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Atmospheric Chemistry and Physics Discussions

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 #2

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This is an interesting study that makes comprehensive use of a unique dataset (ATom) to evaluate a series of models. The multi-model approach is particularly valuable for pinpointing model deficiencies in these remote environments. The authors present a thorough series of comparisons, however the conclusions are not well supported. This is primarily due to the reliance on an analysis to separate POA from SOA in the measurements which is not very well justified. More work is needed to expand this analysis (see below for suggestions), or remove it and alter the text accordingly, before the manuscript would be acceptable for publication.

1. Figure 1: This figure is unclear and not sufficiently discussed in the main text. What does "distribution of studies" used as the x-axis of Figure 1b mean? – a more exact definition of what is plotted should be provided. In addition, the quantitative discussion of these AeroCom results in the abstract is unclear (line 37) – what does "factor of 400-1000" imply – that the spread of the means is of this range? This could more clearly be given as a percentage of the mean or median model, or as phrased in lines 100-103 as "model dispersion" in orders of magnitude. The manuscript does not fully discuss what is shown in Figure 1b.

2. Section 2.1 would benefit from a bit more discussion of the methodology in selecting these models and the differences in their configurations. Are they all standard configurations (i.e. as downloaded), including emissions used, if not why were different parameters chosen? The level of detail in the description of the various models is quite uneven –the authors should ensure that the same information is provided for all models. Finally, are simulations performed and sampled to match the spatial location of the ATom aircraft (with emissions and meteorology matched to the year of the measurements)?

3. The manuscript is missing any discussion of the role of POA treatment in these comparisons. It's not 100% clear from Section 2.1 (e.g. no info provided on POA for ECHAM-HAM, GC10-TOMAS, or any of the CESM configurations), but it appears that all of these simulations use non-volatile POA. A number of modeling studies have implemented a semi-volatile treatment of POA since Robinson et al. (2007). It seems like a major weakness to draw general conclusions on OA model performance when using a series of models which do not represent the semi-volatile nature of POA. It would be nice to see the authors add such a simulation to their suite, but if this proves impractical at this stage of the work, the manuscript should be altered considerably to acknowledge the gaps in the POA treatment and how this may have a substantial impact on the comparisons and conclusions drawn here. The lack of discussion of the simulation (and emissions) of POA also somewhat undercuts the discussion of Section

4.3. It's clear that models are underestimating the observed OM:OC during ATom, but if the models are over-estimating the POA to begin with (perhaps because it's all assumed to be non-volatile?) then this could be a compensating bias related solely to how POA is treated.

4. Some information on model configurations is missing that would be important for comparing model performance (could potentially be added to Table 1): what is the assumed OM:OC ratio, what are the global emission totals for key precursors (isoprene, monoterpenes, POA, etc.)?

5. The estimation of the POA fraction in Section 3.2 is not well supported. First, the manuscript is missing a discussion of the uncertainty on the fBB from PALMS (lines 340-342). Second, the numbers in Table S1 do not support the averages used in the text, for example EFs for urban sources range over an order of magnitude (0.16-15.4) and the authors appear to have simply averaged these values, which seems highlight inappropriate. The example provided by the authors of using a single ratio of BB from Andraea (2019) leading to a POA fraction of > 100% in African plumes also illustrates the inappropriate application of a single number. EFs range significantly with fuel type, combustion conditions, and location; use of any single value is likely to lead to uncertainties that would vastly outweigh the value of the analysis. A more appropriate approach might be to take a lower limit set of EFs and an upper limit set of EFs, and bracket the POA estimation using first one and then the other. Absent such an analysis, this POA estimate seems unreliable and the results of Section 4 are highly questionable. The analysis of Figure S9 seems to go in this direction, but the range in EFs in this Figure do not represent the full range of values shown in Table S1. Given that all the conclusions in Section 4.4 hinge on this analysis, perhaps the authors could expand this discussion: describe the range in fBB values, and then the calculated POA contributions (from FF and BB separately) estimated for all the ATom data. In order to explore the uncertainties in their methodology, the authors could also apply the same analysis to the model output of [BC] and assumed EFs (use first the same EFs as

used in the measurement analysis and then the EFs used in the model) to see how an estimated POA_model would compare to the simulated POA. This could pinpoint whether flaws in methodology for estimating POA or flaws in the model simulation of POA dominate.

6. Lines 813-821: Figure 10 seems interesting, but it feels like an aside. The details of how these models treat inorganics (including nitrate, ammonium, sea salt, and dust) and the relevant emissions, which would be necessary to understand these differences are not included in the manuscript. Thus, the authors should either eliminate this text in favor of a more focused discussion of the OA results (as suggested in point #5 above), or substantially enhance the model description section to include the relevant details.

Minor Comments

1. The mixed capitalization in the title is a bit odd.

2. Line 69: The authors might consider re-phrasing. The word "major" implies a larger role in RF than OC contributes in the AR5 assessment cited (i.e. GHG dominate the RF, and even amongst aerosols, the effect of OC is considerably less than the inorganics or BC).

3. Line 92: Hodzic et al (2016) do not use the "same field campaigns" – rather they use a subset of those previously analyzed by Heald et al. (2011) with some additional campaigns.

4. Lines 87-91: Pai et al., ACPD, 2019 provides a more recent evaluation of the standard GEOS-Chem model configurations (including comparisons with ATom) which should be discussed here and perhaps elsewhere in the manuscript, particularly as they do not see the same bias away from source that was highlighted in previous studies (Heald et al., 2011; Hodzic et al., 2016).

5. Table 1: Why are dust and seasalt sizes included here, and why are they listed in the sub-micron only? Dust and sea salt go well into the 10's of um in model simulations.

6. Lines 198-199 and 756-757: Marais et al. (2016) replace the isoprene VBS with their mechanism for isoprene SOA. Please clarify whether isoprene SOA in your simulations follows this or whether it includes both that from the VBS of Pye et al. (2010) as well as that produced using the mechanism of Marais et al. (2016), which might lead to double-counting of isoprene SOA.

7. Line 201: does "with the exception of the treatment of isoprene SOA" imply that photolytic removal does not apply to isoprene SOA in GC12-DYN?

8. Line 249-250: does this imply that CESM2-DYN uses the same SOA yield parameters, photolytic loss, and updated Henry's law constants as GC12-DYN? If not, please clarify which differ.

9. Line 275: "in a climatological way" is not defined here. Suggest remove as the later text describes how the model is sampled.

10. Figure S1 should be included in the main text given that it shows a central comparison of ATom-2 with the models

11. Section 3.1: The measurement description section should include the detection limits and uncertainties on the AMS data during ATom and how this might impact the comparisons. I noted that some of this is given in lines 415-423, but it seems like this belongs earlier in the measurement description section, or at least that the authors could refer the reader to this later discussion in their manuscript, so that they know it will be addressed.

12. Line 329: could the authors be more explicit? Does this imply that biomass burning OA from Africa is larger in size than typical?

13. Lines 330-339: what is the size range of the aerosols detected by the PALMS instrument?

14. Line 339: unclear. Why is the AMS transmission function applied to the PALMS data?

15. Line 343: Given that fBB from PALMS is a derived quantity and not a direct measurement the statement that the PALMS fBB "is more useful as a particle tracer" is a bit bold and requires a citation. Or the language should be softened to "may be more useful"

16. Lines 346-349: These sentences seem to conflate primary and biomass burning, which are not necessarily the same thing. If the implication is that the analysis assumes no SOA from biomass burning (such as suggested by Hodshire et al., 2019), that assumption should be stated explicitly here.

17. Line 382: what are the units on the POA? In units of carbon or was an OM:OC applied?

18. Line 391: EFs range orders of magnitude and these ranges in both the model and measurements are being compared. It's not clear that "no significant bias is apparent", they could easily differ by a factor of two on average – perhaps the authors rather mean something like "the ranges in values are consistent".

19. Section 3.3: The authors have focused on the means for their model-measurement comparisons. It would be useful to examine whether this is an appropriate metric – are the distributions skewed? Can the models capture the shape of the distribution? Might a comparison of medians in Section 4 provide different results?

20. Lines 435-436: could you provide the range of MBL heights and tropopause heights along the flight tracks used here?

21. Figure 2b: please provide either total number of points or percentages of total dataset for the categories should here.

22. Line 472: suggest inserting the word "likely" to "less polluted than ATom-1, likely due to a" since you haven't definitely compared emissions or source contributions.

23. Line 497-498: The statement "It should be noted.." is surprising. The authors haven't shown any analysis for this and Figures 2a and S1 clearly show elevated OA in

the North Pacific which seems likely associated with Asian source. Could the authors explain?

24. Line 537: The NMB in Table 2 for CESM1-CARMA is given as -33.2%, so the -20-30% range seems incorrect.

25. Table 2 indicates that the NMB for all models is positive for ATom-2. I didn't see this surprising result discussed in the text.

26. Line 539-541: This statement is incorrect as it only appears to apply to CESM. According to Table 2, while GC12-DYN is slightly less biased than GC12-REF for ATom-1, the reverse is true for ATom-2.

27. Section 4.4: The POA to OA ratio is derived, not directly measured (e.g. line 685)

28. Lines 726-728, and 737-738: given these statements why does Figure 8 not include a comparison of BC with and without in-cloud removal?

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