Point to point responses to Reviewers

"Emission inventory of air pollutants and chemical speciation for specific anthropogenic sources based on local measurements in the Yangtze River Delta region, China" by Jingyu An et al.

Reviewer #1's comments:

1. *As mentioned above, my main concern is that the scope of the paper could be expanded a little bit, and would be focused more on the recent progress of method and data. That might be more helpful for the whole research community instead of local scientists. It invites more review on published work for the YRD region, and more comparison and discussion with other inventories. Some examples include: NMVOC emission estimation: As major source of NMVOC, there are many working procedures involved with VOC release for given type of chemical plant. Some more detailed methodologies were suggested and applied in the region. How did the authors evaluate the quality and feasibility of the more complicated methods for the region NH3 emission estimation: Similarly, different methods have been conducted for the region (Atmos. Chem. Phys., 20, 4275–4294, 2020), and how did the authors analyze the advantage of various methods? Some more examples include agricultural machines (Environmental Pollution 266 (2020) 115075). It might not be necessary for the authors to recalculate the emissions, but a more careful review and discussion for the method choice and further improvement should be sufficient.*

Re: Thanks for the comment. We agree with the comment that the method of emission inventory compilation is improving in recent years. In the revised manuscript, we supplemented some reviews and discussions in the introduction and method sections. Please see the following changes.

Changes in manuscript:

(1) **Section 1, lines 72-78:** We added the reviews on the recent progress of method and data and rewrote this section to be: "In the last five years, only individual province (Zhou et al., 2017) and some sources were updated with the progress of method and data. Fan et al. (2016) established a high-resolved ship emission inventory for 2010 base on Automatic Identification System (AIS) data over the YRD region and East China Sea. Huang et al. (2018a) developed a non-road machinery emission inventory for 2014 based on local surveys in the cities of YRD. Zhang et al. (2020) further developed a "grid-based" (30 × 30 m) inventory of agricultural machinery with daily emissions for 2015 by combining satellite data, land and soil information, and in-house investigation. Wang et al. (2018b) established an emission inventory

of civil aviation for landing take-off (LTO) cycles for 2017. Yang and Zhao (2019) estimated air pollutant emissions from open biomass burning for 2005–2015 using three (traditional bottom-up, fire radiative power (FRP), and constraining) approaches. Zhao et al. (2020) developed a NH_3 emission inventory for 2014 based on dynamic emission factors (EFs) and activity data integrating the local information of soil, meteorology, and agricultural processes. These studies provided novel methods for emission estimation and expanded our understanding on the emissions over the YRD region. However, with the implementation of air pollution prevention and control measures, $PM_{2.5}$ pollution in the YRD region has improved significantly in recent years as the region's energy, industry, and vehicle structures have been modified accordingly (Zheng et al., 2016; Wang et al., 2017a; Zhang et al., 2017a). A comprehensive update of activity levels and sources in the YRD region could assist with accurate air quality simulations and emission reduction measures."

New references:

Zhang, J., Liu, L., Zhao, Y., Li, H., Lian, Y., Zhang, Z., Huang, C., and Du, X.: Development of a high-resolution emission inventory of agricultural machinery with a novel methodology: A case study for Yangtze River Delta region, Environ. Pollut., 266, 115075, 2020.

Zhao, Y., Yuan, M., Huang, X., Chen, F., and Zhang, J.: Quantification and evaluation of atmospheric ammonia emissions with different methods: a case study for the Yangtze River Delta region, China, Atmos. Chem. Phys., 20, 4275-4294, 2020.

(2) **Section 2.3, line 152-155:** According to the reviewer's comment, we explained the segment-based method for large-point source emission estimation and discussed the advantages of this method. The specific modifications are as follows: "For large point sources, we established a segment-based emission estimation method based on local surveys. For example, we subdivided the ferrous metal manufacturing industry into raw material yard, iron making (including sintering, pelletizing, and blast furnace), steel making (including converter and electric furnace), casing steel, rolling steel, and ferroalloy production. The petroleum refining industry was subdivided into eight segments including process devices, equipment leak, storage tank, bulk loading, flare, wastewater treatment, cooling tower, and petrochemical furnace. The activity data and EFs of each segment were both derived from on-site surveys and measurements. Emissions from industrial solvent-use sources were calculated using the mass balance method based on the consumption and NMVOC content of solvents, such as paints, coatings, inks, adhesives, thinners, etc. Small amounts of NMVOC remaining in products, wastewater and waste were not

considered in this calculation. The solvent consumption and their VOC content of large point sources were mainly from field surveys and then extended to similar industries and solvent varieties."

(3) **Section 2.3, line 159-163:** We added some discussions on the difference between the methods in this study and other novel method published in previous studies. The specific modifications are as follows: "Non-road machinery emissions were estimated from the NONROAD model (US EPA, 2010), which was based on fuel consumption and fuel-based EFs. Fuel consumption was calculated from the population, working hours and fuel consumption rate per hour derived from local survey in typical cities like Shanghai and Hangzhou. The method was introduced in our previous study (Huang et al., 2018a). Limited by the data source, we haven't achieved a daily-resolved emission estimation of agricultural machinery introduced by Zhang et al. (2020), which may cause higher uncertainty on its total amount and temporal and spatial distribution."

2. *Another issue is the comparison between inventories. Different data and methods resulted in discrepancy in emission estimation, as well as the spatial distribution. The authors compared the emission levels of this work compared with the national inventory MEIC in particular, but how about some other information, like temporal and spatial distribution?*

Re: Thanks for the comment. In the revised manuscript, we supplemented some discussions on the spatial distribution. Considering the temporal distribution is not the focus of this study, we haven't covered too much. Please see the following changes.

Changes in manuscript:

(1) **Section 3.1.3:** We added a paragraph at the end of this section to compare the spatial distribution of our study with the MEIC and previous studies in the YRD region. Please see the details as follows: "Previous studies have shown that the unit-based bottom-up approach based on local activity data can improve the spatial distribution of emission inventories (Zhao et al., 2015; Zheng et al., 2017; Zhao et al., 2018; Zheng et al., 2019). The spatial distribution of major air pollutants obtained in this study is consistent to theother unit-based inventories based on local surveys. For example, the distribution of NMVOC emissions is consistent with that obtained from the on-site surveys in Jiangsu Province (Zhao et al., 2017); the distribution of NH3 emissions is also consistent with the results using dynamic emission factors and localized information (Zhao et al., 2020). Compared with the national-scale inventory like the MEIC, this study has improved the distribution along the Yangtze

River and Hangzhou Bay where large point sources were denser, and it also reduced the misjudgment of NOx and NMVOC emission hotspots in the northern and southern areas, as shown in Figure S1. The distribution of NH_3 emissions was also improved in the northern areas of the region and in the city centers with more localized EFs of mobile and agriculture sources."

New references:

Zhao, Y., Qiu, L. P., Xu, R. Y., Xie, F. J., Zhang, Q., Yu, Y. Y., Nielsen, C. P., Qin, H. X., Wang, H. K., Wu, X. C., Li, W. Q., and Zhang, J.: Advantages of a city-scale emission inventory for urban air quality research and policy: the case of Nanjing, a typical industrial city in the Yangtze River Delta, China, Atmos. Chem. Phys., 15, 12623–12644, 2015.

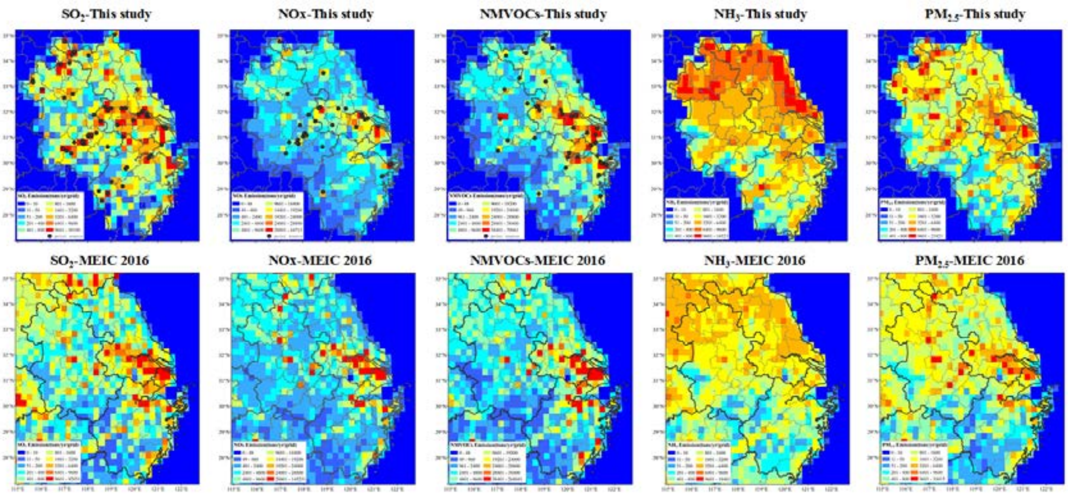

Figure S1. Comparisons of spatial distributions of SO_2, NO_x, NMVOCs, NH_3, and $PM_{2.5}$ emissions between this study and the MEIC. The black dots represent for large point sources.

Moreover, as the authors indicate in the introduction, the region experienced dramatic change for the past years, how did they evaluate the data for 2017 compared to earlier years?

Re: Thanks for the comment. We have added some discussions on the comparison of different years. Please see the following changes.

Changes in manuscript:

(1) **Section 3.1.1:** We added a paragraph at the end of this section to compare the results for 2017 with the earlier years. Please see the details as follows: "Compared with our previous inventory for 2014 (Li et al., 2019; Ni et al., 2020), SO_2, NO_x, PM_{10}, and $PM_{2.5}$ emissions in the YRD region have decreased by 47%, 15%, 20%, and

24%, respectively, which were consistent with the trends of regional air quality improvement (SO_2 44 %; NO_2 5%; PM_{10} 22%; $PM_{2.5}$ 27%). However, it should be noted that the approach of emission estimation in this study has made a number of localized corrections in terms of emission factors and activity data. For example, CO, NMVOC, and NH_3 emissions have increased significantly compared to 2014, which mainly because more point sources were included in this study and more localized EFs, which were generally higher than those in previous studies, were applied to estimate NO_x, CO, NMVOC, and NH_3 emissions from solvent-use, motor vehicles, non-road machinery, and agricultural sources. Next, it is necessary to estimate the emission inventories by the same approach for different years to evaluate the changes in air pollutant emissions in recent years."

New references:

Ni, Z., Luo, K., Gao, Y., Gao, X., Jiang, F., Huang, C., Fan, J., Fu, J., and Chen, C.: Spatial–temporal variations and process analysis of O_3 pollution in Hangzhou during the G20 summit, Atmos. Chem. Phys., 20, 5963–5976, 2020.

3. *Some data sources were unclear. For example, the environment statistics did not provide all the information used in the emission calculation for point sources. Did the authors make more on-site investigations or surveys?*

Re: Yes. As the reviewer mentioned, the environment statistics didn't provide all the information like the aftertreatment technologies and their efficiencies, especially for NMVOCs. The information was mainly obtained from on-site investigations in typical cities in the YRD region. In the revised manuscript, we supplemented some discussions. Please see the following changes.

Changes in manuscript:

(1) **Section 2.4, line 189:** We added some sentences to explain how we obtain other information on point sources. Please see the details as follows: "However, the database didn't include all the information like the technologies of NMVOC removal and their efficiencies, especially for the median and small-size factories. To obtain more detailed information, we have conducted more on-site investigations on the removal technologies and efficiencies of industrial sources in typical cities including Shanghai, Hangzhou, Suzhou, etc. According to the investigations, we classified the proportions of removal technologies and efficiencies to different industrial sectors and then extended them to the entire region."

Biomass burning was not reported, how did the authors estimate the activity data *“based on statistics” ?*

Re: Thanks for the comment. Biomass burning was reported in this study, but only the emissions from household biomass-fueled stoves were included. We have made some explanation in the revised manuscript. Please see the following changes.

Changes in manuscript:

(1) **Section 2.4, line 201:** We added some description on the estimation method of the activity data for biomass burning emissions in this study. Please see the details as follows: “The biomass burning emissions in this study only included the emissions from household biomass-fueled stoves. Their activity data was estimated based on the crop yields and grain straw ratios combined with the proportions of household burning in each city. The crop yields were obtained from the statistical yearbooks; the grain straw ratios and the proportions of household burning were derived from the surveys from agricultural department. In 2017, the average household burning ratio of various types of straw was about 12% (3%–16%), 3% in developed cities such as Shanghai; the highest ratios (16%) were in the cities of Anhui Province; and the ratios in other cities were about 12%.”

Could the authors also provide the emission contribution of point sources by species and region (province) ?

Re: Sure. We have uploaded the gridded emissions of air pollutants from various sources for the YRD region developed by this study at a horizontal resolution of 4 × 4km and a summary table of emissions by cities and sources. Please see the download link (https://doi.org/10.6084/m9.figshare.13340648) in the “Data availability” section.

4. *Please provide more information on the method of uncertainty analysis. How did the authors evaluate the bias of each category of parameters? At least the information needs to be given in the Supplement.*

Re: Thanks for the comment. We have supplemented some explanation on the method of uncertainty analysis in the revised manuscript. Please see the following changes.

Changes in manuscript:

(1) **Section 2.8:** To supplement more information the method of uncertainty analysis, the whole paragraph was rewritten to be: “Uncertainty was mainly derived from the activity data and EFs. The coefficients of variation of the activity data and EFs for each source were classified into seven grades in the range of 2%–100% using expert

judgment. The coefficient of variation for the activity data was determined according to the data source. Environmental statistical data with specific source information was assigned the lowest coefficient of uncertainty (2%), while activity data estimated from the statistical yearbooks, such as biomass burning, was assigned the highest uncertainty value (98%). The coefficients of uncertainty for other activity data sources were assigned to be 18%, 34%, 50%, 66%, and 82% in turn. The principle for assignments of the coefficients of variation for EFs was the same as the activity level. EFs derived from local measurements in the YRD region with large samples were assigned the lower coefficients of uncertainty (18%), while those from USEPA or EMEP/EEA datasets were assigned higher coefficients (98%). Then the uncertainty of each pollutant from each emission source can be combined by Eq. (3–5). A detailed description of the analytical methods used can be found in our previous study (Huang et al., 2011).

$$CV = \frac{U}{E} = 1.96 \times \sqrt{(1 + C_a^2) \times \left(1 + C_f^2\right) - 1} \quad (3)$$

$$U_j = \sqrt{\sum_k U_{j,k}^2} \quad (4)$$

$$U = \sqrt{\sum_j U_j^2} \quad (5)$$

where, *CV* is the coefficient of variation of the emission rate, *E* is the emission rate, *U* is the uncertainty of the emission source, C_a is the uncertainty of activity data, C_f is the uncertainty of EF, *j* and *k* represent for pollutant and emission source, respectively."

5. *It is great that the authors made detailed evaluation (validation might not be a proper word) with CMAQ modeling and provided the results in the supplement. However, the discussion in the main text seems descriptive. Could you be more specific on the reasons for the relatively big discrepancy due to emission data, and also suggest the possible direction for future improvement on emission estimation?*

Re: Thanks for the comment. We have added more discussions in section 3.3 "Model validation". Please see the following changes.

Changes in manuscript:

(1) **Section 3.3, line 512-527:** We rewrote the second paragraph in Section 3.3 to further describe the results of model validation Please see the details as follows: "Figure 8 shows a comparison of the simulated and observed concentrations for

SO_2, NO_2, $PM_{2.5}$, PM_{10}, O_3, and CO for cities in the YRD region in January and July 2017. The simulated concentration distribution of the different pollutants was consistent with the observed results, indicating that the updated EI generally reflected the distribution of air pollution sources in the YRD region. Comparatively, agreement between the simulated concentration distribution and the observed results for the cities in the central areas of the YRD region was stronger than cities of the northern and southern border areas. This was mainly because border areas were more susceptible to the effects of emissions from areas outside the region, which resulted in greater deviation of the simulation results. A statistical analysis of the hourly concentrations obtained from the model for the pollutants in each city can be found in Table S7 of the supporting information. Figure 9 shows the mean fractional error (MFE) and the mean fractional bias (MFB) between the simulated and observed daily average concentrations in the cities of the region. Overall, the MFB and MFE of simulation and observation results of all the pollutants in January and July were all within the criteria (MFB $\leq \pm 60\%$, MFE $\leq 75\%$) of model performance recommended by Boylan and Russell (2006), and most of them were with the performance goals (MFB $\leq \pm 30\%$, MFE $\leq 50\%$), which indicated that the EI in this study could reflect the air pollution in winter in the YRD region. In July, the MFB and MFE of O_3 and $PM_{2.5}$ model performance all fell within the criteria range. However, the simulation results of primary pollutants like SO_2, NO_2, PM_{10} and CO were somewhat underestimated. Especially for SO_2 and CO, nearly half of the cities had MFBs lower than -60%, and the cities with large deviations were mainly concentrated in peripheral areas of the YRD region (such as Huangshan, Chizhou, Xuancheng, Lishui, etc.). These cities generally had higher contributions of area emissions from residential and agriculture sources instead of large point industrial sources. The activity data of these sources usually had higher uncertainties and would easily cause the deviation of emission estimation. For example, the underestimation of the amount of residential coal combustion would undoubtedly lead to a severely low estimate of SO_2 and CO emissions. However, since $PM_{2.5}$ and O_3 pollution were more regional, their simulation results were less affected by insufficient local activity data in these cities. Conducting more detailed on-site investigations to obtain more accurate activity data is the key to further improving the performance of EI in the future."

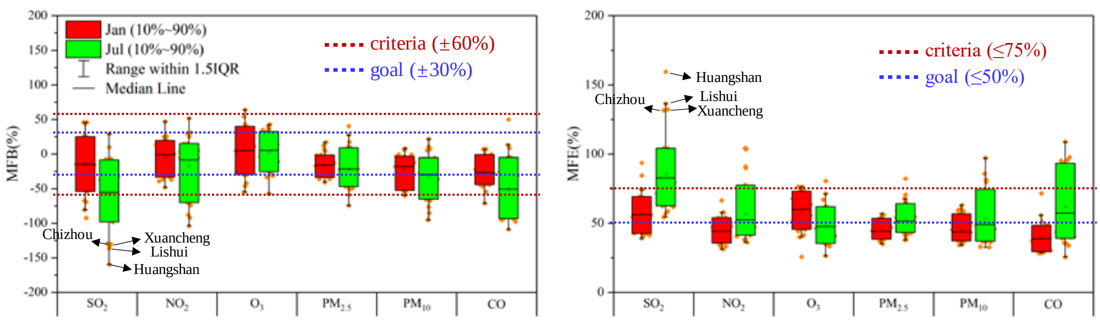

Figure 9. MFB (a) and MFE (b) between the simulation and observation data for daily average concentrations of various pollutants of the cities in the YRD region in January and July 2017

New references:

Boylan, J. W., and Russell, A. G.: PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models, Atmos. Environ., 40, 4946-4959, 2006.

Emery, C., Liu, Z., Russell, A. G., Odman, M. T., Yarwood, G., and Kumar, N.: Recommendations on statistics and benchmarks to assess photochemical model performance, J. Air Waste Manag. Assoc., 67, 582-598, 2017.

6. *Language should be improved as well. Some English expression is not correct.*

Re: Thanks for the comment. The grammar and vocabulary in the manuscript has been polished by a native speaker. Please see the modifications in the revised manuscript.

Reviewer #2's comments:

1. *Method. Since this study updates an emission inventory developed by the same group before, the method section should put more focus on the new features of the updated emission inventory compared to the last version. 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 this paper. In my opinion, only an update of emission inventory for another year without any novel method or data source cannot be published as a research article in ACP.*

Re: Thanks for the comment. In the revised manuscript, we added a comparison with the methods and data sources in our previous study and provided a detailed table to show the differences in this study. Please see the following changes.

Changes in manuscript:

(1) **Section 2.3, line 512-527:** We added a paragraph at the end of this section to compare the methods and data sources in this study with our previous study. Please see the details as follows: "The emission estimation method of this study has been improved on the basis of our previous study (the latest version was for 2014) (Li et al., 2019; Ni et al., 2020). Table 1 shows the differences between the methods and data sources of this study and the previous. First, the source category has been refined from the third-level 135 categories to the fourth-level 2812 categories. Among them, large point sources such as iron & steel and petroleum refining sectors were further subdivided into different emission segments. Secondly, in addition to

the environmental statistics data, the activity data has been refined through local investigations on the removal technologies and efficiencies, operating hours, and working conditions of industrial and mobile sources including motor vehicles and non-road machinery; emissions from ships and aircrafts, which were not considered in our previous study, were estimated based on dynamic activity data like AIS provided by local department. In terms of the EFs, most of them were corrected based on local measurements."

Table 1. Comparison of the methods and data sources in this study with our previous study.

Methods/Data sources	This study	Our previous study
Source classification	2812 source categories, subdivided into four levels, detailed to emission segments for large point sources	135 source categories, subdivided into three levels
Activity data		
Stationary combustion sources	Based on environmental statistics	Based on environmental statistics
Industrial process sources	Based on environmental statistics and local investigation on removal technologies and efficiencies	Based on environmental statistics
Industrial solvent-use sources	Based on environmental statistics and local investigation on solvent types and consumption	Based on environmental statistics
Motor vehicles	Based on city statistics and local activity surveys	Based on city statistics and local activity surveys
Non-road machinery	Based on city statistics and local activity surveys	Not considered
Ships	Based on AIS data	Not considered
Aviation aircraft	Based on LTO cycles from department surveys	Not considered
Dust sources	Estimated based on city statistics	Estimated based on city statistics
Oil storage and transportation sources	Based on city statistics	Based on city statistics
Residential sources	Based on city statistics	Based on city statistics
Waste treatment and disposal sources	Based on city statistics	Based on city statistics
Livestock and poultry breeding	Based on city statistics	Based on city statistics
N-fertilizer application	Based on city statistics	Based on city statistics
Biomass burning	Estimated based on city statistics	Estimated based on city statistics
EFs		
Stationary combustion sources	Based on literature surveys	Based on literature surveys
Industrial process sources	Updated the EFs for major segments of iron & steel and petroleum refining sectors based on local measurements	Based on literature surveys

Industrial solvent-use sources	Estimated by solvent contents of different solvent types from local investigations	Based on literature surveys
Motor vehicles	IVE model corrected by local measurements	IVE model
Non-road machinery	NONROAD model corrected by local measurements	Not considered
Ships	Based on local measurements	Not considered
Aviation aircraft	Recommended by ICAO	Not considered
Dust sources	Based on literature surveys	Based on literature surveys
Oil storage and transportation sources	Estimated based on local investigations	Based on literature surveys
Residential sources	Based on local investigations and measurements	Based on literature surveys
Waste treatment and disposal sources	Based on literature surveys	Based on literature surveys
Livestock and poultry breeding	Based on local measurements	Based on literature surveys
N-fertilizer application	Based on local measurements	Based on literature surveys
Biomass burning	Based on local measurements	Based on literature surveys

The method section lacks the descriptions of WRF-CMAQ model configurations and the estimation methods of OFP and SOAP.

Re: Thanks for the comment. In the revised manuscript, we supplemented the description of the WRF-CMAQ model configurations and the estimation methods of OFP and SOAP in Section 2.9 “model configurations” and Section 2.10 “Estimation of O_3 and SOA formation potentials”. Please see the following changes.

Changes in manuscript:

(1) **Section 2.9:** “To verify the reliability of the EI, we used CMAQ (version 5.3) to simulate the concentrations of SO_2, NO_2, $PM_{2.5}$, PM_{10}, O_3, and CO in the YRD region for January and July 2017, and compared these with the observation data for each city in the region. The meteorological field for the CMAQ model was obtained from the WRF (version 3). The EI developed in this study was then used to produce an emission system for the YRD region while emissions beyond the YRD were obtained from the MEIC 2016. The anthropogenic data was then combined with biogenic data obtained from the Model for Emissions of Gases and Aerosol from Nature modelling system (version 2.10) as the final input for the EI of the model. Figure S1 and Table S6 show the domain and settings for the model system. Detailed information is provided in Section 6 of the Supporting information.”

(2) **Section 2.10:** “To characterize the regional O_3 and SOA formation contributions of different NMVOC species and their sources, we used the O_3 formation potential (OFP) and SOA formation potential (SOAP) methods of estimation. OFP and SOAP were obtained

from the sum of the individual NMVOC species emissions multiplied by the maximum incremental reactivity (MIR) and SOA yield, respectively. MIR and SOA yield for individual NMVOC species were obtained from previous studies (Carter, 1994; Wu and Xie, 2017). The estimation methods were shown in Eq. (6) and (7).

$$OFP_i = \sum_{j=1} E_{i,j} \times MIR_j \tag{6}$$

$$SOAP_i = \sum_{j=1} E_{i,j} \times Y_j \tag{7}$$

where, OFP_i and $SOAP_i$ are the ozone formation potential and SOA formation potential of source *i*, respectively, $E_{i,j}$ is the VOC emission of species *i*, MIR_j is the maximum increment reactivity for the *j*th chemical species, Y_j is the SOA yield for the *j*th chemical species."

2. *Result. The manuscript in its current format just briefly describes the new inventory by source sector but does not provide any discussions on the improvement of the new emission inventory. Figure 8 only shows a map of modelled air pollutant concentrations with observation stations on it. It is difficult to say the simulated results are consistent with observed values. Table S7 provides statistical results of model performance in each city, which should be included in the main text using a few figures. And the evaluation part in the main text should be rewritten accordingly.*

Re: Thanks for the comment. To discuss on the improvement of the new emission inventory, we rewrote the Section 3.1.1 in the revised manuscript. In addition, we have supplemented more discussions in section 3.3 "Model validation". Please see the following changes.

Changes in manuscript:

(1) **Section 3.1.1:** "Compared with our previous inventory for 2014 (Li et al., 2019; Ni et al., 2020), SO_2, NO_x, PM_{10}, and $PM_{2.5}$ emissions in the YRD region have decreased by 47%, 15%, 20%, and 24%, respectively, which were consistent with the trends of regional air quality improvement (SO_2 44 %; NO_2 5%; PM_{10} 22%; $PM_{2.5}$ 27%). However, it should be noted that the approach of emission estimation in this study has made a number of localized corrections in terms of emission factors and activity data. For example, CO, NMVOC, and NH_3 emissions have increased significantly compared to 2014, which mainly because more point sources were included in this study and more localized EFs, which were generally higher than those in previous studies, were applied to estimate NO_x, CO, NMVOC, and NH_3 emissions from solvent-use, motor vehicles, non-road machinery, and agricultural sources. Next, it is necessary to estimate the emission inventories by the same approach for

different years to evaluate the changes in air pollutant emissions in recent years."

(2) **Section 3.3, line 512-527:** We rewrote the second paragraph in Section 3.3 to further describe the results of model validation Please see the details as follows: "Figure 8 shows a comparison of the simulated and observed concentrations for SO_2, NO_2, $PM_{2.5}$, PM_{10}, O_3, and CO for cities in the YRD region in January and July 2017. The simulated concentration distribution of the different pollutants was consistent with the observed results, indicating that the updated EI generally reflected the distribution of air pollution sources in the YRD region. Comparatively, agreement between the simulated concentration distribution and the observed results for the cities in the central areas of the YRD region was stronger than cities of the northern and southern border areas. This was mainly because border areas were more susceptible to the effects of emissions from areas outside the region, which resulted in greater deviation of the simulation results. A statistical analysis of the hourly concentrations obtained from the model for the pollutants in each city can be found in Table S7 of the supporting information. Figure 9 shows the mean fractional error (MFE) and the mean fractional bias (MFB) between the simulated and observed daily average concentrations in the cities of the region. Overall, the MFB and MFE of simulation and observation results of all the pollutants in January and July were all within the criteria (MFB $\leq \pm 60\%$, MFE $\leq 75\%$) of model performance recommended by Boylan and Russell (2006), and most of them were with the performance goals (MFB $\leq \pm 30\%$, MFE $\leq 50\%$), which indicated that the EI in this study could reflect the air pollution in winter in the YRD region. In July, the MFB and MFE of O_3 and $PM_{2.5}$ model performance all fell within the criteria range. However, the simulation results of primary pollutants like SO_2, NO_2, PM_{10} and CO were somewhat underestimated. Especially for SO_2 and CO, nearly half of the cities had MFBs lower than -60%, and the cities with large deviations were mainly concentrated in peripheral areas of the YRD region (such as Huangshan, Chizhou, Xuancheng, Lishui, etc.). These cities generally had higher contributions of area emissions from residential and agriculture sources instead of large point industrial sources. The activity data of these sources usually had higher uncertainties and would easily cause the deviation of emission estimation. For example, the underestimation of the amount of residential coal combustion would undoubtedly lead to a severely low estimate of SO_2 and CO emissions. However, since $PM_{2.5}$ and O_3 pollution were more regional, their simulation results were less affected by insufficient local activity data in these cities. Conducting more detailed on-site investigations to obtain more accurate activity data is the key to further improving the performance of EI in the future."

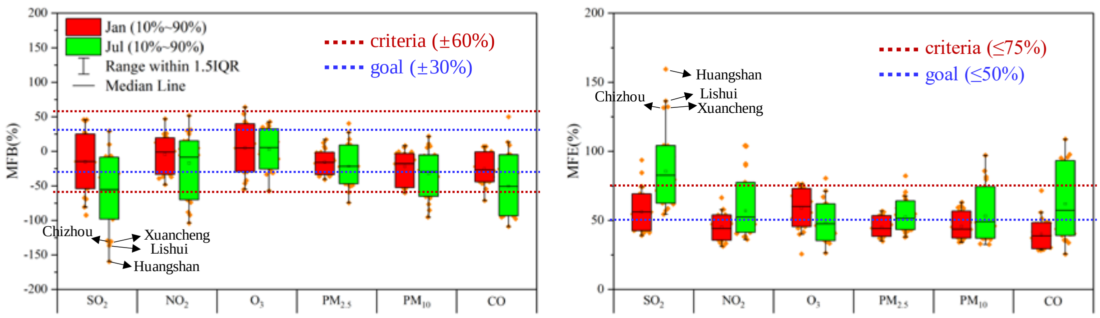

Figure 9. MFB (a) and MFE (b) between the simulation and observation data for daily average concentrations of various pollutants of the cities in the YRD region in January and July 2017

New references:

Boylan, J. W., and Russell, A. G.: PM and light extinction model performance metrics, goals, and criteria for three-dimensional air quality models, Atmos. Environ., 40, 4946-4959, 2006.

Emery, C., Liu, Z., Russell, A. G., Odman, M. T., Yarwood, G., and Kumar, N.: Recommendations on statistics and benchmarks to assess photochemical model performance, J. Air Waste Manag. Assoc., 67, 582-598, 2017.

3. *Data availability. Emission inventories are an important input to air quality models. Can a way for people to access the gridded emissions data by source sector that be included in the manuscript? This will allow other researchers to replicate and build on the modeling results if they wish. The authors now only provided gridded maps of total emissions, which are not enough to drive an air quality model. I suggest that the authors upload gridded emission maps by source sector at a regular spatial and temporal resolution, and also provide summary tables of emissions by city and source (i.e., emissions by source in each city).*

Re: Sure. We have uploaded the gridded emissions of air pollutants from various sources for the YRD region developed by this study at a horizontal resolution of 4 × 4km and a summary table of emissions by cities and sources. Please see the download link (https://doi.org/10.6084/m9.figshare.13340648) in the “Data availability” section.