

Responses to the Comments of Reviewer 2

(1) With the abstract, I thought the highlight of this manuscript should be that authors have incorporated ISORROPIA-lite into the chemical transport model therefore letting the 3-D models capable of considering impacts of aerosol water associated with organic aerosol (ALW_org) on the partitioning of semi-volatile vapors which excited me for a while because this is really important, however this was already done in Kakavas et al. (2022). Impacts of increased aerosol water on PM₁ aerosols including their chemical compositions was also evaluated in Kakavas et al. (2022) although focused region in Kakavas et al. (2022) is Europe.

Kakavas et al. (2022) focused on the development of ISORROPIA-lite and the comparison of the predictions of a CTM using ISORROPIA-lite vs. ISORROPIA-II. The simulations carried out in Kakavas et al. (2022) was for one late-spring early summer month in Europe – which is very limited in scope, just enough to showcase the new capability of the model, evaluate its computational efficiency and the potential importance of adding organic water uptake. Most of the related conclusions were tentative given that only one warm month was simulated.

The present work thoroughly analyzes organic water uptake impacts over one simulated year (not just one month as done in Kakavas et al., 2022) and in quite a different geographical area (US here versus Europe in Kakavas et al., 2022). There are significant differences, but also similarities in the predicted changes and effects of SOA water. In the revised paper, we first stress the differences between the two studies but then proceed with discussions on their similarities.

(2) In view of this, this manuscript should advance further the scientific understanding of the significant roles of ALW_org in atmospheric chemistry simulations, however, this manuscript just looks like a report of a sensitivity test of hygroscopicity parameter kappa over United states. The presentations of the results focus only on percentage increase/decrease of PM₁ levels and aerosol water in different regions or sites, no insightful analysis was done. Most importantly, the model performance of SOA simulations was not evaluated against observations at all. It was well known that the performance of current chemistry models in simulating the heterogeneous/multi-phase formations of SOA is not satisfactory (Miao et al., 2020) and might significantly underestimate SOA mass concentrations in regions that SOA formations associated with heterogeneous/multi-phase reactions prevail, therefore numbers reported in this study might not be convincing at all.

We have followed the advice of the reviewer and extended the scientific discussion of the factors that contribute to the increase of the predicted SOA water, both as a fraction of the total but also as absolute concentrations. We also focus more on the specific periods and areas where these increases are predicted.

We do discuss the evaluation of the model predictions for both PM_{2.5} total concentration and composition. Given the small changes on average to these due to the inclusion of the SOA water the PM_{2.5} performances does not change, but it was very good to start with. The performance of PMCAMx regarding annual average OA is actually quite good in these simulations with a fractional bias of 5% and a fractional error of 26% in the 306 stations in the US. For daily average concentrations the performance is also quite encouraging with a fractional bias of 15% and a fractional error of 56%. This information is provided in the revised paper. We do stress now that the major change in the model predictions is on the aerosol water concentrations.

(3) In addition, as demonstrated by authors, the variations of organic aerosol hygroscopicity (Kappa_OA) was also very important, however, the usage of Kappa_OA were not discussed. In general, discussions of this manuscript are very casual, and literature reviews about significant roles of ALW_org in atmospheric chemistry simulations is poor, for example, previous achievements regarding important roles of ALW_org are not discussed at all (Pye et al., 2017; Jathar et al., 2016; Li et al., 2020).

We have followed the advice of the reviewer and extended our discussion and choices of the organic aerosol hygroscopicity parameter providing the corresponding references. We have also added a paragraph about the importance of the aerosol water in atmospheric chemistry simulations.

Specific comments

(4) L40-42, The number of literatures with quantitative determination of aerosol water is relatively small, therefore, following references should also de included here: (Bian et al., 2014; Deetz et al., 2018; Kuang et al., 2018; Wu et al., 2018; Gopinath et al., 2022).

We have added a brief discussion of the previous efforts to measure the atmospheric aerosol water. These include the papers suggested by the reviewer plus a few more from our group.

(5) L48 Li et al. (2019) should be included here.

We have included this reference at this point of the manuscript.

(6) L51 Kuang et al. (2020).

The reference has been added.

(7) L59 Secondary aerosol formations.

We have rephrased this sentence.

References

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