



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

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14 **Abstract:** A high-resolution air pollutant emission inventory in the Yangtze River Delta
15 (YRD) region was updated for the year 2017 using emission factors and chemical
16 speciation mainly from local measurements in this study. The inventory includes 424
17 NMVOC species and 43 PM_{2.5} species, which can be subdivided into 259 specific
18 source categories. The total emissions of SO₂, NO_x, CO, NMVOCs, PM₁₀, PM_{2.5}, and
19 NH₃ in the YRD region in 2017 are 1,552, 3,235, 38,507, 4,875, 3,770, 1,597, and 2,467
20 Gg, respectively. SO₂ and CO emissions are mainly from boilers, accounting for 49%
21 and 73%, respectively. Mobile sources dominate the NO_x emissions and contribute 57%
22 of the total. VOC emissions mainly come from industrial sources, occupying 61%. Dust
23 sources take up to 55% and 28% of PM₁₀ and PM_{2.5} emissions, respectively.
24 Agricultural sources account for 91% of NH₃ emissions. Major PM_{2.5} species are OC,
25 Ca, Si, PSO₄ and EC, accounting for 9.0%, 7.0%, 6.4%, 4.6% and 4.3% of total PM_{2.5}

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26 emissions. The main species of VOCs are aromatics, accounting for 25.3%. OVOCs
27 contribute 21.9% of total VOC emissions. Toluene has the highest comprehensive
28 contribution to ozone and SOA formation potentials, and the others are 1,2,4-
29 trimethylbenzene, m,p-xylene, propylene, ethene, o-xylene, ethylbenzene and so on.
30 Industrial process and solvent use sources are the main sources of ozone and SOA
31 formation potential, followed by motor vehicles. Among industrial sources, chemical
32 manufacturing, rubber & plastic manufacturing, appliance manufacturing and textile
33 have made relatively outstanding contributions. The inventory can provide scientific
34 guidance for future joint control of air pollutants in the YRD region, China.

35 **Key words:** emission inventory; PM_{2.5} species; VOC species; the Yangtze River Delta
36 region; air pollutant emissions

37 1. Introduction

38 Air pollutant emissions from anthropogenic sources have attracted wide attentions
39 due to their adverse impacts on air quality (Monks et al., 2009), human health (Guan et
40 al., 2016; Requia et al., 2018), and climate change (Fiore et al., 2012). Air pollutants
41 include gaseous compounds, such as sulfur dioxide (SO₂), nitrogen oxides (NO_x),
42 carbon monoxide (CO), nonmethane volatile organic compounds (NMVOCs),
43 ammonia (NH₃), etc., and particles with different sizes including PM₁₀ and PM_{2.5},
44 whose aerodynamic diameter less than 10 and 2.5 μm. NMVOCs and PM_{2.5} are
45 aggregates of various chemical compositions. NMVOCs contains thousands of species
46 such as alkanes, alkenes, aromatics and oxygenated organic compounds (OVOCs), and
47 is the key precursor of ozone (O₃) and secondary organic aerosols (SOA). PM_{2.5} is
48 composed of a complex mixture, including sulfate (SO₄²⁻), nitrate (NO₃⁻), ammonium
49 (NH₄⁺), organic carbon (OC), element carbon (EC), and various elements, which
50 degrades visibility and threaten public health (Qiao et al., 2014; Liang et al., 2016).

51 Emission inventory (EI) is a key fundamental for air pollution source
52 apportionment, air quality forecasting and decision-making of air pollutant control
53 measures. In the last two decades, emission inventories have been improved both in



54 global and regional scales. According to recently reported inventories, anthropogenic
55 emissions still show growing trends in global scale (Janssens-Maenhout et al., 2015;
56 Klimont et al., 2017; Crippa et al., 2018; Hoesly et al., 2018). China's air pollutant
57 emission intensity is at a higher level in the world due to the increasing energy
58 consumption, urbanization and motorization. However, China's emissions are
59 undergoing dramatic changes especially in key regions, such as the Jing-Jin-Ji (JJJ),
60 Yangtze River Delta (YRD), and Pearl River Delta (PRD) regions, with the efforts of
61 air pollution prevention and control measures in these years (Cai et al., 2018; Zheng et
62 al., 2018). Updating the EI has become very necessary.

63 The YRD region is located in East China and covers Jiangsu, Zhejiang, Anhui, and
64 Shanghai, which has the most intensive economy, population and transportation and
65 results in its highest emission level in China. According to the new released data by the
66 Multi-resolution EI for China (MEIC, <http://meicmodel.org/>), the emission intensities
67 per unit area of SO₂, NO_x, NMVOCs, PM_{2.5}, and NH₃ in the YRD region are 2.3, 4.5,
68 5.2, 3.4, and 3.0 times of the national average. We have established an EI for the core
69 cities in the YRD region in 2007 (Huang et al., 2011). After that, Fu et al. (2013) updated
70 the EI for Jiangsu, Zhejiang, and Shanghai in the YRD region in 2010. In the last five
71 years, only individual provinces or part of sources were updated in the YRD region
72 (Fan et al., 2016; Zhou et al., 2017; Huang et al., 2018a; Wang et al., 2018b; Chen et
73 al., 2019; Yang and Zhao, 2019). Due to the implementation of air pollution prevention
74 and control measures, PM_{2.5} pollution in the YRD region has been significantly
75 alleviated, and the regional energy, industry and vehicle fleet are undergoing great
76 changes in recent years (Zheng et al., 2016; Wang et al., 2017a; Zhang et al., 2017a).
77 Updating the activity levels for detailed sources in the YRD region can help to simulate
78 air quality and guide emission reduction measures more accurately.

79 Besides of activity levels, speciation profiles of PM_{2.5} and NMVOC emissions are
80 also very important to improve the performance of chemical transport models (CTMs)
81 in simulating O₃ mixing ratios and PM_{2.5} concentrations. Source profiles from USEPA's



82 SPECIATE database has been commonly used to conduct source apportionment and
83 create speciated EI for air quality modeling since the 1990s (USEPA, 2009; Simon et
84 al., 2010). However, emission characteristics of anthropogenic sources have
85 considerable difference between different regions. Differences in fuel properties,
86 operating conditions, raw materials, and after-treatment techniques can result in
87 inconsistent speciation profiles for PM_{2.5} and NMVOCs. A previous study indicates that
88 using the speciation profiles from SPECIATE database leads to relatively poor model
89 performance for trace elements at an urban site in Beijing, China (Ying et al., 2018).
90 The emission estimates for individual NMVOC species differ between one and three
91 orders of magnitude for some species when different sets of speciation profiles are used,
92 which will lead to significant deviations in O₃ and SOA simulation (Li et al., 2014;
93 Zhao et al., 2017; Stroud et al., 2018; Wang et al., 2018c). In view of its importance to
94 model performance, detailed and observation-based emissions of individual speciated
95 PM_{2.5} and NMVOCs have become critical.

96 In this study, we updated an anthropogenic air pollutant EI in the YRD region for
97 the year of 2017 using the emission factors (EFs) and PM_{2.5} and NMVOCs speciation
98 profiles mainly derived from local measurements. The pollutants include SO₂, NO_x, CO,
99 NMVOCs, PM₁₀, PM_{2.5}, and NH₃. In addition, 424 individual NMVOC species
100 including alkanes, alkenes, aromatics, haloalkanes, and OVOCs and 43 PM_{2.5} species
101 including OC, EC, ions, and elements were included in the inventory. To obtain detailed
102 sources of emissions, the EI was refined to 259 specific source categories in 4 levels
103 based on the fuel types, industrial sectors, equipment types, and emission level, etc.
104 Finally, the EI was validated using Community Multiscale Air Quality (CMAQ) model
105 and observations in the YRD region in 2017.

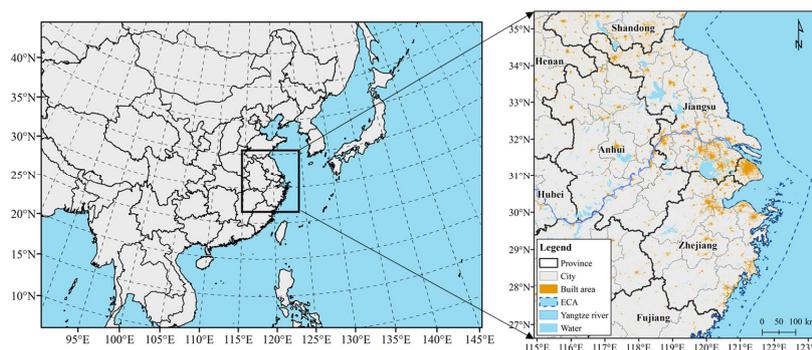
106 **2. Materials and methods**

107 **2.1 Domain of this study**

108 The YRD region in this study covers three provinces, including Jiangsu, Zhejiang,
109 and Anhui provinces, as well as Shanghai municipality. The region has a land area of



110 approximately 350,400 km², accounting for 3.7% of the whole China. However, the
111 whole region produced a gross domestic product (GDP) of 2,893 billion USD, about
112 24% of the total national GDP in 2017, and growing at a rate of about 9.3% per year in
113 the last decade (NBSC, 2018). Correspondingly, the region consumed 717.8 million tce
114 of energy, about 17% of the national total in 2017. Coal is the main energy type in this
115 region, contributing about 60% of total energy consumption (NBSC, 2018). The
116 automobile population reached 40.9 million in 2017, occupying 19.6% of the total in
117 China. The region also has a high concentration of traditional industries, producing
118 13.9%, 11.3%, 9.0%, 18.2% and 19.1% of the total products of gasoline, diesel, coke,
119 cement, and crude steel in China in 2017 (NBSC, 2018). Figure 1 shows the domain of
120 the YRD region in this study. The waters within the dashed line on the right figure are
121 China's ship emission control areas. The ship emissions mentioned in this study are the
122 summary of emissions in this region and inland waters in the YRD region.



123
124

Figure 1. The domain of the YRD region in this study.

125 2.2 Sources classification

126 A total of 241 categories of emission sources in 4 levels were divided in this study.
127 The first level is divided into 9 major sources, including stationary combustion sources,
128 industrial process sources, industrial solvent-use sources, mobile sources, dust sources,
129 oil storage and transportation sources, residential sources, waste treatment and disposal
130 sources, and agricultural sources. The second level has a total of 36 source categories,



131 mainly based on combustion facilities and industrial, transportation, residential, and
132 agriculture sectors. The third-level classification is mainly based on fuel, product and
133 material types, and contains a total of 127 categories. The fourth-level classification
134 includes combustion types, emission segments, and control levels. Detailed
135 classification is shown in Table S1 in the support information.

136 2.3 Emission estimation methods

137 The emissions of SO₂, PM₁₀, and PM_{2.5} from stationary combustion sources are
138 calculated using the mass balance method by Eq. (1) and (2). Other pollutant emissions
139 are calculated using the EF method, as shown in Eq. (3).

$$140 \quad E_{\text{SO}_2} = 2 \times S \times F \times C_s \times (1 - \eta_{\text{SO}_2}) \quad (1)$$

$$141 \quad E_{\text{PM}} = A \times F \times C_A \times P_{\text{ratio}} \times (1 - \eta_{\text{PM}}) \quad (2)$$

$$142 \quad E_{i,j} = AL_j \times EF_{i,j} \times (1 - \eta_{i,j}) \times 10^{-3} \quad (3)$$

143 Where, E_{SO_2} and E_{PM} represent the emissions of SO₂ and PM₁₀ or PM_{2.5} (t). S and A
144 represent fuel sulfur content and ash in fuel (%). F is the fuel consumption (t). C_s and
145 C_A are the conversion efficiencies from sulfur and ash to SO₂ and PM (%). P_{ratio} is the
146 mass percentage of PM₁₀ or PM_{2.5} in total PM. η_{SO_2} and η_{PM} represent the removal
147 efficiency of SO₂ and PM₁₀ or PM_{2.5}. $E_{i,j}$ represents the emissions of pollutant i from
148 source j (t). AL_j is the activity data of source j , such as fuel consumption, product output,
149 and raw material consumption, etc. $EF_{i,j}$ is the EF of pollutant i from source j (kg per
150 activity data). $H_{i,j}$ is the removal efficiency of pollutant i from source j .

151 Emissions from the industrial process sources are calculated using the EF method
152 shown in Eq. (3). Emissions from industrial solvent-use sources are calculated using
153 the mass balance method based on the consumption and VOC content of solvents, such
154 as paints, coatings, inks, adhesives, thinners, etc. A small amount of VOC remaining in
155 products, wastewater and waste was not considered in this calculation.

156 For motor vehicles, we use the International Vehicle Emission (IVE) model to
157 calculate the emissions. However, the EFs and activity data including driving conditions,



158 fleet composition, vehicle mileage travels (VMT), and meteorological parameters in the
159 model were localized via real-world measurements and surveys in this study. Non-road
160 machinery emissions are estimated with reference to the NONROAD model (USEPA,
161 2010), which is based on the fuel consumption and fuel-based emission factors. The
162 amount of fuel consumption is calculated based on the population, working hours and
163 fuel consumption rate per hour. Ship emissions are estimated using the approach based
164 on the Automatic Identification System (AIS) data. The detailed method has been
165 reported by Fan et al. (2016). Civil aviation aircraft source refers to aircraft emissions
166 under the land take-off (LTO) cycles, which include four operating modes, like
167 approaching, taxiing, taking-off, and climbing. SO₂ emission from civil aviation aircraft
168 source is estimated using mass balance method. The sulfur content in aviation fuel is
169 0.068%, which is the default value provided in a previous study (Wayson et al., 2009).
170 NO_x, CO, and NMVOC emissions are estimated using the EF method, which multiplied
171 the fuel consumption rate by the EFs. PM emission is calculated using the FOA3.0
172 method (Wayson et al., 2009). The rated thrust and working hour of the aircraft in each
173 LTO mode are referenced to the recommended parameters by the International Civil
174 Aviation Organization (ICAO). The climbing mode specified by the ICAO refers to the
175 altitude of about 1 km from the end of take-off to the top of the boundary layer. However,
176 the height of boundary layer in the actual atmosphere will change with the
177 meteorological conditions. In this study, a meteorological model (WRF-v3.9.1) was
178 used to simulate the boundary layer height to correct the time of climbing mode.
179 Detailed description of the methodology for aviation emission estimation is provided
180 in our previous study (Wang et al., 2018b).

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

184 2.4 Activity data sources

185 The activity data related to the industrial sources (including stationary combustion



186 sources, industrial process and solvent-use sources) of this study are mainly from the
187 2017 Environmental Statistics Database, which contains the information on fuel
188 consumption, product output, raw material consumption, and removal technology and
189 efficiencies. There are nearly 30,000 major point sources in the YRD region in the
190 database. Considering that environmental statistics do not include all industrial sources,
191 we take the difference between the total fuel consumption and product output in the
192 statistical yearbook and the sum of environmental statistics for each city as an area
193 source. To improve the accuracy of mobile source emissions, a number of local surveys
194 on the activity data (such as population, vehicle or machine type, fuel type, and
195 emission standard, etc.) were conducted for motor vehicles, non-road machinery, and
196 aviation aircrafts. The activity data of ships come from the AIS data for the East China
197 Sea in 2017. The activity data of area sources are derived from the statistical yearbooks
198 of cities in the YRD region. For the sources whose activity data are not recorded in the
199 statistical yearbooks (such as the number of construction sites, civil solvent usage,
200 catering, biomass burning, etc.), we make some estimations based on statistical data,
201 such as population, building area, and crop yield, etc. Table S2 in the support
202 information summarizes the emission estimation methods and activity data sources for
203 various sources and their reliability levels.

204 2.5 Determination of emission factors

205 The EFs of each specific emission source were determined by local measurements
206 (or surveys) in the YRD region, domestic EI guidebook of China (MEP, 2014), and
207 those recommended in USEPA's AP-42 (USEPA, 2002) and European's EMEP datasets
208 (EEA, 2013) in turn. To minimize the uncertainty of the EI, this study localizes the EFs
209 of 80 source categories, which include the majority of anthropogenic emission sources,
210 such as coal-fired power plants and boilers (Yao et al., 2009; Zhao et al., 2010; Wang
211 et al., 2011; Lou, 2014; Sun, 2015; Xu et al., 2018), petroleum refining and ferrous
212 metal manufacturing (Guo et al., 2017), gasoline and diesel vehicles (Huang et al., 2016;
213 Huang et al., 2017; Huang et al., 2018b; Huang et al., 2018c), non-road machinery (Fu



214 et al., 2012; Fu et al., 2013; Ge et al., 2013; Qu et al., 2015; Li et al., 2016), and
215 emissions from cooking (Wang et al., 2018a; Gao et al., 2019), livestock and poultry
216 breeding (Chen, 2017; Zhou, 2019), N-fertilizer application (Chen et al., 2017; Xia et
217 al., 2018), and biomass burning (Tang et al., 2014), etc. The NMVOC EFs for some
218 evaporation loss sources, like industrial and residential solvent-use sources and oil
219 storage and transportation sources, are estimated based on the results of field surveys
220 of some typical sources in the YRD region. For the sources that have not been measured
221 or investigated, the EFs recommended in the EI guidebook of China (MEP, 2014) are
222 preferred, followed by the recommended factors in the USEPA's AP-42 (USEPA, 2002)
223 and European's EMEP datasets (EEA, 2013). The EFs for each emission source and
224 their references are provided in Table S1 in the supporting information.

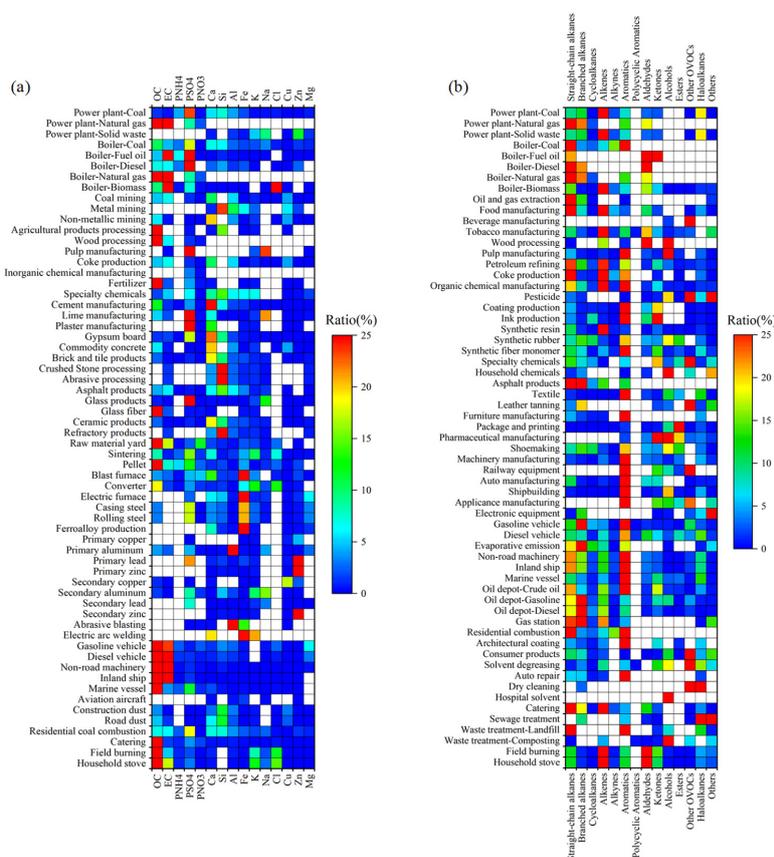
225 2.6 PM_{2.5} and NMVOC speciation

226 PM_{2.5} and NMVOC emissions are further split into individual species to simulate
227 PM_{2.5} chemical components and O₃ mixing ratios in the atmosphere. There are 43
228 chemical species in PM_{2.5}, including OC, EC, sulfate (PSO₄), nitrate (PNO₃),
229 ammonium (PNH₄) and 36 elemental components such as Na, Mg, K, Ca, Al, and Si,
230 etc. Additional species such as particulate water (H₂O), noncarbon organic matter
231 (NCOM), metal bound oxygen (MO), and other unspciated PM_{2.5} (PMO) are
232 calculated according to the method introduced by Reff et al. (2009). There are 424
233 species of VOCs, including 96 alkanes, 45 alkenes and alkynes, 44 aromatics, 164
234 OVOCs, 43 haloalkanes, and 32 other organic compounds.

235 The method for determining the PM_{2.5} and NMVOC source profiles is similar to
236 that for EFs. The results of local measurements are prioritized in this study, followed
237 by domestic measurements in previous studies, and finally the USEPA's SPECIATE4.4
238 database (Hsu et al., 2014). To enhance the representativeness of source profiles in the
239 inventory, the PM_{2.5} chemical compositions of 34 sources and the NMVOC chemical
240 compositions of 64 sources were localized according to the measurements in the YRD
241 region. The source categories of localization for PM_{2.5} profiles include power plants,



242 coal-fired boilers, ferrous metal manufacturing, gasoline and diesel vehicles, non-road
243 machinery, ships, catering, and biomass burning, etc. (Zheng et al., 2013; Tang et al.,
244 2014; Huang et al., 2016; Xu et al., 2018). The localized NMVOC sources include coal
245 combustion, gasoline and diesel vehicles, ships, catering, biomass burning, and the
246 majority of industrial process and solvent-use sources, like petroleum refining, coke
247 production, chemical manufacturing, textile, furniture manufacturing, package and
248 printing, auto manufacturing, shipbuilding, and architectural coating, etc. (Wang et al.,
249 2014a; Wang et al., 2014b; Wang et al., 2016; Wang et al., 2017b; Wang et al., 2017c;
250 Huang et al., 2018d; Gao et al., 2019). Detailed information for the references, samples,
251 and sampling and analytical methods for the sources are represented in Table S3. For
252 the species which cannot be analyzed by the analytical methods, we supplement the
253 mass fractions of these species from the SPECIATE database. Figure 2 and Figure 3
254 show the PM_{2.5} and NMVOC speciation profiles of major sources in the YRD region,
255 respectively.



256

257 **Figure 2.** The speciation profile of PM_{2.5} (a) and NMVOCs (b) for major emission sources.

258 **2.7 Spatial distributions**

259 Emissions from industrial sources, including power plants, boilers, industrial
 260 process and solvent-use sources, were allocated based on their latitude and longitude
 261 coordinates from Environmental Statistics Database. Vehicle emissions were
 262 determined based on the mileage sharing of various vehicle types on different levels of
 263 roads. The composition of traffic flow on different levels of roads was obtained from
 264 the surveys in Shanghai and Hangzhou (Huang et al., 2015; Yang et al., 2017). The
 265 approach of spatial allocation for road dust was consistent with vehicle emissions. The
 266 spatial distribution of emissions from non-road machinery varies in different ways
 267 depending on the type of machinery. The emissions from construction and agricultural
 268 machinery were allocated according to the built-up and farmland areas in the 2015 land



269 use data released by European Space Agency (ESA) ([https://www.esa-landcover-](https://www.esa-landcover-cci.org/)
270 [cci.org/](https://www.esa-landcover-cci.org/)). Emissions from port and factory machinery, and airport ground handling
271 equipment were allocated according to their latitude and longitude coordinates.
272 Emissions from residential sources were allocated based on 1 km resolved population
273 distribution data. Those of agriculture sources were allocated based on the farmland
274 areas in the land-use data.

275 2.8 Uncertainty analysis

276 The uncertainty is mainly derived from the activity data and EFs in the EI. In this
277 study, we classify the coefficients of variation of the activity data and EFs of each
278 source into seven grades in the range of 2%–100% based on expert judgment. The
279 coefficient of activity data is determined based on the data source. The environmental
280 statistic data with individual source information is assigned the lowest coefficient of
281 uncertainties, while the estimated activity data based on statistical yearbooks such as
282 biomass burning are assigned the highest values. The EFs derived from local
283 measurements in the YRD region with large samples are assigned the lowest values,
284 while those from USEPA's or European's datasets are assigned high coefficients.
285 Detailed analysis method can be found in our previous study (Huang et al., 2011).

286 3. Results and discussion

287 3.1 Emission and source contributions

288 3.1.1 Emissions and their comparisons with previous studies

289 The total emissions of SO₂, NO_x, CO, NMVOCs, PM₁₀, PM_{2.5}, and NH₃ in the
290 YRD region for the year of 2017 were 1,552, 3,235, 38,507, 4,875, 3,770, 1,597 and
291 2,467 Gg, respectively. If ship emissions were not included, the air pollutant emissions
292 above would be 1,437, 2,936, 38,486, 4,867, 3,754, 1,583 and 2,467 Gg, respectively.
293 Detailed information of air pollutant emissions for each city is shown in Table S1 in the
294 Supplement.

295 Table 1 shows the emissions in the YRD region estimated in this study and their



296 comparisons with previous studies. SO₂ emissions were close to the result in MEIC
297 2016, and were much lower than those reported in other studies in the past few years.
298 Emission reductions on coal-fired facilities including power plants and boilers were the
299 main reason for the significant decline in SO₂ emissions (Zheng et al., 2018). NO_x
300 emissions were generally lower than the results in previous studies. Some modeling and
301 satellite studies verified that the NO_x emissions in previous studies were overestimated
302 partly due to the failure to consider the improved NO_x control measures for power sector
303 (Zhao et al., 2018; Sha et al., 2019). The NO_x emission factors for coal-fired power
304 plants and boilers in this study were derived from local measurements which were
305 generally lower than those in previous studies, so the NO_x emissions from power sector
306 were 47% lower than MEIC. CO emissions were higher than MEIC's results but close
307 to those reported by Sun et al. (2018a). NMVOC emissions for key sources in this study
308 were individually estimated base on "bottom-up" method, so the estimates were lower
309 than the others who used "top-down" approach. Another reason is the majority of
310 emission factors selected in this study were detailed into different process segments,
311 which are generally lower than the comprehensive factors for whole industrial sectors
312 in the previous studies. Since dust sources were not included in MEIC inventory, PM₁₀
313 and PM_{2.5} emissions estimated in this study were 1.7 and 0.5 times higher than the
314 results in MEIC, respectively. A previous study has pointed out that the existing NH₃
315 emissions in China were underestimated mainly due to the underestimate of NH₃
316 emission rates from fertilizer application and livestock and missing of some emission
317 sources (Zhang et al., 2017). Therefore, we used the local measured NH₃ emission
318 factors for fertilizer application and part of livestock breeding in the YRD region instead
319 in this study. Another difference came from transportation sector. NH₃ emissions from
320 transportation sector were 2.8 times higher than those in MEIC when localized NH₃
321 emission factors form light-duty gasoline vehicles (Huang et al., 2018) were used in
322 this study. In addition, NH₃ slip from selective catalyst reduction (SCR) devices in
323 power sector was also considered in this study. However, this emission source has not



324 been included in previous studies.

325 **Table 1.** Air pollutant emissions in the YRD region in this study and their comparisons with other
 326 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	1437	2936	38486	4867	3754	1583	2467
	MEIC	2016	1136	3753	19560	5527	1374	1025	1153
	Simayi et al., 2019	2016				4984			
	Sun et al., 2018a	2015	3050	4160	30210	5490			
	Zhang et al., 2017b	2015							1632
	Wu et al., 2018	2013				6198			
Shanghai	This study	2017	57	225	1393	418	124	56	54
	MEIC	2016	168	345	1192	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
	Wu et al., 2018	2013				838			
	Fu et al., 2013	2010	260	453		422	86	59	65
Jiangsu	This study	2017	619	1165	17309	2056	1440	577	1093
	MEIC	2016	468	1586	8191	2128	516	388	532
	Simayi et al., 2019	2016				2024			
	Sun et al., 2018a	2015	1230	1700	13780	2000			
	Zhang et al., 2017b	2015							703
	Wu et al., 2018	2013				2240			
	Zhou et al., 2017	2012	1142	1642	7680	1747	1394	941	1100
	Fu et al., 2013	2010	1126	1257		1759	619	401	976
Zhejiang	This study	2017	339	676	7036	1484	775	308	363
	MEIC	2016	280	867	3779	1671	219	151	159
	Simayi et al., 2019	2016				1624			
	Sun et al., 2018a	2015	730	980	5110	1810			
	Zhang et al., 2017b	2015							257
	Wu et al., 2018	2013				2214			
	Fu et al., 2013	2010	762	1067		1641	301	184	398
Anhui	This study	2017	422	869	12748	910	1415	642	957
	MEIC	2016	221	954	6398	1045	570	435	437
	Simayi et al., 2019	2016				608			
	Sun et al., 2018a	2015	540	1010	9070	1100			
	Sun et al., 2018b	2015	434	688				323	422
	Zhang et al., 2017b	2015							622
	Wu et al., 2018	2013				906			



327 3.1.2 Source contributions

328 Figure 3 shows the contributions of emission sources divided by different source
329 categories (a), industrial sectors (b), and mobile source types (c). Detailed information
330 of the emissions from each source was provided in Table S5. SO₂ and CO emissions
331 were mainly from boilers, accounting for 49% and 73% of the total, respectively.
332 Notably the emission contributions of power plants were much lower than those in other
333 inventories (MEIC, <http://meicmodel.org/>; Zhou et al., 2017), resulting mainly from the
334 significant reduction in power plant emissions due to the implementation of ultra-low
335 emission reduction measures in recent years (Wu et al., 2019; Zhang et al., 2019).

336 Mobile sources dominated the NO_x emissions in the YRD region, which
337 contributed 57% of the total. This estimate was generally higher than the proportion of
338 mobile sources in MEIC and other inventories (Zhou et al., 2017; Sun et al., 2018a).
339 Emission control measures for power plants played an important role in reducing their
340 contributions on NO_x emissions. In addition, other studies did not include ship
341 emissions, which accounted for 16% of NO_x emissions from mobile sources in the YRD
342 region, as shown in Figure 3(c). Another reason that cannot be ignored was the NO_x
343 emission factors from gasoline and diesel vehicles were modified based on local
344 measurements in this study, which were generally higher than those recommended by
345 MEP (2014). Some real-world measurements based on portable emission measurement
346 system (PEMS), on-road chasing, and tunnel experiments also indicate that the NO_x
347 emissions from vehicles in China were higher than expected due to the existence of
348 high-emitting vehicles (Wu et al., 2012; Huang et al., 2017; Song et al., 2018; Wen et
349 al., 2019).

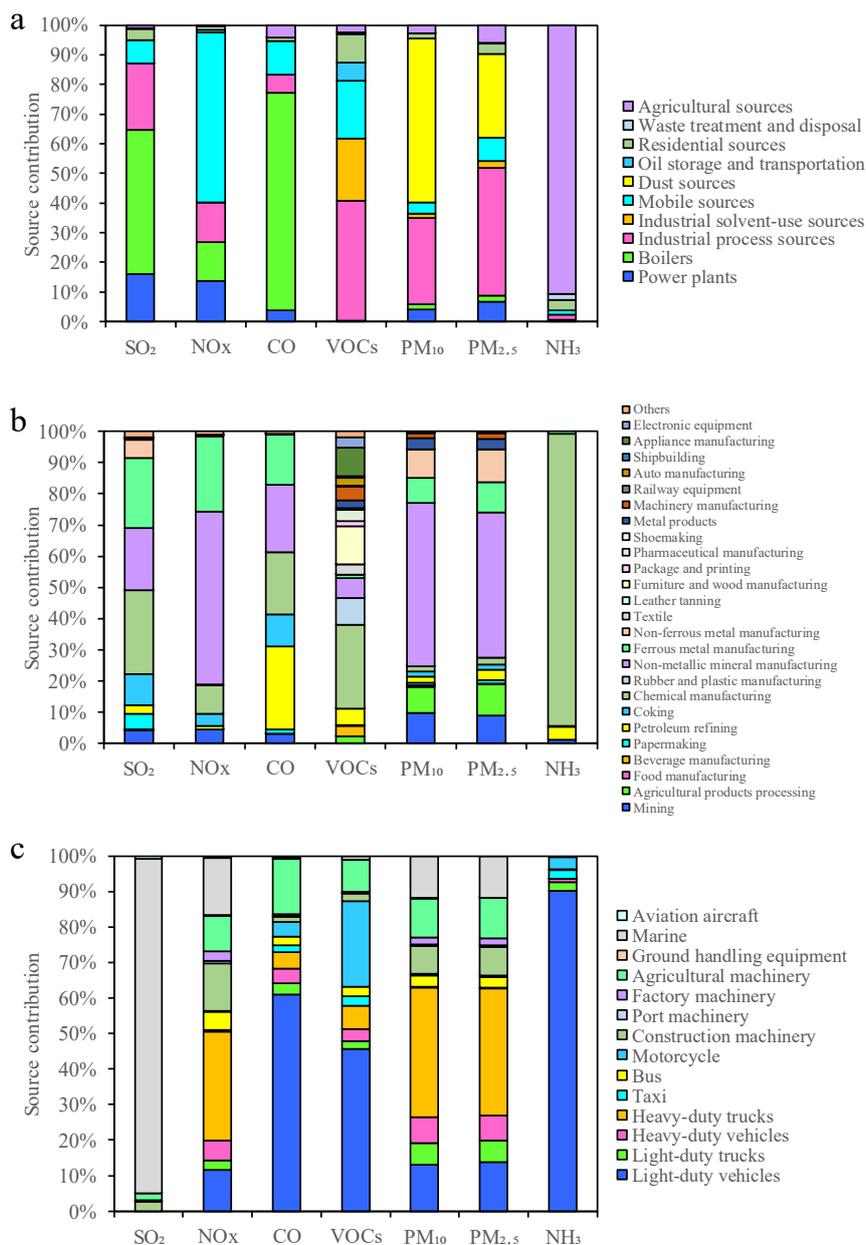
350 VOC emissions were mainly contributed by industrial sources, accounting for 61%
351 of the total, of which industrial process and solvent-use sources accounted for 34% and
352 27%, respectively. Mobile and residential sources contributed 20% and 10%,
353 respectively. Dust sources were main contributors to PM₁₀ and PM_{2.5} emissions,
354 occupied 55% and 28%, respectively. Agricultural sources contributed up to 91% of



355 NH₃ emissions. In addition, residential and mobile sources contributed 3% and 1% of
356 NH₃ emissions, respectively. Although NH₃ emission factors from vehicles have been
357 considered in this study, their NH₃ emission was still significantly lower than
358 agricultural sources. However, vehicle emissions were mainly concentrated in urban
359 areas, and their contribution to NH₃ emissions in urban areas would be considerable.

360 Industrial SO₂ and CO emissions mainly came from five major sectors, including
361 petroleum refining, coking, chemical manufacturing, non-metallic mineral
362 manufacturing, and ferrous metal manufacturing. Non-metallic mineral manufacturing
363 and ferrous metal manufacturing dominated the industrial NO_x, PM₁₀, and PM_{2.5}
364 emissions. The top five sectors of industrial VOCs emissions in the YRD region were
365 chemical manufacturing, Furniture and wood manufacturing, Appliance manufacturing,
366 rubber and plastic manufacturing, and non-metallic mineral manufacturing, accounting
367 for 27%, 12%, 9%, 9%, and 6% of the total, respectively. Chemical manufacturing
368 contributed the majority of industrial NH₃ emissions in the YRD region.

369 The YRD region has the largest port group in the world, so the emissions from the
370 transportation of ships and heavy-duty trucks dominate the mobile source emissions.
371 Among them, ships accounted for 94%, 16%, 12%, and 12% of mobile source SO₂,
372 NO_x, PM₁₀, and PM_{2.5} emissions, and heavy-duty trucks occupied 31%, 37%, and 36%
373 of mobile source NO_x, PM₁₀, and PM_{2.5} emissions, respectively. Light-duty vehicles
374 contributed significantly to CO, VOCs, and NH₃ emissions, accounting for 61%, 46%,
375 and 90%, respectively. Non-road machinery accounted for 27%, 18%, 12%, 21%, and
376 22% of NO_x, CO, VOCs, PM₁₀, and PM_{2.5} emissions from mobile sources, respectively.
377 Construction and agriculture machinery were major contributors.



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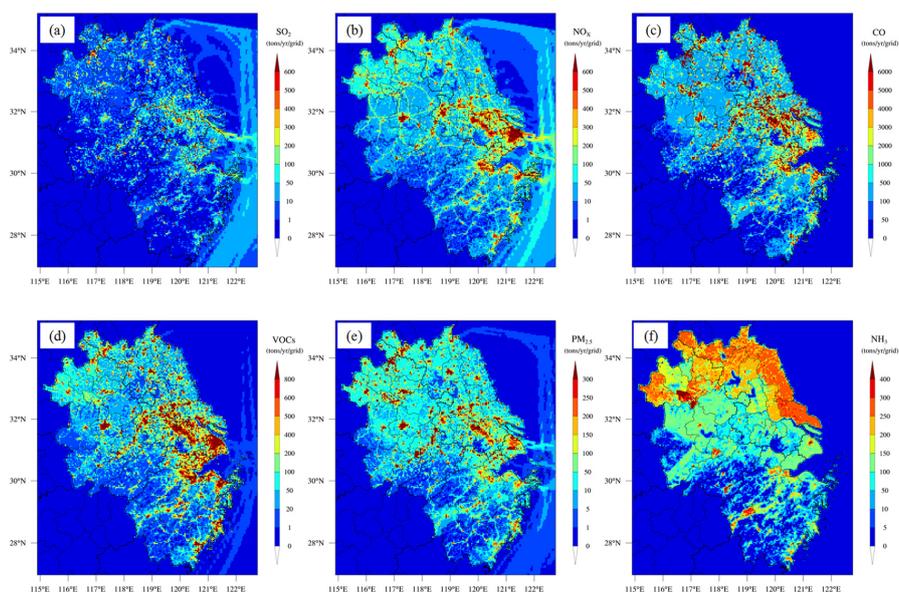
381

Figure 3. Source contributions of major air pollutant emissions in the YRD region. (a) Divided by major source categories; (b) Divided by detailed industrial sectors; (c) Divided by detailed mobile sources.



382 3.1.3 Spatial distribution

383 Fig. 4 shows the spatial allocation of SO₂, NO_x, CO, NMVOCs, PM_{2.5}, and NH₃
384 emissions in the YRD region. SO₂ emissions were mainly concentrated in the Yangtze
385 River and East China Sea estuary, where ships were densely populated. SO₂ emissions
386 along the Yangtze River and in the cities of northern Anhui and Jiangsu provinces were
387 also dense, mainly from power plants and boilers in these regions. The spatial
388 distribution of NO_x and NMVOCs was similar, mainly concentrated along the Yangtze
389 River and Hangzhou Bay, where the industries and logistics were most developed. CO
390 and PM_{2.5} emissions were mainly concentrated in the built-up areas of cities due to
391 intensive road traffic and human activities such as construction sites. NH₃ emissions
392 were relatively high in northern Anhui and Jiangsu provinces, resulting mainly from
393 their developed agriculture. The contribution of NH₃ emissions from residential and
394 mobile sources has led to higher NH₃ emission densities for large cities such as
395 Shanghai.

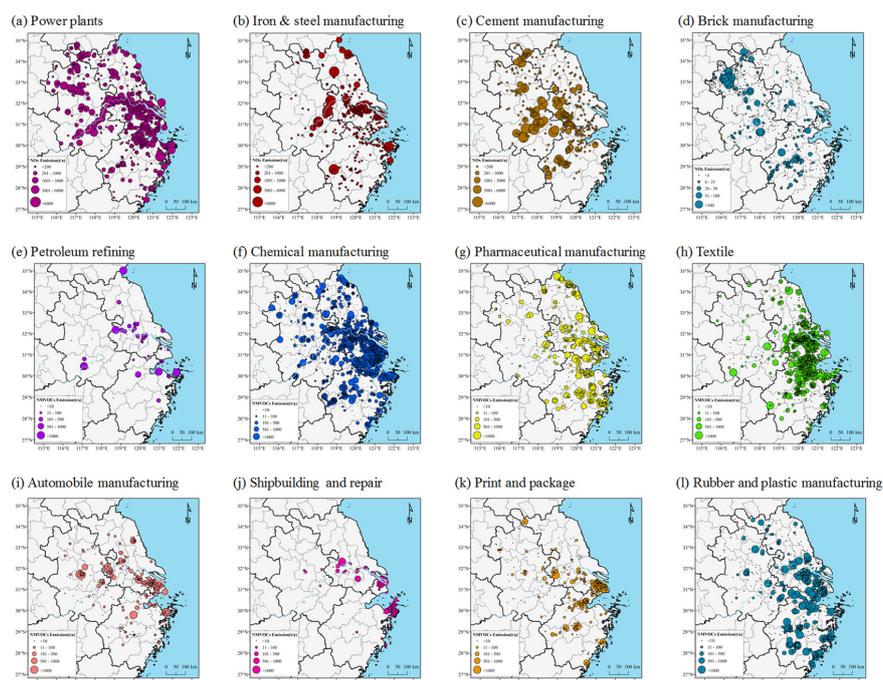


396
397 **Figure 4.** Spatial distribution of major air pollutant emissions in the YRD region. (a)~(f) refer to
398 SO₂, NO_x, CO, VOCs, PM_{2.5}, and NH₃ in turn.

399 Figure 5 shows the spatial distribution of major industrial sectors of NO_x and



400 VOCs emissions in the YRD region. There were large differences in the spatial
401 distribution of different industrial sectors. The power plants were mainly distributed
402 along the Yangtze River and Hangzhou Bay and the northern part of the YRD region.
403 The iron & steel manufacturing sector was concentrated along the Yangtze River.
404 Cement and brick manufacturing sectors were mainly distributed in the western and
405 northern regions of the YRD. In comparison, the key sectors of VOC emissions (Figure
406 5(e)~(l)) were mainly concentrated in the central and eastern regions of the YRD,
407 including Shanghai, Suzhou, Wuxi, Changzhou, Nanjing, Hangzhou, Ningbo, Jiaxing,
408 and Shaoxing, etc., which also had the strongest NO_x emission intensities in the YRD
409 region. High intensities of NO_x and VOC emissions are the key factors leading to
410 serious pollution of ozone and secondary particulate matter in this region (Li et al., 2018;
411 Li et al., 2019). Refining the specific industrial sectors of emissions can help to find out
412 the detailed sources inducing air pollution.



413
414

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



415 3.1.4 Uncertainty assessment

416 The inventory was compiled using a “bottom-up” approach based on local
417 emission factors and activity data in the region. The activity data of industrial sources,
418 including fuel consumption, sulfur content, ash content, raw material used, and control
419 efficiency, were collected from Environmental Statistics Database. Emission factors
420 from some key sources, such as coal-fired power plants and boilers, iron & steel
421 manufacturing, gasoline and diesel vehicles, non-road machinery, catering, and
422 agricultural sources, etc., have been modified based on the local measurements. These
423 all help to reduce the uncertainty of the emission estimates. Table 2 shows the
424 uncertainties of major sources at the 95% confidence interval in this inventory. The
425 average uncertainties of emissions from the YRD region were estimated as -29 to 36%
426 for SO₂, -28 to 33% for NO_x, -42 to 75% for CO, -44 to 68% for NMVOCs, -36 to 62%
427 for PM₁₀, -30 to 46% for PM_{2.5}, and -58 to 117% for NH₃. The uncertainty of this
428 inventory was reduced compared to our previous inventory for the YRD region (Huang
429 et al., 2011).

430 The uncertainty assessment indicates that the stationary combustion sources
431 including power plants and boilers were more reliable, because the emissions were
432 estimated based on the detailed activity data and local measurements. The uncertainties
433 of major industrial sectors, such as ferrous metal manufacturing, non-ferrous metal
434 manufacturing, and non-metallic mineral manufacturing, were greatly improved when
435 using detailed emission estimation approach for different process segments. In
436 comparison, the emissions from chemical manufacturing still have large uncertainties
437 since there are a large number of process segments and unorganized emissions. The
438 uncertainties of emissions from vehicles and non-road machinery in this study mainly
439 came from the activity data. Although their population could be obtained from the
440 statistical yearbooks, their mileage travels or working hours were still difficult to
441 estimate accurately. Dust emissions including construction and road dust have much
442 higher uncertainties due to less information of their activity data and emission factors



443 was available. Most of the area sources, like residential and agricultural sources, were
 444 estimated based on the activity data from statistical yearbooks, resulting in higher
 445 uncertainties of their emission estimates. Overall, using of emission estimation
 446 approach based on refined process segments and local measurements can help to reduce
 447 the uncertainties of EI. However, more detailed activity data and accurate emission
 448 factors are still very critical to improve the EI in the future.

449 **Table 2.** Uncertainty assessment of major emission sources in the YRD region.

Sources	SO ₂	NO _x	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%)

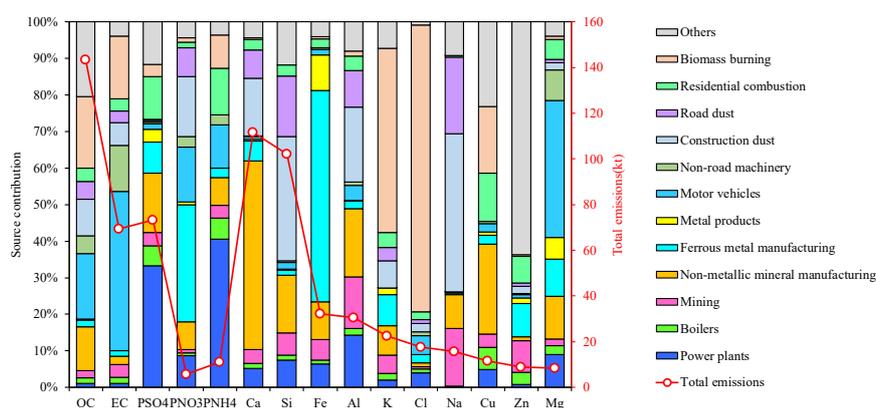
450 3.2 PM_{2.5} and VOC species emissions

451 3.2.1 PM_{2.5} species

452 Figure 6 shows the emissions and source contributions of major PM_{2.5} species in
 453 the inventory. OC, Ca, Si, PSO₄, and EC were top five components in primary PM_{2.5} in
 454 the YRD region, accounting for 9.0%, 7.0%, 6.4%, 4.6%, and 4.3% of PM_{2.5} emissions,
 455 respectively. There were large differences in the emission contributions of different
 456 PM_{2.5} species. Among the industrial sources, non-metallic mineral manufacturing
 457 sector had largest contributions to Ca, Si, and Al emissions, accounting for 51.6%,



458 15.9%, and 18.8% of these species, respectively. Ferrous metal manufacturing was the
459 main source of Fe emissions, accounting for 57.9%. Vehicles was major contributors to
460 OC and EC emissions, taking up 18.0% and 43.5%, respectively. K and Cl emissions
461 mainly came from biomass burning, accounting for 50.4% and 78.5%, respectively.
462 Construction dust was also an important source of PM_{2.5} components, accounting for
463 15.9%, 34.1% and 20.4% of Ca, Si, and Al emissions, respectively.



464
465 **Figure 6.** Emissions and source contributions of major PM_{2.5} species in the YRD region.

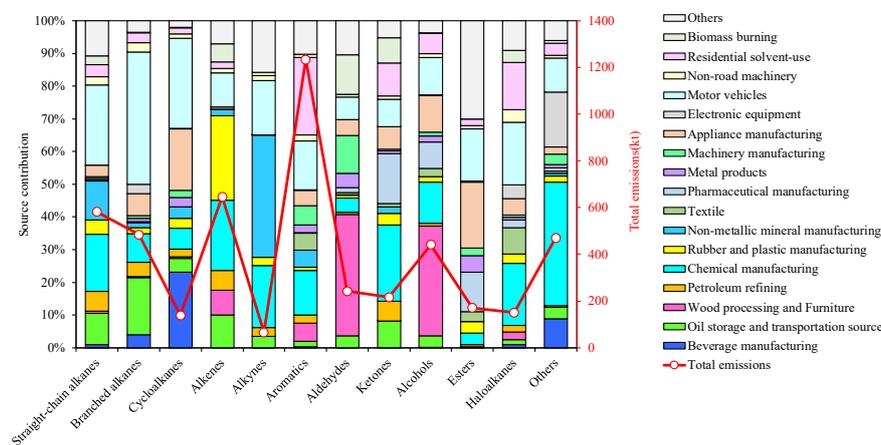
466 3.2.2 VOC species

467 Figure 7 shows VOC species emissions and their source contributions. The
468 aromatics dominated the VOC species, accounting for 25.3% of the total VOC
469 emissions in the YRD region, followed by the alkanes, occupying 24.7%. Among them,
470 the straight-chain, branched, and cycloalkanes took up 11.9%, 9.9%, and 2.8%,
471 respectively. OVOCs also accounted for a considerable proportion of VOC emissions
472 in the YRD region, about 21.9% of the total. Among them, the aldehydes, ketones,
473 alcohols and esters took up 5.0%, 4.4%, 9.0% and 3.5%, respectively. In addition, the
474 haloalkanes occupied about 3.1% of the total VOC emissions. The aromatics were also
475 dominant species in VOC emissions in the YRD region in a previous study reported by
476 Wu et al. (2017), even higher (40%) than the proportion in ours. The proportion of
477 OVOCs was quite close to our study, while the proportion of alkenes and haloalkanes
478 were generally lower than ours.



479 The chemical manufacturing accounted for a considerable proportion of various
480 VOC species in the YRD region, accounting for 12.7%, 21.5%, 13.7%, and 10.8% of
481 the alkanes, alkenes, aromatics, and OVOC emissions, respectively. Industrial solvent-
482 use sources, including furniture and wood processing, textile, package and printing,
483 pharmaceutical manufacturing, metal products, auto manufacturing, and appliance
484 manufacturing, etc., were also an important source of VOC emissions in this region,
485 which occupied 29.3% and 33.3% of the aromatics and OVOC emissions, while
486 residential solvent-use sources accounted for 23.7% and 4.9% of the aromatics and
487 OVOC emissions in the YRD region. Motor vehicles also have a very important
488 contribution to various VOC species in the YRD region, occupying 31.2%, 10.4%,
489 15.1%, and 10.5% of the alkanes, alkenes, aromatics, and OVOC emissions in the
490 region. Biomass burning contributed 12.0% of the aldehyde emissions, although it
491 accounted for only 2.5% of the total VOC emissions in the region.

492 Overall, the refinement of VOC source profiles can help to provide an important
493 support for assessing the impacts of VOC emissions on ambient air quality in the region.
494 However, there are still considerable differences of VOC composition in different
495 studies. It is necessary to strengthen the verification of VOC species emissions in the
496 future.



497
498 **Figure 7.** Emissions and source contributions of major VOC species in the YRD region.



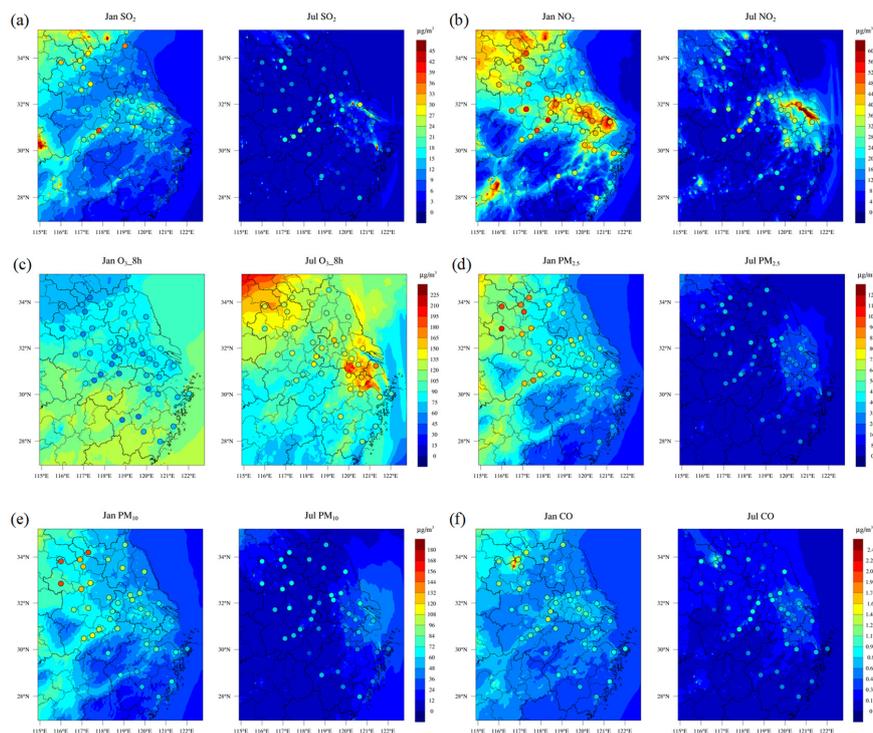
499 3.3 Model validation

500 To verify the reliability of the emission inventory, we used the Community
501 Modeling and Analysis System (CMAQ version 5.3) to simulate the concentrations of
502 SO₂, NO₂, PM_{2.5}, PM₁₀, O₃, and CO in the YRD region in January and July 2017, and
503 compared with the observation data in each city of the region. The Weather Research
504 and Forecast (WRF) version 3 supplied the meteorological field for CMAQ model. The
505 emission inventory developed in this study was used to produce the emission system in
506 the YRD region while emission beyond YRD was supplied by Multiresolution Emission
507 Inventory for China (MEIC-2016) (<http://www.meicmodel.org>). The anthropogenic
508 data was then combined with the biogenic data from Model for Emissions of Gases and
509 Aerosol from Nature (MEGAN version 2.10) as the final input of emission inventory
510 for the model. Figure S1 and Table S6 show the domain and settings of model system,
511 respectively. Detailed information is provided in the Supporting information Section 6.

512 Figure 8 compares the simulated concentration for SO₂, NO₂, PM_{2.5}, PM₁₀, O₃, and
513 CO in January and July 2017 in the YRD region with those of the observation data. The
514 simulated concentration distribution of various pollutants is consistent with the
515 observation results, which indicates the updated inventory generally reflects the
516 distribution of air pollution sources in the YRD region. Comparatively, the consistency
517 between the simulated concentration distribution and the observed results of the cities
518 in the central areas of the YRD region is stronger than those of the northern and southern
519 border areas. This is mainly because the concentration in the border areas is more
520 susceptible to the effects of emissions from the outer areas, which leads to greater
521 simulation results deviation. Detailed statistical results of the model performance for
522 simulating various pollutants in each city is shown in Table S7 of the supporting
523 information. Overall, the simulation results of O₃ were relatively high in January, while
524 SO₂, NO₂, PM_{2.5}, PM₁₀, and CO were relatively low. While in July, except that the O₃
525 simulation concentrations were slightly higher than the observed results, the average
526 NO₂ simulation values were consistent with the measured averages, other pollutants



527 were relatively lower.



528

529 **Figure 8.** Comparisons of simulated and observed (circles) monthly average concentrations of
530 SO₂, NO₂, daily maximum 8-hour of O₃ (O_{3_8h}), PM_{2.5}, PM₁₀, and CO in cities in the YRD region
531 in January and July 2017.

532 3.4 Ozone and SOA formation potentials

533 To characterize the regional ozone and SOA formation contributions of different
534 VOC species and their sources, we used ozone formation potential (OFP) and SOA
535 formation potential (SOAP) methods to estimate. OFP and SOAP are the sum of
536 individual VOC species emissions multiplied by maximum incremental reactivity (MIR)
537 and SOA yield, respectively. The MIR and SOA yield of individual VOC species was
538 referenced from previous studies (Carter, 1994; Wu and Xie, 2017).

539 Figure 9 shows the OFP and SOAP contributions from major VOC species,
540 emission sources, and industrial sectors in the region. In terms of individual species,
541 toluene is the most important species for both OFP and SOAP, which contributed 45.0%

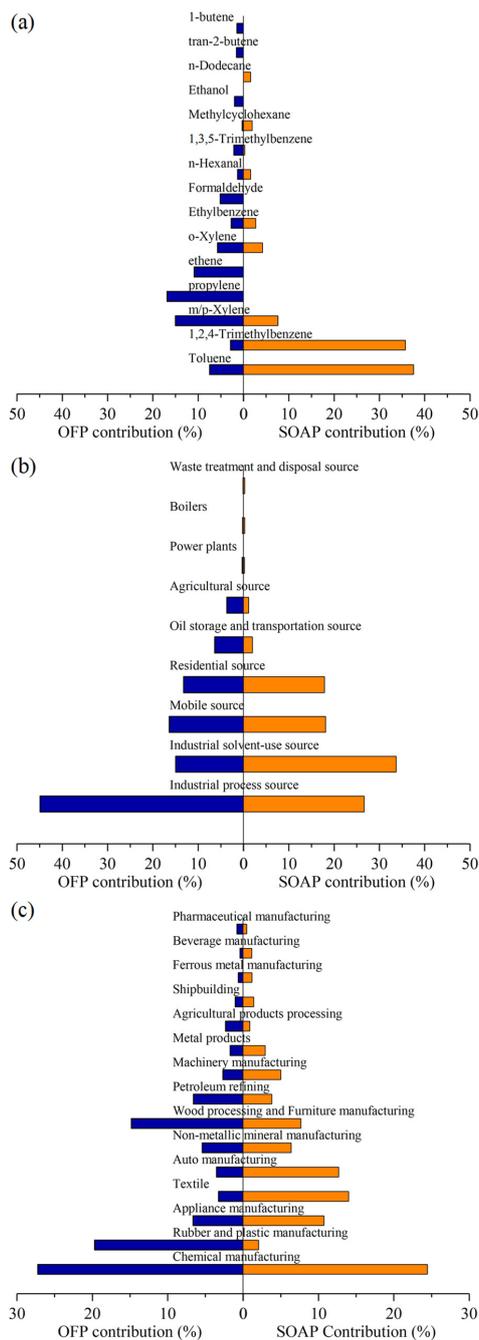


542 of the total (7.5% OFP and 37.5% SOAP), followed by 1,2,4-trimethylbenzene,
543 contributing 29.1% of the total (2.1% OFP and 27.0% SOAP). Others were m/p-xylene,
544 propylene, ethene, o-xylene, ethylbenzene, etc., their contributions to both OFP and
545 SOAP were 14.9%, 16.6%, 10.7%, 5.7%, and 2.7% in turn. Their OFP contribution was
546 relatively more prominent.

547 Industrial process sources dominated the OFP and SOAP in the region, which
548 contributed 44.9% and 26.7%, respectively. Industrial solvent-use sources followed,
549 with OFP and SOAP contributions of 15.0% and 33.8%, and their contribution to SOAP
550 even exceeded the industrial process sources. The contributions of motor vehicles to
551 regional OFP and SOAP were 13.9% and 13.5%, respectively, which was close to those
552 from residential solvent-use sources. These two sources were major contributors of
553 ozone and SOA formation in urban areas.

554 There are four major industrial sectors with significant potential contribution to
555 ozone and SOA production in the YRD region. The chemical manufacturing sector
556 contributed 16.4% and 14.8% of OFP and SOAP, respectively. The second was rubber
557 & plastic manufacturing sector, with a SOAP contribution rate of 11.8%, while its OFP
558 was relatively low, about 1.2%; the third and fourth were appliance manufacturing and
559 textile sectors, accounting for 10.5% and 10.4% of both OFP and SOAP contributions.

560 Based on the above, it can be concluded that the reduction of aromatic emissions
561 from industrial and vehicular sources were of vital importance for the YRD region,
562 especially for the high reactivity species, such as toluene, xylene, and trimethylbenzene,
563 etc., which should be the top priority on VOCs pollution control in the region.



564

565

566

Figure 9. Ozone and SOA formation potentials from different (a) VOC species, (b) emission sources, and (c) industrial sectors.



567 4. Conclusions

568 A high-resolution air pollutant emission inventory in the YRD region was updated
569 using emission factors mainly from local measurements in this study. In addition to the
570 conventional pollutants, 424 NMVOCs and 43 PM_{2.5} components were also included.
571 Source categories were divided into 4 levels and 259 specific sources. The results
572 indicate that the total emissions of SO₂, NO_x, CO, NMVOCs, PM₁₀, PM_{2.5}, and NH₃ in
573 the YRD region in 2017 are 1,552, 3,235, 38,507, 4,875, 3,770, 1,597, and 2,467 Gg,
574 respectively. Overall, the SO₂ and NO_x emissions estimated in this study are lower than
575 the existing EIs such as MEIC. The substantial reductions in power plants and boilers
576 in recent years are the main reason. The VOC emissions is also slightly lower than the
577 results of the previous studies, which is mainly due to the fact that this study uses
578 emission factors refined to the process segments, which are usually lower than the
579 comprehensive emission factors. Due to the consideration of dust sources, PM₁₀ and
580 PM_{2.5} emissions are 1.7 times and 0.5 times higher than MEIC, respectively. The NH₃
581 emissions of this study are estimated using localized emission factors, and the results
582 are significantly higher than those of previous studies.

583 SO₂ and CO emissions are mainly from boilers, accounting for 49% and 73% of
584 the total. Mobile sources dominate the NO_x emissions from anthropogenic sources in
585 the YRD region, accounting for 57% of the total. VOC emissions mainly come from
586 industrial sources, accounting for 61%. The main industrial sectors are chemical
587 manufacturing and solvent-use sources like furniture manufacturing, appliance
588 manufacturing, textile, package and printing, and machinery manufacturing. 55% and
589 28% of PM₁₀ and PM_{2.5} come from dust sources, respectively. Agricultural sources
590 account for 91% of NH₃ emissions.

591 Major PM_{2.5} species emitted from anthropogenic sources in the YRD region are
592 OC, Ca, Si, PSO₄ and EC, which account for 9.0%, 7.0%, 6.4%, 4.6% and 4.3% of total
593 primary PM_{2.5} emissions. The main species of VOCs are aromatics, accounting for
594 25.3%. OVOCs also occupy a relatively high proportion, accounting for 21.9%. Among



595 them, aldehydes, ketones, alcohols, and esters account for 5.0%, 4.4%, 9.0% and 3.5%,
596 respectively. Toluene has the highest comprehensive contribution to ozone and SOA
597 formation potentials, and the others are 1,2,4-trimethylbenzene, m,p-xylene, propylene,
598 ethene, o-xylene, ethylbenzene and so on. Industrial process and solvent use sources
599 are the main sources of ozone and SOA formation potential, followed by motor vehicles.
600 Among industrial sources, chemical manufacturing, rubber & plastic manufacturing,
601 appliance manufacturing and textile have made relatively outstanding contributions.

602 In recent years, the ambient air quality in the YRD region has improved
603 significantly. At the same time, the contributions of air pollutant emissions have also
604 been subtly changing in these years. The emissions of primary pollutants such as SO₂
605 and NO_x from power plants and boilers have dropped significantly, but the contribution
606 of mobile sources has become increasingly prominent, and the emissions of reactive
607 organic compounds from industrial sources are still at a high level, resulting in
608 outstanding secondary pollution issues. We hope that the EI in detailed sources and
609 species established in this study can provide scientific guidance for future joint control
610 of air pollutants in the YRD region, China.

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615 Fund Project (Grant No. 2019-01).

616 **Data availability**

617 The gridded emissions of air pollutants for Yangtze River Delta developed by this
618 study at a horizontal resolution of 4 × 4km can be downloaded from website
619 (<https://doi.org/10.6084/m9.figshare.12720938>). Additional related data is available
620 upon request by contacting the corresponding author (Cheng Huang;
621 huangc@saes.sh.cn).



622 **Author contribution**

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

628 **Competing interests**

629 The authors declare that they have no conflict of interest.

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