before 1960 and 10% of the value was adopted in

1990. Emission factors between 1960 and1990 were interpolated.

- Republic of Korea and Taiwan: Default emission factors were used before 1975 and 10% of the value was adopted in 2005. Emission factors between 1975 and 2005 were interpolated.

Settings of emission controls

For iron and steel production, emission controls were only considered for PM species. Settings and assumptions for reduction of emissions in China by abatement equipment adopted in REASv3 are summarized in Table 4.3. For other countries, the same settings for combustion emissions in iron and steel industry were adopted. (See Table 3.15 in Sect. S3.2.4.)

Table 4.3. Settings and assumptions of emission controls of PM species for iron and steel production in China.

Countries	Settings and assumption
China	● Referring Wu et al. (2017), reduction rates of $PM_{10}/PM_{2.5}$ for sinter production, pig iron, BOF, and EF in 2000, 2005, 2010, and 2015 were assumed as follows ➢ Sinter: 0.780/0.592, 0.892/0.809, 0.946/0.916, and 0.956/0.939 ➢ Pig iron: 0.850/0.715, 0.910/0.844, 0.954/0.936, and 0.961/0.945 ➢ BOF: 0.850/0.715, 0.870/0.758, 0.955/0.937, and 0.959/0.943 ➢ EF: 0.782/0.568, 0.834/0.678, 0.900/0.815, and 0.977/0.968 ● It was assumed that reduction rates were zero in 1980 and values between 1980, 2000, 2005, 2010, and 2015 were interpolated.

S4.2.2 Non-ferrous metal production

In REASv3, emissions of SO_2, PM_{10}, and $PM_{2.5}$ were estimated using production amounts of copper, zinc, lead, and aluminum.

SO_2

Default emission factors were taken from Kato and Akimoto (1992) as follows:

- Copper: 2.0 kt/kt- produced
- Zinc: 1.0 kt/kt-produced
- Lead: 0.32 kt/kt-produced

In some countries, SO_2 emitted from non-ferrous metal plants were collected and used for materials of sulphuric acid. In that case, the amounts of collected SO_2 need to be reduced from SO_2 emissions calculated by default emission factors. In REASv3, amounts of sulphuric acid produced using SO_2 collected from non-ferrous metal plants were obtained from the Sulphuric Acid Association of Japan based on reports of International Fertilizer Industry Association, the British Sulphur Cooperation Limited, Sulphuric Acid Notebook of Japan, and Kato et al. (1991). In addition, the same reduction rates of SO_2 by emission control equipment for non-ferrous metal industry were adopted.

PM_{10} and $PM_{2.5}$

Default emission factors t/kt-produced were obtained from Lei et al. (2011) for China and Klimont et al. (2002b) for other countries as follows:

China:

- Copper, Zinc, and Lead: 276.0 for PM_{10} and 246.0 for $PM_{2.5}$
- Aluminum (primary): 26.51 for PM_{10} and 18.28 for $PM_{2.5}$
- Aluminum (secondary): 6.98 for PM_{10} and 5.20 for $PM_{2.5}$

Other countries:

- Copper, Zinc, and Lead: 13.8 for PM_{10} and 12.3 for $PM_{2.5}$
- Aluminum (primary): 27.26 for PM_{10} and 18.5 for $PM_{2.5}$
- Aluminum (secondary): 6.97 for PM_{10} and 5.195 for $PM_{2.5}$

For emission controls, the same settings for combustion emissions in industry sectors were adopted except for China. (See Table 3.15 in Sect. S3.2.4.) For China, reduction rates were assumed as follows:

- Referring Zhao et al. (2014), reduction rates of PM_{10}/$PM_{2.5}$ in 2010 and 2015 were 0.910/0.882 and 0.945/0.906, respectively and values between 2010 and 2015 were interpolated.
- Trends of reduction rates between 1980 and 2010 were assumed to be the same as settings for combustion emissions in other industries. (See Table 3.15 in Sect. S3.2.4.)

S4.2.3 Cement production

In REASv3, emissions of CO_2 and PM species for all countries and those of NO_x and CO for Japan were estimated using production amounts of cement. For emission of NO_x and CO in Japan and those of PM species in China and Japan, emission factors for combustion emissions were described in Sects. S3.2.2, S3.2.3 and S3.2.4, respectively. In this sub-section, emission factors for non-combustion emissions were described.

Default emission factor of CO_2 was 0.52 t/t-clinker produced based on IPCC (2006). Clinker to cement ratios were roughly assumed as follows:

- China: 0.72 before 2005 and 0.6 in 2015 based on Gao et al. (2017). Values between 2005 and 2015 were interpolated.
- India: 0.83 before 1990 and 0.77 after 2005 based on Barcelo (2014). Values between 1990 and 2015 were interpolated.
- Japan: 0.85 base on Cement handbook (Japan Cement Association, 2019)
- Others: 0.9 before 1990 and 0.85 after 2005 based on Barcelo (2014). Values between 1990 and 2015 were interpolated.

For PM species, default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC t/kt-produced were assumed as follows:

- China: 34.3, 9.8, 0.0588, and 0.098 were taken from Hua et al. (2016) and Lei et al. (2011).
- Others: 16.0, 4.64, 0.0278, and 0.0464 were derived from AP-42 (US EPA, 1995) and Lei et al. (2011).

For emission controls, the same settings for combustion emissions in cement industry were adopted except for China. (See Table 3.15 in Sect. S3.2.4.) For China, reduction rates were assumed as follows:

- Referring Hua et al. (2016), reduction rates of PM_{10}/$PM_{2.5}$ during 1980-2012 were estimated for each year. Values were 0.565/0.218, 0.586/0.250, 0.746/0.527, and 0.973/0.916 in 1980, 1990, 2000, and 2012, respectively.
- It was roughly assumed that reduction rates of PM_{10}/$PM_{2.5}$ in 2015 were 0.98/0.97 and zero in 1975. Values between 1975 and 1980 and those between 2010 and 2015 were interpolated.

S4.2.4 Lime production

In REASv3, emissions of CO_2 and PM species were estimated using production amounts of lime. Default emission factors of CO_2 were taken from IPCC (2006) and those of PM species were derived from Klimont et al. (2002b) and Kupiainen and Klimont (2004) as follows:

- CO_2: 750 t/kt-produced
- PM_{10}: 12.0 t/kt-produced
- $PM_{2.5}$: 1.4 t/kt-produced
- BC: 0.028 t/kt-produced
- OC: 0.014 t/kt-produced

For emission controls of PM species, the same settings for combustion emissions in industry sectors were adopted except for China. (See Table 3.15 in Sect. S3.2.4.) For China, reduction rates were assumed as follows:

- Referring Zhao et al. (2014), reduction rates of PM_{10}/$PM_{2.5}$ in 2010 and 2015 were 0.766/0.670

and 0.782/0.697, respectively and values between 2010 and 2015 were interpolated.

- Trends of reduction rates between 1985 and 2010 were assumed to be the same as settings between 1980 and 2005 for combustion emissions in other industries. (See Table 3.15 in Sect. S3.2.4.)

S4.2.5 Brick production

In REASv3, emissions of CO and PM species were estimated using production amounts of brick.

For CO, note that emissions in China, Japan, Republic of Korea, and Taiwan were estimated using fuel consumption as described in Sect. S3.2.3. For other countries, emissions were estimated with production amounts of brick and emission factor 2.0 t/kt-produced was taken from Weyan et al. (2014).

For PM species, default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC t/kt-produced were assumed as follows:

- China: 0.71, 0.27, 0.108, and 0.0945 were taken from Lei et al. (2011).
- Japan, Republic of Korea, and Taiwan: Emission factors of tunnel kiln 0.4773, 0.18, 0.002, and 0.0035 were obtained from Klimont et al. (2017).
- Others: Emission factors of Bull's trench kiln 0.5, 0.19, 0.15, and 0.007 were based on Weyant et al. (2014).

For emission controls of PM species, the same settings for combustion emissions in industry sectors were adopted except for China. (See Table 3.15 in Sect. S3.2.4.) For China, reduction rates were assumed as follows:

- Referring Zhao et al. (2014), reduction rates of PM_{10}/$PM_{2.5}$ in 2010 and 2015 were 0.425/0.208 and 0.362/0.143, respectively and values between 2010 and 2015 were interpolated.
- Trends of reduction rates between 1985 and 2010 were assumed to be the same as settings for combustion emissions in other industries. (See Table 3.15 in Sect. S3.2.4.)

S4.2.6 Sulphuric acid production

In REASv3, emissions of SO_2 were estimated using production amounts of sulphuric acid. Default emission factors were taken from Kato et al. (1991) as follows:

- 20.0 t/kt-produced for China, Japan, Republic of Korea, and Taiwan
- 33.0 t/kt-produced for other countries.

For emission controls, the same settings for combustion emissions in large industries were adopted for Japan, Republic of Korea, and Taiwan and those for other industries were applied for China. For other countries, no emission controls were considered.

S4.2.7 Carbon black production

In REASv3, emissions of NMVOC and PM species were estimated using production amounts of carbon black. Default emission factor of NMVOC was taken from Klimont et al. (2002a) and those of PM species were derived from Klimont et al. (2002b) and Kupiainen and Klimont (2004) as follows:

- NMVOC: 90 t/kt-produced
- PM_{10}: 1.60 t/kt-produced
- $PM_{2.5}$: 1.44 t/kt-produced
- BC: 1.10 t/kt-produced
- OC: 0.00 t/kt-produced

For emission controls of PM species, the same settings for combustion emissions in industry sectors were adopted for all countries. (See Table 3.15 in Sect. S3.2.4.)

S4.2.8 Other transformation sectors

Coke ovens

In REASv3, emissions of CO, NMVOC, CO_2, and PM species were estimated using production amounts of coke oven coke.

For CO, emission factors were taken from Streets et al. (2006) as follows:

- 1.6 t/kt-produced for machinery coke ovens
- 15.6 t/kt-produced for indigenous coke ovens

Production amounts of coke oven coke in different technologies were only considered for China. Ratios of production amounts between machinery and indigenous coke ovens in each province in 2005 and 2006 were taken from China Industrial Economy Statistics Yearbook (National Bureau of Statistics, 2006-2007) and were extrapolated based on national ratios during 1990-2011 obtained from Huo et al. (2012a). It was roughly assumed that ratios of machinery coke ovens in 1970 were zero and gradually increased from 2011 to 2015. Data between 1970 and 1990 were interpolated. Due to lack of information, emission factors for machinery coke ovens were adopted for all other countries. As described in Sect. S3.2.3, emission factors were assumed to include contribution from combustion emissions.

Default emission factors of NMVOC was taken from Klimont et al. (2002a) and that of CO_2 was obtained from IPCC (2006) as follows:

- NMVOC: 1.44 t/kt-produced
- CO_2: 560 t/kt-produced

For PM species, default emission factors of PM_{10}, $PM_{2.5}$, BC, and OC t/kt-produced were assumed as follows:

- China: 8.79, 5.22, 1.57, and 1.83 were taken from Lei et al. (2011).
- Others: 3.36, 2.00, 0.75, and 0.54 were taken from Klimont et al. (2002b) and Kupiainen and Klimont (2004).

As described in Sect. S3.2.4, emission factors were assumed to include contribution from combustion emissions. For emission controls of PM species, the same settings for combustion emissions in iron and steel industry were adopted except for China. (See Table 3.15 in Sect. S3.2.4.) For China, reduction rates were assumed as follows:

- Referring Zhao et al. (2014), reduction rates of $PM_{10}/PM_{2.5}$ in 2010 and 2015 were estimated for machinery and indigenous coke ovens as follows:
  - Machinery: 0.773/0.560 and 0.803/0.624 in 2010 and 2015, respectively.
  - Indigenous: 0.193/0.140 and 0.200/0.156 in 2010 and 2015, respectively.
  - Values between 2010 and 2015 were interpolated.
- Trends of reduction rates between 1985 and 2010 were assumed to be the same as settings for combustion emissions in other industries. (See Table 3.15 in Sect. S3.2.4.)

Petroleum refineries

In REASv3, emissions of SO_2, NMVOC and PM species were estimated using consumption amounts of crude oil in oil refinery industry. Default emission factors were derived from Kato and Akimoto (1992) for SO_2, Klimont et al. (2002a) for NMVOC, Klimont et al. (2002b) and Kupiainen and Klimont (2004) for PM species as follows:

- SO_2: 0.46S t/kt (S: Sulfur contents in fuel in wt%)
- NMVOC: 2.34 t/PJ
- PM_{10}: 1.20 t/kt
- $PM_{2.5}$: 0.96 t/kt
- BC: 0.00015 t/kt
- OC: 0.00 t/kt

For emission controls of SO_2 and PM species, the same settings for combustion emissions in industry sectors were adopted for all countries. (See Table 3.15 in Sect. S3.2.4.)

S4.2.9 Speciation of NMVOC emissions

Emission factors described in Sect. S4.2 were for total NMVOC. In REASv3, total NMVOC emissions were allocated to 19 NMVOC species categories defined in Sect. S2.1. The speciation was conducted based on speciation profiles for each sub-sector provided by D. G. Streets (private communication) generally based on Klimont et al. (2002a) used for REASv1 and REASv2. The speciation profiles were commonly used for all countries and periods.

S5. Non-combustion sources of NMVOC

In this section, activity data, emission factors, and their sources used to estimate evaporative NMVOC emissions in REASv3 are described. See Sect. S2.4.3 for sub-sector categories defined in REASv3. For Japan, NMVOC emissions from evaporative sources were derived from the Ministry of the Environment Japan (MEOJ, 2017a) and thus, activity data and emission factors of Japan were not compiled in REASv3 (see Sect. S5.3.1 for Japan).

S5.1 Activity data

In REASv3, activity data of REASv2 during 2000-2008 estimated based on Klimont et al. (2002a) were used as “default”.

S5.1.1 Extraction processes

In REASv3, emissions from gas production and distribution, oil production and handling, petroleum refineries, service stations, and transport and depots are included in those from extraction processes. Data sources and treatments of activity data for each sub-sector category used in REASv3 were summarized in Table 5.1.

Table 5.1. Data sources and treatments of activity data for sub-sectors of extraction processes.

Sub-sector categories	Data sources and treatments of activity data
Gas production and distribution	● Activity data: Natural gas production ● Data sources and treatments: ➢ China: Regional data during 1985-2015 were taken from the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017). Before 1985, data were extrapolated to 1971 using the International Energy Agency (IEA) World Energy Balances (IEAWEB) (IEA, 2017) and to 1950 using Mitchell (1998). ➢ India: National total data were obtained from IEAWEB and extrapolated to 1950 using Mitchel (1998). For regional distribution, weighting factors were calculated using regional data taken from TERI (2013, 2018). ➢ Other countries: National total data were derived from IEAWEB or the United Nations (UN) Energy Statistics Database (UN, 2016) and

	extrapolated to 1950 using Mitchel (1998).
Crude oil production and handling	● Activity data: Crude oil production ● Data sources and treatments: ➢ China: Regional data during 1950-2015 were derived from China Data Online. ➢ India: National total data were obtained from IEAWEB and extrapolated to 1950 using Mitchel (1998). For regional distribution, weighting factors were calculated using regional data taken from TERI (2013, 2018). ➢ Other countries: National total data were derived from IEAWEB or the UN Energy Statistics Database (UN, 2016) and extrapolated to 1950 using Mitchel (1998).
Petroleum refineries	● Activity data: Consumption of crude oil in petroleum refineries ● Data sources and treatments: See Sect. S3.1.
Service stations	● Activity data: Consumption of gasoline in road transport sector ● Data sources and treatments: See Sect. S3.1.
Transport and depots	● Activity data: Consumption of gasoline and diesel in road transport sector ● Data sources and treatments: See Sect. S3.1.

S5.1.2 Solvent use

In this sub-section, activity data of NMVOC evaporative emissions from solvent use except for printing (See Sect. S5.1.3) and paint application (See Sect. S5.1.4) were described. Data sources and treatments of activity data for each sub-sector category used in REASv3 were summarized in Table 5.2. (See Sect. S4.1.7 for data sources of GDP used in this sub-section.)

Table 5.2. Data sources and treatments of activity data for sub-sectors of solvent use.

Sub-sector categories	Data sources and treatments of activity data
Dry cleaning	● Activity data: Textiles cleaned ● Data sources and treatments: ➢ China: National total data in 2012 were taken from Wu et al. (2016) and extrapolated during 1950-2015 using trends of GDP. For regional distribution, urban population (see descriptions for domestic use of solvents in this table) were used as weighting factors.

	➢ India: National data in 2010 were based on Sharma et al. (2015) and extrapolated during 1950-2015 using trends of GDP. For regional distribution, urban population were used as weighting factors. ➢ Other countries: Default data were used and extrapolated during 1950-2015 using trends of GDP.
Degreasing operation	● Activity data: Solvent used ● Data sources and treatments: ➢ China: National total data in 2005 were taken from Wei et al. (2008). Regional distribution and extrapolation during 1950-2015 were conducted based on GDP. ➢ Other countries and regions: Default data were used during 2000-2008 and extrapolated during 1950-2015 using trends of GDP.
Vehicle treatment	● Activity data: Cars registered ● Data sources and treatments: See Sect. S6.1.1.
Domestic use of solvents	● Activity data: Urban and rural population ● Data sources and treatments: ➢ China: National and regional total population were obtained from China Data Online. Regional urban population data were calculated using proportion of urban population during 2005-2015 in China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016) and the proportion data in 2005 for each region were used to estimated urban population before 2004. Then rural population in each region during 1950-2015 were calculated. ➢ India: National total population were taken from UN (2018). Regional ratios and proportion of urban population during 1951-2011 were estimated using data in Indiastat. Then, urban and rural population in each region were calculated. ➢ Other countries: National urban and rural population during 1950-2015 were derived from UN (2018). For Taiwan, population data were taken from Worldometer (https://www.worldometers.info/).
Asphalt blowing	● Activity data: Asphalt produced ● Data sources and treatments: ➢ China: National total data in 2012 were taken from Wu et al. (2016) and extrapolated to 1950 using trends of Bitumen consumption in IEAWEB and GDP. Regional distribution was based on GDP. ➢ Other countries and regions: National and regional data were taken

	from default and extrapolated to 1950 using trends of Bitumen consumption in IEAWEB and GDP.
Paint production	● Activity data: Paint produced ● Data sources and treatments: ➢ China: National total data during 2011-2013 were taken from Zheng et al. (2017). ➢ Other countries and regions: National data were taken from Industrial Commodity Statistical Yearbook. ➢ All countries and regions: Extrapolation for missing data and regional distribution were based on GDP.
Ink production	● Activity data: Ink produced ● Data sources and treatments: ➢ China: National total data during 2011-2013 were taken from Zheng et al. (2017). ➢ Other countries and regions: National data were taken from Industrial Commodity Statistical Yearbook. ➢ All countries and regions: Extrapolation for missing data and regional distribution were based on GDP.
Tire production	● Activity data: Tire produced ● Data sources and treatments: ➢ China: National total data during 2011-2013 were taken from Zheng et al. (2017). ➢ India: National data in 2010 were derived from Sharma et al. (2015). ➢ Other countries: National data were taken from Industrial Commodity Statistical Yearbook. ➢ All countries and regions: Extrapolation for missing data and regional distribution were based on GDP.
Synthetic rubber production	● Activity data: Synthetic rubber produced ● Data sources and treatments: ➢ China: National total data during 2011-2013 were taken from Zheng et al. (2017). ➢ India: National data in 2010 were derived from Sharma et al. (2015). ➢ Indonesia: National data in 2010 were obtained from Permadi et al. (2017). ➢ Other countries: National data were taken from Industrial Commodity Statistical Yearbook.

	➢ All countries and regions: Extrapolation for missing data and regional distribution were based on GDP.
Textile industry	● Activity data: Textile produced ● Data sources and treatments: ➢ China: National total data during 2011-2013 were derived from Zheng et al. (2017). ➢ Other countries and regions: National and regional data were taken from default. ➢ All: Extrapolation for missing data and regional distribution for China were based on GDP.
Preservation of wood	● Activity data: Wood treated ● Data sources and treatments: ➢ All: National and regional data were taken from default and extrapolated during 1950-2015 using trends GDP.
Adhesive application	● Activity data: Adhesive consumed ● Data sources and treatments: ➢ China: National total data in 2005 and 2010 were taken from Wei et al. (2008; 2011). ➢ India: National data in 2010 were derived from Sharma et al. (2015). ➢ Indonesia: National data in 2010 were obtained from Permadi et al. (2017). ➢ Other countries: National data were taken from default. ➢ All countries and regions: Extrapolation for missing data and regional distribution were based on GDP.

S5.1.3 Printing

In REASv3, NMVOC evaporative emissions from following four printing activities are considered: packing, offset printing, publication, and screen printing. Activity data are ink consumption for each purpose. In this sub-section, data sources and treatments of activity data used in REASv3 were described.

National total ink consumption data were calculated as default for this sub-section using production, export, and import amounts taken from Industrial Commodity Statistical Yearbook and missing data were extrapolated based on GDP. For China, national total ink consumption in 2005, 2010, and 2012 were derived from Wei et al. (2008, 2011) and Wu et al. (2016) and interpolated during 2005 and 2012. Before 2005 and after 2012, the data were extrapolated based on the default data. For Indonesia, national total ink consumption data in 2010 were obtained from Permadi et al. (2017) and extrapolated during 1950-2015 based on the default data. For India, national ink consumption amounts in 2010 are available for packing, offset printing, publication, and screen printing in Sharma et al. (2015). The data were extrapolated during 1950-2015 based on the default data. For distribution of total ink consumption to each purpose except for India and regional distribution of national total data in China and India, activity data of REASv2 during 2000-2008 were used as weighting factors. Before 1999 and 2009, data in 2000 and 2008 were used respectively.

S5.1.4 Paint application

In REASv3, NMVOC evaporative emissions from paint application were considered for following purposes: architecture, domestic usage, automobile manufacture, vehicle refinishing, and other industrial applications. In this sub-section, data sources and treatments of activity data used in REASv3 were described.

National total paint consumption data during 2000-2009 were taken from a report of Information Research Limited and missing data were extrapolated during 1950-2015 based on GDP. For China, national total paint application data in 2005, 2010, and 2012 were derived from Wei et al. (2008, 2011) and Wu et al. (2016) and interpolated during 2005 and 2012. Before 2005 and after 2012, the data were extrapolated based on GDP. For India and Indonesia, national total paint consumption data in 2010 were obtained from Sharma et al. (2015) and Permadi et al. (2017), respectively and extrapolated during 1950-2015 based on GDP. The total paint consumption data were distributed to each purpose described above except for automobile manufacture using activity data of REASv2 during 2000-2008 as weighting factors. Before 1999 and after 2010, data in 2000 and 2008 were used respectively.

For automobile manufacture, activity data are production number of small and large vehicles. Production data of passenger vehicles (treated as small vehicles), bus and trucks (considered as large vehicles) in Asian countries during 2013-2015 were derived from the Japan Automobile Manufacture Association, Inc. (http://www.jama-english.jp/). Data of India and Republic of Korea were extrapolated to 1999 using data taken from Global Note (https://www.globalnote.jp/). Production number of passenger and duty vehicles were obtained from Michell (1998) and missing data were interpolated. For China, regional data during 1980-2015 were obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016) and extrapolated to 1950 using national data in China Data Online.

S5.1.5 Chemical industry

Activity data of NMVOC evaporative emissions from chemical industry were described in this sub-section. Data sources and treatments for each sub-sector category used in REASv3 were summarized in Table 5.3. (See Sect. S3.1 for energy consumption in chemical industry sub-sector and Sect. S4.1.7 for data sources of GDP used in this sub-section.)

Table 5.3. Data sources and treatments of activity data for sub-sectors of Chemical industry.

Sub-sector categories	Data sources and treatments of activity data
Ethylene production	● Activity data: Ethylene produced ● Data sources and treatments: ➢ China: Regional data during 2004-2015 were extrapolated to 1978 using national data both obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016). The data were extrapolated to 1950 based on total energy consumption in chemical industry sub-sector. ➢ India: National data in 2010 were derived from Sharma et al. (2015) and Industrial Commodity Statistical Yearbook during 1983-2003. Data between 2003 and 2010 were interpolated and missing data were extrapolated based on total energy consumption in chemical industry sub-sector. For regional distribution, the default data were used as weighting factors. ➢ Other countries and regions: National data before 1983 were taken from Industrial Commodity Statistical Yearbook and TOZAI BOEKI TSUSHINSHA (2014a). Missing data were interpolated and extrapolated based on total energy consumption in chemical industry.
Polyethylene production	● Activity data: `Polyethylene produced ● Data sources and treatments: ➢ China: National data before 1985 were taken from Industrial Commodity Statistical Yearbook and TOZAI BOEKI TSUSHINSHA (2014b). For regional distribution, data of ethylene were used as weighting factors. ➢ Other countries and regions: National data before 1983 were taken from Industrial Commodity Statistical Yearbook and TOZAI BOEKI TSUSHINSHA (2014a). For regional distribution in India, the default

	data were used as weighting factors.
Styrene production	● Activity data: Styrene produced ● Data sources and treatments: ➢ National data during 2008-2013 in China and those during 2009-2015 were obtained from TOZAI BOEKI TSUSHINSHA (2014b; a). Extrapolation during 1950-2015 and regional distribution for China and India were conducted based on data of ethylene.
Polystyrene production	● Activity data: Polyethylene produced ● Data sources and treatments: ➢ China: National data in 2010 were obtained from Wei et al. (2011). The data were extrapolated to 1950 and distributed to each region using data of ethylene. ➢ India: National data in 2010 were derived from Sharma et al. (2015). The data were extrapolated to 1950 and distributed to each region using data of ethylene. ➢ Other countries and regions: National data before 1983 were taken from Industrial Commodity Statistical Yearbook and TOZAI BOEKI TSUSHINSHA (2014a). Missing data were interpolated and extrapolated based on data of ethylene.
Polyvinylchloride production	● Activity data: Polyvinylchloride produced ● Data sources and treatments: ➢ China: National data during 2008-2013 were obtained from TOZAI BOEKI TSUSHINSHA (2014b). The data were extrapolated to 1950 and distributed to each region using data of ethylene. ➢ India: National data in 2010 were derived from Sharma et al. (2015). The data were extrapolated to 1950 and distributed to each region using data of ethylene. ➢ Other countries and regions: National data before 1983 were taken from Industrial Commodity Statistical Yearbook and TOZAI BOEKI TSUSHINSHA (2014a). Missing data were interpolated and extrapolated based on data of ethylene.
Propylene production/ Polypropylene production	● Activity data: Propylene produced/Polypropylene produced ● Data sources and treatments: ➢ China: National data during 2008-2013 were obtained from TOZAI BOEKI TSUSHINSHA (2014b) and extrapolated to 1950 using data of ethylene.

	➢ Other countries and regions: National data before 1983 were taken from Industrial Commodity Statistical Yearbook and TOZAI BOEKI TSUSHINSHA (2014a). Missing data were interpolated and extrapolated based on data of ethylene. Regional distribution for China and India were conducted also based on data of ethylene.
Storage of organic chemicals	● Activity data: Total production of organic chemicals ● Data sources and treatments: See descriptions for organic chemicals in this table.
Polyvinylchloride processing	● Activity data: Polyvinylchloride produced ● Data sources and treatments: The same as for "Polyvinylchloride production"
Polystyrene processing	● Activity data: Polyethylene produced ● Data sources and treatments: The same as for "Polystyrene production"
Carbon black	● Activity data: Carbon black produced ● Data sources and treatments: See Sect. S4.1.7.

S5.1.6 Other industry

In this sub-section, activity data of NMVOC evaporative emissions from other industrial processes were described. Data sources and treatments for each sub-sector category used in REASv3 were summarized in Table 5.4. (See Sect. S4.1.7 for data sources of GDP used in this sub-section.)

Table 5.4. Data sources and treatments of activity data for sub-sectors of other industry.

Sub-sector categories	Data sources and treatments of activity data
Bread production	● Activity data: Bread produced ● Data sources and treatments: ➢ China: National total data in 2012 were taken from Wu et al. (2016). ➢ India: National data in 2010 were derived from Sharma et al. (2015). ➢ Other countries: National data were taken from Industrial Commodity Statistical Yearbook. ➢ All countries and regions: Extrapolation for missing data were based on population (see descriptions for domestic use of solvents in Sect. S5.1.2). For regional distribution of China and India, the default data were used as weighting factors.
Beer production	● Activity data: Beer produced

	● Data sources and treatments: ➢ China: Regional data during 1983-2015 were obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016) and extrapolated to 1950 using Mitchell (1998). ➢ Other countries: National data after 2006 were taken from Brewers Association of Japan (http://www.brewers.or.jp/english/index.html) and before 1993 were obtained from Mitchell (1998). For regional distribution of India, the default data were used as weighting factors.
Coke production	● Activity data: Coke produced ● Data sources and treatments: See Sect. S4.1.8.
Asphalt production	● Activity data: Asphalt produced ● Data sources and treatments: See Sect. S5.1.2 (Asphalt blowing).
Crude steel production	● Activity data: Crude steel produced ● Data sources and treatments: See Sect. S4.1.1.
Hot rolled steel production	● Activity data: Hot rolled steel produced ● Data sources and treatments: See Sect. S4.1.1.
Pulp and paper production	● Activity data: Paper pulp produced ● Data sources and treatments: ➢ China: Regional data during 1983-2015 were obtained from China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016) and extrapolated to 1950 using China Data Online. ➢ Other countries: National data were taken from FAOSTAT (http://www.fao.org/faostat/en/). For regional distribution of India, the default data were used as weighting factors.

S5.1.7 Waste disposal

In REASv3, evaporative NMVOC emissions from disposal of municipal wastes were considered and those of industrial wastes were not included due to lack of information. Activity data are amounts of municipal wastes. Data sources and treatments of activity data used in REASv3 were summarized in Table 5.5. (See Sect. S5.1.2 (Domestic use of solvents) for data sources of population used in this sub-section.)

Table 5.5. Data sources and treatments of activity data for waste disposal.

Countries and regions	Data sources and treatments of activity data
China	Regional amounts of municipal wastes after 2003 were derived from China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016) and extrapolated to 1950 using number of population.
India	National total data in 2000, 2005, 2010, and 2015 were taken from Niyati (2015) and those in 2012 were obtained from UN Environment Programme (2017). The data were interpolated, extrapolated during 1950-2015, and distributed to each region based on number of population.
Rep. of Korea	National data during 1994-2004 were taken from Shragge and An (2014) and those in 2012 were obtained from UN Environment Programme (2017). The data were interpolated and extrapolated during 1950-2015 based on number of population.
Taiwan	National data during 2003-2015 were taken from Environmental Protection Administration (https://www.epa.gov.tw/eng/2C04F91E41A2000B/) and extrapolated during 1950-2015 using number of population
Thailand	National data during 1993-2002 were taken from Chiemchaisri et al., (2007) and extrapolated during 1950-2015 using number of population
Other countries	National data were obtained from UN Environment Programme (2017) and missing data were extrapolated during 1950-2015 based on number of population.

S5.2 Emission factors

In this section, emission factors for non-combustion sources of NMVOC for each sub-category are described. Note that emission controls were not considered for non-combustion emissions of NMVOC in REASv3.

S5.2.1 Extraction processes

Emission factors for following sub-sectors were taken from Klimont et al. (2002a) and the same settings were used for all countries and regions as well as for all target years of REASv3:

- Gas production
- Gas distribution
- Oil production

- Oil handling
- Petroleum refinery
- Service stations
- Transport and depots (gasoline/diesel)

S5.2.2 Solvent use

In this sub-section, emission factors for solvent use except for printing and paint use are described. Sources and settings of emission factors are summarized in Table 5.6.

Table 5.6. Sources and settings of emission factors for sub-sectors of solvent use.

Sub-sector categories	Sources and settings of emission factors
Dry cleaning	● Sources: Data for existing and new installations in Klimont et al. (2002a) ● Settings: The value for existing installations was commonly used for all target countries and periods except for Rep. of Korea and Taiwan where the same value was used before 2000. For Rep of Korea and Taiwan, it was assumed that all installations in 2020 are new and ratios of existing and new installations were changed linearly between 2000 and 2020. Based on the assumption emission factors during 2001 and 2015 were calculated.
Degreasing operation	● Sources and settings are the same as those “Dry cleaning”.
Vehicle treatment	● Sources: Default data and settings until 2030 in Klimont et al. (2002a) ● Settings: The Default value was used before 2000. After 2001, data in 2000 and those assumed in 2030 in Klimont et al. (2002a) were interpolated. These settings are commonly adopted for all countries.
Domestic use of solvents	● Sources: Default emission factors and settings until 2030 for rural and urban population in Klimont et al. (2002a) ● Settings: Emission factors for rural and urban population were estimated by the same methodology for “Vehicle treatment” and adopted for all countries.
Asphalt blowing	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Paint production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.

Ink production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Tire production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Synthetic rubber production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Textile industry	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Preservation of wood	● Sources and settings are the same as those “Dry cleaning”.
Adhesive application	● Sources: EEA (2016) ● Settings: The value was used for all target countries and periods.

S5.2.3 Printing

Klimont et al. (2002a) provides emission factors of packaging, offset printing, publication, and screen printing for existing and new installations. The same assumption for sub-sectors such as dry cleaning described in Sect. S5.2.2 was used in RESv3.1. as follows:

- The values for existing installations were commonly used for all target countries and periods except for Rep. of Korea and Taiwan where the same value was used before 2000.
- For Rep of Korea and Taiwan, it was assumed that all installations in 2020 are new and ratios of existing and new installations were changed linearly between 2000 and 2020. Based on the assumption emission factors during 2001 and 2015 were calculated.

S5.2.4 Paint use

In this sub-section, emission factors for paint use for architecture, domestic usage, automobile manufacture, vehicle refinishing, and other industrial applications are described. Sources and settings of emission factors are summarized in Table 5.7.

Table 5.7. Sources and settings of emission factors for sub-sectors of paint use.

Sub-sector categories	Sources and settings of emission factors
Architecture	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Domestic use	● Sources: Klimont et al. (2002a)

	● Settings: The value was used for all target countries and periods.
Vehicle refinishing	● Sources: Data for existing and new installations in Klimont et al. (2002a) ● Settings: The value for existing installations was commonly used for all target countries and periods except for Rep. of Korea and Taiwan where the same value was used before 2000. For Rep of Korea and Taiwan, it was assumed that all installations in 2020 are new and ratios of existing and new installations were changed linearly between 2000 and 2020. Based on the assumption emission factors during 2001 and 2015 were calculated.
Automobile manufacturing	● Sources: Range of emission factors depending on the proportion of vehicle types in Klimont et al. (2002a) ● Settings: The lowest and highest values of the range were used for small and large vehicles, respectively. See Sect. S5.1.4 for the definitions of vehicle sizes here.
Other industrial application	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.

S5.2.5 Chemical industry

In this sub-section, emission factors for chemical industry are described. Sources and settings of emission factors are summarized in Table 5.8.

Table 5.8. Sources and settings of emission factors for sub-sectors of chemical industry.

Sub-sector categories	Sources and settings of emission factors
Ethylene production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Polyethylene production	● Sources: Klimont et al. (2002a) ● Settings: Average of emission factors for low and high-density polyethylene production were used for all target countries and periods.
Styrene production	● Sources: EEA (2016) ● Settings: The value was used for all target countries and periods.
Polystyrene production	● Sources: EEA (2016) ● Settings: The value was used for all target countries and periods.
Polyvinylchloride production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.

Propylene production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Polypropylene production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Storage of organic chemicals	● Sources: Klimont et al. (2002a) ● Settings: Emission factors of EEA (2016) include contribution from the storage. In REASv3, 10 percent of the value was used for all target countries and periods.
Polyvinylchloride processing	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Polystyrene processing	● Sources: EEA (2016) ● Settings: The value was used for all target countries and periods.
Carbon black	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.

S5.2.6 Other industry

In this sub-section, emission factors for non-combustion emissions from other industry are described. Sources and settings of emission factors are summarized in Table 5.9.

Table 5.9. Sources and settings of emission factors for sub-sectors of other industry.

Sub-sector categories	Data sources and treatments of activity data
Bread production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Beer production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Coke production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Asphalt production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.
Crude steel production	● Sources: Klimont et al. (2002a) ● Settings: The value for steel production was used for all target countries and periods.
Hot rolled steel production	● Sources: Klimont et al. (2002a) ● Settings: The value for rolling mills was used for all target countries and

	periods.
Pulp and paper production	● Sources: Klimont et al. (2002a) ● Settings: The value was used for all target countries and periods.

S5.2.7 Waste disposal

In REASv3, the emission factor for landfills for waste disposal in Klimont et al. (2002a) were adopted for all activity data (amounts of municipal wastes) described in S5.1.7.

S5.2.8 Speciation of NMVOC emissions

Emission factors described in Sect. S5.2 were for total NMVOC. In REASv3, total NMVOC emissions were allocated to 19 NMVOC species categories defined in Sect. S2.1. The speciation was conducted based on speciation profiles for each sub-sector provided by D. G. Streets (private communication) generally based on Klimont et al. (2002a) used for REASv1 and REASv2. The speciation profiles were commonly used for all countries and periods.

S5.3 Other emission inventories included in REASv3

S5.3.1 Japan

In REASv3, evaporative emissions of individual NMVOC species from sub-sectors in Japan during 2000-2015 were obtained from the Ministry of the Environment of Japan (MOEJ, 2017a). Information for regional distribution was also available in MOEJ (2017a). Emissions of the individual species were aggregated to 19 NMVOC species categories defined in Sect S2.1. Before 1999, data in 2000 were extrapolated based on trend factors related to each sub-sector as described in Table 5.10.

Table 5.10. Sources and treatments of trend factors for sub-sectors of NMVOC evaporative emissions in Japan

Sub-sector categories	Data sources and treatments of trend factors
Natural gas production	● Trend factors: Natural gas production ● Data sources and treatments: Data during 1960-2000 were derived from IEAWEB and extrapolated to 1950 using trends taken from the Historical Statistics of Japan (Japan Statistical Association, 2006).

Coke production	● Trend factors: Coke produced ● Data sources and treatments: See Sect. S4.1.8.
Petroleum refinery	● Trend factors: Consumption of crude oil in petroleum refineries ● Data sources and treatments: See Sect. S3.1.
Service stations	● Trend factors: Consumption of gasoline in road transport sector ● Data sources and treatments: See Sect. S3.1.
Transport and depots	● Trend factors: Consumption of gasoline and diesel in road transport sector ● Data sources and treatments: See Sect. S3.1.
Dry cleaning	● Trend factors: Number of facilities ● Data sources and treatments: Data during 1963-2000 were taken from Japan Cleaning Journal (http://www.nicli.co.jp/stat-sisetu.html) and extrapolated to 1950 using values of shipments for industrial organic chemicals obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006) were used as trend factors.
Detergents usage in industry	● Trend factors: Values of shipments of detergents for industries ● Data sources and treatments: Data during 1960-2000 were obtained from Yearbook of Chemical Industry Statistics (Ministry of Economy, Trade and Industry, Japan, https://www.meti.go.jp/statistics/). Before 1960, values of shipments for industrial organic chemicals obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006) were used as trend factors.
Adhesive application	● Trend factors: Adhesive produced ● Data sources and treatments: Data during 1960-2000 were obtained from Yearbook of Chemical Industry Statistics (Ministry of Economy, Trade and Industry, Japan, https://www.meti.go.jp/statistics/). Before 1960, values of shipments of industrial organic chemicals obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006) were used as trend factors.
Asphalt blowing	● Trend factors: Asphalt produced ● Data sources and treatments: Data during 1950-2000 were derived from the Historical Statistics of Japan (Japan Statistical Association, 2006).
Rubber production	● Trend factors: Rubber produced ● Data sources and treatments: Production amounts and values of shipments for rubber products were taken from the Historical Statistics of Japan (Japan Statistical Association, 2006).

Synthetic leather production	● Trend factors: Synthetic leather produced ● Data sources and treatments: Data during 1985-2000 and those for all leather products before 1984 were obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006).
Protection of fishing net	● Trend factors: Fishing net produced ● Data sources and treatments: Data were obtained from Yearbook of Current Production Statistics (Ministry of Economy, Trade and Industry, Japan, https://www.meti.go.jp/statistics/) and the Historical Statistics of Japan (Japan Statistical Association, 2006).
Ink application	● Trend factors: Values of shipments by publishing, printing and allied industries ● Data sources and treatments: Data were obtained from Yearbook of Chemical Industry Statistics (Ministry of Economy, Trade and Industry, Japan, https://www.meti.go.jp/statistics/) and the Historical Statistics of Japan (Japan Statistical Association, 2006).
Paint application	● Trend factors: Values of shipments by paint industries of manufacturing ● Data sources and treatments: Data during 1960-2000 were obtained from Yearbook of Chemical Industry Statistics (Ministry of Economy, Trade and Industry, Japan, https://www.meti.go.jp/statistics/). Before 1960, production of synthetic paints obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006) were used.
Other solvent use	● Trend factors: Values of shipments of industrial organic chemicals ● Data sources and treatments: Data during 1950-2000 were obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006)
Chemical industry	● Trend factors: Petrochemicals produced ● Data sources and treatments: Data during 1960-2000 were obtained from Yearbook of Chemical Industry Statistics (Ministry of Economy, Trade and Industry, Japan, https://www.meti.go.jp/statistics/) were extrapolated to 1950 using values of shipments of industrial organic chemicals obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006) were used as trend factors.
Food production	● Trend factors: Values of shipments by food industries of manufacturing ● Data sources and treatments: Data during 1950-2000 were obtained from the Historical Statistics of Japan (Japan Statistical Association, 2006).
Pesticide application	● Trend factors: Pesticide produced ● Data sources and treatments: Data during 1950-2000 were taken from

	Japan Crop Production Association (https://www.jcpa.or.jp/qa/a5_12.html).
Others	● Trend factors: GDP ● Data sources and treatments: See Sect. S4.1.7

S5.3.2 Republic of Korea

For Republic of Korea, first, NMVOC (including 19 individual species) emissions from evaporative sources were tentatively estimated using activity data and emission factors described in Sects. S5.1 and S5.2, respectively. Then, emissions from extraction processes, solvent use including printing and paint application, and industrial processes in both chemical and other industries, and waste disposal were obtained from the National Institute of Environmental Research (http://airemiss.nier.go.kr/mbshome/mbs/airemiss/index.do) during 1999-2015. Finally, the tentatively estimated emissions for each sub-sector were adjusted by ratios between the aggregated emissions of the National Institute of Environmental Research and those of the tentative estimation. For example, tentative emissions from dry cleaning were adjusted by factors calculated for solvent use. Before 1999, the tentative emissions were adjusted using the factors for the year 1999. Note that emissions from combustion sources for Republic of Korea were originally estimated in REASv3.

S6. Road transport

S6.1 Activity data

S6.1.1 Annual mileage

In REASv3, exhaust emissions from road vehicles were estimated based on annual distances vehicles are driven (annual mileage) and corresponding emission factors (amounts of air pollutants per distance driven). The annual mileages were calculated by number of vehicles and annual distances traveled for each vehicle type. The number of vehicles was obtained from national and international statistics and related literatures. However, available vehicle categories in the data are different among countries and regions. In addition, information for categories of different fuel types such as gasoline and diesel and annual distances traveled for each vehicle type is limited. In Table 6.1, data sources and assumptions to estimate historical annual mileage data are provided.

Table 6.1. Data sources and settings of number of vehicles and annual distance travelled for each country and region in REASv3.

(a) China

Number of vehicles	● Data sources: ➢ Regional data of large/medium/small/mini passenger vehicles and heavy/medium/light/mini trucks during 1985-2015 were taken from China Statistical Yearbook and extrapolated to 1950 using number of civil motor vehicles in each region in China Data Online. ➢ For motorcycles, national total during 1991-2015 were taken from IRF (1990-2018) and distributed to each region and extrapolated to 1950 using the number of civil motor vehicles in each region. ● Vehicle categories: ➢ For data based on China Statistical Yearbook, large/medium and small/minicar passenger vehicles were treated as buses and cars, respectively. For trucks, heavy/medium and light/mini vehicles were treated as heavy and light trucks, respectively. For distribution of fuel types, data in He et al. (2005) were used for cars and those in Yan and Crookes (2009) were used for buses and trucks. ➢ No classification was done for motorcycles and it was assumed that only gasoline was used in motorcycles.
Annual distance travelled	● Settings of annual distance travelled for each vehicle type were based on Huo et al. (2012b).

(b) Hong Kong

Number of vehicles	● Data sources: ➢ Data of passenger cars, buses, trucks, and motorcycles during 1964-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline, diesel, and LPG passenger cars, taxis, buses, and light and heavy trucks, and motorcycles. For relative ratios of vehicles numbers of each fuel type, in addition to data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time, data in A clean air plan for Hong Kong (Environment Bureau, 2013) and consumption amounts of LPG in road transport sector in the International Energy Agency (IEA) World Energy Balances (IEAWEB) (IEA, 2017) were used.

Annual distance travelled	● Settings of Singapore were used in REASv3.

(c) Macau

Number of vehicles	● Data sources: ➢ Data of passenger cars, buses, trucks, and motorcycles during 1994-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of fuel consumption in the United Nations (UN) Energy Statistics Database (UN, 2016). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Hong Kong in REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Settings of Singapore were used in REASv3.

(d) India

Number of vehicles	● Data sources: ➢ Regional data of passenger cars, taxis, jeeps, buses, light trucks, heavy trucks, trailers, light motor vehicles, and motorcycles during 2001-2015 were taken from TERI (2013, 2018) and extrapolated to 1950 using trends of national data for cars & jeeps & taxis, buses, trucks, and motorcycles obtained from Indiastat. ● Vehicle categories: ➢ In general, passenger cars, taxis, jeeps, light motor vehicles, and motorcycles assumed to consume gasoline and for buses, trucks, and trailers, the fuel type is assumed to be diesel. For Delhi and Mumbai (in Maharashtra), number of CNG cars, taxis, and buses in 2010 were assumed based on Sahu et al. (2014) and extrapolated using IEAWEB. ➢ According to Baidya and Borken-Kleefeld (2009), there are large differences between registered number of vehicles and those actually circulating on the road. Relative ratios of vehicle numbers in operation to registered ones were taken from Prakash and Habib (2018) and Baidya and Borken-Kleefeld (2009).

Annual distance travelled	● Settings of annual distance travelled for each vehicle type were based on Prakash and Habib (2018) and Pandey and Venkataraman (2014).

(e) Japan

Annual mileages	● Data sources: ➢ National annual mileages for each vehicle type (including different fuel types) among different vehicle speed categories were derived from reports of Pollutants Release and Transfer Register (METI, 2003-2017) during 2001-2015 and extrapolated to 1950 using trends of annual distances travelled for aggregated vehicle types in the Historical Statistics of Japan (Japan Statistical Association, 2006). Vehicle types were further divided into detailed categories using number of vehicles provided in the report of the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC 2012a). ➢ For regional distribution of national data, weighting factors during 1960-2015 were calculated using annual distances travelled of aggregated vehicle types in annual reports of road transport statistics (MLIT, 1961-2016). Before 1960, data in 1960 were used. ● Vehicle categories: ➢ Vehicle types include passenger cars (gasoline and LPG), light, medium and heavy trucks (gasoline and diesel), buses (gasoline and diesel), special purpose vehicles (gasoline and diesel), and several sizes of motorcycles. Trucks, buses, and special purpose vehicles were further divided into different weight categories.

(f) Republic of Korea

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1976-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ➢ Number of LPG and CNG vehicles in 2010 were taken from a report of European Commission (Alternative fuels and infrastructure in seven non-EU markets) and the Gas Vehicles Report, respectively and extrapolated using trends of fuel

	consumption in IEAWEB. ● Vehicle categories: ➢ Vehicle types include passenger cars (gasoline, diesel, and LPG), buses (gasoline, diesel, and CNG), light and heavy trucks (gasoline and diesel), rural vehicles, and several sizes of motorcycles. For relative ratios of number of gasoline and diesel vehicles, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Settings of Singapore were used in REASv3 except for motorcycles which were taken from Jang et al. (2010).

(g) Democratic People's Republic of Korea

Number of vehicles	● Data sources and vehicle categories: ➢ Number of gasoline and diesel vehicles for passenger cars, buses, light and heavy trucks, rural vehicles, and motorcycles in 2000 were taken from REASv1 generally based on Streets et al. (2003) and extrapolated using trends of gasoline and diesel oil consumption in road transport in IEAWEB.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(h) Mongolia

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1950-2015 were obtained from National Statistics Office of Mongolia (https://www.en.nso.mn/). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(i) Taiwan

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1976-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles in National Statistics of Taiwan (https://eng.stat.gov.tw/mp.asp?mp=5). ➢ Number of LPG vehicles in 2010 were estimated based on ratios of vehicle numbers and fuel consumption in Rep. of Korea and extrapolated using trends of fuel consumption in IEAWEB. ● Vehicle categories: ➢ Vehicle types include passenger cars (gasoline, diesel, and LPG), buses (gasoline and diesel), light and heavy trucks (gasoline and diesel), and motorcycles. For relative ratios of number of gasoline and diesel vehicles, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Settings of Singapore were used in REASv3.

(j) Brunei

Number of vehicles	● Data sources and vehicle categories: ➢ National data of passenger cars, buses, trucks, and motorcycles during 2010-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of fuel consumption in IEAWEB and those of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(k) Cambodia

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1990-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of fuel consumption in IEAWEB and those of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(l) Indonesia

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1950-2015 were obtained from Statistics Indonesia (https://www.bps.go.id/linkTableDinamis/view/id/1133/). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, rural vehicles, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Indonesia provided in Clean Air Asia (2012) were used.

(m) Laos

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1987-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of fuel consumption in IEAWEB and those of number of vehicles for aggregated vehicle types in

	Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Laos provided in Clean Air Asia (2012) were used.

(n) Malaysia

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1963-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, rural vehicles, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Malaysia provided in Clean Air Asia (2012) were used.

(o) Myanmar

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1993-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of fuel consumption in IEAWEB and those of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003)

	and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(p) Philippines

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1981-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, rural vehicles, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Philippines provided in Clean Air Asia (2012) were used.

(q) Singapore

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1981-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Singapore provided in Clean Air Asia (2012) were used.

(r) Thailand

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1967-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline, diesel, LPG, and CNG passenger cars, buses, and light and heavy trucks, rural vehicles, and motorcycles. For relative ratios of vehicles numbers of each fuel type, in addition to data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time, data in Chollacoop et al. (2011) and consumption amounts of LPG and CNG in road transport sector in IEAWEB were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Thailand provided in Clean Air Asia (2012) were used.

(s) Vietnam

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 2007 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of fuel consumption in IEAWEB and those of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, small and large buses, light and heavy trucks, rural vehicles, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Vietnam provided in Clean Air Asia (2012) as well as Manh et al. (2011) were used.

(t) Afghanistan

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1975-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of India in REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(u) Bangladesh

Number of vehicles	● Data sources: ➢ National data of passenger cars, taxis, jeeps, buses, trucks, rural vehicles and motorcycles during 2000-2015 were obtained from Statistical Yearbook of Bangladesh (2013-2016) and extrapolated to 1950 using trends number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline, diesel, and CNG passenger cars, taxis, jeeps, small and large buses, light and heavy trucks, rural vehicles and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, Wadud and Khan (2011) as well as data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Bangladesh provided in Clean Air Asia (2012) were used.

(v) Bhutan

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1994-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles taken from

	Statistical Yearbook of Bhutan (http://www.nsb.gov.bt/publication/publications.php?id=3) ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

(w) Nepal

Number of vehicles	● Data sources and vehicle categories: ➢ National data of passenger cars/jeeps, 3 wheeler vehicle, taxis, micro, mini and medium buses, mini and medium trucks, pickup and motorcycles in 2013 and trends during 1990-2012 for aggregated vehicle types were derived from Malla (2014). ➢ Malla (2014) provided fuel types and ratios of operational to registered vehicle for each vehicle type. ➢ Before 1990, data during 1950-2015 were estimated based on trends of fuel consumption data in IEAWEB.
Annual distance travelled	● Settings of annual distance travelled for each vehicle type were based on Malla. (2014).

(x) Pakistan

Number of vehicles	● Data sources: ➢ National data of motor cars/jeeps, taxis, buses, trucks, motorcycles, 3 wheeler vehicles during 2001-2012 were taken from Pakistan Statistical Yearbook (http://www.pbs.gov.pk/publications/) and were extrapolated to 1963 and 2015 using trends of number of vehicles in IRF (1976-2018). ● Vehicle categories: ➢ Vehicle types include gasoline, diesel, and CNG passenger cars, taxis, mall and large buses, light and heavy trucks, rural vehicles and motorcycles. For relative ratios of gasoline, diesel, and CNG vehicle numbers, Khan and Yasmin (2014) as well as data of Streets

	et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Pakistan provided in Clean Air Asia (2012) were used.

(y) Sri Lanka

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1963-2015 were obtained from IRF (1976-2018) and extrapolated to 1950 using trends of number of vehicles for aggregated vehicle types in Mitchell (1998). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of Streets et al. (2003) and REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type in Sri Lanka provided in Clean Air Asia (2012) were used.

(z) Maldives

Number of vehicles	● Data sources: ➢ National data of passenger cars, buses, trucks, and motorcycles during 1991-2015 were obtained from IRF (1976-2018). ● Vehicle categories: ➢ Vehicle types include gasoline and diesel passenger cars, buses, light and heavy trucks, and motorcycles. For relative ratios of gasoline and diesel vehicle numbers, data of India in REASv2 generally based on GAINS ASIA at that time were used.
Annual distance travelled	● Annual vehicle kilometer travelled per vehicle type averaged in Asia provided in Clean Air Asia (2012) were used.

S6.1.2 Fuel consumption

In REASv3, emissions of SO_2 and CO_2 were calculated using fuel consumption amounts. In order to estimate emissions from each vehicle type, total fuel consumption in road transport sector (see Sect. S3.1.2) needs to be distributed to each type of vehicles. The distributions were performed in each country and region based on weighting factors which were products of annual mileages (see S6.1.1) and fuel efficiencies of each vehicle type. In this sub-section, the fuel efficiencies used in REASv3 are described.

The fuel efficiencies were taken from Clean Air Asia (2012) for following countries: Indonesia, Laos, Malaysia, Philippines, Singapore, Thailand, Vietnam, Bangladesh, Nepal, Pakistan, and Sri Lanka. For Republic of Korea, Taiwan, Hong Kong, and Macau, data of Singapore were used the same as for the annual distance travelled. For North Korea, Mongolia, Brunei, Cambodia, and Myanmar, averaged data of Southeast Asian countries in Clean Air Asia (2012) were used. Similarly, averaged values of South Asian countries in Clean Air Asia (2012) were used for Afghanistan, Bhutan, and Maldives. For China, the fuel efficiencies were derived from Yan and Crookes (2009). For Japan, fuel consumption in vehicle type are available in each region after 2009. Before 2008, the data in 2009 were extrapolated using trend of annual mileages for each vehicle type in each region and used as weighting factors to distribute regional fuel consumption in road transport to each vehicle type.

S6.2 Emission factors of exhaust emissions

S6.2.1 NO_x, CO, NMVOC, and PM species

In REASv3, emission factors of NO_x, CO, NMVOC, and PM species for exhaust emissions from road vehicles were estimated as follows:

1. Emission factors of each vehicle type in a base year (different from country to country) were estimated.
2. Trends of the emission factors for each vehicle type were estimated considering the timing of road vehicle regulations in each country and the regions and the ratios of vehicles production years.
3. Emission factors of each vehicle type during 1950-2015 were calculated using those of base years and the corresponding trends.

The information of road vehicle regulations in each country and regions were taken from Clean Air Asia (2014). For the ratios of vehicle production years, due to lack of information, data for Macau derived from Zhang et al. (2016) were used for Hong Kong, Republic of Korea, and Taiwan and those from Japan Environmental Sanitation Center and Suuri Keikaku (2011) for Vietnam were used for other countries and regions. Then, trends of emission factors were estimated using the above data and information with values of Europe and United States standards. Finally, emission factors used to estimate emissions were calculated for each vehicle type.

In this sub-section, ranges of emission factors during 1950-2015 used in REASv3 were presented in Tables 6.2-6.5 for following major vehicles types: CARG, CARD, LDTG, LDTD, HDTG, HDTD, BUSG, BUSD, and MC (CAR: Passenger cars, LDT: Light duty trucks, HDT: Heavy duty trucks, BUS: Buses, MC: Motorcycles, G: Gasoline vehicles, and D: Diesel vehicles). For PM species, referring Klimont et al. (2002b) and Bond et al. (2004), ratios of $PM_{2.5}$, BC, and OC to PM_{10} were assumed as follows:

- $PM_{2.5}/PM_{10}$: 0.95 for gasoline and light diesel vehicles, 1.0 for heavy diesel vehicles, and 0.9 for LPG and CNG vehicles.
- BC/PM_{10}: 0.34 for gasoline vehicles and 0.66 for diesel vehicles.
- OC/PM_{10}: 0.36 for gasoline vehicles and 0.21 for diesel vehicles.
- BC and OC emissions from LPG and CNV vehicles were neglected.

Note that emissions from road vehicles in Japan were estimated by different methodology as described in Sect. S.6.2.4.

Table 6.2. Emission factors of NO_x, CO, NMVOC (NMV), and PM_{10} for exhaust emissions from road vehicle in China. Unit is g/km (expressed as NO_2 for NO_x).

g/km	CARG[c]	LDTG	LDTD	HDTG	HDTD
NO_x	0.25-2.70 (0.53)[a]	0.23-3.00 (0.53)[a]	2.22-5.00 (2.85)[a]	0.78-2.18 (1.91)[a]	5.41-9.03 (7.65)[a]
CO	2.72-29.7 (5.93)[a]	3.17-40.0 (8.01)[a]	1.20-9.46 (1.89)[a]	5.26-81.6 (16.3)[a]	1.95-27.2 (5.44)[a]
NMV	0.33-1.89 (0.66)[a]	0.41-3.53 (0.88)[a]	0.37-2.50 (0.75)[a]	0.24-4.00 (1.47)[a]	0.32-1.47 (0.98)[a]
PM_{10}	0.013-0.019 (0.016)[a]	0.012-0.021 (0.016)[a]	0.075-0.37 (0.15)[a]	0.042-0.17 (0.081)[a]	0.13-0.63 (0.29)[a]
g/km	BUSG	BUSD	MC	BUS(LPG)[b]	BUS(CNG)[b]
NO_x	0.92-2.14 (1.91)[a]	5.75-8.79 (7.65)[a]	0.17-0.29 (0.22)[a]	2.60	5.70
CO	6.34-81.6 (16.3)[a]	2.43-27.2 (5.44)[a]	8.64-25.2 (12.9)[a]	1.00	12.0
NMV	0.40-4.00 (1.47)[a]	0.37-1.37 (0.98)[a]	2.41-5.45 (3.59)[a]	0.70	1.40

PM_{10}	0.050-0.15 (0.081)[a]	0.16-0.55 (0.29)[a]	0.060-0.16 (0.10)[a]	0.033	0.033

a. Emission factors in 2010 used as based data estimated referring Wu et al. (2011), Huo et al. (2012b; 2012c), Zhao et al. (2012), Zhang et al. (2013), and Xia et al. (2016). b. ABC Emission Inventory Manual (Shrestha et al., 2013). c. CARD was not categorized.

Table 6.3. Emission factors of NO_x, CO, NMVOC (NMV), and PM_{10} for exhaust emissions from road vehicle in India. Unit is g/km (expressed as NO_2 for NO_x).

g/km	CARG[c]	LDTG	LDTD	HDTD[c]	BUSD[c]
NO_x	0.98-2.70 (1.79)[a]	1.28-2.70 (2.24)[a]	5.22-9.00 (6.77)[a]	7.81-12.80 (11.3)[a]	5.70-9.08 (8.16)[a]
CO	1.62-9.01 (3.50)[a]	2.27-10.3 (4.00)[a]	2.80-8.12 (4.00)[a]	4.40-14.8 (11.9)[a]	5.24-14.3 (11.9)[a]
NMV	0.41-2.06 (0.80)[a]	0.58-2.91 (1.13)[a]	0.53-1.19 (1.13)[a]	0.47-1.96 (1.38)[a]	0.51-2.20 (1.09)[a]
PM_{10}	0.13-0.19 (0.18)[a]	0.43-0.68 (0.65)[a]	0.32-1.63 (0.65)[a]	0.55-2.79 (1.41)[a]	0.33-1.26 (072)[a]
g/km	MC	CAR_CNG[b]	BUS_CNG[b]		
NO_x	0.20-0.30 (0.24)[a]	2.10	5.70		
CO	1.98-15.7 (8.04)[a]	4.00	12.0		
NMV	1.63-4.60 (2.46)[a]	0.50	1.40		
PM_{10}	0.025-0.049 (0.030)[a]	0.067	0.067		

a. Emission factors in 2010 used as based data estimated referring Mishra et al. (2014), Sahu et al. (2014), and Pandey and Venkataraman. (2014). b. ABC Emission Inventory Manual (Shrestha et al., 2013). c. CARD, HDTG, and BUSG were not categorized.

Other East Asian countries

Emission factors of Republic of Korea and Taiwan were estimated with high uncertainties based on values of Europe and United States standards, respectively. For Democratic People's Republic of Korea and Mongolia, emission factors used in REASv1 and REASv2 were adopted. Ranges of emission factors are presented in Table 6.4.

Table 6.4. Emission factors of NO_x, CO, NMVOC (NMV), and PM_{10} for exhaust emissions from road vehicle in other East Asian countries. Unit is g/km (expressed as NO_2 for NO_x).

(a) Republic of Korea

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.10-2.70	0.34-0.67	0.10-2.14	0.50-0.90	3.01-5.37
CO	0.41-8.60	0.10-0.57	1.60-14.1	0.17-0.91	8.52-35.2
NMV	0.084-0.92	0.026-0.25	0.12-2.07	0.063-0.15	0.55-3.09
PM_{10}	0.0018-0.0030	0.018-0.20	0.0017-0.0030	0.014-0.28	0.0017-0.014
g/km	HDTD	BUSG	BUSD	MC	
NO_x	3.04-12.0	5.17-8.42	5.59-9.09	0.05-0.43	
CO	0.23-0.94	0.51-1.63	0.25-0.81	4.43-20.1	
NMV	0.066-0.37	0.21-2.8	0.11-0.41	0.64-6.76	
PM_{10}	0.021-0.62	0.012-0.060	0.11-1.01	0.010-0.14	
g/km	CAR/LPG	BUS/CNG			
NO_x	0.056	2.50			
CO	0.62	1.00			
NMV	0.10	0.052			
PM_{10}	0.0012	0.0012			

(b) Taiwan

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.30-2.70	0.55-1.11	0.28-3.10	1.02-1.66	3.62-6.81
CO	1.38-8.60	0.14-0.50	3.64-23.4	2.21-6.26	8.75-45.0
NMV	0.21-2.10	0.045-0.29	0.19-2.84	0.094-0.15	0.92-4.00
PM_{10}	0.0015-0.0020	0.053-0.27	0.0021-0.0030	0.029-0.28	0.0080-0.068
g/km	HDTD	BUSG	BUSD	MC	CAR/LPG
NO_x	3.99-7.50	5.72-9.66	8.74-14.8	0.19-0.39	0.056
CO	0.36-2.19	3.19-13.0	1.19-4.83	2.70-16.4	0.62
NMV	0.12-1.04	0.55-2.50	0.42-1.90	1.18-3.09	0.1
PM_{10}	0.11-0.93	0.023-0.12	0.20-1.05	0.086-0.32	0.0012

(c) Democratic People's Republic of Korea and Mongolia

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	1.79	2.39	3.51	2.58	9.56
CO	69.3	12.1	69.3	12.1	135.0
NMV	3.82	0.16	3.44	0.13	5.25
PM_{10}	0.033	0.34	0.033	0.34	0.066
g/km	HDTD	BUSG	BUSD	MC	
NO_x	24.1	9.56	24.1	0.12	
CO	17.7	135.0	17.7	21.1	
NMV	0.72	1.99	1.99	6.05	
PM_{10}	0.47	0.066	0.47	0.033	

Southeast Asian countries

For Southeast Asian countries, default emission factors were assumed based on Boken et al. (2007) and used as uncontrolled values. Then, emission factors during 1950-2015 were estimated considering effects of regulations. Ranges of emission factors of Southeast Asian countries are presented in the following tables.

Table 6.5. Emission factors of NO_x, CO, NMVOC (NMV), and PM_{10} for exhaust emissions from road vehicle in Southeast Asian countries. Unit is g/km (expressed as NO_2 for NO_x).

(a) Brunei, Cambodia, Laos, and Myanmar

g/km[a]	CARG	CARD	LDTG	LDTD	HDTG
NO_x	2.50	2.77	3.20	3.15	4.00
CO	15.4	1.07	28.0	2.00	45.0
NMV	1.70	0.99	2.40	1.28	4.00
PM_{10}	0.0030	0.23	0.0060	0.63	0.025
g/km[a]	HDTD	BUSG	BUSD	MC	
NO_x	11.7	4.00	14.8	0.15	
CO	3.30	45.0	6.00	15.9	
NMV	2.00	4.00	3.70	4.30	
PM_{10}	0.62	0.025	2.08	0.10	

a. Due to lack of information for regulations, default emission factors were used without changes during 1950-2015 for Brunei, Cambodia, Laos, and Myanmar.

(b) Indonesia

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.79-2.50	1.87-2.77	0.62-3.20	2.42-3.15	2.84-4.00
CO	5.53-15.4	0.61-1.07	9.25-28.0	1.01-2.00	22.6-45.0
NMV	0.61-1.70	0.41-0.99	0.49-2.40	1.28	1.64-4.00
PM_{10}	0.0030	0.099-0.23	0.0060	0.26-0.63	0.0080-0.025
g/km	HDTD	BUSG	BUSD	MC	
NO_x	8.30-11.7	3.05-4.00	11.3-14.8	0.11-0.15	
CO	1.66-3.30	26.8-45.0	3.57-6.00	7.49-15.9	
NMV	0.82-2.00	2.03-4.00	1.88-3.70	2.87-4.30	
PM_{10}	0.20-0.62	0.010-0.025	0.87-2.08	0.045-0.10	

(c) Malaysia

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.17-2.50	2.23-2.77	0.17-3.20	2.32-3.15	3.74-4.00
CO	2.23-15.4	0.56-1.07	5.91-28.0	0.86-2.00	36.4-45.0
NMV	0.19-1.70	0.26-0.99	0.16-2.40	1.28	3.18-4.00
PM_{10}	0.0023-0.0030	0.074-0.23	0.0041-0.0060	0.21-0.63	0.015-0.025
g/km	HDTD	BUSG	BUSD	MC	
NO_x	11.0-11.7	3.8-4.00	14.1-14.8	0.08-0.15	
CO	2.67-3.30	37.5-45.0	5.00-6.00	3.28-15.9	
NMV	1.59-2.00	3.33-4.00	3.08-3.70	1.92-4.30	
PM_{10}	0.37-0.62	0.016-0.025	1.37-2.08	0.025-0.10	

(d) Philippines

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.73-2.50	1.95-2.77	0.56-3.20	2.40-3.15	3.52-4.00
CO	5.24-15.4	0.60-1.07	8.85-28.0	0.97-2.00	36.9-45.0
NMV	0.58-1.70	0.38-0.99	0.45-2.40	1.28	2.73-4.00
PM_{10}	0.0030	0.096-0.23	0.0060	0.25-0.63	0.013-0.025
g/km	HDTD	BUSG	BUSD	MC	
NO_x	10.3-11.7	3.58-4.00	13.3-14.8	0.12-0.15	
CO	2.71-3.30	38.0-45.0	5.07-6.00	7.44-15.9	
NMV	1.37-2.00	2.90-4.00	2.68-3.70	2.86-4.30	
PM_{10}	0.31-0.62	0.014-0.025	1.19-2.08	0.050-0.10	

(e) Singapore

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.24-2.50	1.39-2.77	0.22-3.20	1.69-3.15	1.92-4.00
CO	2.40-15.4	0.25-1.07	5.51-28.0	0.48-2.00	7.86-45.0
NMV	0.27-1.70	0.13-0.99	0.19-2.40	0.45-1.28	0.54-4.00
PM_{10}	0.0027-0.0030	0.039-0.23	0.0051-0.0060	0.073-0.63	0.0036-0.025
g/km	HDTD	BUSG	BUSD	MC	
NO_x	5.61-11.7	2.19-4.00	8.11-14.8	0.10-0.15	
CO	0.58-3.30	12.1-45.0	1.62-6.00	4.71-15.9	
NMV	0.27-2.00	0.92-4.00	0.85-3.70	2.30-4.30	
PM_{10}	0.088-0.62	0.0056-0.025	0.47-2.08	0.039-0.10	

(f) Thailand

g/km	CARG	CARD	LDTG	LDTD	HDTD[a]
NO_x	0.15-2.50	1.52-2.77	0.14-3.20	1.80-3.15	9.36-11.7
CO	2.01-15.4	0.25-1.07	4.16-28.0	0.65-2.00	2.59-3.30
NMV	0.18-1.70	0.14-0.99	0.14-2.40	0.60-1.28	1.21-2.00
PM_{10}	0.0018-0.0030	0.047-0.23	0.0032-0.0060	0.11-0.63	0.23-0.62
g/km	BUSG	BUSD	MC	CAR/LPG[b]	CAR/CNG[b]
NO_x	3.28-4.00	12.1-14.8	0.080-0.15	2.10	2.10
CO	36.0-45.0	4.80-6.00	3.25-15.9	6.05	4.00
NMV	2.56-4.00	2.37-3.70	1.75-4.30	1.84	0.50
PM_{10}	0.011-0.025	0.87-2.08	0.039-0.10	0.067	0.067
g/km	BUS/LPG[b]	BUS/CNG[b]	LDT/CNG[b]	HDT/CNG[b]	
NO_x	5.70	5.70	2.10	5.70	
CO	24.0	12.0	8.00	12.0	
NMV	8.00	1.40	3.50	1.40	
PM_{10}	0.067	0.067	0.067	0.067	

a. HDTG was not categorized. b. ABC Emission Inventory Manual (Shrestha et al., 2013).

(g) Vietnam

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.55-2.50	1.95-2.77	0.43-3.20	2.36-3.15	3.48-4.00
CO	4.31-15.4	0.57-1.07	7.94-28.0	0.92-2.00	35.4-45.0
NMV	0.48-1.70	0.32-0.99	0.35-2.40	1.28	2.60-4.00
PM_{10}	0.0030	0.083-0.23	0.0060	0.23-0.63	0.011-0.025
g/km	HDTD	BUSG	BUSD	MC	
NO_x	10.2-11.7	3.55-4.00	13.1-14.8	0.12-0.15	
CO	2.59-3.30	36.2-45.0	4.82-6.00	6.73-15.9	
NMV	1.30-2.00	2.76-4.00	2.56-3.70	2.74-4.30	
PM_{10}	0.27-0.62	0.012-0.025	1.03-2.08	0.050-0.10	

Other South Asian countries

For Southeast Asian countries except for India, default emission factors were assumed based on Boken et al. (2007) and used as uncontrolled values. Then, emission factors during 1950-2015 were estimated considering effects of regulations. Ranges of emission factors of Southeast Asian countries are presented in Table 6.6.

Table 6.6. Emission factors of NO_x, CO, NMVOC (NMV), and PM_{10} for exhaust emissions from road vehicle in other South Asian countries. Unit is g/km (expressed as NO_2 for NO_x).

(a) Afghanistan, Bhutan, and Maldives

g/km[a]	CARG	CARD	LDTG	LDTD	HDTG
NO_x	2.20	1.45	3.20	4.80	4.00
CO	12.2	1.45	28.0	1.50	45.0
NMV	2.10	1.18	2.40	1.41	4.00
PM_{10}	0.0030	0.26	0.0060	0.34	0.025
g/km[a]	HDTD	BUSG	BUSD	MC	
NO_x	13.6	4.00	15.3	0.20	
CO	3.60	45.0	6.10	15.7	
NMV	2.20	4.00	3.70	4.60	
PM_{10}	0.68	0.025	2.09	0.10	

a. Due to lack of information for regulations, default emission factors were used without changes during 1950-2015 for Afghanistan, Bhutan, and Maldives.

(b) Bangladesh

g/km	CARG[a]	LDTG	LDTD	HDTD[a]	BUSD[a]
NO_x	0.21-2.20	0.21-2.20	3.53-4.80	13.0-13.6	14.8-15.3
CO	1.86-12.2	1.86-12.2	0.65-1.50	2.86-3.60	4.86-6.10
NMV	0.31-2.10	0.31-2.10	1.41	1.83-2.20	3.16-3.70
PM_{10}	0.0030	0.003	0.11-0.34	0.42-0.68	1.33-2.09
g/km	MC	CAR/CNG[a]	LDT/CNG[a]	BUS/CNG[a]	
NO_x	0.13-0.20	2.10	2.10	5.70	
CO	4.38-15.7	4.00	4.00	12.0	
NMV	2.51-4.60	0.50	0.50	1.40	
PM_{10}	0.025-0.10	0.067	0.067	0.067	

a. CARD, HDTG, and BUSG were not categorized.

(c) Nepal

g/km	CARG[a]	LDTG	LDTD	HDTD[a]	BUSG
NO_x	0.56-2.20	0.56-2.20	3.33-4.80	12.3-13.6	0.99-4.00
CO	4.18-12.2	4.18-12.2	0.64-1.50	2.89-3.60	17.2-45.0
NMV	0.69-2.10	0.69-2.10	1.14-1.41	1.72-2.20	1.01-4.00
PM_{10}	0.0024-0.0030	0.0024-0.0030	0.11-0.34	0.38-0.68	0.019-0.025
	BUSD	MC	BUS/LPG[b]		
NO_x	14.2-15.3	0.16-0.20	0.20		
CO	5.00-6.10	7.21-15.7	3.90		
NMV	3.04-3.70	2.83-4.60	0.77		
PM_{10}	1.30-2.09	0.072-0.10	0.00		

a. CARD and HDTG were not categorized. b. Malla (2014).

(d) Pakistan

g/km	CARG[a]	LDTG[a]	HDTD[a]	BUSG	BUSD
NO_x	1.47-2.20	1.65-3.20	9.75-13.6	2.42-4.00	11.7-15.3
CO	8.38-12.2	16.8-28.0	1.83-3.60	30.2-45.0	3.66-6.10
NMV	1.44-2.10	1.26-2.40	1.02-2.20	2.44-4.00	2.05-3.70
PM_{10}	0.0030	0.0060	0.25-0.68	0.025	0.96-2.09

g/km	MC	CAR/CNG[b]
NO_x	0.18-0.20	2.10
CO	11.5-15.7	4.00
NMV	3.83-4.60	0.50
PM_{10}	0.072-0.10	0.067

a. CARD, LDTD, and HDTG were not categorized. b. ABC Emission Inventory Manual (Shrestha et al., 2013).

(e) Sri Lanka

g/km	CARG	CARD	LDTG	LDTD	HDTG
NO_x	0.65-2.20	1.05-1.45	0.57-3.20	3.65-4.80	4.00
CO	4.23-12.2	0.83-1.45	8.97-28.0	0.73-1.50	45.0
NMV	0.73-2.10	0.46-1.18	0.45-2.40	1.41	4.00
PM_{10}	0.0030	0.11-0.26	0.0060	0.13-0.34	0.025

g/km	HDTD	BUSG	BUSD	MC
NO_x	13.6	4.00	15.3	0.16-0.20
CO	3.60	45.0	6.10	7.52-15.7
NMV	2.20	4.00	3.70	3.09-4.60
PM_{10}	0.68	0.025	2.09	0.053-0.10

Cold start emissions

In REASv3, cold start emissions were roughly estimated for NO_x, CO, NMVOC, and PM species using the following equation:

$$E_{COLD} = \sum_i \{NV_i \times ADT_i \times EF_{HOTi} \times \beta_i(T) \times F_i(T)\}$$

where, E_{COLD} is the cold start emission, i is the vehicle type, NV is the number of vehicles in operation, ADT is the annual distance traveled, EF_{HOT} is the emission factor for the hot emission, β is the fraction of distance traveled driven with a cold engine or with the catalyst operating below the light-off temperature, and F is the correction factor of EF_{HOT} for cold start emission. β and F are functions of temperature T and assumed based on EEA (2016) as follows:

- $\beta = 0.33182 - 0.004966 \times T$
- F for gasoline vehicles
  - $1.14 - 0.006 \times T$ for NO_x
  - $3.7 - 0.09 \times T$ for CO
  - $2.8 - 0.06 \times T$ for NMVOC
- F for diesel vehicles
  - $1.3 - 0.013 \times T$ for NO_x
  - $1.9 - 0.03 \times T$ for CO
  - $3.1 - 0.09 \times T$ for NMVOC
  - $3.1 - 0.1 \times T$ for PM species
- F for LPG vehicles
  - $0.98 - 0.006 \times T$ for NO_x
  - $3.66 - 0.09 \times T$ for CO
  - $2.24 - 0.06 \times T$ for NMVOC

For T, monthly averaged temperature at surface were calculated using NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html). Therefore, cold start emissions were estimated in each month assuming daily traffic volumes were unchanged during the target year. In addition, effects of regulations on cold start emission were not considered in REASv3.

S6.2.2 NH_3

Exhaust emissions of NH_3 only from gasoline vehicles were roughly estimated in REASv3. Emission factors were obtained from Kannari et al. (2001) as follows:

- 0.0221 g/km for passenger cars
- 0.0211 g/km for buses
- 0.0108 g/km for light trucks
- 0.0146 g/km for heavy trucks
- 0.0068 g/km for motorcycles

S6.2.3 SO_2 and CO_2

For SO_2 and CO_2, emissions were estimated based on fuel consumption in REASv3 except for Japan (see Sect. S6.2.4). SO_2 emissions were calculated using sulfur contents in fuels in gasoline and diesel consumed in road transport sector, assuming sulfur retention in ash is zero. Default settings of sulfur contents were taken from REASv1 and REASv2 described in Sect S3.2.1 and update with information obtained from Clean Air Asia (2011), Wang and Hao (2012), etc. The data for gasoline and diesel oil used in REASv3 are summarized in Table 6.7.

Table 6.7. Sulfur contents in gasoline and diesel oil for road vehicles used in REASv3.

Countries	Settings and data sources
China	● Gasoline referring Wang and Hao (2012): ➢ Beijing: 0.15/0.1/0.08/0.05/0.015/0.005 in 1950-1999/2000/2001-2003/2004/2005-2007/2008-2015 ➢ Shanghai 0.15/0.1/0.08/0.05/0.005 in 1950-1999/2000/2001-2004/2005-2008/2009-2015 ➢ Guangdong 0.15/0.08/0.05/0.015 in 1950-1999/2000-2003/2004/2005-2015 ➢ Others: 0.15/0.1/0.08/0.05 in 1950-1999/2000-2002/2003-2005/2006-2015 ● Diesel referring Clean Air Asia (2011)[b]: ➢ Beijing, Shanghai, and Guangdong 0.5/0.2/0.05/0.35/0.05 in 1950-2001/2002-2003/2004/2005-2007/2008-2015 ➢ Hong Kong 0.5/0.5-0.05/0.05/0.005/0.001 in 1950-1989/1990-1996/1997-2001/2002-2006/2007-2015 ➢ Others 0.5/0.2/0.125/0.35/ in 1950-2001/2002-2004/2005-2009/2010-2015
India	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ Delhi 1.0/0.5/0.25/0.05/0.035/0.05 in 1950-1995/1996-1999/2000/2001-2004/2005-2009/2010-2015 ➢ Others 1.0/0.5/0.25/0.05/0.035 in 1950-1995/1996-1999/2000/2001-2009/2010-2015

Republic of Korea	● Gasoline: REASv1 and REASv2[a] ● Diesel referring Clean Air Asia (2011)[b]: ➢ 0.4/0.25/0.05/0.043/0.01/0.003/0.0015 in 1950-1989/1990-1994/1995-2002/2003/2004-2005/2006/2007-2015
Taiwan	● Gasoline: REASv1 and REASv2[a] ● Diesel referring Clean Air Asia (2011)[b]: ➢ 0.8/0.8-0.3/0.3/0.05/0.035/0.01/0.005 in 1950-1988/1989-1996/1997-1998/1999-2001/2002-2003/2004-2007/2008-2015
Cambodia	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 0.8/0.8-0.2/0.2/0.15 in 1950-1989/1990-1996/1997-2003/2004-2015
Indonesia	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 1.0/1.0-0.5/0.5/0.35/0.035 in 1950-1989/1990-1996/1997-2004/2005-2015
Malaysia	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 0.5/0.3/0.05 in 1950-1997/1998-2001/2002-2015
Philippines	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 0.5/0.2/0.05 in 1950-2000/2001-2003/2004-2015
Singapore	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 0.5/0.5-0.3/0.3/0.05/0.005 in 1950-1989/1990-1996/1997/1998-2005/2008-2015
Thailand	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 1.0/1.0-0.05/0.05/0.035/0.005 in 1950-1989/1990-1999/1999-2003/2004-2011/2012-2015
Vietnam	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 1.0/0.05 in 1950-2006/2007-2015
Bangladesh	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 1.0/1.0-0.5/0.5 in 1950-1989/1990-1996/1997-2015

Pakistan	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 1.2/1.2-1.0/1.0/0.7 in 1950-1989/1990-1996/1997-2001/2002-2015
Sri Lanka	● Gasoline: REASv1 and REASv2[a] ● Diesel: referring Clean Air Asia (2011)[b]: ➢ 1.0/0.5/0.175/0.05/0.005 in 1950-2002/2003/2004-2006/2007-2011/2012-2015
Others	● Gasoline and: diesel: REASv1 and REASv2[a]

a. Settings of "REASv1 and REASv2" are as follows:

- Data of REASv1 and REASv2 were used in 1980-1999 and 2000-2008, respectively.
- Data in 1980 and 2008 were used before 1979 and after 2009, respectively.

b. Settings before 1995 were taken from REASv1 and after 1996 were based on Clean Air Asia (2011).

For CO_2, emissions were simply calculated by consumption amounts of fuels (gasoline, diesel, liquefied petroleum gas, and natural gas) and the corresponding emission factors taken from IPCC (2006).

S6.2.4 Japan

Emissions of NO_x, CO, NMVOC, NH_3, CO_2, and PM species in Japan were estimated using following data and information:

- Emission factors for different speed ranges and production years
- Regulations for vehicle emissions and their phase-in periods
- Ratios of number of vehicles of different ages
- Traffic volumes by the speed ranges

Emission factors and information of regulations were obtained from JPEC (2012a). Data of vehicle ages were taken from NILIM (2012). See Sect. 6.1.1 for other data. For SO_2, emission factors after 2005 were estimated by the same methodologies for the other species and those before 2004 were adjusted based on regulation of sulfur contents in gasoline and diesel oil.

Ranges of net emission factors during 1950-2015 used in REASv3 were presented in Table 6.8 for following vehicle categories: CARG, CARD, LDTG, LDTD, MDTG, MDTD, HDTG, HDTD, BUSG, LBUSD, MBUSD, HBUSD, LSPCG, HSPCG, LSPCD, HSPCD, SMC, and MC (CAR: Passenger cars, LDT: Light duty trucks, MDT: Middle duty trucks, HDT: Heavy duty trucks, LBUS: Light buses, MBUS: Middle buses, HBUS: Heavy Buses, LSPC: Light special purpose vehicles, HSPC: Heavy special purpose vehicles, SMC: Small motorcycles, MC: Motorcycles, G: Gasoline vehicles, and D: Diesel vehicles). Note that each vehicle category includes several seizes of vehicles especially trucks and buses. Therefore, ranges of net emission factors in Table 6.8 were caused not only by regulations, but also differences of vehicle types in each category.

Table 6.8. Ranges of net emission factors for Japan used in REASv3. Unit is g/km (expressed as NO_2 for NO_x).

g/km	CARG	CARD	LDTG	LDTD	MDTG
SO_2	0.00085-0.012	0.0015-1.48	0.00097-0.032	0.0011-3.20	0.0091-0.014
NO_x	0.062-3.49	0.18-3.77	0.21-19.3	0.24-9.04	0.052-6.12
CO	1.13-21.3	0.23-1.00	3.09-60.5	0.30-3.01	1.13-24.9
NMV	0.033-2.90	0.017-0.19	0.020-6.07	0.020-1.47	0.015-2.79
NH_3	0.015-0.033	-	0.018-0.090	-	0.016-0.049
CO_2	130-190	159-249	128-535	163-411	140-240
PM_{10}[a]	-	0.037-0.13	-	0.0094-0.65	-
$PM_{2.5}$[a]	-	0.037-0.13	-	0.0094-0.65	-
BC	-	0.015-0.053	-	0.0050-0.34	-
OC	-	0.012-0.041	-	0.0023-0.16	-
g/km	**MDTD**	**HDTG**	**HDTD**	**BUSG**	**LBUSD**
SO_2	0.0013-3.42	0.0022-0.03	0.0040-16.6	0.0013-0.03	0.0014-2.01
NO_x	0.72-9.68	0.10-20.1	4.17-46.9	0.071-19.8	0.82-6.30
CO	0.23-3.11	3.92-52.8	0.48-15.1	2.98-54.6	0.29-1.85
NMV	0.021-1.47	0.028-5.23	0.11-7.14	0.039-5.45	0.035-1.03
NH_3	-	0.028-0.090	-	0.022-0.087	-
CO_2	184-446	339-511	612-2127	197-500	192-266
PM_{10}[a]	0.014-0.69	-	0.055-3.35	-	0.022-0.44
$PM_{2.5}$[a]	0.014-0.69	-	0.055-3.35	-	0.022-0.44
BC	0.0081-0.39	-	0.031-1.89	-	0.010-0.20
OC	0.0030-0.15	-	0.011-0.70	-	0.0062-0.12
g/km	**MBUSD**	**HBUSD**	**LSPCG**	**LSPCD**	**HSPCG**
SO_2	0.0023-3.56	0.0040-7.66	0.00091-0.014	0.0014-1.34	0.0022-0.030
NO_x	2.02-10.1	4.83-21.7	0.042-6.07	0.23-2.36	0.10-20.0
CO	0.48-3.29	0.75-7.09	0.70-25.1	0.050-0.83	4.04-53.5
NMV	0.12-1.59	0.17-3.43	0.015-2.81	0.010-0.18	0.029-5.30
NH_3	-	-	0.012-0.049	-	0.028-0.091
CO_2	352-456	619-982	140-241	159-246	339-512

PM_{10}[a]	0.056-0.72	0.094-1.55	-	0.026-0.13	-
$PM_{2/5}$[a]	0.056-0.72	0.094-1.55	-	0.026-0.13	-
BC	0.025-0.32	0.041-0.68	-	0.015-0.071	-
OC	0.015-0.20	0.026-0.42	-	0.0020-0.010	-

g/km	HSPCD	SMC	MC
SO_2	0.0014-7.42	0.00027-0.0025	0.00036-0.0056
NO_x	0.72-21.0	0.13-0.511	0.048-0.59
CO	0.24-6.77	7.01-14.9	17.5-24.2
NMV	0.022-3.22	0.16-2.76	0.13-5.85
NH_3	-	-	-
CO_2	189-951	19.7-50.3	49.3-97.8
PM_{10}[a]	0.015-1.50	-	-
$PM_{2/5}$[a]	0.015-1.50	-	-
BC	0.0083-0.85	-	-
OC	0.0011-0.11	-	-

a. It was assumed that emissions of PM species were only from diesel vehicles and ratios of $PM_{2.5}/PM_{10}$ were 1.0 for all vehicle categories.

For cold start emissions, ratios of cold start and hot emissions for each vehicle type for SO_2, NO_x, CO, NMVOC, and PM species were estimated based on the JEI-DB (JPEC 2012a, b, c; 2014). Then, cold start emissions for each vehicle type were calculated by the hot emissions and the corresponding ratios. Note that the ratios were adopted for all target years without changes which means that effects of regulations on cold start emissions were not considered in REAS3.

S6.3 Evaporative emissions

In REASv3, evaporative emissions from gasoline vehicles except for Japan were estimated using the following equation:

$$E_{EVP} = \sum_i \{NV_i \times EF_{EVPi}(T) \times 365\}$$

where, E_{EVP} is the evaporative emission, i is the vehicle type, NV is the number of vehicles in operation, and EF_{EVP} is the emission factor as a function of surface temperature T. Settings of EF_{EVP} g/vehicle/day were taken from EEA (2016) as follow:

- T: around 20 to 30 °C
    - 14.6 for Passenger cars
    - 22.2 for Light duty vehicles

- 7.50 for Motorcycles

- T: around 10 to 20 ℃
  - 7.80 for Passenger cars
  - 12.7 for Light duty vehicles
  - 4.60 for Motorcycles
- T: around 0 to 10 ℃
  - 5.7 for Passenger cars
  - 9.3 for Light duty vehicles
  - 3.4 for Motorcycles
- T: less than 0 ℃
  - 4.0 for Passenger cars
  - 6.5 for Light duty vehicles
  - 2.6 for Motorcycles

The same as for the cold start emissions, evaporative emissions were estimated each month based on monthly averaged temperature at surface calculated using NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html).

For Japan, evaporative emissions from running loss, hot soak loss and diurnal breaking loss in 2000, 2005, and 2010 were obtained for 6 sub-regions in Japan defined in Sect. S2.3 from the JEI-DB (JPEC 2012a, b, c; 2014). Data between 2000 (2005) and 2005 (2010) were interpolated and those before 2000 and after 2000 were extrapolated using the following trend factors:

- Running loss: Trends of traffic volumes of gasoline vehicles
- Diurnal breaking loss: Trends of number of gasoline vehicles
- Hot soak loss: Trends of emissions from gasoline vehicles roughly estimated by number of gasoline vehicles and corresponding emission factors obtained from JPEC (2012a).

S6.4 Speciation of NMVOC emissions

Emission factors of NMVOC described in Sects. S6.2 and S6.3 were for total NMVOC. In REASv3, total NMVOC emissions were allocated to 19 NMVOC species categories defined in Sect. S2.1. The speciation was conducted based on speciation profiles for exhaust emissions from each vehicle type and evaporative emissions provided by D. G. Streets (private communication) based on Klimont et al. (2002a) used for REASv1 and REASv2. The speciation profiles were commonly used for all countries and periods.

S7. Other transport

S7.1 Sub-sectors included in REASv3

In REASv3, emissions from railway, pipeline transport and non-specified sectors defined in the International Energy Agency (IEA) World Energy Balances (IEA, 2017) were included in transport sector except for road transport. Aviation and navigation are out of scope of REASv3.

S7.2 Activity data

Activity data in other transport sectors are fuel consumption which was described in Sect. S3.1.

S7.3 Emission factors

Table 7.1 summarized emission factors for diesel oil and heavy fuel oil combustion in railway sector. For emission factors of other sources and speciation of NMVOC species, settings for fuel combustion in industry sector were used as default. Note that emission controls were not considered emissions from other transport sectors in REASv3.

Table 7.1. Emission factors for diesel oil and heavy fuel oil combustion in railway. Unit is t/PJ (expressed as NO_2 for NO_x).

	Diesel oil	Heavy fuel oil
NO_x[a]	900	1249
CO[a]	250	1000
NMVOC[a]	200	110
NH_3[a]	0.16	0.01
CO_2[b]	74100	77400
PM_{10}[c]	102	143
$PM_{2.5}$[c]	96.4	135
BC[c]	44.0	58.5
OC[c]	25.0	39.0

a. ABC Emission Inventory Manual (Shrestha et al., 2013). b. IPCC (2006). c. Klimont et al. (2002b) and Kupiainen and Klimont (2004) for PM species.

S7.4 Speciation of NMVOC emissions

Emission factors of NMVOC described in Sect. S7.3 were for total NMVOC. In REASv3, total NMVOC emissions were allocated to 19 NMVOC species categories defined in Sect. S2.1. The speciation was conducted based on speciation profiles for exhaust emissions from each vehicle type and evaporative emissions provided by D. G. Streets (private communication) based on Klimont et al. (2002a) used for REASv1 and REASv2. The speciation profiles were commonly used for all countries and periods.

S8. Non-combustion sources of NH_3

S8.1 Manure management

S8.1.1 Methodology

In REASv3, gridded emissions from manure management were developed based on following procedures except for Japan (see Sect. S8.1.5 for Japan):

1. Gridded emissions of REASv1 (Yamaji et al., 2004) in 2000 were used for based data.
2. Emissions in each country and region during 1950-2015 were estimated using numbers of livestock as activity data (see Sect. S8.1.2) and emission factors for each livestock (see Sect. S8.1.3).
3. Spatial allocation factors of emissions in target years were created using the base data and ratios of emission amounts in each grid between target years and 2000 obtained from the Emission Database for Global Atmospheric Research (EDGAR) version 4.3.2 during 1970-2012 (Crippa et al., 2016). Before 1970 and 2012, data in 1970 and 2012 were used, respectively.
4. Annual gridded emissions data in each country and region during 1950-2015 were developed using the base data described in No.1, ratios of emissions between target years and the base year based on the trends of emissions estimated in No.2, and the spatial allocation factors for each country and region in target years developed in No.3. Note that emission values estimated in No.2 were not directly used in REASv3.
5. Monthly gridded data during 1950-2015 were created using the annual gridded emission data developed in No.4 and monthly allocation factors for each country and region (see Sect. S8.1.4).

Note that in REASv3, emissions from animal manures utilized for fertilizers are not included in manure management, but in fertilizer application (see Sect. S8.2).

S8.1.2 Activity data

As described in Sect. S8.1.1, activity data to estimate NH_3 emissions from manure management of livestock are number of livestock. In REASv3, contributions from following livestock were included: buffalo, dairy cows, other cattle, swine, goats, sheep, horses, camels, mules and asses, broilers, ducks, geese, laying hens, and turkeys. National data were derived from FAOSTAT (http://www.fao.org/faostat/en/) during 1961-2015 and extrapolated to 1950 using Mitchell (1998). For China, weighting factors for regional distribution were calculated during 1987-2015 based on China Statistical Yearbook (National Bureau of Statistics of China, 1986–2016). The weighting factors in 1987 were used for the data before 1986. For regional weighting factors for India, data during 1997-2012 were estimated based on Livestock Census (http://www.dahd.nic.in/about-us/divisions/statistics/) and the weighting factors in 1997 and 2012 were used before 1997 and 2012, respectively.

S8.1.3 Emission factors

Annual emission factors of manure management were taken from EEA (2016) for emissions from housing, storage and yards for all countries and regions except for China. For China, regional monthly emission factors were estimated based on those for manure spreading from Xu et al. (2016) and ratios of emission factors between manure management and manure applied to soils from EEA (2016). The emission factors were commonly used for all target years.

S8.1.4 Monthly allocation factors

For China, as descried in Sect. S8.1.3, monthly emission factors were estimated. For other countries and regions, monthly allocation factors were calculated based on relationships between monthly weighting factors of Japan from the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC, 2012b; 2014) and monthly averaged temperature.

S8.1.5 Japan

In REASv3, gridded emissions from manure management were developed based on following procedures:

1. Monthly gridded emissions of JEI-DB (JPEC, 2012b; 2014) in 2000 were used as based data before 2002 and those in 2005 were used after 2003.
2. Emissions in 47 prefectures during 1950-2015 were estimated using numbers of livestock as activity data and corresponding emission factors for each livestock.
3. Monthly gridded emissions data in each prefecture during 1950-2015 were developed using the base data described in No.1, ratios of emissions between target years and the base year based on the trends of emissions estimated in No.2. Note that emission values estimated in No.2 were not directly used in REASv3.
4. Monthly gridded data during 1950-2015 were created by adding data of each prefecture developed in No.3.

For Japan, contributions from following livestock were included: dairy cows, other cattle, fattening pigs, other hogs, sheep, goats, broilers, and layers. Data of each prefecture during

1960-2015 were obtained from the statistics of Ministry of Agriculture, Forestry and Fisheries (https://www.maff.go.jp/j/tokei/kouhyou/tikusan/) and extrapolated using Historical Statistics of Japan (Japan Statistical Association, 2006). Emission factors were taken from EEA (2016) for housing, storage and yards the same as for other countries and regions.

S8.2 Fertilizer application

S8.2.1 Methodology

In REASv3, gridded emissions from fertilizer were developed based on following procedures except for Japan (see Sect. S8.2.5 for Japan):

1. Gridded emissions of REASv1 (Yan et al., 2003) in 2000 were used for based data.
2. Emissions from both synthetic fertilizer and animal manure used as fertilizer in each country and region during 1950-2015 were estimated. Those from synthetic fertilizer were calculated using amounts of applied synthetic fertilizer (see Sect. S8.2.2) and emission factors for each fertilizer type (see Sect. S8.2.3) and those from animal manure were estimated based on number of livestock (see Sect. S8.1.2) and emission factors for each livestock (see S8.2.3).
3. Spatial allocation factors of emissions in target years were created using the base data and ratios of amounts of synthetic nitrogen fertilizer applied to each grid between target years and 2000 obtained from Nishina et al. (2017) during 1961-2010. Before 1961 and 2010, data in 1961 and 2010 were used, respectively.
4. Annual gridded emissions data in each country and region during 1950-2015 were developed using the base data described in No.1, ratios of emissions between target years and the base year based on the trends of emissions estimated in No.2, and the spatial allocation factors for each country and region in target years developed in No.3. Note that emission values estimated in No.2 were not directly used in REASv3.
5. Monthly gridded data during 1950-2015 were created using the annual gridded emission data developed in No.4 and monthly allocation factors for each country and region (see Sect. S8.2.4).

S8.2.2 Activity data

Synthetic fertilizer

As described in Sect. S8.2.1, activity data to estimate NH_3 emissions from synthetic fertilizer are applied amounts of synthetic fertilizer. In REASv3, contributions from following synthetic fertilizer were included: ammonium nitrate, ammonium phosphate, ammonium sulphate, ammonium sulphate nitrate, ammonium bicarbonate, calcium ammonium nitrate, calcium nitrate, sodium nitrate, urea, other nitrogen fertilizer, and other complex fertilizer.

For China, national data of different fertilizers were taken from FAOSTAT (http://www.fao.org/faostat/en/) and Fu et al. (2017) during 1982-2015. The data were extrapolated to 1950 and regionally distributed based on total consumption of chemical fertilizer obtained from China Data Online. For India, national data for each fertilizer type were taken from FAOSTAT during 1982-2015 and regionally distributed using state-wise consumption of nitrogen fertilizers obtained from Indiastat. The data were extrapolated to 1961 using national consumption data of total nitrogen fertilizer in India from FAOSTAT and to 1950 based on global nitrogen fertilizer consumption from Hammond and Matthews (1999). Using the same procedures for India, national data of other countries and regions for each fertilizer type were derived from FAOSTAT and were extrapolated based on national and global nitrogen fertilizer consumption data.

Animal manure

Activity data for emissions from animal manure used as fertilizer are numbers of livestock and the same data for manure management described in Sect. S8.1.2 were used.

S8.2.3 Emission factors

Synthetic fertilizer

Annual emission factors of ammonium nitrate, ammonium phosphate, ammonium sulphate, calcium ammonium nitrate, and urea were based on EEA (2016). In REASv3, data for normal pH and temperate climate were adopted. For ammonium bicarbonate, emission factor was obtained from Yan et al. (2003). For other fertilizers including ammonium sulphate nitrate, calcium nitrate, sodium nitrate, other nitrogen fertilizer, and other complex fertilizer, data of other straight N compounds in EEA (2016) were used. The emission factors were commonly used for all target years.

Animal manure

Annual emission factors of from animal manure used as fertilizer were taken from EEA (2016) for emissions from following manure application for all countries and regions except for China. For China, regional monthly emission factors were taken from Xu et al. (2016). The emission factors were commonly used for all target years.

S8.2.4 Monthly allocation factors

Synthetic fertilizer

For China, monthly allocation factors in REASv3 were estimated based on monthly application nitrogen ratio taken from Xu et al. (2015). The data were used commonly for each grid in China during 1950-2015. For other countries and regions, first, monthly allocation factors were calculated using N fertilizer application amounts for each country and region obtained from Nishina et al. (2017) during 1961-2010. In the calculated monthly factors, there are cases that some months have high factors, whereas the others have almost zero. In REASv3, the highest monthly factor was set at 0.2 and the factors of all months were adjusted accordingly referring to Janssens-Maenhout et al. (2015). The modified monthly factors during 1961-2010 were commonly used for each country and region and data in 1961 and 2010 were used before 1960 and 2011, respectively.

Animal manure

For China, as descried in Sect. S8.2.3, monthly emission factors were estimated. For other countries and regions, monthly allocation factors were calculated based on relationships between monthly weighting factors of Japan from JEI-DB (JPEC, 2012b; 2014) and monthly averaged temperature.

S8.2.5 Japan

In REASv3, gridded emissions from fertilizer application were developed based on following procedures:

1. Monthly gridded emissions of JEI-DB (JPEC, 2012b; 2014) in 2000 were used as based data before 2002 and those in 2005 were used after 2003.
2. Emissions from both synthetic fertilizer and animal manure used as fertilizer in 47 prefectures during 1950-2015 were estimated. Those from synthetic fertilizer were calculated using amounts of applied synthetic fertilizer and emission factors for each fertilizer type and those from animal manure were estimated based on number of livestock and emission factors for each livestock.
3. Monthly gridded emissions data in each prefecture during 1950-2015 were developed using the base data described in No.1, ratios of emissions between target years and the base year based on the trends of emissions estimated in No.2. Note that emission values estimated in No.2 were not directly used in REASv3.

4. Monthly gridded data during 1950-2015 were created by adding data of each prefecture developed in No.3.

Synthetic fertilizer

Activity data for emissions from synthetic fertilizer were applied amounts of synthetic fertilizers. National data of different fertilizers were derived from FAOSTAT during 1971-2002 and extrapolated during 1960-2015 and distributed to 47 prefectures based on data in Fertilizer Statistics Yearbook (Newspaper department of Japan Fertilizer Association), Handbook of Fertilizer (Association of Agriculture and Forestry Statistics) and statistics provided by Japan Fertilizer & Ammonia Producers Association (http://www.jaf.gr.jp/en.html). The data were extrapolated to 1950 based on global nitrogen fertilizer consumption from Hammond and Matthews (1999).

Animal manure

Activity data for emissions from animal manure used as fertilizer are numbers of livestock and the same data for manure management described in Sect. S8.1.5 were used. Emission factors were taken from EEA (2016) for manure applied to soils the same as for other countries and regions. The emission factors were commonly used for all target years.

S8.3 Industrial production

In REASv3, NH_3 emissions from industrial processes for production of ammonia, ammonium nitrate, and urea (fertilizers) are considered. National production amounts of ammonia during 1990-2015 were obtained from Minerals Yearbook (USGS). For China, data before 1990 were taken from Vroomen (2013). Data of Japan before 1990 were derived from the Historical Statistics of Japan (Japan Statistical Association, 2006). For other countries, data were extrapolated based on trends of production capacity obtained from World Nitrogen Survey (Constant and Sheldrick, 1992). For urea and ammonium nitrate, data of Japan were derived from Handbook of Fertilizer (Association of Agriculture and Forestry Statistics). For China, national production amounts of urea obtained taken from Vroomen (2013). Other national data of urea and ammonium nitrate were estimated from IFASTAT (https://www.ifastat.org/) and World Nitrogen Survey (Constant and Sheldrick, 1992). For regional distribution of Japan, weighting factors were developed using reginal shipment data for chemical industrial products obtained from Ministry of Economy, Trade and Industry (https://www.meti.go.jp/statistics/tyo/kougyo/index.html). For China, regional production ratios of urea in 2015 were used as weighting factors. For India, national data were distributed to each region using total energy consumption in chemical industry developed based on methodologies described in Sect. S3.1.

Emission factors for industrial process emissions from production of ammonia, and urea were derived from Shrestha et al. (2013). For ammonium nitrate, median of the range provided in Shrestha et al. (2013) were used. The emission factors were adopted for all target countries and periods.

S8.4 Human

NH_3 emissions from human perspiration and respiration were included in REASv3. Activity data are number of total population in each country and region. See descriptions for domestic use of solvents in Sect. S5.1.2 for data sources of total population. Emission factors were taken from Kannari et al. (2001) and adopted for all target countries and periods.

S8.5 Latrines

In REASv3, emissions from latrines were estimated based on number of population in no sewage service areas. For Japan, data were obtained from Mizuochi (2012) and MOEJ (Ministry of Environment of Japan) (2017b). Due to lack of information, corresponding data in other countries and regions were roughly estimated based on the following assumptions referring Kanamori and Hijioka (2013):

- Rep. of Korea, Taiwan, Singapore, Hong Kong, and Macau: ratios of population in sewage service areas were 95 percent of Japan
- Beijing and Shanghai: ratios of population in sewage service areas were 60 percent of Japan
- Other countries and regions: ratios of population in sewage service areas were one-third of Japan

Emission factor for latrines was taken from Vallack and Rypdal (2012) which was half value provided in EEA (2016) and adopted for all target countries and periods.

S9. Spatial and temporal distribution

S9.1 Grid allocation factors

S9.1.1 Population distribution

In REASv3, spatial distributions of total population were used as default grid allocation factors. In addition, urban and rural population distributions were also used for spatial allocation factors for several sectors (see Sects. S9.1.2, S9.1.3, S9.1.4, and S9.1.6). HYDE 3.2.1 (Klein Goldwijk et al. 2017) provides total, urban, and rural population data with $5' \times 5'$ in 1950, 1960, 1970, 1980, 1990, 2000, 2005, 2010, and 2015. REASv3 used the total, urban, and rural population data of HYDE 3.2.1 as weighting factors to create grid allocation factors for .$0.25° \times 0.25°$ data. The data of missing years were created by interpolation.

S9.1.2 Power plants

As described in Sects. S2.5 and S3.1.6, REASv3 treats large power plants as point sources and information of longitude and latitude were provided with emissions from each power plant. The locations of power plants were surveyed using internet services such as Industry About (https://www.industryabout.com/), Global Energy Observatory (http://globalenergyobservatory.org/), and search engines based on names of units, plants, and companies derived from the World Electric Power Plants Database (WEPP) (Platts, 2018). Emissions form area sources were distributed to grid cells using based on total population distribution. For Japan, emissions from area sources were gridded using grid allocation factors for other industries (see Sect. S9.1.7).

S9.1.3 Iron and steel industry

In REASv3, iron and steel plants were not treated as point sources, but grid allocation factors for iron and steel industry were developed as follows:

1. Major iron and steel plants including names, production capacities, and start years of operations were surveyed using Minerals Yearbook (USGS), websites of iron and steel plants, and internet search engines. For plants without information of production capacity and start years of operations, small values were assumed for production capacities by referring to other plants in each country and region and the data were used for all target years to estimate grid allocation factors.
2. Locations of the surveyed plants were searched using internet services such as Industry About

(https://www.industryabout.com/), websites of iron and steel plants, and Google Map.

3. Grid allocation factors were created for each target year based on longitude and latitude and production capacity of each plant in operation used as weighting factors.

One problem of these grid allocation factors is that not all emissions in iron and steel industry sector were from plants considered in above procedures. In REASv3, 80% of both combustion and non-combustion emissions from iron and steel industry were allocated to grid cells using the grid allocation factors developed here. For the other 20%, emissions were distributed to grid cells based on total population distribution except for Japan where grid allocation factors for other industries (see Sect. S9.1.7) were used.

S9.1.4 Cement industry

The same as for iron and steel plants, in REASv3, cement plants were not treated as point sources, but grid allocation factors for iron and steel industry were developed as follows:

4. Major cement plants including names, production capacities, and start years of operations were surveyed using Minerals Yearbook (USGS), websites of cement plants, and internet search engines. For plants without information of production capacity and start years of operations, small values were assumed for production capacities by referring to other plants in each country and region and the data were used for all target years to estimate grid allocation factors.
5. Locations of the surveyed plants were searched using internet services such as Industry About (https://www.industryabout.com/), websites of cement plants and Google Map.
6. Grid allocation factors were created for each target year based on longitude and latitude and production capacity of each plant in operation used as weighting factors.

Also, the same as for the case of iron and steel plants, one problem of these grid allocation factors is that not all emissions in cement industry sector were from plants considered in above procedures. In REASv3, 80% of both combustion and non-combustion emissions from cement industry were allocated to grid cells using the grid allocation factors developed here. For the other 10%, emissions were distributed to grid cells based on total population distribution except for Japan where grid allocation factors for other industries (see Sect. S9.1.7) were used.

S9.1.5 Road transport

Grid allocation factors for road transport sector were created from other emission inventory datasets. For Japan, gridded emission data of the Japan Auto-Oil Program (JATOP) Emission Inventory-Data Base (JEI-DB) (JPEC 2012a, c; 2014) 2000, 2005, and 2010 were used to create grid allocation factors for each target species. For the year between 2000 and 2005/2005 and 2010, the

JEI-DB data were interpolated. Before 2000 and after 2010, the JEI-DB data for 2000 and 2010 were used, respectively. For other countries and regions, grid allocation factors for each species were created using gridded emission data of road transport sector of the Emission Database for Global Atmospheric Research (EDGAR) version 4.3.2 (Crippa et al., 2016) during 1970-2012. Before 1970 and after 2012, data for 1970 and 2012 were used, respectively.

S9.1.6 Domestic sectors

Residential fuel combustion

For China, emissions from residential fuel combustion were estimated in urban and rural areas separately. They were distributed to grid cells based on rural and total population distribution, respectively. For other countries and regions, emissions from fuel combustion were estimated for total residential sector. For emissions from coal fuels, kerosene, and biofuels combustion, grid allocation factors developed using rural population distribution were used. For other fuels, emissions were distributed to grid cells based on total population distribution.

Commercial and public services (fuel combustion)

Emissions were distributed to grid cells based on urban population distribution.

Agriculture and forestry (fuel combustion)

Emissions were distributed to grid cells based on rural population distribution.

NMVOC non-combustion emissions related to residential activities

Emissions from dry cleaning and waste disposal were distributed to grid cells based on urban and rural population distributions, respectively. For those from domestic use of solvents and paint, grid allocation factors developed using total population distribution were used.

NH_3 emissions related to human biological phenomenon

Emissions from human perspiration and respiration were distributed to grid cells based on total population distribution and those from latrines were gridded using grid allocation factors developed using rural population distribution.

S9.1.7 Others

For all other sources which were not included in descriptions in Sects. S9.1.2-6, emissions were allocated to grid cells based on total population distribution except for Japan. Grid allocation factors for the other sources of Japan were summarized in Table 9.1.

Table 9.1. Data sources and treatments for grid allocation factors for Japan for sources not described in Sects. S9.1.2-S9.1.6.

Sector categories	Data sources and treatment
Non-ferrous metal industry	● Longitude and latitude, start years of operations, and production capacities of copper, zinc, lead, and aluminium plants surveyed using Minerals Yearbook (USGS), websites of non-ferrous metal plants, and internet search engines. ● Using the same methodology for iron and steel industry described in Sect. S9.1.3, grid allocation factors were developed for copper, zinc, lead, aluminium, and total non-ferrous metal sectors independently. Data for total non-ferrous metal sector include points of all non-ferrous metal plants. ● Emissions from non-combustion sources were estimated for each metal sector and corresponding grid allocation factors were used. For combustion sources, grid allocation factors for total non-ferrous metal sector were used. ● Similar to the methodology for iron and steel, 80% of emissions from non-ferrous metal industry sectors were allocated to grid cells using the grid allocation factors developed here. For the other 20%, emissions were distributed to grid cells based on grid allocation factors for other industries (see “Other industry” of this table).
Other industry	● Grid allocation factors for each target species were created based on gridded emission data of JEI-DB (JPEC 2012b, c; 2014) in 2000, 2005 and 2010 for industry sector where contributions from grids including point sources of iron and steel, cement, and non-ferrous metals were excluded. For the year between 2000 and 2005/2005 and 2010, the data were interpolated. Before 1999 and after 2011, the data for 2000 and 2010 were used, respectively.
NMVOC evaporative sources	● Grid allocation factors were created based on gridded emission data of JEI-DB (JPEC 2012b, c; 2014) for NMVOC evaporative sources

	using the same methodology for road transport sector described in Sect. S9.1.5.

S9.2 Monthly variation factors

S9.2.1 Power plants

Data sources and treatment for monthly variation factors used in REASv3 were summarized in Table 9.2.

Table 9.2. Data sources and treatments for monthly variation factors for emissions from power plants used in REASv3.

Countries and regions	Data sources and treatment
China	● Weighting factors: Monthly generated electricity ● Data sources and treatment: ➢ Regional data during 2002-2010 were obtained from China Data Online. Before 2001 and after 2011, the data in 2002 and 2011 were used, respectively. ➢ Estimated monthly variation factors were used for all fuel types.
India	● Weighting factors: Monthly thermal generation of electricity ● Data sources and treatment: ➢ National data during in 2000, 2005, 2010 were taken from Monthly Abstract of Statistics (Ministry of Statistics and Programme Implementation, http://mospi.gov.in/). Data during 2001-2004/2006-2009 were interpolated. Before 1999 and after 2011, the data in 2000 and 2010 were used, respectively. ➢ Estimated monthly variation factors were used for all fuel types and regions.
Japan	● Monthly variation factors were derived from a report of JEI-DB (2014) and used for all fuel types, regions, and periods.
Taiwan	● Weighting factors: Monthly generated electricity ● Data sources and treatment: ➢ National data in 2011 were taken from Monthly Bulletin of Statistics (National Statistics, https://eng.stat.gov.tw/). ➢ Estimated monthly variation factors were used for all fuel types and periods.

Thailand	● Monthly variation factors were derived from Thao Pham et al. (2008) and used for all fuel types, regions, and periods.
Vietnam	● Weighting factors: Monthly generated electricity ● Data sources and treatment: ➢ National data during 2005-2010 were taken from monthly statistics provided by General Statistics Office of Vietnam (https://www.gso.gov.vn/). Before 2004 and after 2011, the data in 2005 and 2010 were used, respectively. ➢ Estimated monthly variation factors were used for all fuel types.

S9.2.2 Industry

Data sources and treatment for monthly variation factors used in REASv3 were summarized in Table 9.3. Note that emissions from industry sub-categories not described in Table 9.3 were distributed to each month using number of dates as weighting factors.

Table 9.3. Data sources and treatments for monthly variation factors for emissions from power plants used in REASv3.

Countries and regions	Data sources and treatment
China	● Weighting factors: Monthly production ● Data sources and treatment: ➢ Regional data of steel and cement during 2002-2010 were derived from China Data Online. Before 2001 and after 2011, the data in 2002 and 2011 were used, respectively. Monthly variations based on steel (cement) production were adopted to both combustion and non-combustion emissions from iron and steel (cement) industry. ➢ National data of coke and sulphuric acid production during 2006-2010 were derived from China Data Online. Before 2005 and after 2011, the data in 2006 and 2010 were used, respectively. The monthly variations based on coke production were adopted to both combustion and non-combustion emissions from coke industry and those for sulphuric acid were used only for non-combustion emissions. ➢ National data of copper, zinc, lead, and aluminum in 2001 and 2002 were obtained from JOGMEC (2002-2003). Before 2000 and after 2003, data in 2001 and 2002 were used. The monthly variations

	based on production of each metal type were adopted to non-combustion emissions from each metal industry. Those for combustion in non-ferrous metal industry were estimated using production amounts of total non-ferrous metals. ➢ For petroleum refinery, monthly variations were calculated based on national monthly processed volume of crude oil derived from China Data Online during 2006-2010. Before 2005 and after 2011, the data in 2006 and 2010 were used, respectively. The monthly variations were adopted to both combustion and non-combustion emissions from petroleum refinery industry including energy sector. ➢ For other industries, monthly variations were calculated using numbers of each month as weighting factors. ➢ Estimated monthly variation factors were used for all fuel types
India	● Weighting factors: Monthly production ● Data sources and treatment: ➢ National data during in 2000, 2005, 2010 were taken from Monthly Abstract of Statistics (Ministry of Statistics and Programme Implementation, http://mospi.gov.in/). Data during 2001-2004/2006-2009 were interpolated. Before 1999 and after 2011, the data in 2000 and 2010 were used, respectively. Following monthly variations were estimated. ✧ Pig iron: Non-combustion emissions from pig iron production ✧ Steel products: Non-combustion emissions from steel production ✧ Total production amounts of iron and steel: Combustion emissions from iron and steel industry ✧ Total production amounts of non-ferrous metals: Combustion and non-combustion emissions from non-ferrous metal industry ✧ Cement: Combustion and non-combustion emissions from cement industry ✧ Non-metallic mineral products (index numbers of industrial production): Combustion and non-combustion emissions from non-metallic minerals industry except for cement and brick. ✧ Sulphuric acid: Non-combustion emissions from sulphuric acid production ✧ Coke: Combustion and non-combustion emissions from coke industry

	✧ Total production amounts of refined petroleum: Combustion and non-combustion emissions from petroleum refinery including energy sector. ➢ Emissions from brick production were allocated to November to June referring Maithel (2013). ➢ Estimated monthly variation factors were used for all fuel types.
Japan	● Monthly variation factors were derived from a report of JEI-DB (JPEC, 2014) and adopted as follows: ➢ Iron and steel industry: Combustion and non-combustion emissions from iron and steel industry ➢ Construction: Combustion emissions from construction industry. ➢ Petroleum refinery: Combustion and non-combustion emissions from petroleum refinery including energy sector. ➢ Gas works: Combustion emissions from manufacture of gaseous fuels including energy sector ➢ Other industry sectors: Settings of monthly variations for other industries in JPEC (2014) are relatively close and in REASv3, their averaged values were adopted to combustion and non-combustion emissions from other industries not included above. ● Estimated monthly variation factors were used for all fuel types and periods.
Republic of Korea	● Weighting factors: Monthly production ● Data sources and treatment: ➢ Pig iron: and crude steel: National data during 2000-2010 were taken from Steel Statistical Yearbook (https://www.worldsteel.org/steel-by-topic/statistics/steel-statistical-yearbook.html). Monthly production amounts of pig iron (crude steel) were used to calculate monthly variations for non-combustion emissions from pig iron (crude steel) production. Monthly variations for combustion emissions from iron and steel industry were estimated based on total production amounts of pig iron and crude steel. Before 1999 and 2011, monthly variations in 2000 and 2010 were used, respectively. ➢ Estimated monthly variation factors were used for all fuel types.
Taiwan	● Weighting factors: Monthly production ● Data sources and treatment:

	➢ Cement: National data in 2011 were taken from Monthly Bulletin of Statistics (National Statistics, https://eng.stat.gov.tw/). Estimated monthly variation factors were used for all fuel types and periods. ➢ Pig iron: and crude steel: National data during 2000-2010 were taken from Steel Statistical Yearbook (https://www.worldsteel.org/steel-by-topic/statistics/steel-statistical-yearbook.html). Monthly production amounts of pig iron (crude steel) were used to calculate monthly variations for non-combustion emissions from pig iron (crude steel) production. Monthly variations for combustion emissions from iron and steel industry were estimated based on total production amounts of pig iron and crude steel. Before 1999 and 2011, monthly variations in 2000 and 2010 were used, respectively. Estimated monthly variation factors were used for all fuel types.
Brunei	● Monthly variations for NMVOC emissions from crude oil production were estimated based on monthly crude oil production during 2000 and 2005 taken from Brunei Economic Bulletin. Before 1999 and 2006, monthly variations in 2000 and 2005 were used.
Indonesia	● Combustion and non-combustion emissions from brick production were mainly allocated to dry seasons during June to September.
Malaysia	● Monthly production amounts during 2008-2010 were taken from Monthly Statistics Bulletin Malaysia and adopted as follows: ➢ Iron and steel: Combustion and non-combustion emissions from iron and steel industry. ➢ Cement: Combustion and non-combustion emissions from cement industry. ➢ Crude oil: NMVOC emissions from crude oil production ➢ Natural gas: NMVOC emissions from natural gas production ➢ Before 2007 and after 2011, monthly variations in 2008 and 2010 were used, respectively. ● Combustion and non-combustion emissions from brick production were mainly allocated to dry seasons during June to September. ● Estimated monthly variation factors were used for all fuel types.
Myanmar	● Combustion and non-combustion emissions from brick production were mainly allocated to dry seasons during December to April.
Philippines	● Monthly variations of emissions were estimated during 2001-2010 based

	on value of production index taken from Philippine Statistics Authority and adopted as follows: ➢ Iron and steel: Combustion and non-combustion emissions from iron and steel industry. ➢ Non-ferrous metal: Combustion and non-combustion emissions from non-ferrous metal industry. ➢ Cement: Combustion and non-combustion emissions from cement industry. ➢ Non-metallic minerals: Combustion and non-combustion emissions from non-metallic minerals industry except for cement. ➢ Refined petroleum products: Combustion and non-combustion emissions from petroleum refinery. ➢ Before 2000 and after 2011, monthly variations in 2001 and 2010 were used, respectively. ● Estimated monthly variation factors were used for all fuel types.
Singapore	● Relative ratios of monthly production in 2006, 2008, and 2009 were estimated based on Monthly digest statistics Singapore and adopted as follows: ➢ Refinery petroleum products: Combustion and non-combustion emissions from petroleum refinery including energy sector. ➢ Non-metallic minerals products: Combustion and non-combustion emissions from cement industry. ➢ Before 2007 and after 2010, data in 2006 and 2009 were used, respectively. Estimated monthly variation factors were used for all fuel types.
Thailand	● Monthly variation factors were derived from Thao Pham et al. (2008) and adopted as follows: ➢ Basic Metal: Iron and steel and non-ferrous metal ➢ Chemicals: Chemical and petrochemical ➢ Non-Metal: Cement, lime, and non-specified non-metallic minerals except for brick ➢ Food & Beverage: Food and tobacco ➢ Paper: Paper, pulp and printing ➢ Wood & Furniture: Wood and wood products ➢ Textile: Textile and leather ➢ Other: Other industry sectors not included above except for brick

	industry whose emissions were mainly allocated to dry seasons during November to May. ● Estimated monthly variation factors were used for all fuel types and periods.
Vietnam	● Monthly variations of emissions were estimated during 2005-2010 based on production amounts taken from General Statistics Office of Viet Nam and adopted as follows: ➢ Cement: Cement: Combustion and non-combustion emissions from cement industry. ➢ Crude oil: NMVOC emissions from crude oil production ➢ Natural gas: NMVOC emissions from natural gas production ➢ Before 2004 and after 2011, monthly variations in 2005 and 2010 were used, respectively. ● Combustion and non-combustion emissions from brick production were mainly allocated to dry seasons during December to March. ● Estimated monthly variation factors were used for all fuel types.
Pakistan	● Weighting factors: Monthly production ● Data sources and treatment: ➢ Pig iron: and crude steel: National data during 2000-2010 were taken from Steel Statistical Yearbook (https://www.worldsteel.org/steel-by-topic/statistics/steel-statistical-yearbook.html). Monthly production amounts of pig iron (crude steel) were used to calculate monthly variations for non-combustion emissions from pig iron (crude steel) production. Monthly variations for combustion emissions from iron and steel industry were estimated based on total production amounts of pig iron and crude steel. Before 1999 and 2011, monthly variations in 2000 and 2010 were used, respectively. ➢ Estimated monthly variation factors were used for all fuel types.
Bangladesh Nepal Sri Lanka	● Monthly variations of India were used for combustion and non-combustion emissions from brick production.

S9.2.3 Road transport

Japan

Monthly variation factors for total emissions from road transport including hot, cold start and NMVOC evaporative emissions were calculated for each region using gridded emission data of JEI-DB (JPEC 2012a, c; 2014) in 2000, 2005, and 2010 for each species. For the year between 2000 and 2005/2005 and 2010, the data were interpolated. Before 1999 and after 2011, the data for 2000 and 2010 were used, respectively.

Other countries and regions

In REASv3, cold start emissions were estimated on a monthly basis using monthly average surface temperature. For hot emissions and NMVOC evaporative emissions, monthly variations were not considered. Annual emissions were distributed to each month using number of date as weighting factors. Data of surface temperature were obtained from NCEP reanalysis data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA (https://psl.noaa.gov/data/gridded/data.ncep.reanalysis.html).

S9.2.4 Residential combustion

Japan

Monthly variation factors for gas fuels, kerosene, and liquefied petroleum gas (LPG) were taken from a report of JEI-DB (2014) and used for all regions and periods. For other fuel types, data for LPG were adopted.

Other countries and regions

In REASv3, monthly variation of emissions from residential combustion was assumed to be correlated to monthly average surface temperature. Based on monthly proportions of coal consumption in Beijing, Tianjin, and Hebei taken from Zhu et al. (2018), indices of residential emissions as functions of monthly average temperature were created. Using the indices, monthly variations of emissions from residential combustion were estimated for each country and region based on monthly average surface temperature.

S9.2.5 Others

NH_3 emissions from human and latrines

Monthly variations for Japan were obtained from JPEC (2014). For other countries and regions, similar to residential combustion described in Sect. S9.2.4, indices of emissions as function of monthly average surface temperature were created using data of JPEC (2014), assuming that NH_3 emissions from human and latrines are correlated to surface temperature. Then, using the indices, monthly variations of emissions from residential combustion were estimated for each country and region based on monthly average surface temperature.

NMVOC emissions from solvent and paint use

Monthly variation of evaporative emissions from solvent and paint use for reach region of Japan were calculated using gridded emission data of JEI-DB (JPEC 2012b, c; 2014) in 2000, 2005 and 2010 for total solvent use. For the year between 2000 and 2005/2005 and 2010, the data were interpolated. Before 1999 and after 2011, the data for 2000 and 2010 were used, respectively. For other countries and regions, annual emissions were distributed to each month using number of date as weighting factors.

Other sources

For other sources not described above, annual emissions were distributed to each month using number of date as weighting factors.

S10. Uncertainties

S10.1 Methodology

In REASv3, uncertainties of emissions were estimated after Streets et al. (2003) and Huang et al. (2011) using the following equation:

$$U_{i,j} = 1.96 \times \sqrt{(1+U_A^2)(1+U_F^2)-1} \times E_{i,j} \quad (1)$$

where $E_{i,j}$ and $U_{i,j}$ represents respectively emission and its uncertainty for sub-sector category j and its activity i, U_A is uncertainty of i and U_F is uncertainty of emission factor for i and j. U_F were generally estimated based on uncertainties of emission factors (U_{EF}) and those of removal ratios (U_R) as follows:

$$U_F = \sqrt{U_{EF}^2 + U_R^2} \quad (2)$$

U_F for SO_2 emissions based on sulfur contents of fuels and ratio of sulfur retention in ash were estimated using the following equation:

$$U_F = \sqrt{U_S^2 + U_{ERS}^2 + U_R^2} \quad (3)$$

where U_S, U_{ERS}, and U_R represent uncertainties of sulfur contents in fuels, ratios of sulfur retention in ash, and removal ratios, respectively. For road transport sectors, activity data is annual mileage which were calculated by number of vehicles and annual distances traveled for each vehicle type. U_A for road transport sector were estimated using following equation:

$$U_F = \sqrt{U_{NV}^2 + U_{ADT}^2} \quad (4)$$

where U_{NV}, and U_{ADT} represent uncertainties of number of vehicles and those of annual distances traveled, respectively.

The uncertainties in emissions from power plants, industries, road transport, other transport, domestic and other sectors, as well as uncertainties in total emissions were calculated for all target species. The uncertainties of different sub-sectors and activities were combined in quadrature and estimated for each country and region. For uncertainties of national emissions in China, India, and Japan, those in their sub-regions were added linearly.

S10.2 Settings of uncertainties of each component

In REASv3, uncertainties in emissions were estimated in 1955, 1985, and 2015 for all species and most sources. In this sub-section, settings of uncertainties of activity data, emission factors, and emission controls and their assumption are described. Note that uncertainties of emissions that were not originally developed in REASv3 (NH_3 emissions from manure management and fertilizer application, and NMVOC evaporative emissions from Japan and the Republic of Korea) were not evaluated.

S10.2.1 Stationary combustion sources

Activity data

Activity data of stationary combustion sources are amounts of fuel consumption in each sub-sector. The data were derived from variety of sources and a lot of treatments were done for missing data as described in Sect. S3.1. Settings of uncertainties of the data were based on the differences of the data sources and following assumption were taken into considered:

- Values of uncertainties were estimated referring EEA (2016) assuming uncertainties of data for Asian countries are generally higher than those of European countries.
- Uncertainties of fossil fuel consumption data are lower for Japan, Republic of Korea, and Taiwan in 2015 and Japan in 1985 compare to other countries and regions. Those for China using the China Energy Statistical Yearbook (CESY) (National Bureau of Statistics of China, 1986, 2001-2017) and those for other countries using the International Energy Agency (IEA) World Energy Balances (IEAWEB) (IEA, 2017) are assumed to be the same.
- Uncertainties of primary biofuels (fuelwood, crop residue, and animal waste) are higher than those of fossil fuels.
- Uncertainties of other fuels such as charcoal and municipal wastes are higher than those of fossil fuels but lower than those of biofuels.
- Uncertainties of data in the United Nations (UN) Energy Statistics Database (UN, 2016) and the UN data, which is a web-based data service of the UN (http://data.un.org/) are higher than those in CESY and IEAWEB. (The data were used for Macau, Laos, Bhutan, Afghanistan, and Maldives in 1955, 1985, and 2015 and Cambodia in 1955 and 1985).
- Uncertainties of data in 2015 are lower than in 1985.
- Uncertainties of fuel consumption in power plants, iron and steel, and cement industries are lower than those in other industries.
- Uncertainties of fuel consumption in residential and other domestic sectors were higher than those in the other industries.
- All fuel consumption data in 1955 were extrapolated using trend factors (see Sect. S3.1). Therefore, the same settings of uncertainties much higher than 1985 are assumed.

Uncertainties of fuel consumption data adopted in REASv3 are summarized in Table 10.1.

Table 10.1. Uncertainties [%] of fuel consumption amounts in 1995, 1985, and 2015 assumed in REASv3. Values in the table (X/Y/Z) are for power plants and iron and steel, and cement industries/other industries/residential and other domestic sectors, respectively.

	Fossil fuels	Primary biofuels	Other fuels
2015			
Japan	±2/±2/±5	±30/±30/±30	±5/±5/±10
Group A	±2/±2/±5	±30/±30/±30	±5/±5/±10
Group B	±10/±15/±20	±30/±30/±30	±15/±20/±25
Group C	±30/±30/±30	±50/±50/±50	±35/±35/±35
1985			
Japan	±5/±10/±15	±40/±40/±40	±10/±15/±20
Group A	±10/±15/±20	±40/±40/±40	±15/±20/±25
Group D	±15/±20/±25	±40/±40/±40	±20/±25/±30
Group E	±40/±40/±40	±60/±60/±60	±45/±45/±45
1955			
All countries and regions	±50/±50/±50	±70/±70/±70	±55/±55/±55

Group A: Republic of Korea and Taiwan. Group B: Countries and regions except for Japan and those in Group A and C using IEAWEB in 2015. Group C: Macau, Laos, Bhutan, Afghanistan, and Maldives using UNESD in 2015. Group D: Group B – Cambodia. Group E: Group C + Cambodia.

Emission factors

For emission factors, two causes need to be considered for their uncertainties. One is uncertainties in the data themselves. Another is those caused by selections of the data including technologies. Values of uncertainties of emission factors were not available in most literature used in REASv3. In addition, there is no specific way to quantify the uncertainties of emission factors caused by the second reason. In REASv3, uncertainties of emission factors were roughly estimated as summarized in Table 10.2 based on the following assumption:

- Uncertainties of CO_2 and SO_2 for gas and oil combustion are smaller than those of others. (Note that uncertainties for SO_2 here were only for ratios of sulfur in fuels emitted as SO_2 and influences of uncertainties in sulfur contents in fuels were not included.)
- Uncertainties of NO_x for fossil fuel combustion are larger than those of CO_2 and SO_2, but smaller than those of other species.
- The same settings were adopted for CO, NMVOC, and PM species for fossil fuel combustion except for those of PM species for coal combustion in residential sector where their

uncertainties are assumed to be larger than those of other species.

- In general, uncertainties for coal combustion are larger than those for gas and oil combustion.
- Uncertainties for biofuel combustion are much larger than those for fossil fuel combustion. Due to lack of information, the same settings were used for uncertainties of emission factors for other fuels including charcoal and municipal wastes.
- The smallest uncertainties were assumed for power plants, the largest ones were assumed for residential and other domestic sectors, and uncertainties for industry sectors were generally between them.
- For industry sectors, uncertainties for iron and steel, and cement industries were smaller than those for other industry sectors.
- The largest uncertainties were assumed for NH_3 due to lack of limited information.
- The common settings of uncertainties were adopted for all countries and regions. Exceptions were uncertainties of emission factors of NO_x for coal combustion in power plants and those of PM species for coal combustion in China based on Zhang et al. (2007) and Lei et al. (2011) where 10% smaller values than those of other countries and regions were adopted.
- It was assumed that estimated uncertainties for emission factors include effects from limited information of technologies.

Note that except for SO_2 and CO_2, uncertainties for the year 1985 and 1955 were assumed to be 10% and 20% larger than those in 2015, respectively. For SO_2 and CO_2, settings of uncertainties were not changed between 2015, 1985, and 1955.

For SO_2, in addition to uncertainties for ratios of sulfur emitted as SO_2, those in sulfur contents in fuels need to be taken into considered. Uncertainties of the sulfur contents in fuels including effects of regulation (i.e. usage of low sulfur fuels) were assumed based on data sources as follows:

- China:
  - 15%/15%, 15%/20%, and 20%/25% for coal, light and diesel oil, and heavy oil in 2015/1985, respectively.
  - 30% for all fossil fuels in 1955.
- India:
  - 20%/15%, 30%/25%, 20%/20%, and 25%/25% for hard coal, brown coal, light and diesel oil, and heavy oil in 2015/1985, respectively.
  - 30% for all fossil fuels in 1955.
- Japan:
  - 10%/10%, 20%/20%, 15%/15%, and 20%/20% for hard coal, brown coal, light and diesel oil, and heavy oil in 2015/1985, respectively.
  - 30% for all fossil fuels in 1955.

- Republic of Korea and Taiwan
  - 20%/15%, 30%/25%, 20%/15%, and 25%/20% for hard coal, brown coal, light and diesel oil, and heavy oil in 2015/1985, respectively.
  - 30% for all fossil fuels in 1955.
- Others:
  - 20%/15%, 30%/25%, 20%/20%, and 25%/25% for coal, light and diesel oil, and heavy oil in 2015/1985.
  - 30% for all fossil fuels in 1955.

Note that uncertainties in 1985 were smaller than other years for some countries and regions because the data were based on detailed surveys of Kato et al. (1991).

Table 10.2. Uncertainties [%] of emission factors of fuel combustion in 2015 assumed in REASv3. Values in the table (W/X/Y/Z) are for power plants/iron and steel, and cement industries/other industries/residential and other domestic sectors, respectively.

	Coal fuels	Gas and oil fuels	Primary biofuels	Others
SO_2	±15/±20/±25/±30	±10/±10/±10/±10	±75/±75/±100/±125	±75/±75/±100/±125
NO_x	±30[a]/±40/±50/±60	±30/±40/±50/±60	±75/±75/±100/±125	±75/±75/±100/±125
CO	±50/±60/±70/±80	±40/±50/±60/±75	±75/±75/±100/±125	±75/±75/±100/±125
NMVOC	±50/±60/±70/±80	±40/±50/±60/±75	±75/±75/±100/±125	±75/±75/±100/±125
NH_3	±100/±100/±100/±100	±100/±100/±100/±100	±150/±150/±150/±150	±150/±150/±150/±150
CO_2	±15/±15/±15/±15	±10/±10/±10/±10	±50/±50/±50/±50	±25/±25/±25/±25
PM_{10}	±50[a]/±60/±70/±100	±40/±50/±60/±75	±100/±100/±125/±150	±100/±100/±125/±150
$PM_{2.5}$	±50[a]/±60/±70/±100	±40/±50/±60/±75	±100/±100/±125/±150	±100/±100/±125/±150
BC	±50[a]/±60/±70/±100	±40/±50/±60/±75	±100/±100/±125/±150	±100/±100/±125/±150
OC	±50[a]/±60/±70/±100	±40/±50/±60/±75	±100/±100/±125/±150	±100/±100/±125/±150

a. 10% smaller values were adopted for China.

Removal efficiencies

For removal efficiencies, the same as for emission factors, it is necessary to consider uncertainties in the data themselves and those caused by selection of data. In addition, uncertainties in settings of emission controls such as timing of introduction and penetration rates of abatement equipment need to be considered where there is no specific way to quantify the uncertainties neither. In REASv3, uncertainties of removal efficiencies were roughly estimated as summarized in Table 10.3 based on the following assumption:

- Uncertainties of removal efficiencies are assumed to be smaller if settings were generally based on local information and literatures. For example, those for Japan were generally smaller than other countries because their settings were based on domestic information such as MRI (2015) and MOEJ (2000).

- For emission sources where no emission controls were considered, uncertainties of corresponding removal efficiencies were assumed to be zero which means that uncertainties caused by neglecting emission controls were not considered. For example, uncertainties of removal efficiencies were assumed to be zero for all emission sources, species, and countries and regions in 1955.
- For emission sources where introduction rates of abatement equipment were small, uncertainties caused by settings of emission controls were assumed to be small.

Table 10.3. Settings of uncertainties of removal efficiencies adopted in REASv3. Note that uncertainties of removal efficiencies for sources without description here were assumed to be zero.

Countries and regions	Settings of uncertainties of removal efficiencies
China	● SO_2: Uncertainties of removal efficiencies were only estimated in 2015. The values for power plants were assumed to be 20% and those for industry sectors were 10% higher than those for power plants (namely 30%). ● NO_x: The same as for SO_2, uncertainties for power plants were only estimated in 2015 and the values were assumed to be 25% (5% higher than those for SO_2). The same values were adopted for cement industries. ● PM species: Uncertainties for power plants in 2015 and 1985 were assumed to be 15% and 20%, respectively. For industry sectors, 5% higher values were adopted (namely, 20% and 25% for 2015 and 1985, respectively).
India	● SO_2: Only for power plants as point sources with FGD, 20% were adopted for their uncertainties of removal efficiencies. ● PM species: Due to lack of information, 10% higher values were adopted for power plants and industry sectors as follows: In 2015 and 1985, 25% and 30% for power plants and 30% and 35% for industry sectors, respectively.
Japan	● SO_2: Uncertainties of removal efficiencies in both 2015 and 1985 were assumed to be 20% for power plants. For industry sectors, 5% higher values than those for power plants were adopted (namely 25%). ● NO_x: The same settings for SO_2 were used for both power plants and industry sectors in 2015 and 1985. ● PM species: For power plants, uncertainties of removal efficiencies in both 2015 and 1985 were assumed to be 10%. For industry sectors, higher values

	than those of power plants were assumed as follows: 15% and 20% in 2015 and 1985, respectively.
Korea and Taiwan	● SO_2: Uncertainties of removal efficiencies for power plants and industry sectors in 2015 were assumed to be 25% and 30%, respectively. In 1985, assuming relatively lower introduction rates of abatement equipment, 10% lower values than those for 2015 were adopted (15% and 20% for power plants and industry sectors, respectively). ● NO_x: Uncertainties were only estimated for power plants in 2015 and 5% higher values than those for SO_2 were assumed (namely 25%). ● PM species: The same settings for China were adopted.
Thailand	● SO_2: Uncertainties of removal efficiencies were only estimated for power plants in 2015. The values were assumed to be 25%. ● PM species: The same settings for India ware adopted.
Other countries and regions	● SO_2: Only for power plants as point sources with FGD, 20% were adopted for their uncertainties of removal efficiencies. ● PM species: Uncertainties of removal efficiencies were assumed for power plants and industry sectors only for the year 2015. 5% higher values than those for Thailand were adopted (namely, 30% for power plants and 35% for industry sectors.)

S10.2.2 Stationary non-combustion sources: Industrial production and other transformation

Activity data

Activity data for emissions from industrial production and other transformation were such as production amounts of industrial products and input amounts of materials. The same as for the case of fuel consumption data as described in Sect. 10.2.1, uncertainties of the activity data depend on reliability and availability of their data sources and for settings of the uncertainties, following assumptions were taken into considered:

- In the same international statistics, uncertainties of data are lower for Japan, Republic of Kora, and Taiwan in 2015 and Japan in 1985 compare to other countries and regions.
- Uncertainties of activity data estimated by such as interpolation or extrapolation were larger than those directly taken from the statistics and literatures.
- Uncertainties of major industrial products such as metals and cement are smaller than minor ones such as lime and carbon black even tough data were taken from the same international statistics.

Uncertainties of activity data for emissions from industrial products and other transformation adopted in REASv3 are summarize in Table 10.4.

Table 10.4. Settings of uncertainties of activity data for industrial production and other transformation adopted in REASv3. Note that settings for non-combustion sources of NMVOC and NH_3 were described in Sects. S10.2.3 and S10.2.6, respectively.

Sub-sector categories	Settings of uncertainties of activity data
Iron and steel production	● If data were directly taken from data sources, values of uncertainties were assumed as follows: ➢ 5% for Japan, Republic of Korea and Taiwan in 2015 and Japan in 1985 ➢ 10% for other countries and regions in 2015 and 1985 ● For all countries and regions, if data were estimated by interpolation or extrapolation, the uncertainties were assumed to be 15% in 2015 and 1985. ● For the year 1955, the uncertainties were assumed to be 20% for all countries and regions.
Non-ferrous metal production	● The same settings for iron and steel production were adopted.
Cement production	● The same settings for iron and steel production were adopted.
Lime production	● The same criteria for iron and steel production were assumed for differences of settings among countries and regions and years. For values of the uncertainties, 5% higher values than those for iron and steel production were adopted.
Brick production	● Due to lack of available data and information, high uncertainties were assumed as follows: 40%, 50%, and 75% for all countries and regions in 2015, 1985, and 1955, respectively.
Sulphuric acid production	● The same settings for iron and steel production were adopted.
Carbon black production	● The same settings for lime production were adopted.
Coke production	● The same settings for coal consumption in iron and industry in Table 10.1
Petroleum refineries	● The same settings for oil consumption in other industries in Table 10.1.

Emission factors and settings of emission controls

Causes of uncertainties of emission factors and settings of emission controls for industrial production and other transformation sectors were basically the same as for those for fuel combustion. Table 10.5 summarizes the settings of uncertainties and related assumptions for emission factors and emission controls adopted in REASv3. Note that except for SO_2 and CO_2, uncertainties of emission factors for the year 1985 and 1955 were assumed to be 10% and 20% larger than those in 2015, respectively, the same as the case for fuel combustion sources. Settings of uncertainties for SO_2 for non-ferrous metal production and CO_2 were not changed between 2015, 1985, and 1955.

Table 10.5. Settings of uncertainties of emission factors in 2015 and emission controls for industrial production and other transformation adopted in REASv3. Values were commonly used for all countries and regions unless otherwise indicated.

Sub-sector categories	Settings of uncertainties of emission factors and emission controls
Iron and steel production	● Emission factors: 40% for CO, 15% for CO_2, and 60% for PM species. ● Emission controls: Settings for fuel combustion in iron and steel industry were adopted. For China, the uncertainties in 2015 were assumed to be 10% because the settings were based on local information of Wu et al. (2017).
Non-ferrous metal production	● Emission factors: 20% for SO_2 and 60% for PM species. ● Emission controls: Settings for fuel combustion in other industries were adopted for PM species. For SO_2, considering uncertainties in collection amounts for sulphuric acid production, high uncertainties of 30% were assumed for the years 2015 and 1985.
Cement production	● Emission factors: ➢ Japan: As described in Sect. S3.2 and S4.1.3, NO_x, CO, and PM species from fuel consumption in cement kilns were estimated using local information of cement production in each kiln type. Therefore, values of uncertainties for NO_x, CO, and PM species were assumed to be 20% lower than those for default settings in Table 10.2. ➢ PM species: 60% was adopted except for China and Japan. For China, because settings were based on local information of Lei et al. (2011) and Wu et al. (2017), 20% lower values were adopted (namely 40%). ➢ CO_2: Considering uncertainties in clinker to cement ratios, relatively high uncertainties (20%) was adopted. ● Emission controls: Settings for fuel combustion in cement industry were

	adopted. For China, the uncertainties in 2015 were assumed to be 10% because the settings were based on local information of Hua et al. (2016).
Lime production	● Emission factors: 15% and 60% were adopted for CO_2 and PM species, respectively. ● Emission controls: The same settings for fuel combustion in other industries were adopted.
Brick production	● Emission factors: ➢ CO: 60% were adopted for countries and regions where emissions were estimated using amounts of brick production. ➢ PM species: 60% were adopted. ● Emission controls: The same settings for fuel combustion in other industries were adopted.
Sulphuric acid production	● Emission factors: 20% were adopted for SO_2. ● Emission controls: The same settings for fuel combustion in other industries were adopted.
Carbon black production	● Emission factors: 60% was adopted for PM species. ● Emission controls: The same settings for fuel combustion in other industries were adopted.
Coke production	● Emission factors: 40% for CO, 15% for CO_2, and 60% for PM species. ● Emission controls: The same settings for fuel combustion in iron and steel industry were adopted.
Petroleum refineries	● Emission factors: 20% for SO_2 and 60% for PM species. ● Emission controls: The same settings for fuel combustion in other industries were adopted.

S10.2.3 Non-combustion sources of NMVOC

Basically, causes of uncertainties of activity data and emission factors for non-combustion sources of NMVOC are the same as for those for combustion sources, industrial production and other transformation described in Sects. S10.2.1 and S10.2.2. Due to lack of available data and information, the uncertainties for non-combustion sources of NMVOC were generally assumed to be larger than other sources as described in this sub-section. Note that emission controls of NMVOC were not considered in REASv3 and influences of their uncertainties were neglected. In addition, note that uncertainties of non-combustion sources of NMVOC emissions in Japan and Republic of Korea which depended on other inventories were not estimated.

Extraction processes

Activity data for extraction processes were taken from energy statistics. The settings of the uncertainties were assumed to be the same as for those of gas and oil fuels in power plants, iron and steel, and cement industries in Table 10.1. For emission factors, the uncertainties were assumed to be 70% except for petroleum refinery where lower uncertainty of 50% was assumed. The settings for emission factors were commonly used for all countries and regions for all target years.

Solvent use

As described in Sect. S5.1, activity data of solvent use in REASv3 were based on limited available statistics and literatures and if appropriate data were not available, activity data of REASv2 during 2000-2008 were used as default. In addition, missing data were often estimated by extrapolation of GDP. Considering the above limitations, relatively high uncertainties were assumed for activity data as follows:

- If activity data were directly based on available statistics and literatures, the uncertainties were assumed to be 20%.
- If activity data were derived by interpolated or extrapolated from the available statistics and literatures, the uncertainties of them were assumed to be 30%, 40% and 50% in 2015, 1985, and 1955, respectively.
- If activity data were based on default, the uncertainties of them were assumed to be 40% for data of 2015 and 50% for those of 1985 and 1955.
- For vehicle treatment, activity data are number of registered vehicles and their uncertainties were described in Sect. S10.2.4.
- For domestic use of solvents, activity data are number of urban and rural populations. Considering uncertainties in urban and rural population ratios, the uncertainties were assumed to be 10% higher than those of total population number described in Sect. S10.2.6.
- For paint use for automobile manufacturing, activity data were production number of vehicles. For activity data directly taken from statistics in 2015 and 1985, the uncertainties were assumed to be 10% and 20%, respectively. If activity data were derived by interpolated or extrapolated from the available statistics and literatures, the uncertainties of them were assumed to be 20%, 30% and 50% in 2015, 1985, and 1955, respectively.

For emission factors, uncertainties of 70% were commonly used for all sub-categories, countries and regions, and target years.

Chemical industry

Uncertainties of activity data for chemical industry were assumed basically same procedures for those of solvent use as follows:

- If activity data were based on available statistics and literatures, the uncertainties of them were assumed to be 15%, 25%, and 40% for the years of 2015, 1985, and 1955, respectively. Considering the availability of statistics and literatures, values of uncertainties were assumed to be lower than those of solvent use.
- If activity data were based on default, the uncertainties of them were assumed to be 40% for data of 2015 and 50% for those of 1985 and 1955.
- For carbon black production, see Table 10.4 for settings of the uncertainties.

For emission factors, uncertainties of 70% were commonly used for all sub-categories, countries and regions, and target years. For carbon black production, lower uncertainties of 50% was assumed.

Other industry

Activity data of other industry are production amounts of bread, beer, coke, asphalt, crude steel, hot rolled steel, and pulp and paper. Uncertainties of them were assumed as follows:

- For bread, beer, and asphalt, pulp and paper production, the uncertainties of activity data were assumed based on the same procedures for chemical industry.
- For coke, crude steel, and hot rolled steel production, see Table 10.4 for settings of the uncertainties.

For emission factors, uncertainties of 70% were commonly used for all sub-categories, countries and regions, and target years except for coke, crude steel and hot rolled steel production where lower uncertainties of 50% was assumed.

Waste disposal

Activity data of waste disposal sector are amounts of municipal wastes and their uncertainties were assumed based on available data sources as follows:

- If data were directly taken from national or international statistics and literatures, the uncertainties were assumed to be 30% in 2015 and 40% in 1985.
- If data were estimated by interpolation or extrapolation, the uncertainties were assumed to be 40% for 2015 and 50% for 1985 and 75% for 1955.

For emission factors, uncertainties of 80% were commonly used for all sub-categories, countries and regions, and target years.

S10.2.4 Road transport

Activity data

Activity data of emissions from road transport were number of vehicles and annual distance travelled for NO_x, CO, NMVOC, NH_3, and PM species. Uncertainties of number of vehicles which were also used for estimation of NMVOC evaporative emissions were assumed based on data sources as follows:

- For data based on detailed national statistics, the uncertainties were assumed to be 10% for passenger cars, 15% for buses and motor cycles, and 20% for trucks. If data were interpolated or extrapolated based on the detailed national statistics, uncertainties in 1985 (1955) were assumed to be 15% (30%) for passenger cars, 20% (40%) for buses and motor cycles, and 25% (40%) for trucks.
- For data based on IRF (1976-2018), the uncertainties were assumed to be 20% for passenger cars, 25% for buses and motor cycles, and 30% for trucks. If data were interpolated or extrapolated based on international statistics, uncertainties in 1985 (1955) were assumed to be 25% (40%) for passenger cars, 30% (50%) for buses and motor cycles, and 35% (50%) for trucks.
- For data based on IRF (1976-2018) and national information, the uncertainties were assumed to be 15% for passenger cars, 20% for buses and motor cycles, and 25% for trucks. If data were interpolated or extrapolated based on national or international statistics, uncertainties in 1985 (1955) were assumed to be 20% (30%) for passenger cars, 25% (40%) for buses and motor cycles, and 30% (40%) for trucks.

Similarly, uncertainties of annual distance travelled were assumed based on data sources as follows:

- For data based on national information, the uncertainties were assumed to be 15% for passenger cars and motor cycles and 20% for buses and trucks.
- For data based on national data in Clean Air Asia (2012), the uncertainties were assumed to be 20% for passenger cars and motor cycles and 30% for buses and trucks.
- For other data such as average of Asian data in Clean Air Asia (2012), the uncertainties were assumed to be 30% for passenger cars and motor cycles and 40% for buses and trucks.
- For Japan where annual mileage data were directly obtained from literatures and statistics, uncertainties of the annual mileages were assumed to be 10%/15%/25% for cars, buses, and trucks and 15%/20%/30% for motorcycles and special purpose vehicles in 2015/1985/1955.

For SO_2 and CO_2, emissions from road transport were estimated based on fuel consumption as described in Sect. S6.2.3. The uncertainties of activity data were assumed to be the same values for oil consumption in power plants, iron and steel, and cement industries in Table 10.1.

Emission factors

For emission factors of exhaust emissions from road vehicles, uncertainties were estimated as summarized in Table 10.6 based on the following assumptions:

- The lowest uncertainties were assumed for Japan where detailed local information was available for estimation of emission factors.
- Uncertainties of emission factors for China and India referring studies of national emission inventories were also smaller than those of other countries and regions.
- Uncertainties of emission factors were assumed to be smaller for NO_x and larger for PM species and those of CO and NMVOC were between them. Due to lack of information, uncertainties for NH_3 were assumed to be the largest.
- The same settings were adopted for all vehicle types except for rural vehicles and special purpose vehicles where 10% higher uncertainties were considered.

Table 10.6. Uncertainties [%] of emission factors of exhaust emissions in 2015/1985/1955 for NO_x, CO, NMVOC, NH_3, and PM species. All data were commonly adopted for all vehicle types except for rural vehicles and special purpose vehicles where 10% higher uncertainties were used.

	China and India	Japan	Others
NO_x	±25/±35/±45	±20/±30/±40	±30/±40/±50
CO	±35/±45/±55	±30/±40/±50	±40/±50/±60
NMVOC	±35/±45/±55	±30/±40/±50	±40/±50/±60
NH_3	±100/±100/±100	±75/±75/±75	±100/±100/±100
PM species	±45/±55/±65	±40/±50/±60	±50/±60/±70

For CO_2 and SO_2, the uncertainties of emission factors were assumed to be 10% which are the same settings for stationary combustion. In addition, for SO_2, uncertainties of sulfur contents in gasoline and diesel oil were also taken from settings for stationary combustion provided in Sect. S10.2.1.

For estimation of NMVOC evaporative emissions, simple methodology of EEA (2016) were adopted as described in Sect. S6.3. Therefore, high uncertainties of 100% were assumed for the emission factors.

S10.2.5 Other transport

As described in Sect. S7, other transport sector includes railway, pipeline transport and non-specified sectors defined in the IEAWEB. Settings of the uncertainties of their activity data and emission factors were the same as for those of fuel combustion in other industries.

S10.2.6 Non-combustion sources of NH_3

In REASv3, for non-combustion sources of NH_3, uncertainties of emissions from fertilizer production, human, and latrines were estimated. The uncertainties of activity data and emission factors were estimated by the same procedures for those of NMVOC. Settings and assumptions for the uncertainties are described in this sub-section. Note that emissions from manure management and fertilizer application were not originally estimated and thus, their uncertainties were not estimated.

Fertilizer production

As described in Sect. S8.3, activity data of NH_3 emissions from fertilizer production considered in REASv3 are ammonia, ammonium nitrate, and urea and the uncertainties were assumed as follows:

- Ammonia: In 2015, data were taken from Minerals Yearbook (USGS) and their uncertainties were assumed to be 15%. For the year 1985, the uncertainties were assumed to be 20% for China and Japan where national trend factors were available and those for other countries were assumed to be 30%. In 1955, the uncertainties were assumed to be 40% for all countries and regions.
- Ammonium nitrate: For Japan where national statistics were available, the same settings for ammonia were adopted. For other countries, higher uncertainties were assumed as 30%, 40%, and 50%, respectively.
- Urea: For China and Japan where national statistics were available, the same settings for ammonia were adopted. For other countries, the settings for ammonium nitrate were used.

For emission factors, uncertainties of 50% were commonly used for all sub-categories, countries and regions, and target years.

Human perspiration and respiration

Activity data of NH_3 emissions from human perspiration and respiration is number of total population and the uncertainties were assumed as follows:

- Similar to the case for IEAWEB, low uncertainties of 2% were assumed for Japan in 2015 and 1985 and Republic of Korea and Taiwan in 2015.
- For others, uncertainties were assumed to be 5% in 2015 and 1985 and 10% in 1955.

For emission factors, uncertainties of 50% were commonly used for all sub-categories, countries and regions, and target years.

Latrines

Activity data of NH_3 emissions from latrines are number of population in no sewage service areas. Available data were very limited except for Japan and the uncertainties were assumed as follows:

- Uncertainties for Japan were assumed to be 10% in 2015 and 1985 and 30% in 1955.
- For other countries and regions, uncertainties were assumed to be 30%, 40%, and 50% in 2015, 1985, and 1955, respectively.

For emission factors, uncertainties of 70% were commonly used for all sub-categories, countries and regions, and target years.

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Supplement of

Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3

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Supplementary figures

Figures S1, S3, S5, S7, S9, and S11 show emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors during 1950-2015 in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA), respectively. See Fig. 1 for the definitions of SEA, OEA, and OSA. (Sectors for (a)-(h): PP = Power plants, IND = Industry, ROAD = Road transport, OTRA = Other transport, RESI = Residential, and ODOM = Other domestic; Sectors for (i): CMB = Combustion, ROAD = Road transport (including both tail pipe and evaporative emissions), INDPRC = Industrial processes, EXT = Extraction processes, PAINT = Paint use, SLV = Solvent use, and WST = Waste treatment; Sectors for (j): CMB = Combustion, MM = Manure management, FER = Fertilizer application, HUMAN = Human perspiration and respiration, LTRN = Latrines, and INDPRC = Industrial processes.)

Figures S2, S4, S6, S8, S10, and S12 provide emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type during 1950-2015 in China, India, Japan, SEA, OEA, and OSA, respectively. See Fig. 1 for the definitions of SEA, OEA, and OSA. Note that emissions from non-combustion sources are not included in (i) NMVOC and (j) NH3 to show contributions from fuel types clearly because majority of their emissions are from non-combustion sources. (Fuel types: COAL = Primary coal, DC = Secondary coal, NGAS = Natural gas, OGAS = Other gas fuels, LF = Light oil fuels, MD = Diesel oil, HF = Heavy oil fuels, BF = Biofuels, OTH = Other fuels, NCMB = Non-combustion sources, and CEMK = combustion emissions from cement kilns (only for Japan). Notes: For CO emissions from pig iron, crude steel, and sinter production for all countries, those from brick production except for China, Japan, Republic of Korea, and Taiwan, emissions of PM species from sinter and pig iron production for China, and those from brick production for all countries and regions estimated based on their production amounts, both combustion and non-combustion emissions are included in NCMB here. In Japan, emissions from cement production were estimated not by fuel consumption, but based on production amounts of cement in each kiln type. Therefore, contributions from total emissions from cement kiln combustion are included in CEMK.)

Figure S13 illustrates grid maps of annual emissions of CO_2 and PM_{10} in 1965 and 2015.

Figures S14 and S15 compare CO, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, and OC emissions in REASv3 with other published estimates for China and India, respectively. Note that IM means estimates by inverse modeling and [A][B][C] of Jiang et al. (2017) are estimates based on A: MOPITT Column, B: MOPITT Profile, and C: MOPITT Lower Profile, respectively.

Figures S16-S19 compare emissions of SO_2, NO_x, BC, CO, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, and OC in REASv3 with other published estimates for Japan, SEA, OEA, and OSA, respectively. See Fig. 1 for the definitions of SEA, OEA, and OSA. Note that IM means estimates by inverse modeling and [A][B][C] of Jiang et al. (2017) are estimates based on A: MOPITT Column, B: MOPITT Profile, and C: MOPITT Lower Profile, respectively.

Figures S20 compares Asia total emissions and relative ratios of those from China, India, Japan, SEA, OEA, and OSA for CO, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, and OC among REASv3, CEDS, and EDGARv4.3.2. See Fig. 1 for definitions of SEA, OEA, and OSA.

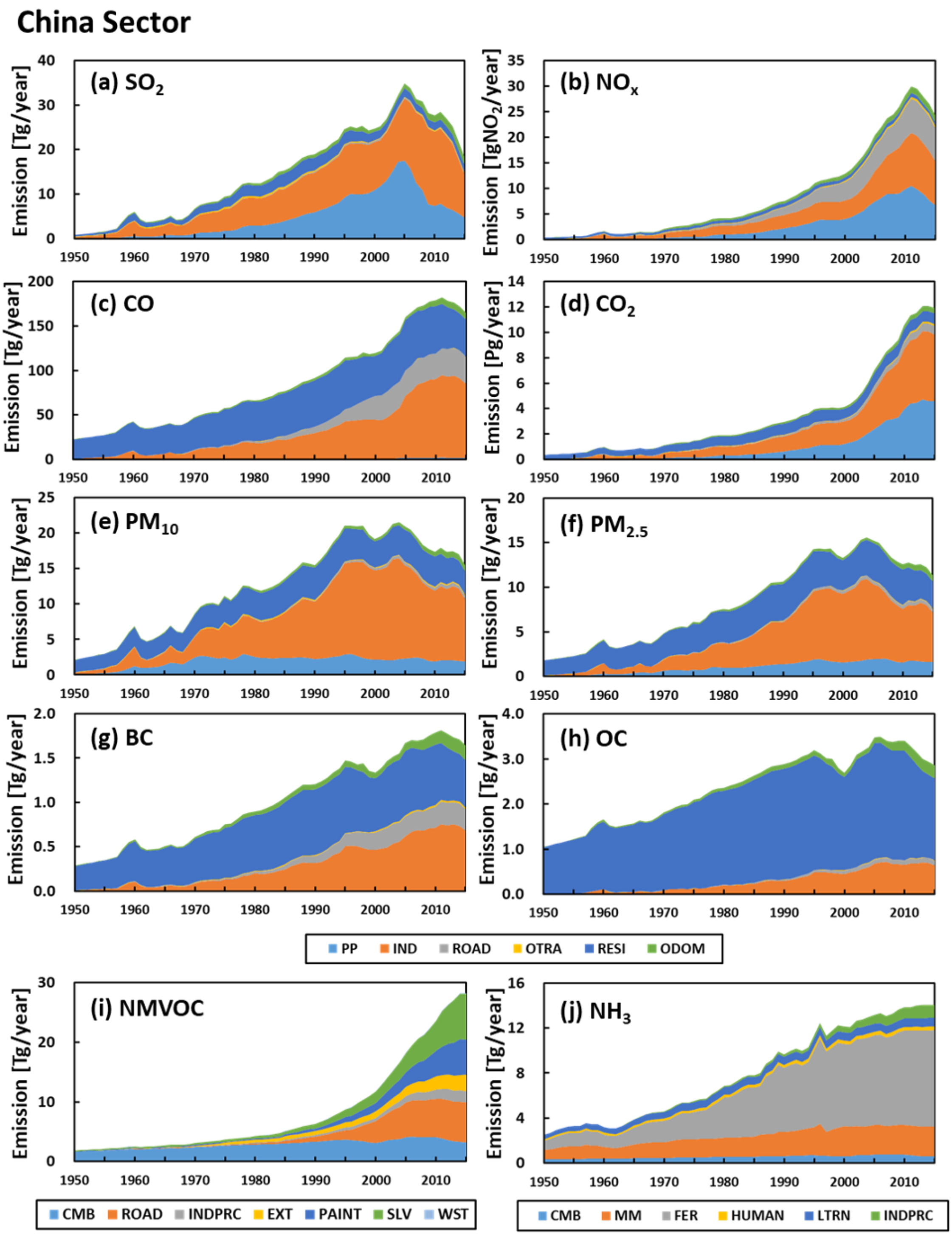

Figure S1. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors in China from 1950 to 2015.

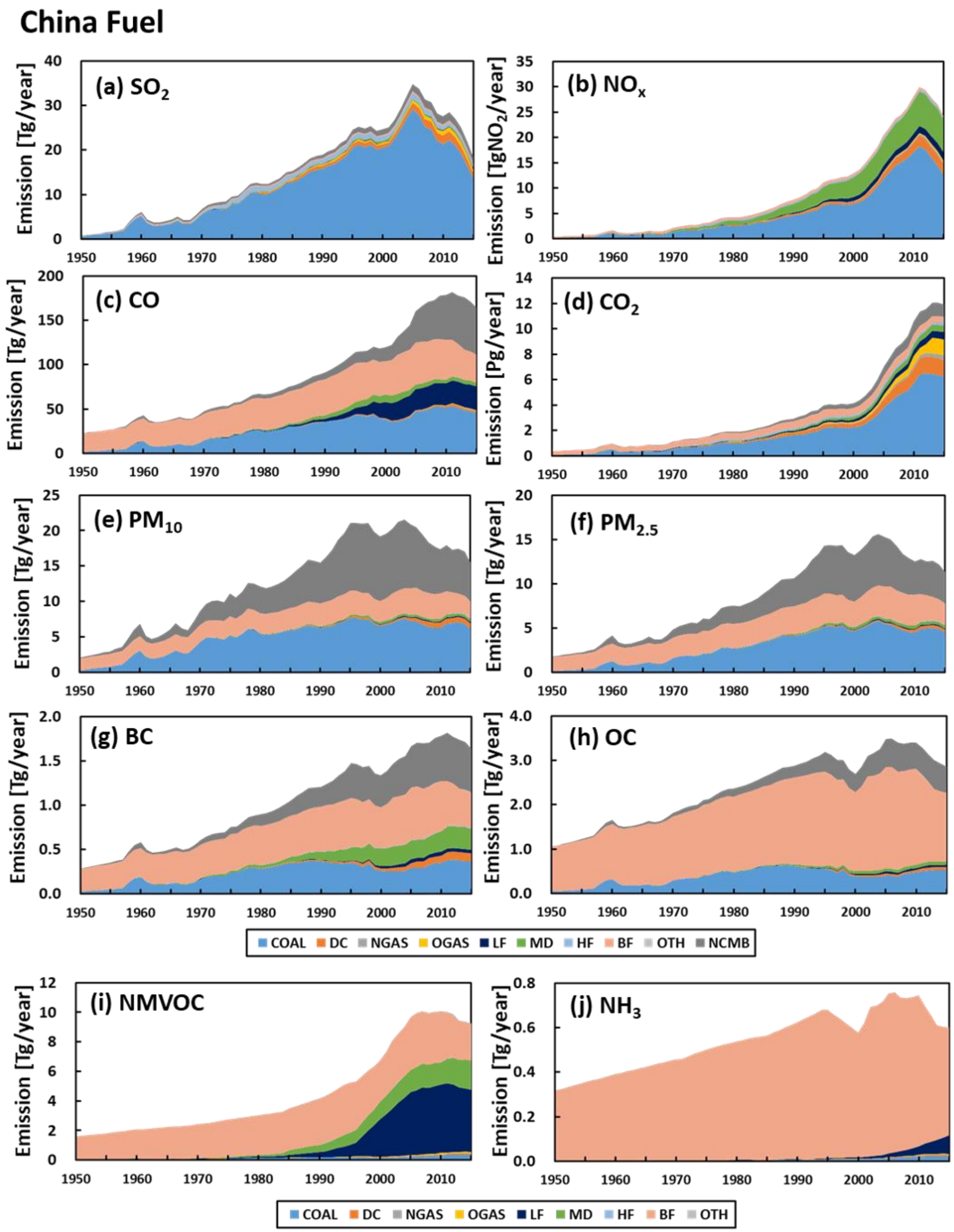

Figure S2. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type in China from 1950 to 2015.

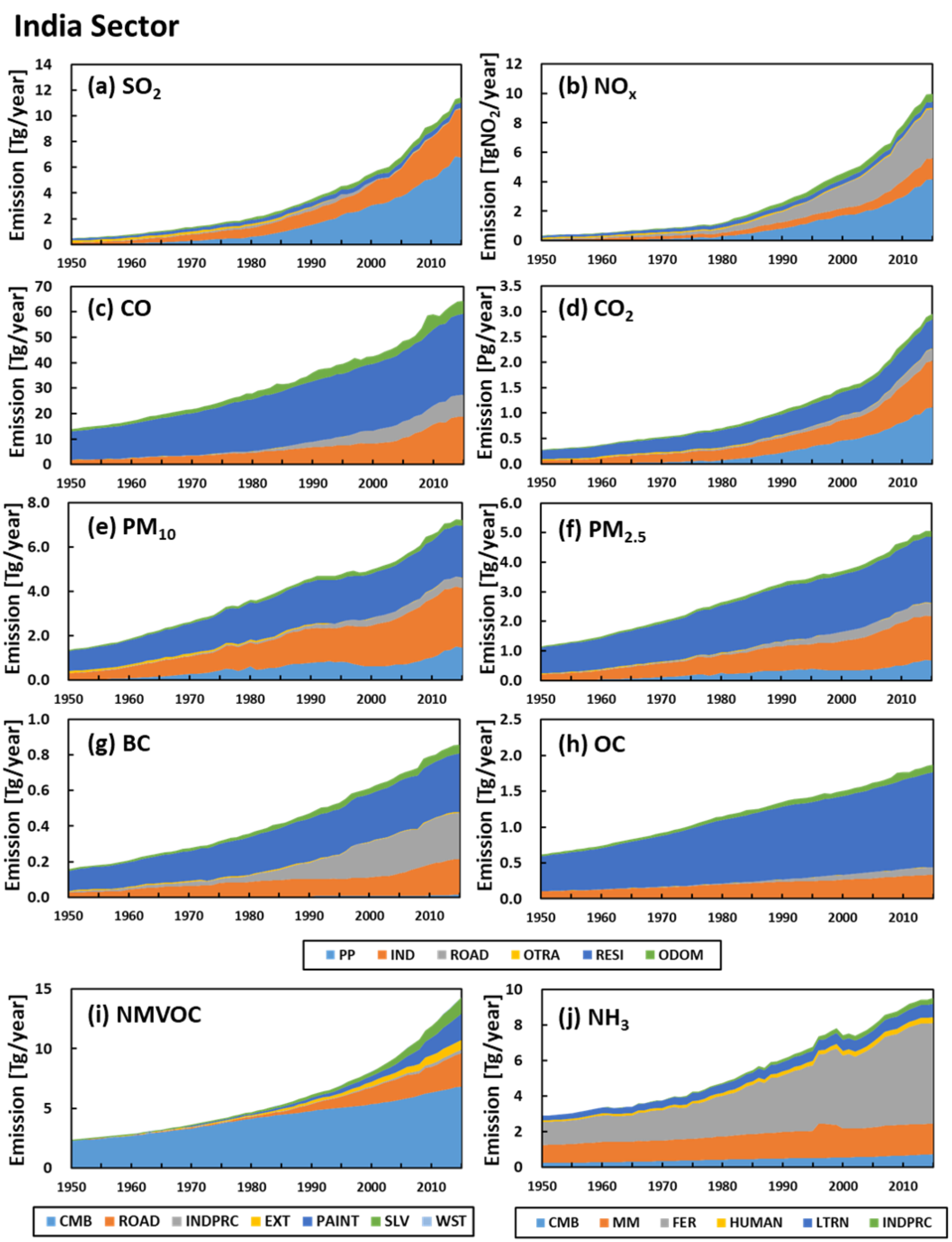

Figure S3. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors in India from 1950 to 2015.

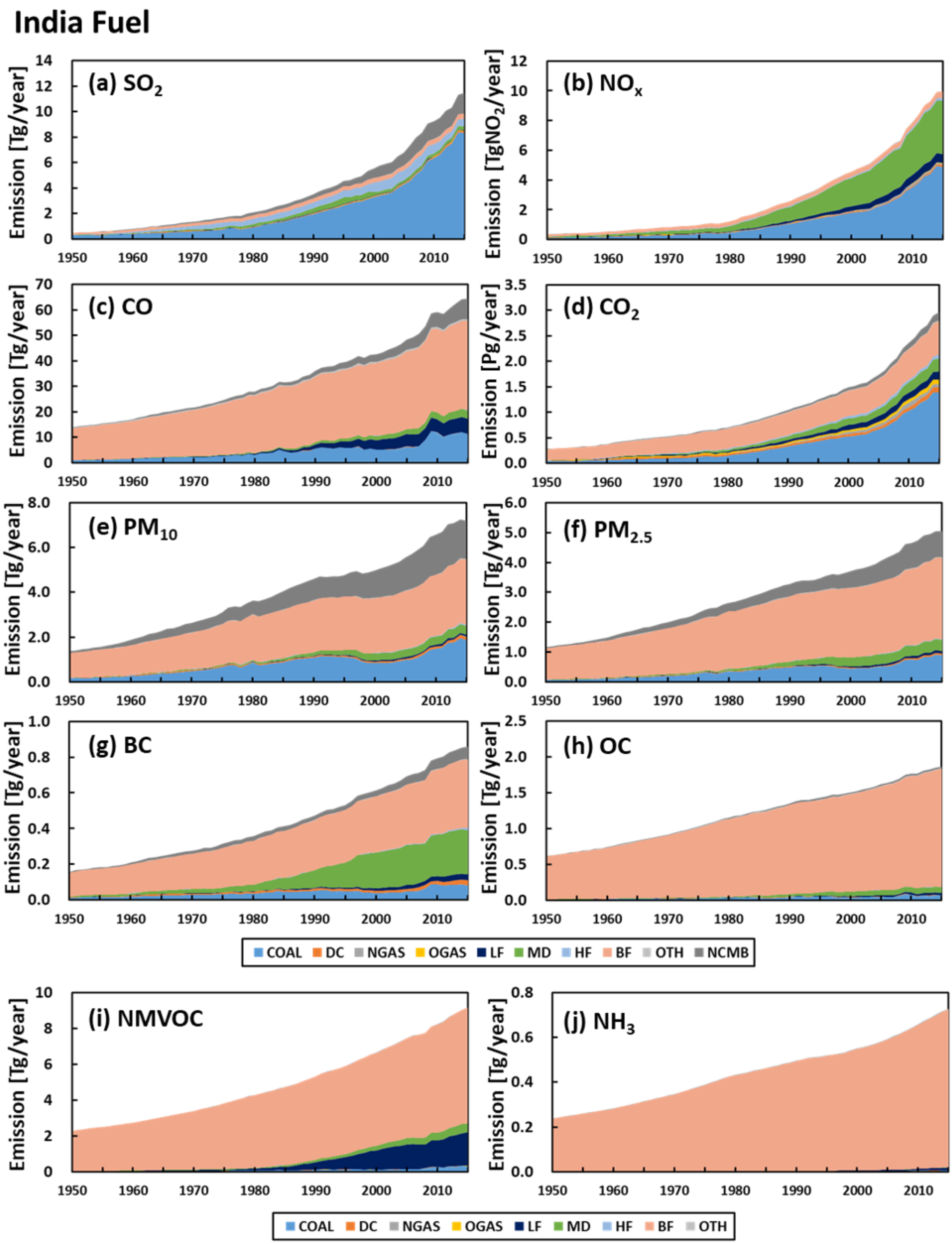

Figure S4. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type in India from 1950 to 2015.

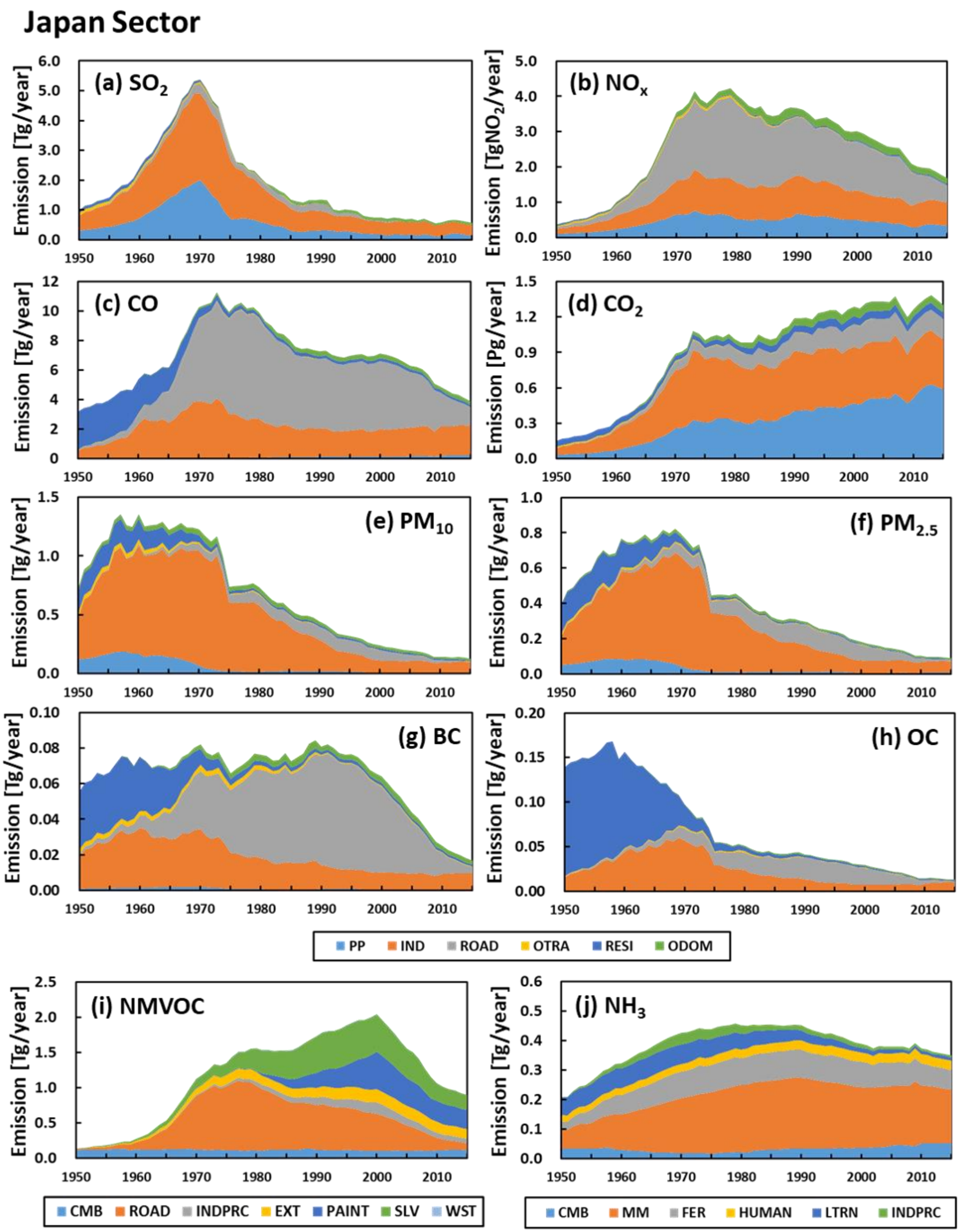

Figure S5. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors in Japan from 1950 to 2015.

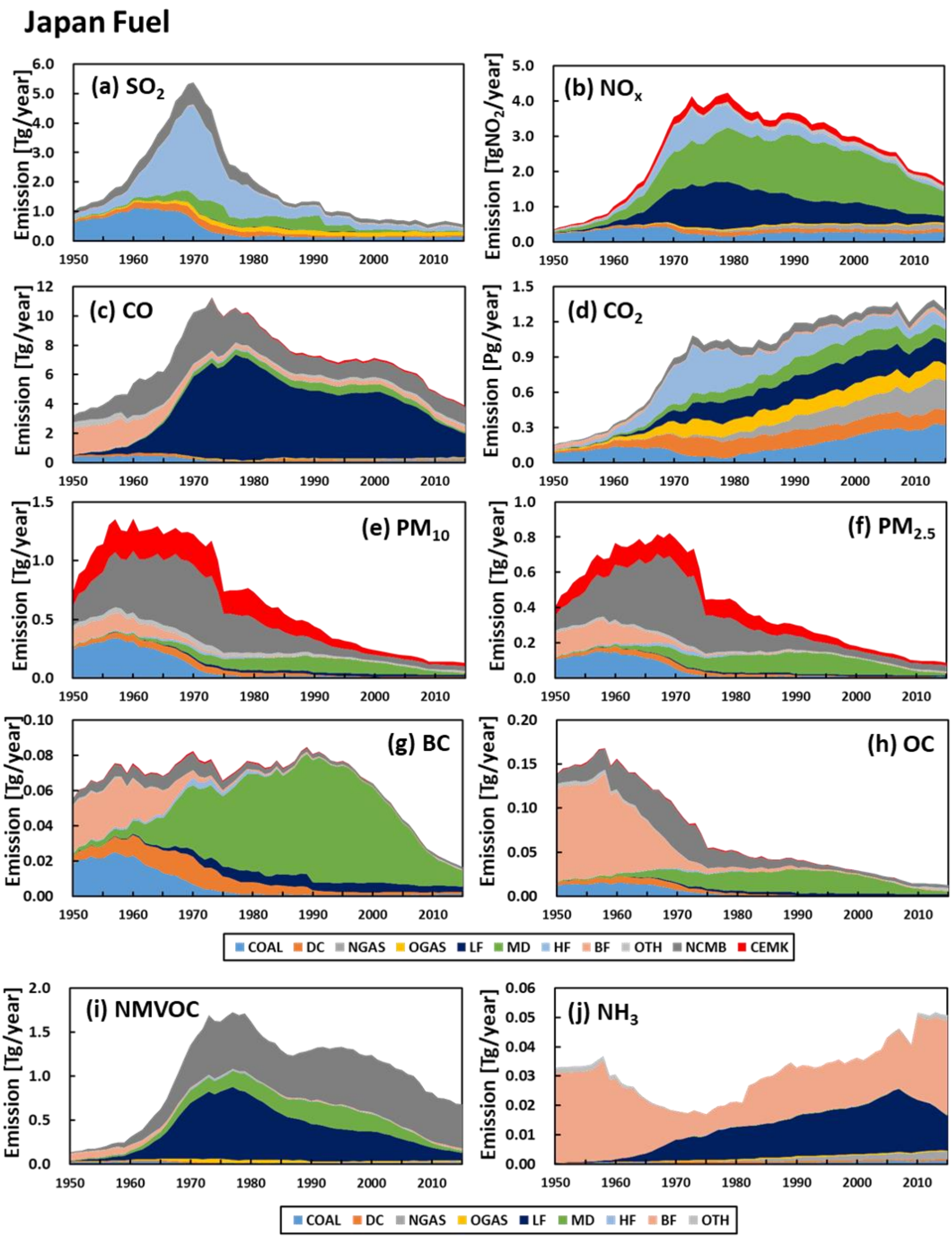

Figure S6. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type in Japan from 1950 to 2015.

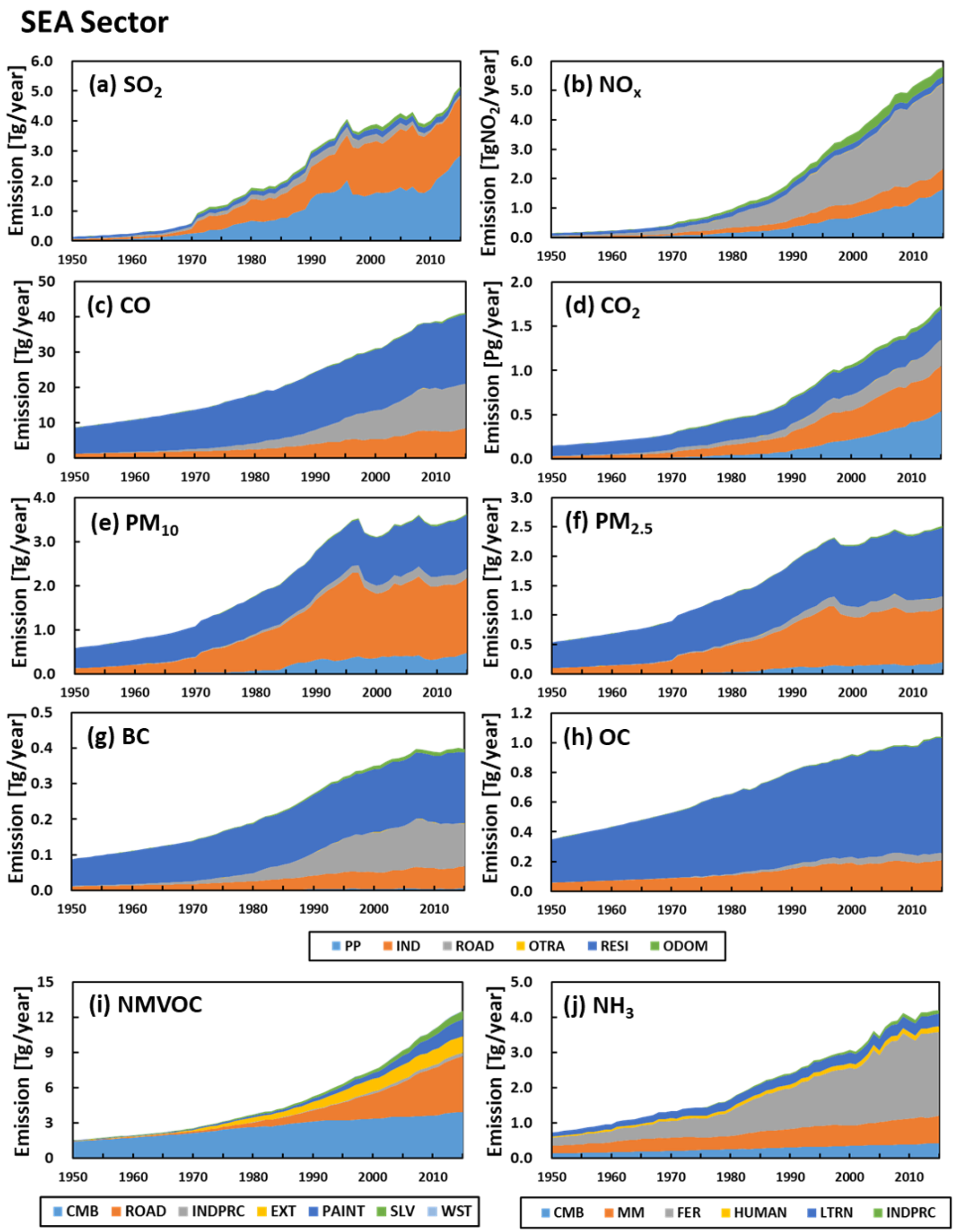

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Figure S7. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors in SEA from 1950 to 2015. See Fig. 1 for the definitions of SEA.

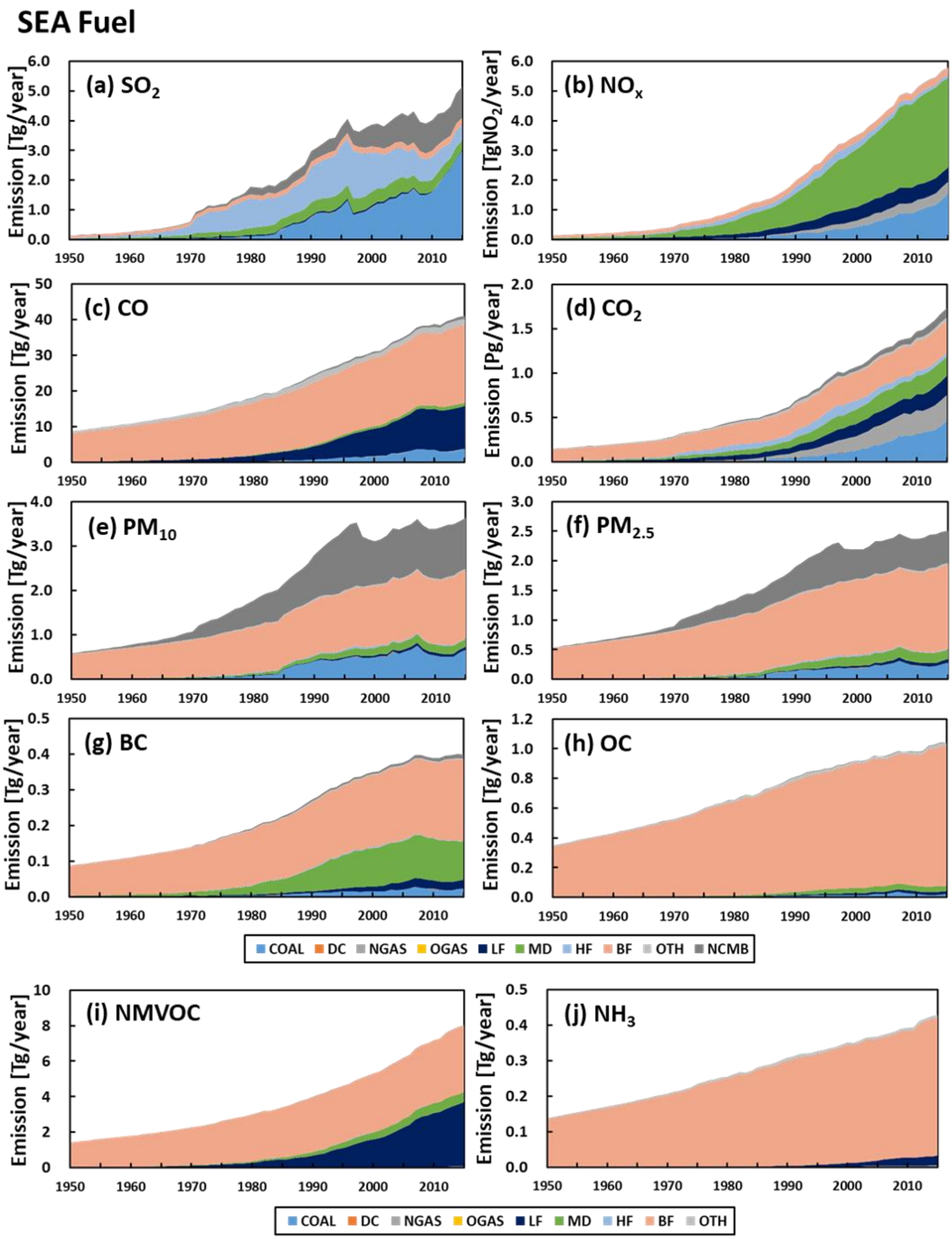


Figure S8. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type in SEA from 1950 to 2015. See Fig. 1 for the definitions of SEA.

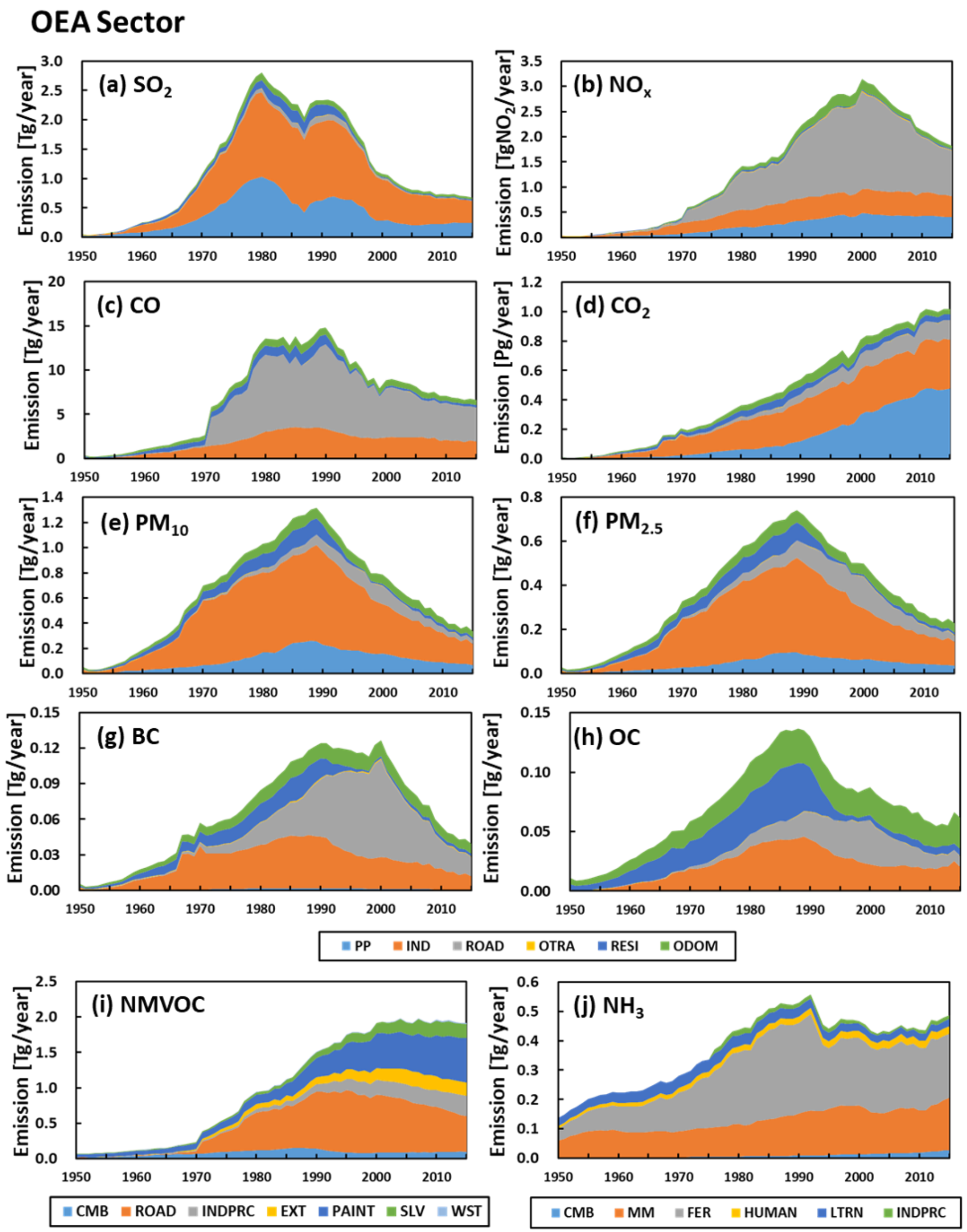


Figure S9. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors in OEA from 1950 to 2015. See Fig. 1 for the definitions of OEA.

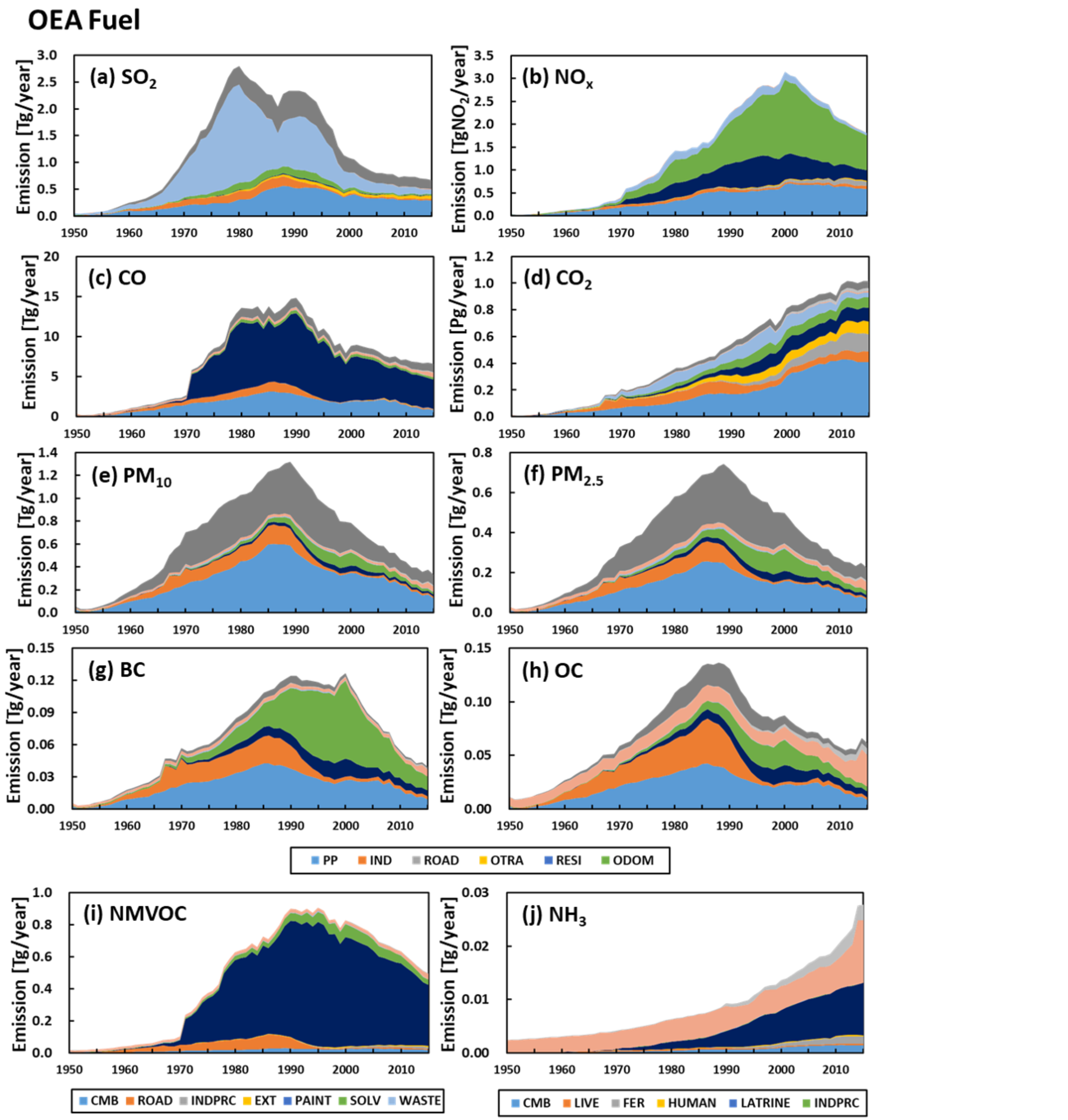


Figure S10. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type in OEA from 1950 to 2015. See Fig. 1 for the definitions of OEA.

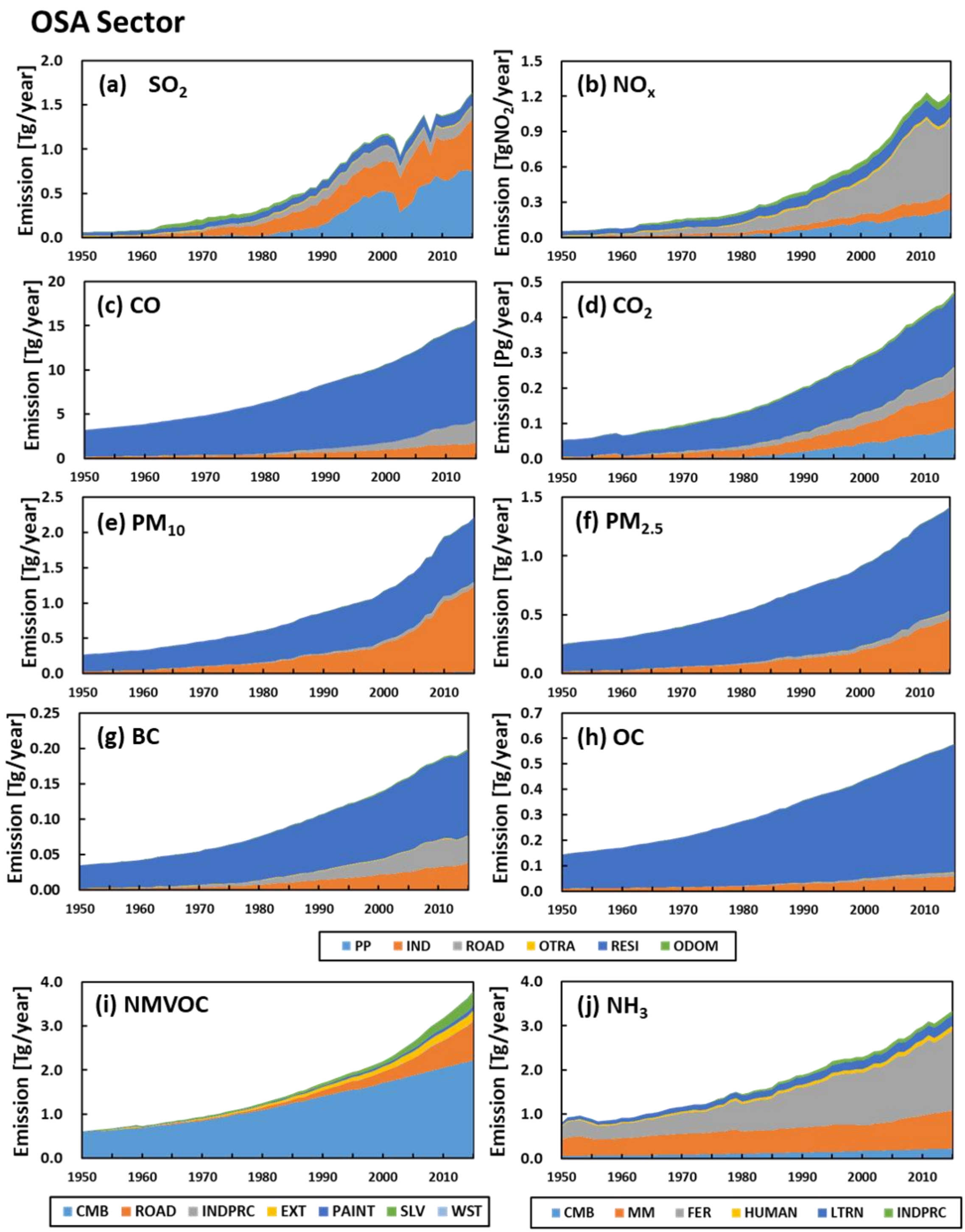


Figure S11. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from major sectors in OSA from 1950 to 2015. See Fig. 1 for the definitions of OSA.

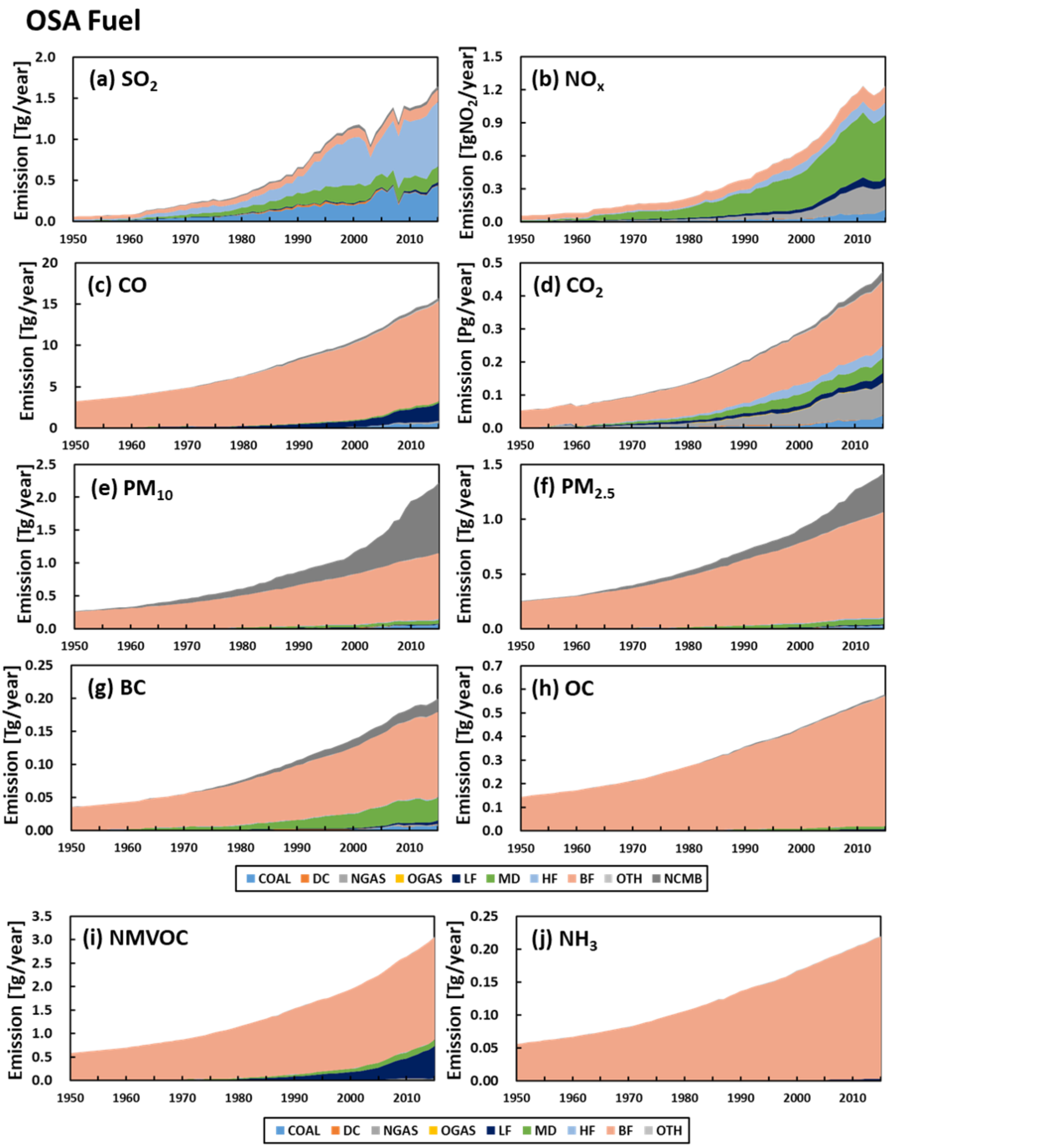


Figure S12. Emissions of (a) SO_2, (b) NO_x, (c) CO, (d) CO_2, (e) PM_{10}, (f) $PM_{2.5}$, (g) BC, (h) OC, (i) NMVOC, and (j) NH_3 from each fuel type in OSA from 1950 to 2015. See Fig. 1 for the definitions of OSA.

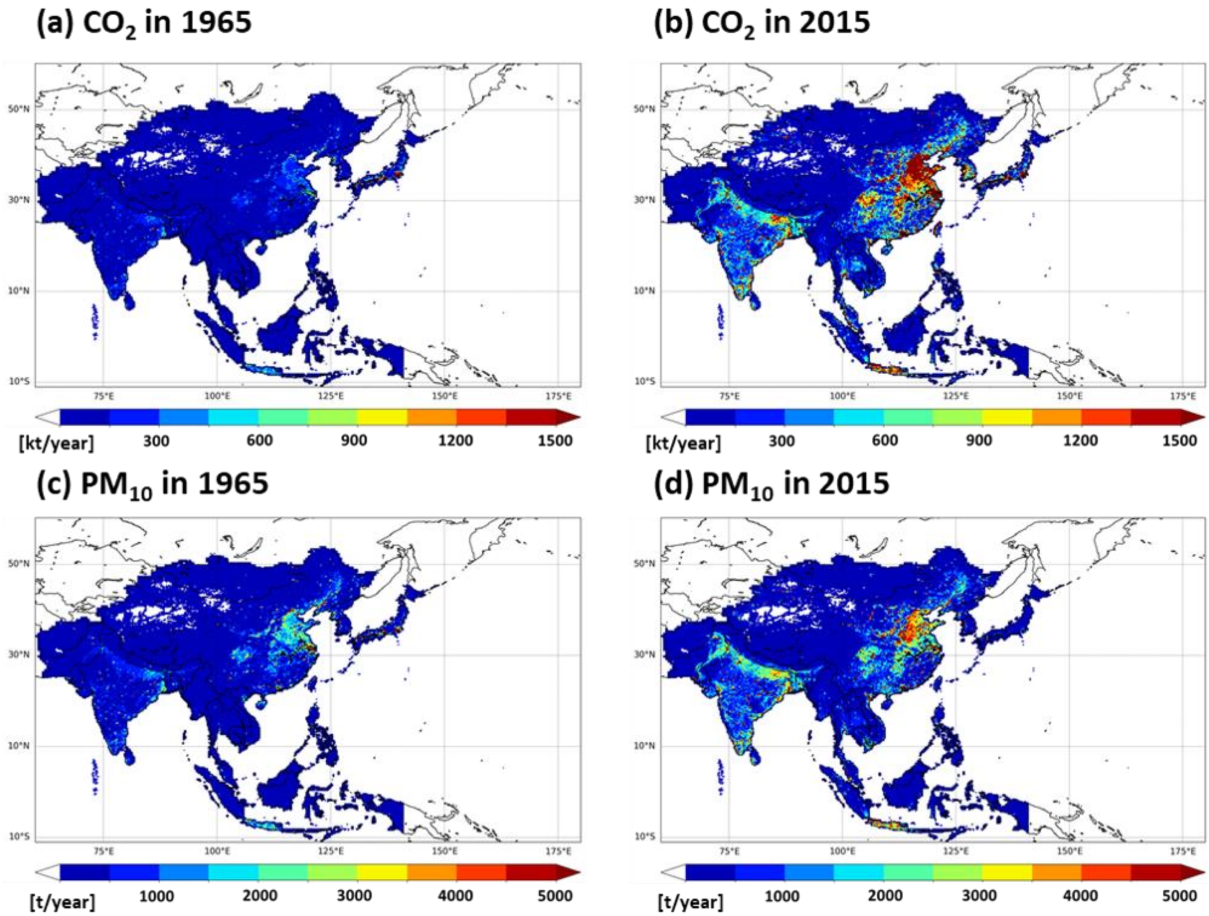


Figure S13. Grid maps of annual emissions of (a, c) CO_2 (kt year^{-1} per grid cell) and (b, d) PM_{10} (t year^{-1} per grid cell) in 1965 (left) and 2015 (right).

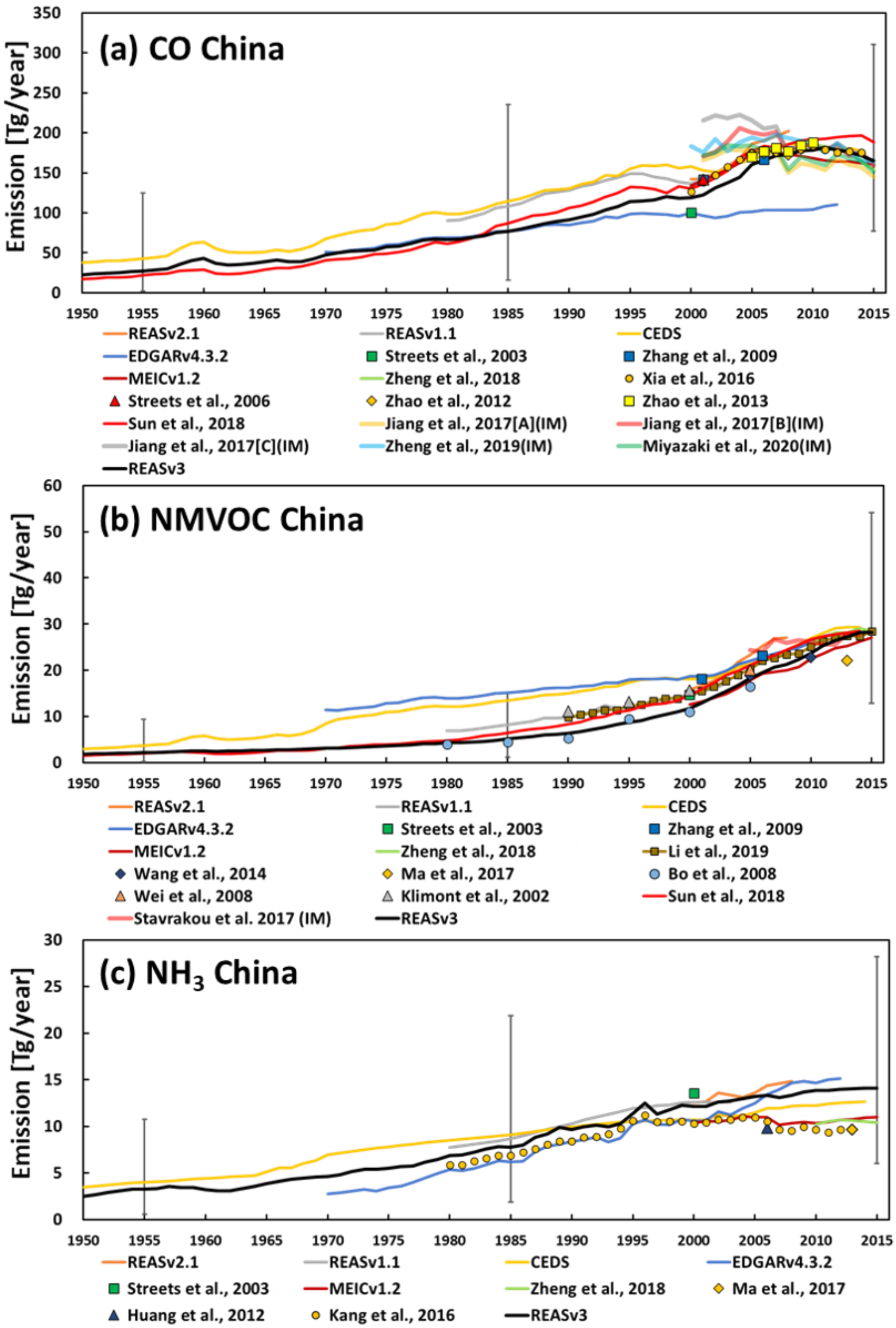
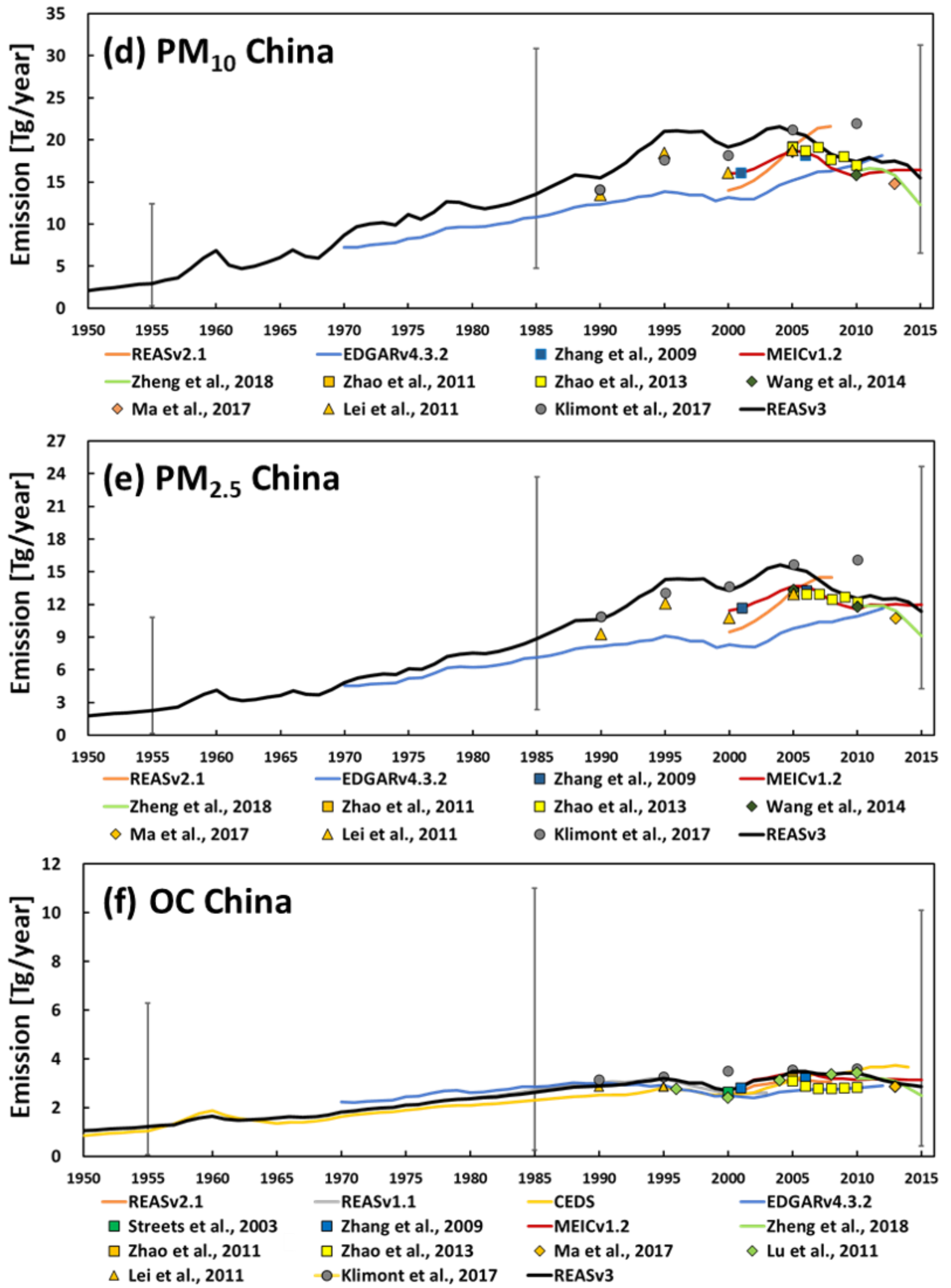


Figure S14. Comparison of (a) CO, (b) NMVOC, (c) NH_3, (d) PM_{10}, (e) $PM_{2.5}$ and (f) BC emissions in China between REASv3 and other studies. Emissions from domestic and fishing ships were excluded from REAS series, CEDS, and EDGARv4.3.2. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.



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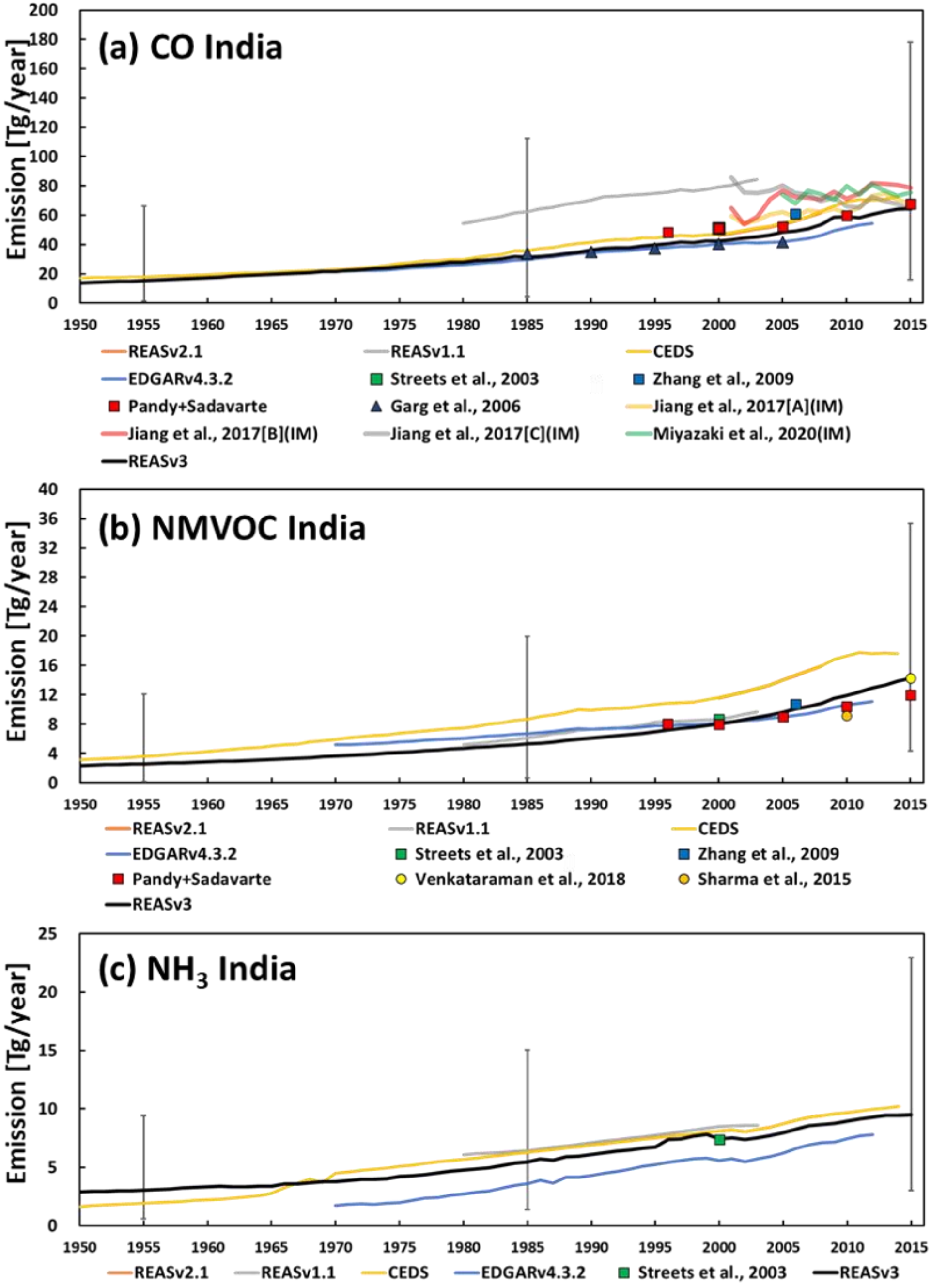

90 **Figure S15.** Comparison of (a) CO, (b) NMVOC, (c) NH_3, (d) PM_{10}, (e) $PM_{2.5}$ and (f) OC emissions in India between REASv3 and other studies. Emissions from domestic and fishing ships were excluded from REAS series, CEDS, and EDGARv4.3.2. Note that values of "Pandy+Sadavarte" are calculated from Pandey and Venkataraman (2014) and Sadavarte and Venkataraman (2014). Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.

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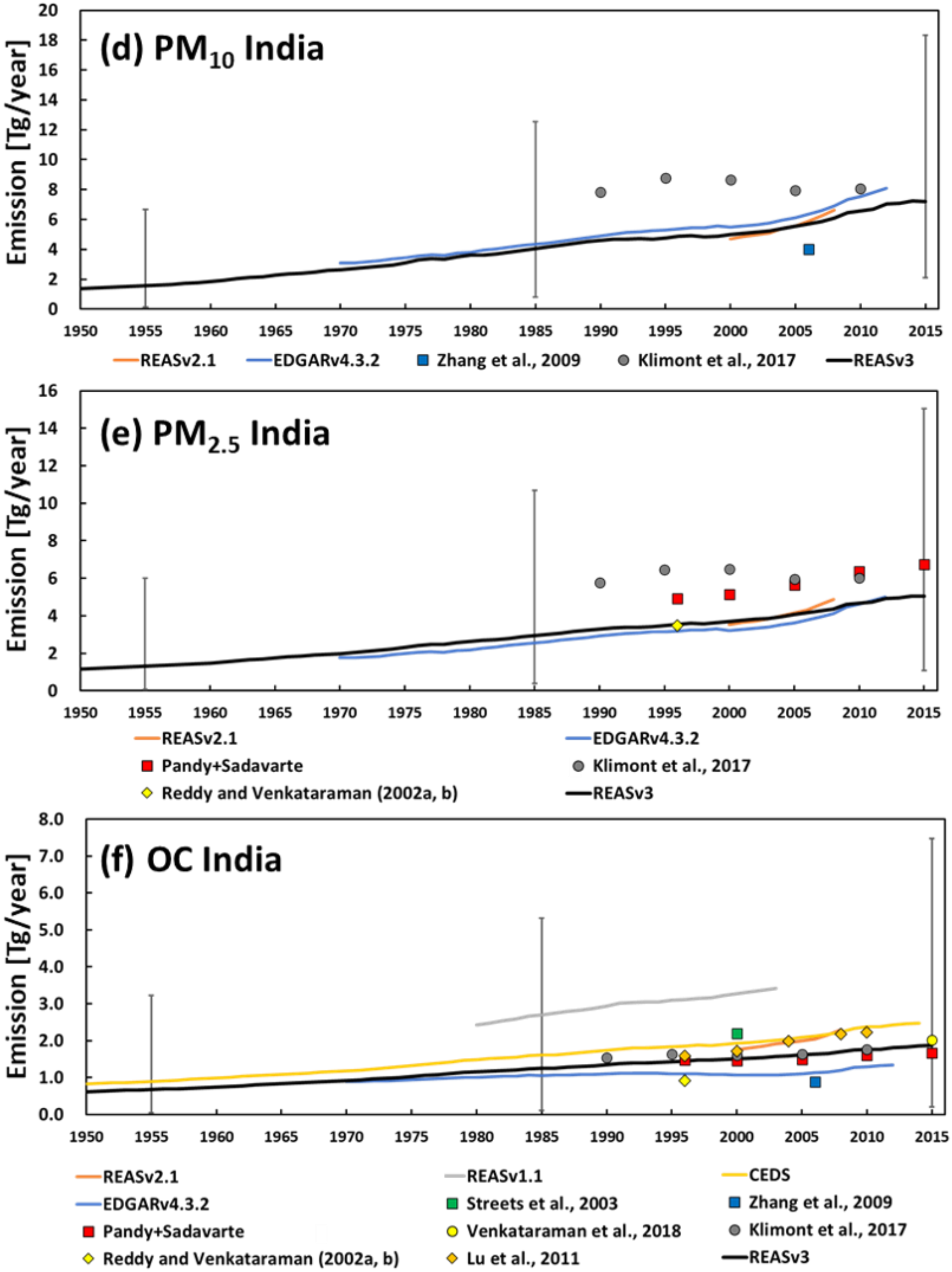

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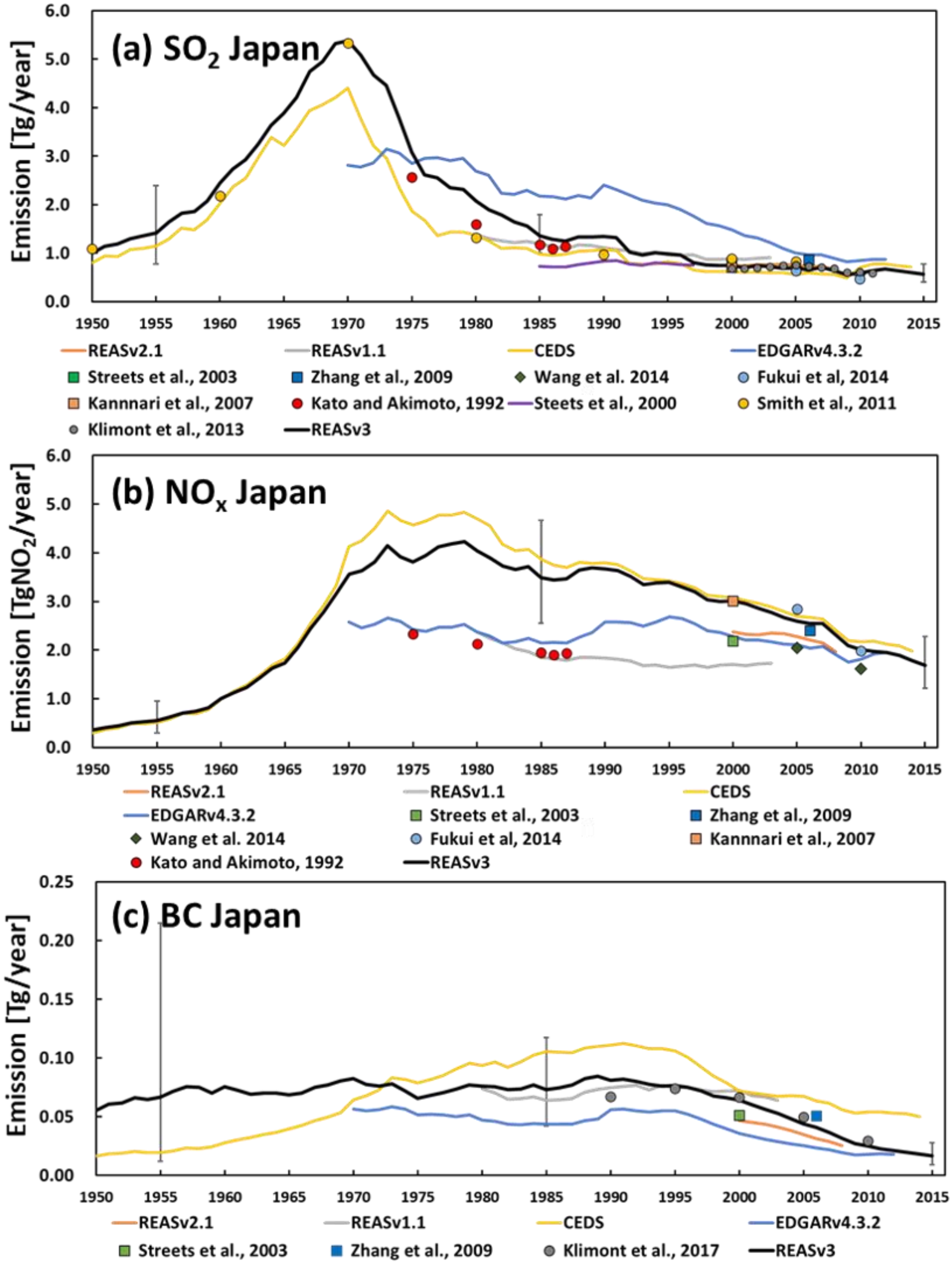
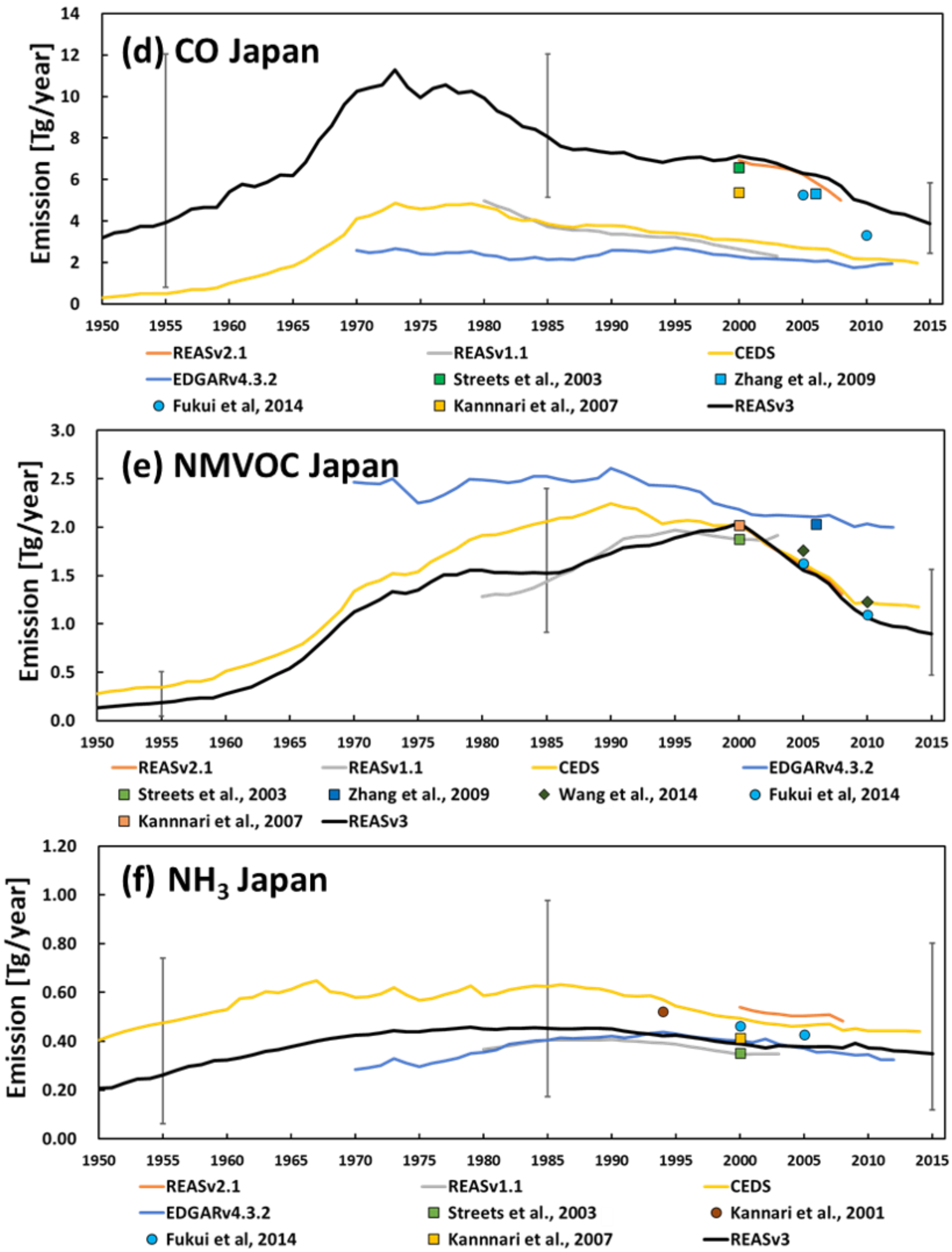


Figure S16. Comparison of (a) SO_2, (b) NO_x, (c) BC, (d) CO, (e) NMVOC, (f) NH_3, (g) PM_{10}, (h) $PM_{2.5}$ and (i) OC emissions in Japan between REASv3 and other studies. Emissions from domestic and fishing ships were excluded from REAS series, CEDS, EDGARv4.3.2, Kannari et al. (2007), and Fukui et al. (2013). Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.



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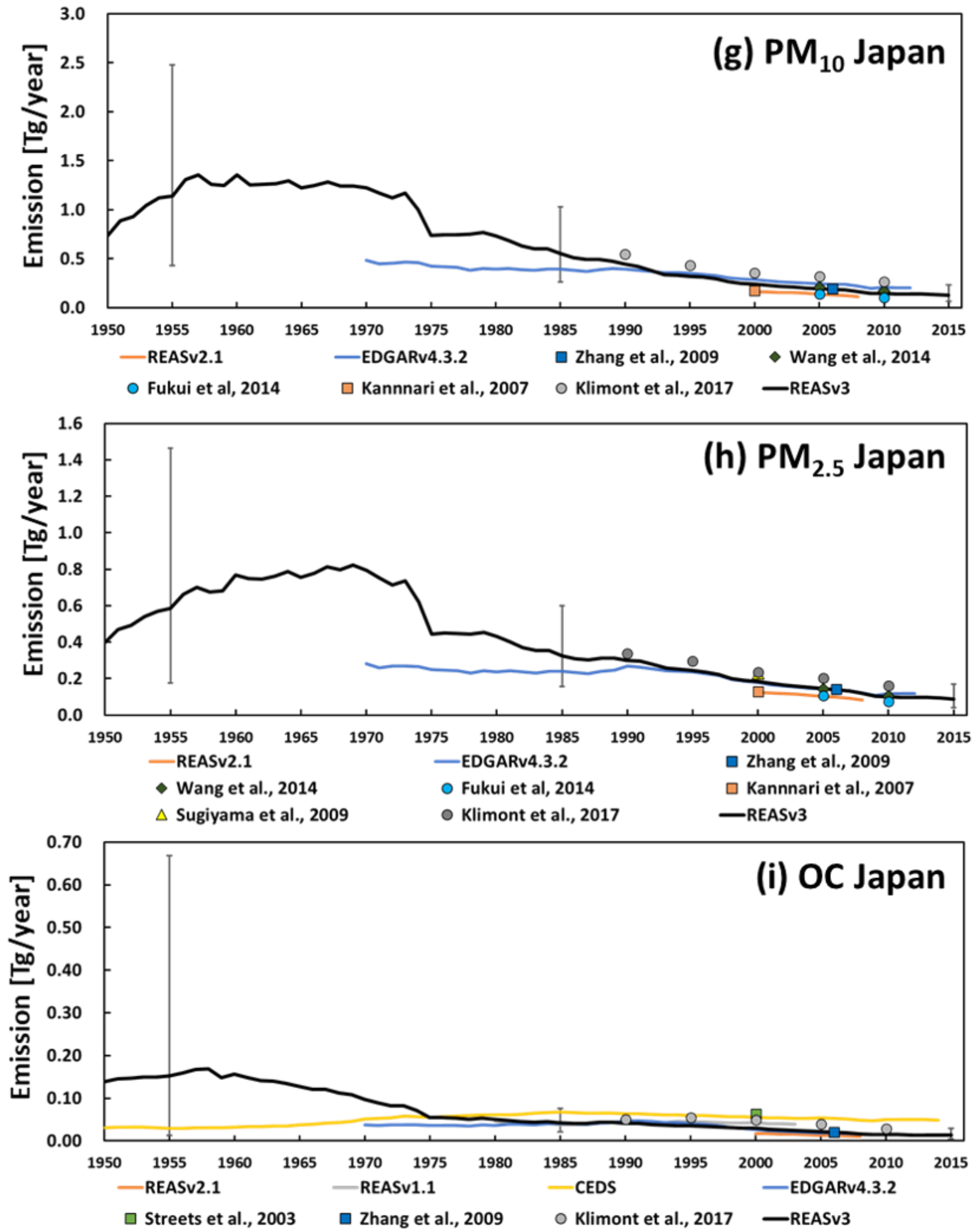

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Figure S16. Continued.

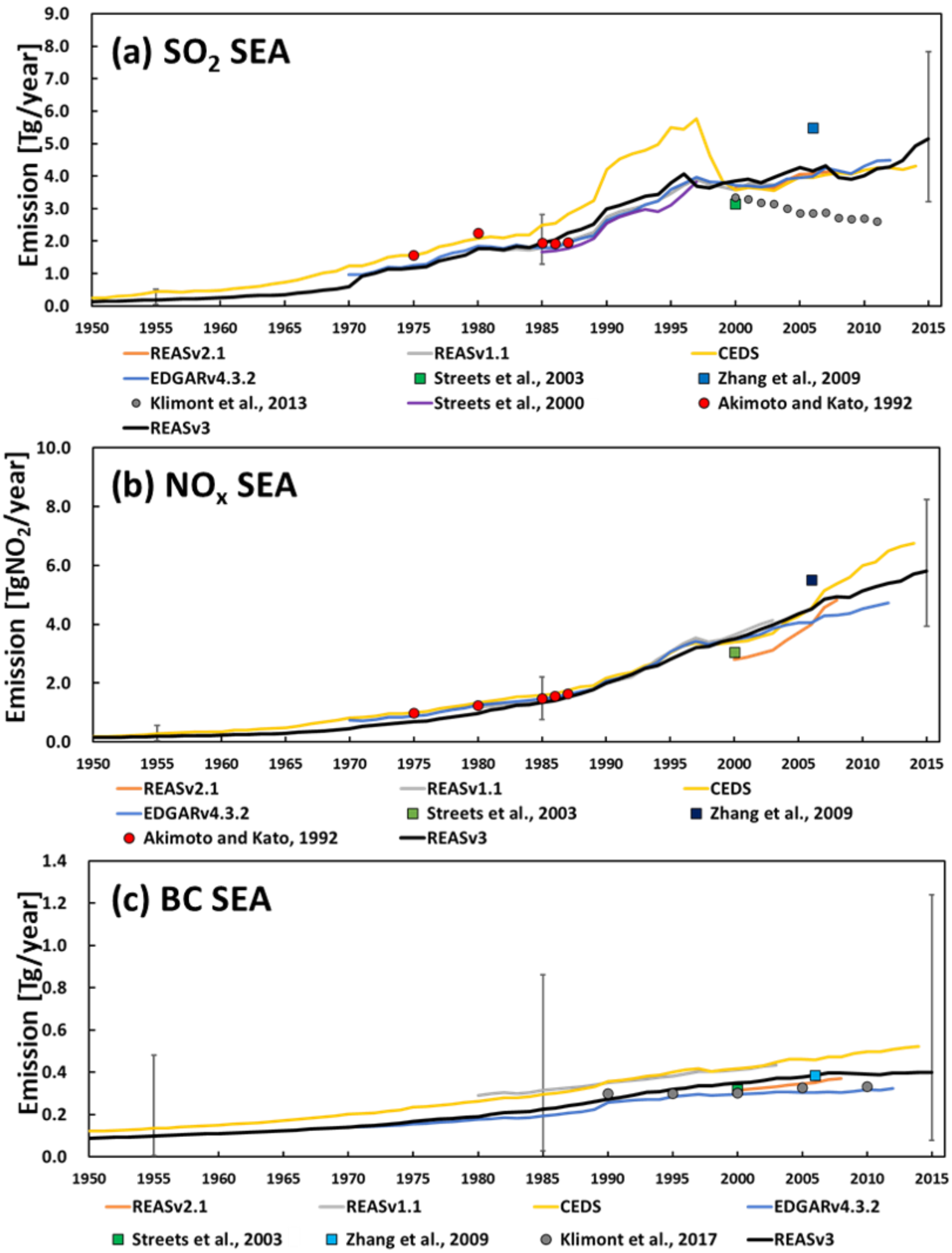
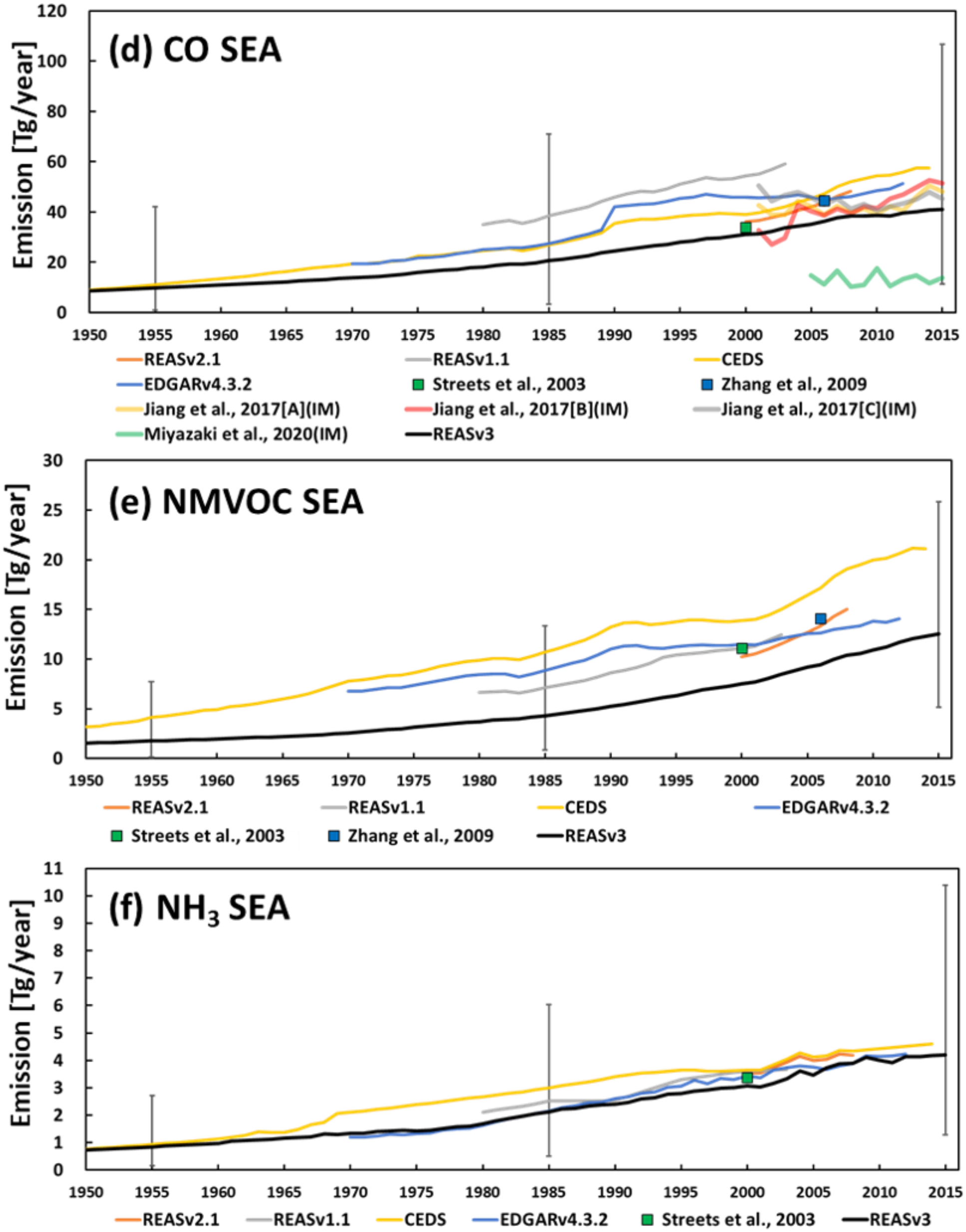


Figure S17. Comparison of (a) SO_2, (b) NO_x, (c) BC, (d) CO, (e) NMVOC, (f) NH_3, (g) PM_{10}, (h) $PM_{2.5}$ and (i) OC emissions in SEA between REASv3 and other studies. Emissions from domestic and fishing ships were excluded from REAS series, CEDS, and EDGARv4.3.2. See Fig. 1 for the definitions of SEA. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.



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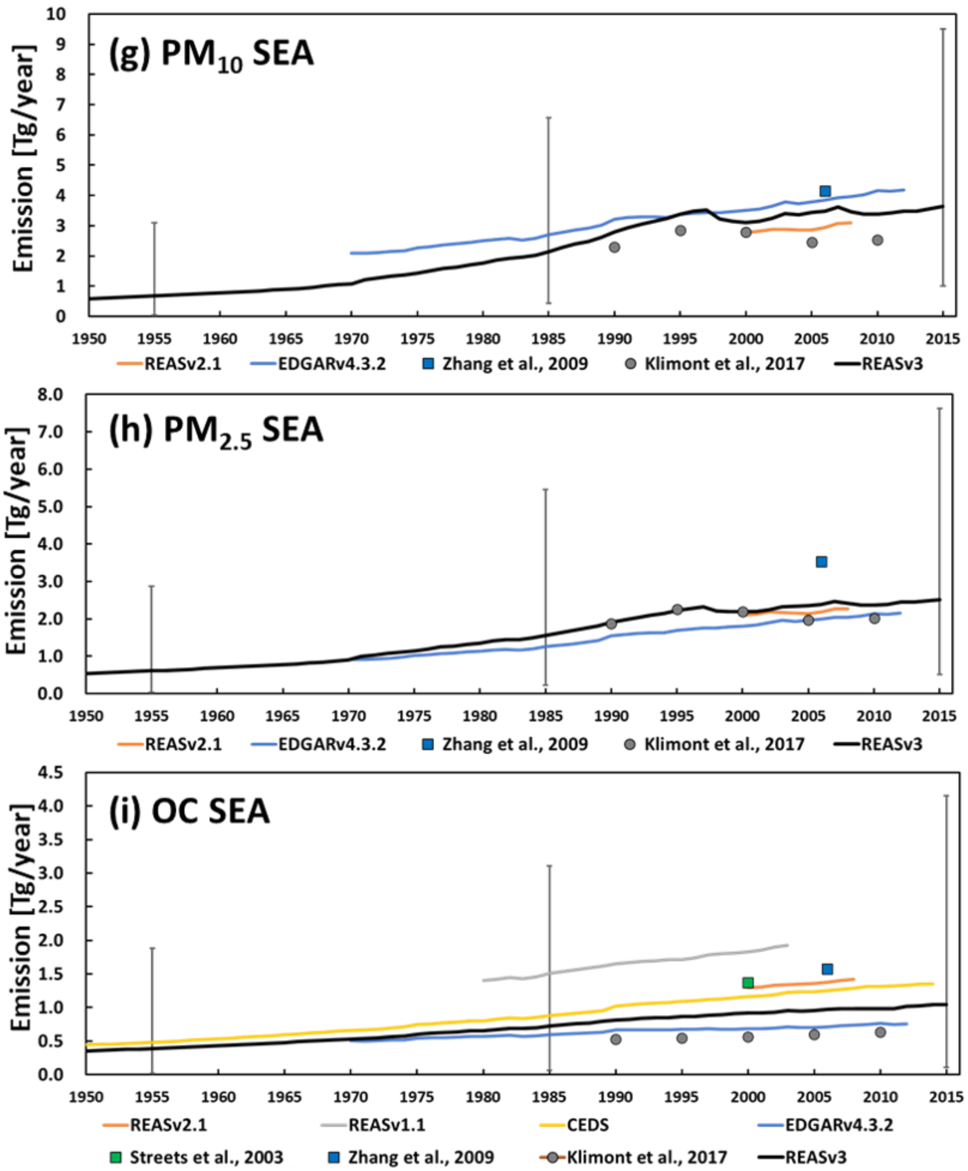

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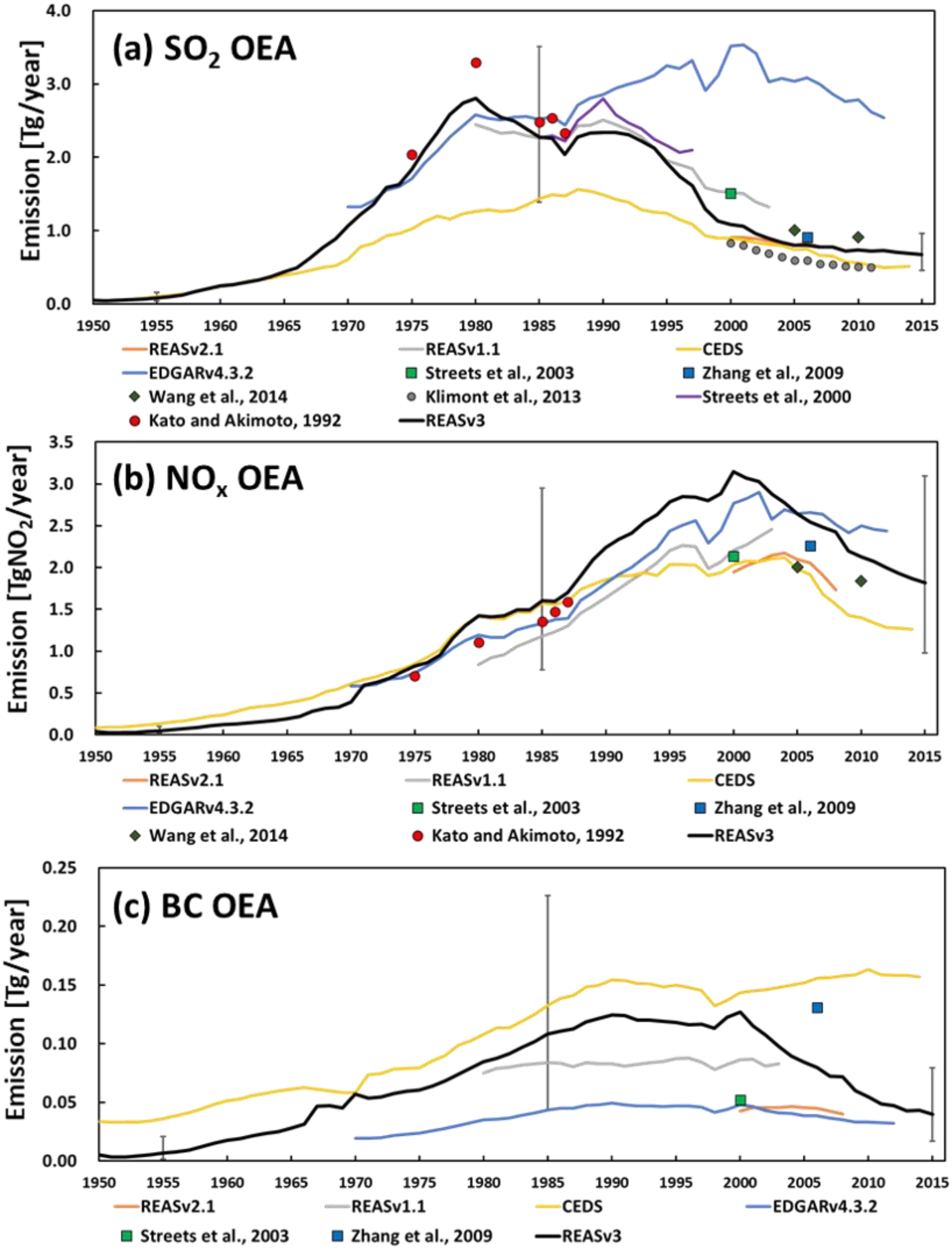
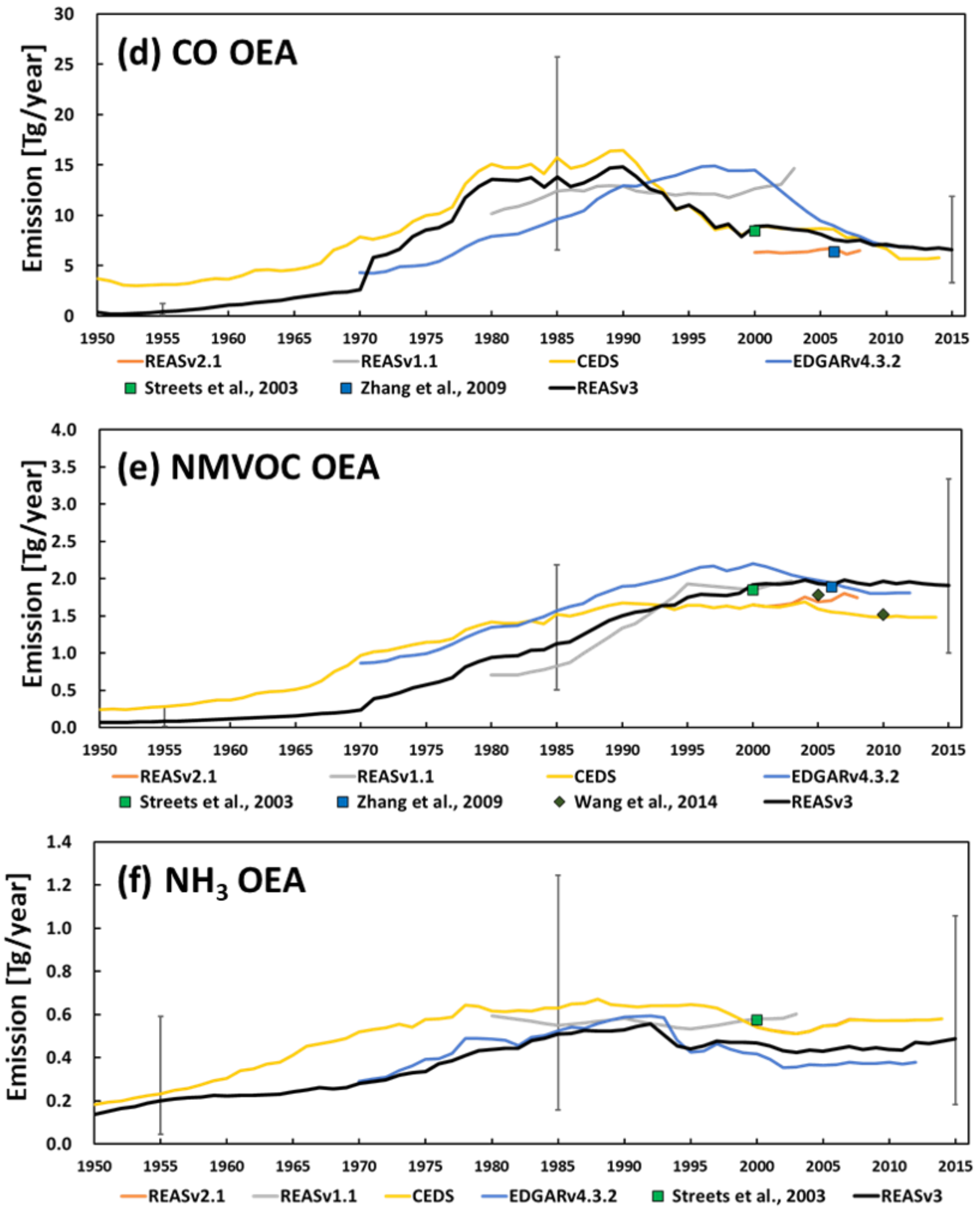


Figure S18. Comparison of (a) SO_2, (b) NO_x, (c) BC, (d) CO, (e) NMVOC, (f) NH_3, (g) PM_{10}, (h) $PM_{2.5}$ and (i) OC emissions in OEA between REASv3 and other studies. Emissions from domestic and fishing ships were excluded from REAS series, CEDS, and EDGARv4.3.2. See Fig. 1 for the definitions of OEA. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.



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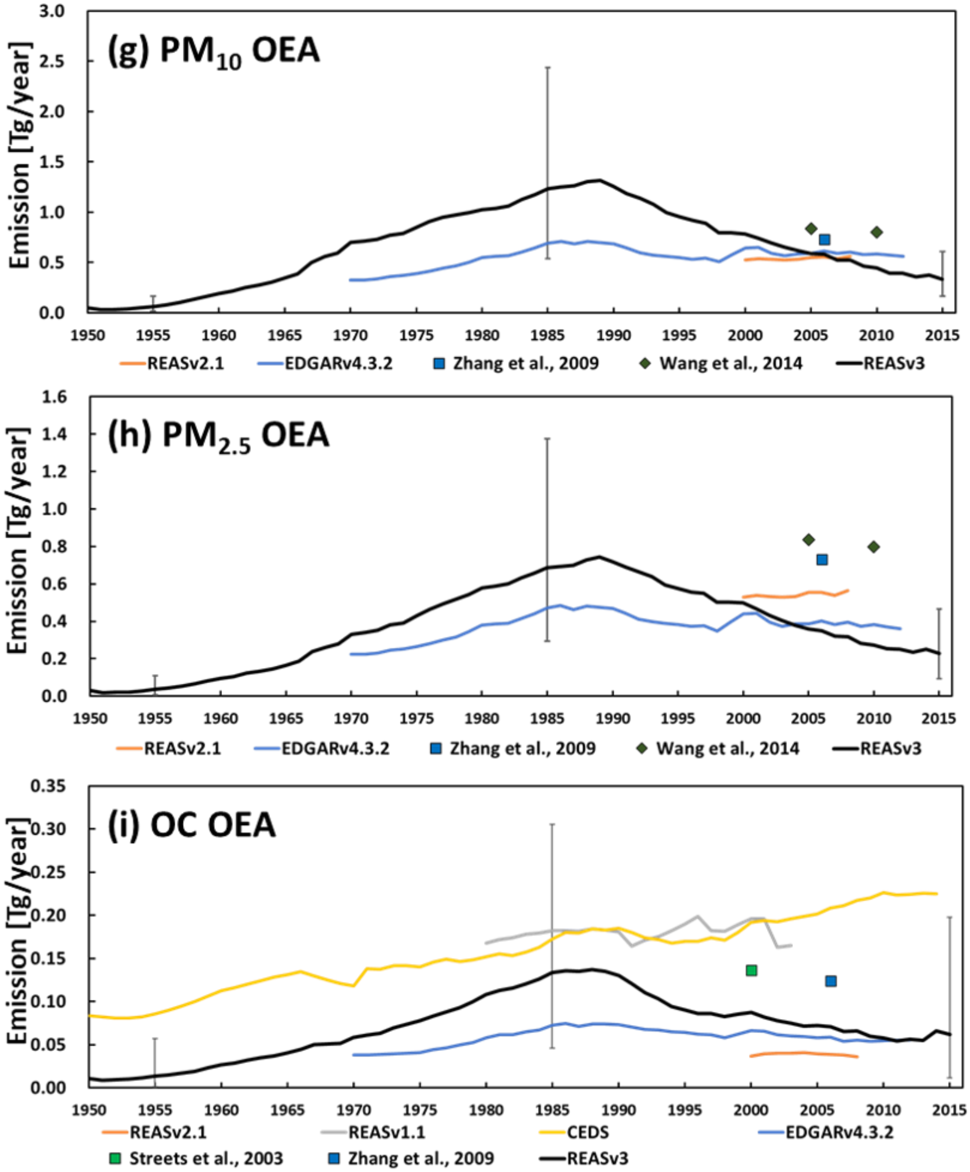

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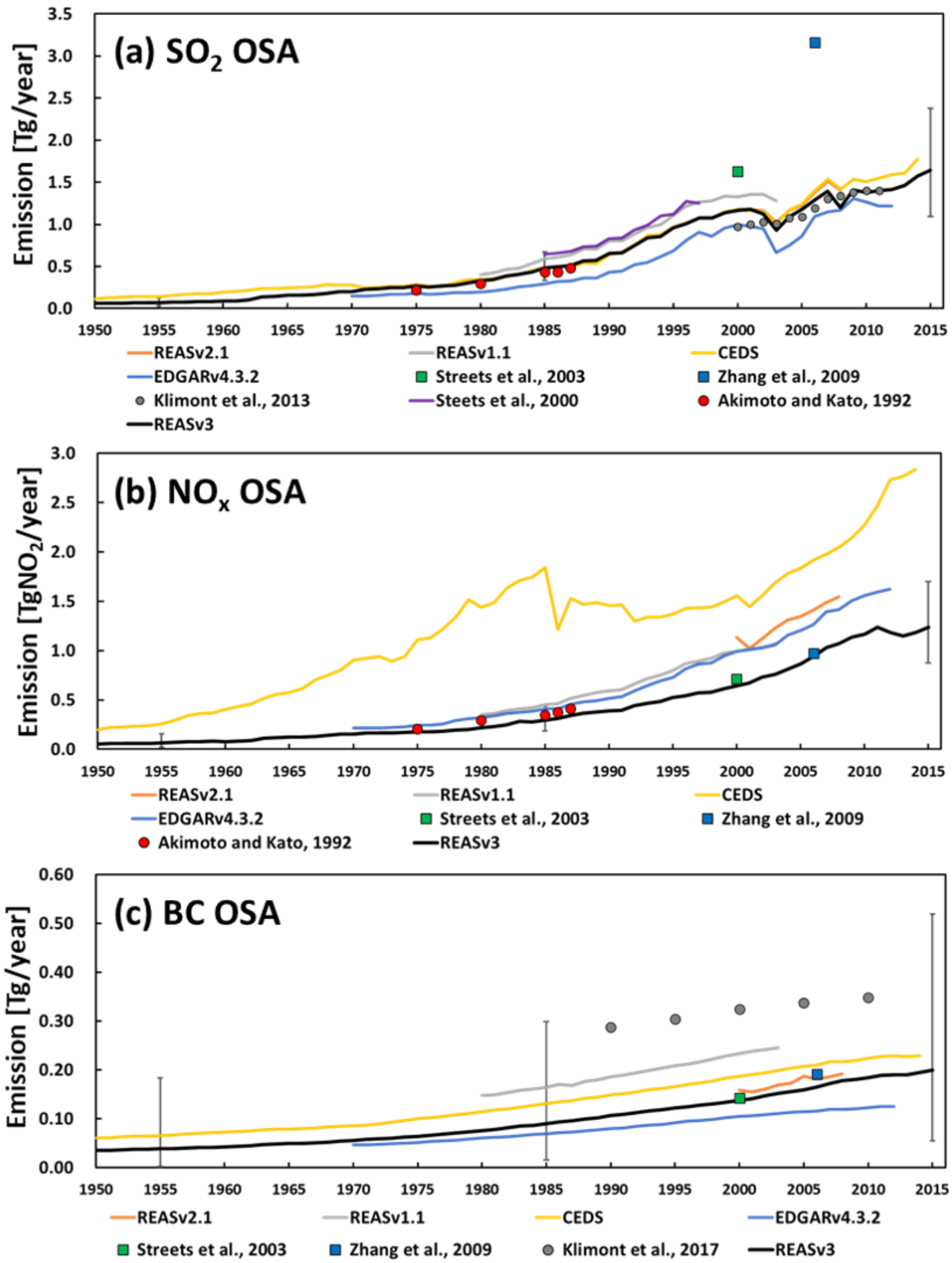
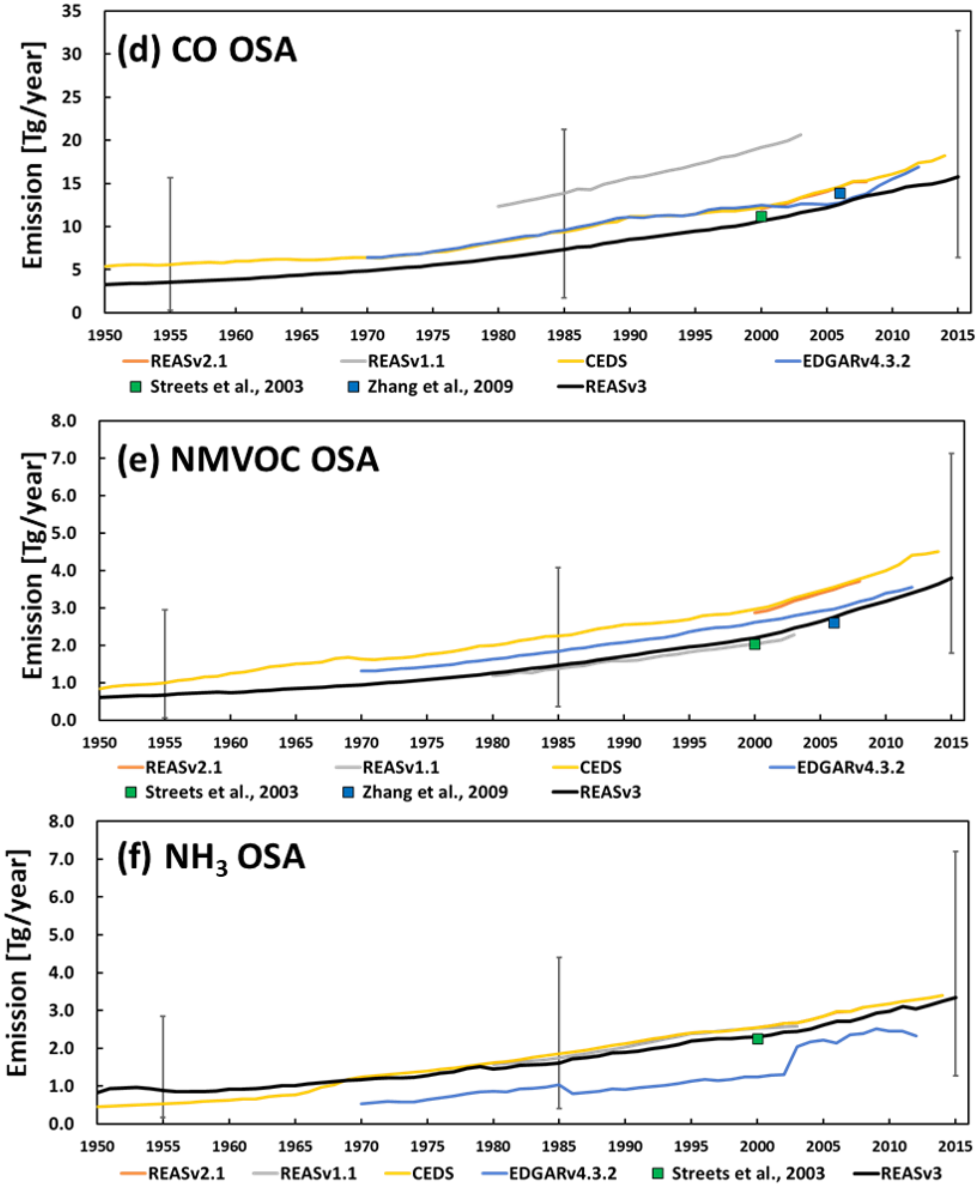


Figure S19. Comparison of (a) SO_2, (b) NO_x, (c) BC, (d) CO, (e) NMVOC, (f) NH_3, (g) PM_{10}, (h) $PM_{2.5}$ and (i) OC emissions in OSA between REASv3 and other studies. Emissions from domestic and fishing ships were excluded from REAS series, CEDS, and EDGARv4.3.2. See Fig. 1 for the definitions of OSA. Error bars indicate the uncertainty range of REASv3 in 1955, 1985, and 2015.



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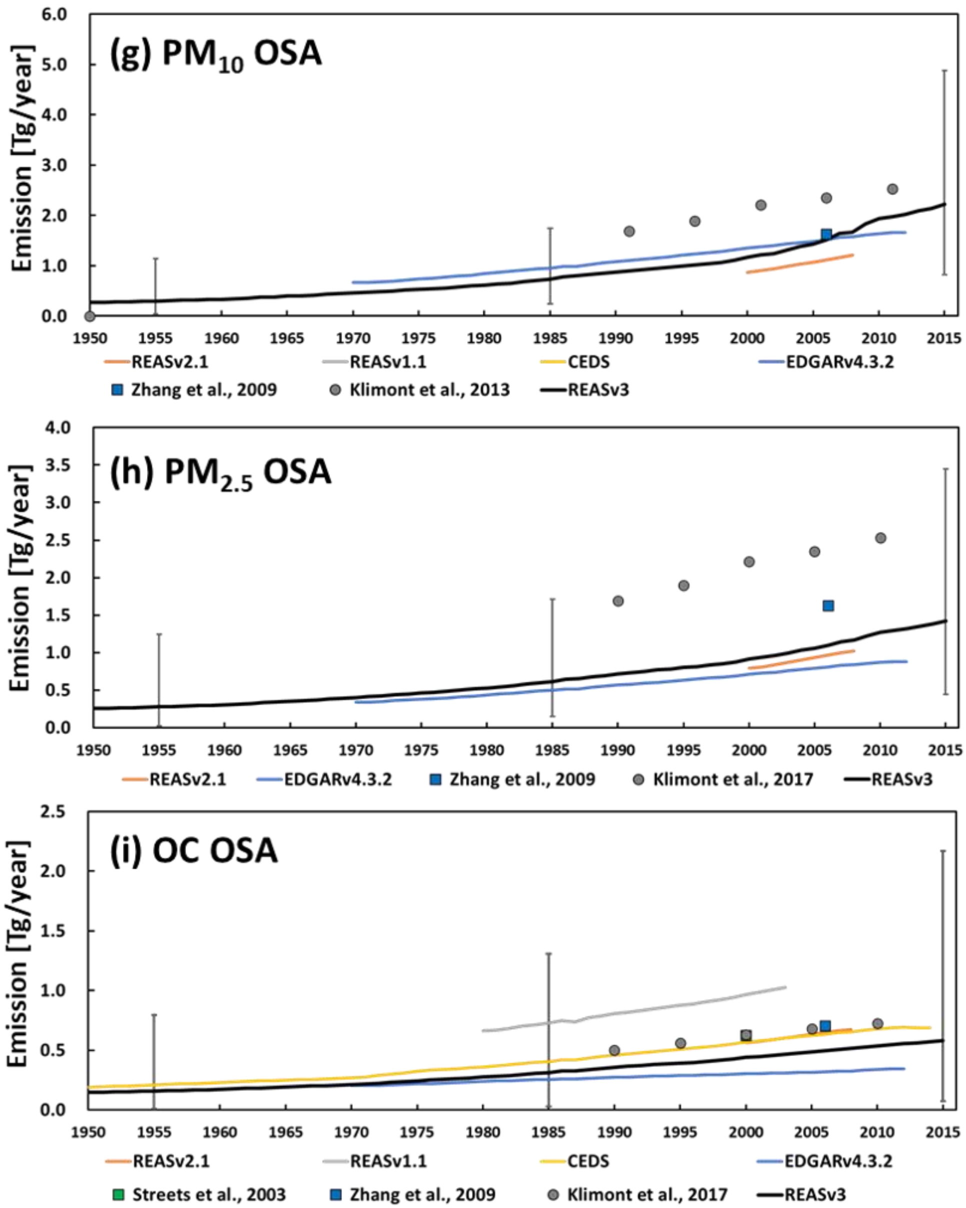

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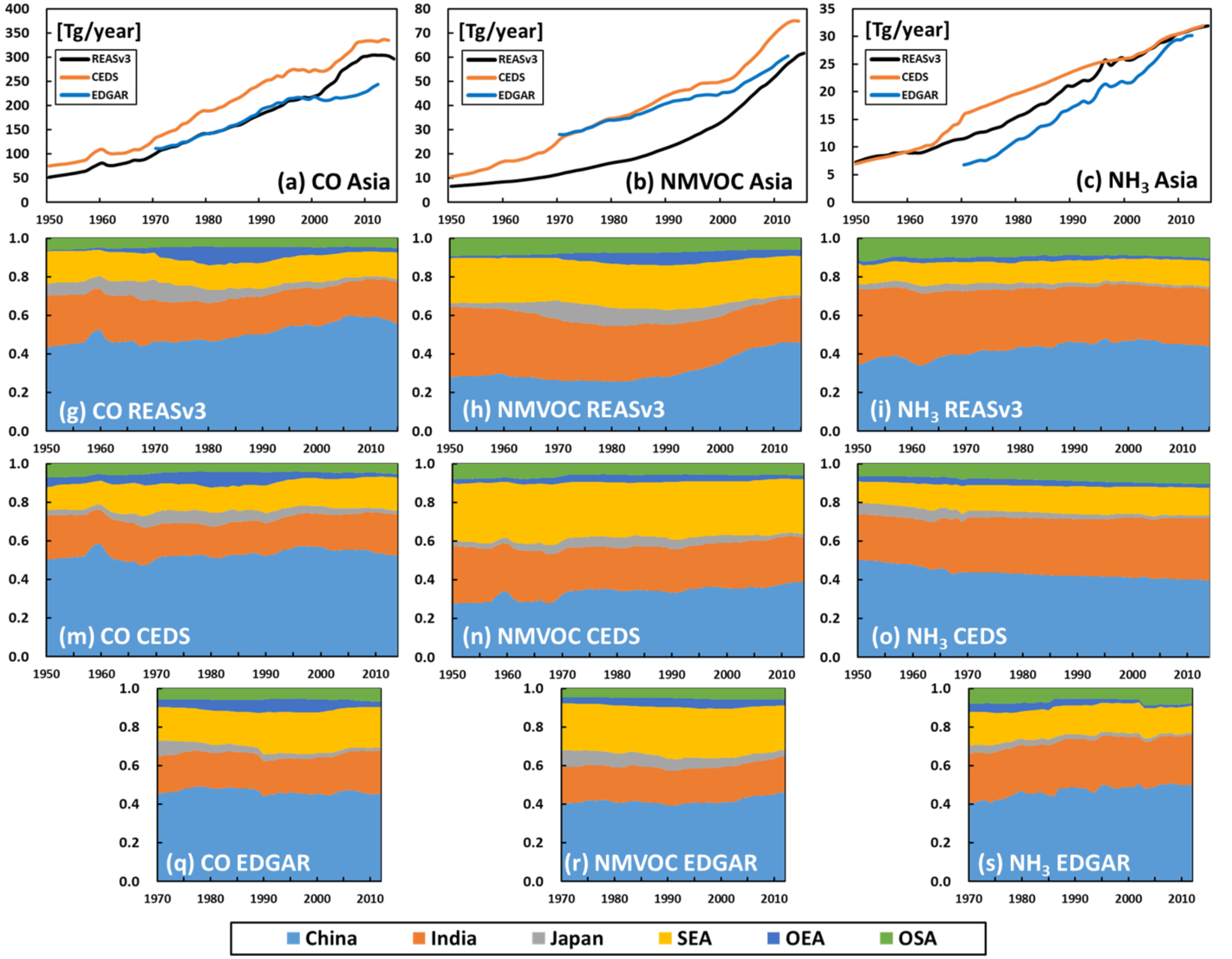

Figure S20. Comparison of trends of (a) CO, (b) NMVOC, (c) NH_3, (d) PM_{10}, (e) $PM_{2.5}$, (f) OC emissions in Asia and relative ratios of emissions from China, India, Japan, SEA, OEA, and OSA for (g, m, q) CO, (h, n, r) NMVOC, (i, o, s) NH_3, (j, t) PM_{10}, (k, u) $PM_{2.5}$, and (l, p, v) OC among (g, h, i, j, k, l) REASv3, (m, n, o, p) CEDS, and (q, r, s, t, u, v) EDGARv4.3.2. See Fig. 1 for the definitions of SEA, OEA, and OSA.

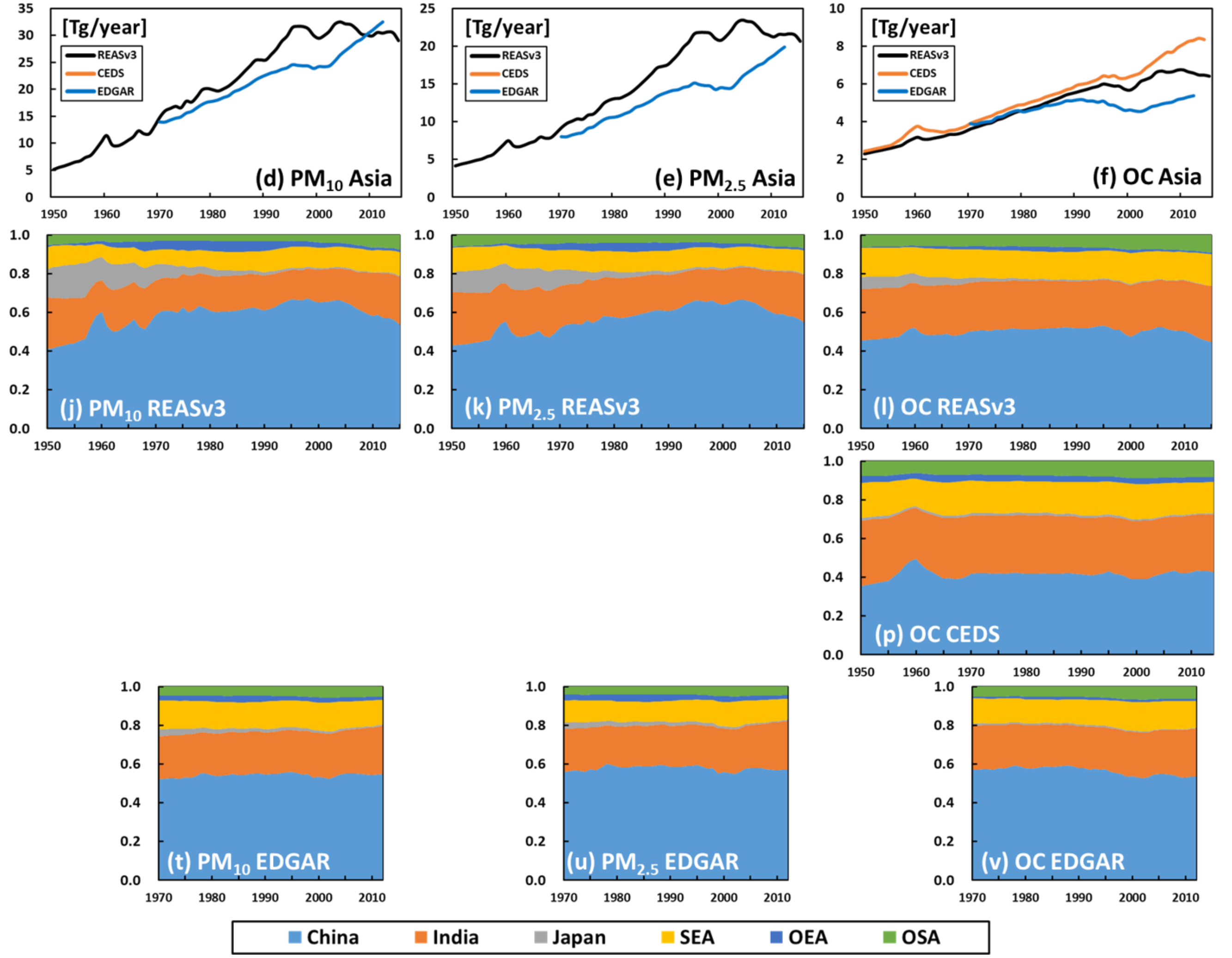

Figure S20. Continued.

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Supplement of

Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3

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Supplementary tables

Table S1 shows uncertainties of emissions of SO_2, NO_x, CO, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC for each sector in China, India, Japan, Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA) in 1955, 1985, and 2015. See Fig. 1 for the definitions of SEA, OEA, and OSA.

Table S2 presents uncertainties of emissions of NMVOC for each sector in China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. See Fig. 1 for the definitions of SEA, OEA, and OSA.

Table S3 provides uncertainties of emissions of NH_3 for each sector in China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. See Fig. 1 for the definitions of SEA, OEA, and OSA.

Table S1. Uncertainties [%] of emissions of SO_2, NO_x, CO, CO_2, PM_{10}, $PM_{2.5}$, BC, and OC for each sector in China, India, Japan, SEA, OEA, and OSA in (a) 1955, (b) 1985, and (c) 2015. Abbreviations for sectors are as follows: PP = Power Plants, IND = Industry, ROAD = Road transport, OTRA = Other transport, and DOM = Domestic. For OTRA of OEA in 1955, no emissions were estimated. See Fig. 1 for the definitions of SEA, OEA, and OSA.

(a) 1955

	SO_2	NO_x	CO	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
China								
PP	±115	±125	±180	±97	±163	±163	±148	±163
IND	±117	±153	±177	±91	±127	±126	±187	±177
ROAD	±73	±102	±91	±53	±128	±129	±131	±124
OTRA	±130	±163	±200	±103	±182	±182	±182	±182
DOM	±170	±303	±316	±168	±361	±365	±359	±370
India								
PP	±93	±117	±142	±83	±147	±152	±139	±153
IND	±111	±171	±240	±137	±259	±283	±273	±323
ROAD	±71	±97	±90	±52	±133	±134	±137	±129
OTRA	±123	±131	±169	±96	±195	±184	±168	±138
DOM	±249	±292	±305	±169	±370	±374	±368	±375
Japan								
PP	±100	±129	±152	±84	±179	±177	±157	±161
IND	±92	±128	±93	±79	±120	±132	±176	±163
ROAD	±45	±49	±72	±25	±77	±77	±79	±77
OTRA	±96	±121	±147	±94	±193	±178	±158	±122
DOM	±92	±160	±234	±107	±256	±301	±308	±331
SEA								
PP	±114	±120	±147	±90	±165	±163	±140	±143
IND	±164	±212	±271	±162	±294	±309	±315	±328
ROAD	±70	±93	±116	±57	±129	±128	±131	±125
OTRA	±130	±163	±200	±103	±200	±200	±200	±200
DOM	±304	±294	±306	±164	±373	±375	±377	±374
OEA								
PP	±96	±138	±171	±97	±181	±180	±171	±163
IND	±100	±131	±157	±87	±152	±153	±162	±171
ROAD	±80	±135	±119	±64	±150	±152	±153	±147
OTRA	-	-	-	-	-	-	-	-
DOM	±224	±202	±254	±106	±276	±304	±282	±319
OSA								
PP	±91	±108	±137	±76	±169	±164	±130	±144
IND	±168	±203	±267	±142	±265	±296	±312	±327
ROAD	±69	±85	±96	±54	±139	±142	±142	±136
OTRA	±112	±144	±172	±86	±160	±160	±159	±162
DOM	±292	±320	±327	±179	±383	±383	±383	±383

(b) 1985

	SO_2	NO_x	CO	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
China								
PP	±48	±62	±119	±37	±110	±109	±94	±108
IND	±57	±93	±127	±37	±102	±94	±148	±144
ROAD	±29	±69	±63	±17	±92	±93	±94	±89
OTRA	±69	±116	±154	±49	±144	±144	±144	±144
DOM	±81	±178	±231	±88	±255	±265	±250	±276
India								
PP	±47	±75	±107	±37	±128	±129	±119	±91
IND	±58	±96	±155	±61	±175	±194	±190	±260
ROAD	±32	±76	±65	±18	±105	±106	±109	±100
OTRA	±61	±80	±106	±39	±155	±132	±112	±103
DOM	±152	±203	±247	±110	±307	±313	±303	±315
Japan								
PP	±51	±69	±86	±19	±107	±103	±88	±142
IND	±50	±78	±88	±26	±101	±112	±115	±146
ROAD	±29	±38	±55	±22	±66	±66	±67	±66
OTRA	±44	±120	±129	±27	±135	±135	±135	±135
DOM	±45	±103	±119	±19	±110	±120	±133	±157
SEA								
PP	±55	±65	±105	±29	±125	±121	±102	±162
IND	±64	±101	±202	±63	±167	±182	±222	±264
ROAD	±30	±68	±86	±18	±87	±87	±90	±85
OTRA	±70	±116	±154	±49	±154	±154	±154	±154
DOM	±181	±195	±253	±107	±314	±317	±316	±317
OEA								
PP	±52	±75	±111	±37	±115	±114	±111	±95
IND	±63	±91	±114	±39	±122	±125	±123	±132
ROAD	±41	±111	±100	±23	±125	±127	±130	±122
OTRA	±59	±116	±135	±44	±135	±135	±135	±135
DOM	±84	±105	±134	±40	±163	±162	±161	±165
OSA								
PP	±56	±58	±81	±26	±102	±101	±88	±91
IND	±55	±96	±184	±58	±169	±184	±203	±258
ROAD	±32	±75	±83	±20	±110	±111	±112	±107
OTRA	±53	±90	±115	±34	±105	±105	±105	±106
DOM	±200	±233	±273	±120	±325	±325	±324	±326

(c) 2015

	SO_2	NO_x	CO	CO_2	PM_{10}	$PM_{2.5}$	BC	OC
China								
PP	±55	±65	±97	±34	±85	±85	±84	±81
IND	±66	±71	±78	±28	±92	±95	±127	±127
ROAD	±22	±41	±45	±15	±65	±65	±71	±60
OTRA	±82	±94	±118	±35	±122	±116	±113	±116
DOM	±71	±129	±204	±58	±235	±245	±227	±259
India								
PP	±60	±55	±106	±32	±102	±101	±93	±189
IND	±54	±74	±103	±37	±147	±160	±152	±241
ROAD	±27	±54	±55	±13	±67	±68	±72	±63
OTRA	±53	±103	±121	±35	±122	±122	±122	±122
DOM	±130	±161	±230	±89	±289	±297	±286	±300
Japan								
PP	±49	±57	±98	±23	±95	±97	±80	±131
IND	±50	±68	±81	±23	±94	±100	±93	±136
ROAD	±21	±27	±37	±13	±51	±51	±52	±51
OTRA	±38	±109	±117	±19	±119	±120	±120	±121
DOM	±47	±82	±111	±13	±107	±106	±116	±121
SEA								
PP	±70	±56	±96	±30	±111	±112	±105	±167
IND	±59	±79	±155	±41	±149	±164	±194	±246
ROAD	±28	±57	±78	±14	±70	±70	±72	±72
OTRA	±53	±103	±122	±35	±122	±122	±122	±122
DOM	±169	±158	±241	±89	±296	±300	±296	±302
OEA								
PP	±51	±47	±81	±26	±93	±90	±83	±80
IND	±65	±82	±109	±34	±108	±108	±110	±116
ROAD	±34	±92	±82	±16	±99	±100	±101	±97
OTRA	±48	±103	±125	±35	±123	±122	±122	±122
DOM	±76	±116	±161	±46	±178	±203	±184	±220
OSA								
PP	±53	±48	±72	±23	±78	±77	±70	±87
IND	±58	±86	±163	±45	±131	±144	±165	±243
ROAD	±27	±55	±79	±15	±81	±82	±87	±81
OTRA	±52	±103	±121	±35	±122	±122	±122	±122
DOM	±243	±202	±259	±100	±311	±311	±310	±312

Table S2. Uncertainties [%] of emissions of NMVOC for each sector in China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. Abbreviations for sectors are the same as in Table S1 except for EXT = Extraction processes, SLV = Solvent and paint use, and WST = Waste treatment. Note that uncertainties of emissions from non-combustion sources in Japan based on MOEJ (2017) were not assessed in this study. For OTRA of OEA in 1955, no emissions were estimated. See Fig. 1 for the definitions of SEA, OEA, and OSA.

	China	India	Japan	SEA	OEA	OSA
1955						
PP	±164	±124	±134	±147	±159	±134
IND	±147	±251	±160	±267	±242	±262
ROAD	±89	±83	±64	±125	±114	±102
OTRA	±200	±131	±117	±200	-	±155
DOM	±327	±324	±223	±326	±294	±329
EXT	±191	±164	-	±184	±216	±180
SLV	±167	±151	-	±167	±146	±155
WST	±245	±245	-	±245	±245	±245
1985						
PP	±103	±102	±84	±94	±105	±77
IND	±109	±159	±100	±169	±121	±148
ROAD	±65	±75	±51	±100	±96	±86
OTRA	±154	±107	±129	±154	±135	±105
DOM	±268	±269	±116	±273	±146	±275
EXT	±130	±117	-	±127	±137	±135
SLV	±161	±173	-	±177	±179	±180
WST	±200	±200	-	±200	±200	±200
2015						
PP	±94	±91	±97	±91	±76	±69
IND	±99	±115	±81	±133	±166	±136
ROAD	±64	±84	±50	±104	±82	±88
OTRA	±111	±122	±117	±122	±122	±122
DOM	±241	±256	±129	±259	±189	±262
EXT	±126	±114	-	±130	±141	±136
SLV	±153	±150	-	±158	±141	±146
WST	±173	±173	-	±192	±188	±200

Table S3. Uncertainties [%] of emissions of NH_3 for each sector in China, India, Japan, SEA, OEA, and OSA in 1955, 1985, and 2015. Abbreviations for sectors are the same as in Table S1 except for MISC = Human (perspiration and respiration) and latrines. Note that uncertainties of emissions from agricultural sources based on REASv1.1 (Yamaji et al., 2004; Yan et al., 2003) and REASv2.1 (Kurokawa et al., 2013; JPEC 2012a, b, c; 2014) were not assessed in this study. For OTRA of OEA in 1955, no emissions were estimated. See Fig. 1 for the definitions of SEA, OEA, and OSA.

	China	India	Japan	SEA	OEA	OSA
1955						
PP	±279	±236	±264	±238	±279	±263
IND	±129	±377	±243	±384	±325	±383
ROAD	±151	±187	±161	±215	±157	±193
OTRA	±260	±249	±250	±260	-	±228
DOM	±384	±382	±360	±375	±380	±383
MISC	±161	±161	±140	±161	±161	±161
1985						
PP	±217	±202	±300	±314	±206	±212
IND	±77	±199	±307	±241	±129	±151
ROAD	±126	±148	±144	±151	±142	±159
OTRA	±213	±185	±207	±213	±213	±205
DOM	±325	±324	±141	±318	±316	±326
MISC	±148	±161	±122	±161	±161	±161
2015						
PP	±192	±297	±290	±280	±158	±218
IND	±79	±157	±263	±185	±114	±138
ROAD	±145	±145	±162	±176	±142	±174
OTRA	±193	±200	±193	±200	±200	±200
DOM	±311	±310	±124	±305	±308	±312
MISC	±137	±137	±108	±137	±137	±137

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Supplement of

Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3

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Differences between REASv3.2 and REASv3.1

1. Introduction

Datasets of Regional Emission inventory in ASia version 3.1 (REASv3.1) were released from REAS data download site (https://www.nies.go.jp/REAS/) together with a publication of Kurokawa et al. (2019) from December 2019. The datasets were revised though the revision processes of Kurokawa et al. (2019) and the updated data are available as REASv3.2 from the data down load site.

The methodology of both versions was the same and the differences between REASv3.2 and REASv3.1 were mostly by correction of errors in data and system used for REASv3.1. The purpose of this document is to present degrees of differences between REASv3.2 and REASv3.1 especially for users who have already used REASv3.1. Note that following acronyms were used in this document: Southeast Asia (SEA), East Asia other than China and Japan (OEA), and South Asia other than India (OSA).

2. SO_2

Figure 1 compares total SO_2 emissions in China, India, Japan, SEA, OEA, and OSA. There were no clear differences except for Japan and OSA. For Japan, large discrepancies were found between 1970 and 1985: 7%, 22%, and 16% to total of REASv3.1 in 1970, 1980, and 1985, respectively. These differences were mainly caused by errors in settings of sulfur contents in heavy fuel oil in the industry sector. For OSA, discrepancies were found after around 2000: 6%, 5%, and 5% in 2005, 2010, and 2015. These differences were caused by errors in settings of sulfur contents in coal in Pakistan.

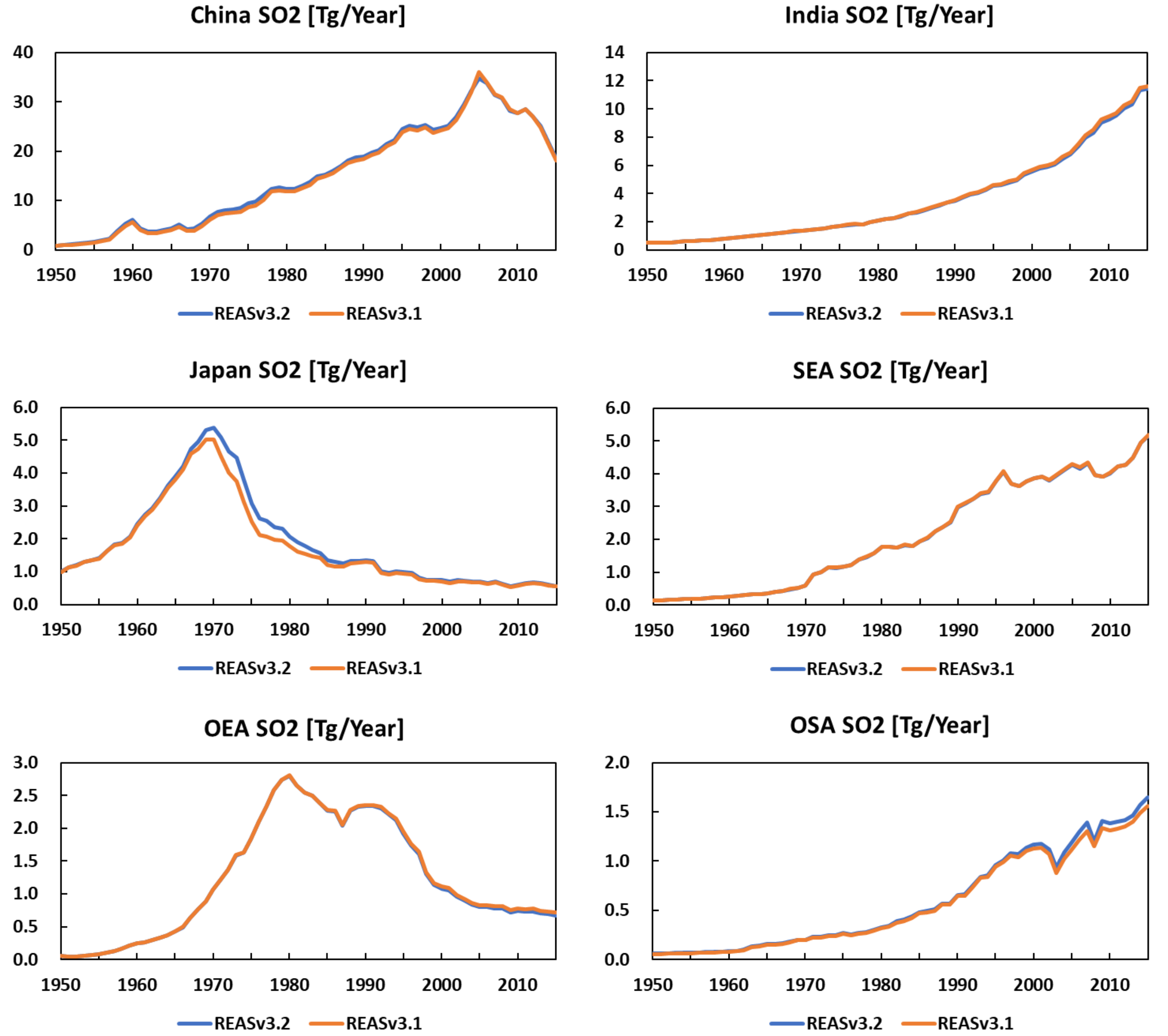


Figure 1. Differences of total SO_2 emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

3. NO_x

Figure 2 compares total NO_x emissions in China, India, Japan, SEA, OEA, and OSA. Somewhat discrepancies were found in all countries and regions. For China, values of REASv3.2 are larger than those of REASv3.1. The largest discrepancies of about 10% to total of REASv3.1 were found around 2000. For other years, the differences were about 7%, 5%, and 3% in 1990, 2010, and 2015. On the other hand, for India, values of REASv3.2 were smaller than REASv3.1. After around 1990, the discrepancies were around 5-7%. Similarly, for Japan, SEA, and OSA, values of REASv3.2 are smaller than those of REAS3.1. For Japan, emissions of REASv3.2 were smaller than REASv3.1 about 5-6% in 1990s and 7-8% after early 2000s. The differences were larger in SEA. The emission amounts of REASv3.2 were smaller about 14-17% from early 1980s to late 1990s and about 10-12% after 2000s. For OSA, after 1980s, values of REASv3.2 were about 5-10% smaller than REASv3.1. On the contrary, for OEA, emissions of REASv3.2 were about 20% higher than REASv3.1 and for other years, the differences were 8%, 18%, 14%, and 4% in 1990, 1995, 2005, and 2010, respectively. The common major causes of these discrepancies in all countries and regions were the road transport sector (Fig. 3). There were errors in settings of the emission factors. Another relatively large differences were in fuel combustion emissions in the agricultural sector of the domestic sector. The emission factors for diesel oil were incorrect except for Japan and higher values were adopted in REASv3.2. The differences in the agricultural sub-sector to total emissions of REASv3.1 were 3-4%, 4-7%, 2-5%, 3-6%, and 4-6% in China, India, OEA, OSA, and SEA, respectively.

4. CO

Figure 4 compares total CO emissions in China, India, Japan, SEA, OEA, and OSA. Relatively large discrepancies were clearly found in China and SEA. For other countries and regions, even though differences of total emissions were small, there were discrepancies in sector level emissions. For China, emissions of REASv3.2 were smaller than REASv3.1. The differences were 6-8%, 9-13%, and 15-20% to total of REASv3.1 during 2013-2015, 2001-2012, and 1970-2000. The causes of these differences were mistakes in settings of emission factors for the industry and road transport sectors. Comparisons of emissions from the industry and road transport sectors between REASv3.2 and REASv3.1 were plotted in Fig. 5. Emissions in SEA of REASv3.2 were also smaller than REASv3.1. The discrepancies were 8-10% after 1995 and 11-14% before 1994. One reason is correction of errors in biofuel data (settings of relative ratios of fuelwood, crop residue, and animal waste in primary solid biofuels) which reduced emissions from the residential sector. Another is errors in settings of emission factors of coal combustion in the industry sector. Similar correction of the emission factors for coal combustion of the industry sector were done for India and OSA. On the other hand, revision of emission factors for motor cycles and correction of emission factors for diesel combustion in the agricultural sub-sector of the domestic sector increased CO emissions of REASv3.2 from REASv3.1 and then, differences in total emissions were small in India and OSA. For Japan and OEA, differences between REASv3.2 and REASv3.1 were generally small for all sectors.

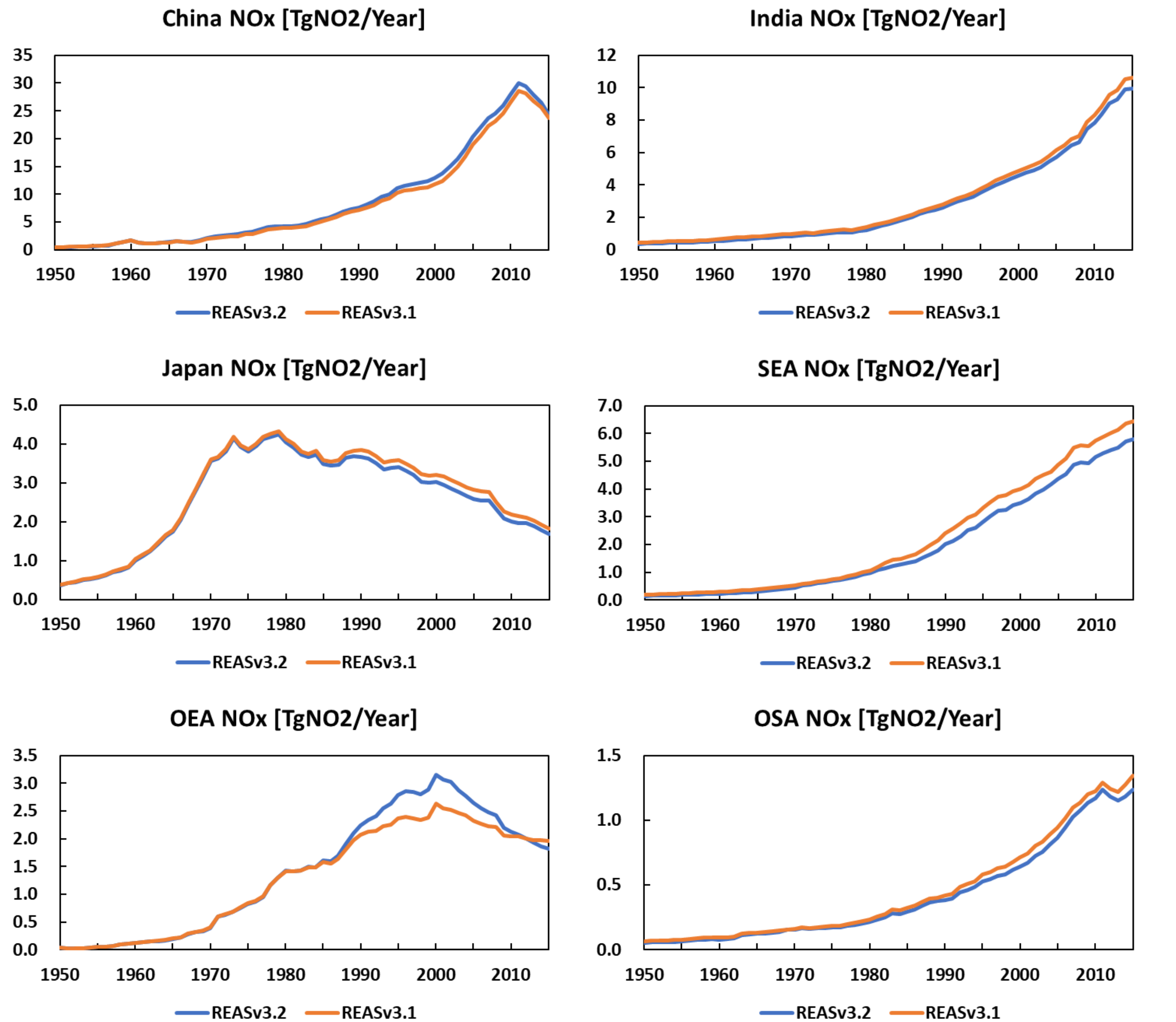


Figure 2. Differences of total NO_x emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

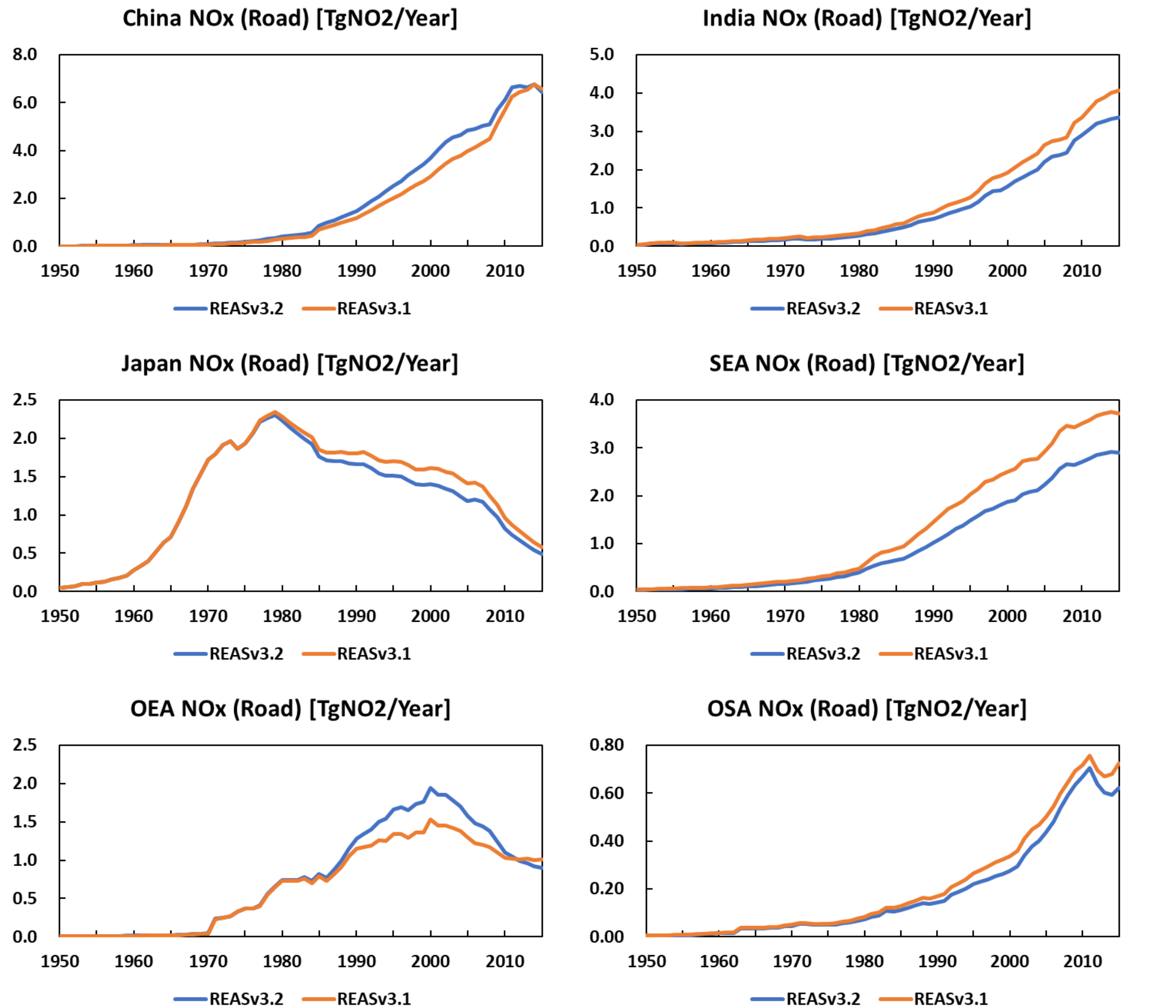


Figure 3. Differences of NO_x emissions from the road transport sector in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (red line) and REASv3.1 (red line).

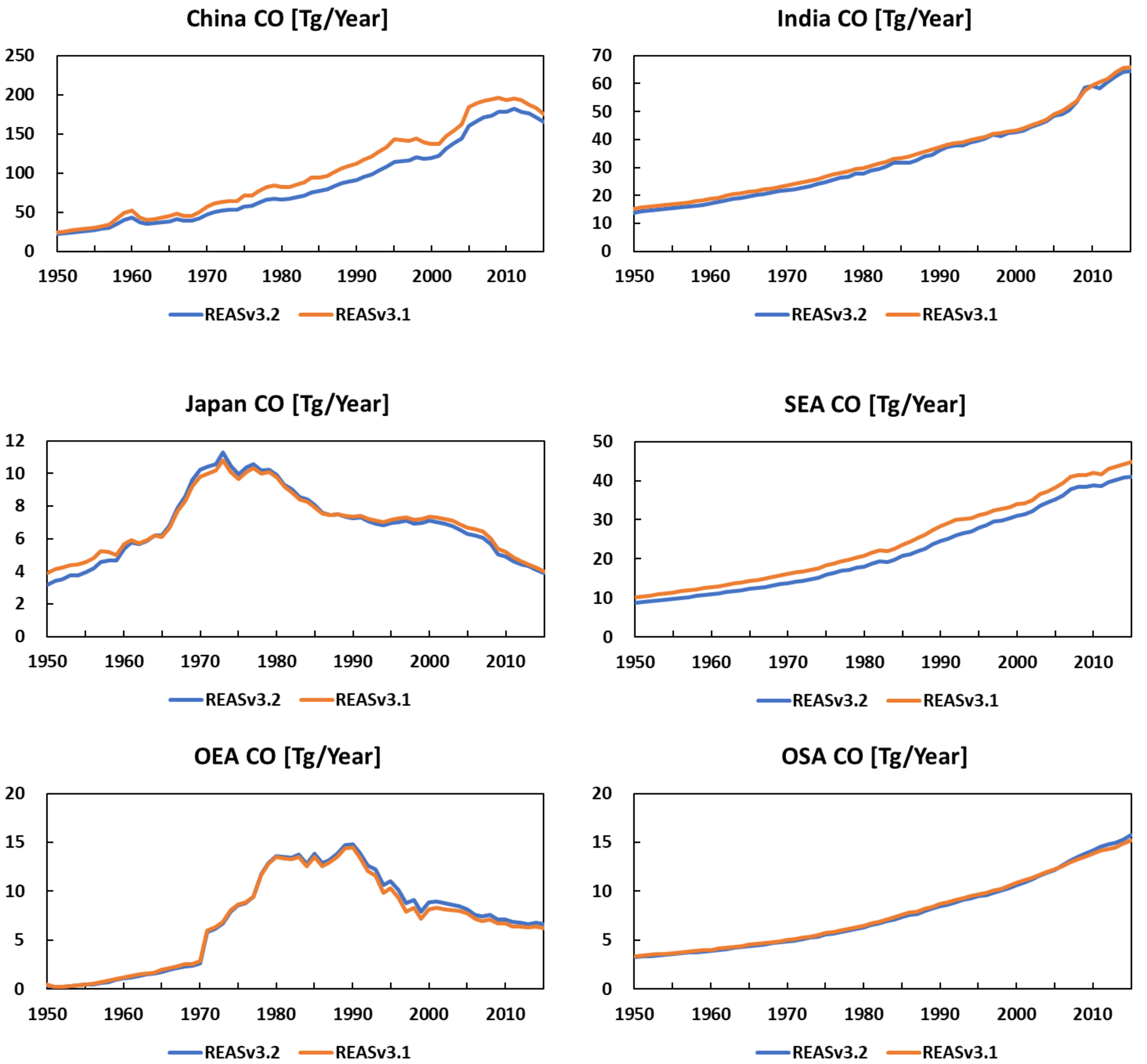


Figure 4. Differences of total CO emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

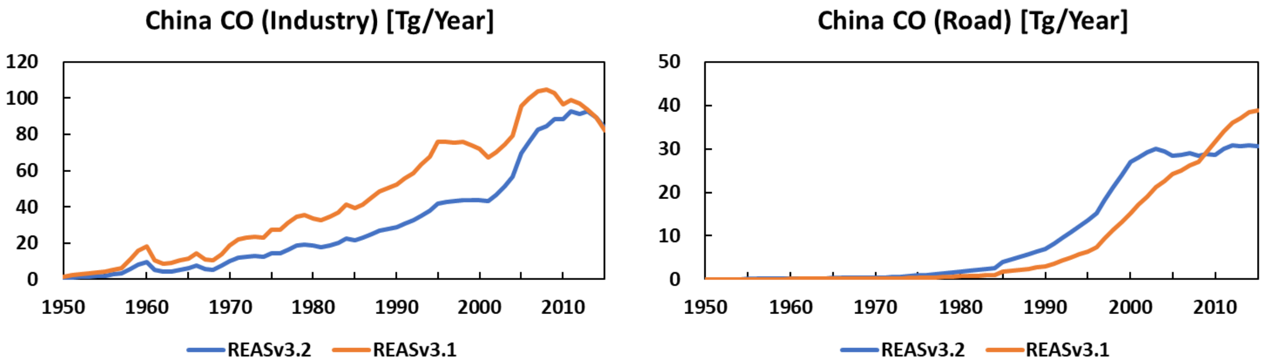


Figure 5. Differences of CO emissions from the industry and road transport sectors in China between REASv3.2 (blue line) and REASv3.1 (red line).

5. NMVOC

Figure 6 compares total NMVOC emissions in China, India, Japan, SEA, OEA, and OSA. Somewhat discrepancies were found in all countries and regions. Two major sources which caused the differences are biofuel combustion and exhaust emissions from road vehicles. In REASv3.2, settings of the emission factors for biofuel and those for road vehicles were revised from REASv3.1. For China, emissions from biofuel combustion in REASv3.2 were smaller than REASv3.1 and the differences to total emissions of REASv3.1 were 20%, 15%, 10%, and 4% in 1985, 1995, 2005, and 2015, respectively. On the other hand, for emissions from road transport, emissions of REASv3.2 were larger during late 1990s and late 2000s and then, after early 2010s, the values of REASv3.2 were smaller than REASv3.1. For India, Japan, and SEA, majority of differences between REASv3.2 and REASv3.1 were caused by revision of biofuel emission factors. Particularly for Japan, relatively large discrepancies before 1965 were mainly caused by revision of emission factors for charcoal combustion. For OEA, the majority of the differences were correction of errors in settings of emission factors for road transport. For OSA, the differences were mainly caused by revision of emission factor for biofuels, but after late 1990s, emissions from road vehicles in REASv3.2 became larger than REASv3.1 by revision of emission factors. Then, differences of total emissions in OSA were becoming smaller after 2005.

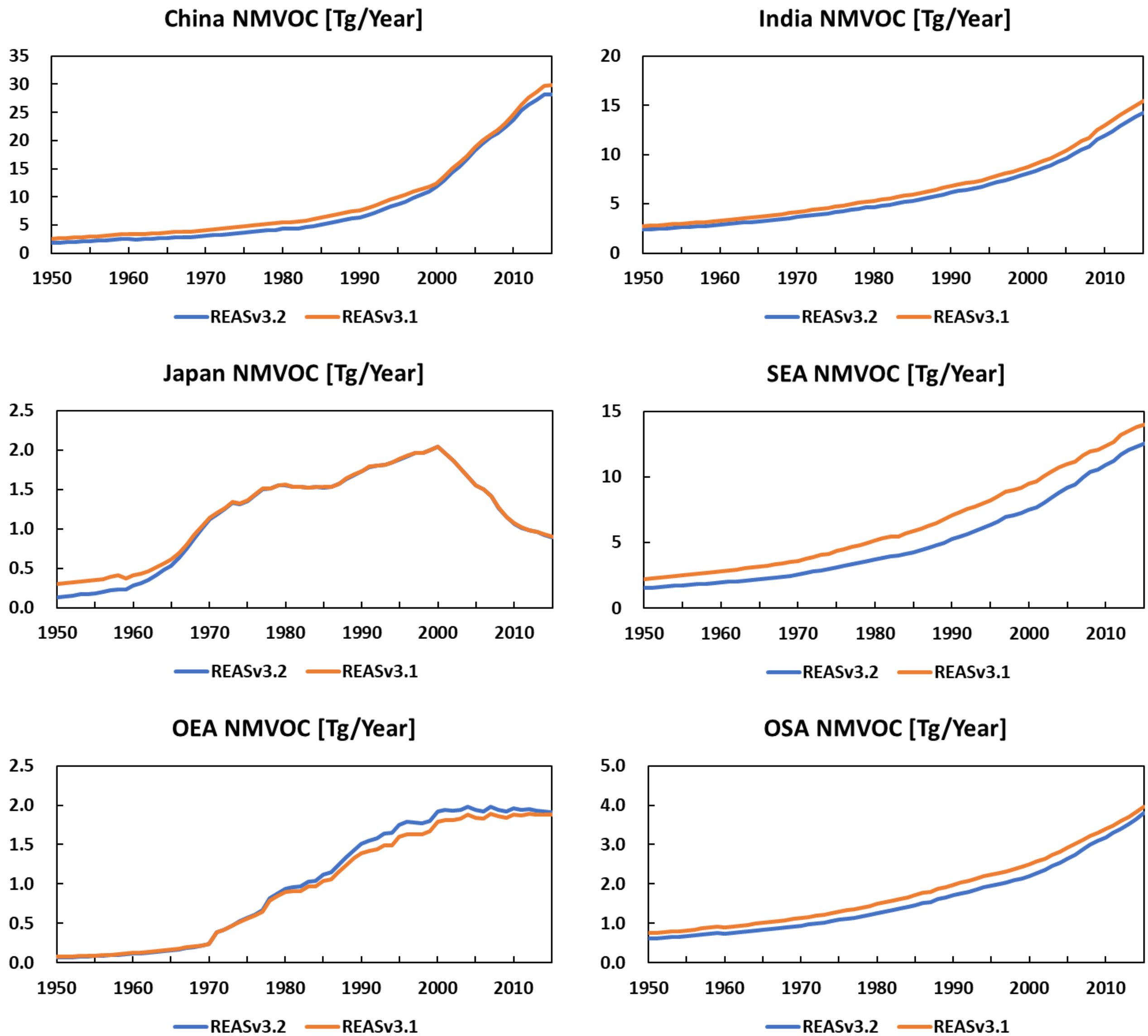


Figure 6. Differences of total NMVOC emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

6. NH_3

Figure 7 compares total NH_3 emissions in China, India, Japan, SEA, OEA, and OSA. For NH_3, differences between REASv3.2 and REASv3.1 were negligible for all countries and regions.

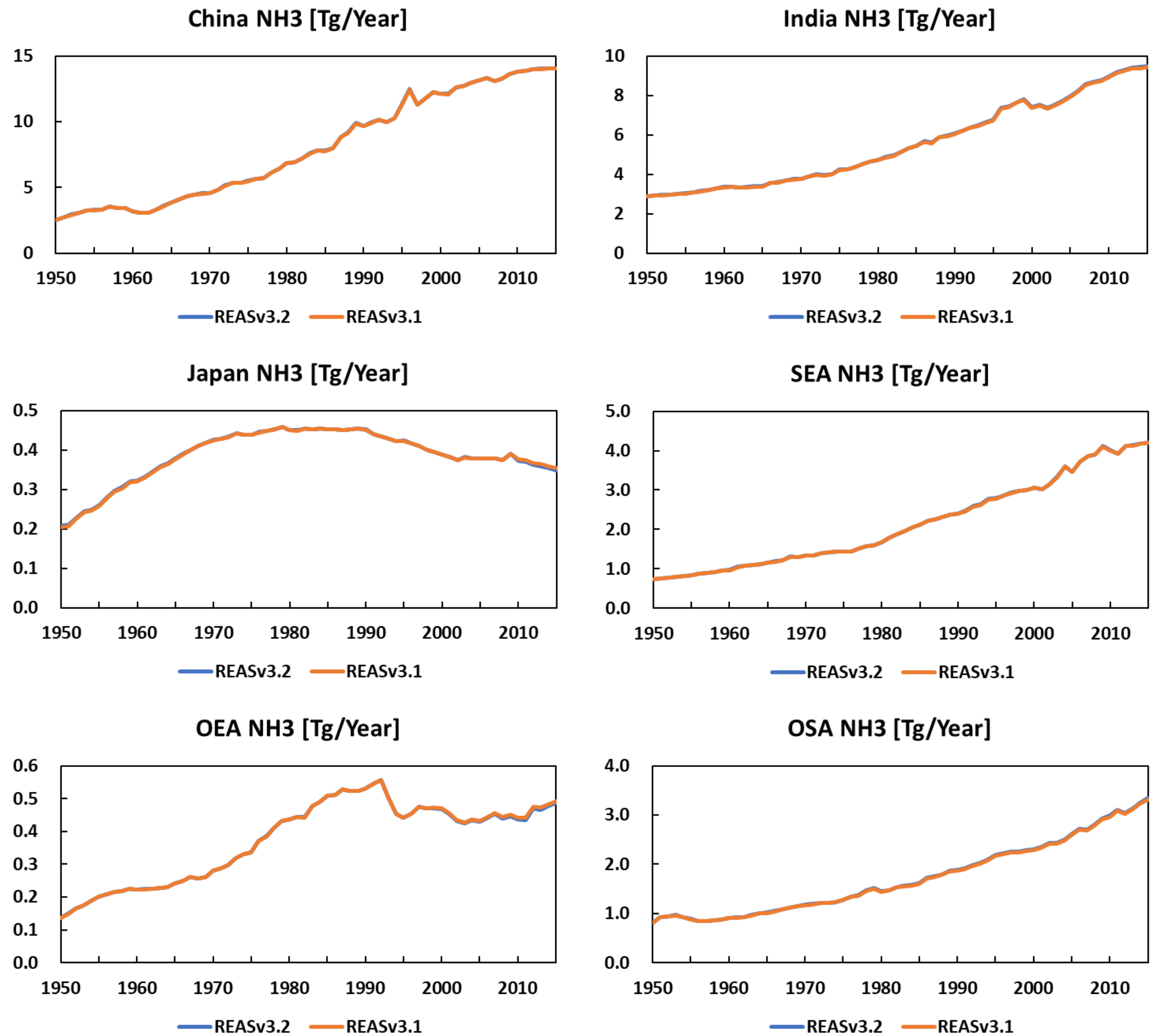


Figure 7. Differences of total NH_3 emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

7. CO_2

Figure 8 compares total CO_2 emissions in China, India, Japan, SEA, OEA, and OSA. For CO_2, differences between REASv3.2 and REASv3.1 were negligible for all countries and regions.

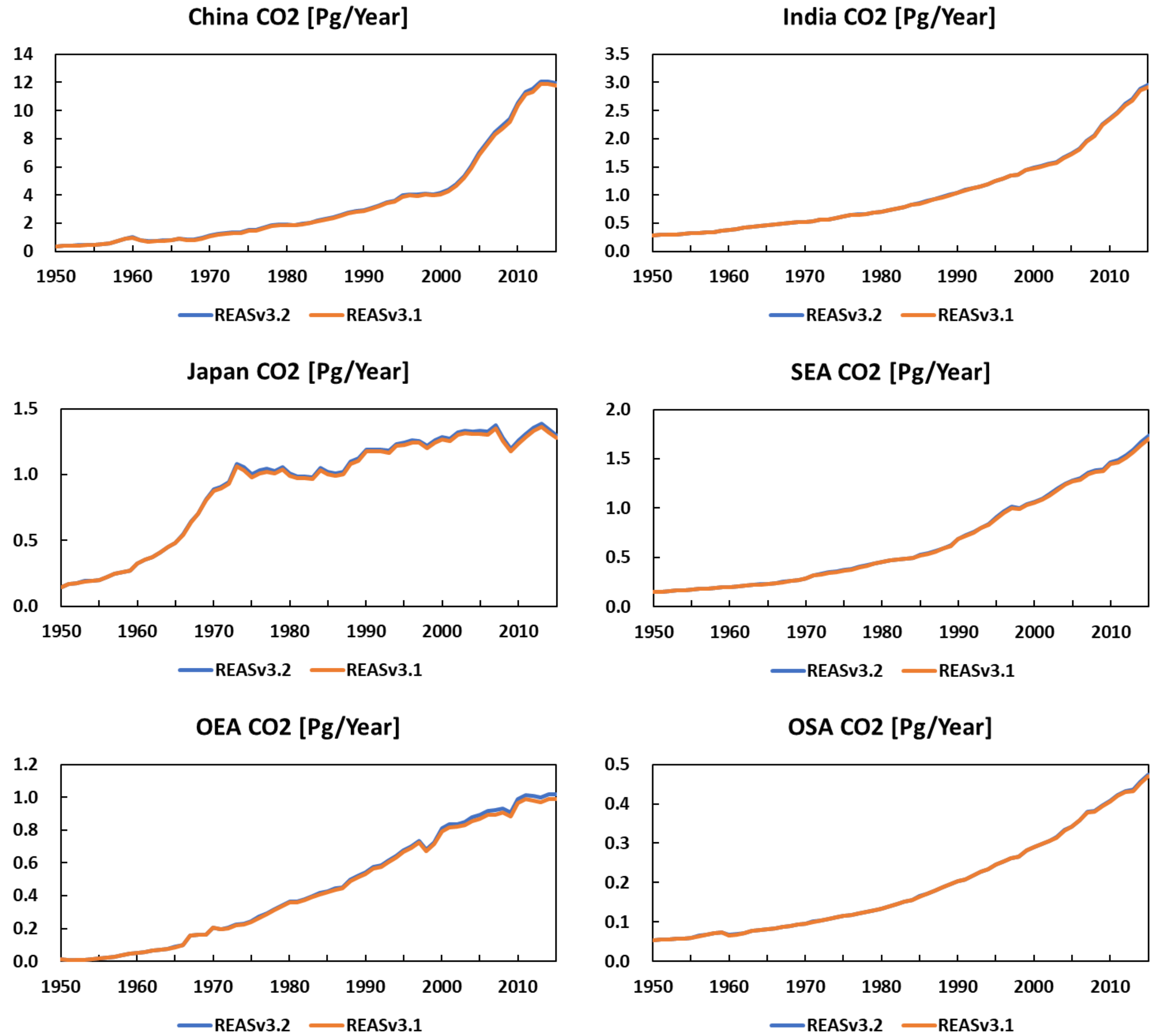


Figure 8. Differences of total CO_2 emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

8. PM_{10} and $PM_{2.5}$

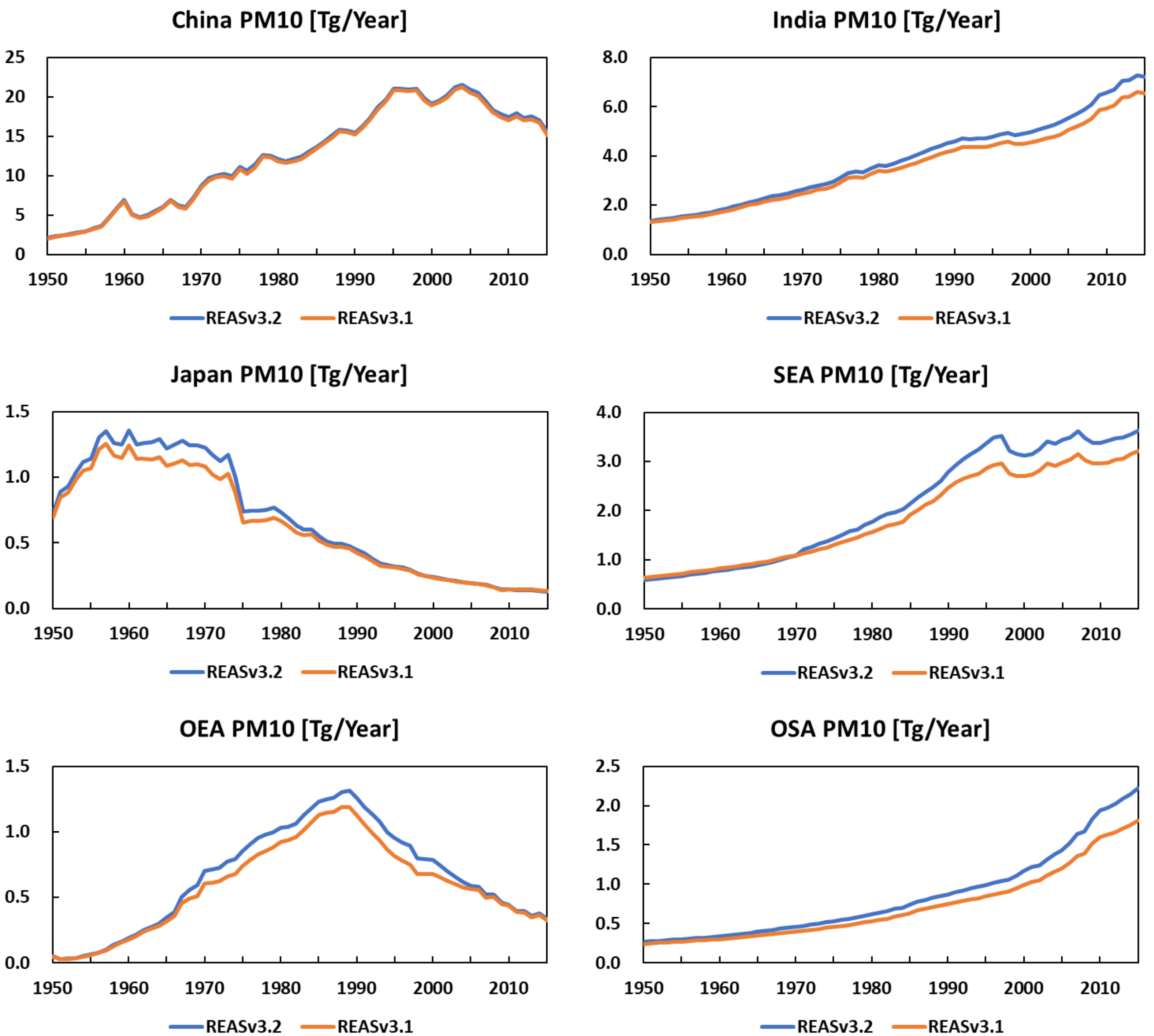
Figures 9 and 10 respectively compare total emissions of PM_{10} and $PM_{2.5}$ in China, India, Japan, SEA, OEA, and OSA. Differences between REASv3.2 and REASv3.1 were negligible for China, but for other countries and regions, there were clear discrepancies. For India, Japan, and OEA, relatively large differences appeared at different timings. Causes of the discrepancies were correction of emission factors for non-combustion emissions from cement production and oil refinery. The different timings depended on increasing effects of activity data and penetration rates of abatement equipment. For SEA and OSA, in addition to differences in the industry sector by the same causes for India, Japan, and OEA, emissions from biofuel combustion in REASv3.2 were smaller in SEA and larger in OSA. These differences were caused by correction of errors in biofuel data (settings of relative ratios of fuelwood, crop residue, and animal waste in primary solid biofuels).

9. BC

Figure 11 compares total emissions of BC in China, India, Japan, SEA, OEA, and OSA. Differences between REASv3.2 and REASv3.1 were negligible for China and Japan, but for other countries and regions, there were clear discrepancies. Emissions of REASv3.2 in India, SEA, and OSA were generally smaller than those of REASv3.1. These were mainly caused by revision of emission factors for biofuel combustion. On the other hand, for SEA, emissions from road transport were increased by correction of emission factors, but amounts of the differences were smaller than those from biofuel combustion. For OEA, emissions of REASv3.2 were smaller than REASv3.1 before 1990 and after 2000. The causes of these discrepancies were correction of emission factors for brown coal in the industry sector before 1990 and revision of emission factors for road vehicles after 2000.

10. OC

Figure 12 compares total emissions of OC in China, India, Japan, SEA, OEA, and OSA. Somewhat discrepancies were found in all countries and regions. For China, the differences were mainly caused by revision of emission factors for diesel oil combustion in the agricultural sub-sectors of the domestic sector. For India, Japan, SEA, and OSA, emissions of REASv3.2 were smaller than those of REASv3.1 due to revision of emission factors for biofuel combustion. The same as for NMVOC, the differences in Japan were mainly caused by revision of emission factors for charcoal combustion. For OEA, emissions of REASv3.2 were larger than REASv3.1 during 1970-2000 due to revision of emission factors for brown coal in the industry sector.



180

Figure 9. Differences of total PM_{10} emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

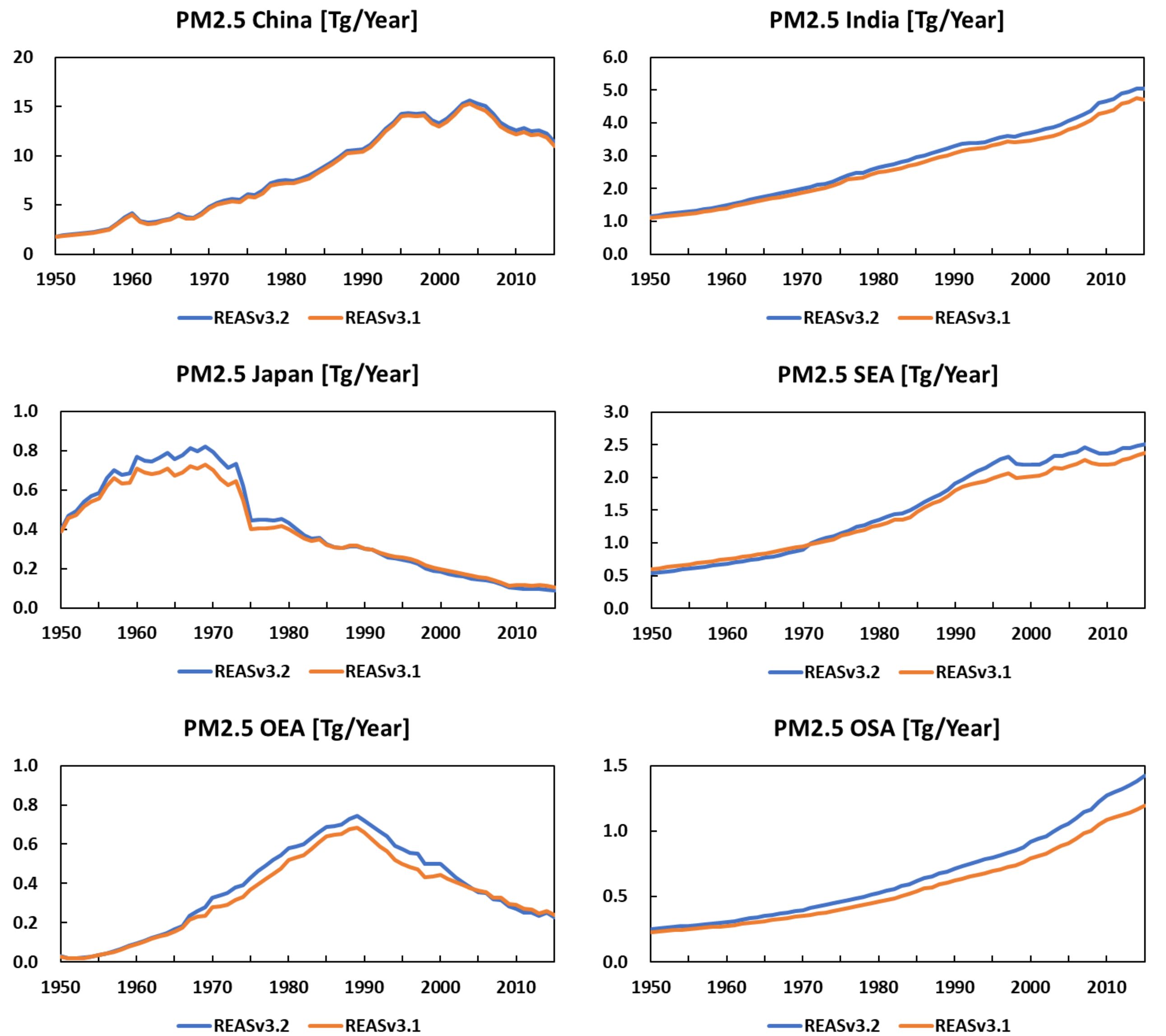


Figure 10. Differences of total $PM_{2.5}$ emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

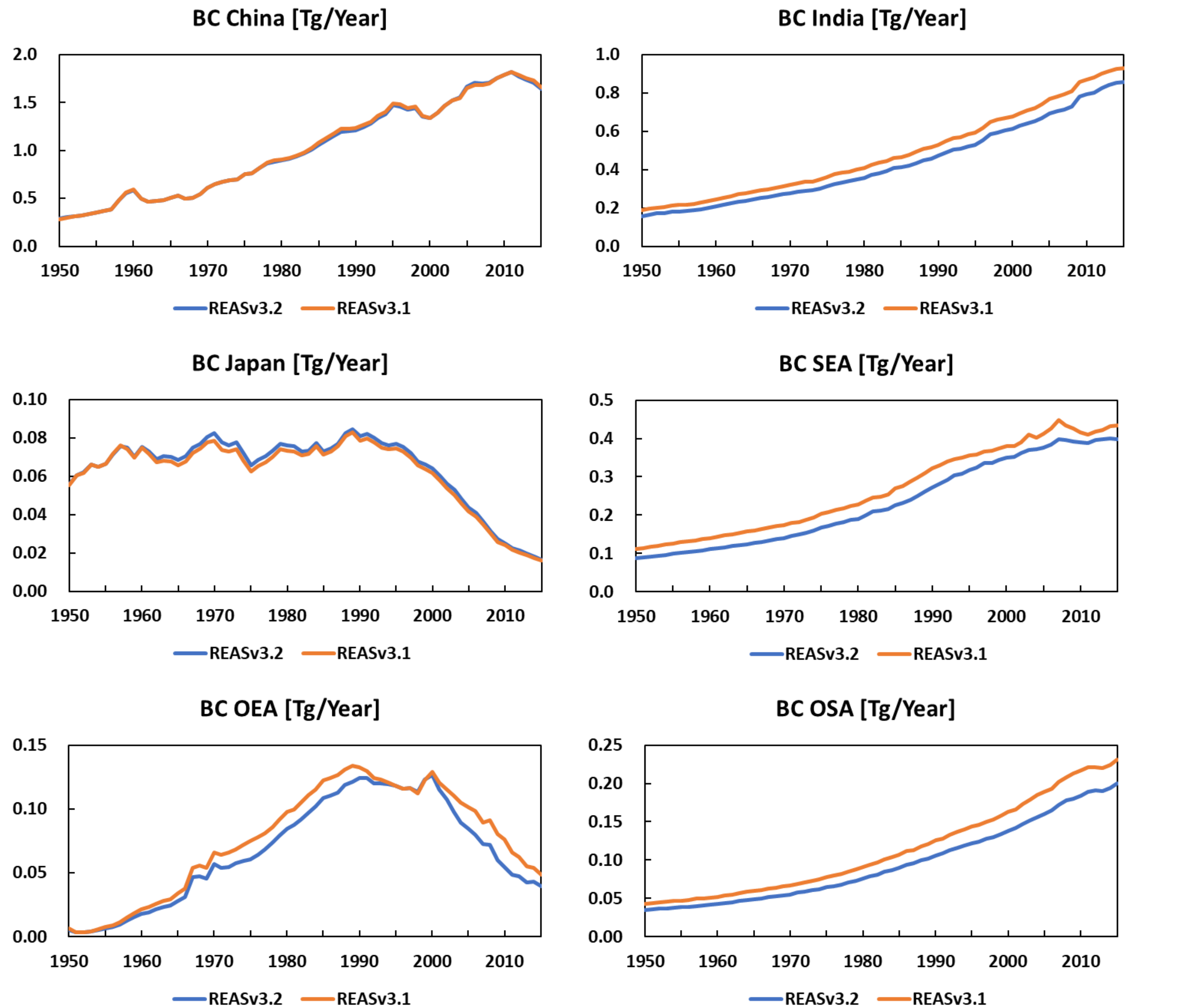


Figure 11. Differences of total BC emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

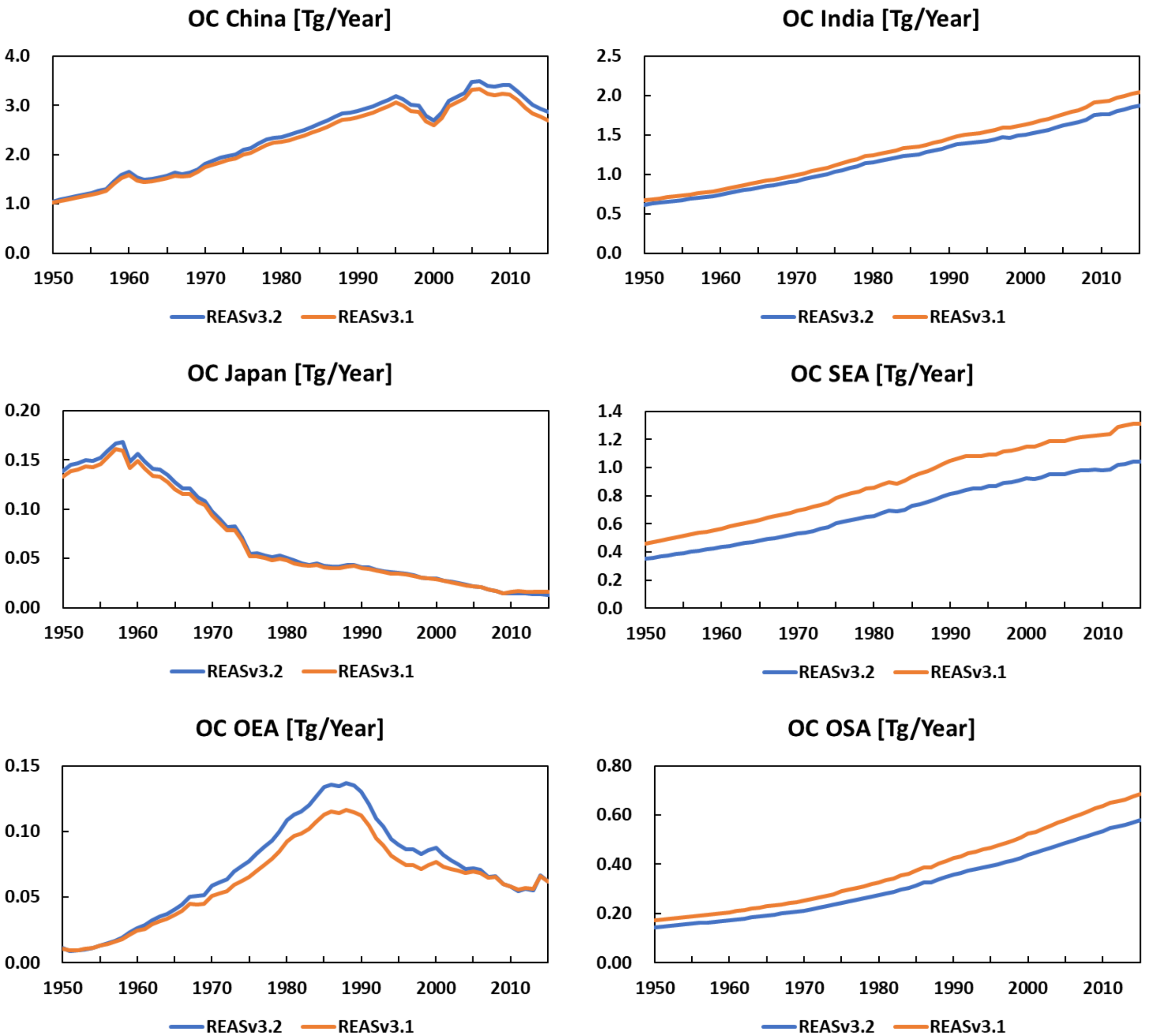


Figure 12. Differences of total OC emissions in China, India, Japan, SEA, OEA, and OSA between REASv3.2 (blue line) and REASv3.1 (red line).

References

Kurokawa, J. and Ohara, T.: Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3.1, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-1122.