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 #3

This study is very interesting because it uses a unique approach to estimate NMVOC emissions in China. In particular, the approach of estimating VOC emissions from adhesives is excellent in terms of the points of view.

Reply: We appreciated it very much for the reviewer's valuable comments. We addressed the concerned raised below to improve the quality of our manuscript. Please see following responses.

It would be good to work on deriving the VOC composition from the literature values, as shown carefully in Figures S3 -12.

Reply: Thanks for the comment. Due to lack of comprehensive VOC source profiles for different categories of the solvent use in China, we therefore derived the VOC compositions by combining available literature values to reduce the uncertainty and avoid bias from individual-specific measurement. See response to Reviewer #2, Comment #3 for more details on our VOC speciation procedure.

It is still necessary to examine the details in order to use these results as input data for an atmospheric model, like CMAQ, but I think it will be useful enough for discussions on understanding the NMVOC emissions of solvents from China.

Reply: We totally agree with the reviewer's that the detailed VOC compositions are urgently needed in the air quality modeling, like using CMAQ as mentioned. However, VOC speciation remains largely uncertain because of lacking local China source profiles. Our study is trying advance our understandings on this aspect. Details on the NMVOC compositions of solvent use are discussed in *Section 3.4 Speciated NMVOCs emissions, OFP and SOAP*. This could provide

implications for atmospheric modeling results. Nonetheless, model-ready emission inventories are still needed in our future studies so as to facilitate air quality modeling.

However, with regard to IVOC, it is a pity that only parameters related to IVOC are listed in Table S6.

Reply: Thanks for the comment. We added the results and discussion related to S/IVOCs in the revised manuscript 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.

In addition, authors mentioned VOC source categories that are not included in this study (L359-360). Do the authors not include them in the total amount as reference or reference data? I think that is one of the reasons of the differences between the VOC emissions from the solvent of REAS v3.1 and this study.

Reply: Yes, the source categories such as pharmaceutical production and edible oil production were not included in this study because of lacking estimation parameters such as *Wvoc* for these sources. It could be one of the reasons for the lower estimates of VOCs emissions in this study than other studies. However, these categories are not significant, contributing less than 5% of the total solvent VOCs emissions (Wei et al., 2008). The difference between REAS v3.2 and this study is probably due to the different emission factors used in the estimation. We added the related discussion "The reason is mainly due to higher emission factors used in solvent use (SLV) and paint use (PAIN) estimates in REASv3.2. Some solvent source categories like pharmaceutical production and edible oil production (Wei et al., 2008) were not included because of lacking estimation parameters such as *Wvoc* for these sources. However, their contributions

are not significant (<5%) to the total solvent use emissions (Wei et al., 2008). in the revised manuscript. Please see Page 12 Line 377-382.

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.

There are some other things that I noticed:

"3.1 Control of NMVOCs emissions" :

Please indicate which of the six categories you are considering applying industrial solvent emission control to. Also, please indicate whether the values are uniform or individually set for all applied categories. For coating, what do the authors think of architectural coating for emission control?

Reply: We considered the emission control for sub-categories of industrial solvent use processes, including industrial solvent use included coatings except architectural coatings, inks, industrial adhesives (woodworking, paper converting, shoemaking, fiber processing, packaging, and labelling), and industrial detergents (Figure S7). For the architectural coating, we did not consider the emission control in this study because VOCs treatment devices are hardly used in China. We added the description in the revised manuscript to clarify. "…control of NMVOCs emissions from the solvent use industrial sources, including coatings except architectural coatings, inks, industrial adhesives (woodworking, paper converting, shoemaking, fiber processing, packaging, and labelling), and industrial detergents considered in this study (Table S7)." Please see Page 8 Line 238-245 in the revised manuscript.

L53: Please write the full terms of OFP and SOAP in the abstract.

Reply: The full terms of OFP and SOAP are added in the revised manuscript.

L122-125: I think it is better to write somewhere that "Level 1" refers to the six categories (coatings, inks, adhesives, pesticides, cleaners and personal care).

Reply: We added "Level 1" in the revised manuscript.

L125-128: I think it is better to explain the subcategories in the order they appear in L122-123.

Reply: We explained the subcategories in the same order as they appeared for the first time.

Please see Page 5 Line 125-131 in the revised manuscript.

L161-164: I think the Wvoc listed in Table S6 was calculated by the authors based on Table S1-S5, but they need to mention that clearly.

Reply: Thanks for pointing out the problem. We added the explanation in the revised manuscript as follows: "Table S1-S5 listed the W_{voc} for different sub-categories of coatings, inks, adhesives, pesticides and cleaners, and personal care products, respectively. Taking architectural coatings as an example, the VOCs content of solvent- and water-based coatings are obtained on two national standards (GB) for VOC emission restrictions in China-GB18582-2008 and GB24408-2009. Averages were used when several values are available from different regions of China and data sources. Those categories lacking *Wvoc* were approximated by the values from similar sources."
Please see Page 6 Line 167-173 in the revised manuscript.

Only insecticide is shown for pesticides in Table S4. Table S6 lists Insecticide, Herbicide, Bactericide and Other. How did the authors set the VOC ratio for pesticide components other than insecticide?

Reply: We are sorry that only the *Wvoc* of insecticide is available from the literature. Accordingly, this value was taken to appropriate the VOC contents of Insecticide, Herbicide, Bactericide and Other in Table S7. We added the explanation with "Those categories lacking *Wvoc* were approximated by the values from similar sources" in the revised manuscript.

L163: It would be nice to note that GB18582-2008 and GB24408-2009 are the national standards (GB) for volatile organic compound (VOC) emission restrictions in China.

Reply: We added the explanation in the revised manuscript.

L170: Eqation => Equation

Reply: It is corrected.

L193-194: The authors applied foreign profiles to the products but are the source of the foreign profiles "Li et al., 2014".? Do the authors see "Lie et al., 2014" as a methodology reference? Please write the reference of the foreign profiles.

Reply: Sorry for the confusing expression. We mainly used the foreign profiles from the US (McDonald et al., 2018), and we cited Li et al., 2014 as a methodology reference for combining the local and foreign profiles. We therefore revised the expression as "Source profiles of solvents use used in this study are obtained by combining domestic profiles (e.g., Wang et al., 2014b; Yuan et al., 2010) and foreign profiles (McDonald et al., 2018), following the methods proposed by Li et al., (2014)." The references of domestic source profiles as well as the foreign profiles are illustrated in the Figure S3-S12. Please see Page 7 Line 205-207 in the revised manuscript.

L305: Please write the full terms of OVOC.

Reply: We added the full term of OVOC with "oxygenated VOC" in the revised manuscript.

L323: SOA => SOAP

Reply: It is corrected.

L340: The reference for REAS v3.1 is incorrect. This reference is REASv2.

Reply: Sorry for the inappropriate citation. We revised the reference with "Kurokawa, J. and Ohara, T.: Long-term historical trends in air pollutant emissions in Asia: Regional Emission inventory in ASia (REAS) version 3, Atmos. Chem. Phys., 20, 12761–12793, https://doi.org/10.5194/acp-20-12761-2020, 2020."

Figure 6: This REAS v3.1 NMVOC is the sum of PAINT and SLV, but the decrease from 2014 to 2015 does not seem to be so large. Since the numerical value of REAS v3.2 has been released, I think it is good to replace it. URL: https://www.nies.go.jp/REAS/

Reply: We replace the REAS v3.1 with REAS v3.2 accordingly.

Figure 5: How did the authors decide on the SOAP for unspecified VOCs?

Reply: We followed the method in McDonald et al. (2018) to allocate a value of 0.11 (n-tetradecane) for the unspeciated VOCs. Please also see parts of Table S8 in McDonald et al.

(2018) as follows. We added a list of VOCs and S/IVOCs species and their MIR and SOA yield in Table S9.

No. C	No. O	CARB Compound [a] (surrogate)	TVOC %	R_{OH} %	SOA %	k_{OH} [b] ppb^{-1} s^{-1}	SOA Yield g g^{-1}	log C* [c] µg m^{-3}	Indoor[d] fraction
14	0	n-tetradecane	0.16	0.10	0.48	0.41	0.11 ± 0.02	5.8	0.47
		Unspeciated							
--	--	diesel	1.4	1.3	9.1	0.28	0.23 ± 0.04	6.5	0.39
--	--	non-oxy IVOCs (n-tetradecane)	1.9	0.7	4.3	0.41	0.11 ± 0.02	5.8	0.22
--	--	oxy IVOCs (n-tetradecane)	3.8	1.4	8.5	0.41	0.11 ± 0.02	5.8	0.22
		∑ =	7.2	3.4	21.9				

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.