Point to point responses to Reviewers

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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.

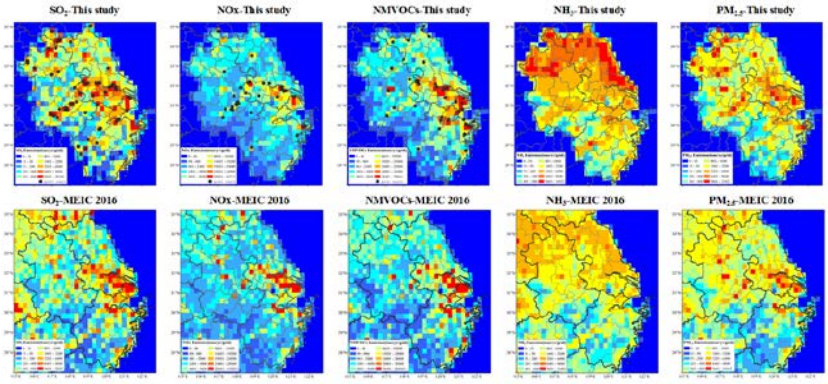

Figure S1. Comparisons of spatial distributions of SO_2, NO_x, NMVOCs, NH_3, and $PM_{2.5}$ emissions between this study and the MEIC. The black dots represent for large point sources.

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).

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

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

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

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."

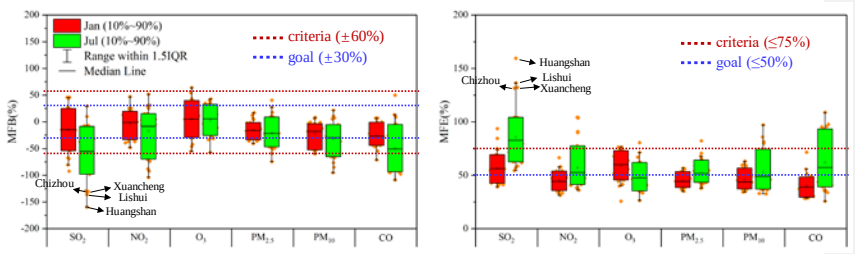

Figure 9. MFB (a) and MFE (b) between the simulation and observation data for daily average concentrations of various pollutants of the cities in the YRD region in January and July 2017

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).

$$OFP_i = \sum_{j=1} E_{i,j} \times MIR_j \quad (6)$$

$$SOAP_i = \sum_{j=1} E_{i,j} \times Y_j \quad (7)$$

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 $\leqslant$ ±60%, MFE $\leqslant$ 75%) of model performance recommended by Boylan and Russell (2006), and most of them were with the performance goals (MFB $\leqslant$ ±30%, MFE $\leqslant$ 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."

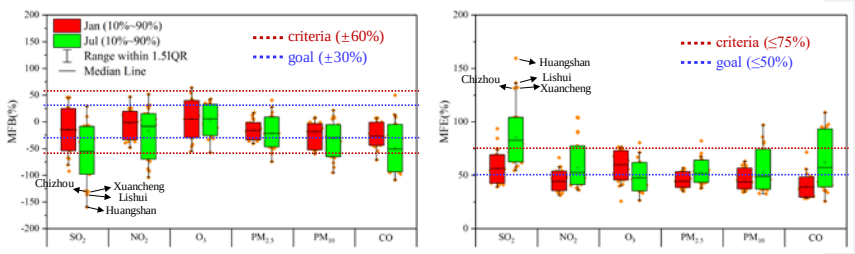

Figure 9. MFB (a) and MFE (b) between the simulation and observation data for daily average concentrations of various pollutants of the cities in the YRD region in January and July 2017

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.

Emission inventory of air pollutants and chemical speciation for specific anthropogenic sources based on local measurements in the Yangtze River Delta region, China

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Abstract: A high-resolution air pollutant emission inventory ~~in~~ for the Yangtze River Delta (YRD) region was updated for ~~the year~~ 2017 using emission factors and chemical speciation based ~~from~~ on local measurements in this study. The inventory ~~includes~~ included 424 non-methane volatile organic compounds (NMVOC) ~~species~~ and 43 fine particulate matter ($PM_{2.5}$) species~~, which can be subdivided~~ from ~~into~~ 259 specific source~~s categories~~. The total emissions of SO_2, NO_x, CO, NMVOCs, PM_{10}, $PM_{2.5}$, and NH_3 in the YRD region in 2017 ~~are~~ were 1,552, 3,235, 38,507, 4,875, 3,770, 1,597, and 2,467 Gg~~,~~ respectively. SO_2 and CO emissions ~~are~~ were mainly from boilers, accounting for 49% and 73% of the total~~, respectively~~. Mobile sources dominated ~~the~~ NO_x emissions ~~and contribute~~contributing 57% ~~of~~ to the total. NMVOC emissions, mainly ~~come~~ from industrial sources, ~~occupying~~ made up 61% of the total. Dust sources ~~take up~~accounted for ~~to~~ 55% and 28% of PM_{10} and $PM_{2.5}$ emissions~~,~~ respectively.

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Agricultural sources accounted for 91% of NH_3 emissions. Major $PM_{2.5}$ species ~~are~~ were OC, Ca, Si, PSO_4 and EC, accounting for 9.0%, 7.0%, 6.4%, 4.6% and 4.3% of total $PM_{2.5}$ emissions respectively. The main species of NMVOCs ~~are~~ were aromatic hydrocarbons, ~~accounting~~ making up~~for~~ 25.3% of the total. Oxygenated volatile organic compounds (OVOCs) contributed 21.9% ~~of~~ to the total NMVOC emissions. Toluene ~~has~~ had the highest comprehensive contribution to ozone (O_3) and secondary organic aerosol (SOA) formation potentials, ~~and~~ while ~~the~~ other~~s~~ NMVOCs included ~~are~~ 1,2,4-trimethylbenzene, m,p-xylene, propylene, ethene, o-xylene, ethylbenzene ~~and so on~~. Industrial process and solvent use sources ~~are~~ were the main sources of O_3~~ozone~~ and SOA formation potential, followed by motor vehicles. Among industrial sources, chemical manufacturing, rubber & plastic manufacturing, appliance manufacturing and textile ~~have~~ made ~~relatively outstanding~~significant contributions. This emission inventory ~~can~~ should provide scientific guidance for future ~~joint~~ control of air pollutants in the YRD region of~~,~~ China.

Key words: emission inventory; $PM_{2.5}$ species; NMVOC species; the Yangtze River Delta region; air pollutant emissions

1. Introduction

Air pollutant emissions from anthropogenic sources have attracted ~~wide~~ considerable attention~~s~~ due to their adverse impact~~s~~ on air quality (Monks et al., 2009), human health (Guan et al., 2016; Requia et al., 2018), and climate change (Fiore et al., 2012). Air pollutants include gaseous compounds~~,~~ such as sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), ammonia (NH_3)~~, etc.~~, and ~~particles~~ particulate matter (PM) defined by ~~with different sizes including PM_{10} and $PM_{2.5}$, whose aerodynamic~~ diameters that are generally less than 10 μm, e.g. PM_{10} and $PM_{2.5}$ μm. ~~NMVOCs and $PM_{2.5}$ are aggregates of various chemical compositions.~~ NMVOCs ~~contains~~ are a large and diverse group of organic compounds,~~thousands of species~~ such as alkanes, alkenes, aromatic hydrocarbons and oxygenated volatile organic compounds (OVOCs)~~,~~. They

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are ~~and is the~~ key precursors of ozone (O_3) and secondary organic aerosols (SOA). $PM_{2.5}$ are complex aggregates ~~is~~ composed of ~~a complex mixture, including~~ e.g. sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic carbon (OC), elemental carbon (EC), and various elements~~,~~. They ~~which~~ can degrade~~s~~ atmospheric visibility and ~~threaten~~ adversely affect ~~public~~ human health (Qiao et al., 2014; Liang et al., 2016).

The ~~E~~emission inventory (EI) is ~~a key fundamental~~an important tool for air pollution source apportionment, air quality forecasting, and decision-making ~~of~~ in air ~~pollutant~~ pollution control ~~measures~~. In the last two decades, ~~emission inventories~~EI have ~~been~~ improved ~~both in~~ on both global and regional scales. According to ~~recently reported inventories~~recent EI, anthropogenic emissions ~~still show growing~~have shown increasing trends ~~in~~ on a global scale (Janssens-Maenhout et al., 2015; Klimont et al., 2017; Crippa et al., 2018; Hoesly et al., 2018). China's air pollutant ~~emission intensity~~EI ~~is~~ were at a higher level ~~in~~ compared with the rest of the world due to ~~the~~ increasing energy consumption, urbanization and ~~motorization~~vehicle population. However, China's emissions are undergoing dramatic changes, especially in key regions~~,~~ such as the Jing-Jin-Ji ~~(JJJ)~~, Yangtze River Delta (YRD), and Pearl River Delta ~~(PRD) regions~~, ~~with~~ which have seen the ~~efforts~~ introduction of air pollution prevention and control measures in ~~these~~ recent years (Cai et al., 2018; Zheng et al., 2018). Consequently, ~~U~~updating the national EI has become ~~very necessary~~an important requirement.

The YRD region ~~is located~~ in ~~Eastern~~ eastern China, which ~~and covers~~ includes areas of Jiangsu, Zhejiang, Anhui, and Shanghai, accounts for a significant proportion of China's population and gross domestic product (GDP). ~~which has the most intensive~~A rapidly growing economy~~,~~ and increased urbanization ~~population and transportation and results~~ have resulted in ~~its~~ the highest emission levels in China for this region. ~~According to the new released d~~Data recently released by the Multi-resolution EI for China (MEIC, http://meicmodel.org/)~~,~~ ~~the~~ showed that emissions ~~intensities~~EI ~~per unit area~~ of SO_2, NO_x, NMVOCs, $PM_{2.5}$, and NH_3, per unit area in the YRD region, ~~are~~ were 2.3, 4.5, 5.2, 3.4, and 3.0 times, respectively, that of the national

average. To understand the emission levels for the YRD region, Huang et al., (2011)~~We have~~ established an EI for the core cities ~~in the YRD region~~ in 2007 ~~(Huang et al., 2011).~~; ~~After that,~~ Fu et al. (2013) updated the EI for Jiangsu, Zhejiang, and Shanghai ~~in the YRD region~~ in 2010~~.~~ and refined the inventory of $PM_{2.5}$ and NMVOC speciation; ~~In~~ in the last five years, only individual province~~s~~ (Zhou et al., 2017) ~~or part of~~ and some sources were updated ~~based on~~with the ~~recent~~ 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 ~~the YRD region in~~ 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.~~(Fan et al., 2016; Zhou et al., 2017; Huang et al., 2018a; Wang et al., 2018b; Chen et al., 2019; Yang and Zhao, 2019).~~ These studies provided novel methods for emission estimation and expanded our understanding on the emissions over the YRD region. However, with ~~Due to~~ the implementation of air pollution prevention and control measures, $PM_{2.5}$ pollution in the YRD region has ~~been~~ improved significantly in recent years~~alleviated, and~~ as the ~~regional~~ region's energy, industry, and vehicle structures ~~fleet are~~ have ~~undergoing great changes~~been modified accordingly ~~in recent years~~ (Zheng et al., 2016; Wang et al., 2017a; Zhang et al., 2017a). A comprehensive ~~Updating~~update of ~~the~~ activity levels ~~for~~and ~~detailed~~ sources in the YRD region ~~can~~ could ~~help to~~assist with accurate ~~simulate~~ air quality simulations and ~~guide~~ emission

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reduction measures ~~more accurately~~.

~~Besides of~~In addition to activity levels, speciation profiles of $PM_{2.5}$ and NMVOC emissions are ~~also very important to~~required to improve the performance of chemical transport models (CTMs) ~~in simulating~~that simulate O_3 mixing ratios and $PM_{2.5}$ concentrations. Source profiles from USEPA~~'s~~ repository of organic gas and PM speciation profiles of air pollution sources (SPECIATE) ~~database has been~~were commonly used ~~to conduct~~ for source apportionment and create speciated EI for air quality modeling since the 1990s (USEPA, 2009; Simon et al., 2010). However, the emission characteristics of anthropogenic sources ~~have~~ can vary ~~considerable~~ considerably ~~difference~~ between different regions. Differences in fuel properties, operating conditions, raw materials, and after-treatment techniques can result in inconsistent speciation profiles for $PM_{2.5}$ and NMVOCs. A previous study ~~indicates~~ indicated that us~~e~~ing of ~~the~~ speciation profiles from the SPECIATE database ~~leads to~~gave relatively poor model performance for trace elements at an urban site in Beijing, China (Ying et al., 2018). The emission estimates for individual NMVOC species ~~differ~~ varied ~~between~~ by one ~~and~~ to three orders of magnitude for some species when different sets of speciation profiles ~~are~~ were used, which ~~will~~ could ~~lead to~~result in significant deviations in the outputs of O_3 and SOA simulations (Li et al., 2014; Zhao et al., 2017; Stroud et al., 2018; Wang et al., 2018c). ~~In view of its importance to model performance~~Consequently, detailed and observation-based emissions of individual speciated $PM_{2.5}$ and NMVOCs have become ~~critical~~essential for accurate CTM.

In this study, we updated an anthropogenic air pollutant EI ~~in the~~for the 2017 YRD region ~~for the year of 2017~~ using ~~the emission factors (~~EFs~~)~~ and $PM_{2.5}$ and NMVOCs speciation profiles mainly derived from local measurements. The pollutants include~~d~~: SO_2, NO_x, CO, NMVOCs, PM_{10}, $PM_{2.5}$, and NH_3~~.~~; ~~In addition,~~the NMVOC comprised 424 individual ~~NMVOC~~ species ~~including~~ such as alkanes, alkenes, aromatic~~s~~ hydrocarbons, haloalkanes~~,~~ and OVOCs; ~~and~~ ~~and~~ 43 $PM_{2.5}$ species ~~including~~containing organic carbon (OC), elemental carbon (EC), ions, and elements

~~were included in the inventory~~. To obtain detailed ~~sources of~~ emissions sources, the EI was refined into ~~259 specific source categories in 4~~ four ~~levels~~categories, broadly based on ~~the~~ fuel types, industrial sectors, equipment types, and emission levels, to give a total of ~~etc~~259 specific emission sources. Finally, the EI was validated using the Community Multiscale Air Quality (CMAQ) modeling system and observations from 2017 in the YRD region ~~in 2017~~.

2. Materials and methods

2.1 Study ~~D~~domain ~~of this study~~

The YRD region in this study covers the three provinces~~,~~ of ~~including~~ Jiangsu, Zhejiang, and Anhui ~~provinces~~, as well as Shanghai municipality. The region has a land area of approximately 350,400 km^2, accounting for 3.7% of the whole of China. ~~However, the whole region~~The ~~produced a gross domestic product (~~GDP~~)~~ for the region was ~~of~~ 2,893 billion USD in 2017, i.e. ~~about~~ ~24% of the total national GDP in ~~2017~~ that year~~,~~ and was growing at a rate of ~~about~~ ~9.3% per year in the last decade (NBSC, 2018). Correspondingly, the region consumed 717.8 million ton coal equivalents~~tce~~ of energy which was~~,~~ ~~about~~ ~17% of the national total in 2017. Coal is the main energy type in ~~this region~~the YRD, contributing ~~about~~ ~60% of the total energy consumption (NBSC, 2018). The automobile population reached 40.9 million in 2017, ~~occupying~~ accounting for 19.6% of the total in China. The region also has a high concentration of traditional industries, producing 13.9%, 11.3%, 9.0%, 18.2% and 19.1% of the total ~~products~~ production of gasoline, diesel, coke, cement, and crude steel respectively in China in 2017 (NBSC, 2018). Figure 1 shows the domain of the YRD region in this study. The coastal waters within the dashed line on the ~~right figure~~expanded image of the region are China's ship emission control areas. The ship emissions ~~mentioned~~ used in this study ~~are~~ were ~~the~~ a combination ~~summary~~ of emissions ~~in~~ from this region and the inland waters ~~in~~ of the YRD region.

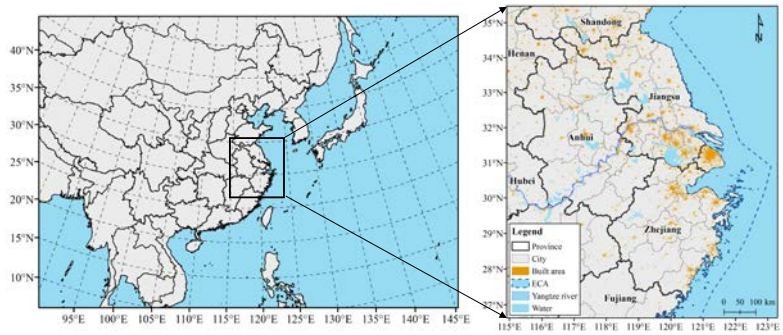

Figure 1. The domain of the YRD region in this study. Coastal waters within the dashed line on the expanded image represent ship emission control areas.

2.2 Sources classification

~~A total of 241 categories of e~~The EI~~mission sources~~ was separated into four~~in 4 levels~~main categories ~~were divided in this study~~. The first ~~level~~ category ~~is~~ was sub-divided into ~~9~~ nine major sources~~,~~ including, stationary combustion ~~sources~~, industrial process ~~sources~~, industrial solvent-use ~~sources~~, mobile ~~sources~~, dust ~~sources~~, oil storage and transportation ~~sources~~, residential ~~sources~~, waste treatment and disposal ~~sources~~, and agricultural ~~sources~~. The second ~~level~~ category ~~has~~ comprised ~~a total of~~ 36 source types ~~categories~~, mainly based on combustion facilities and the industrial, transportation, residential, and agriculture sectors. The third ~~level~~ category comprised 127 sources ~~classification~~ classified~~is~~ mainly ~~based on~~by fuel, product and material types~~, and contains a total of 127 categories~~. The fourth ~~level~~ category included sources ~~classification includes~~by combustion types, emission segments, and control levels. The ~~D~~detailed classification is shown in Table S1 ~~in~~ of the supporting information.

2.3 Emission estimation methods

The emissions of SO_2, PM_{10}, and $PM_{2.5}$ from stationary combustion sources ~~are~~ were calculated using the mass balance method ~~by~~ according to Eq. (1) and (2). Other pollutant emissions ~~are~~ were calculated using the EF method, ~~as shown in~~given by Eq. (3).

$$E_{SO_2} = 2 \times S \times F \times C_s \times \left(1 - \eta_{SO_2}\right) \quad (1)$$

$$E_{PM} = A \times F \times C_A \times P_{ratio} \times (1 - \eta_{PM}) \quad (2)$$

$$E_{i,j} = AL_j \times EF_{i,j} \times \left(1 - \eta_{i,j}\right) \times 10^{-3} \quad (3)$$

Where~~,~~: E_{SO2} and E_{PM} represent the emissions of SO_2 and PM_{10} or $PM_{2.5}$ (t)~~,~~; S and A represent fuel sulfur content and ash in fuel (%)~~,~~; F is the fuel consumption (t)~~,~~; Cs and C_A are the conversion efficiencies from sulfur and ash to SO_2 and PM (%)~~,~~ P_{ratio} is the mass percentage of PM_{10} or $PM_{2.5}$ in total PM~~,~~; η_{SO2} and η_{PM} represent the removal efficiency of SO_2 and PM_{10} or $PM_{2.5}$~~,~~; $E_{i,j}$ represents the emissions of pollutant i from source j (t)~~,~~ AL_j is the activity data of source j, such as fuel consumption, product output, and raw material consumption~~, etc.~~; $EF_{i,j}$ is the EF of pollutant i from source j (kg per activity data)~~,~~; $H_{i,j}$ is the removal efficiency of pollutant i from source j.

Emissions from the industrial process sources ~~are~~ were calculated using the EF method shown in Eq. (3). 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 ~~are~~ were calculated using the mass balance method based on the consumption and NMVOC content of solvents, such as paints, coatings, inks, adhesives, thinners, etc. ~~A~~ ~~s~~Small amounts of NMVOC remaining in products, wastewater and waste ~~was~~ were not considered in this calculation. The solvent consumption~~s~~ and their VOC content of large point sources were mainly from field surveys and ~~are~~then extended to similar industries and solvent varieties.

For motor vehicles, ~~we use~~ the International Vehicle Emission (IVE) model was

used to calculate the emissions. However, the EFs and activity data ~~including~~ for driving conditions, fleet composition, vehicle mileage travel~~s~~ed (VMT), and meteorological parameters in the model were ~~localized~~ restricted to~~via~~ real-world measurements and surveys ~~in this study~~. Non-road machinery emissions ~~are~~ were estimated ~~with reference to the~~from the NONROAD model (USEPA, 2010), which ~~is~~ was based on ~~the~~ fuel consumption and fuel-based ~~emission factor~~EFs. ~~The amount of f~~Fuel consumption ~~is~~ was calculated ~~based on~~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. Ship emissions ~~are~~ were estimated according to the method of Fan et al. (2016) using ~~the~~ an approach based on the ~~Automatic Identification System (~~AIS~~)~~ data~~. The detailed method has been reported by Fan et al. (2016)~~. The ~~C~~civil aviation aircraft source ~~refers to aircraft~~included emissions ~~under the~~defined by the International Civil Aviation Organization (ICAO) for ~~landing take-off (~~LTO~~)~~ cycles, which include~~d four operating modes, like~~ approach~~ing~~, taxi~~ng~~, ~~taking off~~take-off, and climb~~ing~~. SO_2 emission from the civil aviation aircraft source ~~is~~ was estimated using the mass balance method. The sulfur content in aviation fuel ~~is~~ of 0.068%~~, which is~~ was the default value provided in a previous study (Wayson et al., 2009). NO_x, CO, and NMVOC emissions ~~are~~ were estimated using the EF method~~,~~ as~~which~~ the product of~~multiplied the~~ fuel consumption rate ~~by the~~and EFs. PM emission ~~is~~ was calculated using the first-order approximation (FOA3.0) method (Wayson et al., 2009). The rated thrust and working hours of the aircraft in each LTO ~~mode~~ cycle ~~are~~ were referenced ~~to~~ from the ~~recommended~~ parameters recommended by ~~the International Civil Aviation Organization (~~ICAO~~)~~. The climbing mode specified by ~~the~~ ICAO ~~refers~~ referred ~~to the~~an altitude of ~~about~~ ~1 km from the end of take-off to the top of the atmospheric boundary layer. However, since the height of boundary

layer ~~in the actual atmosphere will change with the~~was dependent on meteorological conditions~~.~~, ~~In this study, a~~ The Weather Research and Forecast ~~meteorological model~~ (WRF~~ v3.9.1~~) Model (version 3.9.1) was used to simulate the boundary layer height ~~to~~ and correct the ~~time~~duration of the climbing mode. A ~~D~~detailed description of the method~~ology~~ used for aviation emission estimation is provided in our previous study (Wang et al., 2018b).

Emissions from ~~the~~ other sources (dust~~ source~~, oil storage and transportation ~~source~~, residential~~ source~~, waste treatment and disposal~~ source~~, and agricultural~~ source~~) ~~are~~ were all calculated using the EF method.

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

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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

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Biomass burning	Based on local measurements	Based on literature surveys

2.4 Activity data sources

The aActivity data related to the industrial sources (including e.g. stationary combustion sources, industrial process and solvent-use sources) of this study arewere mainly from the 2017 Environmental Statistics Database, which contains contained the information on fuel consumption, product output, raw material consumption, and removal technology and efficiencies. There are were nearly ~30,000 major point sources in for the YRD region in the database. 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. Considering thatAs environmental statistics do not include all industrial sources, we take derived an area source using the difference between the total fuel consumption and product output in the statistical yearbook and the sum of environmental statistics for each city as an area source. To improve the accuracy of mobile source emissions, a number ofseveral local activity surveys on the activity data (such ase.g. population, vehicle or machine type, fuel type, and emission standard, etc.) were conducted for motor vehicles, non-road machinery, and civil aviation aircrafts. The activity data of ships come was obtained from the AIS data for the East China Sea in 2017. The activity data of area sources are were derived from the statistical yearbooks of cities in the YRD region. For the sSources whose of activity data are not recorded in the statistical yearbooks (such ase.g. the number of construction sites, civil solvent usage, catering, biomass burning, etc.) were, we make some estimations estimated based onfrom statistical data, such as population, building area, and crop yields, etc. 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%. Table S2 in the support information summarizes the emission estimation methods, activity data sources and their data reliability.

2.5 Determination of ~~emission factor~~EFs

The EFs of each specific emission source were determined by local measurements (or surveys) in the YRD region, domestic EI guidebook of China (MEP, 2014), and those ~~recommended~~ given in USEPA~~'s~~ Compilation of Air Pollutant Emissions Factors ~~AP-42~~ (AP-42; USEPA, 2002) and Joint European Monitoring and Evaluation Programme (EMP)/European Environment Agency ~~European's~~ (E~~M~~EA~~P~~) datasets (EEA, 2013) ~~in turn~~respectively. To minimize ~~the~~ uncertainty ~~of~~ in the EI, this study ~~localizes~~ localized the EFs ~~of~~ from 80 source categories~~,~~. This ~~which~~ included the majority of anthropogenic emission sources, such as coal-fired power plants and boilers (Yao et al., 2009; Zhao et al., 2010; Wang et al., 2011; Lou, 2014; Sun, 2015; Xu et al., 2018), petroleum refining and ferrous metal manufacturing (Guo et al., 2017), gasoline and diesel vehicles (Huang et al., 2016; Huang et al., 2017; Huang et al., 2018b; Huang et al., 2018c), non-road machinery (Fu et al., 2012; Fu et al., 2013; Ge et al., 2013; Qu et al., 2015; Li et al., 2016), ~~and~~ emissions from cooking (Wang et al., 2018a; Gao et al., 2019), livestock and poultry breeding (Chen, 2017; Zhou, 2019), N-fertilizer application (Chen et al., 2017; Xia et al., 2018), and biomass burning (Tang et al., 2014)~~, etc~~. ~~The~~ NMVOC EFs for some evaporation loss sources, such as~~, like~~ industrial and residential solvent-use ~~sources and~~, oil storage, and transportation ~~sources~~, ~~are~~ were estimated ~~based on~~from the results of field surveys of ~~some~~ typical sources in the YRD

region. For ~~the~~ sources that could not be~~have not been~~ measured or investigated, the EFs were obtained in the following order of preference:~~recommended in the~~ EI guidebook of China (MEP, 2014)~~> are preferred, followed by the recommended factors in the~~ USEPA~~'s~~ AP-42 (USEPA, 2002)~~> and European's~~ EMEP/EEA datasets (EEA, 2013). The EFs for each ~~emission~~ source and their references are provided in Table S1 ~~in~~ of the supporting information.

2.6 $PM_{2.5}$ and NMVOC speciation

$PM_{2.5}$ and NMVOC emissions ~~are~~ were converted into profiles of~~further split into~~ individual species to simulate $PM_{2.5}$ chemical components and O_3 mixing ratios in the atmosphere. The profile for $PM_{2.5}$ comprised~~There are~~ 43 chemical species ~~in $PM_{2.5}$~~, including OC, EC, particulate sulfate (PSO_4), nitrate (PNO_3), and ammonium (PNH_4) and 36 elemental components such as Na, Mg, K, Ca, Al, and Si, etc. Additional species such as particulate ~~water (H_2O)~~, noncarbon organic matter ~~(NCOM)~~, metal bound oxygen ~~(MO)~~, and other unspeciated $PM_{2.5}$ ~~(PMO)~~ ~~are~~ were calculated according to the method ~~introduced by~~of Reff et al. (2009). The profile for NMVOC ~~There are 424 species of VOCs,~~ includ~~ed~~ing 96 alkanes, 45 alkenes and alkynes, 44 aromatic hydrocarbons, 164 OVOCs, 43 haloalkanes, and 32 other organic compounds.

The method ~~for~~ used to ~~determining~~ determine the $PM_{2.5}$ and NMVOC source profiles ~~is similar to~~followed that used for the EFs and~~.~~ ~~The results of~~data was preferentially selected as follows: local measurements ~~are prioritized in this study, followed by domestic~~ >measurements from ~~in~~ previous domestic studies~~, and finally the~~ >USEPA~~'s~~ SPECIATE 4.4 database (Hsu et al., 2014). To ~~enhance~~ improve the representativeness of source profiles in the inventory, the $PM_{2.5}$ chemical compositions ~~of~~ from 34 sources and the NMVOC chemical compositions ~~of~~ from 64 sources were localized according to the measurements in the YRD region. ~~The s~~Source categories ~~of~~ used for~~localization~~ the localized ~~for~~ $PM_{2.5}$ profiles included power plants, coal-fired boilers, ferrous metal manufacturing, gasoline and diesel vehicles, non-road machinery, ships, catering, and biomass burning~~, etc.~~ (Zheng et al., 2013; Tang et al., 2014; Huang

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et al., 2016; Xu et al., 2018). The ~~localized NMVOC~~ source categories~~s~~ used for the localized NMVOC profiles included coal combustion, gasoline and diesel vehicles, ships, catering, biomass burning, and the majority of industrial process and solvent-use sources, ~~like~~ such as petroleum refining, coke production, chemical manufacturing, textile, furniture manufacturing, package and printing, auto manufacturing, shipbuilding, and architectural coating~~, etc.~~ (Wang et al., 2014a; Wang et al., 2014b; Wang et al., 2016; Wang et al., 2017b; Wang et al., 2017c; Huang et al., 2018d; Gao et al., 2019). Detailed information ~~for the~~concerning references, samples, and sampling and analytical methods for the sources are ~~represented~~ given in Table S3. For ~~the~~those species which ~~cannot~~ could not be ~~analyzed~~ determined by ~~the~~ analytical methods, ~~we supplement~~ the mass fraction~~s~~ data was supplemented with ~~of these species~~ that obtained from the SPECIATE database. Figure 2 ~~and Figure 3~~ show~~s~~ the $PM_{2.5}$ and NMVOC speciation profiles ~~of~~ for the major emission sources in the YRD region~~, respectively~~.

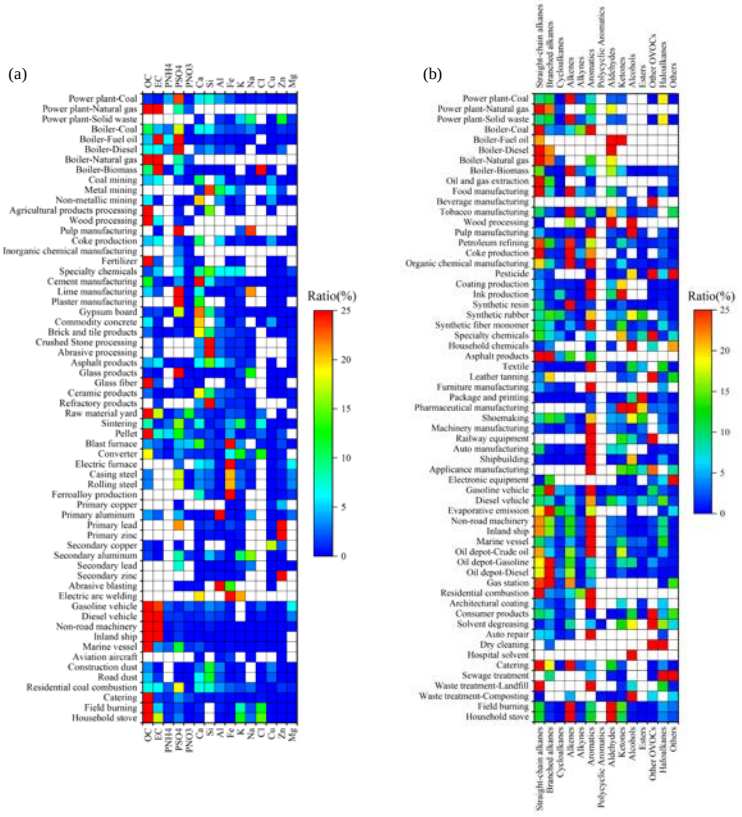

Figure 2. ~~S~~The ~~s~~peciation profiles for the major emission sources~~of~~: (a) $PM_{2.5}$ ~~(a);~~ and (b) NMVOCs ~~(b) for major emission sources~~.

2.7 Spatial distributions

Emissions from industrial sources, e.g. ~~including~~ power plants, boilers, industrial process and solvent-use sources, were allocated from the Environmental Statistics Database based on their ~~latitude and longitude~~geographical coordinates ~~from Environmental Statistics Database~~. Vehicle emissions were ~~determined based~~calculated from ~~on~~ the mileage ~~sharing~~ distributions of various vehicle ~~types~~categories on different ~~levels~~ types of road~~s~~. The composition of traffic flow on different ~~levels~~ types of road~~s~~ was obtained from ~~the~~ previous surveys in Shanghai and Hangzhou (Huang et al., 2015; Yang et al., 2017). The approach of spatial allocation for road dust was consistent with that used for vehicle emissions. The spatial distribution of emissions

from non-road machinery ~~varies~~ varied ~~in different ways~~ depending on the type of machinery. The emissions from construction and agricultural machinery were allocated according to ~~the built up~~urban and farmland areas given in the 2015 land use data released by European Space Agency (ESA) (https://www.esa-landcover-cci.org/). Emissions from port and factory machinery, and airport ground handling equipment were ~~allocated~~ assigned according to their ~~latitude and longitude~~geographical coordinates. Emissions from residential sources were ~~allocated~~ assigned ~~based~~ from population distribution data ~~on~~ with a 1 km ~~resolved~~ resolution~~population distribution data~~. ~~Those of~~Emissions from ~~agriculture~~ agricultural sources were allocated ~~based on~~from ~~the~~ farmland areas in the land ~~-~~use data (ibid.).

2.8 Uncertainty analysis

~~The u~~Uncertainty ~~is~~ was mainly derived from the activity data and EFs ~~in the EI~~.

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~~In this study, we classify t~~The coefficients of variation of the activity data and EFs ~~of~~ for each source were classified into seven grades in the range of 2%–100% ~~based on~~using expert judgment. The coefficient of variation for the activity data ~~is~~ was determined ~~based on~~according to the data source. ~~The e~~Environmental statistical data with ~~individual~~specific source information ~~is~~ was assigned the lowest coefficient of ~~uncertainties~~uncertainty (2%), while ~~the estimated~~ activity data ~~based~~ estimated ~~on~~ from the statistical yearbooks, such as biomass burning, ~~are~~ was assigned the highest uncertainty value~~s~~ (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 ~~of the~~for ~~emission factor uncertainty~~EFs ~~is~~was the same as the activity level. ~~The~~ EFs derived from local measurements in the YRD region with large samples ~~are~~ were assigned the ~~lowest~~ lower coefficients of uncertainty~~values~~ (18%), while those from USEPA~~'s~~ or EMEP/EEA~~European's~~ datasets ~~are~~ were assigned high~~er~~ coefficients (98%). Then the uncertainty of each pollutant from each emission source can be combined by Eq. (3–5). A ~~D~~detailed description of the ~~analysis~~ analytical method~~s~~ used can be found in our previous study (Huang et al.,

2011).

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

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

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

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.

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2.9 ~~Uncertainty analysis~~Model configurations ~~validation~~

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.

2.10 ~~Model configurations~~Estimation of O_3 and SOA formation potentials

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).

$$OFP_i = \sum_{j=1} E_{i,j} \times MIR_j \quad (6)$$

$$SOAP_i = \sum_{j=1} E_{i,j} \times Y_j \quad (7)$$

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

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3. Results and discussion

3.1 Emission and source contributions

3.1.1 Emissions and ~~their~~ comparisons with previous studies

The total emissions of SO_2, NO_x, CO, NMVOCs, PM_{10}, $PM_{2.5}$, and NH_3 in the YRD region for the year of 2017 were 1,552, 3,235, 38,507, 4,875, 3,770, 1,597 and 2,467 Gg, respectively. ~~If~~ Excluding ship emissions ~~were not included~~, the air pollutant emissions ~~above would be~~were 1,437, 2,936, 38,486, 4,867, 3,754, 1,583 and 2,467 Gg, respectively. Detailed information of air pollutant emissions for each city is shown in Table S1 ~~in~~ of the supporting information~~the Supplement~~.

Table ~~1~~ 2 shows the estimated emissions ~~in~~ for the YRD region ~~estimated in this study~~ and their comparison~~s~~ with previous studies. SO_2 emissions were close to ~~the~~ that from~~result in~~ the MEIC in 2016~~,~~ and ~~were~~ much lower than those reported in ~~other studies in the past few years~~2015. Emission reductions ~~on~~ from coal-fired facilities, including power plants and boilers, ~~were~~ could be the main reason for the significant decline in SO_2 emissions (Zheng et al., 2018). NO_x emissions were generally lower than the results ~~in~~ from previous studies. Some modeling and satellite studies verified that the NO_x emissions in previous studies were overestimated, partly due to ~~the failure to consider the improved~~ improvements in NO_x control measures for the power sector

which were not considered at that time (Zhao et al., 2018; Sha et al., 2019). The NO_x ~~emission factor~~EFs for coal-fired power plants and boilers in this study were derived from local measurements which were generally lower than those in previous studies, so the NO_x emissions from the power sector were 47% lower than those from the MEIC. CO emissions were higher than the MEIC~~'s~~ results but ~~close~~ similar to those reported by Sun et al. (2018a). NMVOC emissions for key sources in this study were individually estimated ~~base on~~using the "bottom-up" method, so the estimates were lower than ~~the others who~~those ~~used~~ using the "top-down" approach. ~~Another reason~~In addition, ~~is the majority~~most of the ~~emission factor~~EFs selected in this study were detailed into different process segments, which ~~are~~ were generally lower than the comprehensive ~~factors~~ EFs used for whole industrial sectors in ~~the~~ previous studies. Since dust sources were not included in the MEIC inventory, PM_{10} and $PM_{2.5}$ emissions estimated in this study were 1.7 and 0.5 times higher ~~than the results in MEIC~~, respectively. A previous study ~~has pointed out~~ also showed that ~~the existing~~ NH_3 emissions in China were underestimated, mainly due ~~to the underestimate of NH_3~~the application of lower emission rates from fertilizer applications and livestock and ~~missing~~ the omission of ~~some emission~~other sources (Zhang et al., 2017). Therefore, we used ~~the~~ local measured NH_3 ~~emission factor~~EFs for fertilizer application and ~~part~~ some of the livestock breeding sources in the YRD region ~~instead in this study~~. ~~Another difference came from transportation sector.~~ NH_3 emissions from the transportation sector were also ~~2.8 times~~ higher (by a factor of 2.8) than those from the ~~in~~ MEIC ~~when~~ due to the localized NH_3 ~~emission factor~~EFs ~~form~~ from light-duty gasoline vehicles (Huang et al., 2018) ~~were~~ used in this study. In addition, NH_3 slip from selective catalyst reduction ~~(SCR)~~ devices in the power sector (not included in previous studies) was also considered in this study. ~~However, this emission source has not been included in previous studies.~~

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 NOx, 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.

Table 12. Air pollutant emissions in the YRD region ~~in this study~~ and their comparison~~s~~ with other studies.

Regions	Data source	Base year	Annual air pollutant emissions (Gg/year)						
			SO_2	NO_x	CO	NMVOCs	PM_{10}	$PM_{2.5}$	NH_3
YRD	This study	2017	1,437	2,936	38,486	4,867	3,754	1,583	2,467
	MEIC	2016	1,136	3,753	19,560	5,527	1,374	1,025	1,153
	Simayi et al., 2019	2016				4,984			
	Sun et al., 2018a	2015	3,050	4,160	30,210	5,490			
	Zhang et al., 2017b	2015							1,632
	Our previous study	2014	2,737	3,457	33,118	4,069	4,681	2,085	1,582
	Wu et al., 2018	2013				6,198			
Shanghai	This study	2017	57	225	1,393	418	124	56	54
	MEIC	2016	168	345	1,192	683	69	51	25
	Simayi et al., 2019	2016				728			
	Sun et al., 2018a	2015	550	470	2250	580			
	Zhang et al., 2017b	2015							50
	Our previous study	2014	100	256	1,699	391	197	93	73
	Wu et al., 2018	2013				838			
	Fu et al., 2013	2010	260	453		422	86	59	65
Jiangsu	This study	2017	619	1,165	17,309	2,056	1,440	577	1,093
	MEIC	2016	468	1,586	8,191	2,128	516	388	532
	Simayi et al., 2019	2016				2,024			
	Sun et al., 2018a	2015	1,230	1,700	13,780	2,000			
	Zhang et al., 2017b	2015							703

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	Our previous study	2014	1,002	1,315	12,667	1,560	1,761	779	544
	Wu et al., 2018	2013				2,240			
	Zhou et al., 2017	2012	1,142	1,642	7,680	1,747	1,394	941	1,100
	Fu et al., 2013	2010	1,126	1,257		1,759	619	401	976
Zhejiang	This study	2017	339	676	7,036	1,484	775	308	363
	MEIC	2016	280	867	3,779	1,671	219	151	159
	Simayi et al., 2019	2016				1,624			
	Sun et al., 2018a	2015	730	980	5,110	1,810			
	Zhang et al., 2017b	2015							257
	Our previous study	2014	646	903	9,372	1,346	1,199	508	374
	Wu et al., 2018	2013				2,214			
	Fu et al., 2013	2010	762	1,067		1,641	301	184	398
Anhui	This study	2017	422	869	12,748	910	1,415	642	957
	MEIC	2016	221	954	6,398	1,045	570	435	437
	Simayi et al., 2019	2016				608			
	Sun et al., 2018a	2015	540	1,010	9,070	1,100			
	Sun et al., 2018b	2015	434	688				323	422
	Zhang et al., 2017b	2015							622
	Our previous study	2014	725	983	9,380	772	1,524	706	592
	Wu et al., 2018	2013				906			

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3.1.2 Source contributions

Figure 3 shows the emission contributions of the major air pollutants in the YRD region from the~~contributions of emission sources divided by different~~ main ~~source categories~~ category sources (a), industrial sector sources (b), and mobile sources ~~types~~ (c). Further ~~Detailed~~ detailed information of ~~the~~ emissions from each source ~~was~~ is provided in Table S5. SO_2 and CO emissions were mainly from boilers, accounting for 49% and 73% of the total emissions, respectively. Notably, ~~the~~ emission contributions ~~of~~ from power plants were ~~much~~ significantly lower than those ~~in other~~from previous inventories (MEIC, http://meicmodel.org/; Zhou et al., 2017), resulting mainly from ~~the significant reduction in power plant emissions due to~~ the implementation of ~~ultra-low~~stringent emission reduction measures in recent years (Wu et al., 2019; Zhang et al., 2019).

Mobile sources accounted for ~~dominated~~ the majority (57%) of NO_x emissions in the YRD region~~, which contributed 57% of the total. This estimate~~ which was generally

higher ~~than the proportion of mobile sources~~ contributions given in the MEIC and other ~~inventories~~ EI (Zhou et al., 2017; Sun et al., 2018a). Emission control measures for power plants had demonstrably ~~played an important role in reducing~~reduced ~~their contributions on~~ NO_x emissions. In addition, ~~other studies did not include~~ ship emissions, which had not been included in previous studies, accounted for 16% of NO_x emissions from mobile sources in the YRD region~~,~~ (see ~~as shown in~~ Figure 3(c)~~,~~ 'Marine'). Furthermore, ~~Another reason that cannot be ignored was the~~ NO_x ~~emission factor~~EFs ~~from~~ for gasoline and diesel vehicles used in this study, ~~were modified based on~~derived from local measurements ~~in this study~~, ~~which~~ were generally higher than those ~~recommended~~ given by MEP (2014). Some real-world measurements based on portable emission measurement systems ~~(PEMS)~~, on-road chasing, and tunnel experiments also indicated that the NO_x emissions from vehicles in China were higher than expected, probably due to deficiencies in the type-approval protocols and emission controls~~the existence of high-emitting vehicles~~ (Wu et al., 2012; Huang et al., 2017; Song et al., 2018; Wen et al., 2019).

Industrial sources accounted for the majority of total NMVOC emissions ~~were mainly contributed by industrial sources, accounting for~~ (61%) ~~of the total~~, of which industrial process and solvent-use sources ~~accounted for~~contributed 34% and 27%, respectively. Mobile and residential sources contributed 20% and 10%, respectively. Dust sources were the main contributors to PM_{10} (55%) and $PM_{2.5}$ (28%) emissions~~,~~ ~~occupied 55% and 28%, respectively~~. Agricultural sources ~~contributed up to~~accounted for 91% of NH_3 emissions with minor contributions from~~. In addition,~~ residential (3%) and mobile (1%) sources ~~contributed 3% and 1% of NH_3 emissions, respectively~~. Although NH_3 ~~emission factor~~EFs from vehicles ~~have been~~were considered in this study, their ~~NH_3~~ emissions ~~was still~~were significantly lower than those from agricultural sources. However, vehicle emissions were mainly concentrated in urban areas where~~,~~ ~~and~~ their contribution to NH_3 emissions ~~in urban areas~~ would be considerable.

Industrial ~~–~~emissions of SO_2 and CO ~~emissions~~ were mainly ~~came~~ from the five

major sectors~~, including~~ of petroleum refining, coking, chemical manufacturing, non-metallic mineral manufacturing, and ferrous metal manufacturing. Non-metallic mineral manufacturing and ferrous metal manufacturing dominated the industrial NO_x, PM_{10}, and $PM_{2.5}$ emissions. The top five ~~sectors of~~ industrial sector sources of NMVOCs emissions in the YRD region were chemical manufacturing, ~~Furniture~~ furniture and wood manufacturing, ~~Appliance~~ appliance manufacturing, rubber and plastic manufacturing, and non-metallic mineral manufacturing~~.~~. These ~~accounting~~ accounted for 27%, 12%, 9%, 9%, and 6% of the total NMVOC emissions~~,~~ respectively. Chemical manufacturing contributed to the majority of industrial NH_3 emissions in the YRD region.

The ports and harbors of the YRD region ~~has the largest port~~are collectively the largest group in the world~~,~~ and, as expected, ~~so the~~ emissions from the transportation of ships and heavy-duty trucks dominated the mobile source emissions. ~~Among them,~~ ~~s~~Ships accounted for 94%, 16%, 12%, and 12% of mobile source SO_2, NO_x, PM_{10}, and $PM_{2.5}$ emissions, respectively while ~~and~~ heavy-duty trucks ~~occupied~~ contributed 31%, 37%, and 36% ~~of~~ to mobile source NO_x, PM_{10}, and $PM_{2.5}$ emissions~~,~~ respectively. Light-duty vehicles contributed significantly to CO, NMVOCs, and NH_3 emissions, accounting for 61%, 46%, and 90%, respectively. Non-road machinery accounted for 27%, 18%, 12%, 21%, and 22% of NO_x, CO, NMVOCs, PM_{10}, and $PM_{2.5}$ emissions from mobile sources~~,~~ respectively. Construction and agriculture machinery were also major contributors.

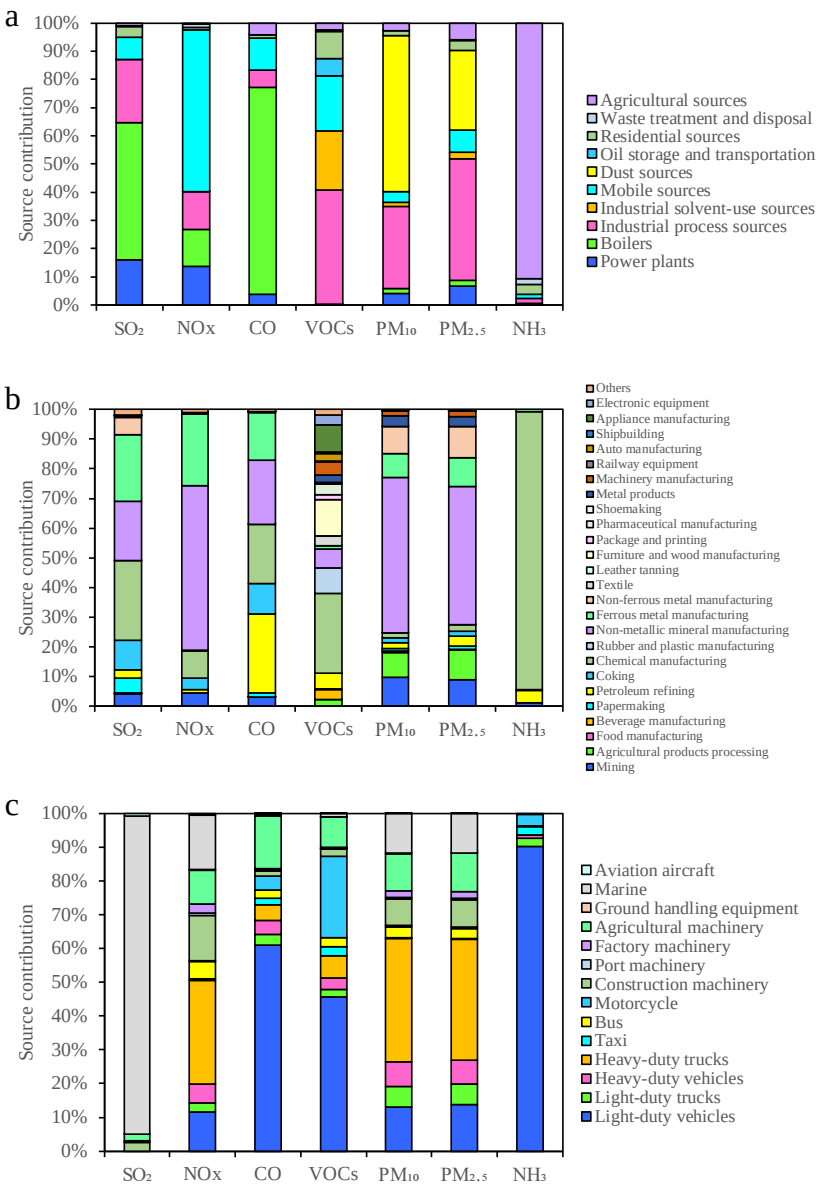

Figure 3. Emission ~~Source~~ contributions of major air pollutant~~s emissions~~ in the YRD region from~~.~~: (a) ~~Divided by major~~ Major ~~source categories~~category sources; (b) ~~Divided by detailed~~ industrial sector~~s~~ sources; (c) ~~Divided by detailed~~ mobile sources.

3.1.3 Spatial distribution

Figure. 4 shows the spatial allocationdistribution of SO_2, NO_x, CO, NMVOCs, $PM_{2.5}$, and NH_3 emissions in the YRD region. SO_2 emissions were mainly concentrated in the high-density shipping regions of the Yangtze River and East China Sea estuary, where ships were densely populated. SO_2 emissions from power plants and boilers along the Yangtze River and in the cities of northern Anhui and Jiangsu provinces were also dense, mainly from power plants and boilers also significant in these regions. The spatial distribution of NO_x and NMVOCs was similar and, mainly concentrated along the Yangtze River and Hangzhou Bay, where the industries industry and logistics were most developed. CO and $PM_{2.5}$ emissions were mainly concentrated in the built upurban areas of cities due to intensive road traffic and human activities such as construction sitework. NH_3 emissions were relatively high in northern Anhui and Jiangsu provinces, resulting mainly from their developed agriculture. Large cities such as Shanghai presented higher NH_3 emission intensities, largely due toThe contributions of NH_3 emissions from residential and mobile sources has led to higher NH_3 emission densities for large cities such as Shanghai.

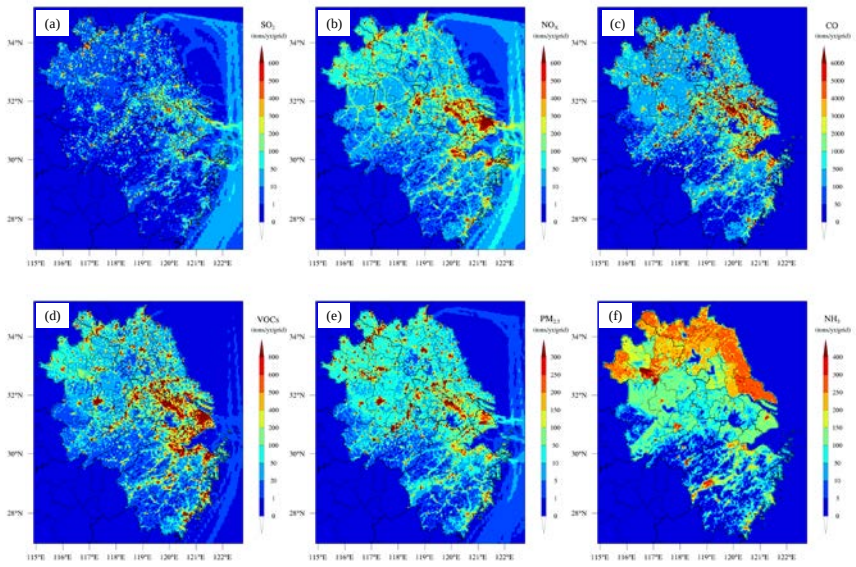

Figure 4. Spatial distribution of major air pollutant emissions in the YRD region~~.~~: (a)~(f) ~~refer to~~Represent SO_2, NO_x, CO, NMVOCs, $PM_{2.5}$, and NH_3 ~~in turn~~respectively.

Figure 5 shows the spatial distribution of ~~major~~ NO_x and NMVOCs from the major industrial sectors ~~of NO_x and VOCs emissions~~ in the YRD region. There were large differences in the spatial distribution of the different industrial sectors. Key sources of NO_x emissions, shown in Figure 5(a)-(d), included: ~~The power~~ Power plants ~~were~~ mainly distributed along the Yangtze River and Hangzhou Bay and the northern part of the YRD region~~.~~; ~~The~~ iron ~~&~~ and steel manufacturing ~~sector was~~ concentrated along the Yangtze River~~.~~; and ~~Cement~~ cement and brick manufacturing ~~sectors were~~ mainly distributed in the western and northern regions of the YRD. ~~In comparison~~Compared with NOx emissions, the key ~~sectors~~ sources of NMVOC ~~emissions~~ (Figure 5(e) ~~-~~(l)) were mainly concentrated in the central and eastern regions of the YRD~~,~~. These ~~including~~ included the regions of Shanghai, Suzhou, Wuxi, Changzhou, Nanjing, Hangzhou, Ningbo, Jiaxing, and Shaoxing~~, etc.~~, which also had the ~~strongest~~ highest NO_x emission intensities in the YRD region. High ~~intensities~~ levels of NO_x and NMVOC ~~emissions are the~~were key factors ~~leading to~~responsible for ~~serious~~ harmful pollution ~~of~~ by ~~ozone and~~ secondary ~~particulate matter~~PM and O_3 in this region (Li et al., 2018; Li et al., 2019). ~~Refining the specific industrial sectors of emissions can help to find out the detailed sources inducing air pollution.~~

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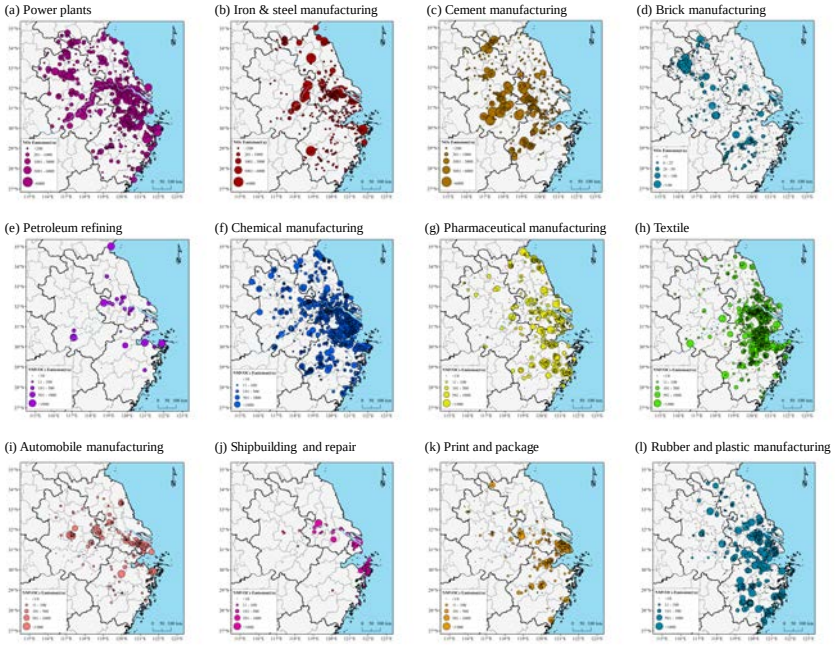

Figure 5. Spatial distribution of the major NO_x and NMVOC emission sources in the YRD region.

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 ~~the~~other 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 NH_3 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 NO_x 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.

3.1.4 Uncertainty assessment

The ~~inventory~~ EI was compiled using ~~a~~ the "bottom-up" approach based on local ~~emission factor~~EFs and activity data ~~in~~ from the region. The activity data ~~of~~ for industrial sources, including fuel consumption, sulfur content, ash content, raw material used, and control efficiency, were collected from the Environmental Statistics Database. ~~Emission factor~~EFs from some key sources, such as coal-fired power plants and boilers, iron ~~&~~ and steel manufacturing, gasoline and diesel vehicles, non-road machinery, catering, and agricultural sources~~, etc.~~, ~~have been~~were modified based on the local measurements. These ~~all~~ measured helped to reduce the uncertainty of the emission estimates. Table ~~2~~ 3 shows the uncertainties of major sources at the 95% confidence interval in this ~~inventory~~EI. The average uncertainties of emissions from the YRD region were estimated as -29 to 36% for SO_2, -28 to 33% for NO_x, -42 to 75% for CO, -44 to 68% for NMVOCs, -36 to 62% for PM_{10}, -30 to 46% for $PM_{2.5}$, and -58 to 117% for NH_3. The overall ~~uncertainty~~ uncertainties ~~of this inventory was~~ were ~~reduced~~ lower compared ~~to~~ with our previous ~~inventory~~ EI for the YRD region (Huang et al., 2011).

The uncertainty assessment ~~indicates~~ indicated that emissions from ~~the~~ stationary combustion sources, such as ~~including~~ power plants and boilers, were more reliable, because the ~~emissions were estimated~~estimates were based on ~~the~~ detailed activity data and local measurements. The uncertainties ~~of~~ for emissions from major industrial sectors, such as ferrous metal manufacturing, non-ferrous metal manufacturing, and non-metallic mineral manufacturing, were ~~greatly~~ significantly improved ~~when using~~ by the detailed emission estimation approach for the different process segments. ~~In comparison~~However, ~~the~~ large uncertainties remained for emissions from chemical manufacturing ~~still have large uncertainties since there are a~~due to the many uncategorized ~~large number of~~ processes and emissions for that sector ~~segments and unorganized emissions~~. The uncertainties ~~of~~ for emissions from vehicles and non-road machinery in this study were mainly ~~came~~ from the activity data. Although their population could be obtained from the statistical yearbooks, ~~their mileage~~ VMT ~~travels~~

~~or~~and working hours ~~were still difficult to~~could not be estimated accurately. Dust emissions ~~including~~ from construction and roads dust ~~have~~ had much higher uncertainties ~~due to~~as ~~less information of~~ their activity data lacked detail and fewer ~~emission factor~~EFs ~~was~~ were available. Most of the area sources, ~~like~~ such as residential and agricultural ~~sources~~, were estimated ~~based on~~from ~~the~~ activity data obtained from the statistical yearbooks, resulting in higher uncertainties ~~of~~ in their emission estimates. Despite these limitations, ~~Overall, using of~~the emission estimation approach, based on refined process segments and local measurements, ~~can help to~~ reduced the overall uncertainties of the EI. However, more ~~detailed~~comprehensive activity data and accurate ~~emission factor~~EFs are still ~~very critical~~required to improve the quality of EIs in the future.

Table ~~2~~3. Uncertainty assessment ~~of~~ for the major emission sources in the YRD region.

Sources	SO_2	NOx	CO	NMVOCs	PM_{10}	$PM_{2.5}$	NH_3
Power plants	(-25%, 28%)	(-33%, 15%)	(-26%, 27%)	(-28%, 22%)	(-24%, 29%)	(-25%, 28%)	(-45%, 76%)
Boilers	(-29%, 38%)	(-23%, 27%)	(-24%, 30%)	(-19%, 23%)	(-24%, 30%)	(-24%, 30%)	(-46%, 56%)
Petroleum refining	(-49%, 84%)	(-45%, 72%)	(-51%, 90%)	(-40%, 57%)	(-53%, 60%)	(-53%, 64%)	(-39%, 62%)
Chemical manufacturing				(-71%, 167%)			
Ferrous metal manufacturing				(-41%, 61%)	(-23%, 48%)	(-12%, 34%)	
Non-ferrous metal manufacturing	(-37%, 78%)	(-42%, 62%)		(-44%, 70%)	(-38%, 60%)	(-52%, 94%)	
Non-metallic mineral manufacturing	(-48%, 75%)	(-46%, 71%)	(-47%, 72%)	(-45%, 69%)	(-44%, 74%)	(-43%, 68%)	
Vehicles		(-38%, 55%)	(-48%, 73%)	(-46%, 69%)	(-50%, 83%)	(-44%, 67%)	(-55%, 98%)
Non-road machinery	(-47%, 75%)	(-44%, 66%)	(-57%, 112%)	(-50%, 86%)	(-46%, 76%)	(-45%, 77%)	
Construction dust					(-56%, 104%)	(-57%, 102%)	
Road dust					(-35%, 71%)	(-43%, 68%)	
Oil storage and transportation sources				(-43%, 69%)			
Residential solvent-use				(-57%, 116%)			
Residential combustion	(-64%, 143%)	(-44%, 79%)	(-70%, 88%)	(-68%, 165%)	(-43%, 66%)	(-43%, 66%)	(-44%, 72%)
Biomass burning	(-66%, 171%)	(-62%, 124%)	(-67%, 153%)	(-65%, 142%)	(-66%, 149%)	(-66%, 152%)	(-68%, 159%)
Livestock and poultry farming							(-67%, 148%)
Fertilizer application							(-78%, 213%)
Overall	(-29%, 36%)	(-28%, 33%)	(-42%, 75%)	(-44%, 68%)	(-36%, 62%)	(-30%, 46%)	(-58%, 117%)

3.2 $PM_{2.5}$ and NMVOC species emissions

3.2.1 $PM_{2.5}$ species

Figure 6 shows the major ~~emissions and~~ source contributions and species

comprisingof major $PM_{2.5}$ species in the inventoryEI. OC, Ca, Si, PSO_4, and EC were top five components in of primary $PM_{2.5}$ in the YRD region, accounting for 9.0%, 7.0%, 6.4%, 4.6%, and 4.3% of $PM_{2.5}$ emissions, respectively. There were large differences in the emission contributions of for the different $PM_{2.5}$ species. Among the industrial sources, the non-metallic mineral manufacturing sector had made the largest contributions to Ca, Si, and Al emissions, accounting for 51.6%, 15.9%, and 18.8% of these species, respectively. Ferrous metal manufacturing was the main source of Fe emissions, accounting for 57.9%. Vehicles was were major contributors to OC and EC emissions, taking up at 18.0% and 43.5%, respectively. K and Cl emissions mainly came from biomass burning, accounting for 50.4% and 78.5%, respectively. Construction dust was also an important source of $PM_{2.5}$ componentsspecies, accounting for 15.9%, 34.1% and 20.4% of Ca, Si, and Al emissions, respectively.

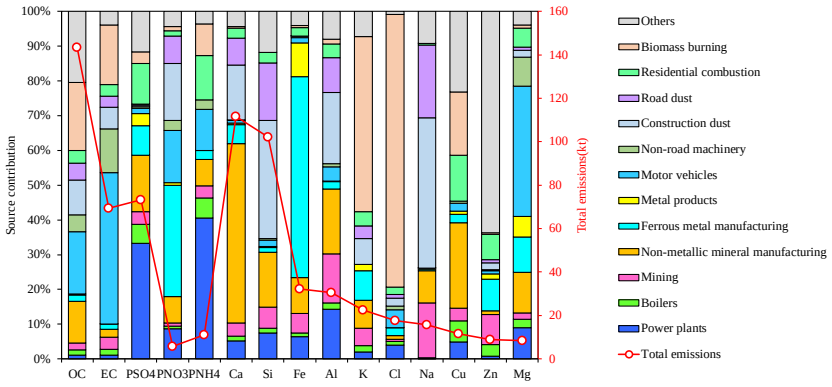

Figure 6. Emissions and Major source contributions and species comprisingof major $PM_{2.5}$ species in the YRD region.

3.2.2 NMVOC species

Figure 7 shows the major VOC species emissions and their source contributions for species comprising NMVOC. The aAromatic hydrocarbons and alkanes were the maindominated the NMVOC species, accounting for ~25.3% each of the total NMVOC emissions in the YRD region, followed by the alkanes, occupying 24.7%. Among the alkanesm, the straight chainlinear, branched, and cycloalkanes took made up 11.9%,

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9.9%, and 2.8% of the total~~,~~ respectively. OVOCs also accounted for a considerable proportion (21.9%) of NMVOC emissions in the YRD region~~, about 21.9% of the total~~. Among the~~se m~~, ~~the~~ aldehydes, ketones, alcohols and esters ~~took~~ made up 5.0%, 4.4%, 9.0% and 3.5% of the total respectively. ~~In addition, the h~~Haloalkanes ~~occupied~~ accounted for~~about~~ 3.1% of the total NMVOC emissions. In the previous study of NMVOC emissions in the YRD region by Wu et al. (2017), ~~The~~ aromatic hydrocarbons were also the dominant species ~~in VOC emissions in the YRD region in a previous study reported by Wu et al. (2017), even higher~~ (40%) ~~than the proportion in ours. The proportion of~~, concentrations of OVOCs ~~was quite close~~were similar ~~to our study~~, while the proportion of alkenes and haloalkanes were generally lower~~ than ours.~~

~~The c~~Chemical manufacturing accounted for a considerable proportion of various NMVOC species in the YRD region, accounting for 12.7%, 21.5%, 13.7%, and 10.8% of the alkane~~s~~, alkene~~s~~, aromatic~~s~~ hydrocarbon, and OVOC emissions~~,~~ respectively. Industrial solvent-use sources, ~~including~~ which included furniture and wood processing, textile~~s~~, ~~package~~ packaging and printing, pharmaceutical manufacturing, metal products, auto manufacturing, and appliance manufacturing,~~ etc.,~~ were also an important source of NMVOC emissions in this region~~,~~. Industrial solvent-use sources ~~which occupied~~accounted for 29.3% and 33.3% of ~~the~~ aromatic~~s~~ hydrocarbon and OVOC emissions respectively, while residential solvent-use sources accounted for 23.7% and 4.9% of ~~the~~ aromatic~~s~~ hydrocarbon and OVOC emissions respectively ~~in the YRD region~~. Motor vehicles also ~~have a very important~~made a significant contribution to ~~various~~ NMVOC species in the YRD region, ~~occupying~~ making up 31.2%, 10.4%, 15.1%, and 10.5% of the total emissions of alkanes, alkenes, aromatics, and OVOC ~~emissions~~ in the region respectively. Biomass burning contributed to 12.0% of ~~the~~ aldehyde emissions, although it accounted for only 2.5% of the total NMVOC emissions in the region.

Overall, the refinement of ~~VOC~~ the source profiles ~~can help~~helped to ~~provide an important support for~~ assess~~ing~~ the impact~~s~~ of NMVOC emissions on ambient air

quality in the region. However, ~~there are still considerable~~ differences ~~of~~ in NMVOC compositions between ~~in different~~ studies remained and~~. It is necessary to strengthen~~ efforts should be made to improve the verification of NMVOC species ~~emissions~~ in ~~the~~ future emissions studies.

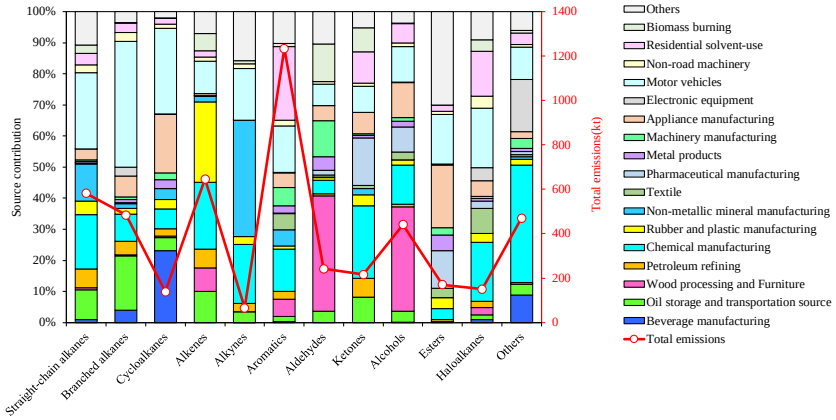

Figure 7. Major ~~Emissions and~~ source contributions of species comprising ~~major~~ NMVOC ~~species~~ in the YRD region.

3.3 Model validation

~~To verify the reliability of the emission inventoryEI, we used the Community Modeling and Analysis System (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 in for January and July 2017, and compared these with the observation data in for each city of in the region. The Weather Research and Forecast (WRF) version 3 supplied the meteorological field for the CMAQ model was obtained from the WRF (version 3). The emission inventoryEI developed in this study was then used to produce the an emission system in for the YRD region while emissions beyond the YRD was were supplied obtained fromby Multiresolution Emission Inventory for Chinathe (MEIC (2016) (http://www.meicmodel.org). The anthropogenic data was then combined with the biogenic data obtained from the Model for Emissions of Gases and Aerosol from Nature~~

~~modelling system (MEGAN version 2.10) as the final input of~~ for the ~~emission inventory~~EI ~~for~~ of the model. ~~Figure S1 and Table S6 show the domain and settings of for the model system, respectively. Detailed information is provided in Section 6 of the Supporting information Section 6.~~

Figure 8 shows a comparison of ~~compares~~ 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 ~~in the YRD region with those of the observation data~~. The simulated concentration distribution of the different~~various~~ pollutants ~~is~~ was consistent with the ~~observation~~ observed results, ~~which indicates~~indicating that the updated ~~inventory~~ EI generally ~~reflects~~ reflected the distribution of air pollution sources in the YRD region. Comparatively, ~~the consistency~~ agreement between the simulated concentration distribution and the observed results ~~of~~ for the cities in the central areas of the YRD region ~~is~~ was stronger than ~~those~~ cities of the northern and southern border areas. This ~~is~~ was mainly because ~~the concentration in the~~ border areas ~~is~~ were more susceptible to the effects of emissions from ~~the outer~~ areas outside the region, which ~~leads to~~resulted in greater deviation of the simulation results ~~deviation~~. A statistical analysis of the ~~results~~hourly concentrations obtained from the ~~Detailed statistical results of the model performance~~ model for ~~simulating various~~ the pollutants in each city ~~is shown~~ 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. ~~Overall, the simulation results of O_3 were relatively high in January, while SO_2, NO_2, $PM_{2.5}$, PM_{10}, and CO were relatively low. While in July, except that the O_3 simulation concentrations were slightly higher than the observed results, the average NO_2 simulation values were consistent with the measured averages, other pollutants were relatively lower.~~

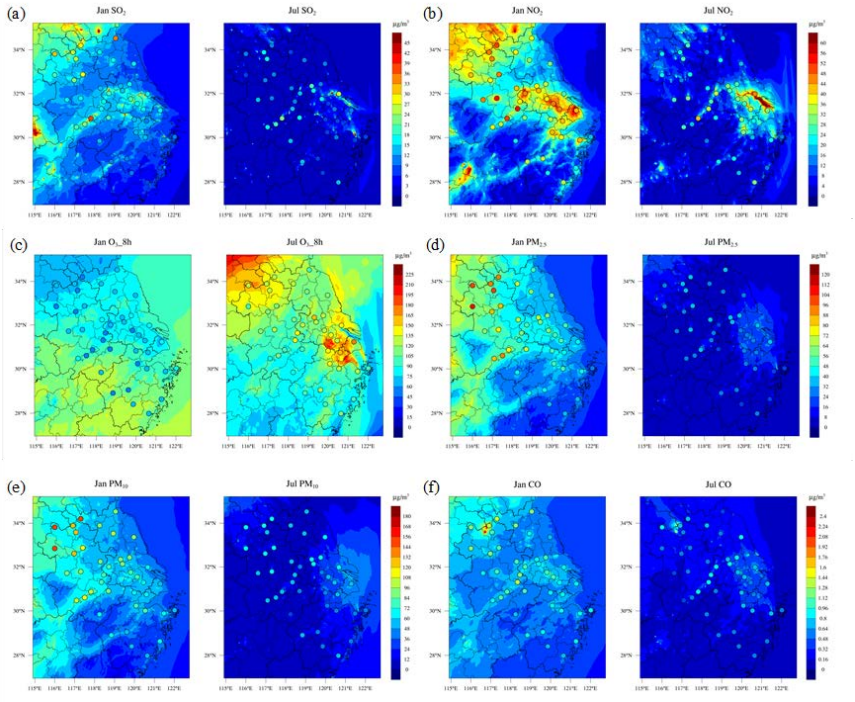

Figure 8. Comparisons of simulated and observed (circles) monthly average pollutant concentrations for cities in the YRD region in January and July 2017: (a) ~~of~~ SO_2~~,~~; (b) NO_2~~,~~; (c) daily maximum 8-hour ~~of~~ O_3 (O_3_8h)~~,~~; (d) $PM_{2.5}$~~,~~; (e) PM_{10}~~,~~; and (f) ~~and~~ CO ~~in cities in the YRD region in January and July 2017~~.

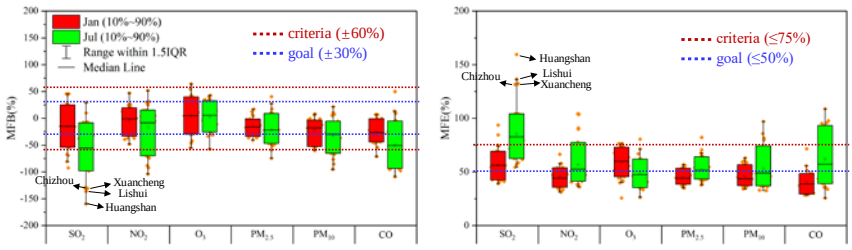

Figure 9. MFB (a) and MFE (b) between the simulation and observation data for daily average concentrations of various pollutants of the cities in the YRD region in January and July 2017

3.4 Ozone and SOA formation potentials

~~To characterize the regional O_3ozone and SOA formation contributions of different NMVOC species and their sources, we used the O_3ozone formation potential (OFP) and~~

~~SOA formation potential (SOAP) methods to of estimate~~estimation~~. OFP and SOAP are were obtained from the sum of the individual NMVOC species emissions multiplied by the maximum incremental reactivity (MIR) and SOA yield, respectively. The MIR and SOA yield of for individual NMVOC species was were referenced obtained from previous studies (Carter, 1994; Wu and Xie, 2017).~~

Figure 9 shows the OFP and SOAP contributions ~~from~~ for major NMVOC species, emission sources, and industrial sectors in the region. ~~In terms of individual species, t~~Toluene ~~is~~ was the most important NMVOC species for both OFP and SOAP~~, which~~and contributed to 45.0% of the total (7.5% OFP and 37.5% SOAP), followed by 1,2,4-trimethylbenzene~~,~~ which ~~contributing~~ contributed to 29.1% of the total (2.1% OFP and 27.0% SOAP). Other~~s~~ NMVOC included~~were~~ m/p-xylene, propylene, ethene, o-xylene, ethylbenzene~~,~~ which gave combined ~~etc., their~~ contributions to ~~both~~ OFP and SOAP ~~were~~ of 14.9%, 16.6%, 10.7%, 5.7%, and 2.7% ~~in turn~~respectively~~.~~; ~~Their~~ OFP was the major contribution ~~was relatively more prominent~~.

Industrial process sources ~~dominated~~ had the highest OFP and SOAP in the region~~,~~ ~~which contributed~~ contributing 44.9% and 26.7%~~,~~ respectively. Industrial solvent-use sources followed, with OFP and SOAP contributions of 15.0% and 33.8%~~.~~; ~~and~~ their contribution to SOAP ~~even~~ exceeded that of the industrial process sources. The contribution~~s~~ of motor vehicles to regional OFP and SOAP were 13.9% and 13.5%~~,~~ respectively, which was close to those from residential solvent-use sources. These two sources were the major contributors ~~of~~ to O_3~~ozone~~ and SOA formation in urban areas.

~~There are f~~Four major industrial sectors could account for most of the OFP and SOAP ~~with significant potential contribution to ozone and SOA production~~ in the YRD region~~.~~: The chemical manufacturing sector contributed 16.4% and 14.8% ~~of~~ to OFP and SOAP~~,~~ respectively~~.~~; ~~The~~ the ~~second was~~ rubber & plastic manufacturing sector~~,~~ had ~~with~~ a SOAP contribution rate of 11.8%~~,~~ ~~while~~ (its OFP was relatively low at~~, about~~ 1.2%); the ~~third and fourth were~~ appliance manufacturing and textile sectors, ~~accounting~~ accounted for 10.5% and 10.4% of ~~both~~ OFP and SOAP contributions

respectively.

Based on the above, it ~~can be~~was concluded that the reduction of aromatic hydrocarbon emissions from industrial and vehicular sources were of ~~vital~~ greatest importance for the YRD region~~,~~. In particular, ~~especially for~~ the high reactivity species, such as toluene, xylene, and trimethylbenzene~~, etc., which~~ should be given ~~the top~~ priority ~~on~~ in NMVOCs pollution control measures for~~in~~ the region.

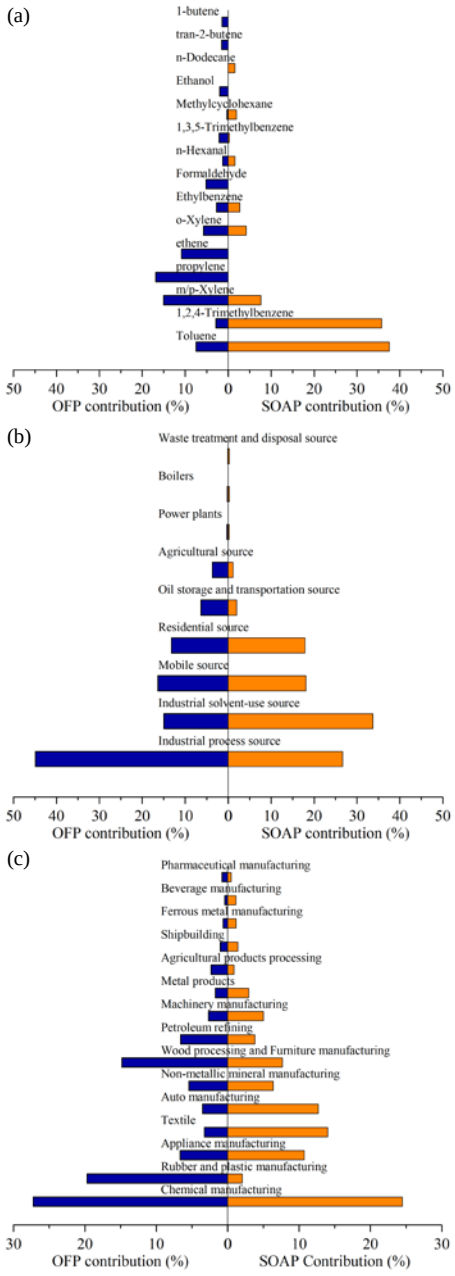

Figure ~~9~~10. OFP~~Ozone~~ and SOAP formation potentials for ~~from~~ different variables: (a) NMVOC species~~,~~; (b) emission sources~~,~~; and (c) industrial sectors.

4. Conclusions

A high-resolution air pollutant ~~emission inventory~~EI ~~in~~ for the YRD region was updated using ~~emission factor~~EFs derived mainly from local measurements ~~in this study~~. In addition to the conventional pollutants, 424 NMVOCs and 43 $PM_{2.5}$ components were also included in the inventory. ~~Source~~ The EI ~~categories~~ was refined ~~were divided~~ into ~~4~~ four main categories ~~levels and~~comprising 259 specific sources. The results indicated that the total emissions of SO_2, NO_x, CO, NMVOCs, PM_{10}, $PM_{2.5}$, and NH_3 in the YRD region in 2017 ~~are~~ were 1,552, 3,235, 38,507, 4,875, 3,770, 1,597, and 2,467 Gg, respectively. Overall, ~~the~~ SO_2 and NO_x emissions estimated in this study ~~are~~ were lower than the ~~existing~~ previous EIs such as the MEIC. ~~The s~~Substantial reductions in emissions from power plants and boilers ~~,~~ in recent years ~~are~~ were a significant factor~~the main reason~~. The NMVOC emissions ~~is~~ were also slightly lower than the results of ~~the~~ previous studies~~,~~. This was ~~which is~~ mainly due ~~to the fact that this study~~ the use~~s~~ of ~~emission factor~~EFs refined to the specific ~~process~~ sectors~~segments~~ in this study, which ~~are~~ were generally ~~usually~~ lower than the comprehensive ~~emission factor~~EFs used elsewhere. ~~Due to the consideration of dust sources,~~ PM_{10} and $PM_{2.5}$ emissions were respectively ~~are~~ 1.7 times and 0.5 times higher than the MEIC, ~~respectively~~due to the inclusion of dust sources. The NH_3 emissions ~~of~~ from this study~~,~~ ~~are~~ estimated using localized ~~emission factor~~EFs, ~~and the results are~~were significantly higher than those of previous studies.

SO_2 and CO emissions ~~are~~ were mainly from boilers in the region, accounting for 49% and 73% of the total. Mobile sources dominated ~~the~~ NO_x emissions from anthropogenic sources ~~in the YRD region~~, accounting for 57% of the total. NMVOC emissions were ~~mainly~~ mostly ~~come~~ from industrial sources, accounting for 61% of the total. The main contributing industrial sectors ~~are~~ were chemical manufacturing and solvent-use sources like furniture manufacturing, appliance manufacturing, textiles, ~~package~~ packaging and printing, and machinery manufacturing. Dust sources were responsible for 55% and 28% of PM_{10} and $PM_{2.5}$ ~~come from dust sources~~, respectively.

Agricultural sources accounted for 91% of NH_3 emissions.

The ~~Major~~main $PM_{2.5}$ species emitted from anthropogenic sources in the YRD region ~~are~~ were OC, Ca, Si, PSO_4 and EC, which accounted for 9.0%, 7.0%, 6.4%, 4.6% and 4.3% of total primary $PM_{2.5}$ emissions respectively. The main species of NMVOCs are aromatic hydrocarbons, accounting for 25.3% of the total. OVOCs also ~~occupy~~ made up a relatively high proportion of NMVOC, accounting for 21.9% of the total. Among ~~them~~these, aldehydes, ketones, alcohols, and esters accounted for 5.0%, 4.4%, 9.0% and 3.5%~~,~~ respectively. Toluene ~~has~~ had the highest ~~comprehensive contribution to ozone~~OFP and SOAP ~~formation potentials, and the others~~while other contributors included~~ are~~ 1,2,4-trimethylbenzene, m,p-xylene, propylene, ethene, o-xylene, ethylbenzene~~ and so on~~. Industrial process and solvent use sources ~~are~~ were the main sources of OFP~~ozone~~ and SOAP~~ formation potential,~~ followed by motor vehicles. Among the industrial sources, chemical manufacturing, rubber & plastic manufacturing, appliance manufacturing and textiles all ~~have~~ made ~~relatively outstanding~~significant contributions.

In recent years, the ambient air quality in the YRD region has improved significantly. At the same time, ~~the~~ contributions ~~of~~ to air pollutants from emissions have ~~also been~~changed subtly ~~changing in these years~~. While ~~The~~ emissions of primary pollutants such as SO_2 and NO_x from power plants and boilers have ~~dropped~~ decreased significantly, ~~but~~ the contribution ~~of~~ from mobile sources ~~has~~ have become ~~increasingly prominent~~important~~,~~. Furthermore,~~and the~~ emissions of reactive organic compounds from industrial sources ~~are~~ were still at a high level, resulting in ~~outstanding~~ elevated secondary pollution issues. ~~We hope that t~~The high-resolution EI ~~in detailed sources and species~~ established in this study ~~can~~should provide scientific guidance for the future ~~joint~~ control of air pollutants in the YRD region~~,~~ of China.

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2018YFC0213800), and the Shanghai Municipal Bureau of Ecology and Environment Fund Project (Grant No. 2019-01).

Data availability

The gridded emissions of air pollutants from various sources for ~~Yangtze River Delta~~the YRD region developed by this study at a horizontal resolution of 4 × 4km and a summary table of emissions by cities and sources can be downloaded from website (https://doi.org/10.6084/m9.figshare.13340648~~https://doi.org/10.6084/m9.figshare.12720938~~). Additional related data is available upon request by contacting the corresponding author (Cheng Huang; huangc@saes.sh.cn).

Author contribution

C.H., H.W. and C.C. designed the research. J.A., Y.H., X.W., R.Y., Q.W., Y.L., and C.X. performed the research. H.W. and S.J. collected the NMVOC species data. L.Q., M.Z., and S.Z. collected the $PM_{2.5}$ species data. C.H., Q.H., and J.L. supported the emission factor data. J.A., Y.H., C.H., X.W., H.L., Y.Z., Y.C., and C.C. analyzed the results. J.A., Y.H, C.H., and X.W. wrote the paper.

Competing interests

The authors declare that they have no conflict of interest.

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