

Interactive comment on “Characterization of Organic Aerosol across the Global Remote Troposphere: A comparison of ATom measurements and global chemistry models” by Alma Hodzic et al.

Anonymous Referee #3

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Hodzic presents a comparison of a large set of global models and observations from the free troposphere with a focus on organic aerosol. I applaud the authors on this large undertaking in terms of number of models and synthesis of observations. The manuscript contains a large amount of information. My major comments are regarding the POA estimation and better illustrating the utility of DYN configuration (which may just be a bit buried).

Major comments:

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1. Section 3.2 and POA estimation: Can the uncertainty in the POA estimates be quantified, ideally with error bars (e.g. in Fig 6 vertical profiles or Fig 8 POA)? Some ideas to consider regarding the POA estimation:

a. There are anthropogenic POA sources that do not have significant amounts of BC. See, for example, Figure 2 of Reff et al. (<https://pubs.acs.org/doi/10.1021/es802930x>) which indicates large emissions of dust associated with anthropogenic activity (road dust, construction dust) that have high OC relative to BC. While fossil fuel-BC may be a general proxy for anthropogenic activity, is the ratio 1.55 reflective of that general behavior? What does the EPA NEI indicate as FFratio including all anthropogenic PM sources (e.g. Fig 2 of Reff et al.)?

b. Can you plot data (obs or model) as a function of $f(\text{BB})$ influenced to get an intercept at $f(\text{BB})=0$ and $f(\text{BB})=1$ for comparison to FFratio and BBratio specified in text?

c. For the calculation of observed POA, should there be small amounts of OA associated with sea spray and dust? You have sea salt and dust observations in Figure 2b and elsewhere that could be used to estimate that POA.

d. Would it be better to label the estimate from equation 1 as “combustion POA”? I would assume models are also more specifically combustion POA?

e. Is non-differential removal of BC and POA appropriate if POA is semivolatile? Consider that near a fire, concentrations could be high enough for IVOCs to be partitioned in to the particle. As the air mass is diluted, POA will decrease more rapidly than a conserved tracer. This may explain why the larger BBratio gives POA >100% (line 378).

f. For model estimates of POA, is hydrophilic OC considered POA or SOA? Can models just label the hydrophilic OC as “SOA” and get the right properties for endpoints (health, climate) of interest?

g. Did you consider using a CTM to verify the method in equation 1 or back calculate

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the model effective FFRatio? Does it reproduce the model POA (hydrophobic OC)?

h. BC is chemically aged. Do measurement techniques measure BC effectively at all atmospheric lifetimes?

i. Line 650: The observation-method is not necessarily an upper-bound limit on the fraction of POA as it does not consider OA emitted in sea spray, dust, and may not consider all anthropogenic forms of POA. Consider rephrasing and/or demonstrating it is a limit by adding error bars by using more conservative (higher) FFRatios.

2. Line 355: Can you elaborate on why the PMF doesn't work to separate SOA and POA? When (what timescale, location, altitude, other factor) does the PMF stop working?

3. Perhaps the strengths of DYN could be better isolated/highlighted (e.g. what lifetimes, OA regimes, POA levels, etc does DYN perform better for?). The abstract statement: "concept of a more dynamic OA...with enhanced removal of primary OA, and a stronger production of secondary OA in global models needed to provide a better agreement with observations" could use more support.

Evaluation seems mixed: Figure 8, indicates GC12 and CESM1-CARMA have reasonable POA (while CESM2-DYN has overestimated POA). Line 803 indicates DYN evaluates better, but that effect seems marginal or secondary to other issues in Figure 9 b at least for GC. RMSE in Table 2 also tends to increase when going from base to DYN treatment in CESM2 and GC12. Figure 7 improvements seem mixed.

Minor comments:

1. Abstract states "OA predictions from AeroCom Phase II...span a factor of 400-1000..." should that be the inter-model variability spans a factor of 400-1000 or the concentrations predicted span that range?

2. Did all models assume nonvolatile POA?

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3. Do OA/OC ratios include consideration of S and N in the form of organosulfates and organonitrates?

4. At times, it was a bit confusing if the AeroCom-II ensemble referred to circa 2014 models or the actual AeroCom II results paired with measurements. On line 103, the "AeroCom results" could be clarified as "AeroCom models" or "models used in AeroCom." Line 277 might be better as "monthly average results of 28 global models" instead of "results of 28 global models."

5. Figure 1: Is "author affiliations" supposed to be in the caption?

6. Line 163: While marine production of OA may be smaller than continental production, it's possible marine production contributes more to concentrations over the oceans than it's global production rate would suggest. I recommend rewording sentence. What concentrations of SOA are predicted for oceanic isoprene sources?

7. How does averaging data allow below detection values to be used?

8. Line 605: qualify the OA/OC of 1.4 as "fossil fuel" combustion as biomass combustion tends to have OA/OC of 1.7.

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