

Atmos. Chem. Phys. Discuss., referee comment RC1
<https://doi.org/10.5194/acp-2022-648-RC1>, 2022
© Author(s) 2022. This work is distributed under
the Creative Commons Attribution 4.0 License.

Comment on acp-2022-648

Anonymous Referee #1

Referee comment on "Predicted and Observed Changes in Summertime Biogenic and Total Organic Aerosol in the Southeast United States from 2001 to 2010" by Brian T. Dinkelacker et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2022-648-RC1>, 2022

Review of "Predicted and Observed Changes in Summertime Biogenic and Total Organic Aerosol in the Southeast United States from 2001 to 2010" by Dinkelacker et al.

This manuscript describes application of the air quality model PMCAMx to the southeastern United States from 2001 to 2010. The authors describe changes in predicted organic aerosol over the time period in response to their simulated changes in anthropogenic emissions of SO₂, NO_x and NMVOCs, in addition to biogenic emissions of isoprene, monoterpenes, and sesquiterpenes. The authors find that NO_x-dependence and partitioning have the largest influence on the anthropogenic controls on bSOA. It's my opinion this conclusion is not robustly defended and I cannot recommend publication in the present form.

My two most pressing overarching comments:

- The discussion of RH dependence for SOA yield in the Introduction is lacking. Further, the lack of water pathways in the SOA model, and sole reliance on semi-volatile partitioning ... and then finding that semi-volatile partitioning in the predominant

controlling mechanism lacks context and the reasoning is flawed.

To my knowledge, Kamens was the first to show that changing RH in a laboratory smog chamber experiment does not necessarily change the amount of liquid water, and that water is the overriding controlling factor. The Kamens paper cited here has liquid water in the title.

For example, in the cited Nguyen paper, there was no seed in the smog chamber experiments. They found no dependence on RH over a wide range. A dramatic change in RH in those experiments did not change liquid water (except maybe on the walls?). Nguyen went on to find in later controlled smog chamber experiments with inorganic seeds, where liquid water in aerosols did change, that liquid water for bSOA formation had the predominant effect and in fact seemed to be necessary. Water mattered much more so than pH in those experiments.
<https://acp.copernicus.org/articles/14/3497/2014/>

It's odd that the water context is not discussed, in particular because the authors want to argue that partitioning is the dominant controlling mechanism, and because water pathways – neglected in this model application – are expected to contribute substantially to SOA in the region according to Carlton and Turpin:
<https://acp.copernicus.org/articles/13/10203/2013/acp-13-10203-2013.html>

- The authors state in multiple locations that they compared their OA predictions to OA from the CSN and IMPROVE networks. This is highly problematic because neither measures OA, they measure OC. Did the authors mistakenly compare their OA predictions to OC measurements or did they use an OM:OC ratio? If they used a conversion factor what was it and was it constant? It is well established that this ratio is changing. Malm finds that the ratio is increasing.
<https://doi.org/10.1029/2019JD031480>. Hand et al.,
<http://vista.cira.colostate.edu/improve/wp-content/uploads/2019/03/Hand2019.pdf> finds that RH problems in the laboratory are likely impacting mass measurements. Depending on the OM:OC ratio method employed, this could impact OA estimates. The authors provide no information on this, and it is difficult to surmise what potential impacts could be.

specific comments:

Line 86: could be primary

Application of MEGAN: did the land description change at all over the time period and if so, how was this reflected? If not, how is that choice justified?

Starting at Line 185: the authors state: "Anthropogenic VOC emissions decreased... which should cause a decrease in biogenic OA due to partitioning effects." This is not necessarily true. Less anthropogenic VOCs will result in less competition for OH and other radicals. Biogenic VOCs that may have blown out of the domain in the older time period simulations, may have opportunity to react and form semi-volatile species in the newer time period simulation.