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Response to Referee #2

Brian T. Dinkelacker et al.

Author 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-AC2, 2022

(1) *Here the authors explore organic aerosol over the southeast United States, an area well known for especially high biogenic emissions. Using the chemical transport model PMCAMx, the authors simulate two summers, one in 2001 and the other in 2010, and compare conditions and results between them. The effects of various aspects of the overall SOA mechanism are discussed, and in some cases quantified. Manuscript text is clear and well composed, and figures are generally effective. However, while the topic itself is worthwhile and deserving of study, in my opinion this work is fundamentally incomplete, lacking a central question and failing to produce any novel conclusions. I do believe there are opportunities available to further develop this manuscript with additional modeling work and analyses, but in its current form I do not support its publication in ACP. Some specific big picture concerns follow.*

We do appreciate the suggestions of the reviewer and we do agree that the central question of the paper was not made clear in the original paper. This question is: "can a chemical transport model including a complete description of gas-phase chemistry that is standard for CTMs, secondary organic formation based on the volatility basis set (semivolatile partitioning effects, NO_x effects on SOA yields, temperature effects, aging reactions) and interactions between primary and secondary SOA based on partitioning theory reproduce in a satisfactory degree the observed OA changes in an environment that is dominated by biogenic emissions and in which significant changes of all anthropogenic emissions have taken place?". There are of course a lot of other directions that could be pursued in this topic, but we believe that the question is intriguing enough and the insights gained by our work valuable enough for the paper to deserve publication after the recommended improvements.

Our responses and changes to the manuscript (in regular font) follow the comments of the referee (in italics).

(2) *Years modeled: By choosing to examine only two total years, the authors limit their ability to draw meaningful conclusions regarding trends and variability. While it is true that changing anthropogenic emission inventories in particular will show a strong signal when comparing between these years, interannual variability in underlying dynamics must be assumed to impact and confound those anthropogenic changes as well as those of*

meteorology-dependent biogenic emissions. To better understand trends in emissions and resulting OA concentrations, a more robust temporal domain (for example including the years between 2001 and 2010 as well) is recommended to help resolve some of these influences.

More years is always better with this type of simulation work. Unfortunately, there are always tradeoffs that come with increasing the number of years that are simulates, the key ones being the development of emissions inventories, meteorology, and resources (computational and human time). As mentioned, the work is a follow up on the study by Skyllakou et al. (2021) which investigated changes in $PM_{2.5}$ and its sources among 1990, 2001, and 2010. Seeking to explain bSOA changes in more detail, we did a deeper dive into the bSOA predictions and focused on the Southeastern US summer due to the prevalence of biogenics here. 1990 was an obvious additional year to use. Unfortunately, measurements during this time period are severely lacking leaving the corresponding analysis meaningless. We do agree that the investigation of the year to year variability of both OA observations and model predictions is a worthwhile scientific objective. We do believe though that it is beyond the scope of the current work. It could be addressed in future studies with the present or other CTMs. We do not believe that including more intermediate years would change our current conclusions. It would just open another set of issues (e.g., the ability of the meteorological model to simulate a specific summer or the uncertainty in reproducing wild fire emissions in another). A discussion of our choice of simulation years and the effects that it may have on the implications of our work, together with suggestions for future work have been added to the revised paper.

(3) *Role and accuracy of meteorological variability: Meteorology and dynamics are inadequately addressed here in general, with mostly qualitative descriptions covering this extremely influential driver of differences in modeled output. Significantly more work here is necessary to better understand exactly how meteorology is affecting both biogenic emissions and precursor transport. Along these same lines, while significant attention is paid to observational comparisons of modeled aerosol, none is given to the WRF output driving emissions and transport. Considering their significance, and the novelty of the dynamics generated here to drive the CTM, this is a glaring omission and worthy of considerable evaluation.*

We do agree with the reviewer about the importance of the meteorological variability between the examined years for OA in the region. To better address the corresponding effects we first include in the paper an analysis and discussion of the effects of meteorology on the emissions of biogenic VOCs. On the topic of transport, we have performed an additional simulation to quantify its role on the predicted OA levels. In this test we have used the 2001 emissions and 2010 meteorology. Negligible change in predicted bSOA was observed in this test. The WRF performance was similar to corresponding applications in other studies. A more detailed discussion of these issues has been added to the revised paper.

(4) *Significance and novelty of conclusions: Perhaps most importantly, on the whole I really struggle to find a key takeaway message contained here. The model output is well presented, with clear maps and figures comparing the two examined years, but I see nothing surprising or helpful in terms of advancing the state of knowledge on the region or on the modeling of OA in general. The NO_x-dependence of SOA yield is removed as a case study, but the reason and value for this is completely unclear to me. It seems trivially obvious to me that keeping only the low-NOx oxidation pathway would increase yields and overall concentrations, and I see no need to confirm this expected result. A major*

rethinking of what questions these simulations are intended to answer is necessary if this work is to make a meaningful contribution to the SOA modeling literature.

We do understand from the comments of both reviewers that the hypothesis tested by this work and its implications were not made entirely clear. Our central hypothesis can be phrased as: a chemical transport model including a complete description of gas-phase chemistry that is standard for CTMs, secondary organic formation based on the volatility basis set (semi-volatile partitioning effects, NO_x effects on SOA yields, temperature effects, aging reactions) and interactions between primary and secondary SOA based on partitioning theory can reproduce in a satisfactory degree the observed OA changes in an environment that is dominated by biogenic emissions and in which significant changes of all anthropogenic emissions have taken place. The simulations performed provide significant support to the hypothesis.

To quantify the effects that the major processes affecting bSOA concentrations that are currently in the model, we formulated sensitivity tests, including the NO_x dependence of the SOA yields mentioned in this comment. These sensitivity tests are meant to clearly illustrate how these levers are pulled in different scenarios, and their contributions to the bSOA prediction. The study is meant to show that these key levers alone produce consistent predictions of bSOA in the Southeast US in response to changes in both anthropogenic and biogenic emissions, and the sensitivity tests are meant to illustrate how the key processes do their job in the model. Even if the results of a single test may seem trivial to some, we aim for transparency with regards to the SOA formation mechanisms in the model and ultimately seek to identify if additional detail is needed.

To address these issues we have rewritten parts of both the Introduction of the paper and the discussion in the Conclusions in an effort to address the comment of the reviewer.

(5) *Lines 64-65: "Plenty of uncertainty still exists regarding the role of isoprene in SOA formation." This is a confusingly broad and poorly explained statement.*

This is a valid point. We have rewritten the statements with additional detail to increase its clarity.

(6) *Lines 135-145: The relevant bins and species included in the model are difficult to parse. For example, I couldn't tell for sure whether there was one species for monoterpenes and one for sesquiterpenes, or just a single species representing both together. A schematic or diagram would be very helpful here.*

There is a single lumped species representing monoterpenes and a second lumped species representing lumped sesquiterpenes (and of course isoprene is its own chemical species in the model as well). There are several bins then for the SOA products (both in the gas and particle phase) which include the products of the oxidation of the aforementioned VOCs. A more detailed description of this aspect of the model has been added to the manuscript.

(7) *Two days seems to me to be an unacceptably short spin-up time, considering the lifetimes of relevant trace gases and precursors.*

The spin-up time in regional models is usually determined by the residence time of the various pollutants inside the modeling domain and not by their lifetimes. Given the size of

our domain and the average wind speed during the simulation period the two days were sufficient to "wash out" most of the initial conditions and to allow the emissions and meteorology to dominate the predicted concentrations. We have tested this for the Eastern US using the Particle Source Apportionment Technology (PSAT) in which the effect of the initial conditions in each simulation is explicitly simulated. This point is now discussed in the paper and the corresponding references are provided.

(8) *The authors note that IMPROVE measurement comparisons were heavily influenced by fires on several days in the modeled domain. Does this mean that biomass burning in general is not included? If they are included, but simply lacked information on those specific fires, more background information on the inventory used is appropriate. If they are not included, this strikes me as a significant problem that should be addressed.*

Biomass burning is of course included in the simulations. However, there was a major fire near a particular sampling site during the 2001 simulated period, that affected dramatically the corresponding OA observations and also the period average. Our analysis showed that the model failed to reproduce the extremely high OA levels observed. This could be due to an underestimation of the emissions or an error in wind direction (the plume can miss the sampling site in the model). Given that this issue was not related to bSOA, we did not include that specific data point in the evaluation. We now explain in more detail this point to avoid misunderstandings.