

The paper reports a study of the impacts of coagulation of particles in biomass burning plumes on the climate impacts of biomass burning aerosol. The study finds that this process, that is not usually included in atmospheric or climate models, reduces the number of cloud droplet forming particles produced by biomass burning by 37% globally. Overall, the study finds that including coagulation of particles in biomass burning plumes reduces the cooling impact of biomass burning aerosol through the aerosol indirect effect, but increases the cooling impact through the direct radiative effect.

This is an important study. The paper is well-written. The model experiments are clearly described and the authors have tested a range of assumptions and datasets. I recommend publication after any minor comments have been addressed.

We thank this reviewer for their helpful and thoughtful review. In particular, we are grateful for this reviewer agreeing to review the paper late in the open discussion and providing comments quickly. Our responses throughout are in italics.

Minor comments:

Section 2.1. What new particle formation scheme and SOA formation did you include in the model? These will control the baseline particle number and the growth rates of particles and so are important for your study.

To clarify this, we have made this addition to Section 2.1: “Nucleation rates are parameterized with binary nucleation (Vehkamaki et al., 2002) and ternary nucleation (Napari et al., 2002) scaled globally by a tuning factor of 10^{-5} (Jung et al., 2010; Westervelt et al., 2013). Secondary organic aerosol includes a 19 Tg yr^{-1} biogenic contribution and a 100 Tg yr^{-1} anthropogenically enhanced contribution correlated with anthropogenic CO emissions (D’Andrea et al., 2013), following the approach of Spracklen et al. (2011).”

Figure 6. Do you report the values averaged over all size distributions? From the figure it looks like the impact of coagulation on the average DRE value is greater than the 4% in the Abstract?

This is a very good point. The 4% value does not adequately capture the effect that sub-grid coagulation of biomass burning aerosol has on the average DRE. The values listed in the conclusions, which is where the 4% value is reported, were only for the default initial size distribution case where D_{pm0} is 100 nm and σ_0 is 2. To clarify this, we added the following sentence, “Sub-grid coagulation increases the biomass burning global-, annual-mean direct radiative effect (DRE) by 4% from -206 mW m^{-2} to -214 mW m^{-2} due to an increase in mass scattering efficiency for the default initial size distribution with an initial median diameter of 100 nm and an initial lognormal modal width of 2 (on average between the external mixing assumption and the internal, core-shell mixing assumption)”.

In Fig. 6, the DRE is not affected very much by the inclusion of sub-grid coagulation of biomass burning aerosol when only the default case (filled square) is considered. However, the sensitivity cases can vary much more. To include the other initial size distribution sensitivity cases, we have added the following to the conclusions: "In our sensitivity cases testing different initial size distributions, described below, the DRE is more affected by the presence or absence of sub-grid coagulation of biomass burning aerosol, changing as much as 22%".

Section 2.2 How would your results depend on parameter uncertainty in equations (1) and (2) on Page 6. The authors should be commended for exploring the uncertainty in the global model inputs/datasets. It is probably beyond scope to explore the impact of uncertainty in these equations, but a short discussion would be useful.

This is a good thing to discuss explicitly. We have added the following text to Section 2.2: "In Sakamoto et al., (2016), these equations (Eqns. 1 and 2 here) explain 77-79% of the variability in Dpm and in their plume simulations. Hence, there are uncertainties in our analyses introduced by the simple form of Eqns. 1 and 2; however, we expect these uncertainties to be smaller than the uncertainties in biomass burning emission inventories, plume overlap, fire size, mixing height, etc.".

Section 2.2 Do you have information on the values of Dpm and model width calculated from equations (1) and (2)? It would be interesting to know the mean values used as input to GEOS-chem as well as spatial and temporal variability.

The spatial variability of the annual-mean values for these two variables was given in Figure 3 (pasted below), though this was not introduced until Section 3.2. We have moved its introduction to Section 2.2 (now becoming Figure 1) with further discussion remaining in Section 3.2.

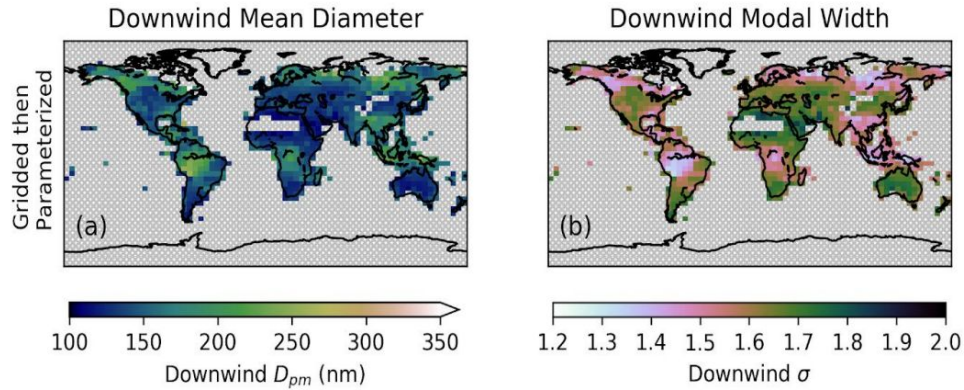


Figure 3: Annual-mean median diameter (a) and modal width (b) for sub-grid-processed biomass burning emissions predicted for 2010 using the Sakamoto et al. (2016) parameterization after 24 hours of sub-grid coagulation with an emitted median diameter of 100 nm and an emitted modal width of 2. Fire (FINNv1.5) and meteorological data is averaged over a $4^{\circ} \times 5^{\circ}$ grid and then that gridded data is run through the Sakamoto et al. (2016) parameterization. The regions with grey cross-hatching are grid-cells with no fire data.

To show temporal variability, we have now plotted the timeseries for the sub-grid-plume processed D_{pm} and σ for the two locations investigated in Figure 5 (Alaska and the Amazon). These timeseries plots are below. In both of these regions, there is day-to-day variability driven by changes in wind, fire size, and mixing depth. There is also an apparent seasonal cycle in the values at both locations, where particles are larger and have a narrower distribution at the peak of the fire season, which is likely driven by larger fire sizes during these times. We have added these figures to the SI and added a brief reference to them in Section 2.2.

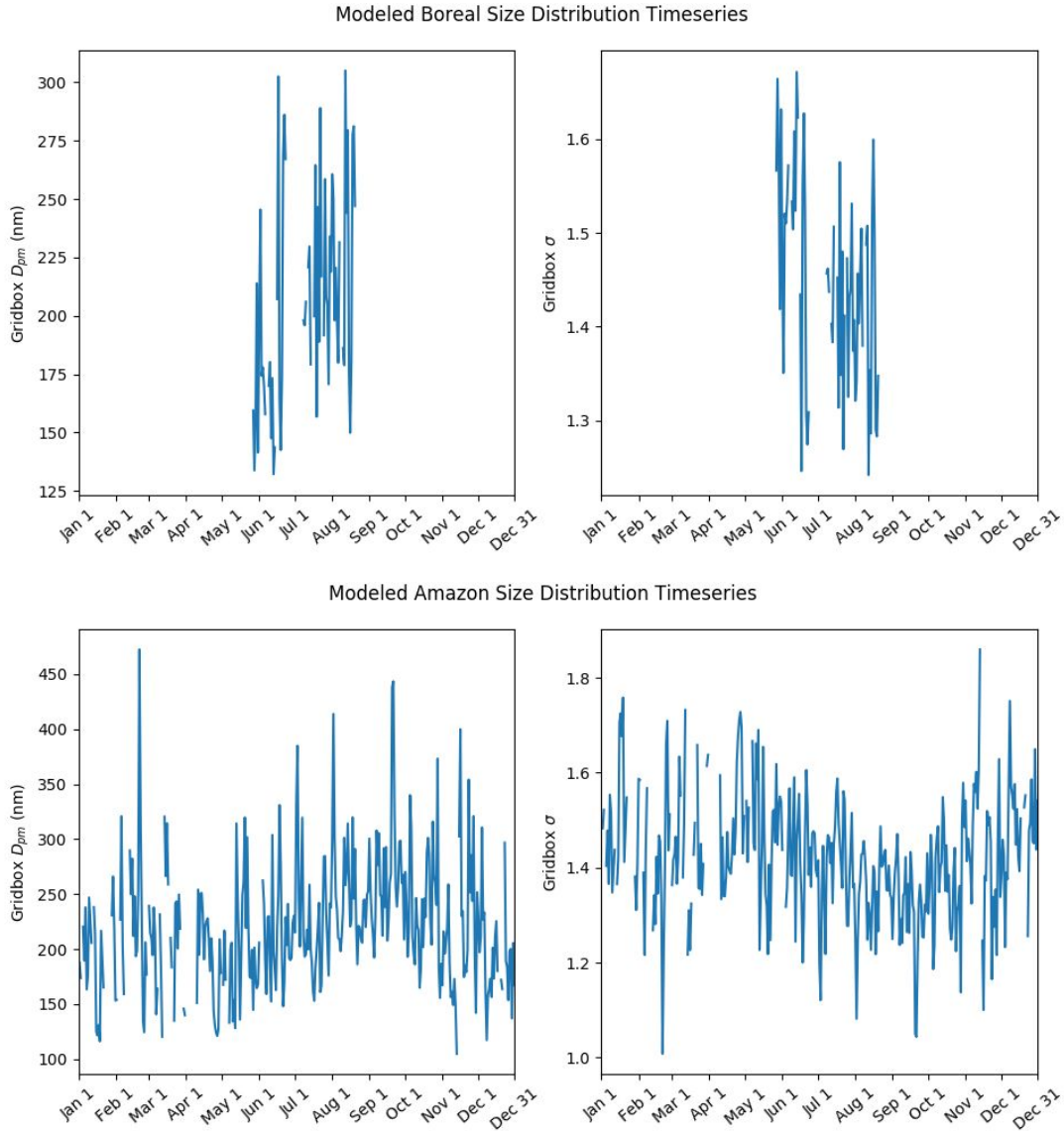


Figure S2: Daily temporal evolution of the predicted grid-resolved median diameter (D_{pm} ; left) and modal width (σ ; right) in a box that spans 8° latitude and 10° longitude for biomass burning emissions predicted for 2010 using FINNv1.5 fire emissions and the Sakamoto et al. (2016) parameterization after 24 hours of sub-grid coagulation with an emitted initial median diameter of 100 nm and an emitted initial modal width of 2. The top plots are centered over Alaska at 62° N, 135° W. Times that are not shown have no fire data in this gridbox. The bottom plots are centered over the Amazon at 6° S, 60° E.

An important point is to what extent the emitted size distribution in the model represents fresh or aged smoke. This is mentioned by the authors in Section 2.2. Could the treatment of in-plume coagulation simply be captured by assuming a larger emitted size? Or does the in-plume calculations allow treatment of important spatial and temporal variability that would be ignored by using a globally uniform value?

Given the spatial and temporal variability in the sub-grid-processed size distributions in Figure 1 (previously Figure 3) and Figure S2, it appears that any fixed global assumption of an “aged” biomass burning size distribution may underestimate regional variability. We have added text to Section 3.2 discussing this: “Given the variability in the sub-grid-processed size distributions in Fig. 1, assuming a single emissions size distribution for biomass burning in coarse grid models may underestimate the variability in biomass burning size distributions between regions.”