

Interactive comment on “The impact of biogenic, anthropogenic and biomass burning emissions on regional and seasonal variations in secondary organic aerosol” by Jamie M. Kelly et al.

Anonymous Referee #2

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Kelly et al. update a global chemistry-climate model (UKCA) for new sources of secondary organic aerosol (SOA) and report on model predictions and the model-measurement comparison from this updated model. They find that, on average, the inclusion of new sources of SOA improves model performance against organic aerosol (OA) mass concentrations, POA-SOA splits, and OA vertical profiles but caution that the model still does not include some major SOA formation pathways and processes (e.g., varying volatility, aqueous chemistry) and remains unconstrained in the southern hemisphere due to a paucity of observations.

Kelly et al. have done an excellent job of reporting results from the model simulations

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and the model-measurement comparison. The manuscript is also very well written and makes for very easy reading. The findings and discussion from this work will be helpful to the community. However, I found that the methods used did not reflect the gaps/uncertainties discussed in the introduction nor come close to the current state-of-the-science for treating SOA formation pathways and processes in atmospheric models. While novel for the UKCA, the sources/formation pathways explored in this work are routine for some of the other global chemistry-climate models and I was struggling to see how this work was novel and offered fresh insights into the SOA budget that haven't been explored in earlier work. This work definitely needs to be published but I am not comfortable recommending publication in Atmospheric Chemistry and Physics given the lack of novelty.

I have listed just a few of my concerns related to the methods:

1. The POA and SOA need to be treated as semi-volatile and reactive to better model the OA mass concentrations and its sensitivity to chemistry and changes in temperature. One example where this would influence one of the findings reported in this work is that SOA monolayers coated on POA could evaporate with dilution/chemistry and transition POA back into its hydrophobic mode. Another complication related to treating the semi-volatile nature of OA that has surfaced recently is if organic particles achieves instantaneous equilibrium with the organic vapors and how the phase state (i.e., diffusion limitations within the particle) might influence the timescales to achieve equilibrium. A semi-volatile treatment should be trivial to include with say a 2-product model.

2. There is plenty of evidence that the chemical lifetimes and SOA mass yields are very different for different SOA precursors (in addition to being a function of the OA mass loading), which can be very easily reflected in this work (regardless of whether the OA is treated as semi-volatile or non-volatile). Furthermore, SOA mass yields need to be corrected for vapor wall loss artifacts experienced in chamber experiments.

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3. Emissions of semi-volatile and intermediate-volatility organic compounds contribute significantly to anthropogenic SOA precursors (and possibly even biogenic SOA precursors) and need to be explicitly modeled. These might help reduce the under-predictions in urban areas and elsewhere. On a related note, it is unclear to me how the model-measurement comparison at urban locations needs to be evaluated. What fraction of the under-prediction can be seemingly attributed to the model resolution?

4. Globally, aqueous processing of organic compounds in aerosol water and clouds is probably a very important source/sink of OA and needs to be included. At the very least, one needs to consider IEPOX, glyoxal, and in-cloud formation of a few dominant organic acids in the model.

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