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 #2

This manuscript presents a long-term NMVOCs emission inventory of solvent use during 2000-2017 in China. Based on a mass (material) balance method, NMVOCs emissions were estimated for six categories, including coatings, adhesives, inks, pesticides, 35 cleaners and personal care products. This paper deal with an important issue of NMVOCs emissions from solvent use, which is a major precursor of ozone and fine particle pollution. Strength of this work is to use of direct activities of VOC-containing product consumption statistics for mass balance-based estimation methodology(eq. 1). Several weaknesses, however, are also found in this paper such as, use of national level statistics, lack of I/S VOC result, non-sector specific control application, and weak discussion in general. I, therefore, think this manuscript need to be improved further to be considered for publication. Followings are my review points and suggestions.

Reply: We would like to thank the reviewer's valuable comments and suggestions. We considered the comments carefully and addressed them point by point as follows.

[Major Comments]

1. Page 5, 12, 13 : Estimation of VOCs emissions using production-consumption data (i.e. mass balance-based estimation) should be more accurate way of estimating VOC emissions, compares to EF-based estimation. The inter-comparison to the various EF-based results (e.g. MEIC, Sun et al., etc.) show higher or lower level of agreement depend on the sectors. I strongly recommend authors to discuss deeper for reason of different agreement levels by sector. If possible, subsector level discussion using Table S13 would help understanding differences among EF-based vs. MB-based approach. I think it would be one of the most important knowledge that this work can contribute.

Reply: Thanks for raising this important issue. The inter-comparison and sub-sector level discussion would help understanding differences among EF-based vs. MB-based approach.

Therefore, we added more discussion on the reasons of differences among the EIs by examining the emission factors, activity data, and source classifications in different studies. Table S13 was also used to facilitate the discussion of subsector-level estimations. Please see some examples of deeper discussion marked in *Red*. Please see Page 12-13 for details.

"Our estimates were peaked in 2014, the same with REASv3.1 whose emissions, however, were much higher. 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). Emissions in EDGARv4.3.2 were significantly higher than our work in early 2000s. However, with much higher annual growth rate of 12% in our work, emissions surpassed those in EDGARv4.3.2 after 2011. Different activity data were used in EDGARv4.3, which was the main reason for the nearly linear increase of solvent use emissions. Compared with the domestic long-term EIs in China, our results were much higher than Sun EI (from 1.6 Tg in 2000 to 5.0 Tg in 2015; 8%) but very close to MEIC (from 2.3 Tg in 2000 to 11.9 Tg in 2017; 10%). The reason for the lower emissions in Sun EI is because of lower EFs, for example, 80 g/kg in Sun EI compared with 620 g/kg in MEIC for architecture interior wall coating (Table S13). Adhesive emissions were not calculated in Sun EI, which was also an important difference. MEIC showed continuously increasing trend after 2014 but a plateau of NMVOCs emissions was found in this study. It is probably because MEIC did not consider the control of NMVOCs in recent years. For the single year estimates, Bo et al. (2008) and Wu et al. (2016) were lower while Wei et al. (2008) was higher than our results.. The reasons for the lower estimates in Bo et al. (2008) and Wu et al. (2016) were mainly due to not including the adhesive emissions, and different methods used to estimate personal care emissions (Table S13). EFs of solvent-based adhesives and inks in Wei et al. (2008) were higher than estimation parameters in our work. Pharmaceutical production and edible oil production were included in Wei et al. (2008) but not in our work. Different types of activity levels and emission factors also resulted in the discrepancy in EIs. In general, different source categories, EFs, and activity data collectively contribute to the differences among the EIs (Table S13)."

Please see Page 12-13 Line 376-400.

2. Page 6 and 10 : Province level emissions would better be presented as a map with provincial VOCs composition graphs on it, in addition to Figure 3. Considering VOCs' atmospheric lifetime, the level of spatial detail(i.e. national level) in emission estimation is limited. Since the top-down allocation(from national to provincial) using the proxy data(Table S7) is not quite accurate methodology, authors need to discuss more on limitations introduced by national level estimation then downscaling, not directly estimated using local(provincial) statistics as in some EF-based estimation.

Reply: As suggested, we added a map with provincial VOCs emissions and compositions in Figure 3.

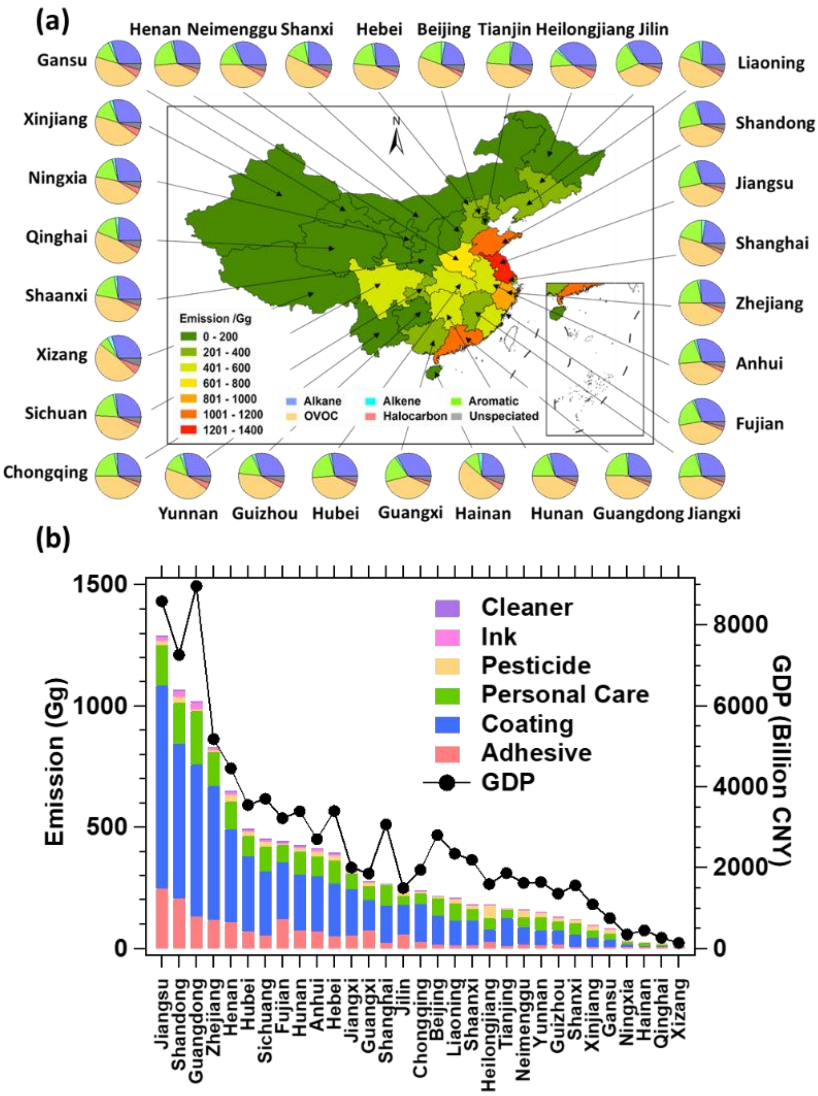


Figure 3. (a) Spatial distributions of solvent use NMVOCs emissions in China and (b) their source contributions in different provinces in 2017**.**

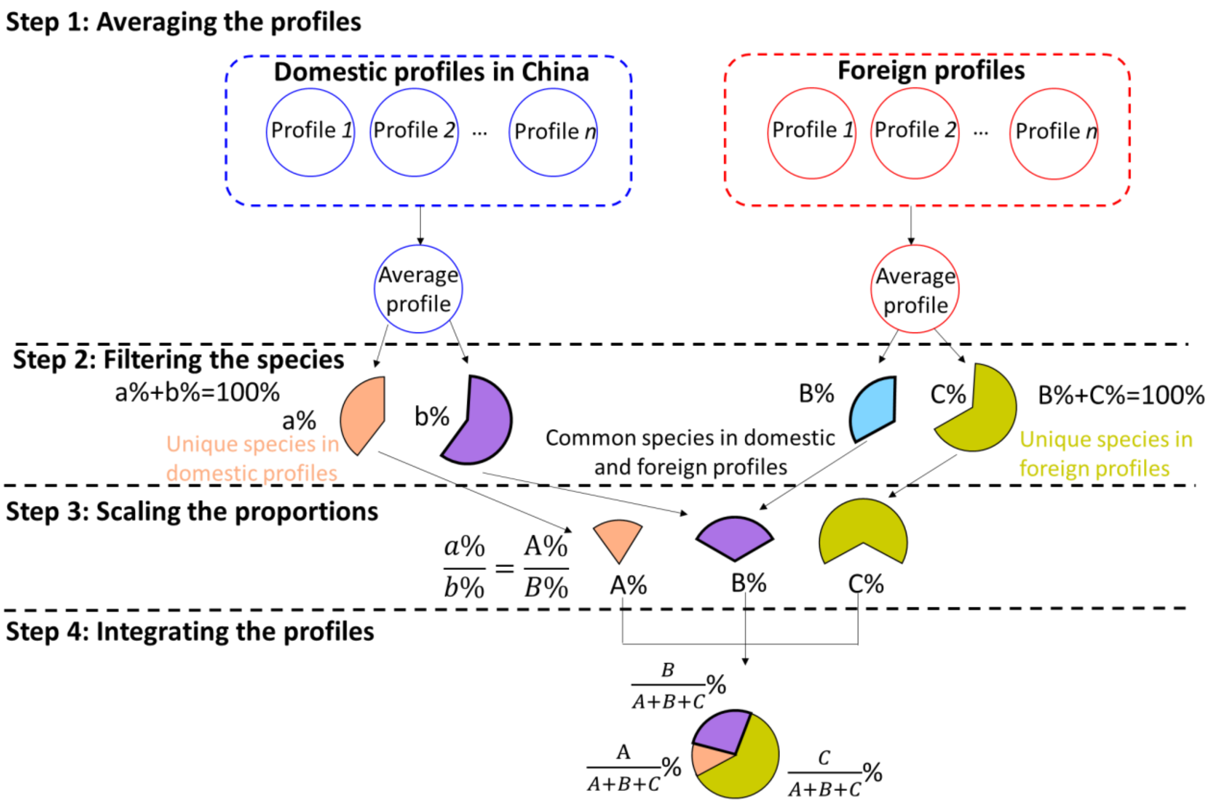
More discussion on limitations of the using the proxy data to downscale from national to provincial emissions were also added in the revised manuscript. "There are limitations of using the proxy data to downscale from national to provincial emissions. For example, the sales value of the solvent products cannot fully represent the locations of solvent use processes. Some products might export from the manufacturing province to other provinces. This introduces the uncertainty in the spatial distribution of the solvent use VOCs emissions. Note that local (provincial) statistics for all the solvent use products are still not comprehensively available in China. Nevertheless, direct estimates using local (provincial) statistics could reduce the errors from downscaling."

Please see Page 7 Line 193-199 in the revised manuscript.

3. Page 7, 10, 11, Text S1 : Chemical speciation should be a very important part of the VOCs estimation. The way they were estimated, however, are not clearly presented and discussed in this manuscript. Authors are required to expand Text S1 and Figure S3-S12 to state more detail data and procedure for the chemical speciation, which can support species-based results in the page 11.

Reply: We extended the description of the procedure and data source for the chemical speciation in Text S1. A flow chart of the step-by-step procedure was added and Figure S3-S12 were elaborated. The more detailed discussion is as follows:

"Source profiles of solvents use used in this study are obtained by combining the domestic and foreign profiles. This compilation of the profiles can make use of the local measurements in China as well as include more comprehensive species, such as S/IVOCs being listed in the foreign profiles. The procedure involves four steps as shown in the following flow chart.



Step 1: A new domestic or foreign profile is formed by averaging weight percentages of NMVOCs groups from multiple source profiles. If some source profiles have OVOC, the treatment of OVOC followed the methods in Wu and Xie (2017) and Li et al. (2014) by averaging the NMHCs and OVOCs proportions, respectively.

Step 2: Common species in the domestic and foreign profiles are identified. Common species may account for different proportions in the domestic and foreign profiles. For example, common species account for b% in the average domestic profile, while B% in the average foreign profile. The remaining unique species account for a% in the domestic profile and C% in the foreign profile. Here a%+b%=100% and B%+C%=100%.

Step 3: We calculate proportion (b%) of common species in the domestic profile, then scale to proportions (B%) of common species in the foreign profile. At the same time, we scale the proportion of unique species in the domestic profile to be A% (=a÷b×B%). Because the foreign profile generally has more comprehensive species and the common species account for a smaller proportion. In order to include more species, we use the proportion of common species in the foreign profile as a reference for scaling.

Step 4: We integrate proportions of common species (B%), unique species in the domestic profile (A%) and unique species in the foreign profile (C%) into a new and complete source profile. Finally, the proportions of unique species in the domestic profile, common species in both the domestic and foreign profiles, and unique species in the foreign profiles are A/(A+B+C)%, B/(A+B+C)%, and C/(A+B+C)%, respectively.

Figure S3-S12 show the procedure and data source of source profiles for architectural coating, furniture coating, automobile coating, other coating, offset printing ink, letterpress printing ink, gravure printing ink, other printing ink, shoemaking adhesive and herbicide. Here, we take the architectural coating as an example to elaborate the detailed procedure for the chemical speciation following the four-step procedure (Figure S3).

Firstly, the domestic profiles of Yuan et al. (2010) and Wang et al. (2014) are averaged to form a new domestic profile, while the foreign profile of McDonald et al. (2018) is used.

Secondly, the common species in the domestic profile and foreign profiles are identified, accounting for 88.8% (b%) and 25.4% (B%), respectively. The unique species account for 11.2% (a%) in domestic profile while 74.6% (C%) in foreign profile.

Thirdly, the proportion of common species in domestic profile (88.8%) is scaled to proportion in foreign profile (25.4%; B%). The proportion of unique species in domestic profile is scaled to be 3.2% (= 11.2%÷88.8×25.4%; A%).

Finally, we normalized the proportions of common species, and unique species in the domestic and foreign profiles to generate the integrated profile."

Please see Text S1 in the Supporting Information.

4. Page 5, 6: Estimation of S/IVOCs are presented in the Eq 1. and 2., but not stated further in the manuscript. Since the importance of I/SVOCs emissions are growing, I recommend adding contents and discussions for them.

Reply: As suggested by the reviewer, we added the contents and discussions for S/IVOCs. The contributions of VOCs and S/IVOCs were added in Figure 4 in the revised manuscript. "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.

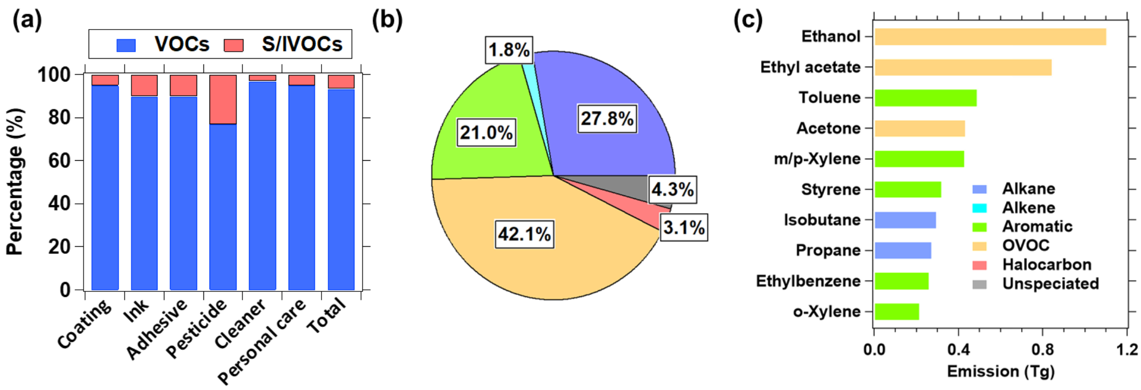


Figure 4. (a) Contributions of VOCs and S/IVOCs, (b) NMVOCs functional group pattern, and (b) the top 10 species in NMVOCs emissions in 2017 from solvent use.

5. Page 8, 9, 13, Figure 2, 9 : VOC emission control stated only for industrial production (i.e. industrial process) in "3.1 Control of NMVOCs emissions" section which is limited. Analysis of emissions control in manufacturing industry would better be sorted by not only control technology (Table S8) but type of industry (i.e sub-sectors). Since much of emissions coming from the consumption process, authors need to add contents and discussion on this process. For example, application of water-based vs. solvent-based paint with respect to national/regional control polices. I suggest to merge control-related contents in the section 4.3 (also, in Figure 9), need to be merged in the section "3.1 Control of NMVOCs emissions."

Reply: We thank the reviewer for pointing out that the emission control would be also sorted by industrial sectors/sub-sectors. However, the information of the control technology for specific sectors are rather limited in China. We have put efforts in looking for more data about the control technology by sub-sector, However, no official and published data are available. We therefore cannot make a more detailed classification at this moment. Nevertheless, more field survey and data collection are needed in our next-step research. Regarding the application of water-based and solvent-based paint with respect to national/regional control policies, as we know, no specific differences are found in applying the control measures in China. This is mainly because control technologies of VOCs emission on solvent use sectors are still developing and not mature in China. As suggested, we added the discussion in the revised manuscript with "control measures are required to be installed for NMVOCs emitting industrial facilities related to solvent use in China. The percentage of solvent use industrial facilities with treatment devices ($C_n$in Equation 1) increases quickly in the recent years. Note that the NMVOCs control technology is still developing and not mature in China. At this time, limited information is available to determine control technology by specific sectors and solvent products." Please see Page 8 Line 240-245 in the revised manuscript.

For the section 3.1 Control of NMVOCs emission, we mainly focus on control measures/technologies on the NMVOCs in recent years due to implementations of the national policy- *Action Plan for Air Pollution Prevention and Control* issued by the State Council of China in 2013. This could also help explain why the solvent use NMVOCs emissions leveled off in recent years (Figure 2). In contrast, in the section 4.3 Implications for NMVOCs control, we are concerning the reduction potential by replacing the solvent-based product with water-based product. Therefore, we make a comparison between two scenarios - Case 1: emissions in 2017, Case 2: emissions in 2017 after solvent-based products replaced by water-based products. This result could give implications for China's policy makers to consider the benefits of using water-based products and in turns the effectiveness in OFP and SOAP reduction. We therefore would like to remain these two parts in the revised manuscript.

6. Page 13: Contents in the "4.2. Comparison with other source" mostly discuss about the cross-sectoral importance changes using other references. I would suggest to shrink and move to introduction and/or conclusion chapters.

Reply: We shrank the discussion of *4.2 Comparison with other source* and moved the content to the *4 Conclusion.* Nevertheless, we kept Figure 8 and moved it to Figure S14 in the Supporting Information. Please see the revised discussion with "Emissions from solvent use grew quickly (with an over five-fold increase) during 2005-2013 and reached a plateau after 2014, which we attribute to the significant industrial expansion in China over the past decades, and effective control on solvent use in recent years (Figure S13). In contrast, combustion and transportation exhibited a decline in the past decade, mainly because of the stringent control of NMVOCs from fuel combustion by industry and on-road vehicles." in Page 15 Line 466-471 in the revised manuscript.

[Minor Comments]

Page 6 : Authors state that "Uncertainty is set to be ±30% if data are directly from official statistics; uncertainty is assumed to be ± 80% if activity data is estimated from other statistical information or reports." How could authors set these values? Form Wei et al, 2011a? Please elaborate more.

Reply: Yes, the values used in this study followed the suggestions by Wei et al. (2011a). They established an evaluation system for uncertainty in activity data, see the table below. We added more elaboration with "We estimate the uncertainty by combining the coefficients of variation (CV, or the standard deviation divided by the mean) of the activity data and the VOCs and S/IVOC contents (Street et al., 2003). According to the accuracy and reliability of the activity data, five-tier evaluation system for uncertainty in activity data was established by Wei et al.

(2011a) as shown in Table S6. We set the uncertainty as ± 30% if data are directly from official statistics and ± 80% if activity data is estimated from other statistical information or reports." in the revised manuscript. Please see Page 6 Line 180-186 in the revised manuscript.

Table S6 Five-tier evaluation system for uncertainty in activity data (Wei et al., 2011a)

Tier	Data source	Uncertainty
I	Directly from official statistics	± 30%
II	Estimated from other statistical information or reports; The data is strongly related; The statistical information or reports is reliable.	± 80%
III	Estimated from other statistical information or reports; The data is strongly related; The statistical information or reports are less reliable.	± 100%
IV	Estimated from other statistical information or reports; The data is less related; The statistical information or reports is reliable.	± 150%
V	The data is less related; The statistical information or reports is less reliable.	± 300%

Reference:

Wei, W., Wang, S., and Hao, J.: Uncertainty analysis of emission inventory for volatile organic compounds from anthropogenic sources in China (in Chinese). Environmental Science, 32, 305-312, 2011a.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S.M., Tsai, N.Y., Wang, M.Q., Woo, J.H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. Journal of Geophysical Research: Atmospheres,108(D21), 8809, doi:10.1029/2002JD003093, 2003.

Page 26: Description for (a) and (b) need to be switched.

Reply: Thanks for the suggestion. We switched the description for (a) and (b).