

# TITLE PAGE

## **Improved provincial emission inventory and speciation profiles of anthropogenic non-methane volatile organic compounds: a case study for Jiangsu, China**

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## ABSTRACT

1  
2 Non-methane volatile organic compounds (NMVOC) are the key precursors of  
3 ozone (O<sub>3</sub>) and secondary organic aerosol (SOA) formation. Accurate estimation in  
4 their emissions plays a crucial role in air quality simulation and policy making. We  
5 developed a high-resolution anthropogenic NMVOCs emission inventory for Jiangsu  
6 in eastern China from 2005 to 2014, based on detailed information of individual local  
7 sources and the field measurements of source profiles of the chemical industry. A total  
8 of 56 NMVOCs samples were collected in 9 chemical plants, and then analyzed with  
9 a gas chromatography-mass spectrometry system (GC-MS). Source profiles of stack  
10 emissions from synthetic rubber, acetate fiber, polyether, vinyl acetate, and ethylene  
11 production, and those of fugitive emissions from ethylene, butanol and octanol,  
12 propylene epoxide, polyethylene and glycol production were obtained. Various  
13 manufacturing technologies and raw materials lead to discrepancies in source profiles  
14 between our domestic field tests and foreign results for synthetic rubber and ethylene  
15 production. The provincial NMVOC emissions were calculated to increase from 1774  
16 Gg in 2005 to 2507 Gg in 2014, and relatively large emission densities were found in  
17 cities along the Yangtze River with developed economy and industry. The estimates  
18 were larger than those from most other available inventories, due mainly to the  
19 complete inclusion of emission sources and to the elevated activity levels from  
20 plant-by-plant investigation in this work. Industrial processes and solvent use were the  
21 largest contributing sectors, and their emissions were estimated to increase  
22 respectively from 461 to 958 and from 38 to 966 Gg. Alkanes, aromatics and  
23 oxygenated VOCs (OVOCs) were the most important species, accounting for  
24 25.9%-29.9%, 20.8%-23.2% and 18.2%-21.0% to annual total emissions respectively.  
25 Quantified with a Monte-Carlo simulation, the uncertainties of annual NMVOCs  
26 emissions vary slightly from years, and the result for 2014 was -41%~+93%,  
27 expressed as 95% confidence intervals (CI). Reduced uncertainty was achieved  
28 compared to previous national and regional inventories, attributed partly to the  
29 detailed classification of emission sources and to the use of information at plant level

30 in this work. Discrepancies in emission estimation were explored for chemical and  
31 refinery sector with various data sources and methods. Compared with  
32 Multi-resolution Emission Inventory for China (MEIC), the spatial distribution of  
33 emissions in this work were more influenced by the locations of large point sources,  
34 and smaller emissions were found in urban region for developed cities in southern  
35 Jiangsu. Besides, discrepancies were found between this work and MEIC in the  
36 speciation of NMVOC emissions under the atmospheric chemistry mechanisms CB05  
37 and SAPRC99. The difference of species OLE1 resulted mainly from the updated  
38 source profile of building paint use, and the differences of other species from the  
39 varied sector contributions to emissions of the two inventories. CMAQ simulation  
40 was applied to evaluate the two inventories, and better performance (indicated by  
41 daily 1h-max O<sub>3</sub> concentrations in Nanjing city) was found for January, April and  
42 October 2012 when the provincial inventory was used.

43

## 44 **1 Introduction**

45 With strong OH and HO<sub>2</sub> radical chemistry reactions, non-methane volatile  
46 organic compounds (NMVOCs) are reported to play crucial roles in formation of  
47 secondary organic aerosols (SOA) and serious photochemical pollution in China,  
48 particularly in developed cities and regions. For example, Huang et al. (2014)  
49 revealed that the contribution of SOA from NMVOC conversion reached 44%-71% to  
50 ambient organic aerosols during a heavy haze period in winter, based on detailed  
51 chemistry composition and source analysis of airborne particles in four important  
52 cities (Beijing, Shanghai, Guangzhou and Xi'an) across the country. Due to intensive  
53 emissions of species with strong atmospheric oxidation capability (indicated as  
54 maximum incremental reactivity, MIR), ozone (O<sub>3</sub>) formation was recognized as  
55 VOC-limited in developed areas including Jing-Jin-Ji (JJJ), Yangtze River Delta  
56 (YRD) and Pearl River Delta (PRD) regions (Geng et al., 2008; Shao et al., 2009;  
57 Zhang et al., 2008; Xing et al., 2011).

58 Given the impacts of NMVOCs on air quality, increasing attentions have been  
59 paid to their sources and emission characteristics. Although natural sources dominate  
60 the emissions at global scale (Guenther et al., 1995; Muller, 1992; GEIA,  
61 [http://eccad.sedoo.fr/eccad\\_extract\\_interface](http://eccad.sedoo.fr/eccad_extract_interface)), the contribution from anthropogenic  
62 sources is elevated at smaller spatial scales, attributed to intensive human activities. In  
63 mainland China, emissions of natural and anthropogenic origin were estimated close  
64 to each other at 10-30 Tg, and anthropogenic emissions were dominated by solvent  
65 use and industrial processes (Tie et al., 2006; Klimont et al., 2002; Streets et al., 2003).  
66 Table S1 in the supplement briefly summarizes the estimations of China's national  
67 NMVOC emissions of anthropogenic origin from various studies. With different  
68 methods and data sources applied, NMVOC emissions in China were estimated to be  
69 doubled from 1990 to 2010, and the contributions of solvent use, non-combustion  
70 industrial processes and transportation were enhanced for recent years. Incorporating  
71 available information at national scale, Tsinghua University developed the  
72 Multi-resolution Emission Inventory for China (MEIC, <http://www.meicmodel.org/>)  
73 and calculated the national total emissions at 23.6 Tg for 2010. Among all the studies,  
74 largest estimations were made in Regional Emission inventory in Asia (REAS, Ohara  
75 et al., 2007; Kurokawa et al., 2013), reaching 28.0 Tg for 2008.

76 At local scale, emissions from anthropogenic sources could be much higher than  
77 natural sources. For example, the anthropogenic NMVOC emissions were estimated  
78 6-18 times those of natural origin in Beijing (Klinger et al., 2002; Wang et al., 2003;  
79 Klimont et al., 2002; Q. Zhang et al., 2009). With information on individual plants  
80 collected, emission inventories for regions with relatively heavy air pollution in China  
81 including JJJ, YRD and PRD have been developed, and differences in sector  
82 contribution were found. Solvent use and transportation were identified as the largest  
83 NMVOC sources in PRD (Zheng et al., 2009), while industrial processes were more  
84 important in YRD (Huang et al., 2011; Fu et al., 2013). Limitation existed in current  
85 regional inventories. First, the information of local sources was still lacking. Although  
86 combustion sources (e.g., power plants) were gradually included in the regional  
87 emission inventory as point sources, the sources that contribute more to NMVOCs

88 including refinery and chemical industry plants were less investigated at local scale,  
89 resulting possibly in big bias in emission estimation. Second, with varied data sources  
90 and methods, large discrepancies might exist between studies in the amount and  
91 spatial pattern of emissions. Such discrepancies were rarely analyzed, and the  
92 uncertainties in emission estimation at local scale were seldom quantified. In  
93 particular, the performances of chemistry transport modeling with various NMVOC  
94 inventories have not been sufficiently evaluated. Moreover, source profiles and  
95 speciation of NMVOC emissions need further improvement. Increasingly, domestic  
96 field measurements have been conducted on chemical profiles of NMVOCs for  
97 typical types of sources including solvent use (Yuan et al., 2010; Zheng et al., 2013),  
98 transportation (Tsai et al., 2012; Huang et al., 2015), residential stoves (Wang et al.,  
99 2009), and biomass burning (Kudo et al., 2014). The effects of those results on  
100 speciation of NMVOC emissions were not fully assessed, except for limited studies  
101 (Li et al., 2014). In addition, the measurements on given sectors such as chemical  
102 engineering are still lacking, and the data from foreign countries had to be used.

103 Under the heavy haze pollution in eastern China (Andersson et al., 2015; Sun et  
104 al., 2015; Wang et al., 2015), series of measures have been conducted particularly on  
105 power and industrial boilers to control the emissions of primary particles and the  
106 precursors of secondary particles such as SO<sub>2</sub> and NO<sub>x</sub> (Zhao et al., 2014). Along  
107 with gradually reduced ambient PM levels in YRD, O<sub>3</sub> pollution becomes a bigger  
108 concern for air quality improvement, motivating the better understanding and  
109 controlling of NMVOC emissions. In this work, we select Jiangsu, a typical province  
110 with intensive refinery and chemical industry in eastern China, to develop and  
111 evaluate the high-resolution emission inventory of anthropogenic NMVOCs. The  
112 geographic location and cities of the province are illustrated in Figure S1 in the  
113 supplement. Field measurements on chemical composition of NMVOC emissions  
114 were conducted to obtain the source profiles of typical chemical engineering  
115 processes. With detailed information of local emission sources collected and temporal  
116 changes tracked, the provincial emission inventory of NMVOC with chemistry  
117 profiles were developed for a ten-year period 2005-2014, and the uncertainties of the

118 emission estimation were quantified. Through a thorough comparison between results  
119 from varied methods and data sources, the discrepancies in emission estimation,  
120 source profiles, and spatial patterns were then evaluated. Finally, chemistry transport  
121 modeling was applied in southern Jiangsu to test the improvement of the provincial  
122 NMVOC inventory.

123

## 124 **2 Data and methods**

### 125 **2.1 Sampling and analysis of NMVOC species from chemical plants**

126 Chemical profiles of NMVOC emissions are still lacking for chemical industry  
127 and oil exploitation and refinery in China, due to a big variety of source categories.  
128 We select nine types of chemical engineering enterprises that are intensively  
129 distributed in Jiangsu to measure the chemistry composition of NMVOC emissions, as  
130 summarized Table S2 in the supplement. To our knowledge, no domestic  
131 measurement on NMVOC speciation has been conducted for those sources yet, and  
132 current work was expected to supplement the domestic source profiles for chemical  
133 and refinery industry. Based on the on-site investigation of main emission processes,  
134 the locations for stack and/or fugitive emission sampling were determined for each  
135 source type (see Table S2 for the details). Note the sampling cannot be conducted for  
136 all the processes in an enterprise due mainly to the limitation of pipeline layout. The  
137 SUMMA canister produced by University of California, Irvine was employed to  
138 collect the air sample. The canister was made out of stainless steel as its inner wall,  
139 and the volume and vacuum pressure range of the canister was 2 L and  $1.1 \times 10^{-4}$  kPa,  
140 respectively. For stack emission measurement, as shown in Figure S2a in the  
141 supplement, the canister was connected with a stainless steel probe, and a filter pipe  
142 filled with glass wool and anhydrous sodium sulfate was applied to remove the  
143 particles and water vapor in the air sample. Under flow control, the sampling time was  
144 roughly 10 minutes until the pressure in the canister reached ambient. For fugitive  
145 emission measurement, the canister was placed 50 meters downwind of the

146 production device, and the sampling time was roughly 8 minutes. Repeated sampling  
147 was conducted for each process to eliminate the bias and a total of 56 samples were  
148 obtained, as shown in Table S2.

149 NMVOC samples were analyzed by one Gas Chromatograph Mass Spectrometer  
150 (GC-MS) system (GC6890/MS5973i, Agilent Technologies, USA), as illustrated in  
151 Figure S2b in the supplement. Firstly, the sample was pumped into a cryogenic  
152 pre-concentrator with a three-stage trapping system. In the first stage, the VOC  
153 species were adsorbed on glass beads at  $-150^{\circ}\text{C}$  and desorbed at  $10^{\circ}\text{C}$ . Vapour water  
154 was converted to solid water and was thus separated from sample. In the second stage,  
155 the sample was trapped on Tenax at  $-30^{\circ}\text{C}$  and desorbed at  $180^{\circ}\text{C}$ , and the target  
156 species were separated from  $\text{CO}_2$  and other compositions in the air. In the third stage,  
157 the sample was focused on a transfer line at  $-160^{\circ}\text{C}$  and heated rapidly to  $70^{\circ}\text{C}$ , and  
158 the species were concentrated. The concentrated VOC was then injected into the gas  
159 chromatograph. The GC oven temperature was initially programmed at  $-50^{\circ}\text{C}$ , and  
160 then increased to  $180^{\circ}\text{C}$  at  $4^{\circ}\text{C}/\text{min}$  and to  $220^{\circ}\text{C}$  at  $15^{\circ}\text{C}/\text{min}$ , holding 3 minutes. The  
161 VOC compounds were separated on a DB-5MS capillary column ( $60\text{ m}\times 0.25\text{ mm}\times 1.0$   
162  $\mu\text{m}$ ) and quantified using a quadrupole mass spectrometer. The mass spectrometer was  
163 operated in SCAN mode and scanned 20-42 amu and 35-270 amu before and after 8.5  
164 minutes, respectively. The ionization method was electron impacting, and the source  
165 temperature was  $230^{\circ}\text{C}$ . The PAMS (Photochemical Assessment Monitoring System)  
166 standard mixture (Spectra Gases Inc., USA) and TO-15 standard mixture (Spectra  
167 Gases Inc., USA) were used to confirm the retention times of different compounds,  
168 and to identify them based on retention time and mass spectrum. The target species  
169 were quantified by using multipoint internal calibration method. To establish  
170 calibration curves, a certified gas mixture containing all the target compounds was  
171 dynamically diluted with pure nitrogen to five concentration gradients using mass  
172 flow controllers. Bromochloromethane, 1,2-difluorobenzene, and chlorobenzene-d5  
173 were chosen as internal standards in samples. The method detection limit was 0.5 ppb  
174 for all species.

175

## 176 2.2 Development of provincial emission inventory

177 Different from national and regional inventories that were based mainly on  
178 energy and economic statistics, detailed information with inter-annual changes for  
179 emission sources in Jiangsu was collected and tracked at plant level from the  
180 multiple-year official Environmental Statistics (the databases of emission sources  
181 compiled by local environmental protection agency), Pollution Source Census (PSC,  
182 internal data of emission sources collected by local environmental protection agency),  
183 and on-site surveys on large emitters conducted by local environmental protection  
184 bureaus. The information included geographic location, types and amounts of raw  
185 materials, types and amounts of products, fuel quality and consumption, and  
186 combustion/manufacturing technology. For 2014, as an example, detailed information  
187 of 6023 plants was obtained, and the locations of those point sources were illustrated  
188 in Figure S1. Incorporating the plant-by-plant information and the energy and  
189 industrial statistics at provincial level, a four-level framework was established,  
190 covering all the anthropogenic NMVOC sources for Jiangsu, as summarized in Table  
191 1. The framework included seven main categories: stationary fossil fuel combustion,  
192 industrial process, solvent use, transportation, oil distribution, biomass burning, and  
193 others. Each main category was further subdivided into subcategories according to  
194 discrepancies in fuel consumption, product manufacturing, or technology application.  
195 The emissions of the province for 2005-2014 were estimated with a bottom-up  
196 method using the following equation:

$$197 \quad E(n) = \sum_i AL(i, n) \times EF(i, n) \quad (1)$$

198 where  $i$  and  $n$  represent the source type and year, respectively;  $E$  is the annual  
199 emissions;  $AL$  is the activity level data; and  $EF$  is the emission factor (i.e., emissions  
200 per unit of  $AL$ ). As summarized in Table 2, emission factors were collected from  
201 extensive literatures and determined as follows with descending priorities: (1) the  
202 results from domestic measurements; (2) the emission limits of local laws and  
203 regulations; (3) the values from expert judgment specific for China; and (4) emission  
204 factors from AP-42 database (USEPA, 2002) and the EMEP/EEA guidebook (EEA,



205 2013) when domestic information was lacking. Details will be discussed by sector in  
206 Section 2.3.

207 The total NMVOC emissions for given source type were then broken down into  
208 individual species using Eq. (2):

$$209 \quad E(i, k) = E(i) \times X(i, k) \quad (2)$$

210 where  $E$  is the emissions;  $i$  and  $k$  represent the source type and individual NMVOCs  
211 species, respectively;  $X$  is chemical profile of NMVOCs (%). To reduce the  
212 uncertainty of source profile from individual measurement, Li et al. (2014) developed  
213 the “composite profiles” for sources where multiple candidate profiles were available,  
214 by revising the OVOCs fraction and averaging the fractions in different profiles for  
215 each species. As a more detailed source classification was applied in this work, some  
216 sources (e.g., biomass open burning, automobile production & repairing, and wood  
217 decoration & wooden furniture paint) were not covered by Li et al. (2014) and thus  
218 the results from SPECIATE were applied instead. We named the combination of Li et  
219 al. (2014) and SPECIATE as a source profile “before updating” hereinafter. While Li  
220 et al. (2014) included the source profiles published before 2011, a series of local  
221 measurements were conducted after then. In this work, therefore, “composite profiles”  
222 were updated following the method by Li et al. (2014). The most recent domestic  
223 results and the measurements conducted in this work were incorporated as summarized  
224 by source type in Section 2.3.

225 To evaluate the atmospheric oxidation capability from NMVOC emissions,  
226 ozone formation potentials (OFPs) were calculated by multiplying the speciated  
227 NMVOC emissions and corresponding MIR values (Carter, 1994). To meet the  
228 requirement of atmospheric modeling, NMVOC emissions were assigned to chemical  
229 mechanism (CB05 and SAPRC99) species by multiplying the emissions of individual  
230 species and mechanism-specific conversion factors using the following equation:

$$231 \quad E(i, m) = \frac{E(i, k)}{M(k)} \times C(k, m) \quad (3)$$

232 where  $E$  is the emissions,  $M$  is the mole weight,  $C$  is the conversion factor, and  $i, m$ ,

233 and  $k$  represent the source type, individual species, and the chemical mechanism  
234 species.

235 The uncertainty of estimated provincial emissions was quantified using a  
236 Monte-Carlo framework (Zhao et al., 2011) for each year. Probability distribution  
237 functions (PDF) were determined for all the parameters involved in the emission  
238 calculation, and 10 000 simulations were performed to estimate the uncertainties of  
239 emissions. The parameters that were most significant in determination of the  
240 uncertainties were identified by source type according to the rank of their  
241 contributions to variance. The detailed information of PDF assumption will be  
242 provided by sector in Section 2.3.

### 243 **2.3 Data sources of emission inventory development by category**

#### 244 **Combustion sources (fossil fuel combustion and biomass burning)**

245 For power sector and heating/industrial boilers, activity data were compiled at  
246 plant level based on the information obtained from Environmental Statistics, PSC and  
247 on-site survey. The annual amount of residential fossil fuel combustion for 2005-2014  
248 and that of biofuel use for residential stoves until 2008 were directly taken from  
249 Chinese official energy statistics (NBS, 2015a). As the data were unavailable for  
250 subsequent years for biofuel, the activity level was calculated as a product of grain  
251 production, waste-to-grain ratio, and the ratio of residual material burned in stoves  
252 according to the government plan of biomass utilization (PGJP, 2009). The biomass  
253 combusted in open fields was calculated as a product of grain production,  
254 waste-to-grain ratio, and the percentage of residual material burned in the field (Wang  
255 and Zhang, 2008; PGJP, 2009), as described in Zhao et al. (2011; 2013). Following  
256 the rules of IPCC (2006), the uncertainties of activity levels were determined  
257 according to the reliability of energy and economy statistics. As shown in Table S3 in  
258 the supplement, normal distributions were assumed with the coefficients of variation  
259 (CV, the standard deviation divided by the mean) determined at 5%, 10%, 20% and  
260 30% for power, industrial, residential fossil fuel and biomass combustion sources,  
261 respectively.

262 As summarized in Table 2, emission factors for power plants and industrial  
263 boilers were taken mainly from Bo et al. (2008). Given the similar designs of boilers  
264 between China and developed countries (Wei et al., 2008), AP-42 database were also  
265 applied when domestic results were lacking. For residential combustion of fossil fuel,  
266 emission factors from domestic tests (Zhang et al., 2000; Wang et al., 2009) were used.  
267 The emission factors for biofuel burning in stoves were from the field test results by  
268 Wang et al. (2009), who measured the NMVOC emission characteristics of multiple  
269 stove-fuel combinations in China and provided the emission factors by province. For  
270 biomass open burning, the local test results by Li et al. (2007) were applied. Chemical  
271 profiles were updated by incorporating domestic measurements for residential fossil  
272 fuel and biofuel burning (Tsai et al., 2003; Liu et al., 2008; Wang et al., 2009; Wang et  
273 al., 2014a), and biomass open burning (Zhang et al., 2013; Kudo et al., 2014). As  
274 measurement results were insufficient for data fitting, uncertainty of emission factor  
275 was evaluated depending on expert judgment for combustion sources (Streets et al.,  
276 2003; Wei et al., 2011). PDF of emission factor was given according to reliability of  
277 data sources and/or the robustness of calculation methods (Wei et al., 2011; also the  
278 case for other sources as indicated below). As summarized in Table S4 in the  
279 supplement, lognormal distributions were assumed with CVs set at 150% and 200%  
280 for fossil fuel and biomass burning, respectively.

## 281 **Industrial processes**

282 Similar with power and industrial combustion, the activity levels of industrial  
283 processes were mainly collected at plant level by source category. However, small  
284 discrepancies existed between the compiled data at plant level and the officially  
285 reported production from economic statistics at provincial level (NBS, 2015b). In  
286 2012, for example, the steel production aggregated from individual plants accounted  
287 for 98% of the provincial total production (Zhou et al., 2017). In this work, therefore,  
288 we treated the individual plants as point sources, and the fraction that was not covered  
289 in plant-by-plant databases as area sources. We assumed the PDFs of industrial  
290 production as normal distribution with CVs at 10% and 20% for point and area

291 sources, respectively.

292       Attributed to a wide variety of manufacturing procedures and complicated VOC  
293 exhaust processes, there were few local tests available on emission factors before,  
294 thus the values from expert judgment (i.e., data from routine investigations reported  
295 by the factory officials to local environmental protection bureaus) and data from  
296 AP-42 and EMEP/EEA had to be applied, as summarized in Table 2. Source profiles  
297 of chemical production including rubber, polyether, and polyethylene were obtained  
298 from our measurements described in Section 2.1. Chemical profiles from most recent  
299 domestic measurements were taken for other sources including iron & steel (Shi et al.,  
300 2015; He et al., 2005; Jia et al., 2009; Tsai et al., 2008; Li et al., 2014), paint and  
301 printing ink production (Zheng et al., 2013), and refineries (Liu et al., 2008; Wei et al.,  
302 2014). For sources without sufficient local tests (e.g., food and wine production),  
303 results from foreign studies were applied including the SPECIATE database by  
304 USEPA (2014), and Theloke and Friedirch (2007). Given the potential large  
305 uncertainties, lognormal distribution with CV set at 500% (i.e., a long-tailed PDF)  
306 was assumed for emission factors for most industrial processes.

### 307 **Solvent use**

308       Although solvent-use enterprises were contained in the plant-by-plant surveys,  
309 many of them failed to report the actual solvent usage. Underestimation in activity  
310 levels and thereby emissions could be expected if the information at plant level was  
311 relied on. As the solvent usage was not directly reported at city level, we followed  
312 Wei (2009) to estimate the activity levels for the sector. The total solvent consumption  
313 at national level was first obtained according to the solvent production and imports &  
314 exports statistics (CNLIC, 2015; GAC, 2015). The provincial level was then  
315 calculated based on the intensities of activities that consume solvent (e.g., building  
316 construction and vehicle production). Finally, the provincial data were allocated to  
317 point sources according to production of individual plants and area sources according  
318 to distribution of industrial GDP. Normal and lognormal distributions with CVs set at  
319 20% and 80% were applied for the activity levels of point and area sources of solvent

320 use, respectively, indicating much larger uncertainty for the latter (Wei, 2009).

321 The VOC contents of solvent were limited by national laws and regulations, as  
322 summarized by Wei et al. (2008). The updating of regulations and their impacts on  
323 inter-annual changes in NMVOC emission factors were considered from 2005 to 2014,  
324 such as GB18583-2003 and GB18583-2008 for indoor painting. Bias needs to be  
325 admitted here and possible underestimation in emissions would be expected for the  
326 sector, as the regulations were not strictly enforced particularly for small solvent use  
327 enterprises and construction sites (area sources). Data from AP-42, EMEP/EEA and  
328 other literatures (Bo et al., 2008; Fu et al., 2013) were applied when local information  
329 was missing, as provided in Table 2. The domestic tests on chemical species (Yuan et  
330 al., 2010; Zheng et al., 2013; Tang et al., 2014; Wang et al., 2014b) were included to  
331 update the source profiles of the sector. For uncertainty of the emission factors,  
332 uniform distribution was tentatively applied, assuming the same probability in a wide  
333 range (Table S4).

### 334 **Transportation**

335 The activity data of off-road transportation for 2005-2010 was taken from Zhao  
336 et al. (2013), and the data for other years were scaled according to the changes in  
337 passenger and freight traffic by rail and shipping, and those in numbers of agricultural  
338 and construction equipments (NBS, 2015c). For on-road transportation, activity data  
339 (total kilometers traveled) by vehicle type and control stage were calculated as the  
340 product of vehicle population and annual average kilometers traveled (VKT). The  
341 fleet composition by control stage was obtained from the survey by local government  
342 (internal data, Zhao et al., 2015), and VKT by vehicle type were determined according  
343 to previous studies (Cai and Xie, 2007; Wang et al., 2008) and the guidebook of  
344 emission inventory development for Chinese cities (He, 2015). While the CV of  
345 transportation activities at national level was estimated at 16% (Zhao et al., 2011),  
346 larger uncertainties were assumed at provincial level (Wei, 2009). We followed Wei  
347 (2009) and assumed normal and lognormal distribution with CVs at 30% and 50% for  
348 on-road and non-road transportation, respectively.

349 For emission factors of off-road transportation, the data from EMEP/EEA and  
350 expert judgment (Wei et al., 2008; Q. Zhang et al., 2009) were adopted, attributed to  
351 lack of domestic tests or relevant standards/regulations. Following He (2015), the  
352 emission factors for on-road vehicles were calculated and adjusted according to local  
353 conditions using Eq. (4):

$$354 \quad EF = BEF \times \varphi \times \gamma \times \lambda \times \theta \quad (4)$$

355 where *BEF* is the base emission factor,  $\varphi$  is the environmental correction factor,  $\gamma$  is  
356 the average traveling speed correction factor,  $\lambda$  is the vehicle deterioration correction  
357 factor, and  $\theta$  is correction factor for other conditions (e.g., vehicle loading and fuel  
358 quality). Domestic tests on chemical compositions (Liu et al., 2008; Tsai et al., 2012;  
359 Huang et al., 2015; Wang et al., 2013; Ou et al., 2014; Gao et al., 2012) were  
360 incorporated to update the source profile of on-road vehicles. Lognormal distributions  
361 with CVs at 150% and 300% were respectively assumed for emission factors of  
362 on-road and off-road transportation.

### 363 **Oil distribution and other sources**

364 For oil distribution, Wei (2009) developed a simplified model to simulate the oil  
365 storage, transport and sale activities based on the provincial energy balance statistics,  
366 and the model was applied in this work to calculate the activity levels for the sector.  
367 Activity data from other sources were directly taken from official provincial statistics  
368 (NBS, 2015b; c). Emission factors of oil distribution and other sources were obtained  
369 from Wei et al. (2008), Shen (2006) and Xia et al. (2014), and lognormal distributions  
370 were assumed for them as summarized in Table S4. The source profiles were obtained  
371 from Liu et al. (2008) and SPECIATE.

## 372 **3 Results**

### 373 **3.1 Source profiles of chemical industry from measurement**

374 NMVOC source profiles of 14 processes (9 for stack emissions and 5 for fugitive  
375 emissions) were obtained from field measurements. With totally 61 species detected

376 by GC-MS systems, the chemistry components were grouped into 6 types (alkanes,  
377 alkenes, halohydrocarbon, aromatics, OVOCs, and others), as illustrated in Figure 1a.  
378 Alkanes were the main species of synthetic rubber industry (SBR, SIS rubber and  
379 SEBS rubber) and the mass fractions were measured over 70%. For production of  
380 cellulose acetate fiber, alkanes, aromatics, and OVOCs were the main species in  
381 process of acetate flake production, while OVOCs dominated in the spinning process.  
382 Resulting from various raw materials applied, the source profiles of the two types of  
383 polyether production differed a lot: the mass fraction of OVOCs was close to 80% for  
384 PPG, while the main species for POP were others, aromatics and halohydrocarbon.  
385 For ethylene production, aromatics were the main composition in the stack gas of  
386 cracking furnace, while alkanes and alkenes dominated the fugitive emissions. For  
387 other types of fugitive emissions, big differences existed in the source profiles  
388 attributed mainly to the various raw materials and chemical reactions.

389 The detailed chemistry compositions for stack and fugitive emissions were  
390 summarized in Tables S5 and S6, respectively. For stack emissions of synthetic rubber  
391 production, cyclohexane was the dominating species, with the mass fractions close to  
392 or above 70% for all types of products. Besides, styrene and acetone were also  
393 important species for SBR and SIS/SEBS rubber, respectively. Used as the solvent in  
394 the chemical reactions, acetone and cyclohexane were considerably emitted during  
395 acetate flake production process, and the mass fraction of acetone reached 70% in the  
396 spinning process. As the raw materials for polyether production, acrylonitrile and  
397 ethylene oxide were the main species emitted from POP and PPG production, with the  
398 mass fractions measured at 43% and 62%, respectively. As the main product,  
399 vinyl acetate was unsurprisingly measured to take 80% of NMVOC emissions from  
400 its production process. For fugitive emissions from ethylene production, the mass  
401 fractions of ethylene, propylene, and n-hexane reached 33%, 26% and 20%,  
402 respectively. 1,2-dichloropropane dominated the emissions from propylene oxide  
403 production, with the mass fraction measured at 65%. For polyethylene and glycol  
404 production, ethylene and xylene were identified as the largest species, with the mass  
405 fractions measured at 42% and 35%, respectively.

406 Since there were very few domestic tests on source profiles of chemical industry,  
407 the results obtained in this work were compared with those available in SPECIATE  
408 for synthetic rubber, ethylene and polyethylene production, as illustrated in Figure  
409 1b-d, respectively. As can be seen in Figure 1b, large discrepancy was found for  
410 source files of SBR between this work and SPECIATE: while cyclohexane was  
411 identified as the main species in this work, SPECIATE included only styrene and 1,3-  
412 butadiene (the raw materials in SBR production). Variation in manufacturing  
413 technologies was the main source of the discrepancy. Emulsion polymerization  
414 technology was considered in SPECIATE, in which a solvent was not used and thus  
415 NMOVC emissions consisted mainly of the volatile raw materials. This work,  
416 however, measured the plants with solution polymerization technology, in which  
417 significant organic solvents would be released during the drying process. As shown in  
418 Figure 1c, both SPECIATE and our measurements on fugitive emissions indicated that  
419 ethylene and isobutene were the important species for ethylene production. In addition,  
420 much larger fractions of propylene and n-hexane were found in this work. Clear  
421 different composition was found for flue gas of cracking furnace, with abundant  
422 species from incomplete combustion. Similar source profiles were found between this  
423 work and SPECIATE for polyethylene production, dominated by ethylene (Figure 1d).

### 424 **3.2 Inter-annual trends and sectoral contribution of NMVOC emissions**

425 As shown in Table 3, the annual emissions of anthropogenic NMVOCs for Jiangsu  
426 were estimated to increase from 1774 to 2507 Gg during 2005-2014, with an average  
427 annual growth rate at 3.9%. Industrial processes and solvent use were identified as the  
428 largest two sectors contributing to the emissions. The emission fractions of the two  
429 types of sources to total anthropogenic emissions were estimated to increase from  
430 26% in 2005 to 38% in 2014, and from 21% to 39%, respectively. In contrast, the  
431 emission contributions from transportation and biomass burning were declining from  
432 18% to 11%, and from 26% to 4%, respectively, attributed mainly to the controlled  
433 motorcycle emissions, replacement of residential biofuel stoves with natural  
434 gas/electricity ones, and the gradual implementation of straw burning prohibition.



435 Relatively small contributions were found for stationary fuel combustion plants, oil  
436 distribution, and other sources, and their collective fractions to total emissions ranged  
437 7-9% during the study period.

438 Figure S3a-c in the supplement provided the inter-annual trends in emissions of  
439 subcategories for industrial processes, solvent use and transportation. The emissions  
440 from industrial processes were estimated to be doubled from 2005 to 2014, and the  
441 inter-annual trend in emissions was well correlated with that in industry GDP (Figure  
442 S3a). The comprehensive investigations on point sources indicated that few measures  
443 were implemented to control NMVOC till 2014, and the increased emissions were  
444 thus mainly driven by the growth of industry activities. Largest growth was found for  
445 synthetic and fine chemical industry, with the emissions elevated from 130 in 2005 to  
446 361 Gg in 2014. Due to enhanced coking industry, the emissions of iron & steel  
447 production were estimated to increase 254% from 27 to 96 Gg.

448 The emissions of solvent use in Jiangsu were calculated to increase 153% from  
449 380 in 2005 to 963 Gg in 2014, and the growth was highly consistent with that of  
450 industry plus construction GDP (Figure S3b). Despite of increased use of water  
451 soluble paint and implementation of emission standards for given processes (e.g., the  
452 VOC content of interior wall paint has been reduced from 250 to 120 g/kg since 2008),  
453 it was still difficult to restrain the emissions under the fast growth of solvent use, as  
454 relevant polices were not widely conducted across the sector. Paint use was the largest  
455 contributor, and its emissions were calculated to increase from 225 in 2005 to 652 Gg  
456 in 2014. The emissions from printing ink increased 355% from 25 to 115 Gg.

457 Although Jiangsu's total vehicle population increased 76% from 2005 to 2014,  
458 the NMVOC emissions of on-road transportation were estimated to decline 31% from  
459 297 to 204 Gg, with the peak emissions at 302 for 2007 (Figure S3c). The  
460 implementation of staged emission standards (State I-V, equal to Euro I-V) on new  
461 vehicles, and reduced motorcycle population were the main reasons for the declining  
462 emissions. For example, emissions of motorcycles decreased 66% from 185 to 64 Gg,  
463 and its contribution to on-road vehicle emissions declined as well from 62% and 31%.  
464 Unsurprisingly, gasoline vehicles dominated the emissions of on-road transportation,

465 with the fraction ranged 81%-87% during the study period.

466 Illustrated in Figure 2 are the spatial distributions of Jiangsu's NMVOC  
467 emissions for various years within a 3×3 km grid system. The emissions of point  
468 sources were directly allocated according to their geographic locations. For other  
469 sources, certain proxies were applied to allocate emissions, including GDP for  
470 industrial area sources and oil distribution, population for solve use area sources, road  
471 net and traffic flow for on-road vehicles, railway and canal net for off-road  
472 transportation, and rural population for biomass burning. High emission intensities  
473 were mainly found in relatively developed cities along Yangtze River in southern  
474 Jiangsu including Nanjing, Suzhou, Wuxi and Yangzhou (see Table S7 for the  
475 emissions by city). In central and northern Jiangsu, large emissions existed in areas  
476 with clustered industrial parks, reflecting the impacts of big plants on spatial pattern  
477 of NMVOC emissions. Comparing the emissions for 2005 and 2014, increased  
478 emissions were commonly found in southern Jiangsu indicating the faster growth of  
479 industry in developed cities (Figure 2d). Moreover, reduced emissions were coincided  
480 with road net distribution, implying the benefits of emission controls on vehicles.

### 481 **3.3 Speciation and OFPs of NMVOC emissions**

482 Table 4 compares the source profiles of this work with those of Li et al. (2014) or  
483 SPECIATE for typical source categories, grouped as alkanes, alkenes, alkynes,  
484 aromatics, OVOCs, and others. Elevated fractions of alkanes are found in this work  
485 for almost all the sources, while the comparisons of other species are less conclusive  
486 between sources. Based on the source profiles, emissions of more than 500 NMVOC  
487 species were calculated and grouped into 12 categories (alkanes, alkenes, alkynes,  
488 aromatics, alcohols, aldehydes, ketones, ethers, acids, esters, halohydrocarbons, and  
489 others). From 2005 to 2014, the mass fractions of alkanes, unsaturated hydrocarbon  
490 (alkenes and alkynes), aromatics, OVOCs (alcohols, aldehydes, ketones, ethers, acids,  
491 and esters), halohydrocarbons, and others were between 26-30%, 13-19%, 21-23%,  
492 18-21%, 3-4%, and 11-12%, respectively.

493 Shown in Figure 3 are the mass fractions of species by source for 2014. Due to

494 varied fuel qualities and combustion conditions, large differences in the speciation of  
495 emissions were found for fossil fuel combustion, transportation and biomass burning.  
496 Dominated by coal combustion, the profile of fossil fuel stationary sources was  
497 relatively simple with little OVOCs and halohydrocarbons, and aromatics were the  
498 largest fraction (45%), followed by alkanes (29%). Alkanes, aromatics and alkenes  
499 were the main species from transportation, with the fractions estimated at 30%, 24%,  
500 and 23%, respectively. For biomass burning, elevated alkenes and less alkanes were  
501 found attributed to the highly incomplete combustion. The mass fractions of alkanes,  
502 aromatics and alkenes from industrial processes were estimated 32%, 16% and 12%.  
503 In particular, relatively close emission fractions were found between species for  
504 chemical industry, the largest emission source of industrial processes: 19%, 15%, 11%,  
505 10% and 10% of aromatics, ketones, alkenes, alkanes, and halohydrocarbons,  
506 respectively. For solvent use, aromatics and alkanes were the most important species  
507 with the fractions estimated at 32% and 22%, respectively, and the collective fraction  
508 of OVOC species reached 27%. Alkanes and aldehydes dominated the emissions of  
509 oil distribution and other sources, which came mainly from the oil evaporation and  
510 residential cooking, respectively.

511 The OFPs from NMVOC emissions in Jiangsu were calculated to increase from  
512 3880 in 2005 to 5200 Gg in 2014, and the ratio of annual OFPs to emissions varied  
513 slightly around 2.1 for the decade. As the chemical profiles of emitted NMVOC vary  
514 between source categories, the OFP to emission ratio for a given source category  
515 could indicate the potential contribution to ozone formation of the category, as a  
516 combined effect of multiple species emitted. The priorities of emission control for  
517 ozone abatement could thus be suggested by the ratio. The ratios for 2014 were  
518 provided by source in Figure 3. With abundant aromatics and alkenes emissions that  
519 were highly active in chemistry, the largest ratio (3.68) was found for fossil fuel  
520 stationary combustion. The ratios of biomass burning and other sources reached 3.0,  
521 attributed to active aromatics and aldehydes emissions, respectively. The lowest ratio  
522 (1.58) was found for oil distribution, as its emissions were dominated by alkanes with  
523 low reactivity. Figure S4 in the supplement provided 25 species with the biggest

524 contributions to OFPs and their emission sources for 2005 and 2014. In 2005, the 25  
525 species were estimated to account for 44% of total NMVOC emissions and 83% of  
526 OFPs (Figure S4a). Xylene, ethylene, and propylene were identified as the most three  
527 important species in terms of OFP. The aromatics species with (e.g., xylene and  
528 toluene) came largely from solvent use and industrial processes, while alkenes species  
529 (e.g., ethylene and propylene) were mainly from industrial processes, biomass burning,  
530 and transportation. Besides, biomass burning was the dominating sources of methyl  
531 glyoxal, methyl alcohol and glyoxal. For 2014, the 25 species were estimated to  
532 account for 38% of total NMVOC emissions and 81% of OFPs, and the contributions  
533 of solvent use and industrial processes to OFPs were clearly elevated (Figure S4b).  
534 The orders of isopropanol and n-butanol that were mostly from solvent use, for  
535 example, were moved forward. Moreover, the contribution of biomass burning  
536 significantly declined, and the glyoxal was not identified as the one of the 25 most  
537 important species to OFPs any more.

### 538 **3.4 Uncertainties of provincial NMVOC emission inventory**

539 The uncertainties of estimated annual NMVOC emissions for Jiangsu 2005-2014  
540 are illustrated in Figure 4, expressed as the 95% confidence intervals (CIs) around the  
541 central estimates. As inter-annual changes were hardly assumed in determination of  
542 probability distributions for parameters, similar uncertainty ranges were thus  
543 calculated for emissions of various years. As shown in Table 5, the uncertainty of  
544 emissions 2014 was quantified at -41%~+93% (95% CI), and biomass burning and  
545 other sources were estimated as the sources with largest uncertainties, followed by  
546 stationary fossil fuel combustion and oil distribution. For most emission  
547 sectors/categories, emission factors were identified as the largest sources of emission  
548 uncertainty, with an exception of solvent use. Resulting from complicated sources of  
549 stack and fugitive emissions, it is generally difficult to conduct comprehensive field  
550 tests on emission factors for industrial and residential sources. As described in Section  
551 2.3, large uncertainties had to be conservatively assumed for them due to limited  
552 domestic samples and to heavy dependence on foreign databases. More measurements

553 on actual emission characteristics are thus recommended to expand data samples for  
554 better evaluating the PDFs and effectively reducing emission uncertainty. Regarding  
555 solvent use for which provincial and city statistics were lacking, the activity data had  
556 to be downscaled from national level leading to possibly big bias in emission  
557 estimation.

558        Provided in Table 5 as well are the uncertainties of national and YRD emissions  
559 from other studies. Note all the studies included for comparison applied Monte-Carlo  
560 simulation except Huang et al. (2011), which calculated the uncertainty of emissions  
561 based on the predetermined CVs of emission factors and activity levels at sector level.  
562 Compared with those results, the emission uncertainties were reduced in this work for  
563 industrial processes, solvent use and transportation. Besides the varied methods and  
564 assumptions of PDFs for relevant parameters, the more detailed classification of  
565 emission sources and adoption of independent emission factors for those sources  
566 should be an important reason. For example, totally 34 vehicle type-control  
567 combinations were taken into account for calculating the on-road vehicle emissions,  
568 and emission factor for each type of combination was assumed independent from  
569 other. In addition, the errors of activity levels for big point sources were significantly  
570 reduced from the detailed investigation and on-site survey at plant level, leading to  
571 smaller uncertainty in emission estimation for industrial and solvent use sources.

## 572 **4 Evaluation of provincial emission inventory**

### 573 **4.1 Chemical and refinery industry emissions from varied data sources and** 574 **methods**

575        As a part of industrial process, chemical and refinery industry was one of the  
576 biggest contributors to anthropogenic NMVOC emissions. We select Nanjing, the  
577 capital city of Jiangsu province, to evaluate the impacts of data sources and methods  
578 on emissions of this category. As described in Sections 2.2 and 2.3, the method used  
579 in this work for provincial inventory improvement incorporated the most available  
580 information from Environmental Statistics, PSC, and on-site surveys (named Method

581 1 here). Besides, two other methods based respectively on data from Environmental  
582 Statistics (Method 2) and economic statistics without any information on individual  
583 plants (Method 3, which was commonly applied in national and regional inventories)  
584 were also applied to calculate the emissions in the city for 2011, and the results with  
585 different methods were compared against each other. Note the emissions of area  
586 sources (i.e., not included in plant-by-plant investigations) in Methods 1 and 2 were  
587 estimated using the same data source as Method 3. Table S8 in the supplement  
588 provides the emissions calculated based on the three data sources by subcategory of  
589 chemical and refinery industry. The emissions estimated using Method 1 were clearly  
590 larger than those using Method 2 or 3, resulting mainly from the incomplete records  
591 of chemical products by environmental or economic statistics. For example, some  
592 basic chemistry products (e.g., ethylene oxide and ethylene glycol) and synthetic  
593 chemical products (e.g., polyether and polyethylene) were not included in current  
594 economic statistics. In addition, although most chemical and refinery plants were  
595 investigated in the Environmental Statistics, only three types of chemical products  
596 were recorded for each plant, much less than the actual (more than one hundred types  
597 for some plants). The omission of chemical product types thus led to underestimations  
598 in NMVOC emissions. With the product types fully covered, Method 1 could even  
599 underestimate the emissions, as the emission factors could not be measured or  
600 determined for all products due to the completed pipe layout or fugitive release.

601 Spatial distributions of the emissions estimated using the three data sources for  
602 Nanjing were illustrated in Figure 5. Similar patterns were found for Method 1 and 2  
603 (Figure 5a and 5b), as the emissions were dominated by the big chemical and refinery  
604 plants. As labeled in Figure 5a, the largest ten plants were estimated to account for  
605 80% of NMVOC emissions of the sector for the whole city. Without detailed  
606 information of individual plants, Method 3 had to apply the proxies (e.g., population  
607 density) to allocate the emissions, and would overestimate the fraction of emissions in  
608 urban downtown (Figure 5c). It could thus be inferred that big discrepancies in spatial  
609 distribution of emissions at small scale might be caused when downscaled from larger  
610 scale without sufficient investigation on local sources, particularly for regions where

611 emissions were dominated by large plants that were gradually moved out of urban  
612 areas.

613 To further examine the emission estimation on individual plants, an alternative  
614 method was applied to calculate the emissions of all manufacturing processes  
615 separately including leaks of hydrocarbon vapors from process equipment (valves,  
616 flanges, seals, etc.), storage of organic liquid, loading and unloading of organic liquid,  
617 and waste water treatment (mentioned as device operation based method herein). As  
618 indicated in Table S9 in the supplement, detailed information of 15 key chemical and  
619 refinery enterprises in Nanjing (i.e., enterprises directly under the control of city  
620 government) were collected and the emissions of those plants were calculated and  
621 compared with the results using the method as described in Sections 2.2 and 2.3  
622 (mentioned as emission factor based method herein). Although the total emissions of  
623 the 15 plants were very close between the two methods, significant discrepancies  
624 existed for individual plants. For example, much larger NMVOC emissions were  
625 calculated for plants 3 and 6 with device operation based method, as the emission  
626 factors for production of chlorobenzene alkylbenzene, and cyclohexanone were  
627 lacking, leading to underestimation by the emission factor based method. As shown in  
628 Figure S6 in the supplement, the differences in emissions from varied calculating  
629 methods for the 15 plants led to moderate changes in spatial distributions of the  
630 chemical and refinery emissions for the city. In general, the device operation based  
631 method could better capture the activities of specific plant and the actual emission  
632 characteristics; however, the method could hardly be applied in a broader scale, as it  
633 depends strongly on the completeness and quality of data collection.

#### 634 **4.2 Changes in speciation of NMVOC emissions**

635 As indicated in Section 2.2, the speciation of NMVOC emissions in Jiangsu were  
636 updated by including our measurements and other most recent domestic tests after  
637 2010, based on the combination profile of Li et al. (2014) and SPECIATE (i.e., the  
638 source profile before updating). The source types we measured accounted for 9-11%  
639 of annual NMOVC emissions from chemical industry and refinery for Jiangsu

640 2005-2014. In particular the contribution of those sources was enhanced in typical  
641 cities with intensive chemical industry. In Nanjing, as an example, the source  
642 categories we measured accounted for 19% of annual emissions from chemical  
643 industry and refinery, and for 10% of the total anthropogenic emissions in 2010.  
644 Figure 6 illustrated the emissions of 445 species (accounting for 99.5% of total  
645 NMVOC emissions) estimated with the source profile before and after updating (this  
646 work) for Jiangsu 2010. As a whole, the difference between emissions of all species  
647 before and after updating was calculated at 281 Gg, i.e., 13% of the total  
648 anthropogenic NMVOC emissions for the province. Due to relatively limited tests  
649 available, the changes in emissions were small for most species when updated profiles  
650 were applied. Relative big changes (over 10 Gg) were found for ethylacetate and  
651 certain aromatics species (benzene, xylene, ethylbenzene, and methylbenzene).  
652 Applying the source profile of paint use measured by Zheng et al. (2013) led to  
653 enhanced ethylacetate. Reduced benzene and methylbenzene, and elevated ethyl  
654 benzene and xylene resulted mainly from the revisions on source profiles of cooking  
655 (Jia et al., 2009; Shi et al., 2015) and solvent use (Wang et al., 2014b; Zheng et al.,  
656 2013). Although incremental information on speciation was obtained through the  
657 on-site measurements and source profile updating in current work, domestic data were  
658 still lacking for many source types, given the big variety of source categories for  
659 industrial process and solvent use. Therefore more efforts on field measurements from  
660 different sectors are still needed in order to establish a more complete database of  
661 chemical profiles for the country and the region.

662 To support the air quality modeling, the emissions of NMVOC species under  
663 CB05 and SAPRC99 mechanisms were calculated using Eq. (4) based on the source  
664 profiles before and after updating, and the results for 2010 were shown in Figure 7  
665 (the speciation for MEIC is illustrated in the figure as well for comparison, as  
666 discussed later in Section 4.3). With source profiles updated based on most recent  
667 measurements, relatively big changes were found for ALDX (see captions of Figure 7  
668 for the detailed meanings) in CB05 and OLE1 in SAPRC99. The revisions on source  
669 profiles of solvent use were the main reason for the changes. For example, the



670 increased ALDX was attributed mainly to the updated profiles of printing ink and  
671 automobile paint use (Zheng et al., 2013; Tang et al., 2014), while increased OLE1  
672 was to that of building coating (Yuan et al., 2010; Wang et al., 2014b) .

### 673 **4.3 Comparisons with other inventories**

674 The total anthropogenic NMVOC emissions in Jiangsu were extracted from other  
675 continental/national/regional inventories and compared with our estimates for various  
676 years in Figure 4. All the results were within the 95% CIs in this work. Except for  
677 REAS that provided 37-77% higher emissions than this work for 2005-2008  
678 (Kurokawa et al., 2013), our estimates were generally 0-18% larger than other studies  
679 in the total emission estimation, attributed mainly to the omission of certain emission  
680 sources in other inventories and to the elevated activity levels from plant-by-plant  
681 investigation in this work. Figure 8 provided the NMVOC emissions by source from  
682 various inventory studies for selected years. As can be seen, the emissions in this  
683 work were 4% and 20% larger than the national inventory for 2005 (Wei et al., 2008)  
684 and regional inventory for 2010 (Fu et al., 2013), respectively. The latter two studies  
685 missed the emissions from the manufacturing processes of certain chemical products.  
686 For example, fermentation alcohol, dye and rubber were not included in Wei et al.  
687 (2008), either glasswork, pesticide or charcoal in Fu et al. (2013). The emissions from  
688 solvent use in this work were larger than those from Bo et al. (2008), attributed to  
689 omission of carpentry coating, pesticide and adhesive using by the latter. The varied  
690 data sources also contributed to the emission discrepancies. For example, Wei et al.  
691 (2008) and Bo et al. (2008) made larger estimates in transportation emissions than us,  
692 as they applied higher values of annual average miles traveled for motorcycles at  
693 national level.

694 The emissions of CB05 and SAPRC 99 species estimated by us and MEIC were  
695 compared in Figure 7. While total NMVOC emissions in this work were 315 Gg or  
696 18% larger than MEIC for 2010, relative changes varied among species and could be  
697 bigger for certain ones. In CB05 mechanism, our results were 46% and 43% smaller  
698 for TOL and XYL but 38% and 59% larger for ETH and ETHA than MEIC (Figure

699 7a), while discrepancies of over 30% (relative to MEIC result) existed for most  
700 species in SAPRC 99 (Figure 7b). Such discrepancies could result either from the  
701 various source profiles, or from the various source contributions in total emissions.  
702 For example, with updated source profile for building coating, much larger OLE1  
703 emissions were estimated in this work than MEIC. Besides the total emissions, the  
704 differed speciations under chemical mechanisms could result in complex impacts on  
705 air quality simulation, which would be discussed in next section.

706 Figure 9 compares the spatial distributions of Jiangsu's NMVOC emissions for  
707 2010 between our results and MEIC. To be consistent in resolution and to ease  
708 visualization, the high-resolution inventory obtained in this work (Figure 2) was  
709 upscaled to  $0.25^{\circ} \times 0.25^{\circ}$ , the same as MEIC. Similar spatial patterns were found for  
710 the two inventories: high emission densities existed in southern Jiangsu with  
711 relatively developed economy and industry. As indicated in Figure 9a, the areas with  
712 big plants and large emissions were consistent with each other, indicating that the  
713 provincial NMVOC emissions were largely influenced by the locations of large point  
714 sources. Figure 9c shows that larger emissions estimated in this work than MEIC were  
715 commonly found in areas with big plants, reflecting the impacts of detailed and  
716 complete investigation on product types and activity levels at plant level on the  
717 emission estimation. Although our result was 18% larger than MEIC in total  
718 anthropogenic NMVOC emissions, lower emissions were found in this work at  
719 downtown Nanjing and the Suzhou-Wuxi-Changzhou city clusters with large  
720 populations. The result implies that downscaling of emissions depending on certain  
721 proxies (e.g., population and economy density) might overestimate the emissions in  
722 urban areas, and detailed information on individual sources should be included if  
723 possible.

#### 724 **4.4 Evaluation of multiple-scale inventories through air quality modeling**

725 The Models-3/Community Multi-scale Air Quality (CMAQ) version 4.7.1 was  
726 applied to test the performances of chemistry transport simulation with various  
727 NMVOC emission inventories for Jiangsu area. As shown in Figure S6 in the

728 supplement, three nested domains (D1, D2 and D3) were set, and the most inner D3  
729 covered the mega city Shanghai and six most developed cities in southern Jiangsu  
730 including Nanjing, Changzhou, Zhenjiang, Wuxi, Suzhou and Nantong, with a  
731 horizontal spatial resolution at 3 km. Chemistry transport simulations were conducted  
732 separately with two inventories, i.e., MEIC and the provincial one developed in this  
733 work, for January, April, July and October 2012. Other model settings on  
734 meteorological simulation, chemistry mechanisms and emissions of natural origin  
735 were the same for the two simulations, as described in Zhou et al. (2017). The first  
736 five days for each month were chosen as spin-up period to provide initial conditions  
737 for later simulations.

738 Figure 10 provides the observed and predicted daily 1h-max O<sub>3</sub> concentrations  
739 for the four months, and Table 6 compares the model performances with MEIC and  
740 our provincial inventory, indicated as normalized mean bias (NMB) and error (NME)  
741 values. As suggested by the minus NMBs for most cases, model usually generated  
742 lower 1h-max O<sub>3</sub> concentrations than observation with either MEIC or provincial  
743 inventory applied, with an exception for April simulation with MEIC applied. The  
744 result thus implied the updated anthropogenic NMVOC emission inventory at  
745 provincial scale was still likely an underestimation of the actual emissions, as YRD  
746 was commonly recognized as VOC-limited region for O<sub>3</sub> formation (Xing et al., 2011).  
747 Compared to MEIC, better model performances (except for July) were generally  
748 achieved when the provincial inventory was applied, indicating the improved  
749 reliability of the detailed bottom-up NMVOC inventory on high-resolution chemistry  
750 transport simulation. In particular, larger emissions were estimated for certain species  
751 with relatively high ozone formation potential (e.g., ethene and ethanol) in the  
752 provincial inventory. It should be noted that the improved ozone simulation was a  
753 combined effect of the updated inventory with revisions on emission estimation,  
754 spatial distribution and source profiles for all the relevant species, and that the impacts  
755 of emission changes for individual species could not be completely validated. More  
756 chemistry transport modeling is further encouraged with intensive sensitivity analysis.

757 The discrepancies between simulation and observation were still large compared

758 to regional studies in North America (Y. Zhang et al., 2009). More efforts on  
759 improving or validating emission inventory at provincial scale are thus in great  
760 needed. Besides careful compilation of emission source information in the bottom-up  
761 method, observation constraint from ground measurements could be used to evaluate  
762 the emission level, source contribution, and speciation of VOC emissions (M. Wang et  
763 al., 2014). Emission uncertainty of NO<sub>x</sub> could also partly explain the discrepancies, as  
764 the NO<sub>x</sub> control measures taken recently could hardly be fully tracked in the emission  
765 inventory development. Besides the limitation of emission input, more analysis on the  
766 impacts of chemical mechanisms and dynamics in the chemistry transport modeling  
767 are also suggested for O<sub>3</sub> prediction in the region.

## 768 **5 Conclusion**

769 Using a bottom-up approach, we developed a high-resolution emission inventory  
770 of anthropogenic NMVOC for Jiangsu province, eastern China, with substantial  
771 detailed information on local sources and source profiles from domestic tests  
772 incorporated. Attributed largely to the elevated contribution from industrial processes  
773 and solvent use, the annual provincial emissions were estimated to increase 41% from  
774 2005 to 2014. Influenced largely by location of big point sources, high emission  
775 densities were found in cities along the Yangtze River. Our estimations were larger  
776 than results from most other available inventories except REAS, due mainly to the  
777 omissions of certain industrial and solvent use sources by other studies and to the  
778 elevated activity levels from plant-by-plant investigation in this work. Benefiting from  
779 more detailed classification and investigation of emission sources, reduced  
780 uncertainties in annual emissions were quantified in this work compared to previous  
781 studies. Varied data sources and methods were of significant impacts on emission  
782 estimation at city/plant level. In particular, downscaling directly from national  
783 inventories would overestimate the fraction of emissions in urban downtown. With the  
784 most recent source profiles from local measurements included, considerable changes  
785 in emissions were found for ethylacetate and certain aromatics species, and the

786 speciation under CB05 and SAPRC99 differed a lot from the national inventory  
787 MEIC. Compared to MEIC, better model performance (indicated by daily 1h-max O<sub>3</sub>  
788 concentrations) were achieved when the improved provincial inventory was used in  
789 CMAQ simulation, although the discrepancies between simulation and observation  
790 need further investigation. As emission controls on NMVOCs started to be  
791 implemented on industrial sources in China (e.g., the application of leak detection and  
792 repair technique in chemical and refinery plants), more field tests were recommended  
793 to better track the temporal changes in emission factors and to reduce the uncertainty  
794 of emission estimation in the future.

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## FIGURE CAPTIONS

Figure 1. NMVOC source profiles by grouped species measured in chemical enterprises (a) and the comparisons with SPECIATE results for synthetic rubber (b), ethylene (c), and polyethylene production (d).

Figure 2. Spatial distribution of Jiangsu's anthropogenic NMVOC emissions (3×3km) for 2005 (a), 2010 (b), and 2014(c); and the differences between 2005 and 2014 (2014 minus 2005, d).

Figure 3. Fractions of NMVOC species and the ratios of OFPs to emission by sector for Jiangsu 2014.

Figure 4. The inter-annual trends in anthropogenic NMVOC emissions in Jiangsu from 2005 to 2014. The grey dotted lines indicate the 95% CIs of emissions estimated in this study. The results from other inventories are illustrated as well for comparison.

Figure 5. Spatial distributions of NMVOC emissions from chemical and refinery industry for Nanjing 2011 (3×3km), estimated using the detailed enterprise information (a), Environmental Statistics (b), and economic statistics without any information on individual sources (c). The black dots indicate the largest ten emitters in the city

Figure 6. Emissions of NMVOC species for Jiangsu 2010 before and after source profile updating.

Figure 7. Emissions of CB05 (a) and SAPRC99 (b) species for Jiangsu 2010. The left and middle columns indicate the results before and after updating of source profiles, and the right columns indicate the results of MEIC. The CB05 species PAR represents paraffin carbon bond, UNR for unreactive parts of molecules, OLE for terminal olefin carbon bond, TOL for toluene and other monoalkylaromatics, XYL for xylene and other polyalkylaromatics, FORM for formaldehyde, ALD2 for acetaldehyde, ETH for ethene, MEOH for methanol, ETOH for ethanol, ETHA for ethane, IOLE for internal olefin carbon bond, ALDX for C3+ aldehydes, NVOL for nonvolatile mass. The

SAPRC 99 species ALK1, ALK2, ALK3, ALK4 and ALK5 represent alkanes and other non-aromatic compounds that react only with OH, and have kOH between 2 and 500, 500 and 2500, 2500 and 5000, 5000 and 10000, greater than 10000 ppm<sup>-1</sup> min<sup>-1</sup>, respectively. ETHENE respect ethane, and OLE1 for alkenes with kOH smaller than 70000 ppm<sup>-1</sup> min<sup>-1</sup>, OLE2 for alkenes with kOH greater than 70000 ppm<sup>-1</sup> min<sup>-1</sup>, ARO1 for aromatics with kOH smaller than 20000 ppm<sup>-1</sup> min<sup>-1</sup>, ARO2 for aromatics with kOH greater than 20000 ppm<sup>-1</sup> min<sup>-1</sup> and NROG for unreactive mass.

Figure 8. Jiangsu's anthropogenic NMVOC emissions by sector for selected years estimated from this work and other inventories. Note emissions in this work are regrouped to be consistent in source categories with Zhang et al. (2009) and MEIC for direct comparison.

Figure 9. Spatial distributions of Jiangsu's anthropogenic NMVOC emissions for 2010 (0.25°×0.25°) by this study (a) and MEIC (b), and the differences between the two inventories (this study minus MEIC, d). The black dots indicate the largest fifty emitters in the province.

Figure 10. Observed and simulated daily 1h-max O<sub>3</sub> concentrations using MEIC and provincial emission inventory in January (a), April (b), July (c) and October (d) in 2012 at the air quality monitoring sites in Nanjing. Note the different y-axis scales between panels.

**Table 1 Categories of anthropogenic NMVOC emission sources.**

Sector	Subsector	Product/fuel/solvent used	Product/technology
Fossil fuel stationary combustion	Power plant	Coal/oil/natural gas/waste/biofuel	N.A.
	Heating and industrial boiler	Coal/coke/oil/natural gas	N.A.
	Residential	Coal/oil/LPG/natural gas	N.A.
Biomass burning	Boiler	N.A.	N.A.
	Stove burning	Crop straw/wood	N.A.
	Open burning	Crop straw	Rice/corn/wheat/other
Industrial process	Iron and steel	Coking	Mechanical/indigenous
		Sinter/pellet/crude steel	N.A.
	Non-metallic mineral	Glass	Flat glass/glass fiber/glass work
		Cement clinker/lime/brick and tile/ceramic	N.A.
	Oil exploitation and refinery	Crude oil exploit/crude oil refinery	N.A.
	Chemical industry	Chemical raw materials: Ethylene/benzene/methanol/acetic acid/synthesis ammonia/phthalic acid/ethylene oxide/vinylacetate/styrene/glycol/octanol/butanol	N.A.
		Synthetic chemical industry: synthetic resin	Polyethylene/polypropylene/Polystyrene/polyvinylchloride
		Synthetic chemical industry: synthetic fiber	viscose/cellulose acetate fiber/nylon fiber
		Synthetic chemical industry: synthetic rubber	N.A.
		Fine chemical industry: pharmaceutical/chemical pesticide/paint/printing ink/adhesive/dye	N.A.
	Food and wine	fermentation alcohol/spirit/beer	N.A.
Bread/cake/biscuit		N.A.	
Vegetable oil extraction		N.A.	

Table 1 (continued)

Sector	Subsector	Product/fuel/solvent used	Product/technology
Industrial process	Plastic and rubber product	Foam plastic/plastic products/tire/other rubber product	N.A.
	Textile industry	Silk/cloth/woolen yarn	N.A.
	Carbon black		N.A.
Solvent use	Paint use	Interior/exterior building coating	Water-based/solvent-based
		Automobile production/repairing	N.A.
		Wood decoration/wooden furniture paint	N.A.
		Anticorrosive paint	N.A.
		Other paint	N.A.
	Printing	New-type/traditional ink	N.A.
	Dyeing		N.A.
	Adhesive use	Shoe making/timber production/other	N.A.
Other solvent use	Agriculture pesticide/Dry cleaning/household solvent use/solvent degreasing	N.A.	
Transportation	On-road	Automobile	Load (light/heavy)-fuel (gasoline/diesel)
		Motorcycle	Gasoline
	Off-road	Train/inland ship/construction machine/tractor/agriculture truck/agriculture machine	N.A.
Oil distribution	Crude oil/gasoline/diesel	Storage/transport/load & unload/gas station	N.A.
Others		Garbage disposal	Burning/landfill/compost
		Cooking fume	N.A.

**Table 2 NMVOC emission factors for combustion sources, industrial processes and solvent use. Unless specifically noted, the units are g/kg-fuel, g/kg-product and g/kg-solvent, respectively.**

Source	Fuel/product/solvent used		EFs in this study	EFs in the references
<b>Combustion</b>				
Fossil fuel	Power plant	Coal	0.15	0.15 <sup>a</sup> ; 0.81 <sup>b</sup> ; 0.02 <sup>c</sup> ; 0.03-0.03 <sup>d</sup> ; 0.02 <sup>e</sup>
		Fuel oil	0.09	3.88 <sup>b</sup> ; 0.04 <sup>c</sup> ; 0.12 <sup>d</sup> ; 0.09-0.14 <sup>e</sup>
		Natural gas (g/m <sup>3</sup> )	0.083	0.12 <sup>c</sup> ; 0.08-0.10 <sup>d</sup> ; 0.08-0.10 <sup>e</sup>
	Heating and industrial boiler	Coal	0.18	0.18 <sup>a</sup> ; 3.95 <sup>b</sup> ; 0.04 <sup>c</sup> ; 0.03-0.03 <sup>d</sup> ; 0.02 <sup>e</sup>
		Fuel oil	0.12	3.88 <sup>b</sup> ; 0.12 <sup>c</sup> ; 0.12 <sup>d</sup> ; 0.09-0.14 <sup>e</sup>
		Natural gas (g/m <sup>3</sup> )	0.094	0.1 <sup>c</sup> ; 0.08-0.10 <sup>d</sup> ; 0.08-0.10 <sup>e</sup>
	Residential	Coal	4.5	0.6 <sup>a</sup> ; 3.08 <sup>b</sup> ; 6.48, 4.98 <sup>g</sup> ; 4.5 <sup>c</sup> ; 6.41 <sup>d</sup>
		Fuel oil	0.35	0.35 <sup>c</sup>
		LPG	5.29	0.17 <sup>a,b</sup> ; 3.28 <sup>g</sup> ; 5.29 <sup>c</sup> ; 66 (g/m <sup>3</sup> ) <sup>e</sup>
		Natural gas (g/m <sup>3</sup> )	0.15	0.13 <sup>c</sup> ; 0.15 <sup>c</sup> ; 0.14 <sup>d</sup> ; 0.18 <sup>e</sup>
Coal gas		0.00044	0.00044 <sup>a</sup>	
Biomass burning	Boiler	Biomass	1.1	0.0015 <sup>e</sup> ; 1.1 <sup>f</sup>
	Stove burning	Wood	3.23	1.09-4.94 <sup>g</sup> ; 3.23 <sup>c</sup> ; 5.3 <sup>e</sup>
		Crop straw	13.77	1.7- 3.0 (corn straw) <sup>g</sup> ; 8.89 (wheat straw) <sup>g</sup> ; 13.77 <sup>c</sup> ; 8.55 <sup>d</sup> ; 5.3 <sup>e</sup>
	Open burning	Rice straw	7.48	7.48 <sup>h</sup>
		Wheat straw	7.48	7.48 <sup>h</sup>
		Corn straw	10.4	10.4 <sup>h</sup>
		Other straws	8.94	Average of straws open burning above
<b>Industrial process</b>				
Iron and steel	Coking/sintering/steel		3.96/0.25/0.06	3.4 <sup>c</sup> ; 3.96 <sup>f</sup> /0.25 <sup>f</sup> /0.06 <sup>f</sup>
Non-metallic mineral	Flat glass/glass fiber/glass work		4.4/3.15/4.4	4.4 <sup>c,f</sup> ; 3.5 <sup>e</sup> /3.15 <sup>a,i</sup> /4.4 <sup>f</sup>
	Cement clinker/lime/brick and tile/ceramic		0.33/0.177/0.13/29.22	0.12 <sup>c</sup> ; 0.018 <sup>d</sup> ; 0.33 <sup>f</sup> /0.177 <sup>f</sup> /0.01 <sup>i</sup> ; 0.2 <sup>c</sup> ; 0.033 <sup>a</sup> ; 0.13 <sup>f</sup> /29 <sup>c</sup> ; 29.215 <sup>a</sup> ; 29.22 <sup>c</sup>
Oil exploitation/refinery			1.42/1.82	1.42 <sup>f</sup> /1.82 <sup>f</sup> ; 1.05 <sup>d</sup> ; 3.54 <sup>e</sup>
Chemical industry	Ethylene/benzene/methanol		0.097/0.1/5.95	0.097 <sup>f</sup> ; 0.6 <sup>d</sup> /0.1 <sup>f</sup> ; 0.25 <sup>j</sup> /5.95 <sup>f</sup>
	Polyethylene/polypropylene		10/8	10 <sup>f,j</sup> ; 0.33 <sup>e</sup> ; 3.4 <sup>d</sup> /8 <sup>f,j</sup> ; 0.35 <sup>e</sup> ; 4 <sup>d</sup> ;
	Polystyrene/polyvinyl chloride		5.4/3	5.4 <sup>f</sup> ; 0.21-3.34 <sup>e</sup> ; 0.12 <sup>d</sup> /3 <sup>f,j</sup> ; 0.33-8.5 <sup>e</sup> ; 0.096 <sup>d</sup>
	Acetic acid/ammonia/phthalic anhydride		1.814/4.72/21	1.81 <sup>i</sup> /4.72 <sup>f,e</sup> /1.1-6.3 <sup>e</sup> ; 1.3-6 <sup>d</sup> ; 21 <sup>j</sup>
	Ethylene oxide/vinyl acetate/styrene		3/4.705/0.223	3 <sup>f</sup> ; 0.98 <sup>e</sup> ; 2 <sup>d</sup> ; 4 <sup>j</sup> /4.705 <sup>i</sup> /0.223 <sup>f</sup> ; 1 <sup>d</sup> ; 3.1 <sup>j</sup>
	Viscose fiber/cellulose acetate fiber/nylon fiber		14.5/73.4/3.3	14.5 <sup>c,f</sup> /73.4 <sup>f</sup> ; 112 <sup>e</sup> /2.13-3.93
	Polyester fiber/acrylic fibers polypropylene fiber		0.7/40/37.1	0.7 <sup>f</sup> ; 0.6 <sup>e</sup> /3.75-40 <sup>e</sup> /37.1 <sup>c</sup>



Table 2 (continued)

Source	Fuel/product/solvent used	EFs in this study	EFs in the references
<b>Industrial process</b>			
Chemical industry	Synthetic rubber	7.17	7.17 <sup>f,e</sup> ; 3.8-8.6 <sup>e</sup> ; 0.27-9 <sup>d</sup> ;
	Pharmaceutical/chemical pesticide	430/20	430 <sup>f,e</sup> ; 300 <sup>d</sup> /20 <sup>k</sup>
	Paint/printing ink/adhesive/dye/pigment	15/50/30/81.4/10	15 <sup>f,e</sup> ; 11 <sup>d</sup> /50 <sup>f</sup> ; 60 <sup>e</sup> /30 <sup>c</sup> ; 20 <sup>f</sup> /81.4 <sup>f</sup> /10 <sup>i</sup>
Plastic and rubber	Foamed plastic /plastic products	120/3.2	770 <sup>c</sup> ; 120 <sup>d</sup> /3.2 <sup>f,j</sup>
	Tire/other rubber products	0.91(kg/tire)/12.5	0.285(kg/tire) <sup>m</sup> ; 0.285(kg/tire) <sup>l</sup> ; 0.91(kg/tire) <sup>f</sup> ; 10 <sup>d</sup> ; 0.659 <sup>i</sup> /12.5 <sup>c</sup>
Food and wine	Fermentation alcohol /white spirit	60/25	218.25 <sup>f</sup> ; 60 <sup>d</sup> /25.35 <sup>l</sup> ; 25 <sup>c,f</sup> ; 150(g/L) <sup>d</sup> ; 33.105(g/L) <sup>i</sup>
	Beer/grape wine	0.25/0.5	0.44 <sup>l</sup> ; 0.4 <sup>c</sup> ; 0.43(g/L) <sup>a</sup> ; 0.25 <sup>f</sup> ; 0.35(g/L) <sup>d</sup> /0.81 <sup>l</sup> ; 0.5 <sup>c,f</sup> ; 0.8(g/L) <sup>d</sup> ; 0.38(g/L) <sup>e</sup>
	Cake and biscuit	1	1 <sup>l,c,d,f</sup>
	Vegetable oil extraction	3.7	3.45-10.35 <sup>l</sup> ; 5.5 <sup>c</sup> ; 3.45 <sup>a</sup> ; 1.57(g/L) <sup>d</sup> ; 4 <sup>i</sup>
Textile industry	Silk/cloth/woolen	10/10/10	10 <sup>b</sup> /10 <sup>b</sup> /10 <sup>b</sup>
Carbon black	N.A.	64.7	90 <sup>m</sup> ; 73.2 <sup>l</sup> ; 64.7 <sup>c</sup> ; 52 <sup>f</sup> ; 0.7 <sup>d</sup> ; 53.57 <sup>e</sup> ;
<b>Solvent use</b>			
Printing	New-type/traditional ink	100/750	100 <sup>f</sup> ;150 <sup>e</sup> /750 <sup>f</sup> ; 650 <sup>c</sup> ,500
Dyeing	-	81.4	81.4 <sup>e,f</sup>
Paint use	Interior wall painting	250; 120*	250 <sup>c</sup> /120 <sup>n</sup>
	Exterior wall painting: aqueous/solvent	120/580	120 <sup>f</sup> /360 <sup>f</sup> ;585 <sup>c</sup> ;580 <sup>n</sup> ;230 <sup>d</sup>
	Vehicle manufacture/refinishing	470/720	460 <sup>n</sup> ; 470 <sup>c</sup> ; 475 <sup>l</sup> /720 <sup>c,d</sup> ; 850 <sup>l</sup>
	Furniture: aqueous/solvent based	250/670	250 <sup>c</sup> /640 <sup>f</sup> ; 637 <sup>n</sup> ; 550-750 <sup>c</sup> ;
	Anticorrosive paint use	442	442 <sup>c</sup>
	Other paint use	240	240 <sup>c</sup>
Adhesive use	Shoe making	670	664 <sup>n</sup> ; 670 <sup>c</sup>
	Timber production	90	88 <sup>n</sup> ; 90 <sup>c</sup>
	Other adhesive use	89	89 <sup>c</sup> ; 85 <sup>l</sup>
Other solvent	Agriculture pesticide	470	368-482 <sup>c</sup> ; 356-576 <sup>f</sup>
	Dry cleaning	0.16**	0.8 <sup>e</sup> ;
	Household solvent use	0.08	0.08 <sup>f</sup> ; 3.7 <sup>d</sup> ; 4.2 <sup>e</sup> ; 0.1 <sup>a</sup>
	Degreasing	0.044	0.044 <sup>a,f</sup>

<sup>a</sup>Bo et al. (2008); <sup>b</sup>Tang and Chen (2002); <sup>c</sup>Wei et al. (2008), Wei(2009), and Wang et al.(2009);

<sup>d</sup>EEA (2013); <sup>e</sup>USEPA (2002); <sup>f</sup>MEP (2014); <sup>g</sup>Zhang et al. (2000); <sup>h</sup>Li et al. (2007); <sup>i</sup>ROC EPA (2009); <sup>j</sup>Fan et al. (2012); <sup>k</sup>Xia et al. (2014); <sup>l</sup>Wang (2006); <sup>m</sup>Klimont et al. (2002); <sup>n</sup>Fu et al. (2013)

\*250 and 120 g/kg for 2005-2007 and 2008-2014, respectively; \*\*Adjusted by per capital income.

**Table 3 Anthropogenic NMVOC emissions in Jiangsu by source from 2005 to 2014 (Gg).**

Source	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014
Fossil fuel combustion	43	43	45	47	48	50	54	54	58	59
Industrial process	461	580	616	626	722	823	747	821	871	958
Transportation	325	321	336	331	331	317	313	302	311	280
Solvent use	380	441	492	495	606	652	697	783	809	966
Oil distribution	35	35	39	42	46	53	56	64	59	53
Biomass burning	458	343	355	147	139	131	126	119	116	110
Other	72	73	74	74	75	76	77	78	78	79
Total	1774	1835	1956	1762	1967	2102	2071	2220	2302	2507

**Table 4 Updated NMVOC source profiles in this study and those from SPECIATE and Li et al. (2014).**

Sector		Species (weight percentage, %)					
		Alkanes	Alkenes	Alkynes	Aromatics	OVOCs	Others
Biomass open burning	This study	14.83	17.24	2.20	10.98	52.58	2.16
	SPECIATE	14.20	20.59	2.62	1.82	57.68	3.08
Coking	This study	25.65	25.00	1.34	25.42	1.43	21.16
	SPECIATE	9.17	39.45	2.14	49.24	0.00	0.00
Iron& steel	This study	25.64	11.23	15.56	13.60	2.36	31.62
	SPECIATE	28.06	16.67	27.72	0.00	0.00	27.56
Paint production	This study	3.90	0.00	0.00	48.61	42.76	4.72
	SPECIATE	0.00	0.00	0.00	80.65	19.35	0.00
Ink production	This study	54.48	15.27	0.00	30.25	0.00	0.00
	SPECIATE	5.00	0.00	0.00	6.50	63.00	25.50
Refinery	This study	77.02	12.10	0.00	10.89	0.00	0.00
	Li et al., 2014	67.57	9.44	0.91	1.96	0.00	20.12
Furniture painting	This study	6.15	0.14	0.00	61.61	29.23	2.87
	SPECIATE	0.00	0.00	0.00	30.00	51.42	18.58
Architecture painting	This study	28.69	9.39	0.00	61.92	0.00	0.00
	Li et al., 2014	27.48	4.71	0.00	67.81	0.00	0.00
Vehicle varnish paint	This study	3.65	0.55	0.06	59.09	32.02	4.64
	Li et al., 2014	2.14	0.82	0.08	96.96	0.00	0.00
Printing	This study	29.54	2.90	0.69	16.64	44.89	5.33
	Li et al., 2014	14.78	2.79	0.66	11.03	26.96	43.78

Table 4 (continued)

Sector		Species (weight percentage, %)					
		Alkanes	Alkenes	Alkynes	Aromatics	OVOCs	Others
Diesel vehicle	This study	44.30	22.41	1.79	20.10	11.39	0.00
	Li et al., 2014	14.88	11.60	0.83	8.99	48.99	14.71
Motorcycle	This study	41.36	23.36	2.11	28.50	4.68	0.00
	Li et al., 2014	45.88	32.18	0.56	21.37	0.00	0.00

**Table 5 Uncertainties of anthropogenic NMVOC emissions (expressed 95% CI around the central estimates) and the most significant two parameters contributing to the uncertainties by sector for Jiangsu 2014. The percentages in the parentheses indicate the contributions of the parameters to the variance of emissions. The uncertainties from other inventories are provided for comparisons.**

Source Category	Uncertainty					Parameters contributing most to uncertainty	
	This work	Wei (2009)	Bo et al. (2008)	Fu et al. (2013)	Huang et al. (2011)	This work	
	Provincial scale	National scale	National scale	Regional scale	Regional scale	Provincial scale	
Fuel stationary combustion	-66%, +190%	-	-	-	-	<i>EF</i> Power plant, coal (68%)	<i>EF</i> Industrial boiler, coal (6%)
Industrial process	-58%, +152%	-88%, +283%	-	-57%, +152%	-60%, +152%	<i>EF</i> Tire (23%)	<i>EF</i> Coking (13%)
Solvent use	-68%, +131%	-82%, +223%	-	-60%, +147%	-59%, +150%	<i>AL</i> External wall paint (20%)	<i>EF</i> Other paint (14%)
Transportation	-51%, +117%	-86%, +261%	-	-	-	<i>EF</i> Inland ship (13%)	<i>EF</i> Construction machine (8%)
Oil distribution	-66%, +162%	-	-	-	-	<i>EF</i> Crude oil storage (27%)	<i>EF</i> Gasoline sale (23%)
Biomass burning	-76%, +499%	-	-	-	-	<i>EF</i> Straw-stove (74%)	<i>R</i> Straw burning in stove <sup>1</sup> (5%)
Other	-98%, +490%	-	-	-	-	<i>EF</i> Cooking (84%)	<i>EF</i> Garbage burning (14%)
Total	-41%, +93%	-51%, +133%	-36%, +94%	-52%, +105%	-53%, +113%		

<sup>1</sup> The ratio of straw burned in stove as biofuel

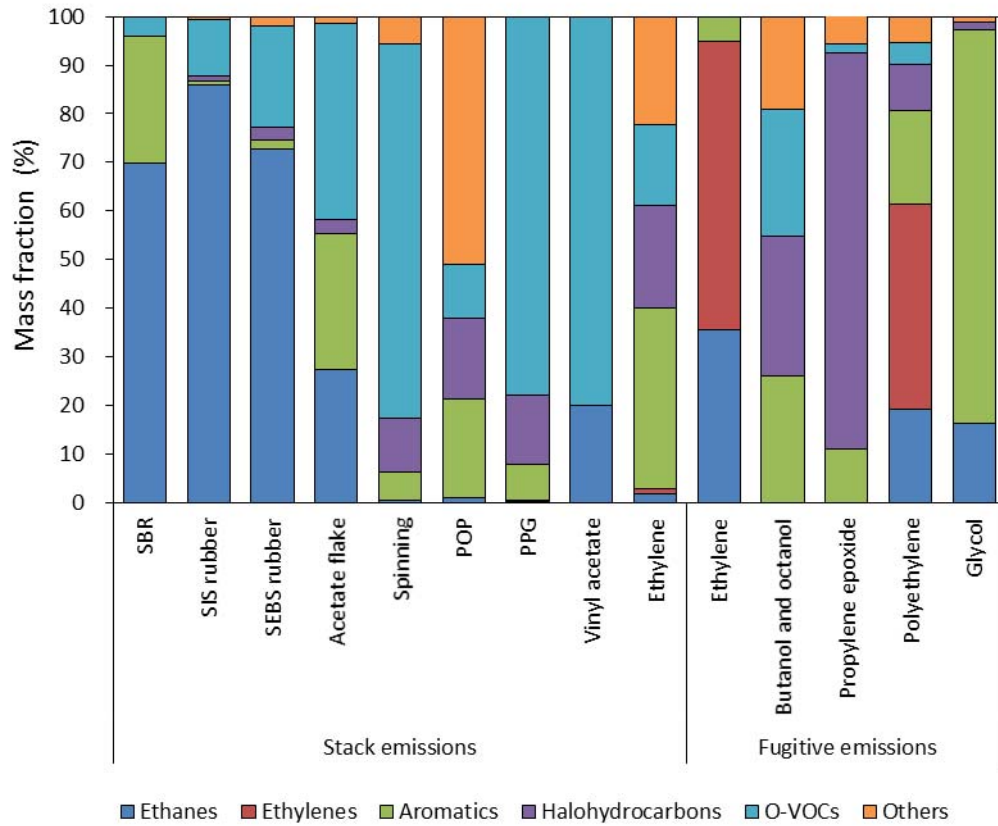
**Table 6 Model performance of daily 1h-max O<sub>3</sub> concentrations using MEIC and provincial inventory for January, April, July and October 2012 in Nanjing.**

	Provincial emission inventory		MEIC	
	NMB <sup>1</sup>	NME <sup>1</sup>	NMB	NME
January	-21%	34%	-58%	59%
April	-26%	38%	35%	55%
July	-28%	33%	-23%	29%
October	-20%	26%	-50%	50%

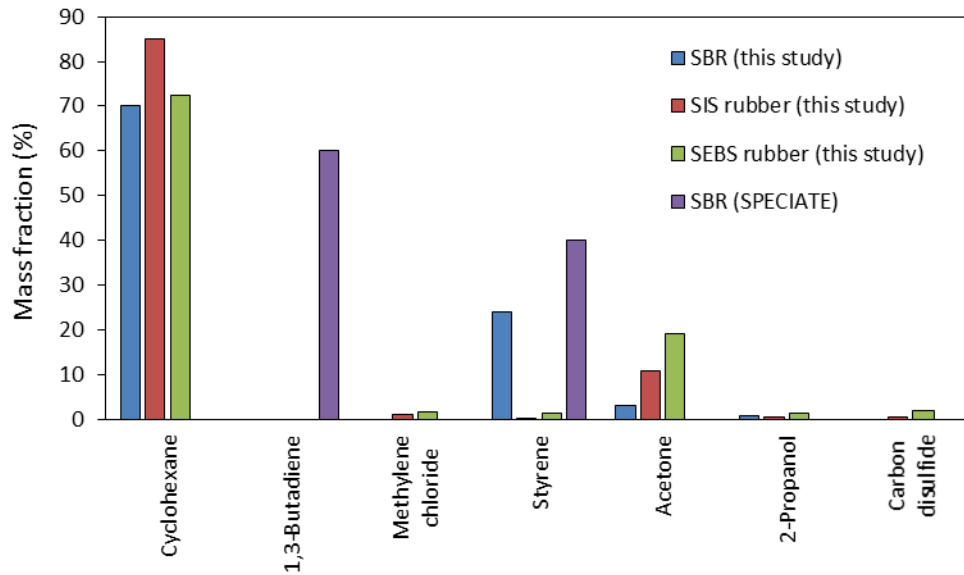
<sup>1</sup> Normalized mean bias (NMB) and error (NME) were calculated as following equations ( $P_i$  and  $O_i$  indicate the results from modeling prediction and observation, respectively):

$$NMB = \frac{\sum_{i=1}^n (P_i - O_i)}{\sum_{i=1}^n O_i} \times 100\% \quad ; \quad NME = \frac{\sum_{i=1}^n |P_i - O_i|}{\sum_{i=1}^n O_i} \times 100\%$$

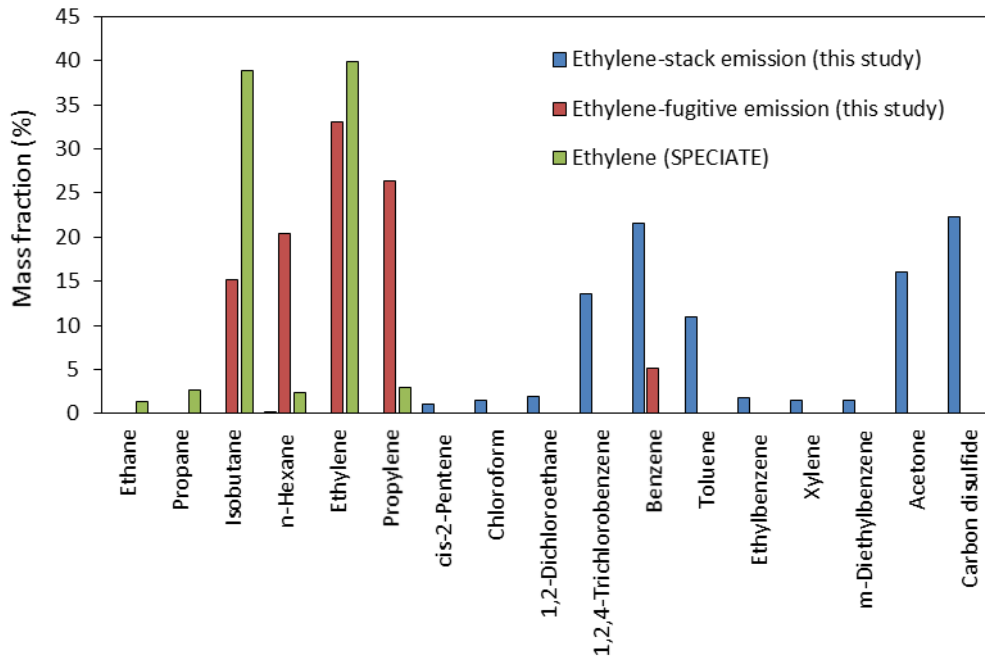
**Figure 1**



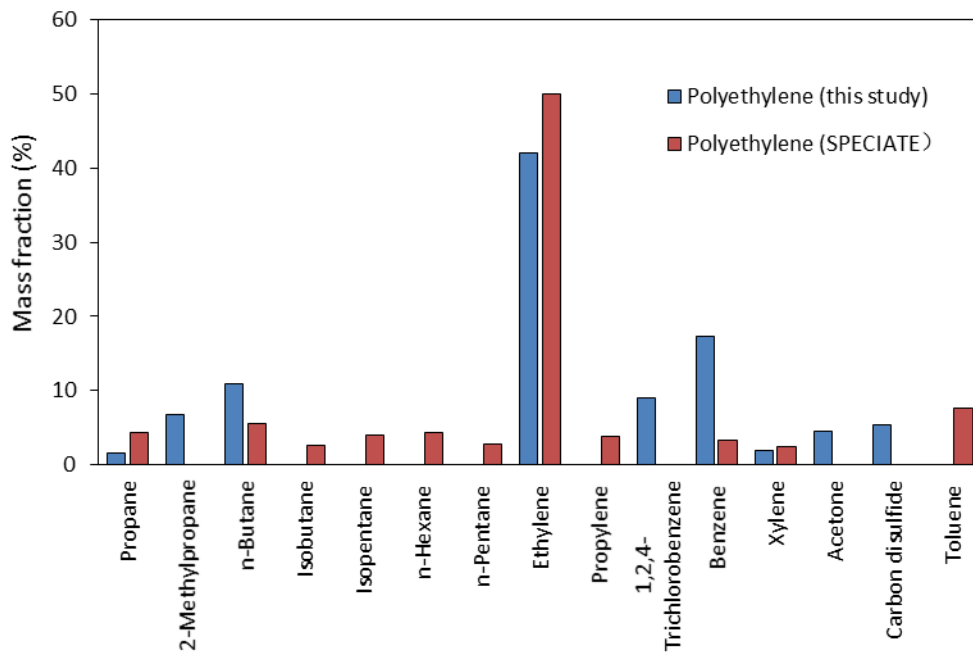
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(b)



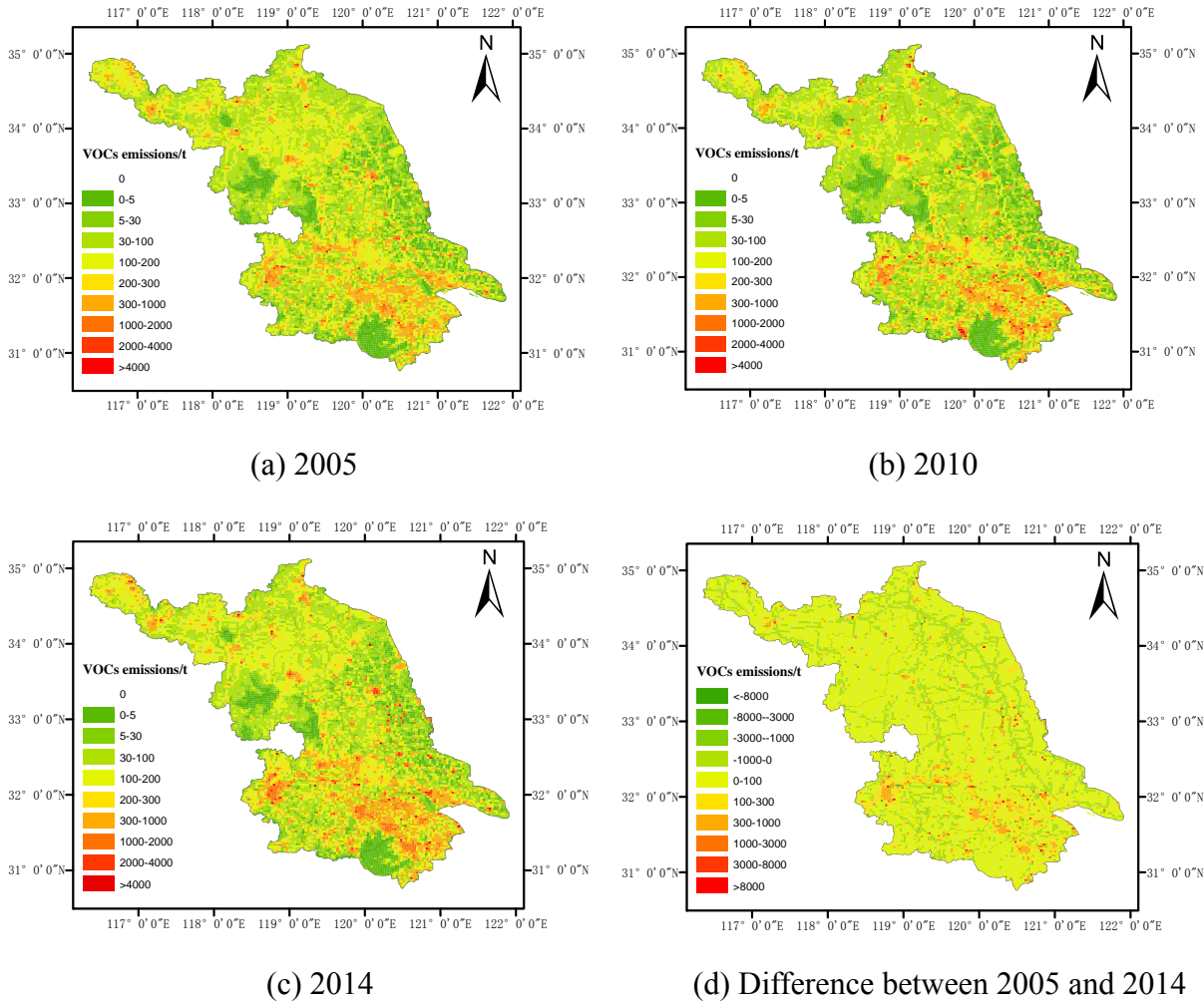
(c)



(d)



**Figure 2**



**Figure 3**

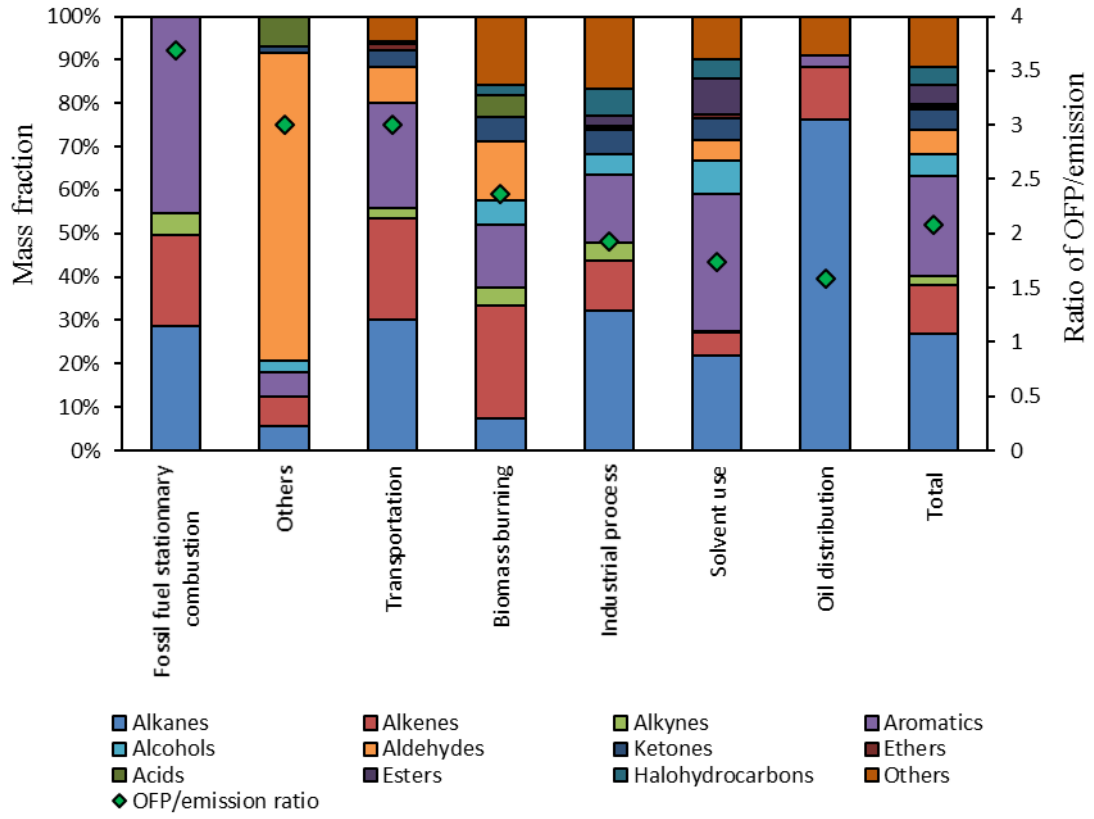


Figure 4

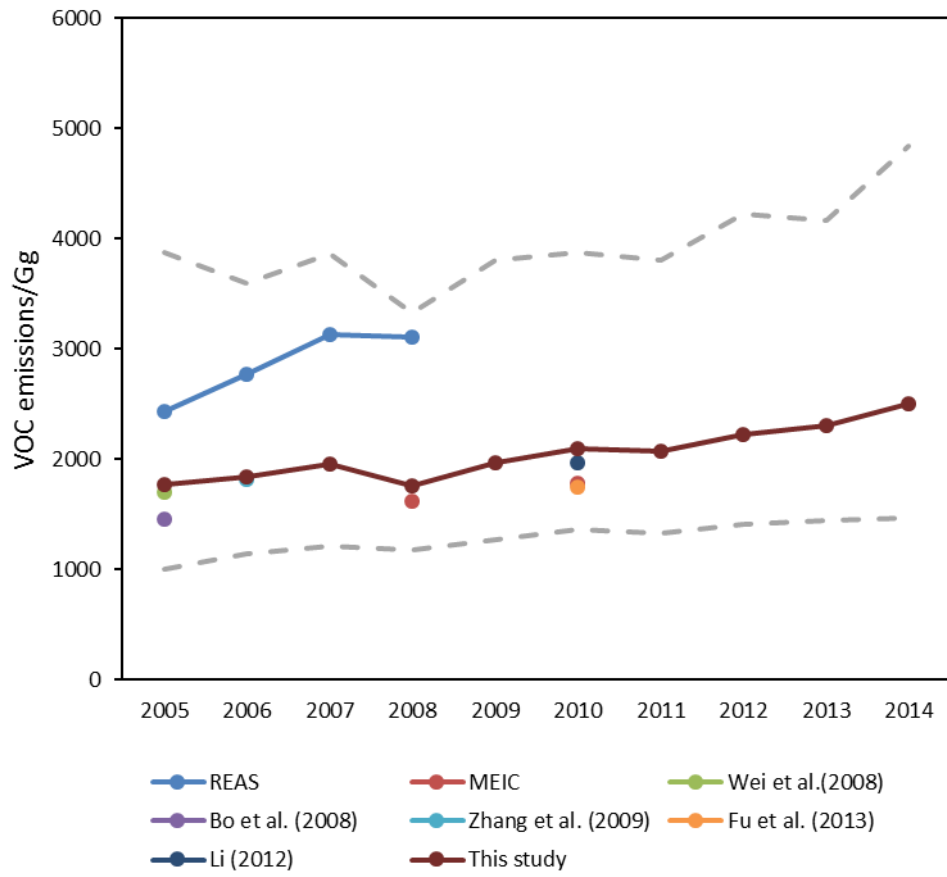
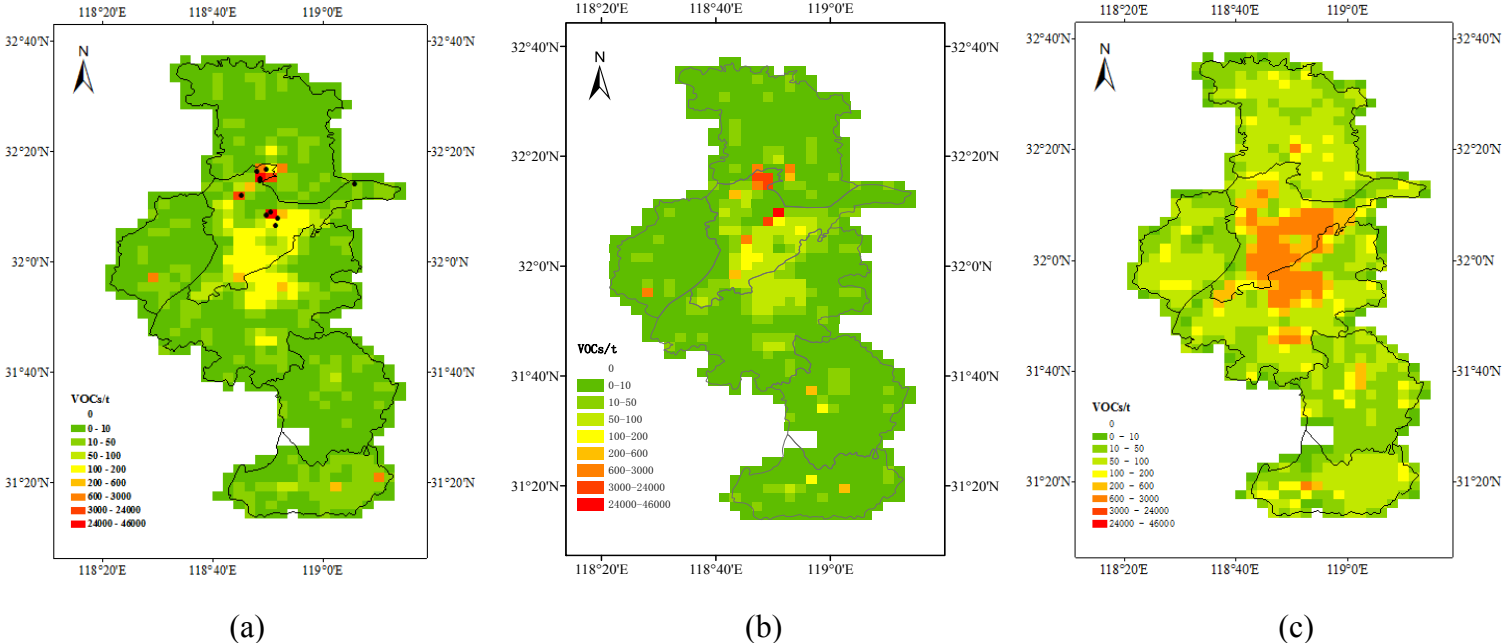
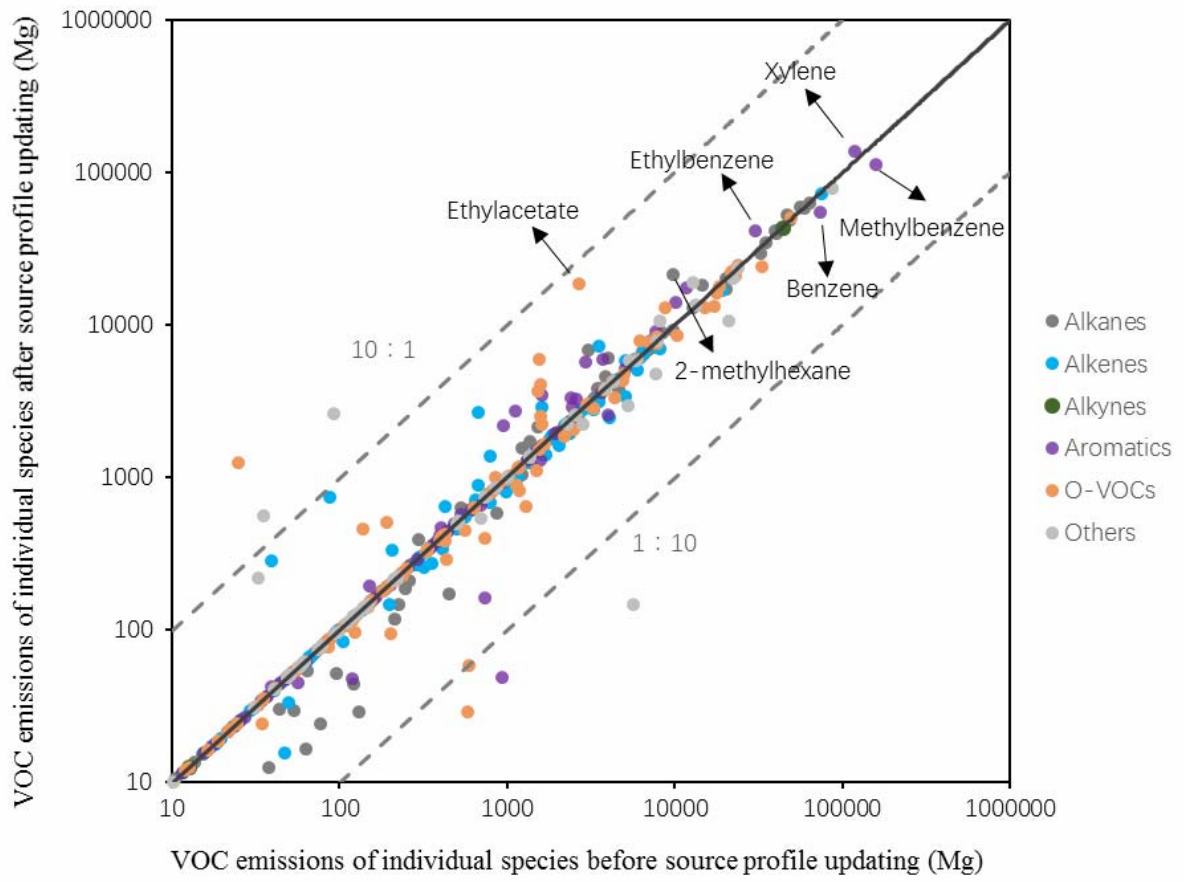


Figure 5

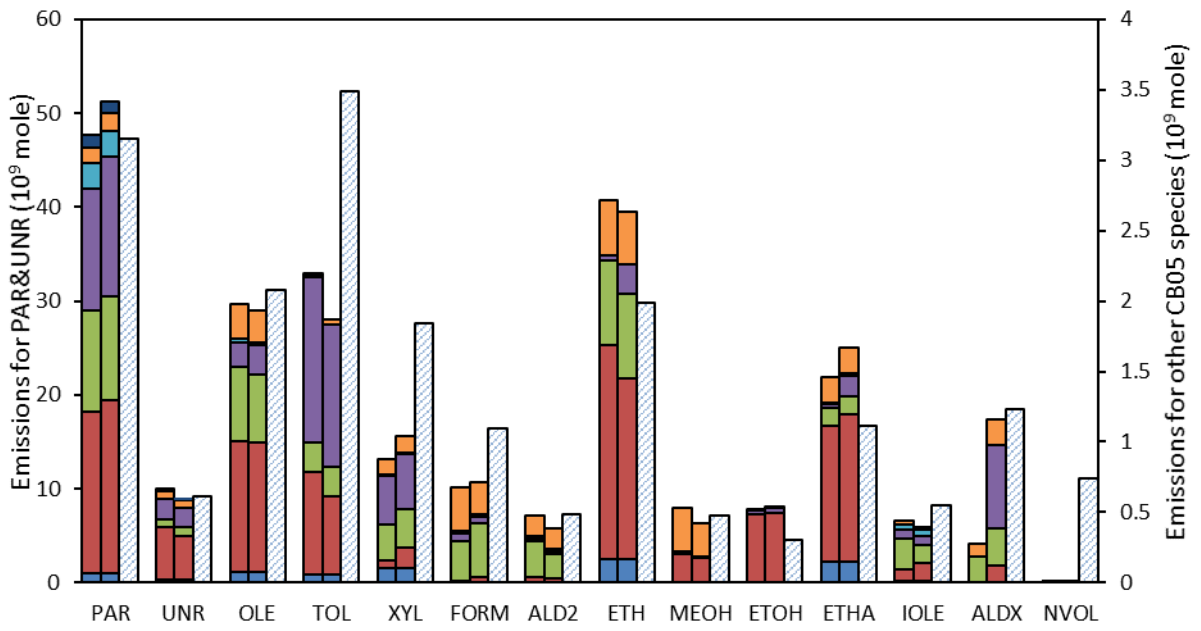


**Figure 6**

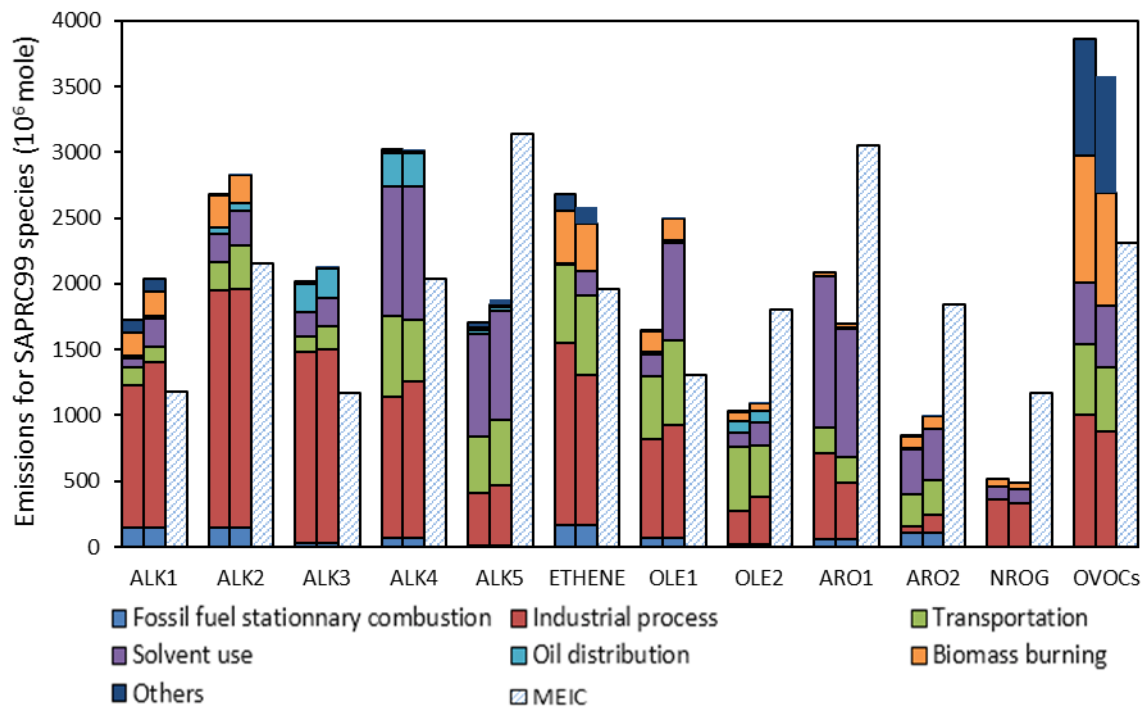


**Figure 7**

(a) CB05



(b) SAPRC99



**Figure 8**

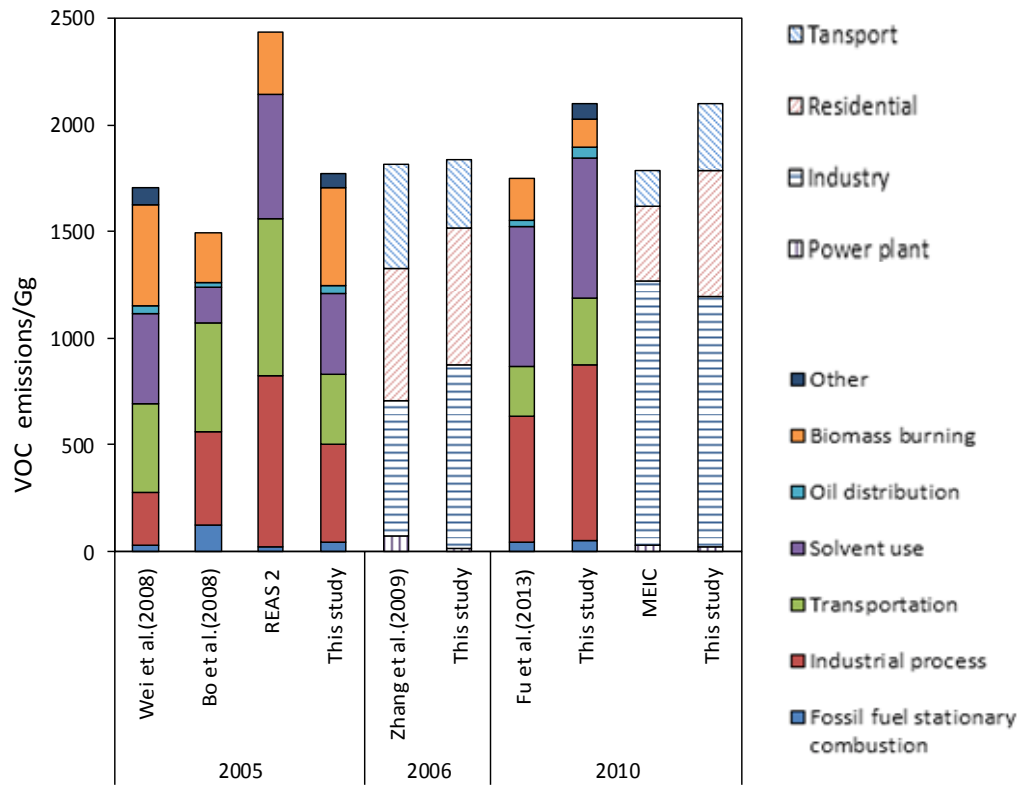
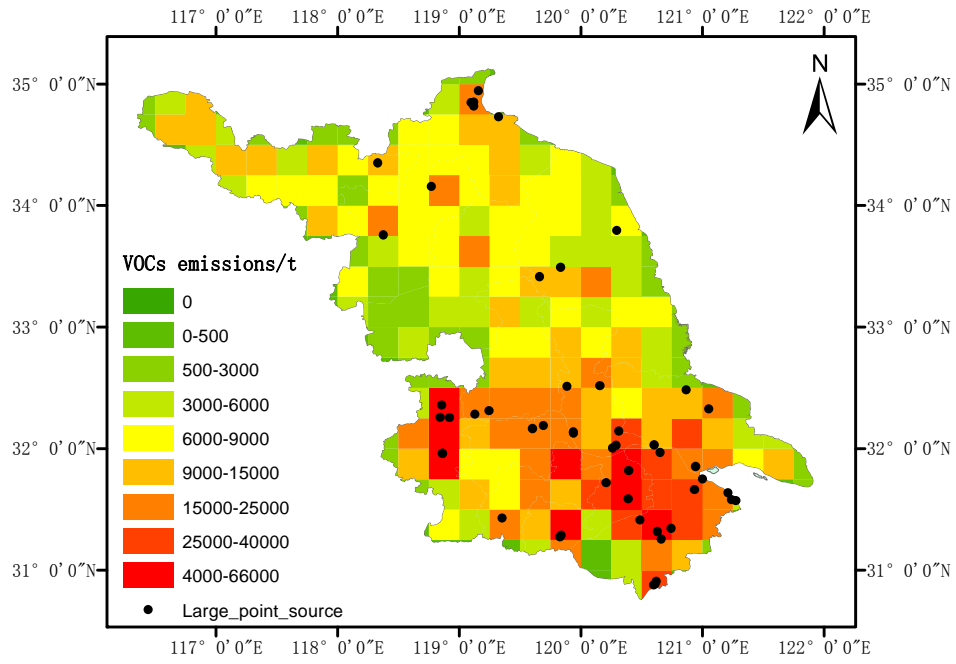
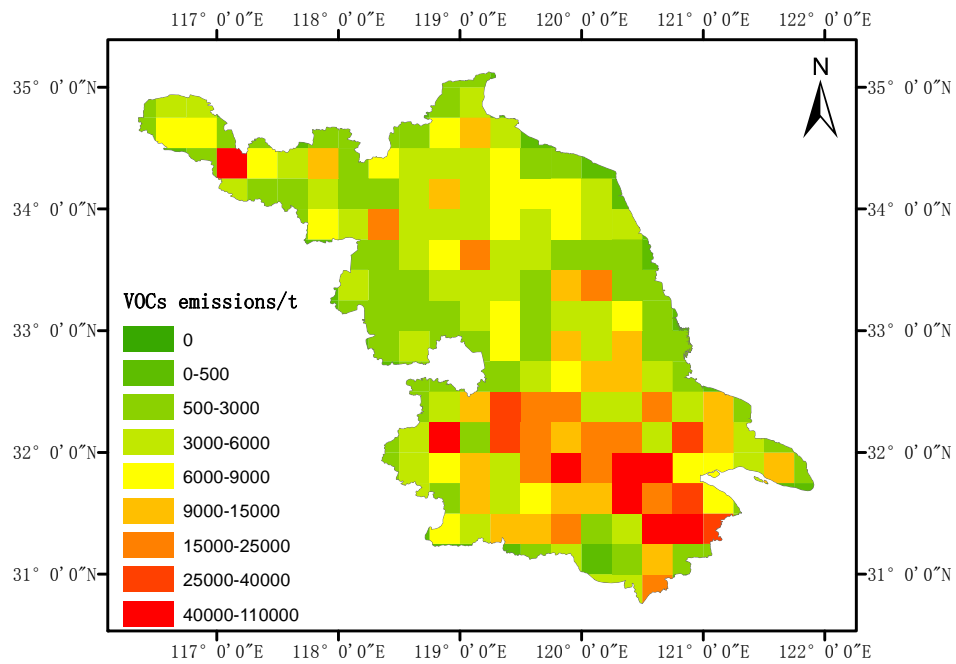


Figure 9



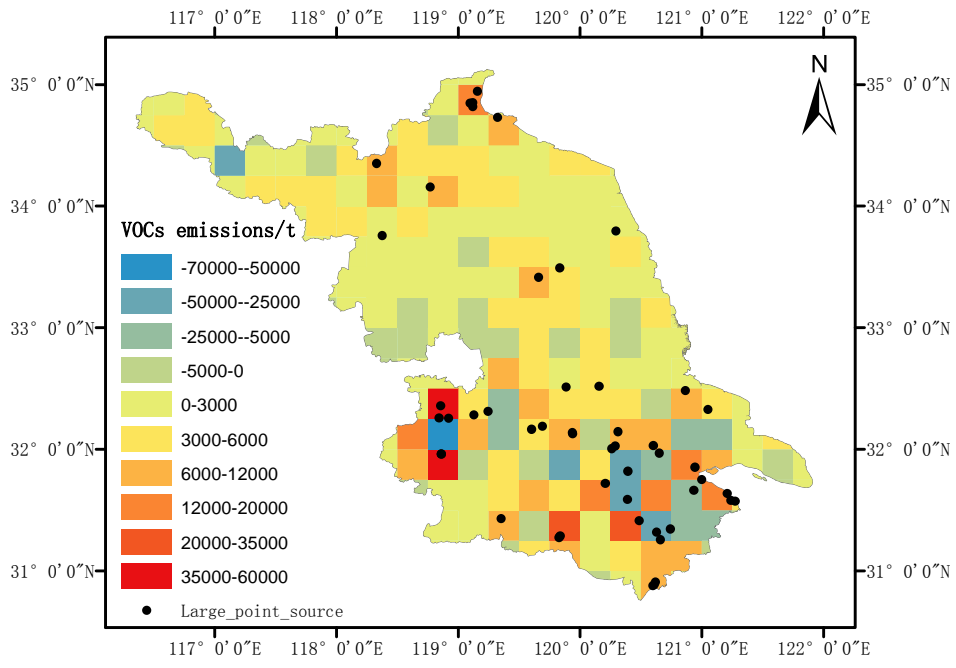
(a)



(b)

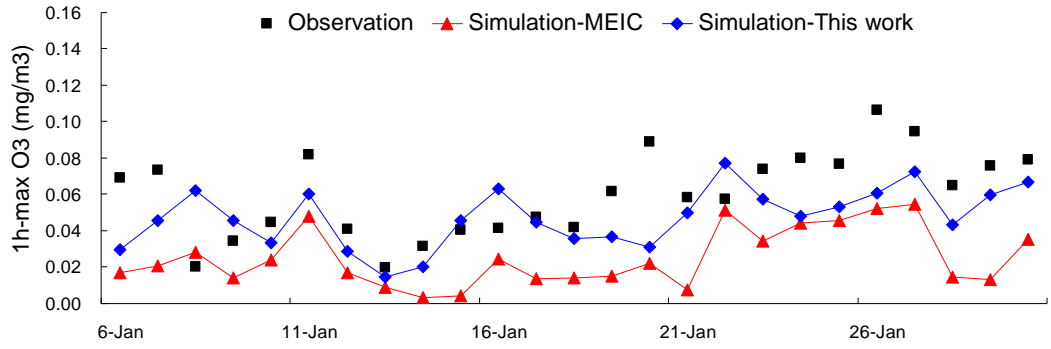


Figure 9 (continued)

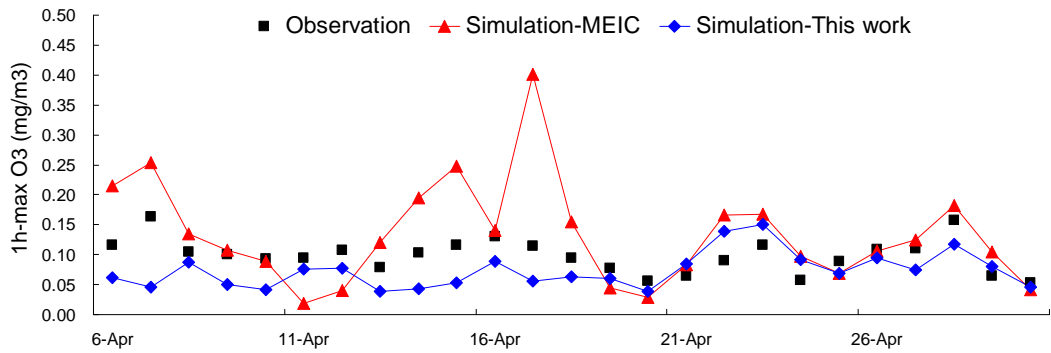


(c)

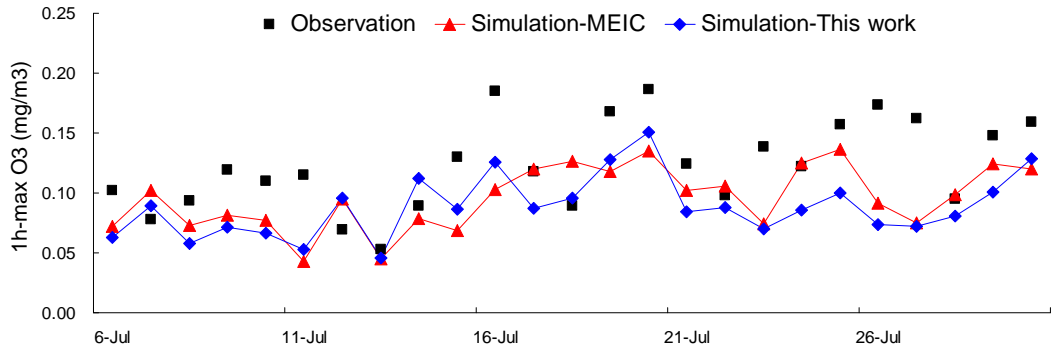
Figure 10



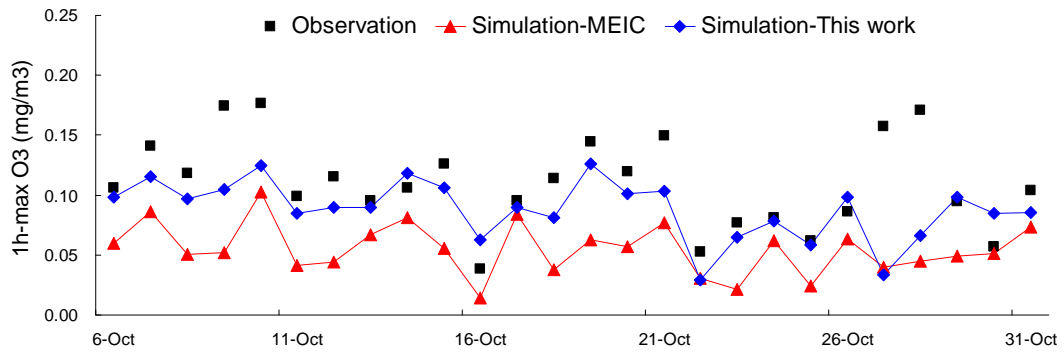
(a)



(b)



(c)



(d)