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Interactive comment on "Improved provincial emission inventory and speciation profiles of anthropogenic non-methane volatile organic compounds: a case study for Jiangsu, China" *by* Yu Zhao et al.

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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 Authors: Yu Zhao, Pan Mao, Yaduan Zhou, Yang Yang, Jie Zhang, Shekou Wang, Yanping Dong, Fangjian Xie, Yiyong Yu, Wenqing Li

We thank very much for the valuable comments and suggestions from reviewer 2, which help us improve our manuscript significantly. The comments were carefully con-

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Discussion paper

sidered and revisions have been made in response to suggestions. Following is our point-by-point responses to the comments and corresponding revisions.

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 405-422 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 attached table. 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.

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.

As some manufacturing technologies we measured are quite unique in China, 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.

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 effects 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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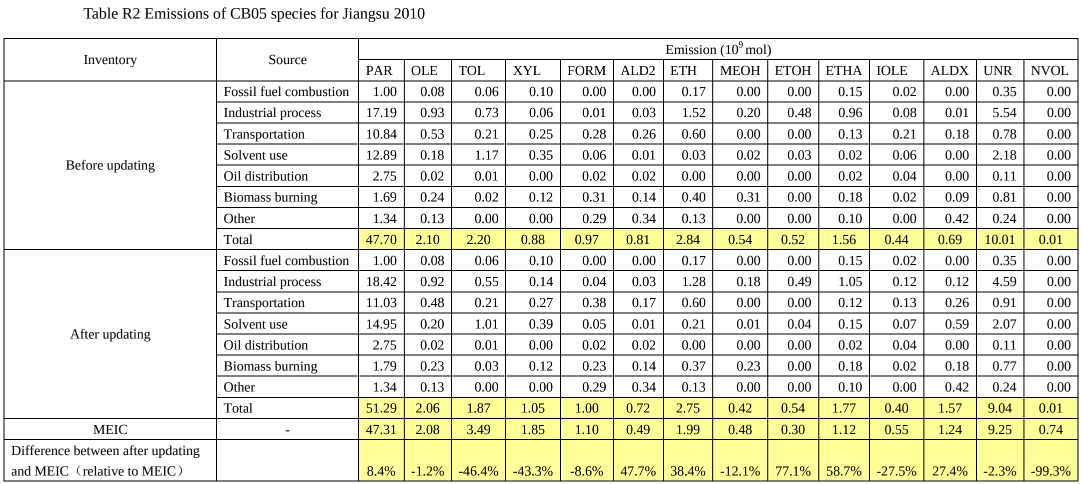

Table R2 Emissions of CB05 species for Jiangsu 2010

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%

Fig. 1.