

Responses to Editor

We are thankful to the editor for the thoughtful comments and suggestions.

We have revised the manuscript accordingly. Listed below are our point-by-point responses in blue to the comments. The modified parts in the revised manuscript are highlighted in yellow.

Editor report on the revised version of acp-2021-136 “Measurement report: Vertical distribution of “biogenic and anthropogenic secondary organic aerosols in the urban boundary layer over Beijing during late summer”

Line numbers refer to the annotated ‘track-change’ manuscript.

We thank the editor’s thoughtful comments. All suggestions have been addressed below.

p. 1, l. 31: replace ‘undertaking’ by ‘undertaken’

Modified. (on page 1 line 31)

p. 2, l. 2: replace ‘radiation force’ by ‘radiative forcing’

Modified. (on page 2 line 2)

p. 2, l. 10ff: These references seem somewhat outdated and should be amended by more recent ones. Please take a look at more recent ones and select those that are relevant to your discussion and should be included, e.g. – but not limited to - (Hodzic et al., 2016, 2020; Huang et al., 2020; Liu et al., 2021; Paiet al., 2020)

We have amended the references. Please see “(An et al., 2019; Hodzic et al., 2016; Nault et al., 2021).” in the revised manuscript. (on page 3 line 11)

p. 2, l. 32: ‘heterogeneous chemical and gas-to-particle reactions’ should be replaced by ‘heterogeneous chemical reactions and gas-to-particle conversion’

Modified. (on page 2 lines 32-33)

p. 3, l. 17: Add ‘good’ or ‘improved’ before ‘air quality’

Modified. (on page 3 line 17)

p. 5, l. 11: ‘...are about half to several hundred meters away from the sampling site.’ – It is not clear what ‘half’ relates to in this context.

We have deleted “half to” and changed this sentence to “Some high buildings are several hundred meters away from the sampling site” in the revised manuscript. (on page 5 lines 11-12)

p. 5, l. 27: 51.1%% - redundant ‘%’ sign

Modified. (on page 5 line 26)

P. 6, l. 5: Define DHOPA

Thanks. DHOPA is the abbreviation of 2,3-Dihydroxy-4-oxopentanoic acid. We have defined DHOPA in the revised manuscript. (on page 6 line 4)

p. 6, l. 7: ‘3-acetuldipic acid’ – please clarify this species name. It does not show up in Tables S1 or S2 and does not seem to be a common name. I assume you might mean acetyl adipic acid? (If so, please also correct Figure 4k)

Sorry for the mistake. We have changed this word to “3-acetyl adipic acid” in the manuscript. (on page 6 line 7). We have also changed Figure 4k. (on page 21)

p. 8, l. 15-22: I am not sure that I understand this new text, in particular the last sentence. If conclusions based on the 2-MT/triol ratio as derived from lab measurements are rather ambiguous, why do the large differences in observed ratios here lead to the conclusion that more vertical SOA measurements are needed? Obviously the 2-MT/triol ratio is not useful in concluding on SOA formation and modification processes.

Thanks for the editor’s comments. The ratios of 2-MTs/C₅-alkene triols from lab measurements are not very useful for concluding SOA formation and modification processes. This discussion in our work is to express the obvious difference of 2-MTs to C₅-alkene triols at three heights and explain the reasons for this difference. However, we didn’t find reasonable reasons to explain this result. We have modified this sentence “Hence, it is still not clear the meaning of the ratio 2-MTs to C₅-alkene triols. However, the large differences of 2-MTs / C₅-alkene triols values at three heights highlight the significance of studying vertical profiles of SOA, and more field investigations are needed.” to “Hence, it is difficult to understand the different ratios of 2-MTs to C₅-alkene triols at three heights, and more field investigations on the vertical profiles of SOA are needed.” (on page 8 lines 19-20)

We hope this expression can be clear to the readers.

p. 9, l. 15: ‘formation of E2’ should be ‘air masses during E2’ (or similar)

This sentence aims to explain the formation of E2. We have changed this to “the E2 is largely influenced by regional transport” in the revised manuscript. (on page 9 line 12)

p. 10, l. 2: Please add a reference to the statement “ASOA is a larger contributor to the loading of SOA and the formation of air pollution in urban areas.”

We have added two references “(An et al., 2019; Fan et al., 2020)” in the revised manuscript. (on page 10 line 31)

p. 10, l. 14: Do you mean ‘biogenic aerosols’ by ‘natural aerosols’?

The “natural aerosols” was used in the reference (Shrivastava et al., 2019), so we used natural aerosols in the manuscript.

p. 10, l. 14-17: It is not clear how this text relates to your findings. Did you also observe higher BSOA formation when anthropogenic factors were enhanced? Are you referring

to effects such as enhanced BSOA yields at high NO_x levels or similar? Make the connection of the literature studies to your findings here clearer.

Thanks for the editor's comments. We found that the anthropogenic tracer DHOPA showed moderate correlations with some biogenic SOA tracers. The concentrations of some BSOA and ASOA tracers increased simultaneously (Figure S6). We referred to these literature studies to suggest the interactions between ASOA and BSOA.

According to the editor's comments, it is likely improper expression. To make the connection of the literature studies to the results in our study clearer, we changed these sentences to "Previous studies have reported that urban pollution can enhance the formation of natural aerosols (Shrivastava et al., 2019); the existence of aromatic compounds can lead to high loading of α -pinene-derived SOA (Shilling et al., 2012; Zelenyuk et al., 2017). These moderate correlations also suggest that the anthropogenic sources are related to biogenic sources, and their interaction mechanisms still need more investigation." in the revised manuscript. (on page 10 lines 12-15)

p. 11, l. 21: 'We found that the fractions of ASOC decreased and Iso_SOC increased for the emission controls' – This sentence is not clear. Should it read '...as a response to the emission controls'? Please clarify.

We have modified this sentence to "We found that the fractions of ASOC decreased and Iso_SOC increased as a response to the emission controls." in the revised manuscript. (on page 11 line 19)

Data availability: Please note our guidelines for Measurement Reports that state that 'The data presented in measurement reports must be openly accessible in accordance with the EGU data policy.'

https://www.atmospheric-chemistry-and-physics.net/about/manuscript_types.html

Please make the data available in a public data repository

https://www.atmospheric-chemistry-and-physics.net/policies/data_policy.html

We have added a way to get the dataset. Please see "The atmospheric particulate matter data used for analysis are available in the Supplementary Material, and the data are also available upon request from the corresponding author Pingqing Fu (fupingqing@tju.edu.cn). The dataset can also be found online (at <https://10.11922/sciencedb.00069>). The DOI will be valid automatically as soon as ScienceDB publishes." in the revised manuscript. (on page 12 lines 22-25).

Responses to Referee 2

The authors have fairly thoroughly addressed the concerns raised in my previous review. I find the revised manuscript more compelling in terms of the conclusions it draws regarding regional vs. local transport. There are a few relatively minor issues noted below, which should be addressed prior to publication.

We are thankful to the reviewer for the thoughtful comments and suggestions.

There remain some typos and odd English, As far as I can tell, none of the authors are from an English-speaking institution, so that is understandable, but it might be useful to a service proofread for English. Examples of these issues include:

The language of this manuscript has been edited by International Science Editing (<http://www.internationalscienceediting.com>).

P1L31: "have been undertaking"

We have corrected it to "have been undertaken". (on page 1 line 31)

P3L1: "It is meaningful in learning"

We have corrected it to "It is meaningful to learn the SOA properties and probe its behaviors in the atmosphere.". (on page 2 line 31)

P3L25: "street road"

We have corrected it to "traffic road". (on page 3 line 25)

P5L11: "half to several hundred meters away"

We have corrected it to "several hundred meters away from the sampling site". (on page 5 line 11-12)

P6L29: "Terrestrial plantations"

We have corrected it to "Terrestrial vegetation". (on page 6 line 28)

P7L11-12: "concentrations of ... concentrations"

We have deleted the second concentrations.

P7L28: "influenced by multi-factors"

We have corrected it to "many causes". (on page 7 line 28)

Minor comments:

P2L3: "IPCC", though a well-known abbreviation should probably still be defined. Similarly "the IPCC report" is fairly informal. Probably the report itself should just be cited, as opposed to a link to the IPCC website.

Thanks for the reviewer. We have defined "IPCC" and modified this sentence to "these

impacts were all shown in the Intergovernmental Panel on Climate Change (IPCC) report (IPCC, 2014).” (on page 2 lines 3-4)

P3L24-25: The phrase "(between 3- and 4-ring)" would probably be very confusing to someone lacking knowledge of Beijing's layout. Maybe changed to "(between the 3rd and 4th Ring Road)" or something like that.

We have modified it to “between the 3rd and 4th ring Road”. (on page 3 lines 24-25)

P5L26: The authors use WSOC and OC concentrations to determine that aerosol are well mixed, then go on to describe differences in composition at each height. Wouldn't differences in composition imply that the aerosols are indeed not completely well mixed? Thanks for the reviewer's comments. We seriously considered the question of the reviewer. We think that it is improper to determine the mixed properties of the aerosols with the average concentrations of WSOC and OC. Hence, we deleted this sentence “indicating that aerosols were well mixed within the boundary layer” in the revised manuscript.

P7L5: The increase in concentration is not uniform, it is driven by some of the species (particularly 2-MTs, I think). It is thus a bit odd to discuss it as a whole since the reasons for it are potentially specific to those species. For instance, if it is 2-MTs (which are almost entirely particle phase) regional transport is likely more dominant, but if it is driven by pinic acid (which partitions based on vapor pressure) then the lower temperatures may be more important.

Thanks for the reviewer's comments. We can feel the reviewer's rigorous attitude towards research. As mentioned by the reviewer, it is a bit inappropriate simply to discuss the vertical properties of the average concentrations. Hence we have changed this sentence “The increase of their concentration with height is potentially linked to the regional transport and gas-to-particle processes of semi-volatile VOCs due to lower temperatures at the upper layers (Goldstein et al., 2009). Moreover, vertical convection transport and BVOCs emission sources cannot be ignored” to “The vertical distribution properties of BSOA tracers are related to complicated factors, such as regional transport and ambient temperatures influencing different BSOA species (Goldstein et al., 2009)” in the revised manuscript. (on page 7 lines 3-4)

P7L9: My original suggestions for 2-MT_{eryth} were not literal, rather I was trying to convey that "eryth" would be a subscript (but indicating this is limited by the textbox into which I enter my report). Using "_eryth" is fairly non-standard, though could be used if the editor is okay with it.

We have changed “eryth” and “threi” to subscript. Please see “2-MT_{eryth}” and “2-MT_{threi}” in the revised manuscript.

P8L13: I don't know the work of Ding et al., but are the authors certain quantification was performed the same way? A difference in 2-MT or 2-MGA quantification (for which authentic standards are lacking) could bias the ratio. It wouldn't impact the

conclusions drawn within this dataset, but it complicates comparisons to other work. The quantification of 2-MT and 2-MGA in our work is different from Ding et al. As the reviewer mentioned that it complicates comparisons to other work. Hence, we have deleted this sentence “The higher values of 2-MTs / 2-MGA in this study than that in a previous study in Beijing at the ground level (average 1.7) (Ding et al., 2014) suggests an efficient reduction of NO_x by the strict emission controls.” in the revised manuscript.

P8L16: I really don't see where in Wang et al. 2005 they suggest C5-ATs are converted to 2-MTs, could the authors please point me toward that claim in that work that they are referencing? Everything I see in there is that C5-ATs are formed from IEPOX (which can also be converted to 2-MTs)

Thanks for the reviewer's comments. The claim in Wang et al. 2005 is that “C5-alkene triols hint at the formation of epoxydiol derivatives of isoprene, which can be regarded as intermediates in the formation of 2-MTs from isoprene.”. We are sorry that we have misunderstood the claim of the reference. We have modified this sentence “C5-alkene triols have been suggested to convert into 2-MTs” to “Both C₅-alkene triols and 2-MTs can be formed from epoxydiol (IEPOX) derivatives of isoprene.” in the revised manuscript. (on page 8 line 14)

P10L27-28: Given the relatively small fraction of OC accounted for the SOC tracer apportionment, do the authors have any thoughts or speculation about what the remaining portion may be? Is it all POA, or is it just due to uncertainty in the tracer apportionment? Relatedly, sesquiterpene and monoterpene SOC are presented as roughly similar - that is fairly unusual in most places, with monoterpenes usually dominating. Do the authors think this is real, or just an artifact of the tracer apportionment? Are there any other papers that have observed this?

Thanks. The small fraction of estimated SOC in OC is attributed to several reasons. The uncertainty in the tracer apportionment is one reason. Another reason is that POC should account for a large fraction of OC. We think the most important reason is that many other sources of SOC are not estimated in our study. Such as 2-methyl-3-buten-2-ol (MBO), which is emitted by pine trees, is a potential precursor of BSOA tracer (Zhang et al., 2012). Some organic sulfates and nitrates in aerosols also account for a large fraction of OC (Wang et al., 2019; Wang et al., 2021).

We think the similarities of sesquiterpene and monoterpene SOC in our study are real. A previous study mentioned that the contribution of sesquiterpene SOC is larger than monoterpene SOC in total suspended particles (TSP) over Beijing (Li et al., 2018). Another study also mentioned that the contribution of sesquiterpene and monoterpene SOC is similar in fine particles in summer over Tianjin (Fan et al., 2020).

References

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