Responses to Reviewers' Comments

In the revised manuscript, we carefully addressed the comments made by the reviewer and clarified the expressions. For ease of review, our responses (in **blue** text) are given point by point to the comments raised by the reviewers (in **black** text). Also, the changes in the manuscript were marked in **red** text.

Reviewer#1

This study concerns on the solvent use emissions that has been one of the most popular topics in the VOCs emission research. In particular, the volatile chemical products (VCPs) are considered as the emerging source of urban NMVOCs. However, understanding of NMVOCs emissions from VCPs are still lacking in China. This work addressed this important problem by establishing a detailed emission NMVOCs inventory of solvent use (including six VCPs, i.e., coatings, adhesives, inks, pesticides, cleaners and personal care products) for China based on the mass balance technique. The authors found that NMVOC emissions from solvent use increased rapidly from 2000 to 2014 but leveled off thereafter due to control measures implemented on the solvent-related factories in China. Personal care products become an increasing important source NMVOCs. Speciated emissions, OFP and SOAP as well as the comparisons with previous studies are also analyzed in detail. Overall, this manuscript is well presented and within the scope of ACP. The methods are solid, and the results are informative. It can be accepted for publication after the following comments are addressed.

Reply: We would like to thank the reviewer's valuable and helpful comments. We considered the comments carefully with point-to point responses as follows.

1. Line 122-123: Why the authors considered the six types of organic solvent products. Are there any other products that could be the sources of NMVOCs?

Reply: Thanks for the comment. The six types of the organic solvent products, i.e., coatings, adhesives, inks, pesticides, cleaners and personal care products are most widely used in the industrial and residential activities. They are in line with the recent study in the United States by McDonald et al. (2018). Some other solvent products are also used in pharmaceutical production

and cooking. However, their emissions are minor (<5%) compared with those considered in this study (Wei et al., 2008). Therefore, the six types of solvent product with available estimation parameters and activity data are estimated for China.

Reference

Wei, W., Wang, S. X., Chatani, S., Klimont, Z., Cofala, J., and Hao, J. M.: Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China, Atmospheric Environment, 42, 4976-4988, https://doi.org/10.1016/j.atmosenv.2008.02.044, 2008.

2. Line 139: In equation (1), emissions of S/IVOC are calculated. Are the NMVOCs included S/IVOCs in this study? Please clarify across the entire manuscript.

Reply: Yes, the S/IVOC emissions are calculated. In this study, the total NMVOCs in products is divided into VOCs and S/IVOCs, considering volatilization of VOCs and S/IVOCs respectively. This is in line with classification in the previous study (McDonald et al., 2018). To clarify the VOCs and S/IVOCs emissions, we added the related contents as follows: "The total NMVOCs emissions can be divided into VOCs and S/IVOCs according to Equation (1), contributing 93% and 7%, respectively (Figure 4a). Among the solvent use products, pesticides emitted the largest contribution (23%) of S/IVOCs, followed by inks (10%), adhesives (10%), coatings (5%), personal care products (5%), and cleaners (4%). This was because of larger S/IVOCs content ($W_{S/IVOC,i}$>20%) in pesticides compared with other products (Table S7). As pesticides emissions were much smaller than coatings and adhesives (Figure 2), total S/IVOCs emissions were not significant (<10% of total NMVOCs emissions). Nevertheless, estimates of S/IVOCs emissions exhibit large uncertainties because of the lack of local measurements of S/IVOCs content in chemical products used in China."

Please see Page 11 Line 328-336 in the revised manuscript.

Reference:

McDonald, Brian C., de Gouw, Joost A., Gilman, Jessica B., Jathar, Shantanu H., Akherati, Ali, Cappa, Christopher D., Jimenez, Jose L., Lee-Taylor, Julia, Hayes, Patrick L., McKeen, Stuart A., Cui, Yu Yan, Kim, Si-Wan, Gentner, Drew R., Isaacman-VanWertz, Gabriel, Goldstein, Allen H., Harley, Robert A., Frost, Gregory J., Roberts, James M., Ryerson, Thomas B., and Trainer, Michael: Volatile chemical products emerging as largest petrochemical source of urban organic emissions, Science, 359, 760, 10.1126/science.aaq0524, 2018.

Line 146-147: Why only the control of NMVOCs emissions from industrial solvent use are considered? Are there any other control measures implemented for the residential sectors in China?

Reply: For the industrial solvent use, exhaust treatment facilities are required to be installed after the *Action Plan for Air Pollution Prevention and Control* was issued by the State Council of China in 2013. As a result, the percentage of solvent use industrial facilities with treatment devices (C_n in Equation 1) increases quickly in the recent years driven by the national policy. However, for the residential sectors such as personal care and daily cleaner, end-of-pipe treatment are hardly implemented in China. Therefore, control of industrial solvent use is considered in this study. We added the following discussion to clarify this problem: "Only end-of-pipe control of NMVOCs from industrial solvent use is considered in this study, while residential emissions such as personal care products and daily cleaners, VOCs treatment is not implemented in residential and commercial buildings in China."

Please see Page 5 Line 149-152 in the revised manuscript.

Line 367-368: "Ink emissions were much higher in MEIC, while similar results were found for Sun EI and this study (Figure 7b)." What are the reasons for these differences?

Reply: The reason is mainly because different emission factors were used. In the Sun EI, inks are divided into conventional inks and new-type inks, with their emission factors of 750g/kg and 100g/kg, respectively. Our work divided inks into different categories, such as solvent-based inks, water-based inks, and other low-emission inks, with their VOC content (W_{voc}) of about 60%-63%, 13%-23% and 1%-4% (Table S7). Therefore, high-emission and low-emission inks were considered in Sun EI and our study. However, in the MEIC, a universal value of emission factor (540g/kg) was used for ink emissions. This is the main reason why ink emissions were much higher in MEIC than Sun EI and this study. To further elaborate the differences in ink emissions among the emission inventories, we added the discussion in the revised manuscript: "The reason is mainly because low-emission and high-emission inks were considered in both Sun EI and this study, resulting in much lower estimates than MEIC that adopted a high and universal emission factor." Please see Page 13 Line 407-409 in the revised manuscript.

Line 409-412: “Coatings contribute...solvent-based coatings are dominant ...the wide use of water-based solvent…” The solvent-based coatings and water-based adhesives/inks are large. What are their fractions, respectively? Please clarify here.

Reply: We are sorry for this unclear expression. We revised the sentences as “Coatings contribute most to NMVOCs emission, OFP and SOAP reduction because of the dominant proportion (74%) of solvent-based products in industrial coating. In contrast, the reductions of adhesives and inks emissions, OFP and SOAP are minor due to the wide use of low VOC content products, accounting for 82% of total adhesives and 65% of inks.”

Please see Page 14 Line 437-441 in the revised manuscript.

Some other minor comments:

Line 372-374: This sentence is too long.

Reply: The sentence is shortened. It is revised as “Pesticides emissions showed a similar trend between Sun EI and this study, but lower than estimates in MEIC (Figure 7d). There was a significant decrease in 2017 in our work due to that the production of pesticides had decreased and export had increased (Figure S2).” Please see Page 13 Line 413-415 in the revised manuscript.

Line 397: The wording “increase less” is confusing.

Reply: It is removed in the revised manuscript.

Figure 4: The size or positions of the left and right panels can be modified.

Reply: We revised Figure 4 accordingly.

Line 355: “The reasons for …. activity data.” This sentence is not completed.

Reply: It is revised “The reasons for the lower estimates in Bo et al. (2008) and Wu et al. (2016) were mainly due to excluding the adhesive emissions and different methods used to estimate personal care emissions (Table S13).” accordingly. Please see Page 13 Line 394-396 in the revised manuscript.

Line 357: between-> from.

Reply: It is revised accordingly.

Line 394-396: “Compared with 2000…industrial process in MEIC in 2017.” This sentence is not completed.

Reply: The sentence is removed in the revised manuscript.