Main revisions and response to reviewers' comments

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

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We thank very much for the valuable comments and suggestions from the two reviewers, which help us improve our manuscript significantly. The comments were carefully considered and revisions have been made in response to suggestions. Following is our point-by-point responses to the comments and corresponding revisions.

Reviewer #1

0. *This paper describes the development and updates of a bottom-up regional anthropogenic emissions inventory for the Jiangsu province in China. Source profiles of industrial facilities were improved compared to previous inventories through measurements at the facility level of NMVOCs from canister samples. Differences to other inventories are discussed and regional CMAQ modeling studies with the various inventories were performed and it was found that using the current inventory improves, albeit still underestimates, ozone predictions. Improving anthropogenic emission inventories is the main pathway to improving air quality and climate modeling and forecasting, especially in areas such as China, which makes this paper a very valuable contribution. Unfortunately, the paper is difficult to read because of necessary information hidden in the SI, important data sources missing from the* *description and generally inconsistent description of the different source types and as a result this paper needs major changes, before it is acceptable.*

Response and revisions:

We appreciate the reviewer’s positive remarks on the contribution of the work. Regarding the weakness pointed out by the reviewer, we have improved the manuscript accordingly. Part of the information in the original supplement has been revised and moved to the main text (please see details in our response to Q1), and data sources of emission inventory development have been clearly described in order as suggested by the reviewer (please see details in our response to Q3 and Q4). The description of different source types has been revised to avoid confusion and inconsistency. Please see the details in the following response and revision list to reviewer’s comment.

1. *My main issue with this paper is that the description of the inventory development in chapters 2.2 and 2.3 is pretty unclear. First of all, the essential information of the source types is in the SI and not in the main text. A paper needs to be understandable even without reading the SI in detail and that is certainly not the case here. Tables S1 and S3 should be combined into one and moved to the main text. I also think that Tables S4, S5 and S6 should be combined and moved to the main text. Then the source types and the data sources for each type need to be discussed in more detail.*

Response and revisions:

We thank and agree with the reviewer’s comment. As suggested by the reviewer, we have moved Table S3 in the original supplement to the revised main manuscript (Table 1). The data sources of each type have been discussed in order in a new Section 2.3 in the revised manuscript.

We have also merged Tables S4-S6 in the original supplement and moved the new table to the revised main manuscript (Table 2). Table S1 summarized the national emission estimates for the country by various studies, while the work focuses mainly on the provincial inventory. Therefore we kept Table S1 in the revised supplement.

2. *I would like to see a discussion on why these source types were chosen. They are the same as in previous inventories, but is this a good choice? Can there improvements be made at this level already? Next it should be discussed what source types are most important, making big changes to a very small source type, is not going to change the total emissions significantly. Besides, emissions in China and Jiangsu in particular are dominated by solvent use and industrial processes, which is not the case in other regions and this needs to be pointed out early.*

Response and revisions:

We thank the reviewer's comment. The framework of national and regional emission inventories was established based on China's energy and economic statistics. In this work, the framework for Jiangsu provincial inventory was updated and revised based on the plant-by-plant information from various data sources (including Environmental Statistics, Pollution Source Census, and on-site surveys as we stated in the text). Therefore the source categories were not "chosen" but were included in a framework that covered all the existing industrial processes for the provinces. We admit that there would be omission attributed to data limitation, but the omitted was supplemented as area sources. We have added the discussion **in lines 177-190 in the revised manuscript**.

For the importance of source types, we expected there were discrepancies in various provinces, and it could not be clearly explored unless detailed source information was incorporated. Therefore, we discussed the issue for Jiangsu in Section 3.2 in the revised manuscript where the emissions by source type were calculated and provided. At current stage, it is difficult to make a thorough emission comparison between studies, because there were not so many national and regional inventories which provided emissions and data sources at such a detailed source category level. We obtained available information and compared the results at coarser sector level **in lines 674-693 in the revised manuscript**. We also agree with the reviewer that emissions in China and Jiangsu in particular are dominated by solvent use and industrial processes, and we have stated the case early **in lines 64-66 in the revised manuscript**.

3. *What is completely missing is a description of the data sources for the activity level data and a comparison to the previous emission inventories. It is not clear to me what was taken from the literature and what is really new here from this work in terms of activity data. These are essential for building this inventory and some of it is hinted on in Figure S3, but needs to be discussed here.*

Response and revisions:

We thank the reviewer's comment. At the beginning of Section 2.2 **(lines 177-188 in the revised manuscript)**, we first stressed the improvement in activity data compared to previous national and regional inventories: the inclusion of detailed plant-by-plant information through thorough investigations on available databases (i.e., the accuracy of activity data for point sources were improved), more local information on traffic fleet, and the improved framework with more detailed classification of emission source categories. The subsequent changes in emission estimation and spatial distribution were then discussed in Section 4.1 and 4.3. We followed the reviewer's suggestion and provided detailed description on the data sources of activity levels by sector in the revised Section 2.3. However, we should admit that the methods of activity data collection from previous studies had also to be applied for certain sources (e.g., solvent use and oil distribution), as the whole framework of emission inventory was comprehensive and detailed activity data were currently unavailable for all the source categories at provincial (or smaller) scale.

4. *The discussion of the inventory development would benefit from re-organizing. I suggest discussing each source type in order and include activity factor, emission factor, speciation, source profile and uncertainty. As is, in chapters 2.2 to 2.5 the source types are covered very inconsistent, which means that a lot of information is missing. All of this means some major re-organization of the first half of the paper, but without it cannot be judged what is new and important from this work. The rest of the paper was often difficult to follow, because it was not clear what all was included in the source type analysis.*

Response and revisions:

We thank the reviewer's comment. Following the reviewer's suggestion, the section of inventory development has been re-organized. In the revised Section 2.2, the principle of emission inventory development was described including the methods of emission calculation, source profile estimation, and uncertainty analysis. In particular, at the beginning of Section 2.2 we have stressed the improvement in data sources compared to previous national and regional inventories. Original Sections 2.3-2.5 have been deleted in the revised manuscript. Instead, a new Section 2.3 has been added, which in order discussed the detailed methods and data sources of activity level estimation, data sources of emission factors and chemical profiles, and the probability distribution functions (PDFs) by source category. We have also revised and updated the Tables 1 (Table S3 in the original supplement), 2 (Tables S4-6 in the original supplement), and S4 (Table S8 in the original supplement) to eliminate the inconsistency in source category description.

We need also to clarify a case in Figure 8, the comparison of sector emissions between different inventories. The sector classification for comparison between our work and Zhang et al. (2009)/MEIC was indeed inconsistent with other, because the latter two studies used a different classification. For direct comparison, therefore, we had to regroup the emissions of our work to be consistent with them in source categories. We have stated this in the revised caption of Figure 8.

5. *Page 6 line 125: Why did you choose these specific facilities? Were there no data on them?*

Response and revisions:

We thank the reviewer's important comment. Currently the chemical profiles of NMVOC emissions are still lacking for many source categories of chemical and refinery industry in China. The source types we selected for measurements were intensively distributed in Jiangsu province, and no domestic measurement has been conducted yet for those sources to our knowledge. Even in SPECIATE the chemical profiles were available for three processes, i.e., synthetic rubber, ethylene and polyethylene production. To improve the completeness of local source profiles of chemical industry, therefore, we chose those facilities for measurement. We have discussed this **in lines 126-134 in the revised manuscript.**

6. *Page 7 lines 146-150: Please explain why all these different stages were necessary for the GC analysis.*

Response and revisions:

We thank the reviewer's comment. In the first stage, vapour water was converted to solid water and was thus separated from sample. In the second stage the target species were separated from CO_2 and other compositions in the air. The sample with target species was then concentrated in the third stage for GC analysis. We have added the explanation **in lines 154-159 in the revised manuscript.**

7. *Page 9 line 207-212: The priorities for the emission factor determination need more detail. Are the emission limits for laws and regulations strictly enforced? If so, they can be used as an upper limit. If not, experience from other regions shows that* *these regulations rarely have much correlation with actual emissions. Also, what do you mean with expert judgment? Are those literature values or estimates from industry officials? Please explain!*

Response and revisions:

We thank and agree the reviewer's comment. We (and previous studies) had to rely on emission limits of relevant regulations to estimate emissions for solvent use due mainly to lack of domestic test results. We have indicated **in lines 324-327 in the revised manuscript** that the regulations were not strictly enforced particularly for small solvent use enterprises and construction sites (area sources). Therefore, bias needs to be admitted here and possible underestimation in emissions would be expected for the sector. We have also clarified **in lines 294-295 in the revised manuscript** that emission factors from "expert judgment" indicated the data from routine investigations reported by the factory officials to local environmental protection bureaus.

8. *Page 12 line 303: How much of the total emissions were covered with these measurements? Do they represent and help update much of the emissions? The source profile update in Figure 6 seems significant only for a few species?*

Response and revisions:

We thank the reviewer's important comment. The source types we measured accounted for 9-11% of annual NMOVC emissions from chemical and refinery categories (Oil exploitation and refinery, chemical raw materials, synthetic chemical industry, and fine chemical industry in Table 1) for Jiangsu 2005-2014. Attributed to a wide variety of source categories for chemical and refinery industry, the sources we measured did not dominate the emissions of the sector. However we mean the measurements improved the inventory (particularly for source profile) as there was no domestic measurement result on chemical composition published before on those sources, and it was the first time that local information was incorporated in the emission inventory development for those sources. In particular the contribution of the sources to emissions was enhanced in typical cities with intensive chemical industry. In Nanjing (the capital city of the province), for example, the source categories we measured accounted for 19% of annual emissions from chemical and refinery industry, and for 10% of the total anthropogenic emissions in 2011. Better information on chemical compositions of NMVOC emissions was expected to be available when the local measurements on source profile were conducted and incorporated for the city. Such information, however, would not be available until the entire emission inventory was developed. Therefore we added the information **in lines 638-643 in the revised manuscript,** after Section 3.2 that describes the NMVOC emissions by source type.

Regarding Figure 6, as we described **in lines 635-638 in the revised manuscript,** both our measurements and other domestic studies after 2010 were incorporated for source profile updating. We have stated that relatively big changes in emissions (over 10 Gg) were found for certain compositions including ethylacetate and aromatics species (benzene, xylene, ethyl benzene, and methyl benzene). To clarify the impact, we further calculated the total difference between emissions of all species before and after updating, and it reached 281 Gg, i.e., 13% of the total emissions for the province in 2010. We have added the discussion **in lines 646-648 in the revised manuscript.**

9. *Figure 6: The units in this Figure cannot be correct.*

Response and revisions:

We thank the reviewer’s reminder and the error has been corrected in the revised Figure 6.

10. *Page 14 line 353: Can you indicate how much the measurements changed the inventory? Were they an essential improvement compared to the previous inventories?*

Response and revisions:

We thank the reviewer's comment. As the measurements targeted on the source profile (i.e., the fractions of chemical species), incorporation of the results could hardly lead to changes in total NMVOC emission estimation. However, as we responded to Q8, the source types we measured accounted for 9-11% of NMVOC emissions from chemical and refinery categories in the province, and the fraction could be even larger in typical city. Better information on NMVOC emission speciation could thus be available when the measurement results were applied for source profile updating compared to previous inventories. The improvement could also be evaluated through air quality modeling for certain species (e.g., ozone) as we presented in Section 4.4, although further analysis is still needed (please also see our response to Q15).

We also need to acknowledge the big variety of source categories for chemical and refinery industry in China. At current stage measurements on chemical sources are still lacking for many source types. Although incremental contribution could be expected through current work, more efforts on field measurments from different research groups are still needed in order to obtain a more complete database of chemical profiles for China's chemical and refinery industry. We have added the discussion **in lines 655-661 in the revised manuscript.**

11. *Page 15 line 382: I assume the units are g/kg?*

Response and revisions:

We thank the reviewer's reminder and the error has been corrected in the revised manuscript.

12. *Page 15 lines 396-405: Can you explain what "certain proxies" means?*

Response and revisions:

"Certain proxies" indicate the parameters used for allocating the emissions from sources other than point sources, including GDP, population, road net and traffic flow, and railway and canal net. We revised the sentence **in lines 468-472 in the revised manuscript** to make it clearer:

"For other sources, certain proxies were applied to allocate emissions, including GDP for industrial area sources and oil distribution, population for solve use area sources, road net and traffic flow for on-road vehicles, railway and canal net for off-road transportation, and rural population for biomass burning."

13. *Page 17 line 437-442: Please explain why the OFP to emissions ratio is important.*

Response and revisions:

OFP is used for evaluating the capability of ozone formation through atmospheric chemical reactions for individual VOC species. As the chemical profiles of emitted VOC vary between source categories, the OFP to emission ratio for a given sector (or source type) indicates the potential contribution to ozone formation for the sector (or source type), as a combined effect of multiple NMVOC species emitted from it. The ratio could thus provide scientific suggestion of emission control for policy makers, e.g., the emission control needs to be preferentially considered for sectors with large OFP/emissions. We have explained the issue **in lines 513-517 in the revised manuscript.**

14. *Page 17 lines 444-448: You need to provide the information about the most important species in the main text as well.*

Response and revisions:

We thank the reviewer's reminder. **In lines 526-527 in the revised manuscript** we have added the information as required: "Xylene, ethylene, and propylene were identified as the most three important species in terms of OFP."

15. *Page 17 lines 450-458: What are the most important species in the modeling shown later in the manuscript?*

Response and revisions:

We thank the reviewer's important comment. **In lines 750-752 in the revised manuscript,** we have indicated that the updated provincial inventory provided larger emission estimates for certain species with relatively high ozone formation potential including ethene and ethanol. Such revisions were expected to improve the ozone simulation with CMAQ as we described in Section 4.4. We should admit, however, that the impacts of emission changes for individual species on ozone simulation could not be completely confirmed in current work. The improved ozone simulation presented here was a combined effect of an updated inventory with revisions on emission estimation, spatial distribution and source profiles for all relevant species. A much more detailed chemistry transport modeling study with intensive sensitivity analysis is needed in order to further figure out the impacts of individual species (as well as factors other than emission input). We mean it is beyond the scope of the current work, and we will keep conducting the relevant analysis in the future. We have discussed this **in lines 752-756 in the revised manuscript.**

16. *Page 17 lines 460-470: The text does not provide enough information on how the uncertainties are estimated, because of the many things that are not included in the inventory development section. It is therefore difficult to judge, if the emission factors are indeed the largest uncertainty. Can you indicate in this chapter, what would be the most helpful for improving the uncertainties. Would actual emissions measurements be critical?*

Response and revisions:

We thank and agree the reviewer's comment. As we respond to Q4 of the reviewer, we have reorganized the inventory development section carefully, and more detailed description on uncertainty analysis has been added into the revised manuscript. We have briefly described the principles of uncertainty quantification **in lines 235-242, Section 2.2**, and we have added the detailed methods and data sources of determination of probability distribution function (PDF) for emission factors and activity levels by source category in Section 2.3. **In lines 273-277 in the revised manuscript,** we have admitted that uncertainty of emission factor was evaluated depending on expert judgment due to insufficient data support from local measurments, and that PDF of emission factor was given according to reliability of data sources and/or the robustness of calculation methods following the rules of previous work (Streets et al., 2003; Wei et al., 2011). As suggested by the reviewer, we have indicated **in lines 552-554, Section 3.4 of the revised manuscript** that actual emissions measurements would be helpful for reducing the uncertainty, since expanded data samples would better support the determination of emission factors through data fitting instead of conservative assumption.

17. *Page 19: The omission of for example basic chemistry production in the activity data seems to be a clear indicator that the current emission inventory is more likely a lower limit of the emissions, because some of the activity is just not captured.*

Response and revisions:

We thank the reviewer's comment. As we stated, the basic chemistry production was not captured in economic statistics, upon which the Method 3 was based. Method 3 was actually applied by most previous national and regional inventories attributed mainly to lack of detailed emission source data, and underestimation in emissions can be expected as judged by the reviewer. In this work, Method 1 that incorporated the most available information from Environmental Statistics, Pollution Source Census, and on-site surveys was applied, thus the errors from activity data omission for certain sources were corrected. We mean the comparisons between different methods just highlighted the improved estimation in emissions by this work. **In lines 578-581 in the revised manuscript**, we stressed that Method 1 was applied in this work to provide the best available emission information. **In lines 598-600 in the revised manuscript**, we also admitted that even Method 1 could lead to emission underestimation as it is difficult to cover all the process in emission factor determination, particularly for the fugitive release.

18. *Page 19 line 520: What is your indication that the emissions in downtown are actually overestimated? It is clear that this method has a large uncertainty, but this does not necessarily mean an overestimation. Is this because the large point source emitters are not located in downtown and you include those in the downscaling method?*

Response and revisions:

We thank and agree the reviewer's comment. We mean the uncertainty came mainly from the method of spatial allocation of emissions. Attributed to lack of detailed information of emission source locations, Method 3 in general allocated the emissions based on the density of economy and population, and it would thus lead to larger emission estimation in downtown area with relatively high density of economic activities and population, compared to suburban area. In this case of Nanjing, however, the large point sources (e.g., chemical industry and refinery plants) were not located in downtown, and discrepancy between the estimation and actual emission distribution would occur. As the total emissions in Method 3 could be underestimated due to omission of certain source categories, we agree with the reviewer that the absolute emission levels might not be necessarily overestimated, and we revised the words as "would overestimate the fraction of emissions in urban downtown" **in line 607 in the revised manuscript**.

19. *Page 21 line 556 (and elsewhere): Thylacetate is really not a very common chemical and I am surprised about the large atmospheric emissions. I think a discussion is warranted on what this compound is and why it is produced in such large quantities. I am wondering, if you meant ethyl acetate, which is pretty common?*

Response and revisions:

We thank the reviewer's reminder and admitted the typo error. Yes it should be ethyl acetate, and we have corrected it in the revised manuscript.

20. *Page 21 line 562-571: Overall the changes are not very large. What are the changes in OFP and total emissions and how much influence does the update have on ozone modeling?*

Response and revisions:

We thank the reviewer's reminder. The relative changes actually varied among species and could be big for certain species. Please see the detailed data in Table R1. The total NMVOC emissions estimated in this work was 315 Gg (i.e., 18%) larger than MEIC for 2010. We have added the information **in lines 695-697 in the revised manuscript**. Unfortunately the OFP for MEIC could not be calculated as the original information of chemical species was unavailable. The influence of updated emission inventory on ozone modeling was described in Section 4.4. As we responded to Q15, current work could not totally disentangle the effects of source profile updating and other changes in emission inventory (e.g., total amount and spatial distribution of emissions), and we will continue the research in the future.

Table R1 Emissions of CB05 species for Jiangsu 2010

Inventory	Source	Emission (10^9 mol)													
		PAR	OLE	TOL	XYL	FORM	ALD2	ETH	MEOH	ETOH	ETHA	IOLE	ALDX	UNR	NVOL
Before updating	Fossil fuel combustion	1.00	0.08	0.06	0.10	0.00	0.00	0.17	0.00	0.00	0.15	0.02	0.00	0.35	0.00
	Industrial process	17.19	0.93	0.73	0.06	0.01	0.03	1.52	0.20	0.48	0.96	0.08	0.01	5.54	0.00
	Transportation	10.84	0.53	0.21	0.25	0.28	0.26	0.60	0.00	0.00	0.13	0.21	0.18	0.78	0.00
	Solvent use	12.89	0.18	1.17	0.35	0.06	0.01	0.03	0.02	0.03	0.02	0.06	0.00	2.18	0.00
	Oil distribution	2.75	0.02	0.01	0.00	0.02	0.02	0.00	0.00	0.00	0.02	0.04	0.00	0.11	0.00
	Biomass burning	1.69	0.24	0.02	0.12	0.31	0.14	0.40	0.31	0.00	0.18	0.02	0.09	0.81	0.00
	Other	1.34	0.13	0.00	0.00	0.29	0.34	0.13	0.00	0.00	0.10	0.00	0.42	0.24	0.00
	Total	47.70	2.10	2.20	0.88	0.97	0.81	2.84	0.54	0.52	1.56	0.44	0.69	10.01	0.01
After updating	Fossil fuel combustion	1.00	0.08	0.06	0.10	0.00	0.00	0.17	0.00	0.00	0.15	0.02	0.00	0.35	0.00
	Industrial process	18.42	0.92	0.55	0.14	0.04	0.03	1.28	0.18	0.49	1.05	0.12	0.12	4.59	0.00
	Transportation	11.03	0.48	0.21	0.27	0.38	0.17	0.60	0.00	0.00	0.12	0.13	0.26	0.91	0.00
	Solvent use	14.95	0.20	1.01	0.39	0.05	0.01	0.21	0.01	0.04	0.15	0.07	0.59	2.07	0.00
	Oil distribution	2.75	0.02	0.01	0.00	0.02	0.02	0.00	0.00	0.00	0.02	0.04	0.00	0.11	0.00
	Biomass burning	1.79	0.23	0.03	0.12	0.23	0.14	0.37	0.23	0.00	0.18	0.02	0.18	0.77	0.00
	Other	1.34	0.13	0.00	0.00	0.29	0.34	0.13	0.00	0.00	0.10	0.00	0.42	0.24	0.00
	Total	51.29	2.06	1.87	1.05	1.00	0.72	2.75	0.42	0.54	1.77	0.40	1.57	9.04	0.01
MEIC	-	47.31	2.08	3.49	1.85	1.10	0.49	1.99	0.48	0.30	1.12	0.55	1.24	9.25	0.74
Difference between after updating and MEIC（relative to MEIC）		8.4%	-1.2%	-46.4%	-43.3%	-8.6%	47.7%	38.4%	-12.1%	77.1%	58.7%	-27.5%	27.4%	-2.3%	-99.3%

21. *Page 21 line 576: The qualification of the REAS inventory as extremely high should be removed. "Extremely high" is a very subjective term and should not be used. Please be quantitative. Besides, it seems to me that this bottom up inventory provides more likely a lower limit for the emission, because there is a large potential for processes not being included. Also the modeling shows that ozone is still under-predicted, which also points to low emissions in this inventory.*

Response and revisions:

We thank and agree the reviewer's comment. We have revised the text with quantitative comparison "Except for REAS that provided 37%-77% higher emissions than this work for 2005-2008" **in line 677 in the revised manuscript**. As we response to Q17, the errors from activity data omission for certain sources were corrected in the updated provincial inventory in this work. Nevertheless, we agree with the reviewer that the under-predicted ozone still implied the possible underestimation in VOC emissions. We have stressed this **in lines 743-746 in the revised manuscript**.

22. *Page 22 line 581: similar type of comment: "much larger emissions" is a very subjective term, so please be quantitative. I actually also disagree with the statement, the inventories are pretty close to each other, clearly within the stated uncertainties.*

Response and revisions:

We thank and agree the reviewer's comment. Subject term has been deleted and quantitative information has been provided as "the emissions in this work were 4% and 20% larger than the national inventory for 2005 (Wei et al., 2008) and regional inventory for 2010 (Fu et al., 2013), respectively" **in lines 682-684 in the revised manuscript**.

23. *Chapter 4.4: I think the modeling shows that the inventory is still likely an underestimation of the actual emissions and I think this should be pointed out in the text.*

Response and revisions:

We thank and agree the reviewer's comment. **In lines 743-746 in the revised manuscript**, we have stated "the updated anthropogenic NMVOC emission inventory at provincial scale was still likely an underestimation of the actual emissions".

24. *Page 24 line 639: Please give a reference for the statement about VOC-limited.*

Response and revisions:

We thank the reviewer's reminder and a reference (Xing et al., 2011) has been given in the revised manuscript.

25. *Technical Comments*

Response and revisions:

We thank the reviewer's reminder and all the corrections have been made as suggested by the reviewer.

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Reviewer #2

0. *The paper describes a bottom-up development and evaluation of a highly-resolved regional emission inventory for NMHCs in the area of Jiangsu, a strong industrialized region in Eastern China. The reference period is almost 10 years. The quantification of chemical processes is based on the determination of realistic source profiles for industrial activities by near-source measurements. The authors provide an extensive work. The paper is divided into 4 parts: first part describes the inventory methodology, second part compares the newly released emission inventory to other downscaled emission inventories regarding absolute emissions and speciated emissions; third part uses the CMAQ model to test the ability of the model to reproduce hourly maximum ozone concentrations with the new emission inventory. Evaluation of emission inventories is important for improving the simulation and forecast of air quality and climate and is unfortunately often neglected.*

Response and revisions:

We appreciate the reviewer's remarks on the importance of the work.

1. *While the science is of relevant atmospheric interest, I have major concerns about the paper: the paper is not easy to read and the reader gets easily lost. For instance, the authors often go back and forth with figures and associated discussion (i.e., Figure 7). In several parts or sections, the paper relies on information reported in the Supplement Material which often makes the paper hard to follow, especially regarding the development of the emission inventory.*

Response and revisions:

We thank the reviewer's comment. In the revised manuscript, we moved important information in the original supplement to the main text, including Table S3 that illustrated the framework of emission inventory development and classification of emission source categories (Table 1 in the revised manuscript), and Table S4-S6 that summarized the emission factors by source category (a merged Table 2 in the revised manuscript). The section of emission inventory development has been re-organized. The revised Section 2.2 described the principles of emission inventory development including the methods of emission calculation, source profile estimation, and uncertainty analysis. In particular, the general improvement in data sources compared to previous national and regional inventories was stressed at the beginning of the section. Original Sections 2.3-2.5 have been deleted in the revised manuscript. Instead, a new Section 2.3 has been added, which in order discussed the detailed methods and data sources of activity level estimation, data sources of emission factors and chemical profiles, and the probability distribution functions (PDFs) by source category. We have also moved the original Table 1 to Section 3.3 (Table 4 in the revised manuscript) as it actually described the results of chemical profiles by source category. We expect such revisions make the text clearer and easier to read. To be concise, Figure 7 illustrated the distributions of chemical species under different mechanisms both in this work and in other inventories, thus we had to discuss the figure in the speciation section (Section 4.2) and comparison section (Section 4.3). We have revised the text **in lines 665-666 in the revised manuscript**, and hopefully it would help to clarify the case.

2. *The sampling and analysis strategy in the field is not described and motivated. Line 103: the choice was put on the speciation of chemical industries. The sampling strategy and the representativeness of emission measurements should be detailed.*

Response and revisions:

We thank the reviewer's important comment. Currently the chemical profiles of NMVOC emissions are still lacking for many source categories of chemical and refinery industry in China. The source types we selected for measurements were intensively distributed in Jiangsu province, and no domestic measurement has been conducted yet for those sources to our knowledge. Even in SPECIATE the chemical profiles were available for three processes, i.e., synthetic rubber, ethylene and polyethylene production. To improve the completeness of local source profiles of chemical industry, therefore, we chose those source categories for measurement. We have discussed this **in lines 126-134 in the revised manuscript.**

3. *Some sections do not provide reliable information. Lines 514-525: The authors compare the spatial distribution of emissions from industrial activities from three different methods including one without any information on individual plants, which uses proxies like population density. They show that the spatial allocation from this method is wrong and not representative of local characteristics. One could wonder whether such result could have been predictable.*

Response and revisions:

We thank the reviewer's comment. The motivation of the comparison was to reveal the discrepancies in emission estimation and spatial distribution between inventories with different data sources. In actual, the method with little information on individual plants, as mentioned by the reviewer, was commonly applied in national and regional inventories at larger spatial scales, and emission downscaling was generally adopted when high-resolution inventory was needed for air quality simulation or other purposes. We have pointed this **in line 583 in the revised manuscript.** Through the comparison conducted in this work, we illustrated that such method could lead to big uncertainty in allocation of emissions and thereby air quality simulation, at least for cities like Nanjing whose emissions were dominated by big industrial plants. Therefore we mean the work highlighted the necessity of careful investigation on individual emission sources when the accuracy of local inventory became a big concern for both scientific community and China's policy makers of air pollution control.

4. *The interpretation of the figures are incomplete or approximate. Regarding the various emission inventory evaluation, it looks like the improvement with the update is not so clear in term of absolute concentration, reactivity and spatialization.*

Response and revisions:

We thank the reviewer's comment. In the revised manuscript, we have provided more quantitative information in the interpretation of figures, particularly for Figures 4, 6, 7, and 8. The differences in emission estimation and chemical profiles between various inventories have been stressed with detailed data support. Subject terms have also been deleted to avoid confusion. Please check the revisions **in lines 646-661, 676-684, and 695-700 in the revised manuscript.**

5. *Lines 476-486: the discussion on uncertainty comparison should be revised or at least clarified. Differences in uncertainties between inventories in Table 3 could be also due to the way uncertainties are estimated or the spatial resolution. Indeed the authors give the impression that the uncertainty of the new released inventory is better. It might be for the wrong reason.*

Response and revisions:

We thank and agree the reviewer's comment. Among all the studies we include for comparison, Huang et al. (2011) applied a different method to calculate the uncertainty other than Monte-Carlo simulation, and it would lead to difference in uncertainty estimation. We have added the discussion **in lines 559-564 in the revised manuscript**. Regarding the spatial scale, as current study focused only on the total emissions, the discrepancies in resolution (and thereby the uncertainty in spatial distribution of emissions) were not covered here. We agree with the reviewer that uncertainties could differ for inventories at different spatial scales. We expect such difference resulted mainly from the various levels of details for emission source information, as we have discussed in the section.

6. *Line 551-561: Comparing the updated speciation of VOCs to the SPECIATE emission profiles is relevant. It is surprising to see that the updated VOC speciation profiles are not so different from the foreign SPECIATE database excepted aromatics and ethylacetate. It would be also relevant and interesting to compare database by only considering the measured profiles.*

Response and revisions:

We thank the reviewer's comment. We admit there was a confusion in the original text. The source profile before updating was not directly taken from SPECIATE but a combination of Li et al. (2014) and SPECIATE. Li et al. (2014) made a comprehensive source profile for China based on domestic measurement results published before 2010. As a more detailed source classification was used in this work, some sources were not covered by Li et al. (2014) and thus the results from SPECIATE were applied. In this work, as we indicated in Sections 2.2 and 2.3 of the revised manuscript, the domestic measurements after 2010 and the local measurements by us were incorporated to update the source profile. As the newly-added measurements were not sufficient enough, not very big differences were found between source profiles between and after updating except for aromatics and ethylacetate. We have clarified this **in lines 211-224 in the revised manuscript.**

Among the sources we measured, the chemical profiles of synthetic rubber, ethylene and polyethylene production were available in SPECIATE. As suggested by the reviewer, therefore, we have compared the profiles between our results and SPECIATE for the three source categories, as shown in Figure 1b-d. Relevant discussions have been provided **in lines 406-423 in the revised manuscript.**

7. *Figure 4: the comparison with other emission inventories for the same spatial domain reveals a quite good consistency regarding absolute values and trends. As stated by the authors all the results are within the 95% confidence limits. From this* *figure, it seems that differences are not statistically different. However the authors keep insisting on such differences.*

Response and revisions:

We thank and agree with the reviewer's comment. We have deleted the subject term and conducted quantitative comparisons in total emissions between different inventories **in lines 676-684 in the revised manuscript.** We admit that the differences between our study and most other inventories were not large except for REAS. As shown in Figure 8, however, emissions of different sectors varied between studies attributed to the various data sources and calculation methods. Therefore, we kept the analysis on the different emissions by sector, and tried to reveal the effects of data sources on NMVOC emission estimation for typical source categories.

8. *Figure 7: The difference between speciation is not so significant after updating and finally raises the question of the usefulness of the updating except for aldehydes. This deserves some discussion.*

Response and revisions:

We thank the reviewer's comment. The relative changes actually varied among species and could be big for certain species. Please see the detailed data in the Table R2. For example, the updated provincial inventory provided larger emission estimates for ethene and ethanol with relatively high ozone formation. We have added the information **in lines 695-700 in the revised manuscript.** As we indicated in the section 4.4 (CMAQ evaluation), the updated speciation of NMVOC emissions were expected to improve the ozone simulation with chemistry transport modeling. However, we also admitted that the improved ozone simulation was a combined effects of an updated inventory with revisions on total emission estimation, spatial distribution and source profiles for all relevant species. Current work could not totally disentangle the effects of source profile updating and other changes in emission inventory. A detailed chemistry transport modeling study with sensitivity analysis is needed in order to further figure out the impacts of individual species, and we will keep conducting the relevant analysis in the future. We have discussed this **in lines 752-756 in the revised manuscript.**

Table R2 Emissions of CB05 species for Jiangsu 2010

Inventory	Source	Emission (10^9 mol)													
		PAR	OLE	TOL	XYL	FORM	ALD2	ETH	MEOH	ETOH	ETHA	IOLE	ALDX	UNR	NVOL
Before updating	Fossil fuel combustion	1.00	0.08	0.06	0.10	0.00	0.00	0.17	0.00	0.00	0.15	0.02	0.00	0.35	0.00
	Industrial process	17.19	0.93	0.73	0.06	0.01	0.03	1.52	0.20	0.48	0.96	0.08	0.01	5.54	0.00
	Transportation	10.84	0.53	0.21	0.25	0.28	0.26	0.60	0.00	0.00	0.13	0.21	0.18	0.78	0.00
	Solvent use	12.89	0.18	1.17	0.35	0.06	0.01	0.03	0.02	0.03	0.02	0.06	0.00	2.18	0.00
	Oil distribution	2.75	0.02	0.01	0.00	0.02	0.02	0.00	0.00	0.00	0.02	0.04	0.00	0.11	0.00
	Biomass burning	1.69	0.24	0.02	0.12	0.31	0.14	0.40	0.31	0.00	0.18	0.02	0.09	0.81	0.00
	Other	1.34	0.13	0.00	0.00	0.29	0.34	0.13	0.00	0.00	0.10	0.00	0.42	0.24	0.00
	Total	47.70	2.10	2.20	0.88	0.97	0.81	2.84	0.54	0.52	1.56	0.44	0.69	10.01	0.01
After updating	Fossil fuel combustion	1.00	0.08	0.06	0.10	0.00	0.00	0.17	0.00	0.00	0.15	0.02	0.00	0.35	0.00
	Industrial process	18.42	0.92	0.55	0.14	0.04	0.03	1.28	0.18	0.49	1.05	0.12	0.12	4.59	0.00
	Transportation	11.03	0.48	0.21	0.27	0.38	0.17	0.60	0.00	0.00	0.12	0.13	0.26	0.91	0.00
	Solvent use	14.95	0.20	1.01	0.39	0.05	0.01	0.21	0.01	0.04	0.15	0.07	0.59	2.07	0.00
	Oil distribution	2.75	0.02	0.01	0.00	0.02	0.02	0.00	0.00	0.00	0.02	0.04	0.00	0.11	0.00
	Biomass burning	1.79	0.23	0.03	0.12	0.23	0.14	0.37	0.23	0.00	0.18	0.02	0.18	0.77	0.00
	Other	1.34	0.13	0.00	0.00	0.29	0.34	0.13	0.00	0.00	0.10	0.00	0.42	0.24	0.00
	Total	51.29	2.06	1.87	1.05	1.00	0.72	2.75	0.42	0.54	1.77	0.40	1.57	9.04	0.01
MEIC	-	47.31	2.08	3.49	1.85	1.10	0.49	1.99	0.48	0.30	1.12	0.55	1.24	9.25	0.74
Difference between after updating and MEIC (relative to MEIC)		8.4%	-1.2%	-46.4%	-43.3%	-8.6%	47.7%	38.4%	-12.1%	77.1%	58.7%	-27.5%	27.4%	-2.3%	-99.3%

9. *The simulation with the CMAQ model. The model outputs not only depend on the representation of emissions but also on the representation of chemistry and dynamics. The authors should take these two drivers as well to explain potential differences with the observed ozone-hourly maximum. Note that we also see some differences between seasons. Emissions might not be the only limitation. The get free from dilution effects or chemistry effects the authors could rather use ratios.*

Response and revisions:

We thank and agree the reviewer's important comment. Since the current work focused mainly on the emission inventory revision and its subsequent impacts on air quality modeling, the model configurations were same on parameters other than emission input, including the chemistry mechanisms, meteorology condition and dynamics as mentioned by the reviewer. Therefore we believe that the discrepancy in ozone concentrations between the two simulations using MEIC and updated provincial inventory came most from the varied estimations on emission level, source profiles, and spatial distribution of emissions. We have stated this **in lines 733-735 in the revised manuscript**.

We also agree with the reviewer that emission input was not the only limitation in chemistry transport modeling. To further figure out the impacts of chemistry and dynamics on ozone simulation, however, a more detailed sensitivity analysis will be needed on relevant parameters. We mean it is beyond the scope of the current work, and we will keep conducting the relevant analysis in the future. We have discussed such limitation **in lines 765-767 in the revised manuscript.**

10. *Specific comments: Figure 2: characters are not visible; Line 337: explain why SPECIATE is used. There are other database (European) that might be also relevant. Please explain why using SPECIATE. Line 439: explain the use and atmospheric relevancy of the OFP/emission ratio*

Response and revisions:

We thank reviewer’s comment and reminder.

The characters in Figure 2 are now visible.

To our knowledge, no domestic measurement has been conducted yet for those sources we selected. Few test results for the same source types were reported in European databases such as Theloke and Friedirch (2007), thus direct comparison was not conducted. Even in SPECIATE the chemical profiles were available for three processes, i.e., synthetic rubber, ethylene and polyethylene production, and we compared our results with SPECIATE for the three types.

OFP is used for evaluating the capability of ozone formation through atmospheric chemical reactions for individual VOC species. As the chemical profiles of emitted VOC vary between source categories, the OFP to emission ratio for a given sector (or source type) indicates the potential contribution to ozone formation for the sector (or source type), as a combined effect of multiple NMVOC species emitted from it. The ratio could thus provide scientific suggestion of emission control for policy makers, e.g., the emission control needs to be preferentially considered for sectors with large OFP/emissions. We have explained this issue **in lines 513-517 in the revised manuscript.**

11. *To conclude. This paper is of importance and I would like to highlight the extensive work that has been accomplished. However, given my comments above, I would not recommend publication. I would encourage the author to submit again their manuscript after improving the clarity of the paper (organization) and the accuracy of conclusions. The authors should also reduce the length of the paper as some parts are not supportive.*

Response and revisions:

Again we appreciate the reviewer’s remarks on the importance of the work. Regarding the weakness pointed out by the reviewer, we have improved the manuscript accordingly. In particular we have reorganized the text and provided more detailed information on emission inventory development to make the manuscript clear. Important data have been moved from the original supplement to the main text. The discussion of results has also been improved to avoid subjective statement and confusion, according to the reviewer’s comments and suggestion. Please see the details in the response and revision list to the reviewer’s comment. We have also tried our best to be concise in text. As much detailed information on methods and data sources (including relevant tables) has been added into the revised manuscript, however, the length of paper could hardly be reduced. We hope the revision could meet the quality standard of publication in Atmos. Chem. Phys.

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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

Non-methane volatile organic compounds (NMVOC) are the key precursors of ozone (O_3) and secondary organic aerosol (SOA) formation. Accurate estimation in their emissions plays a crucial role in air quality simulation and policy making. We developed a high-resolution anthropogenic NMVOCs emission inventory for Jiangsu in eastern China from 2005 to 2014, based on detailed information of individual local sources and the field measurements of source profiles of the chemical industry. A total of 56 NMVOCs samples were collected in 9 chemical plants, and then analyzed with a gas chromatography-mass spectrometry system (GC-MS). Source profiles of stack emissions from synthetic rubber, acetate fiber, polyether, vinyl acetate, and ethylene production, and those of fugitive emissions from ethylene, butanol and octanol, propylene epoxide, polyethylene and glycol production were obtained. Various manufacturing technologies and raw materials lead to discrepancies in source profiles between our domestic field tests and foreign results for synthetic rubber and ethylene production. The provincial NMVOC emissions were calculated to increase from 1774 Gg in 2005 to 2507 Gg in 2014, and relatively large emission densities were found in cities along the Yangtze River with developed economy and industry. The estimates were larger than those from most other available inventories, due mainly to the complete inclusion of emission sources and to the elevated activity levels from plant-by-plant investigation in this work. Industrial processes and solvent use were the largest contributing sectors, and their emissions were estimated to increase respectively from 461 to 958 and from 38 to 966 Gg. Alkanes, aromatics and oxygenated VOCs (OVOCs) were the most important species, accounting for 25.9%-29.9%, 20.8%-23.2% and 18.2%-21.0% to annual total emissions respectively. Quantified with a Monte-Carlo simulation, the uncertainties of annual NMVOCs emissions vary slightly from years, and the result for 2014 was -41%~+93%, expressed as 95% confidence intervals (CI). Reduced uncertainty was achieved compared to previous national and regional inventories, attributed partly to the detailed classification of emission sources and to the use of information at plant level

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in this work. Discrepancies in emission estimation were explored for chemical and refinery sector with various data sources and methods. Compared with Multi-resolution Emission Inventory for China (MEIC), the spatial distribution of emissions in this work were more influenced by the locations of large point sources, and smaller emissions were found in urban region for developed cities in southern Jiangsu. Besides, discrepancies were found between this work and MEIC in the speciation of NMVOC emissions under the atmospheric chemistry mechanisms CB05 and SAPRC99. The difference of species OLE1 resulted mainly from the updated source profile of building paint use, and the differences of other species from the varied sector contributions to emissions of the two inventories. CMAQ simulation was applied to evaluate the two inventories, and better performance (indicated by daily 1h-max O_3 concentrations in Nanjing city) was found for January, April and October 2012 when the provincial inventory was used.

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1 Introduction

With strong OH and HO_2 radical chemistry reactions, non-methane volatile organic compounds (NMVOCs) are reported to play crucial roles in formation of secondary organic aerosols (SOA) and serious photochemical pollution in China, particularly in developed cities and regions. For example, Huang et al. (2014) revealed that the contribution of SOA from NMVOC conversion reached 44%-71% to ambient organic aerosols during a heavy haze period in winter, based on detailed chemistry composition and source analysis of airborne particles in four important cities (Beijing, Shanghai, Guangzhou and Xi'an) across the country. Due to intensive emissions of species with strong atmospheric oxidation capability (indicated as maximum incremental reactivity, MIR), ozone (O_3) formation was recognized as VOC-limited in developed areas including Jing-Jin-Ji (JJJ), Yangtze River Delta (YRD) and Pearl River Delta (PRD) regions (Geng et al., 2008; Shao et al., 2009; Zhang et al., 2008; Xing et al., 2011).

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

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

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

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Under the heavy haze pollution in eastern China (Andersson et al., 2015; Sun et al., 2015; Wang et al., 2015), series of measures have been conducted particularly on power and industrial boilers to control the emissions of primary particles and the precursors of secondary particles such as SO_2 and NO_X (Zhao et al., 2014). Along with gradually reduced ambient PM levels in YRD, O_3 pollution becomes a bigger concern for air quality improvement, motivating the better understanding and controlling of NMVOC emissions. In this work, we select Jiangsu, a typical province with intensive refinery and chemical industry in eastern China, to develop and evaluate the high-resolution emission inventory of anthropogenic NMVOCs. The geographic location and cities of the province are illustrated in Figure S1 in the supplement. Field measurements on chemical composition of NMVOC emissions were conducted to obtain the source profiles of typical chemical engineering processes. With detailed information of local emission sources collected and temporal changes tracked, the provincial emission inventory of NMVOC with chemistry profiles were developed for a ten-year period 2005-2014, and the uncertainties of the emission estimation were quantified. Through a thorough comparison between results from varied methods and data sources, the discrepancies in emission estimation, source profiles, and spatial patterns were then evaluated. Finally, chemistry transport modeling was applied in southern Jiangsu to test the improvement of the provincial NMVOC inventory.

2 Data and methods

2.1 Sampling and analysis of NMVOC species from chemical plants

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

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NMVOC samples were analyzed by one Gas Chromatograph Mass Spectrometer (GC-MS) system (GC6890/MS5973i, Agilent Technologies, USA), as illustrated in Figure S2b in the supplement. Firstly, the sample was pumped into a cryogenic pre-concentrator with a three-stage trapping system. In the first stage, the VOC species were adsorbed on glass beads at –150°C and desorbed at 10°C. Vapour water was converted to solid water and was thus separated from sample. In the second stage, the sample was trapped on Tenax at –30°C and desorbed at 180°C, and the target species were separated from CO_2 and other compositions in the air. In the third stage, the sample was focused on a transfer line at –160°C and heated rapidly to 70°C, and the species were concentrated. The concentrated VOC was then injected into the gas chromatograph. The GC oven temperature was initially programmed at –50°C, and then increased to 180°C at 4°C/min and to 220°C at 15°C/min, holding 3minutes. The VOC compounds were separated on a DB-5MS capillary column (60 m×0.25 mm×1.0 µm) and quantified using a quadrupole mass spectrometer. The mass spectrometer was operated in SCAN mode and scanned 20-42 amu and 35-270 amu before and after 8.5 minutes, respectively. The ionization method was electron impacting, and the source temperature was 230°C. The PAMS (Photochemical Assessment Monitoring System) standard mixture (Spectra Gases Inc., USA) and TO-15 standard mixture (Spectra Gases Inc., USA) were used to confirm the retention times of different compounds, and to identify them based on retention time and mass spectrum. The target species were quantified by using multipoint internal calibration method. To establish calibration curves, a certified gas mixture containing all the target compounds was dynamically diluted with pure nitrogen to five concentration gradients using mass flow controllers. Bromochloromethane, 1,2-difluorobenzene, and chlorobenzene-d5 were chosen as internal standards in samples. The method detection limit was 0.5 ppb for all species.

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2.2 Development of provincial emission inventory

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

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

where *i* and *n* represent the source type and year, respectively; *E* is the annual emissions; *AL* is the activity level data; and *EF* is the emission factor (i.e., emissions per unit of *AL*). As summarized in Table 2, emission factors were collected from extensive literatures and determined as follows with descending priorities: (1) the results from domestic measurements; (2) the emission limits of local laws and regulations; (3) the values from expert judgment specific for China; and (4) emission

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$$E(n)=\sum_i AL(i,n)\times$$

(1)

where *i* and *n* represent the source type and year, respectively; *E* is the annual emissions; *AL* is the activity level data; and *EF* is the emission factor (i.e., emissions per unit of *AL*).

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删除的内容: in the supplement, the emission sources were classified into a four-level framework, including factors from AP-42 database (USEPA, 2002) and the EMEP/EEA guidebook (EEA, 2013) when domestic information was lacking. Details will be discussed by sector in Section 2.3.

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

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

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

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

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

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where E is the emissions, M is the mole weight, C is the conversion factor, and i, m, and k represent the source type, individual species, and the chemical mechanism species.

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

2.3 Data sources of emission inventory development by category

Combustion sources (fossil fuel combustion and biomass burning)

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

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respectively.

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

Industrial processes

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

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删除的内容: The information included geographic location, types and amounts of raw materials, types and amounts of products, fuel quality and consumption, and combustion/manufacturing technology. For 2014, as an example, detailed information of 6023 plants was obtained, with the locations of those point sources illustrated in Figure S1.

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production as normal distribution with CVs at 10% and 20% for point and area sources, respectively.

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

Solvent use

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

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删除的内容：Activity data of other emission sources were generally collected at provincial or city level based on official statistical yearbooks. In particular, a

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20% and 80% were applied for the activity levels of point and area sources of solvent use, respectively, indicating much larger uncertainty for the latter (Wei, 2009).

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

Transportation

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

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on-road and non-road transportation, respectively.

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

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

where BEF is the base emission factor, φ is the environmental correction factor, γ is the average traveling speed correction factor, λ is the vehicle deterioration correction factor, and θ is correction factor for other conditions (e.g., vehicle loading and fuel quality). Domestic tests on chemical compositions (Liu et al., 2008; Tsai et al., 2012; Huang et al., 2015; Wang et al., 2013; Ou et al., 2014; Gao et al., 2012) were incorporated to update the source profile of on-road vehicles. Lognormal distributions with CVs at 150% and 300% were respectively assumed for emission factors of on-road and off-road transportation.

Oil distribution and other sources

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

3 Results

3.1 Source profiles of chemical industry from measurement

NMVOC source profiles of 14 processes (9 for stack emissions and 5 for fugitive

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2.3 Determination of emission factors

Based on extensive literature survey, the NMVOC emission factors were determined as follows with descending priorities: (1) the results from domestic measurements; (2) the emission limits of local laws and regulations; (3) the values from expert judgment specific for China; and (4) emission factors from AP-42 database (USEPA, 2002) and the EMEP/EEA guidebook (EEA, 2013) when above information was lacking. The emission factors applied in this work are summarized by sector in Tables S4-S6 in the supplement.

The emission factors for power plants and industrial boilers were taken mainly from Bo et al. (2008). Given the similar designs of boilers between China and developed countries (Wei et al., 2008), the emission factors in AP-42 database were also applied when domestic results were lacking. For residential combustion of fossil fuel, the emission factors from domestic tests (Zhang et al., 2000; Wang et al., 2009) were used as

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emissions) were obtained from field measurements. With totally 61 species detected by GC-MS systems, the chemistry components were grouped into 6 types (alkanes, alkenes, halohydrocarbon, aromatics, OVOCs, and others), as illustrated in Figure 1a. Alkanes were the main species of synthetic rubber industry (SBR, SIS rubber and SEBS rubber) and the mass fractions were measured over 70%. For production of cellulose acetate fiber, alkanes, aromatics, and OVOCs were the main species in process of acetate flake production, while OVOCs dominated in the spinning process. Resulting from various raw materials applied, the source profiles of the two types of polyether production differed a lot: the mass fraction of OVOCs was close to 80% for PPG, while the main species for POP were others, aromatics and halohydrocarbon. For ethylene production, aromatics were the main composition in the stack gas of cracking furnace, while alkanes and alkenes dominated the fugitive emissions. For other types of fugitive emissions, big differences existed in the source profiles attributed mainly to the various raw materials and chemical reactions.

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

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fractions measured at 42% and 35%, respectively.

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

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3.2 Inter-annual trends and sectoral contribution of NMVOC emissions

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

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gas/electricity ones, and the gradual implementation of straw burning prohibition. Relatively small contributions were found for stationary fuel combustion plants, oil distribution, and other sources, and their collective fractions to total emissions ranged 7-9% during the study period.

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

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

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

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Unsurprisingly, gasoline vehicles dominated the emissions of on-road transportation, with the fraction ranged 81%-87% during the study period.

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

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3.3 Speciation and OFPs of NMVOC emissions

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

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Shown in Figure 3 are the mass fractions of species by source for 2014. Due to varied fuel qualities and combustion conditions, large differences in the speciation of emissions were found for fossil fuel combustion, transportation and biomass burning. Dominated by coal combustion, the profile of fossil fuel stationary sources was relatively simple with little OVOCs and halohydrocarbons, and aromatics were the largest fraction (45%), followed by alkanes (29%). Alkanes, aromatics and alkenes were the main species from transportation, with the fractions estimated at 30%, 24%, and 23%, respectively. For biomass burning, elevated alkenes and less alkanes were found attributed to the highly incomplete combustion. The mass fractions of alkanes, aromatics and alkenes from industrial processes were estimated 32%, 16% and 12%. In particular, relatively close emission fractions were found between species for chemical industry, the largest emission source of industrial processes: 19%, 15%, 11%, 10% and 10% of aromatics, ketones, alkenes, alkanes, and halohydrocarbons, respectively. For solvent use, aromatics and alkanes were the most important species with the fractions estimated at 32% and 22%, respectively, and the collective fraction of OVOC species reached 27%. Alkanes and aldehydes dominated the emissions of oil distribution and other sources, which came mainly from the oil evaporation and residential cooking, respectively.

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

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low reactivity. Figure S4 in the supplement provided 25 species with the biggest contributions to OFPs and their emission sources for 2005 and 2014. In 2005, the 25 species were estimated to account for 44% of total NMVOC emissions and 83% of OFPs (Figure S4a). Xylene, ethylene, and propylene were identified as the most three important species in terms of OFP. The aromatics species with (e.g., xylene and toluene) came largely from solvent use and industrial processes, while alkenes species (e.g., ethylene and propylene) were mainly from industrial processes, biomass burning, and transportation. Besides, biomass burning was the dominating sources of methyl glyoxal, methyl alcohol and glyoxal. For 2014, the 25 species were estimated to account for 38% of total NMVOC emissions and 81% of OFPs, and the contributions of solvent use and industrial processes to OFPs were clearly elevated (Figure S4b). The orders of isopropanol and n-butanol that were mostly from solvent use, for example, were moved forward. Moreover, the contribution of biomass burning significantly declined, and the glyoxal was not identified as the one of the 25 most important species to OFPs any more.

3.4 Uncertainties of provincial NMVOC emission inventory

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

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domestic samples and to heavy dependence on foreign databases. More measurements on actual emission characteristics are thus recommended to expand data samples for better evaluating the PDFs and effectively reducing emission uncertainty. Regarding solvent use for which provincial and city statistics were lacking, the activity data had to be downscaled from national level leading to possibly big bias in emission estimation.

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

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4 Evaluation of provincial emission inventory

4.1 Chemical and refinery industry emissions from varied data sources and methods

As a part of industrial process, chemical and refinery industry was one of the biggest contributors to anthropogenic NMVOC emissions. We select Nanjing, the capital city of Jiangsu province, to evaluate the impacts of data sources and methods on emissions of this category. As described in Sections 2.2 and 2.3, the method used in this work for provincial inventory improvement incorporated the most available

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

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

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scale without sufficient investigation on local sources, particularly for regions where emissions were dominated by large plants that were gradually moved out of urban areas.

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

4.2 Changes in speciation of NMVOC emissions

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

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

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

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profiles of solvent use were the main reason for the changes. For example, the increased ALDX was attributed mainly to the updated profiles of printing ink and automobile paint use (Zheng et al., 2013; Tang et al., 2014), while increased OLE1 was to that of building coating (Yuan et al., 2010; Wang et al., 2014b) .

4.3 Comparisons with other inventories

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

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The emissions of CB05 and SAPRC 99 species estimated by us and MEIC were compared in Figure 7. While total NMVOC emissions in this work were 315 Gg or 18% larger than MEIC for 2010, relative changes varied among species and could be bigger for certain ones. In CB05 mechanism, our results were 46% and 43% smaller

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for TOL and XYL but 38% and 59% larger for ETH and ETHA than MEIC (Figure 7a), while discrepancies of over 30% (relative to MEIC result) existed for most species in SAPRC 99 (Figure 7b). Such discrepancies could result either from the various source profiles, or from the various source contributions in total emissions. For example, with updated source profile for building coating, much larger OLE1 emissions were estimated in this work than MEIC. Besides the total emissions, the differed speciations under chemical mechanisms could result in complex impacts on air quality simulation, which would be discussed in next section.

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

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4.4 Evaluation of multiple-scale inventories through air quality modeling

The Models-3/Community Multi-scale Air Quality (CMAQ) version 4.7.1 was applied to test the performances of chemistry transport simulation with various

NMVOC emission inventories for Jiangsu area. As shown in Figure S6 in the supplement, three nested domains (D1, D2 and D3) were set, and the most inner D3 covered the mega city Shanghai and six most developed cities in southern Jiangsu including Nanjing, Changzhou, Zhenjiang, Wuxi, Suzhou and Nantong, with a horizontal spatial resolution at 3 km. Chemistry transport simulations were conducted separately with two inventories, i.e., MEIC and the provincial one developed in this work, for January, April, July and October 2012. Other model settings on meteorological simulation, chemistry mechanisms and emissions of natural origin were the same for the two simulations, as described in Zhou et al. (2017). The first five days for each month were chosen as spin-up period to provide initial conditions for later simulations.

Figure 10 provides the observed and predicted daily 1h-max O_3 concentrations for the four months, and Table 6 compares the model performances with MEIC and our provincial inventory, indicated as normalized mean bias (NMB) and error (NME) values. As suggested by the minus NMBs for most cases, model usually generated lower 1h-max O_3 concentrations than observation with either MEIC or provincial inventory applied, with an exception for April simulation with MEIC applied. The result thus implied the updated anthropogenic NMVOC emission inventory at provincial scale was still likely an underestimation of the actual emissions, as YRD was commonly recognized as VOC-limited region for O_3 formation (Xing et al., 2011). Compared to MEIC, better model performances (except for July) were generally achieved when the provincial inventory was applied, indicating the improved reliability of the detailed bottom-up NMVOC inventory on high-resolution chemistry transport simulation. In particular, larger emissions were estimated for certain species with relatively high ozone formation potential (e.g., ethene and ethanol) in the provincial inventory. It should be noted that the improved ozone simulation was a combined effect of the updated inventory with revisions on emission estimation, spatial distribution and source profiles for all the relevant species, and that the impacts of emission changes for individual species could not be completely validated. More chemistry transport modeling is further encouraged with intensive sensitivity analysis.

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The discrepancies between simulation and observation were still large compared to regional studies in North America (Y. Zhang et al., 2009). More efforts on improving or validating emission inventory at provincial scale are thus in great needed. Besides careful compilation of emission source information in the bottom-up method, observation constraint from ground measurements could be used to evaluate the emission level, source contribution, and speciation of VOC emissions (M. Wang et al., 2014). Emission uncertainty of NO_X could also partly explain the discrepancies, as the NO_X control measures taken recently could hardly be fully tracked in the emission inventory development. Besides the limitation of emission input, more analysis on the impacts of chemical mechanisms and dynamics in the chemistry transport modeling are also suggested for O_3 prediction in the region.

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5 Conclusion

Using a bottom-up approach, we developed a high-resolution emission inventory of anthropogenic NMVOC for Jiangsu province, eastern China, with substantial detailed information on local sources and source profiles from domestic tests incorporated. Attributed largely to the elevated contribution from industrial processes and solvent use, the annual provincial emissions were estimated to increase 41% from 2005 to 2014. Influenced largely by location of big point sources, high emission densities were found in cities along the Yangtze River. Our estimations were larger than results from most other available inventories except REAS, due mainly to the omissions of certain industrial and solvent use sources by other studies and to the elevated activity levels from plant-by-plant investigation in this work. Benefiting from more detailed classification and investigation of emission sources, reduced uncertainties in annual emissions were quantified in this work compared to previous studies. Varied data sources and methods were of significant impacts on emission estimation at city/plant level. In particular, downscaling directly from national inventories would overestimate the fraction of emissions in urban downtown. With the most recent source profiles from local measurements included, considerable changes in emissions were found for ethylacetate and certain aromatics species, and the speciation under CB05 and SAPRC99 differed a lot from the national inventory MEIC. Compared to MEIC, better model performance (indicated by daily 1h-max O_3 concentrations) were achieved when the improved provincial inventory was used in CMAQ simulation, although the discrepancies between simulation and observation need further investigation. As emission controls on NMVOCs started to be implemented on industrial sources in China (e.g., the application of leak detection and repair technique in chemical and refinery plants), more field tests were recommended to better track the temporal changes in emission factors and to reduce the uncertainty 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^{-1} min^{-1}, respectively. ETHENE respect ethane, and OLE1 for alkenes with kOH smaller than 70000 ppm^{-1} min^{-1}, OLE2 for alkenes with kOH greater than 70000 ppm^{-1} min^{-1}, ARO1 for aromatics with kOH smaller than 20000 ppm^{-1} min^{-1}, ARO2 for aromatics with kOH greater than 20000 ppm^{-1} min^{-1} and NROG for unreactive mass.

Figure 8. Jiangsu's anthropogenic NMVOC emissions by sector for selected yeas 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_3 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
Combustion				
Fossil fuel	Power plant	Coal	0.15	0.15[a]; 0.81[b]; 0.02[c]; 0.03-0.03[d]; 0.02[e]
		Fuel oil	0.09	3.88[b]; 0.04[c]; 0.12[d]; 0.09-0.14[e]
		Natural gas (g/m^3)	0.083	0.12[c]; 0.08-0.10[d]; 0.08-0.10[e]
	Heating and industrial boiler	Coal	0.18	0.18[a]; 3.95[b]; 0.04[c]; 0.03-0.03[d]; 0.02[e]
		Fuel oil	0.12	3.88[b]; 0.12[c]; 0.12[d]; 0.09-0.14[e]
		Natural gas (g/m^3)	0.094	0.1[c]; 0.08-0.10[d]; 0.08-0.10[e]
	Residential	Coal	4.5	0.6[a]; 3.08[b]; 6.48, 4.98[g]; 4.5[c]; 6.41[d]
		Fuel oil	0.35	0.35[c]
		LPG	5.29	0.17[a,b];3.28[g]; 5.29[c]; 66 (g/m^3)[e]
		Natural gas (g/m^3)	0.15	0.13[c]; 0.15[c]; 0.14[d]; 0.18[e]
		Coal gas	0.00044	0.00044[a]
Biomass burning	Boiler	Biomass	1.1	0.0015[e]; 1.1[f]
	Stove burning	Wood	3.23	1.09-4.94[g]; 3.23[c]; 5.3[e]
		Crop straw	13.77	1.7- 3.0 (corn straw)[g], 8.89 (wheat straw)[g]; 13.77[c] ; 8.55[d]; 5.3[e]
	Open burning	Rice straw	7.48	7.48[h]
		Wheat straw	7.48	7.48[h]
		Corn straw	10.4	10.4[h]
		Other straws	8.94	Average of straws open burning above
Industrial process				
Iron and steel	Coking/sintering/steel		3.96/0.25/0.06	3.4[c]; 3.96[f]/0.25[f]/0.06[f]
Non-metallic mineral	Flat glass/glass fiber/glass work		4.4/3.15/4.4	4.4[c,f]; 3.5[e]/3.15[a, i]/4.4[f]
	Cement clinker/lime/brick and tile/ceramic		0.33/0.177/0.13/ 29.22	0.12[c]; 0.018[d]; 0.33[f]/0.177[f]/0.01[i]; 0.2[c]; 0.033[a]; 0.13[f]/29[c]; 29.215[a]; 29.22[c]
Oil exploitation/refinery			1.42/1.82	1.42[f]/1.82[f]; 1.05[d]; 3.54[e]
Chemical industry	Ethylene/benzene/methanol		0.097/0.1/5.95	0.097[f]; 0.6[d]/0.1[f]; 0.25[j]/5.95[f]
	Polyethylene/polypropylene		10/8	10[f,j]; 0.33[e]; 3.4[d]/8[f,j]; 0.35[e]; 4[d];
	Polystyrene/polyvinyl chloride		5.4/3	5.4[f]; 0.21-3.34[e]; 0.12[d]/3[f,j]; 0.33-8.5[e]; 0.096[d]
	Acetic acid/ammonia/phthalic anhydride		1.814/4.72/21	1.81[i]/4.72[f,e]/1.1-6.3[e]; 1.3-6[d]; 21[j]
	Ethylene oxide/vinyl acetate/styrene		3/4.705/0.223	3[f]; 0.98[e]; 2[d]; 4[j]/4.705[i]/0.223[f]; 1[d]; 3.1[j]
	Viscose fiber/cellulose acetate fiber/nylon fiber		14.5/73.4/3.3	14.5[c,f]/73.4[f]; 112[e]/2.13-3.93
	Polyester fiber/acrylic fibers polypropylene fiber		0.7/40/37.1	0.7[f]; 0.6[e]/3.75-40[e]/37.1[c]

Table 2 (continued)

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

[a] Bo et al. (2008); [b] Tang and Chen (2002); [c] Wei et al. (2008), Wei(2009), and Wang et al.(2009); [d] EEA (2013); [e] USEPA (2002); [f] MEP (2014); [g] Zhang et al. (2000); [h] Li et al. (2007); [i] ROC EPA (2009); [j] Fan et al. (2012); [k] Xia et al. (2014); [l] Wang (2006); [m] Klimont et al. (2002); [n] 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.

	Uncertainty					Parameters contributing most to uncertainty	
Source Category	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%	-	-	-	-	$EF_{\text{Power plant, coal}}$ (68%)	$EF_{\text{Industrial boiler, coal}}$ (6%)
Industrial process	-58%, +152%	-88%, +283%	-	-57%, +152%	-60%, +152%	EF_{Tire} (23%)	EF_{Coking} (13%)
Solvent use	-68%, +131%	-82%, +223%	-	-60%, +147%	-59%, +150%	$AL_{\text{External wall paint}}$ (20%)	$EF_{\text{Other paint}}$ (14%)
Transportation	-51%, +117%	-86%, +261%	-	-	-	$EF_{\text{Inland ship}}$ (13%)	$EF_{\text{Construction machine}}$ (8%)
Oil distribution	-66%, +162%	-	-	-	-	$EF_{\text{Crude oil storage}}$ (27%)	$EF_{\text{Gasoline sale}}$ (23%)
Biomass burning	-76%, +499%	-	-	-	-	$EF_{\text{Straw-stove}}$ (74%)	$R_{\text{Straw burning in stove}}$[1] (5%)
Other	-98%, +490%	-	-		-	EF_{Cooking} (84%)	$EF_{\text{Garbage burning}}$ (14%)
Total	-41%, +93%	-51%, +133%	-36%, +94%	-52%, +105%	-53%, +113%		

[1] The ratio of straw burned in stove as biofuel

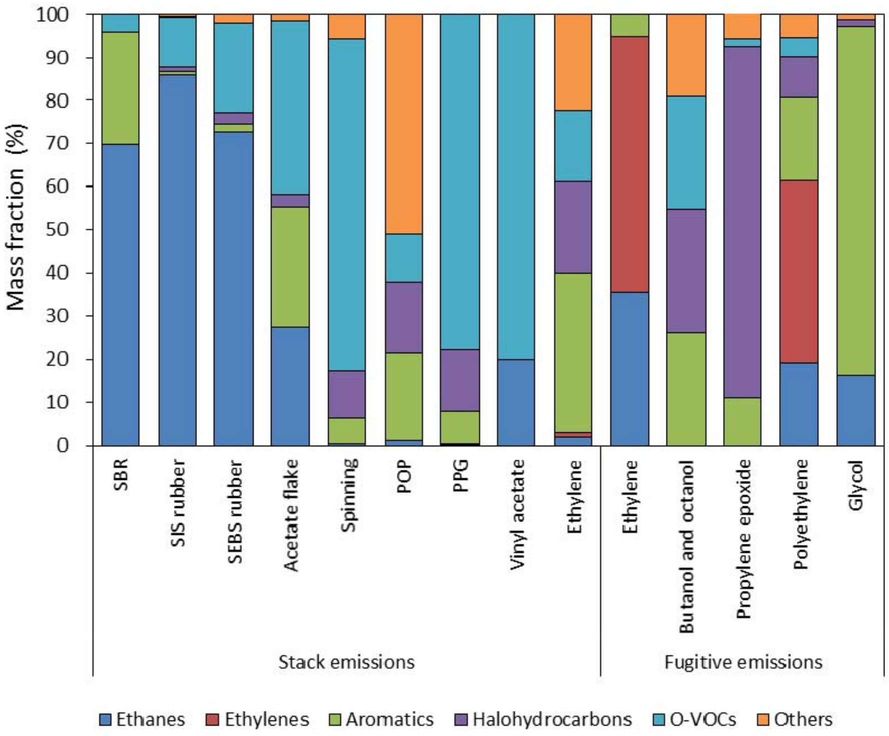
Table 6 Model performance of daily 1h-max O_3 concentrations using MEIC and provincial inventory for January, April, July and October 2012 in Nanjing.

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

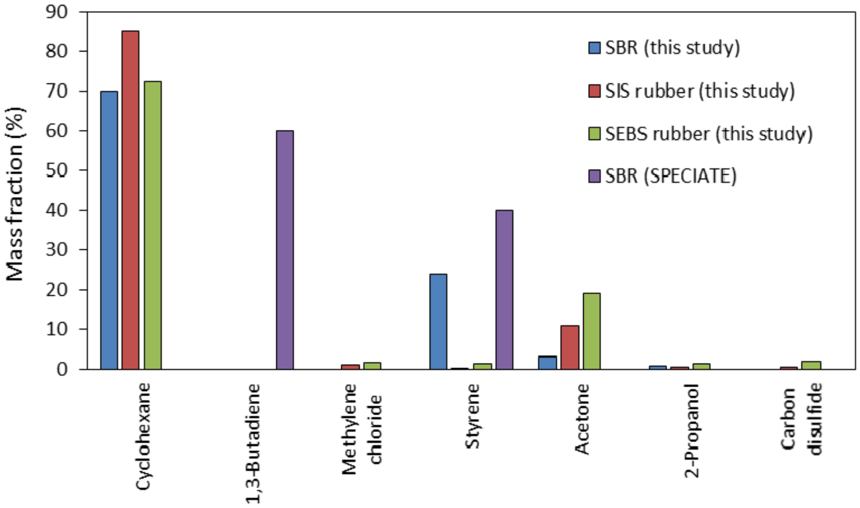
[1] Normalized mean bias (NMB) and error (NME) were calculated as following equations (P_i and O_i indicate the results from modeling prediction and observation, respectively):

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

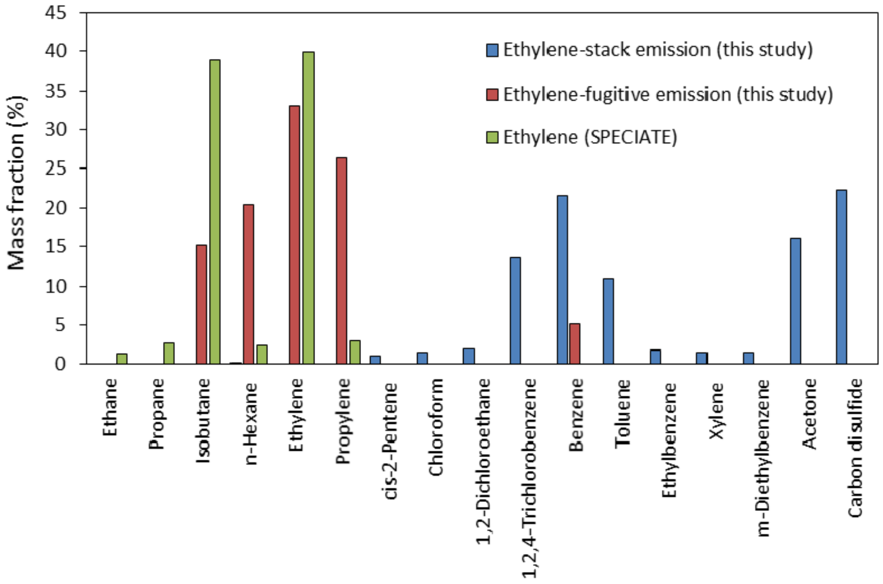
Figure 1


Mass fraction (%)
100
90
80
70
60
50
40
30
20
10
0
SBR
SIS rubber
SEBS rubber
Acetate flake
Spinning
POP
PPG
Vinyl acetate
Ethylene
Ethylene
Butanol and octanol
Propylene epoxide
Polyethylene
Glycol
Stack emissions
Fugitive emissions
Ethanes
Ethylenes
Aromatics
Halohydrocarbons
O-VOCs
Others

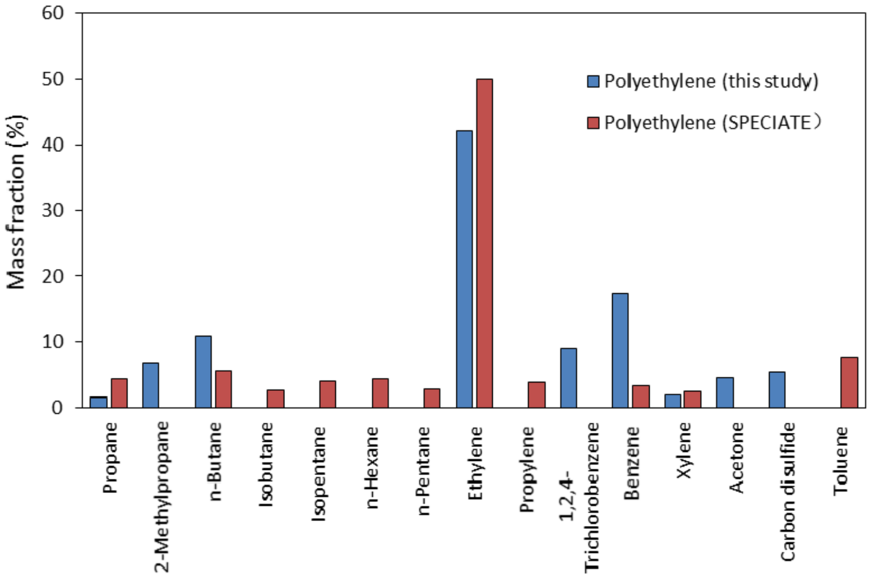
(a)


Mass fraction (%)
90
80
70
60
50
40
30
20
10
0
SBR (this study)
SIS rubber (this study)
SEBS rubber (this study)
SBR (SPECIATE)
Cyclohexane
1,3-Butadiene
Methylene chloride
Styrene
Acetone
2-Propanol
Carbon disulfide

(b)

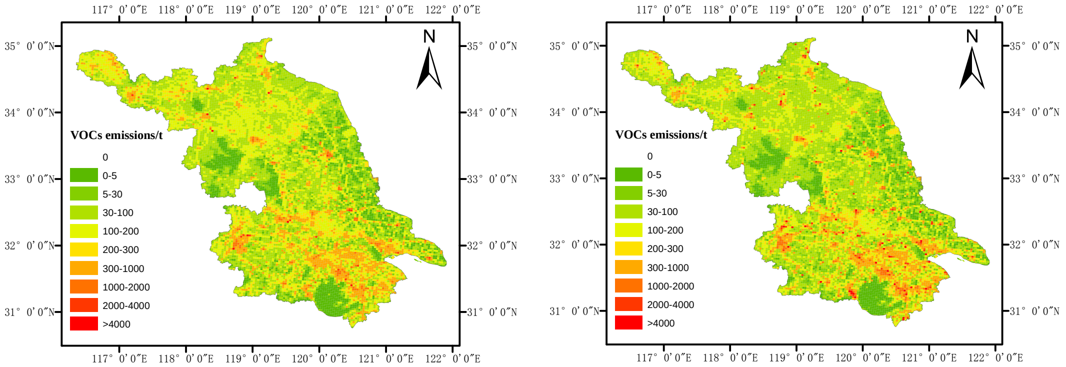

45
40
35
30
25
20
15
10
5
0
Mass fraction (%)
Ethylene-stack emission (this study)
Ethylene-fugitive emission (this study)
Ethylene (SPECIATE)
Ethane
Propane
Isobutane
n-Hexane
Ethylene
Propylene
cis-2-Pentene
Chloroform
1,2-Dichloroethane
1,2,4-Trichlorobenzene
Benzene
Toluene
Ethylbenzene
Xylene
m-Diethylbenzene
Acetone
Carbon disulfide

(c)


60
50
40
30
20
10
0
Mass fraction (%)
Polyethylene (this study)
Polyethylene (SPECIATE）
Propane
2-Methylpropane
n-Butane
Isobutane
Isopentane
n-Hexane
n-Pentane
Ethylene
Propylene
1,2,4-Trichlorobenzene
Benzene
Xylene
Acetone
Carbon disulfide
Toluene

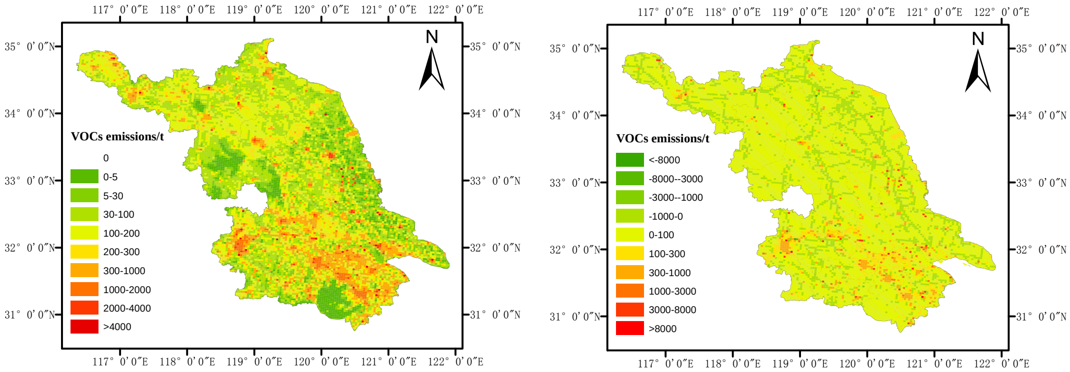
(d)

Figure 2



(a) 2005

(b) 2010



(c) 2014

(d) Difference between 2005 and 2014

Figure 3

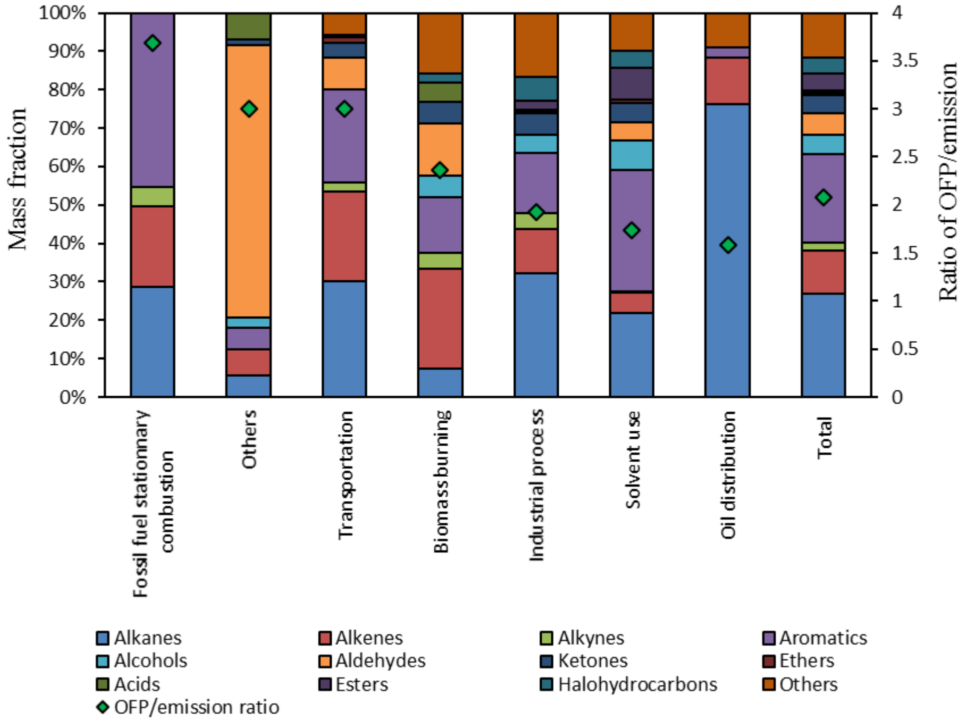

100%
90%
80%
70%
60%
50%
40%
30%
20%
10%
0%
Mass fraction
4
3.5
3
2.5
2
1.5
1
0.5
0
Ratio of OFP/emission
Fossil fuel stationary combustion
Others
Transportation
Biomass burning
Industrial process
Solvent use
Oil distribution
Total
Alkanes
Alkenes
Alkynes
Aromatics
Alcohols
Aldehydes
Ketones
Ethers
Acids
Esters
Halohydrocarbons
Others
OFP/emission ratio

Figure 4

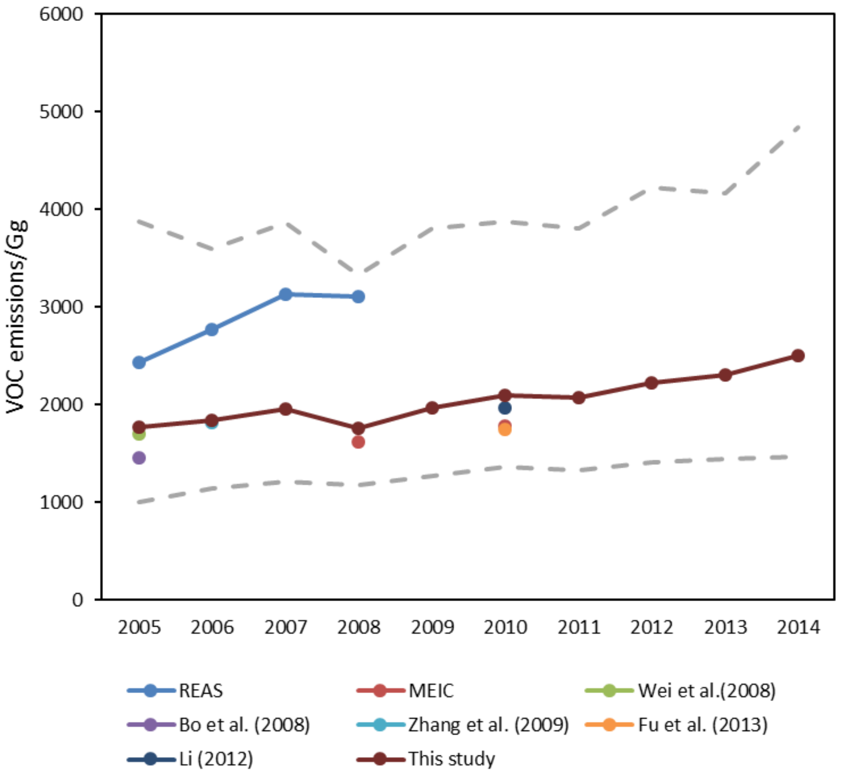
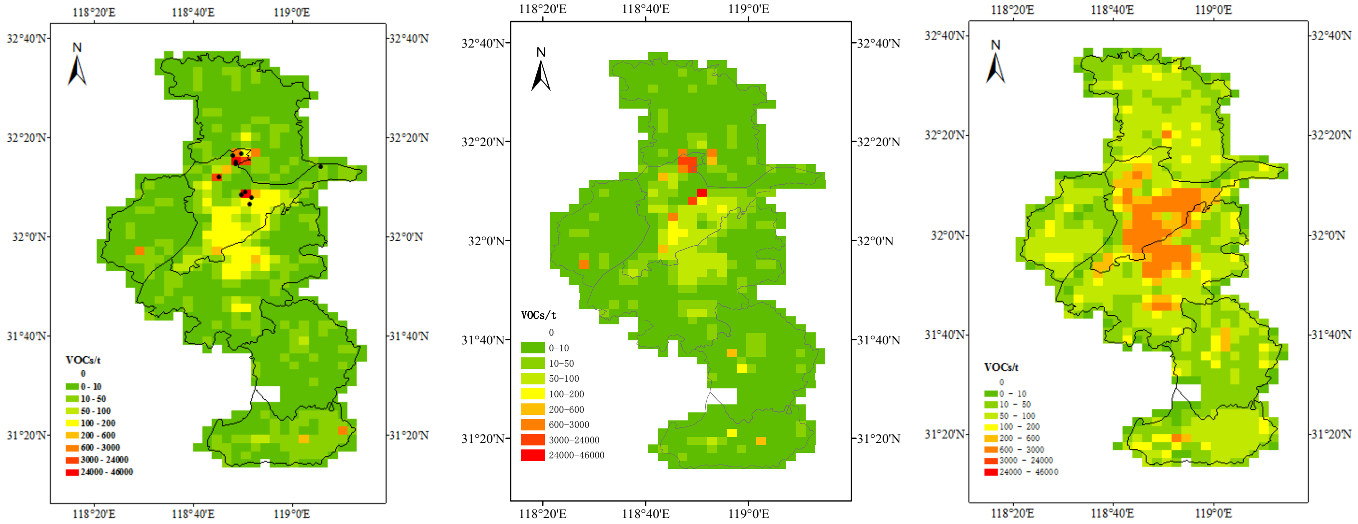

6000
5000
4000
3000
2000
1000
0
VOC emissions/Gg
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
REAS
MEIC
Wei et al.(2008)
Bo et al. (2008)
Zhang et al. (2009)
Fu et al. (2013)
Li (2012)
This study

Figure 5


118°20′E
118°40′E
119°0′E
32°40′N
32°20′N
32°0′N
31°40′N
31°20′N
N
VOCs/t
0
0 - 10
10 - 50
50 - 100
100 - 200
200 - 600
600 - 3000
3000 - 24000
24000 - 46000

(a) (b) (c)

Figure 6

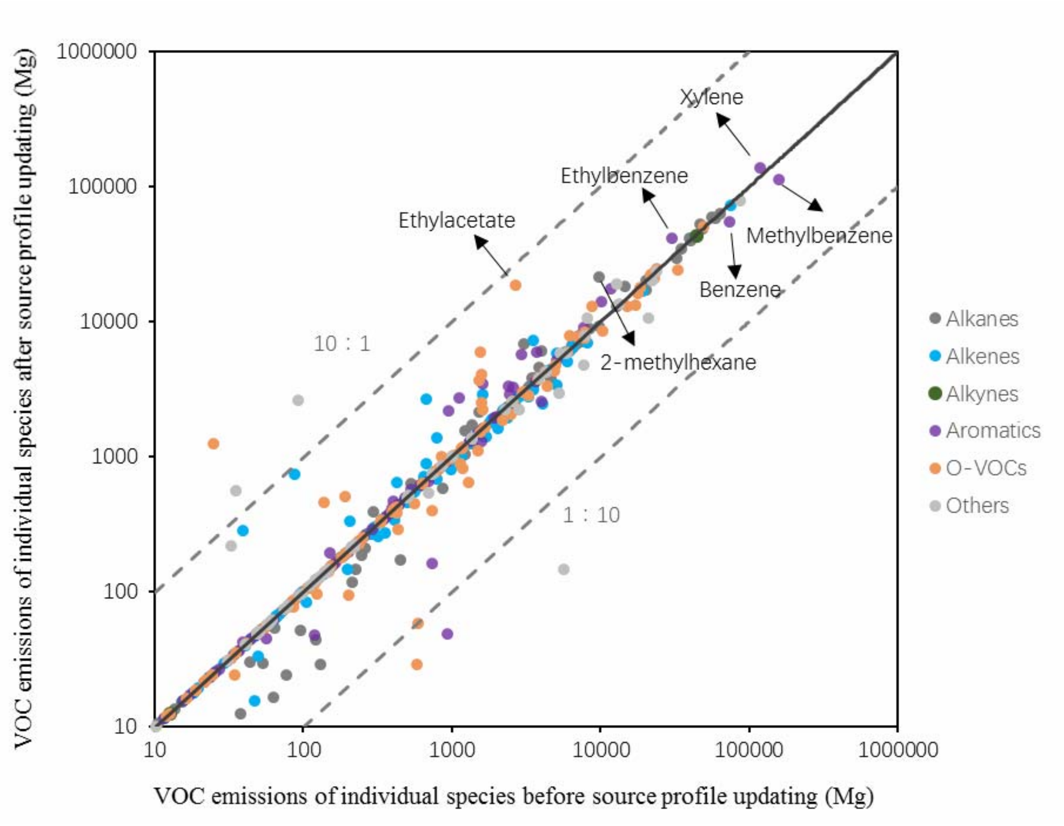
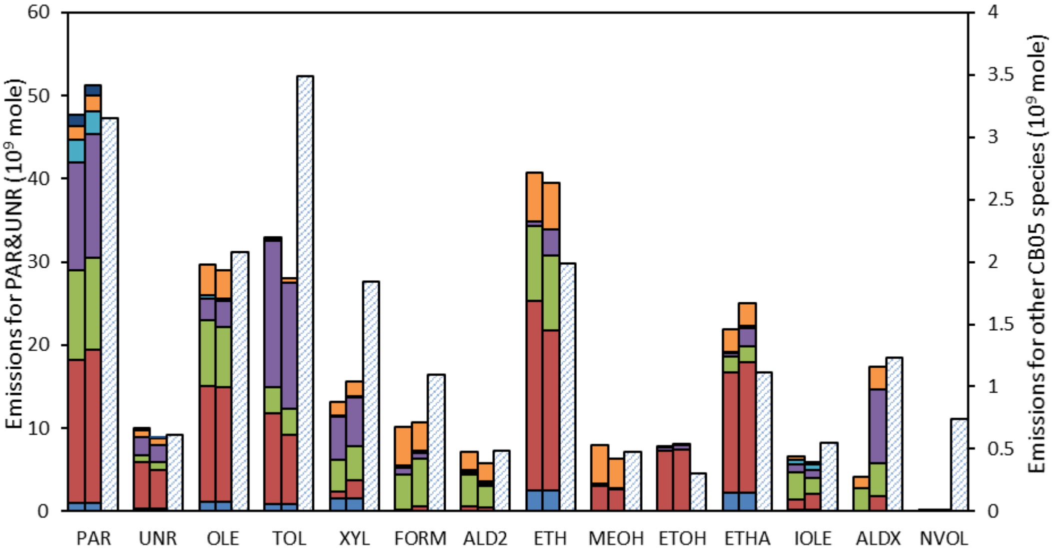

VOC emissions of individual species after source profile updating (Mg)
1000000
100000
10000
1000
100
10
10
100
1000
10000
100000
1000000
VOC emissions of individual species before source profile updating (Mg)
Xylene
Ethylbenzene
Ethylacetate
Methylbenzene
Benzene
2-methylhexane
10 : 1
1 : 10
● Alkanes
● Alkenes
● Alkynes
● Aromatics
● O-VOCs
● Others

Figure 7

(a) CB05



(b) SAPRC99

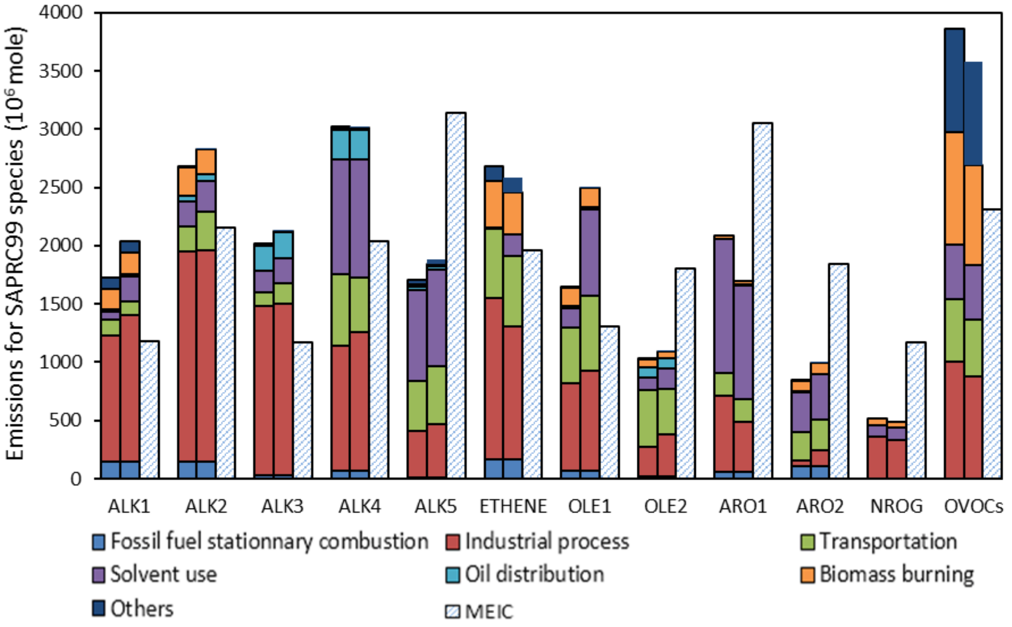


Figure 8

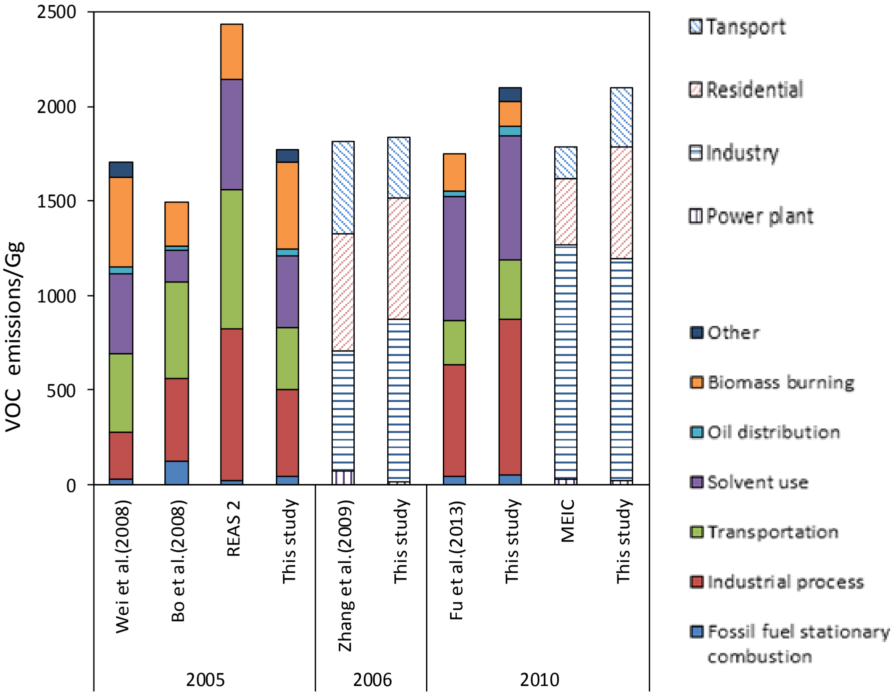
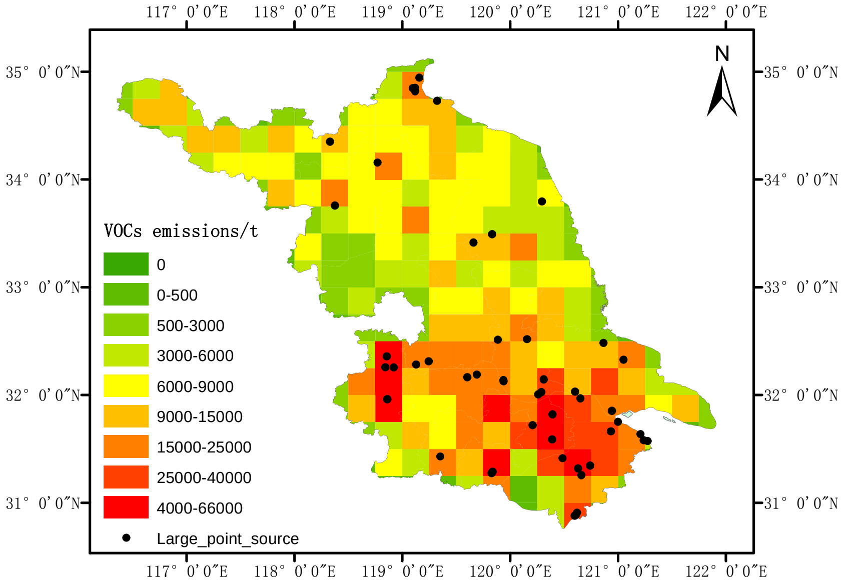
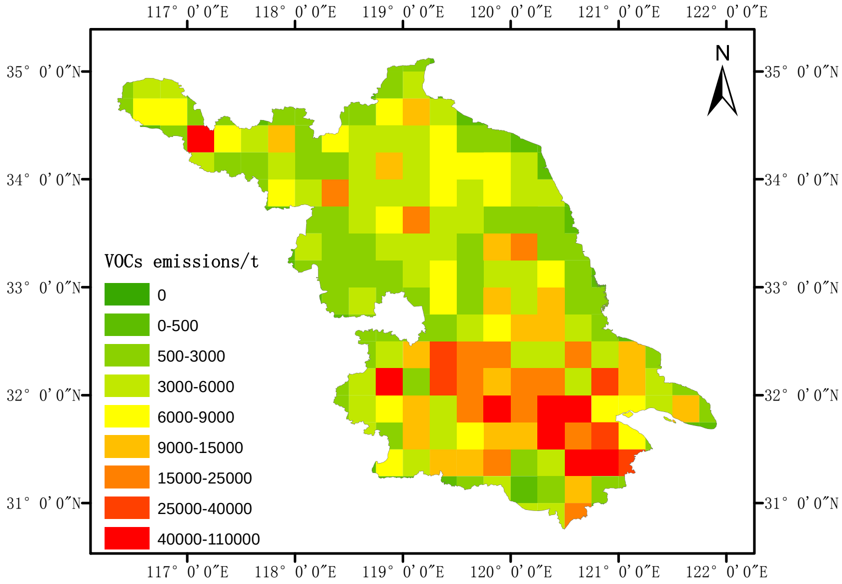

2500
2000
1500
1000
500
0
VOC emissions/Gg
Wei et al.(2008)
Bo et al.(2008)
REAS 2
This study
Zhang et al.(2009)
This study
Fu et al.(2013)
This study
MEIC
This study
2005
2006
2010
Tansport
Residential
Industry
Power plant
Other
Biomass burning
Oil distribution
Solvent use
Transportation
Industrial process
Fossil fuel stationary combustion

Figure 9

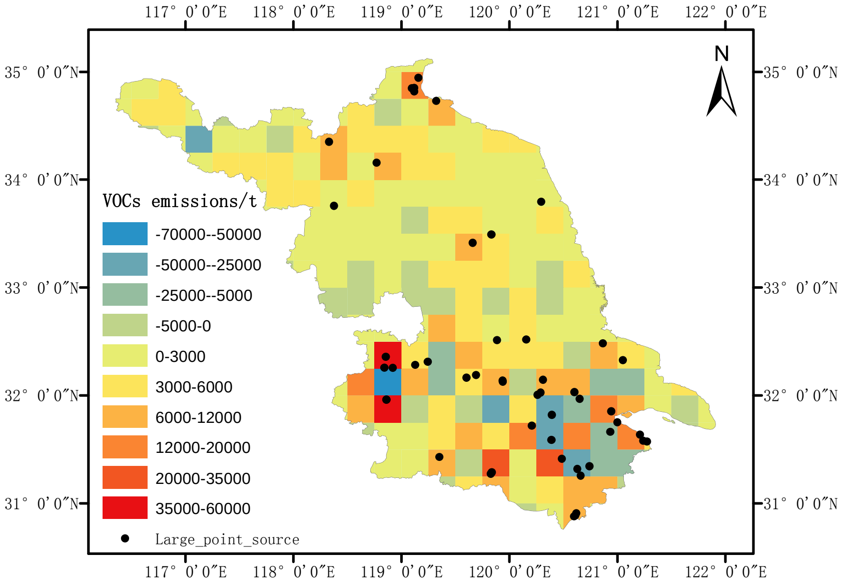

117° 0′0″E
118° 0′0″E
119° 0′0″E
120° 0′0″E
121° 0′0″E
122° 0′0″E
35° 0′0″N
34° 0′0″N
33° 0′0″N
32° 0′0″N
31° 0′0″N
N
VOCs emissions/t
0
0-500
500-3000
3000-6000
6000-9000
9000-15000
15000-25000
25000-40000
4000-66000
Large_point_source

(a)


117° 0′0″E
118° 0′0″E
119° 0′0″E
120° 0′0″E
121° 0′0″E
122° 0′0″E
35° 0′0″N
34° 0′0″N
33° 0′0″N
32° 0′0″N
31° 0′0″N
N
VOCs emissions/t
0
0-500
500-3000
3000-6000
6000-9000
9000-15000
15000-25000
25000-40000
40000-110000

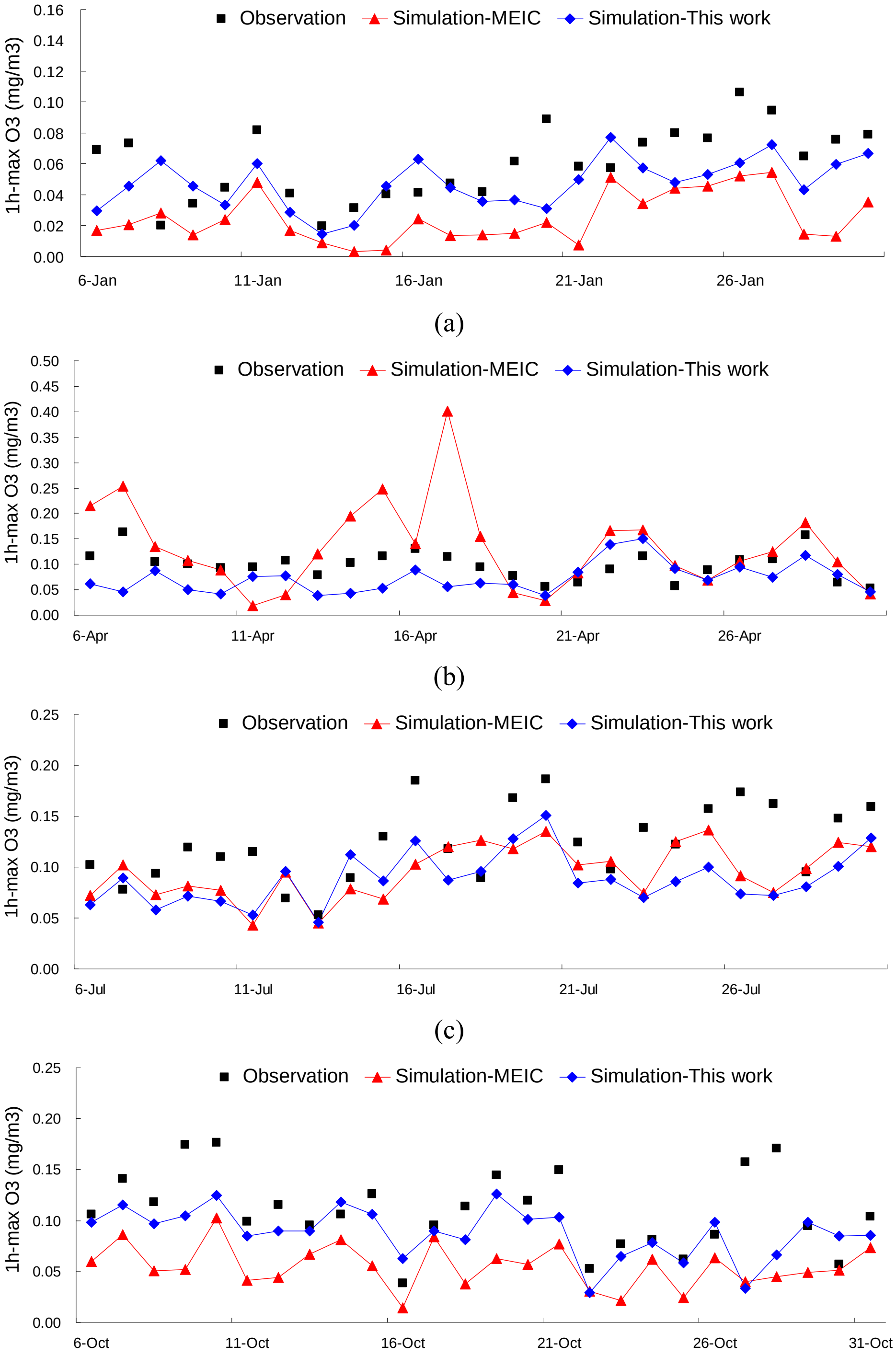
(b)

Figure 9 (continued)



(c)

Figure 10



(a)

(b)

(c)

(d)

2.3 Determination of emission factors

Based on extensive literature survey, the NMVOC emission factors were determined as follows with descending priorities: (1) the results from domestic measurements; (2) the emission limits of local laws and regulations; (3) the values from expert judgment specific for China; and (4) emission factors from AP-42 database (USEPA, 2002) and the EMEP/EEA guidebook (EEA, 2013) when above information was lacking. The emission factors applied in this work are summarized by sector in Tables S4-S6 in the supplement.

The emission factors for power plants and industrial boilers were taken mainly from Bo et al. (2008). Given the similar designs of boilers between China and developed countries (Wei et al., 2008), the emission factors in AP-42 database were also applied when domestic results were lacking. For residential combustion of fossil fuel, the emission factors from domestic tests (Zhang et al., 2000; Wang et al., 2009) were used, as shown in Table S4. For industrial processes, there were very few local tests on NMVOC emission factors conducted before, thus the values from expert judgment and data from AP-42 and EMEP/EEA had to be applied, as summarized in Table S5. For solvent use, the VOC contents of given products were limited by national laws and regulations, as summarized by Wei et al. (2008). The updating of regulations and their impacts on inter-annual changes in NMVOC emission factors were considered from 2005 to 2014, such as GB18583-2003 and GB18583-2008 for indoor painting. Data from AP-42, EMEP/EEA and other literatures (Bo et al., 2008; Fu et al., 2013) were also applied when local information was missing. As a result, the emission factors by source type are summarized in Table S6. Emission factors of oil distribution were taken directly from Wei et al. (2008) and Shen (2006).

For off-road transportation, the data from EMEP/EEA and expert judgment (Wei et al., 2008; Q. Zhang et al., 2009) were adopted, attributed to lack of domestic tests or relevant standards/regulations. Following He (2015), the emission factors for on-road vehicles were calculated and adjusted according to local conditions using Eq. (2):

$$EF=BEF\times\varphi\times\gamma\times\lambda\times\theta \tag{2}$$

where *BEF* is the base emission factor, φ is the environmental correction factor, γ is the average traveling speed correction factor, λ is the vehicle deterioration correction factor, and θ is correction factor for other conditions (e.g., vehicle loading and fuel quality).

The emission factors for biofuel burning in stoves were from the field test results by Wang et al. (2009), who measured the NMVOC emission characteristics of multiple stove-fuel combinations in China and provided the emission factors by province. For biomass open burning, the local test results by Li et al. (2007) were applied, as summarized in Table S4.

2.4 Speciation of NMVOC emissions

The total NMVOC emissions for given source type were broken down into individual species using Eq. (3):

$$E(i,k)=E(i)\times X(i,k) \tag{3}$$

where *E* is the emissions; *X* is chemical profile of NMVOCs (%); *i* and *k* represent the source type and individual NMVOCs species, respectively.

For chemical processes such as rubber, polyether, and polyethylene production, the measurement results of this work were used. The chemical profiles for other sources were mainly taken from domestic measurements, including residential fossil fuel and biomass burning (Tsai et al., 2003; Liu et al., 2008; Wang et al., 2009; Zhang et al., 2000; Wang et al., 2014a), biomass open burning (Zhang et al., 2013; Kudo et al., 2014), on-road vehicles (Liu et al., 2008; Tsai et al., 2012; Huang et al., 2015; Wang et al., 2013; Ou et al., 2014; Gao et al., 2012), iron & steel (Shi et al., 2015; He et al., 2005; Jia et al., 2009; Tsai et al., 2008; Li et al., 2014), paint and printing ink production (Zheng et al., 2013;), solvent use (Yuan et al., 2010; Zheng et al., 2013; Tang et al., 2014; Wang et al., 2014b) and refineries (Liu et al., 2008; Wei et al., 2014).

For sources without sufficient local measurements, results from foreign studies were applied including the SPECIATE database by USEPA (2014), and Theloke and Friedirch (2007).

To reduce the possibly large uncertainty of source profile from individual measurement, Li et al. (2014) developed the "composite profiles" for sources where multiple candidate profiles were available, by revising the OVOCs fraction and averaging the fractions in different profiles for each species. While Li et al. (2014) included the source profiles published before 2011, considerable local measurements were conducted after then. In this work, therefore, "composite profiles" were updated following the method by Li et al. (2014), and the most recent source profiles from domestic results were contained. Table 1 compares the source profiles of this work with those of Li et al. (2014) or SPECIATE for typical sectors, grouped as alkanes, alkenes, alkynes, aromatics, OVOCs, and others. Elevated fractions of alkanes are found in this work for almost all the sources, while the comparisons of other species are less conclusive between sources.

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

$$E(i,m)=\frac{E(i,k)}{M(k)}\times C(k,m) \tag{4}$$

where *E* is the emissions, *M* is the mole weight, *C* is the conversion factor, and *i*, *m*, and *k* represent the source type, individual species, and the chemical mechanism species.

2.5 Uncertainty analysis

The uncertainty of estimated provincial NMVOC emissions for each year were quantified using Monte-Carlo simulation, as described in Zhao et al. (2011). As shown in Table S7 in the supplement, normal distributions were assumed for activity levels, with varied coefficients of variation (CV, the standard deviation divided by the mean) determined for different source types, according to the reliability of energy and economy statistics (IPCC, 2006; Zhao et al., 2011). Due to lack of sufficient measurement results to support the data fitting, the determination of probability distributions for emission factors depended largely on expert judgment (Streets et al., 2003; Zhao et al., 2011; Wei et al., 2011). As summarized in Table S8 in the supplement, the uniform distributions were conservatively assumed for most solvent use sources based on limited data points, while lognormal distributions with large CVs for other sources. Ten thousand simulations were performed to estimate the uncertainties of emissions, and the parameters that were most significant in determination of the uncertainties were identified by source type according to the rank of their contributions to variance.