The authors would like to thank Anonymous Referee #2 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.

Major 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.

Reply:

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.

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.

Reply:

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 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.**

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.

Reply:

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.

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.

Reply:

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.**

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.

Reply:

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.

Minor comments:

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

Reply:

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

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

Reply:

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

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.

Reply:

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

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

Reply:

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

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.

Reply:

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.