Reply to the 2nd review of “Can Semi-Volatile Organic Aerosols Lead to Less Cloud Particles?”

We would like to thank the reviewer for their efforts in evaluating our submission and providing constructive comments. Please find below our answers to all points raised. The original reviewer’s comments are in black font, and our replies are in blue. Changes in the text are *italicized*.

In this paper the authors discuss results from a box-model sensitivity study comparing changes in parameterized cloud droplet numbers to a representation of semi-volatile organic partitioning. Before consideration for publication, there are a number of issues that need addressing that are raised below.

1. Having read the original paper on the MATRIX-VBS model, I couldn’t tell whether, since you are prescribing aqueous solubility, water is explicitly included in the organic partitioning simulations? If not, there is an inconsistency between prescribing a ’kappa’ value [which will not stay constant unless 100% solubility is assumed] and assuming completely ’dry’ partitioning unless all organics are thus actually assumed to have zero aqueous solubility. If so, please describe how you have accounted for varying solubility, and presumably molecular weight, in the new partitioning simulations. With varying solubility per bin, how would you then account for the influence of one VBS bin on the other in-line with mixing thermodynamics? If you do include water in the partitioning simulations, how might you account for this in equilibrium partitioning at 100%RH? How does the fixed linear change with solubility map to the VBS source VOCs used in a host model? For example, the use of experimentally determined RH variation in Kappa from isoprene and monoterpene SOA experiments has been shown to have significant impacts on two state-of-the-art climate model forcing estimates [Microphysical explanation of the RH dependent water affinity of biogenic organic aerosol and its importance for climate N. Rastak et al. https://doi.org/10.1002/2017GL073056]

All aerosol populations in the simulations (including organic-containing aerosol populations) include water as a component, however, water is not considered in the partitioning of organics. They are also separate processes: one is to get organics on the aerosols due to partitioning, another is to grow aerosols with water. We do not take any Henry solubility into account when calculating partitioning, we use the Pankow parameterization (Pankow, 1994) which does not take into account water. For the cloud parameterization we calculate kappa from the chemical composition of the populations. This is not 100% consistent with each other, but it is the state of the art in global climate models. This would certainly be an interesting question to pursue, but is beyond the

scope of this study. As for kappa per VBS bin, we have a constant kappa per VBS bin listed in Table 1 of the manuscript, and the molecular weight of the VBS species are considered constant.

To include this detail, we have included the following in line 85 of our text: "*....we assumed a linear increase of solubility with decreasing volatility (Jimenez et al., 2009). Since we use Pankow type partitioning (Pankow, 1994), water is not considered in the partitioning process. In addition, we do not use different kappa/RH relationships per organic species, which was found to be important for biogenic SOA (Rastak et al., 2017).*"

4. It would seem the crux of the conclusions rests on the above process description and how the ARG parameterization takes that information to predict cloud droplet number. ARG would not capture partitioning through the humidity life-cycle, so please elaborate on the link between partitioning within the VBS model at any given RH to feeding parameters into ARG.

In the model, partitioning occurs before activation as a distinct, not synchronous, process. During activation when we feed information into the ARG scheme, there's no partitioning anymore. As we mentioned in the previous answer, water is not considered, so activated particles, which are essentially cloud droplets now, will not be part of any further partitioning. Since this is a box model, the activated particles are calculated as a diagnostic variable in the model, not a prognostic one. Our next paper will be a global model study, where this link can be explored further. Please also see response to comment 1 from Reviewer #1.

5. The title is certainly a question worth asking. However I wonder whether results from a model sensitivity study that, whilst interesting, rests on a framework that does not apparently capture process level phenomena which would influence results can be used to deliver an answer. Starting with responses to the questions above, I would suggest the following statement requires re-phrasing: 'We expect that implementing the improved box model in the global scale that includes a two moment cloud microphysical scheme (Morrison and Gettelman, 2008; Gettelman and Morrison, 2015) would more accurately represent aerosol-cloud interactions, which will be our focus on a follow up study. Thus it would offer us valuable insights on how the addition of organic partitioning would change cloud activation in the global atmosphere and its implications for climate.' There is no indication that process representation within this study has improved on any previous. Conflicting implications on process combinations restrict this evaluation.

This model captures process level phenomena for aerosol microphysics with consideration of nucleation, condensation, and coagulation, all of which affect the activated number concentration calculated by the model at any given time. The new model has in addition organic aerosol partitioning, a process previously missing from the original version of the model, adding an extra process that affects aerosol microphysics. However, we will need to use the global model coupled with cloud microphysics to look at process level phenomena that would affect clouds following aerosol activation. We have also re-phrased the statement above with, "*We expect that implementing the improved box model in the global scale that includes a two moment cloud microphysical scheme (Morrison and Gettelman, 2008; Gettelman and Morrison, 2015) would more accurately represent aerosol-cloud interactions, which will be our focus on a follow up study.Thus it would offer us valuable insights on how the addition of process level phenomena in aerosol microphysics, as applied here for the organics partitioning, would affect cloud microphysics in the global atmosphere and its implications for climate.*" in the manuscript.

For the comment that "There is no indication that process representation within this study has improved on any previous. Conflicting implications on process combinations restrict this evaluation.", we would like to point out that this study isn't an evaluation of the old or the new model. The partitioning process is quite uncertain and the VBS framework is heavily tuned against certain measurements. There are a lot of degrees of freedom in the system and it cannot be evaluated properly in any process-level study without a chamber simulation designed exactly for that purpose, which this paper is not trying to do. In order to test model skill for its climate implications, the evaluation should be performed on a global scale, which we are doing at the moment and plan to publish soon. We will evaluate whether this would improve the model or not, but it is important to note that partitioning affects microphysics in complex ways that are not easy to estimate without a model simulation, and that's what makes this study valuable.

6. To more accurately represent aerosol- cloud interactions through an attempt to account for organic solubility and volatility, more detail is needed before publication. The alternative, of course, is to not present this as an improved representation but deliver it as an existing model sensitivity study which might be better suited to publication in Geoscientific Model Development

We would like to emphasize that we do not study aerosol-cloud interactions here, only the initial phase of cloud formation. Again, we do not claim that the new model has improved in terms of results, but it is different from that of the original model. And as

mentioned in responses to the questions above, it has certainly improved in terms of processes.

References:

Pankow, J. F.: An absorption model of gas/particle partitioning of organic compounds in the atmosphere, Atmos. Environ., 28, 185–188, 1994.

Rastak, N., Pajunoja, A., Navarro, J. C. A., Ma, J., Song, M., Partridge, D. G., Kirkevag, A., Leong, Y., Hu, W. W., Taylor, N. F., Lambe, A., Cerully, K., Bougiatioti, A., Liu, P., Krejci, R., Petaja, T., Percival, C., Davidovits, P., Worsnop, D. R., Ekman, A. M. L., Nenes, A., Martin, S., Jimenez, J. L., Collins, D. R., Topping, D. O., Bertram, A. K., Zuend, A., Virtanen, A., and Riipinen, I.: Microphysical explanation of the RH-dependent water affinity of biogenic organic aerosol and its importance for climate, Geophys. Res. Lett., 44, 5167-5177, 10.1002/2017gl073056, 2017.