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Comment on acp-2022-545

Anonymous Referee #1

Referee comment on "An evaluation of biomass burning aerosol mass, extinction, and size distribution in GEOS using observations from CAMP^{2}Ex" by Allison B. Marquardt Collow et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-545-RC1, 2022

This manuscript presents a thorough evaluation of the performance of the GEOS global atmospheric chemistry model with the GOCART aerosol parameterization. The model output is compared with in situ, airborne measurements of aerosol compositional, microphysical, and optical properties made in the vicinity of the Philippines as part of the CAMP2EX project in 2019.

The output from the model is compared with the observations over project-wide averages, and for some specific flights in biomass burning smoke, and includes both extensive and some intensive parameters. The importance of assimilation of AOD values is examined, as are the effects of a newer convective parameterization scheme. While not comprehensive, this analysis is enough to identify discrepancies in the assumptions regarding aerosol composition, hygroscopicity, and size distribution in the GOCART parameterization. These discrepancies are large enough to be a concern, and indicate that perhaps the GOCART approach, with its one-size-fits-all static parameterization of the properties of different aerosol components, should be replaced with a more physically based and interactive aerosol parameterization.

This manuscript is suitable for publication in ACP following revisions that fall somewhere between major and minor (I've described them as minor). I have one significant comment below that I would definitely like to see addressed prior to publication, followed by a number of minor comments. Despite the number of comments, this is a generally well written paper, and an interesting and enjoyable one to read.

Major Comment:

Much of the analysis relies on comparison of the model output with extinction values at ambient RH. These are referred to as "measurements", but in fact they are derived values that rely implicitly on an underlying assumption regarding aerosol hygroscopicity. As documented in the headers of the data files from the LARGE measurements for this

campaign, scattering (which vastly dominates extinction here) was measured at two fixed RH values: ~20% and ~80% RH. The ambient scattering is then calculated by assuming a functional form (the gamma parameterization), which is just a power law fit. In other words, the ambient scattering (hence extinction) is based on fitting a parameter to the two measurement points, then extrapolating to the ambient RH. The error in this approach is probably not large for RH values lying between or close to the two measurement RH values (20% and 80%). However, in the CAMP2EX profiles, the ambient RH exceeds 90% in the upper half of the boundary layer, and this is where the greatest contribution to extinction lies. In this high-RH region, the power-law hygroscopic growth curve pitches very sharply upwards, and small errors in the assumed shape of this growth curve can amplify to very large errors in calculated ambient extinction. So this comparison is not optimal, because the ambient extinction values will have large, and unknown, uncertainties in the high-RH region that dominates AOD.

One of the goals of the comparison of the measured and modeled extinction is to determine if the model hygroscopic growth is consistent with the measurements. To accomplish this, it might make more sense to compare the model to the measurements for the actually measured extinction values at the 20% ("dry") and 80% ("wet") conditions. That way the comparison is not between the modeled values and those derived from the measurements with an assumed shape to the hygroscopic growth curve. Alternatively, you could plot the full f(RH) curve the model would produce (for a given location and time), from 20% RH to ~95% RH, and compare that with the same curve provided by the gamma parameter calculated from the LARGE measurements. This would be informative, and would allow you to compare the extinction at the measured 20% and 80% RH values as well as look at the response at higher RH values. The ratio of these two curves would indicate where there might be relative biases (although whether these lie in the model or in the gamma parameterization assumption might not be so clear).

Minor Comments:

There are a few places where there are typos or where the manuscript could be edited for clarity.

1) Lines 81-85. The FIMS and HSRL2 are introduced by name, but the text doesn't say what they measure. That information appears in lines 100-104. I was trying to figure out what a FIMS measured throughout the text between these lines. Suggest moving the latter section up to lines 81-85.

2) Line 117. Change "hydroscopic" to "hygroscopic".

3) Line 123. Define sigma as the geometric standard deviation.

4) Line 130. Change to, "The underlying meteorology from GEOS is used for horizontal and vertical transport and deposition of all of the aerosol species, as well as wind-driven emissions of dust and sea salt."

5) Line 193. Define "lidar ratio".

6) Lines 211 and 223. "Optical array" has a particular meaning in optical design. Suggest changing to "optical properties instruments" or something similar.

7) Line 213. Change to "fine particles that are efficiently sampled".

8) Lines 222-234. Here is where you could expand on the model/measurement comparison of extinction as a function of RH and focus on the actual full frh curve or compare the extinctions at 20% and 80% RH.

9) Line 241. I don't understand this sentence. "Analysis increment"?

10) Line 258. Please add a comma between "region" and "or".

11) Line 273. Please change to, ". . .represented that percentage of organic carbon well; however, it struggled with. . . ."

12) Line 281. In the heading for Table 2, I believe this in not "total aerosol mass" because it excludes BC.

13) Line 299. I believe the values for the extinction measurements are 20% and 80% RH, at least according to the file headers.

14) Line 313. Saying the GEOS values for SSA are "below 0.96" doesn't do justice to the magnitude of the discrepancy, which is quite large. Maybe say they range from "0.9 to 0.96". The co-albedos disagree by a factor of 2-4; this is quite large in the context of direct radiative effects.

15) Line 323-324. It's true that the extinction could be juiced up by making the modal

diameter larger and the width wider, but the assumed standard deviation of ~2+ is already quite a bit larger than literature values would support. You might want to lead into the next sentence by saying, "We will examine the in situ measurements to see if such changes could be justified. We begin by looking at . . . ."

16) Line 337. The sentence beginning "The primary peak" is confusing. Please define what you mean by the "primary peak? It's being "shifted toward a larger radius" than what? Are these the measured or GEOS size distributions you're talking about? Please clarify.

17) Line 368. I'm not sure what "mass piling up at the top of the PBL" means. I don't think mass (of air or of aerosol) can "pile up", at least not without increasing air density a lot!

18) Lines 391-395. These sentences are unclear. What "concern" about the size distribution could be "rectified"? Please be specific, e.g., "agreement between modeled and measured extinction at high RH could be improved if the modeled size distribution had a larger modal radius and/or a larger standard deviation. However, these adjustments are not supported by the FIMS size distribution measurements. In fact, the observed mode radius is in excellent agreement. . . ."

19) References. Please ensure that all references are compliant with Copernicus formatting guidelines. For example, some of the references (e.g., Burton et al., 2012) have capitalized titles, while most do not. This is a result of reference management software, which always needs to be thoroughly checked manually.

20) References. I'm not sure Schill et al. is citable--it's an unpublished conference presentation.

21) Figures. The figures are generally very nicely done, clearly labeled. However, I'm not sure of the ability of those with color impairment to read the shaded vertical profiles (e.g., Figs. 3-6). Also, in these figures the horizontal lines indicating HSRL2 MLH and GOES PBLH are difficult to discern; the black and blue colors are quite close. Could one line be made dotted? In Fig. 4 you might need a 3rd line type for clarity.

22) Figures 13, 14. It might be nice to fit a lognormal to the FIMS peaks, then you could directly compare the mode diameter and standard deviation with the model. My eye says that the standard deviation for the measurements is less than the very broad modeled values, but I can't be sure without fitting. I much prefer the aspect ratio of Fig. 14 to that of Fig. 13; could Fig. 13 be made with side-by-side plots, rather than vertically stacked ones? This would make Fig. 13 and 14 look more similar.