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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Uncertainties in global aerosols and climate effects due to biofuel emissions" *by* J. K. Kodros et al.

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We thank reviewer 2 for their constructive response. We have extracted the critical comments by the reviewer and presented them in italics in-line below.

Although the study includes even 18 sensitivity simulations, the manuscript is written in such form that the main results can usually be extracted with ease. Nevertheless, I would suggest minor revisions to help the reader. Currently, supplementary information is not provided with the manuscript. I feel that all the material provided is relevant to the goal of the manuscript. However, perhaps some of the more technical results could be moved to supplementary part, as they could still be used in the main texts as now. E.g. Figure 6, 8, 10 and 11 could be possibly moved to supplementary

We feel Figures 6 and 8 should remain in the main text. Figure 6 offers a strong visualization of the range of direct effect that result from different mixing state assumptions. This range varies by region. Figure 8 shows how the spatial pattern of the nucleation/condensation suppression feedback influences the global averaged AIE. Figures 10 and 11 have been moved to the supplemental, to help the casual reader. We will upload a version of the document with track changes that shows our modifications.

Section 2.2: If I understand correctly, monthly average cloud fraction is taken as from GEOS5 for DRE and from ISCCP-D2 for AIE? What is the reason for this choice, since it seems to lead to inconsistent assessment of the two radiative effects?

For the AIE we use the method described in Scott et al. (2014) and Spracklen et al. (2011), and wanted to be consistent with cloud fields in these papers. It is possible that using GEOS-5 cloud fields for the direct effect (and model processes such as photolysis rates, and wet removal), we may have somewhat different climate effects than we calculated using the ISCCP fields, but it is unlikely that our major conclusions will change (e.g. what parameters the climate forcings are sensitive to).

Page 10209, line 14 onwards: where are the aerosol concentrations sampled from, cloud bottom?

Cloud droplet number concentrations for each month are calculated in each gridcell from the 3D aerosol distribution values. We have added text to Section 2.2 to clarify this, and other details relating to the calculation of the cloud-albedo AIE.

Page 10209, line 14 onwards: The indirect effect of aerosols is extremely poorly constrained, and in many cases even difficult to analyze from simulations. I think the method for calculating the aerosol indirect effect (Spracklen et al., 2011) contains too many oversimplifications of the complex aerosol-cloud interaction. Additionally, using monthly averages for both aerosol concentrations and cloud properties can lead to potentially large uncertainties, since both fields vary in small timescales and are extremely coupled through e.g. aerosol formation and deposition processes. Apparently, *mixed-phase and ice-clouds are not considered. Although I understand the authors' need to explicitly express the indirect effect in terms of W/m2, I would suggest to report e.g. changes in CCN concentrations instead. If forcing is to be reported in current form, some additional discussion of AIE method in section 2.2 is needed.*

We have added additional explanations on our method in Section 2.2. We have also added the following discussion of limitations in Section 2.2: "This offline determination of the AIE, using monthly averaged aerosol distributions and monthly averaged cloud data does not allow us to capture the effect of variations in either CDNC and cloud properties occurring on shorter timescales and any nonlinearities that arise from considering short-timescale interactions. While this is a simplification, it allows us to understand the relative importance of the uncertainties in aerosol emission and properties explored here."

This discussion is repeated in the Results and Conclusion sections for clarity.

In Table 3, we report changes in N40 and N80. N40 and N80 are often considered proxies for CCN, and the relative sensitivity to N40/N80 is similar to that of CCN. While we agree the use of monthly averages for aerosol concentrations and cloud fields can lead to large uncertainties in the absolute value of the AIE, we feel that this method is sufficient to show a strong sensitivity to uncertainties in aerosol emissions and properties.

Page 10210, line 10: What emission source does "assumed size distribution" relate to? Is this applied to all emissions, including e.g. fossil fuel use and agricultural burning?

Thank you for noting this ambiguity. This is applied to just carbonaceous biofuel emissions. We have amended the text to explicitly state this.

Page 10210, line 15: Why a fixed timescale, and not coupled with simulated chemistry?

We intend to update the aging scheme in GEOS-Chem-TOMAS to be coupled with simulated chemistry. We have not done it yet as it is a non-trivial change. Each of our

15 size sections should have its own calculated timescale. We have added text to the paper that says "this fixed timescale is limitation of our work regarding hygroscopicity; however, our sensitivity to hygroscopicity is very small, so it is unlikely this conclusion will change."

Page 10210, line 19: What method is used for SOA formation, and are there separate assumptions on the precursors and SOA volatilities from biogenic vs. anthropogenic sources?

Sorry we did not state this explicitly, we have added the following line to the text: 'These SOA sources are added upon emission with fixed yields: 0.1 of monoterpene emissions for the biogenic SOA and 0.2 Tg-SOA Tg-CO^{-1} for anthropogenic CO emissions on a mass basis for the anthropogenically enhanced SOA (note that anthropogenic CO is simply being used as a proxy for anthropogenically enhanced SOA rather than an actual precursor).'

Table 5: Although the idea of an overview of main parameters is good in order to summarize the extensive simulation results, I don't feel that the table provides something that couldn't be (or isn't already) said in the text. Why "Emissions size" in DRE and "Emission size distribution" in AIE? Why is composition not at all in AIE?

Table 5 was included to help the casual reader who will not read the paper in detail. We wanted to distill the main points of the paper in a concise way. 'Emission size' should refer to 'Emission size distribution' - we have changed this. Our sensitivity study found that AIE was not sensitive to changes in composition (at least for BC, OA and hygroscopicity). We have added statements for this.

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