

Review of Zheng et al.

This work looks at the modeled vs. observed trends in sulfate and organic aerosol in the southeast US and finds that the model OA is overly dependent on sulfate, due to the impact of sulfate on acid-catalyzed reactive uptake of epoxydiols. The authors use the month-to-month variability in OA during summers to provide a new constraint on SOA formation mechanisms. Overall, this paper is well-written and within the scope of ACP and should be published after minor revisions.

Major comments.

1. Page 8, line 151 – The statement “The 2°x2.5° simulations are adequate when modeling continental boundary layer chemistry (Yu et al., 2016)” is true for ozone and NO_x, but not for isoprene oxidation pathways. Yu et al. showed a large shift from high-NO_x to low-NO_x oxidation (Fig. 3), which would impact IEPOX production and SOA from isoprene. Please address this limitation of your work, and how an overestimate in the high NO_x pathway would impact your results. Possibly a small sensitivity study is warranted to address this issue. Could it be that the model actually would have even more IEPOX SOA at high resolution and the problem would be worse?
2. Is there a reason you didn't use observations past 2013?
3. Could you comment on how the lack of nonvolatile cations in ISORROPIA might impact your calculated aerosol acidity and generally effect your results?
(<https://pdfs.semanticscholar.org/f642/9d2c07179b7624c795aed5bf37b20aa0e2a8.pdf>)

Minor comments.

4. Page 1, line 34 – The statement “Biogenic SOA may account for 60-100% of OA” is confusing, if you are going to call it biogenic SOA, but then discuss anthropogenic influence, maybe call it something else, like biogenic-derived SOA or similar.
 - a. Also, Kim et al., 2015 say that biogenic isoprene and monoterpenes account for 60 % of OA, where do you get the 100% value?
5. Page 5, line 70 – What about the role of NO_x (and nitrate) in regulating aerosol water?
6. Figure S1 – it is implied that the CSN sites are in Figure S1, which only shows SEARCH and IMPROVE.
7. Page 7, line 126 – Where does the baseline model agricultural NH₃ come from?
8. Page 7 line 126 – Do you mean you scale the emissions from 2011 backwards to 2000 and forwards to 2013 using the annual emission totals from EPA? What do you mean by “mapped over 0.1°x0.1°”
9. Page 8, line 134 – The GEOS-Chem complex scheme is also described in great detail by Pai et al., 2020. “An evaluation of global organic aerosol schemes using airborne observations”
10. Page 8 line, 148. It is confusing to dive in and describe sensitivity studies without showing us the problem in Figure 1. I don't think it would be out of place to say that the default model, shown in Figure 1, overestimates the trend in organic aerosol largely due to ... and we run 4 sensitivity studies to address this discrepancy...
11. Page 9, line 154 – It would be helpful to tell the reader that these sensitivity simulations will be described in Sections 2.2.2 etc..
12. Page 9, line 163 – do you mean at “the” or “from the” SOAS2013 campaign (citation?)
13. Do the authors expect that not including non-volatile cations in ISOROPPIA could impact their findings?
14. Page 10, line 178 – Can you provide some evidence/citations for us that there might be such a coating effect? Or is the ethylene glycol supposed to be representative enough of real organic aerosol coating ammonium bisulfate? Please just clarify a little more here.
15. Page 10, line 189 - Please elaborate a little more on the findings of Jo et al., 2019 since this seems important.
16. In Section 2.2.3 – How do your emissions results compare to the NH₃ emissions derived from wet deposition in Paulot et al., 2014? *Paulot F., Jacob, D.J., Pinder R.W., Bash J.O., Travis, K., Henze D.K., Ammonia emissions in the United States, Europe, and China derived by high-resolution inversion of ammonium wet deposition data: Interpretation with a new agricultural emissions inventory (MASAGE_NH3), J. Geophys. Res., 119, 4,343-4,364, 2014.*
17. Page 13, line 241 – Do the sulfate trends refer to Figure 2? If so, please reference it here.

18. Page 11, line 199 – Is this due to improved farming practices as output increases? Or some other explanation? I am surprised NH_3 is not increasing and would assume other readers might appreciate a little more detail.
19. Section 2.2.3 – It would help the reader if there was a plot of the bottom-up and top-down NH_3 emissions.
20. Page 11, line 208 – What surface observations? Please clarify what you mean, are there separate independent NH_3 surface observations you compare against? Again, a plot would be useful here. Same for the seasonal cycle. These can be in the supplement according to the author's preference.
21. Page 11, line 217 – For clarity, please specify that these are “model” IEPOX-SOA, sulfate aerosol etc.
22. Page 13, line 244 – Why not sample the model at the locations of the sites? Would that make a difference to your trend?
23. Figure 1 – can you explain the periods of enhanced OA around 2006 and 2011?
24. Figure 2 – It could be helpful to include the trends on this figure similar to Figure 1.
25. Page 15, line 302 – Figure 3 is difficult to interpret, please provide a little more description of what is shown here, particularly what is meant by “divided by 1 standard deviations.”
26. Page 16, line 325 – Is isoprene interannual variability explained by temperature? Temperature is an important driver of aerosol partitioning, should it not be included in Table S1 and Figure 3? Does temperature explain any of the interannual variation in Figure 1?
27. Page 18, line 376 – I assume that the H^+ value during 2013 SOAS was calculated with a thermodynamic model, although constrained by observations. Please clarify.
28. Page 19, line 378 – Can you comment on why the model agrees much better with SOAS2013 observations and 2013 OA in Figure 1, and what the limitations are of testing your revised IEPOX uptake at the low end of the SO_2 emissions trend?
29. General comment – why not consider trends in ammonium? Particularly to help support your improved NH_3 emissions scenario?
30. Page 22, line 467 – By gas-aerosol partitioning, do you mean reversible uptake of isoprene oxidation products? Could you be more specific on this?
 - a. Generally, the final paragraph that goes through potential reasons for the remaining model discrepancies is very useful but needs additional explanation. The sentence starting “Despite a large MMV in IEPOX-SOA is confusing, please rephrase to more clearly state your meaning.
 - b. Is monoterpene SOA included in the GEOS-Chem simulations? If so, is the lack of MMV captured by the model? I am generally just confused by the discussion here. Are you trying to say that other SOA pathways, not in the model, might have less MMV and therefore improve model performance of the long-term trend?