Comment on “Modeling organic aerosol concentrations and properties during winter 2014 in the northwestern Mediterranean region” by Chrit et al.

acp-2018-149

General Comments:

Chrit et al. have deployed the Polyphemus platform with expanded techniques for simulating organic aerosol formation and aging, particularly from residential heating sources, and applied it to the CharMEx campaign in winter 2014. The study design is generally sound and the sensitivity choices are informative. There is also adequate reporting of direct results. However, I found there to be a lack of further diagnosis or interpretation of results considering the amount of data the authors would have access to from the model. Considering this, I think the paper would be better suited in its current form for GMD, although I think it is of acceptable quality and scope for publication in ACP, given that the authors address specific concerns below.

Specific Comments

1. Section 3.4: I did not understand how profiles 1 and 2 are allocated to individual emissions. It should be made clearer why profiles 1 and 2 can be mismatched for volatility and O:C? Usually, I think of these as tied together by the molecular weight (i.e. carbon number) assumed for each model species of a given volatility. Are the assignments made using some feature from the emissions inputs, or are individual sectors assigned profile 1 or 2 based on some knowledge of their emissions (e.g. waste burning goes with profile 2, offroad diesel goes with profile number 1, etc)? If the latter, can the authors include a table that identifies these assignments? If the former, can the authors better describe what parameters and algorithm are used to make the assignments?
2. After going back to Ciarelli et al. (2017b), I am not convinced they included additional IVOCs, consistent with those being added in this simulation, in their parameterizations. It seems from Table 1 in that paper, that the authors included NTVOCs and also evaporated the existing POA into SVOC and IVOC bins. But I do not think they considered an additional IVOC category. Given this, I am not surprised that simulations here which include additional IVOCs and NTVOCs (S4 and S6) tend to overpredict measurements. I would suggest the authors perform at least one run with R_RH set to 0 for residential heating sources and NTVOC turned on. This will probably look a lot like S5 so if the authors want to adjust the explanation of their cases to avoid doing more simulations, I think that is okay, but some detailed explanation should be added (i.e. R_RH could be defined as adding NTVOC). Note this approach would not be perfect, because the SOA yields for the IVOCs would differ from those Ciarelli et al. (2017b) derived for NTVOC.
3. Are the IVOCs from residential heating assumed to be the same composition (i.e. same SOA yields) as those from vehicle sources? If so, what is this based on?
4. Do the authors have a sense for the variability of wood-burning fuels across the region and how well one volatility distribution would be at simulating their emissions? Are there varying practices for controlling emissions from chimneys or flues that would have an impact on the particle fraction from these sources?

5. The authors make the point that the winter time conditions are not favorable for oxidative aging of SOA or high formation of SOA from VOCs. However, the measurement data show relatively high O:C, out of reach of the model sensitivity cases. Can the authors demonstrate the model's performance for relevant gas-phase oxidants to eliminate that as a factor?
6. I would urge the authors to consider adding more analysis of the relationship between model error and individual sources or chemical descriptions in the model. Are there correlations with other model species that would give some clues as to where the parameterizations are weak or better emissions data are needed (e.g. CO, POA, NOx, etc)? What recommendations do the authors have for future work by experimentalists and other chemical transport model efforts? What pieces of the model description need the most work? One conclusion that comes out is that the results are more sensitive to the volatility distribution than the aging mechanism. I wonder if the authors could emphasize this point as an area in need of further research? Does more work need to be done on constraining the volatility, or on representing the diversity of wood burning fuels and conditions that exist?

Minor Issues/Typos/Suggestions

1. Page 1, line 10: Suggest replacing "whatever the parameterizations" with "in all parameterizations tested".
2. Page 2, line 3: Suggest replacing "primary fraction originates" with "primary fraction originates mostly"
3. Page 2, line 5: evidences should be evidence
4. Page 2, line 8: I think the generally acknowledged IVOC range includes 10^3-10^6 while SVOCs are 0.1-10^3.
5. Page 1, line 16: Add "precursors" to read "main anthropogenic VOC precursors".
6. Page 3, lines 19-22: The 2D-VBS can also accommodate oligomerization pathways, although most transport models don't take it into account.
7. Page 3, line 23: suggest rewording to "scheme that accounts for multigenerational ageing, including functionalization and fragmentation, and that…"
8. Page 3, line 35: Recommend the authors add more description of what the non-traditional VOCs are. In the past, the word nontraditional has been used to identify SOA from IVOCs and SVOC vapors. I was confused at first, but see from the sensitivity case descriptions that these NTVOCs are different compounds.
9. Page 4, line 8: Are studies from 2001 and 2005 still recent? Obviously, this is the authors' call. Maybe everything after 2000 still 'feels' recent? It's certainly more recent than 1975.
10. Page 5, line 11: Are the authors using ISORROPIA v1? Version 2 includes among other things interactions with crustal species. If the model includes version 1, a statement should be added explaining either the unimportance of dust sources during the campaign, and/or the unimportance of crustal cations on organic aerosol concentrations as they are modeled here. The output of ISORROPIA will affect things like water uptake and pH, but most OA models now probably aren't sensitive to parameters like these, at least first- or second-order. Is that true for this model as well?

11. Page 5, line 15-16: The authors reference Chrit et al. (2017) for their grid configuration details, but I think it would still be useful to put it here. What is the grid resolution and layer resolution of the nested and large domains?
12. Page 5, lines 28-30: Is the total [I/S-VOC + POA] equal to 2.5 or 1.5 times the original POA? Could the authors adjust the wording of this sentence to make this clearer?
13. Page 6, line 9: remove "the" to read "at the model cell closest to the station"
14. Figure 1: Could the authors adjust the color scales so it's a bit easier to assess them in relation to each other? For example, 0.01 for the left and 0.05 for the right?
15. Page 7, line 9: Suggest changing "different parameterizations are compared" to "different parameterizations, described in the following sections, are compared".
16. Page 7, line 16: How are the saturation concentrations for the S/I-VOCs chosen? Are they from a previous study? Are they fit to something?
17. Table B1: Why is it that for the SOA vs. POA species, the enthalpies of vaporization are the same even though the molecular weights are higher, the saturation concentrations are somewhat lower and the O/C ratios are somewhat higher? I would guess the SOA species should have larger enthalpies of vaporization.
18. Tables D1 and D2 look to be repeated?
19. Page 11, line 6: Should "SOA" be "POA"?
20. Table 5: What is the uncertainty reflective of? One standard deviation?
21. Page 20, line 24: The authors have cited May et al. 2013a (biomass burning emissions) twice.
22.