Response to the reviewers

Dear reviewers and editors,

Our manuscript entitled "The shifting of secondary inorganic aerosols formation mechanism during haze aggravation: The decisive role of aerosol liquid water (acp-2022-590)" has been revised according to the comments raised by the reviewers. We are very thankful the comments from the reviewers and editors which largely improved the quality of our manuscript. The changes have been marked in the track-changes file. The detailed and point-by-point response to the reviewer comments were detailed below.

Reviewer 2#:

I think the manuscript is much better than before. However, I'm still not sure why the ratios ($PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$) can be used as the proxy for aerosol hygroscopicity. Are there any previous studies that found this conclusion or that this strong conclusion was promoted by the authors? I agree with the authors that the contribution of accumulation mode particles to ALWC dominated among all the aerosol particle modes. However, the hygroscopicity in $PM_{1.0}$ itself will vary significantly under different environments. They observed significant correlations of ALWC with the ratios ($PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$) in this work. The reason, I guess, is that the secondary aerosol formation mainly happens on these $PM_{1.0}$ particles as the surface area and volume of the $PM_{1.0}$ particles are much larger than those of the coarse particles.

Response: Thank you again for your suggestions. Previous works reported aerosol hygroscopicity is typically quantified by the hygroscopic growth factor (HGF) or hygroscopicity parameter (κ). When the particle size increases from 50 nm to several hundreds of nanometers during haze development, the values of HGF and κ typically increase from 1.2 and 0.15 to 1.5 and 0.3, respectively, significantly correlating with inorganic fractions. The increase in aerosol hygroscopicity will result in the acceleration of aqueous chemistry processes and the haze aggravation. We agree with you that secondary aerosol formation mainly happens on these $PM_{1.0}$ particles as the surface area and volume of the $PM_{1.0}$ particles are much larger than those of the coarse particles. Thus, the larger surface area of $PM_{1.0}$ inherently promotes the partitioning of more inorganic fractions on particles. As a result of the co-effects of thermodynamic stability and higher RH, more ALW is required and presents positive feedback toward more components partitioning on the particulates, which in turn promotes the gradual growth of smaller particle sizes and the haze aggravation. Accordingly, the ratios of $PM_{1.0}/PM_{2.5}$ and $PM_{2.5}/PM_{10}$ presented the potential possibility to index the hygroscopic growth of particulate matter. We greatly appreciate your professional suggestion, which helps us to better explain the results in theory. In the revision, this section has been revised in light of your comments.

References

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