

Response to Referee#3

Thanks to your careful reading and their constructive comments and suggestions on our manuscript. The reviewers' comments and suggestions are shown as *italicized font*, our response to the comments is normal font. New or modified text is **in normal font and in blue**. Details are as follows.

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Referee's comments:

The revised manuscript has addressed the majority of my concerns. Upon addressing the remaining few points below, I recommend it for publication in ACP.

In general, the new text needs to be checked for correct language, preferably by a native English-speaking person.

[response]

Thanks for your comments. Please check our point-by-point response.

General comments:

(1) The conclusions regarding wintertime OS underestimation appear reasonable, but the wording could be softened more to avoid implying representativeness for the entire winter season based on a short campaign.

[response]

Thank you for your suggestion. We fully agree that the representativeness of a several-week field campaign for the entire winter season should be carefully qualified. Our intention was to highlight a significant pattern observed during our study period, not to claim it is universally representative of all winter conditions.

In the revised manuscript, we have carefully the description throughout the manuscript, particularly in Section 4. The key findings in this work, such as the underestimation of Aliphatic OSs and the role of specific formation driving factors as characteristics observed in our winter campaign. This clarifies that the conclusions are specific to the conditions we measured, while maintaining the strength and importance reported in this work.

[revised]

Line 36-37:

The formation driving factors of Aliphatic OSs during the field campaign were further investigated.

Line 41-44:

These results reveal a significant underestimation of OSs derived from anthropogenic emissions in wintertime, particularly Aliphatic OSs, highlighting the need for a deeper

understanding of SOA formation and composition in urban environments.

Line 417-420:

In addition, our field campaigns were conducted in three typical different Chinese cities, the effect of these driving factors on the formation of Aliphatic OSs may not be applicable to other cities with different atmospheric conditions.

(5) Thank you for providing more details about the sampling. For how long were the filters prebaked? What was the brand of the RH-resolved sampler?

[response]

Thanks for your comment. The filters were prebaked for 9 hours under the temperature of 550 °C. The RH-resolved sampler is a home-made instrument, the picture of this RH-sampler is shown in Figure R1. The ambient RH is monitored using an RH sensor connected to this instrument. Before sampling, the switch point is set on the control computer, and the RH sensor measures the ambient RH with the time resolution of 1 second during sampling. The sampling channel is selected based on the range of ambient RH. The real-time cumulative sampling volume of each channel is recorded. When sampling terminates, the cumulative sampled volume for each channel, and real-time ambient RH can be exported for further data analysis.

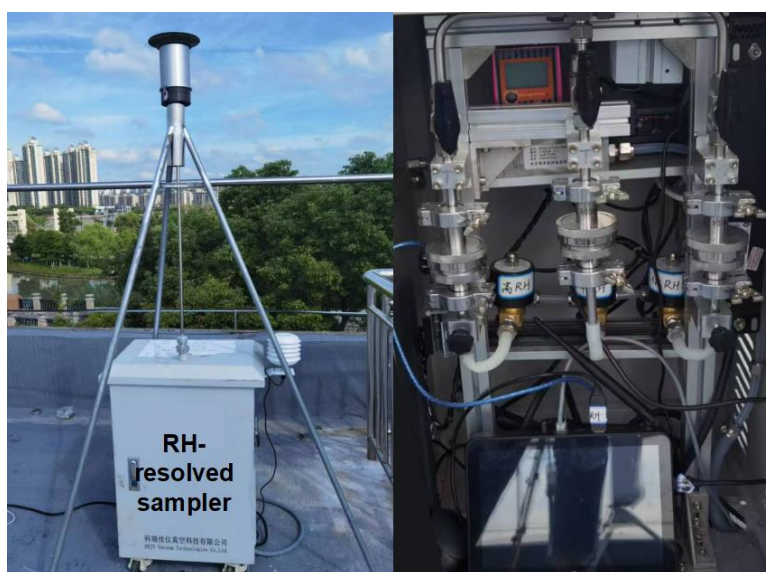


Figure R1 The picture of home-made RH-resolved sampler used in this work

[revised]

Line 108-109:

All quartz fiber filters were pre-baked at 550 °C for 9 hours before sampling to remove the background organic matters.

Specific Comments:

(11) The explanation of site selection based on meteorology and emission characteristics is clear and well justified. But I think the discussion could be further strengthened by briefly clarifying the extent to which the inferred OS formation mechanisms are expected to be generalizable beyond these three cities, and under what conditions they may not apply. This would help readers better understand the broader applicability and limitations of the conclusions.

[response]

Thanks for your suggestion. We agree that clarifying the scope of applicability is crucial. The key driving factors identified in this work, including ALWC, inorganic sulfate, and O_x, are likely operative in other urban environments sharing similar winter conditions characterized by high anthropogenic emissions and moderate-to-high humidity, however, their relative importance and manifestation may differ under contrasting scenarios. In the revised manuscript, we have discussed conditions where our conclusions might not directly apply, such as in summer with strong biogenic emissions, in regions with low aerosol acidity, or in arid cities with persistently low RH.

[revised]

Line 394-399:

It should be noted that though formation driving factors of Aliphatic OSs identified in this work, including ALWC, inorganic sulfate, and O_x, are likely applicable in other urban environments sharing similar winter conditions characterized by high anthropogenic emissions and moderate-to-high humidity. However, their importance may differ in other cities with different atmospheric conditions, like in summer with strong biogenic emissions, in regions with low aerosol acidity, or in arid cities with persistently low RH.

(14) Please note the typo in redisslove -> re-dissolve or re-dissolving

[response]

Revised.

The authors have adequately addressed the remaining comments.

All figures have also been appropriately revised.

In line 23 of the new manuscript "precursor-OS correspondence". It is unclear what you mean, but I assume you just mean "precursor" of the OS.

[response]

We have revised relevant description.

[revised]

Line 25-27:

However, molecular composition, the relationship between OSs and their precursors, and formation driving factors of OSs at different atmospheric conditions have not been fully constrained.