

## Response to Editor

We thank the editor to review our manuscript and particularly for the valuable comments and suggestions that are very helpful in improving the manuscript. Below is our point-by-point response to each comment.

l. 31: 'carboxylic acid with fragmentation' is not clear. Do you mean 'carboxylic acid formation due to fragmentation of larger precursors' or 'fragmentation of carboxylic acids'? Please clarify.

**Response:** Change made. 'carboxylic acid with fragmentation' means the addition of carboxylic acid groups to the site of a C-C bond cleavage, which results in a VK slope of -0.5, according to Ng et al. (2011). To be more accurate, the sentence has been updated in the revised manuscript, which now reads: "Further Van Krevelen (VK) diagram analysis suggests the addition of carboxylic acid groups with fragmentation dominated OA aging in reference days.....".

l. 32: 'was likely transferring the OA evolution close to the processes of addition of alcohol or peroxide groups' is not clear. Do you mean 'likely reflect OA evolution due to the addition of alcohol or peroxide groups'? Please clarify.

**Response:** Change made. In the revised manuscript, this sentence has been updated, which now reads: ".....while the increased aq-OOA contributions during SIA-enhanced periods likely reflect OA evolution due to the addition of alcohol or peroxide groups".

l. 53: replace 'cognition' by 'knowledge'

**Response:** Change made.

l. 60: add 'is' (which is defined as...)

**Response:** Change made.

l. 79: either remove 'are' or replace 'lack' by 'lacking'.

**Response:** Change made. 'are' has been removed in the revised manuscript.

l. 83: replace 'absorption' by 'absorbing'

**Response:** Change made.

l. 89/90: replace 'much consistent with' by 'very similar to'

**Response:** Change made.

l. 117: replace 'gases species' by 'gas species', or remove 'species'

**Response:** Change made. 'species' has been removed in the revised manuscript.

l. 125: Please add reference for the SQUIRREL and PIKA software. To find appropriate citations, please look at [https://cires1.colorado.edu/jimenez-group/wiki/index.php/ToF-AMS\\_Analysis\\_Software](https://cires1.colorado.edu/jimenez-group/wiki/index.php/ToF-AMS_Analysis_Software)

**Response:** We thank the editor's suggestion. The reference for the SQUIRREL and PIKA software has been added in the revised manuscript. The sentence now reads: "The SQUIRREL and PIKA coded in Igor Pro (WaveMetrics) were used to analyze the SP-LToF-AMS data (<https://cires1.colorado.edu/jimenez-group/ToFAMSResources/ToFSoftware/index.html>, last access: 10 July 2022)".

l. 1321: Strictly OM/OC is not an elemental ratio. Better write 'The elemental ratios including oxygen-to-carbon (O/C), and hydrogen-to-carbon (H/C), as well as the organic mass-to-organic carbon (OM/OC) ratio were also analyzed...'

**Response:** Change made. This sentence has been updated as the editor suggested.

l. 150: replace 'increase of' by 'higher'

**Response:** Change made.

l. 163: replace 'transformation of nitrate from gas-phase to particle-phase' by 'partitioning of nitrate to the particle phase'

**Response:** Change made.

l. 201: replace 'much' by 'very'

**Response:** Change made.

l. 217: 'the increase ratio of sulfate or nitrate contribution from reference days to SIA periods' is not fully clear. It might be better to write 'the increased contribution of sulfate or nitrate during SIA periods as compared to reference days ('increase ratio', IR)'

**Response:** Change made. This sentence has been updated as the editor suggested.

l. 239: add 'more' before 'favorable'

**Response:** Change made.

l. 243: replace 'meteorology' by 'meteorological'

**Response:** Change made.

l. 244: remove 'species'

**Response:** Change made.

l. 303: It is not clear what you mean by 'group papers'. Certainly, f44 and f60 were measured

by other groups before – perhaps not at the same location. What do you mean here? Would it be sufficient to shorten the text to ‘(see Fig. 3b, additional data from our previous studies are also shown)’?

**Response:** We thank the editor’s suggestion. It’s sufficient to shorten the text to ‘(see Fig. 3b, additional data from our previous studies are also shown)’, and change made in the revised manuscript.

l. 317: ‘positively’ seems redundant here and could be removed

**Response:** Change made. ‘positively’ has been removed in the revised manuscript.

l. 317, 320: ‘Meanwhile’ is redundant in both sentences

**Response:** Change made. ‘Meanwhile’ has been removed in the revised manuscript.

l. 323: ‘Plots of’ is redundant in a figure caption

**Response:** Change made. ‘Plots of’ has been removed in the revised manuscript.

l. 354: remove ‘The plots of’

**Response:** Change made.

Figure 4: The numbers next to the pie charts are very small. I suggest moving them to a separate panel (4c) and adding the values of the mass concentrations there. It is sufficient in panel a) to write ‘SIA\_P2’, ‘SIA\_P1’ and ‘Reference’ when the additional information is then included in a new, clearer panel c.

**Response:** We thank the editor’s suggestion. In the revised manuscript, Figure 4 has been updated as follows:

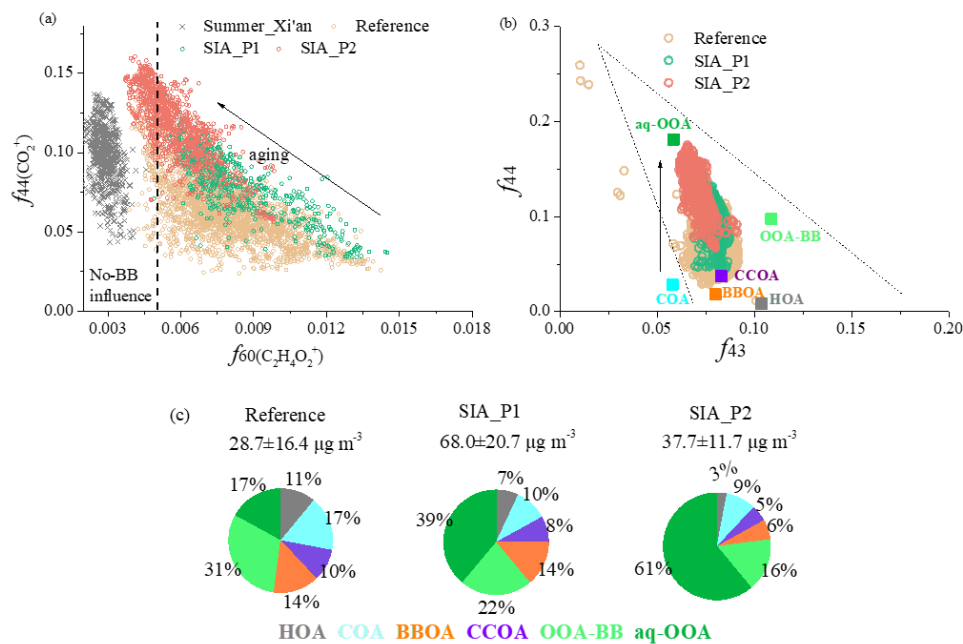


Figure R1.  $f_{44}$  vs  $f_{60}$  during three periods including reference days, SIA\_P1, and SIA\_P2,  $f_{44}$  vs  $f_{60}$  in summer 2019 was also shown for comparison (a),  $f_{44}$  vs  $f_{43}$  during these three periods, the corresponding values of the six OA factors identified in this study are also shown, and the triangle range is from Ng et al. (2010) (b), and the average OA concentration and composition during these three periods (c).

l. 393: replace ‘weak’ by ‘weaker’

**Response:** Change made.

l. 403: specify ‘much more important’ than what? Than during summer? Or than the increase in RH?

**Response:** We thank the editor’s suggestion. The sentence has been updated as: “These results suggest that SIA, compared to RH and ALWC, may play a much more important role in the formation of aq-OOA in winter Xi’an” in the revised manuscript.

l. 422: clarify ‘carboxylic acid with fragmentation (slope=-0.5) dominated OA aging in reference days, and the variation of slope might suggest the transformation of OA from reference days to SIA-enhanced periods, which is likely transferring much close to the processes of addition of alcohol or peroxide groups’ (see my comments to your abstract)

**Response:** Change made. This sentence has been updated to “This suggested the addition of carboxylic acid groups with fragmentation (slope=-0.5) dominated OA aging in reference days, and the variation of slope might suggest the transformation of OA from reference days to SIA-enhanced periods, which likely reflects OA evolution due to the addition of alcohol or peroxide groups (slope  $\approx 0$ ) (Heald et al., 2010; Chen et al., 2015)” according to the editor’s suggestion.

l. 452: Data availability statement: Would it be possible for you to extract the data that were used for your paper (and only those) from the large data base, and provided them in a separate repository? If not, it would be at least helpful if you could give a bit more information in the data availability statement how and where to find the data in the large data base.

**Response:** Change made. In the revised manuscript, we have now extracted the data used for our paper from the large data base, and provided them in a separate repository. The data can be accessible by access and download. This section has been updated as follows:

“Data availability statement: The key data sets are archived at the East Asian Paleo environmental Science Database, National Earth System Science Data Center, National Science & Technology Infrastructure of China ([http://paleodata.ieecas.cn/FrmDataInfo\\_EN.aspx?id=635717f4-2b9a-4edb-9a18-e76497ab9925](http://paleodata.ieecas.cn/FrmDataInfo_EN.aspx?id=635717f4-2b9a-4edb-9a18-e76497ab9925)). For further information please contact the corresponding author (rujin.huang@ieecas.cn)”.