Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-1182-RC2, 2021

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of Secondary Organic Aerosol Simulation Bias in the Community Earth System Model (CESM2.1)" *by* Yaman Liu et al.

Anonymous Referee #2

Received and published: 25 February 2021

Review of Analysis of Secondary Organic Aerosol Simulation Bias in the Community Earth System Model (CESM2.1)

This manuscript presents a comparison between simulated organic aerosol (OA) by the Community Earth System Model (CESM2.1) and measured OA from surface and aircraft observations. The authors demonstrate that simulated OA is over predicted in the summer months, likely due to an overprediction of secondary organic aerosol (SOA). The authors perform a suite of sensitivity simulations, turning off one reaction per simulation in the OA chemical mechanism, and conclude that SOA production through monoterpenes is the likely cause of the simulated OA overestimation in the summer.

The authors also note large OA underpredictions aloft (compared to aircraft observations) and moderate underpredictions at the surface in winter (compared to surface observations). Atmospheric chemical models continue to struggle to accurately represent OA. While there have been a number of previous model/measurement comparison studies on OA, the difficulty in simulating OA warrants additional publications. The subject matter in this study is useful and falls within the scope of ACP. However, I have concerns regarding the extent of the analysis and presentation of findings that prevent me from recommending publication at this time.

General Comments:

1. The argument that the model overestimation of OA at the surface and in summer is caused by an overrepresentation of monoterpene SOA production needs to be better substantiated or the limitations of this assertion better discussed. To be clear, I think the authors make a good suggestion by pointing to the large monoterpene SOA burden and the correlation with model bias. However, the correlation with model bias is not enough to make this argument. For instance, the authors point out that isoprene SOA also has a positive correlation with model bias. I don't completely follow why isoprene SOA was dismissed as a reason (Line 385). Isoprene also appears to have a seasonal cycle that peaks in the summer and the authors note the correlation with isoprene and model bias is also positive. Could the over prediction be due to both monoterpene and isoprene SOA yields? Additionally, the authors state in the Conclusions (L 362) that the other POA and SOA components cannot explain the model bias; however, this does not appear to be explicitly shown in the Results section. I do see that monoterpene SOA dominants the OA composition in the summer (and I agree this is a good candidate for the cause of model overestimation), but I do not see a discussion that the other SOA species could not also contribute to the model overestimation. Again to be clear, I agree that the monoterpene SOA yield is a good suggestion for the cause of the model overestimation, but I feel this argument needs more context. This paper could be improved with a better discussion of this argument and its limitations or an additional

simulation with altered monoterpene yields that reduced the model bias.

2. A large limitation of this study is that it focuses only on SOA production as opposed to any other chemical or physical processes in the model. Could the over estimation of model OA at the surface and underrepresentation of OA aloft point to an issue with vertical transport or removal? What about evaporation of OA - is this included in the model? I believe this paper would benefit from an explicit discussion of this limitation and how it affects the results.

3. The sensitivity simulations turn off chemical reactions one at a time; however, these chemical mechanisms are not necessarily linear (or additive). How does this assumption affect your results?

4. The Introduction Section would benefit from further discussion on SOA oxidation and chemistry (including the VBS scheme) as well as a literature review of previous studies focused on model/measurement comparison of OA. Additionally, a number of statements are lacking citations (see Specific Comments for examples). This topic (of OA representation in models) has been explored previously. As I mentioned at the start of this review, the continued challenge of representing OA in models certainly warrants continued study. That said, I believe this study could be improved by including a literature review of previous measurement/modeling studies in the Introduction. I note that the authors do point to and comment on previous studies in the Methods and Results Sections (which is great). However, I think the manuscript would be improved by clearly discussing relevant previous work in the introduction. This would improve the ability of readers to follow the comparisons in later sections. One such example of this, is that Hodzic et al. (2016), which is cited throughout this work, seems to argue for stronger SOA production rates and stronger SOA sinks. Conversely, this study seems to argue for the opposite. This is an interesting comparison that could use more context.

Specific Comments:

1. I do not think an appropriate color scale was chosen for Figure 1. While red and blue are certainly appropriate for opposite ends of a diverging color scale, it is not immediately obvious to me that green should be opposite yellow. I suggest the authors use a standard diverging color scale (or simply shades of red and blue on each side).

2. The units of the color scale in Figure 1 could be more obvious. I suggest including the units as the color bar label itself or at least in the caption.

3. The legend of Figure 4 is much too small to be readable.

4. The panel labels in the caption of Figure 4 are inconsistent. Sometimes the panel label follows the description while other times it precedes the description.

5. The caption in Figure 7 points to the wrong color (I think it should read “black line” instead of red).

6. The units in Figure 7a are never stated. Please be explicit about units in all figures.

7. Table 3 is difficult to read. Could the different regions be grouped in a more obvious way?

8. Please add units to Table 3.

9. This is a minor comment - the citations need spaces after the semicolon.

10. The sentence at Line 51 is unclear. Are you comparing the model representation to other model processes or stating the reasons why model representation of OA is challenging?

11. Lines 51-57 should be edited or revised for clarity. I am not sure what is meant by these sentences.

12. Citations are needed for the comments on OA and climate impacts (such as on Line 41 and Line 60).

13. Citations are needed for the sentences that begin on Line 49, 50, and 52.

14. Line 80/81 - I think the papers by Donahue et al. (2006) and Robinson et al. (2007) should be included in the citation for the VBS.

15. Line 85 - minor typo with the comma

16. Line 124 - Could you elaborate (briefly) on what a “specified dynamical” simulation is? In addition, please define “FCSD” and “FC2010climo”.

17. L 152 - I do not think the word choice of “critical” simulation bias is correct. Do you mean “large” or “substantial”?

18. L 173 - I suggest changing “descending” trend to “decreasing” trend.

19. L 173 - I think this is a really interesting point that needs a little clarification. Do you mean that the increasing wildfires are leading to increasing trends in observations but the wildfire emissions are not included in the model and so the trend is not represented?

20. L 184 - Is the current study not also influenced by the bias of evaporation of OA off filters as in Hodzic et al. (2016)?

21. Line 321 - Is this an entirely new model configuration and simulation? If so, I recommend including this model configuration in the Methods Section. If I understand correctly, this simulation includes SOA production schemes that were suggested as part of the model bias in the previous paragraph. This seems like an important result that should be given more discussion.

22. Line 356-357 is confusing. Is the second parenthetical placed correctly?

23. Line 368 states “...and photolytic removal processes might be too strong”. I do not follow why this is an argument in support of monoterpene SOA production being too high. It seems like it argues that bias is not entirely due to SOA production rates in contrast to the point of this paragraph.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1182, 2020.