

The authors would like to thank Anonymous Referee #1 for taking his or her time to review our manuscript and for giving very constructive and informative comments. These comments helped us improve the quality and clarity of the manuscript. We revised our manuscript based on them.

General Reply:

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.**

Below are our responses to each comment.

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.

Reply:

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 NH₃.
- In Sect. 2.4.2, Sect. S8.2 of the Supplement was cited for methodologies and data sources for fertilizer application sector for NH₃.
- 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, NH₃, 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.

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 NO_x, 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.

Reply:

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 NH₃, 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.

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.)

Reply:

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.

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.

Reply:

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.

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).

Reply:

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.

For road vehicles, the relationship between the "tentative emission factors" and final

emission factors in section 2.3.3 Emission factors is unclear.

Reply:

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.

One of the significant 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.)

Reply:

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.

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.

Reply:

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.

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.

Reply:

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.

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.

Reply:

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.

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

Reply:

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

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.

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 India 2005–2016: 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.: SO₂ emission estimates using OMI SO₂ 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 SO₂, 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.

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?).

Reply:

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 SO₂ became larger and those of CO₂ 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.