This study used a sub-grid coagulation parameterization for biomass burning plumes in the GEOS-Chem-TOMAS global aerosol microphysical model and showed large impacts of biomass burning sub-grid coagulation on aerosol number concentrations, aerosol size distributions, and aerosol direct and first indirect effects. The authors found sub-grid coagulation reduced the impact of biomass burning aerosols on number concentrations of particles larger than 80 nm by 37% globally and that this reduction changed estimates of aerosol direct and first indirect effects of biomass burning aerosols by 4% (from -206 mW m^{-2} to -214 mW m^{-2}) and by 43% (from -76 mW m^{-2} to -43 mW m^{-2}), respectively. The authors demonstrated that the inclusion of biomass burning sub-grid coagulation significantly reduced the sensitivity of aerosol number concentrations, CCN concentrations, and aerosol-cloud interactions to the treatment of aerosol size distributions at emissions. The topic of this work is interesting and well suited to the scope of Atmospheric Chemistry and Physics. The manuscript is well written and the findings by the authors will be useful for estimating aerosol-climate interactions more accurately. Overall, the manuscript should be accepted by this journal after minor revisions. Some minor comments, which should be addressed before acceptance, are described below.

We thank this reviewer for their helpful and thoughtful review. Our responses throughout are in italics.

Minor comments:

1. Page 1, Lines 23-24:
external mixing –> external mixing of black carbon
internal mixing –> internal mixing of black carbon

Yes, better to be more specific here. We have made this change.

2. Page 2, Lines 20-21:
Please add the following reference: H. Matsui (2016), doi:10.1002/2015JD023998.

We have added this reference.

3. Page 4, Line 24:
Please add a few sentences on the treatment of SOA formation in the global aerosol model.

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)".*

4. Page 4, Lines 23-31:
Please clarify gaseous and aerosol species considered in the biomass burning emissions in the author's global model.

To clarify this, we have made this addition to Section 2.1: "In our simulations, GFED and FINN biomass burning emissions include nitric oxide, carbon monoxide, sulfur dioxide, ammonia, all alkanes except for methane, acetone, methyl ethyl ketone, formaldehyde, acetaldehyde, alkenes with continuous carbon chains longer than two carbons, black carbon aerosol, and organic aerosol. The FINN biomass burning emissions also include hydroxyacetone and glycoaldehyde."

5. Page 6, Line 24:
In equation (1), 84.56 should be 84.576, based on Sakamoto et al. (2016).

We have made this change.

6. Page 7, Line 16:
In Sakamoto et al. (2016), their parameterization is based on their simulations conducted for 5 hours during biomass burning emissions. This parameterization was extended to 24 and 12 hours in the current study. Can the authors justify this extension?

I suggest the authors to confirm this extension does not overestimate sub-grid coagulation rate (because coagulation rate will be slower with time) and to add some discussions to the text.

Yes, we do extrapolate on the fits and this should be explicitly discussed. The Sakamoto fits depend on time to a power of less than 0.5; hence, the coagulation does slow with time in the fits. We have made this addition to Section 2.2: "In Sakamoto et al. (2016), simulations used to develop this parameterization are five hours long, so we are extrapolating their fits. Because of the dependence on time to a power less than 0.5 (Eqns. 1 and 2). The impact of coagulation slows with time, which reduces potential errors associated with extrapolating. To test the sensitivity to this 24-hour assumption we include an additional simulation (SubCoag_12h) where conditions are similar to SubCoag but with 12 hours instead of 24 hours of aging."

7. Page 7, Lines 26-29:
Scatterplots and correlation coefficients may be useful.

We have added these plots to the supplement and made this addition to the main text as a brief discussion: "Further, Figs. S4 and S5 show that most gridboxes report similar D_{pm} and σ using either method, especially at smaller D_{pm} and larger σ values, where most gridboxes lie."

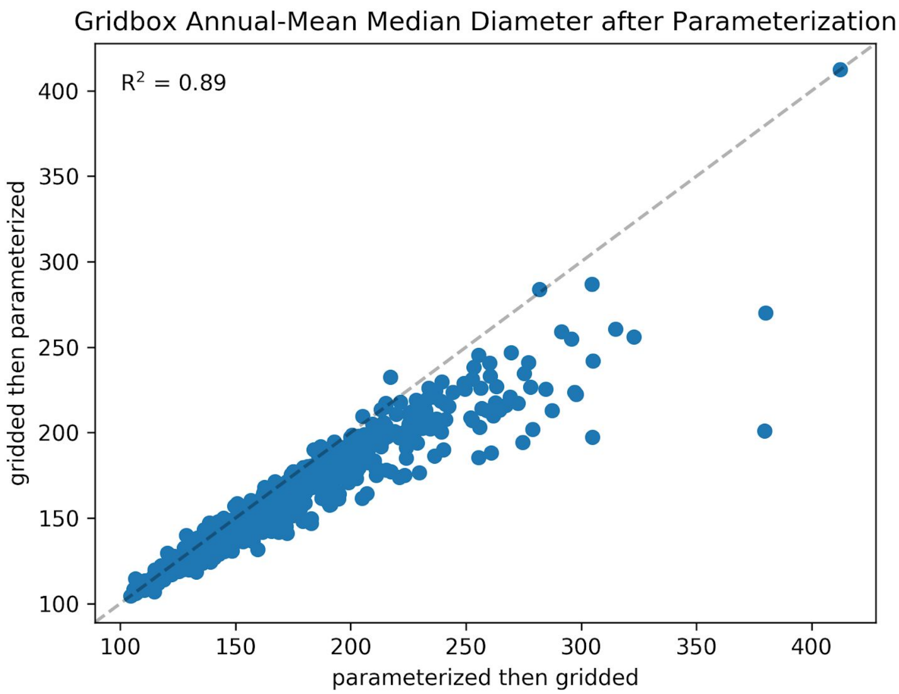

Figure S4: Annual-mean median diameter for biomass burning emissions predicted for 2010 using the Sakamoto et al. (2016) parameterization after 24 hours of sub-grid coagulation with an emitted initial median diameter of 100 nm and an emitted initial modal width of 2. The y-axis shows the resulting median modal width when the fire (FINNv1.5) and meteorological data is averaged over a 4°x5° grid and then that gridded data is run through the Sakamoto et al. (2016) parameterization (what we use in GEOS-Chem in this study). The x-axis shows the results when the individual fires are run through the Sakamoto et al. (2016) parameterization and then the output median modal width is averaged over a 4°x5° grid. Each gridbox globally is represented as a point on the plot. The 1:1 line is in grey dash.

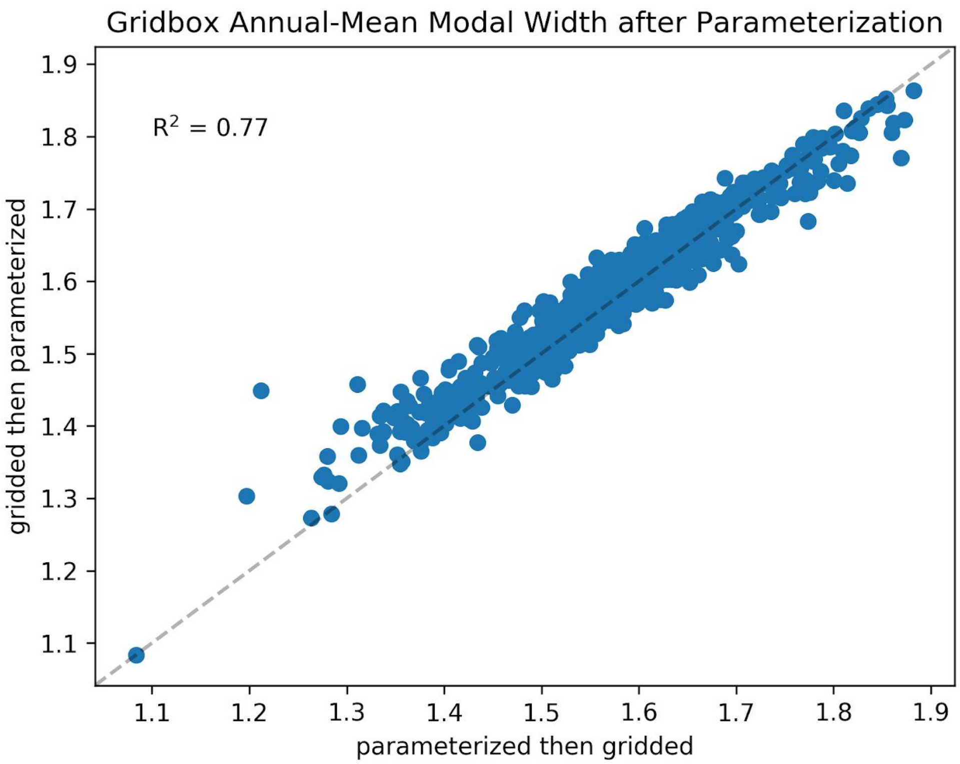

Figure S5: Annual-mean modal width for biomass burning emissions predicted for 2010 using the Sakamoto et al. (2016) parameterization after 24 hours of sub-grid coagulation with an emitted initial median diameter of 100 nm and an emitted initial modal width of 2. The y-axis shows the resulting lognormal modal width when the fire (FINNv1.5) and meteorological data is averaged over a 4°x5° grid and then that gridded data is run through the Sakamoto et al. (2016) parameterization (what we use in GEOS-Chem in this study). The x-axis shows the results when the individual fires are run through the Sakamoto et al. (2016) parameterization and then the output lognormal modal width is averaged over a 4°x5° grid. Each gridbox globally is represented as a point on the plot. The 1:1 line is in grey dash.

8. Page 8, Line 4:
Please add a sentence that the two assumptions of mixing state have the same aerosol number concentrations and size distributions in total.

We have made this addition to Section 2.3: "Both mixing states have the same aerosol number concentrations and size distributions in total."

9. Figure 2 and related figures:
The main focus of this study is the inclusion of sub-grid coagulation. So, I think the difference between SubCoag and noSubCoag is the most important point in this discussion (rather than

the differences from noBB). How about adding plots on the difference between SubCoag and noSubCoag (absolute value or percent)? The authors can add similar difference plots (between SubCoag and noSubCoag) to other figures (Figures 4 and 7-9).

We had debated the idea of making "percent change in the biomass burning impact" plots (i.e. how does the impact of BB on N10 and N80 change by adding sub-grid coagulation of biomass burning aerosol) when writing the paper; however, these plots were challenging to interpret because there regions where N10 and N80 *(1) increase due to biomass burning in both the SubCoag and noSubCoag simulations, (2) decrease due to biomass burning in both simulations, and (3) increase in one simulation and decrease in the other. However, to address this reviewer comment, we have made a version of these plots where we require *both* the SubCoag and noSubCoag simulations to have at least a 1% increase in N10 and N80 (relative to noBB) in a gridbox, and all other gridboxes are masked. We have added these figures (one for the base emissions assumptions and another for the sensitivity emissions assumptions) to the supplement and mentioned them in the main text. In this figure, the interpretation is straightforward; blue colors show that sub-grid coagulation reduces the impact of biomass burning in that location; red colors show that sub-grid coagulation increases the impact of biomass burning in that location (due to microphysics feedbacks: increased nucleation etc).*

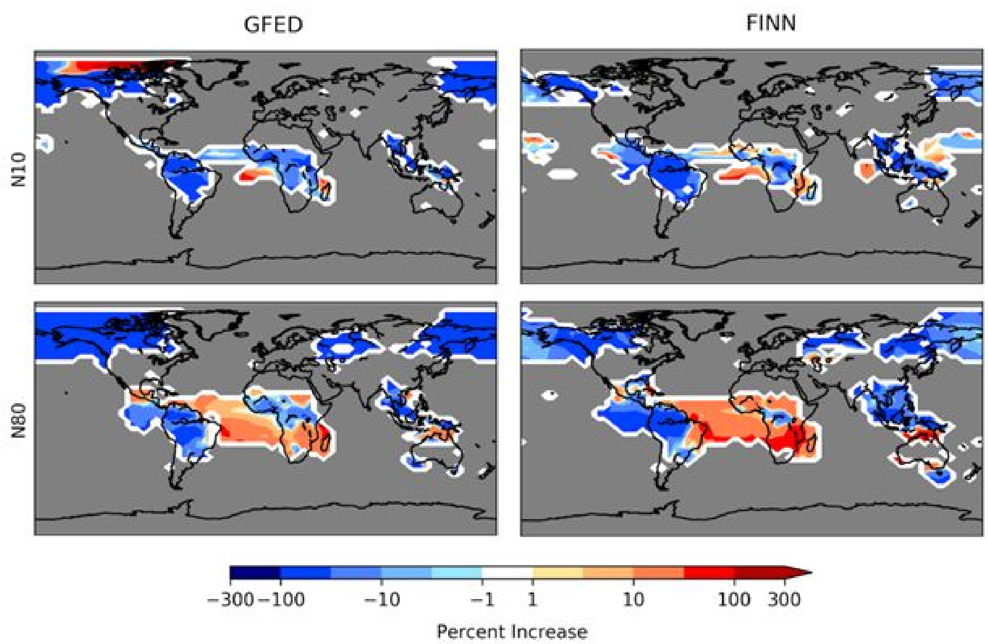

Figure S8: Percent change in the relative contribution of biomass burning to particles larger than 10 nm (N10) and particles larger than 80 nm (N80) due to sub-grid coagulation when using GFED emissions (left) or FINNv1 emissions (right), and an initial median diameter of 100 nm, initial modal width of 2, and coagulation time of 24 hours. Negative values correspond to a reduced impact of biomass burning when sub-grid coagulation is added. Regions where the percent increase of N10 or N80 due to biomass burning is less than 1% (with or without sub-grid coagulation, see Figs. 3 and S7) are shaded in grey.

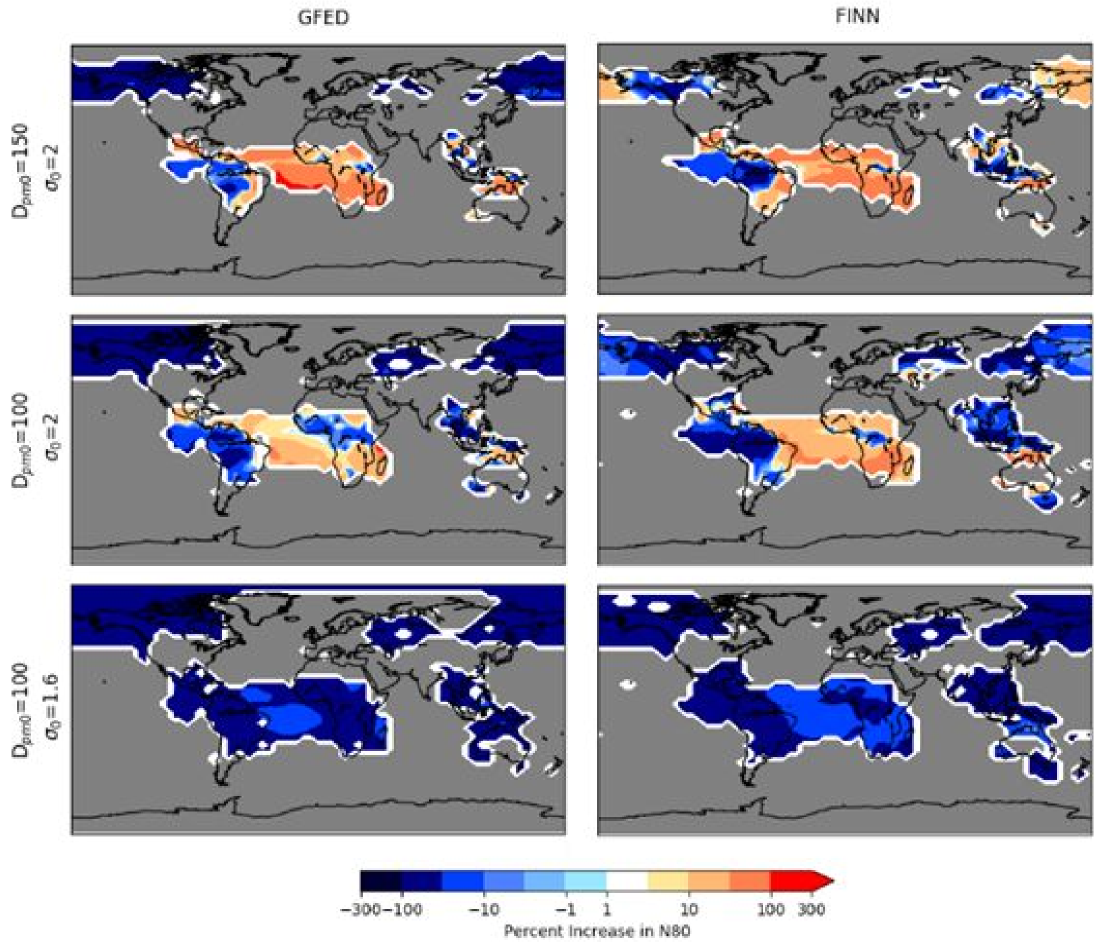

Figure S10: Percent change in the relative contribution of biomass burning to particles larger than 80 nm due to sub-grid coagulation when using GFED emissions (left) or FINNv1 emissions (right) and a coagulation time of 24 hours. Panels in the top row have an emitted median diameter of 150 nm and an emitted modal width of 2. Panels in the middle row have an emitted median diameter of 100 nm and an emitted modal width of 2. Panels in the bottom row have an emitted median diameter of 100 nm and an emitted modal width of 1.6. Negative values correspond to a reduced impact of biomass burning when sub-grid coagulation is added. Regions where the percent increase of N80 due to biomass burning is less than 1% (with or without sub-grid coagulation, see Figs. 4 and S9) are shaded in grey.

We also made analogous "percent change in the biomass burning impact" figures that correspond to the three radiative forcing figures in the main text (Figures 7-9) and also added these figures to the supplement with reference in the main text.

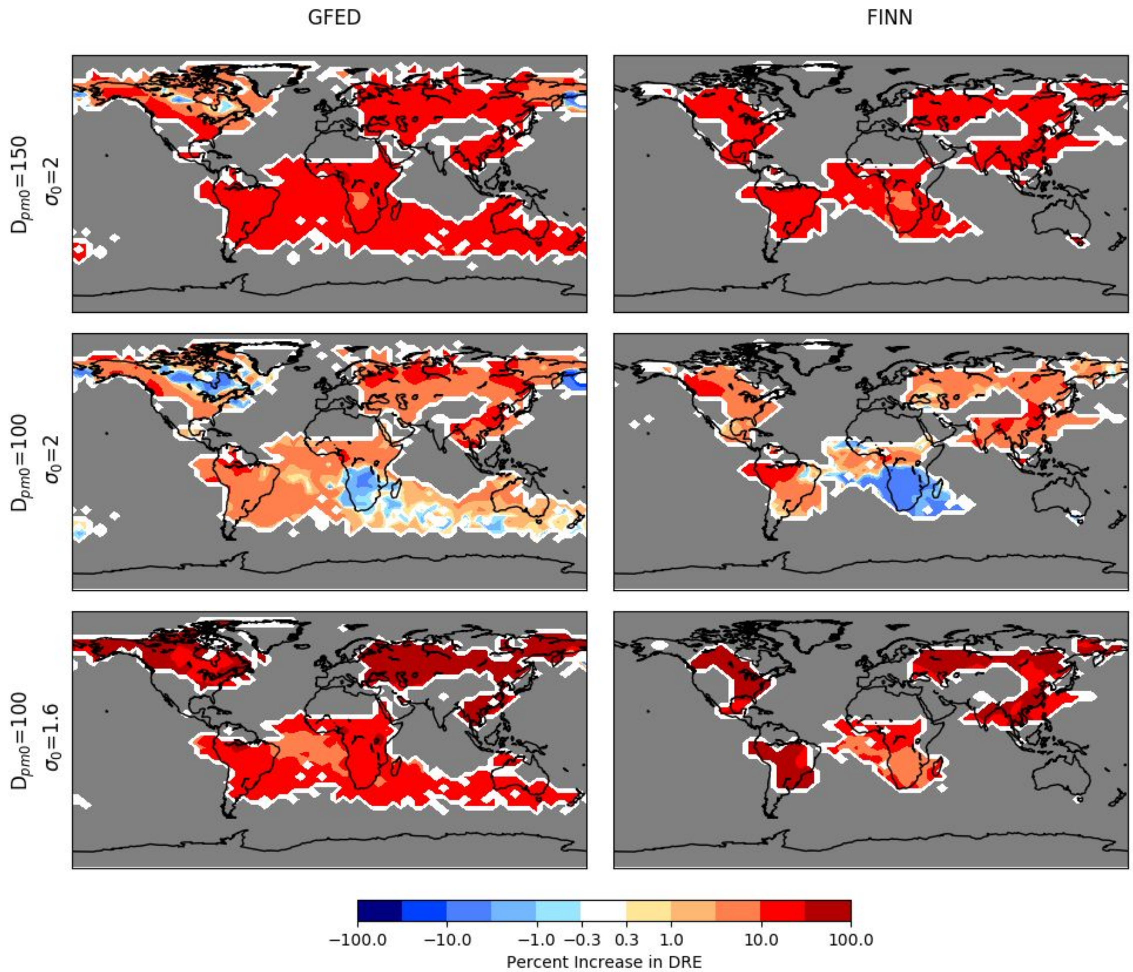

Figure S13: Percent change in all-sky direct radiative effect (DRE) of biomass burning aerosol due to sub-grid coagulation when using the external mixing assumption and GFED emissions (left) or FINNv1 emissions (right) and an assumed in-plume coagulation time of 24 hours. Panels in the top row have an emitted median diameter of 150 nm and an emitted modal width of 2. Panels in the middle row have an emitted median diameter of 100 nm and an emitted modal width of 2. Panels in the bottom row have an emitted median diameter of 100 nm and an emitted modal width of 1.6. Positive (red) values correspond to an increased cooling tendency of biomass burning DRE due to sub-grid coagulation being added. Regions in grey indicate that the DRE due to biomass burning is a more positive value than -100 mW m^{-2} (i.e. there is less than 100 mW m^{-2} of cooling) with or without sub-grid coagulation (see Figs. 7 and S12).

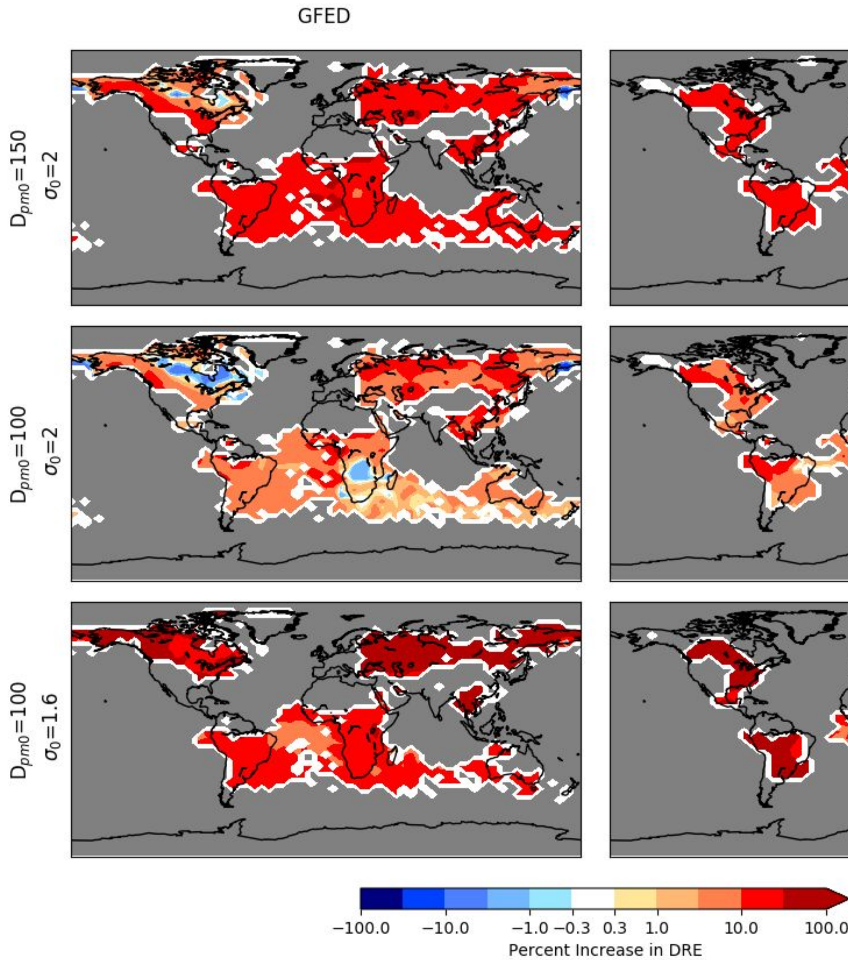

Figure S15: Percent change in all-sky direct radiative effect (DRE) of biomass burning aerosol due to sub-grid coagulation when using the core-shell mixing assumption and GFED emissions (left) or FINNv1 emissions (right) and an assumed in-plume coagulation time of 24 hours. Panels in the top row have an emitted median diameter of 150 nm and an emitted modal width of 2. Panels in the middle row have an emitted median diameter of 100 nm and an emitted modal width of 2. Panels in the bottom row have an emitted median diameter of 100 nm and an emitted modal width of 1.6. Positive (red) values correspond to an increased cooling tendency of biomass burning DRE due to sub-grid coagulation being added. Regions in grey indicate that the DRE due to biomass burning is a more positive value than -100 mW m^{-2} (i.e. there is less than 100 mW m^{-2} of cooling) with or without sub-grid coagulation (see Figs. 8 and S14).

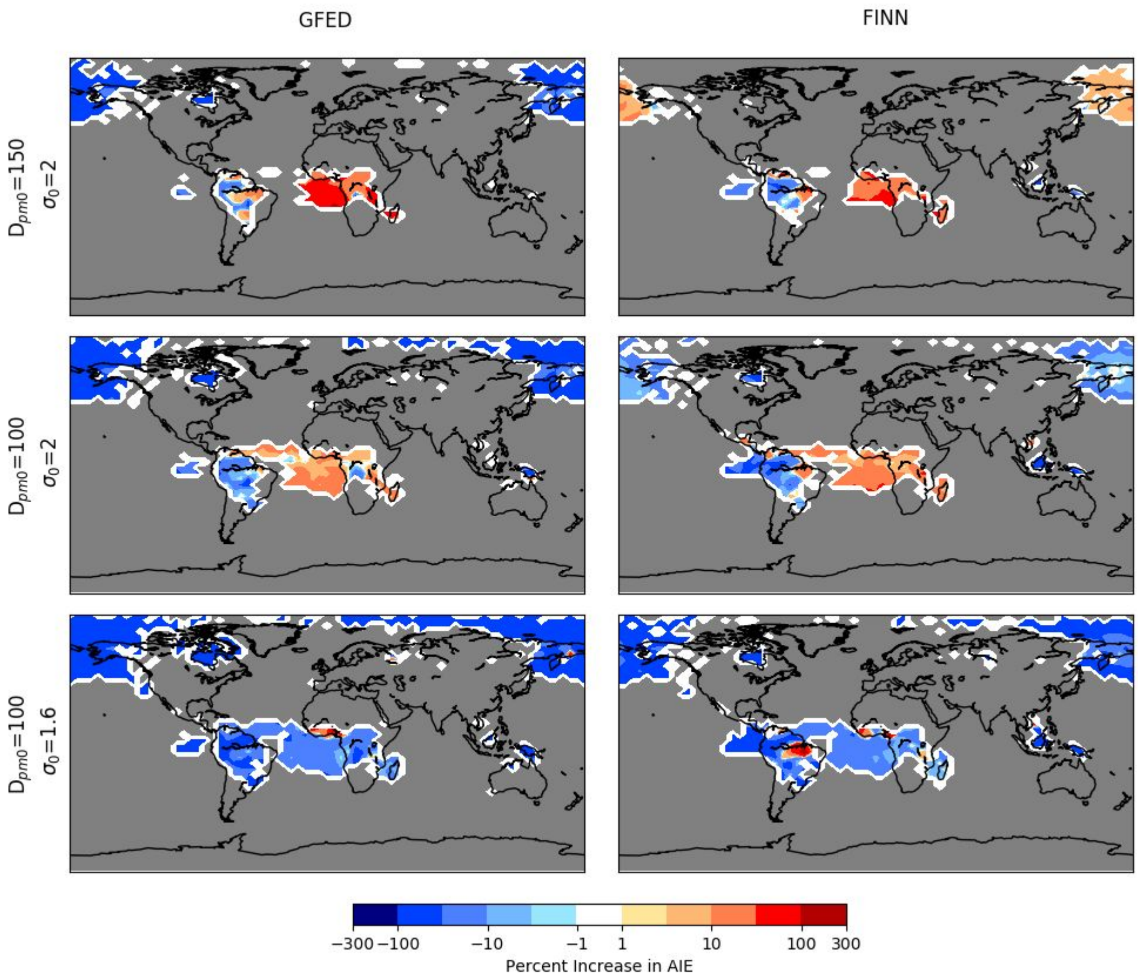

Figure S17: Percent change in cloud albedo aerosol indirect effect (AIE) of biomass burning aerosol due to sub-grid coagulation and GFED emissions (left) or FINNv1 emissions (right) and an assumed in-plume coagulation time of 24 hours. Panels in the top row have an emitted median diameter of 150 nm and an emitted modal width of 2. Panels in the middle row have an emitted median diameter of 100 nm and an emitted modal width of 2. Panels in the bottom row have an emitted median diameter of 100 nm and an emitted modal width of 1.6. Positive (red) values correspond to an increased cooling tendency of biomass burning AIE due to sub-grid coagulation being added. Regions in grey indicate that the AIE due to biomass burning is a more positive value than -100 mW m^{-2} (i.e. there is less than 100 mW m^{-2} of cooling) with or without sub-grid coagulation (Figs. 9 and S16).

10. Figure 6:
This is a nice figure and should be used to summarize conclusions obtained in this study. I suggest the authors to move this figure to the last paragraph of section 3.3 (after Figure 9).

Similar to comment 9, differences between SubCoag and noSubCoag can be added to this figure. Adding them will make the impact of sub-grid coagulation on DRE and AIE clearer.

We have chosen to leave this figure where it currently is because we would like the reader to be able to look at all of the results in context as they read this section, but our intention is for this to be a summary figure with its main discussion coming at the end of the section. We have made this addition to Section 3.3 to clarify: "Figure 6 summarizes our radiative-effect findings with global, annual-average values for each simulation."--> "Figure 6 summarizes our radiative-effect findings with global, annual-average values for each simulation, and we will discuss this figure in detail at the end of this section."

11. Page 19, Line 19 – Page 20, Line 6:
Please clarify how the authors estimated the statistic values in this paragraph (131%, 79%, 64%, 62%, and 49%).

Originally, these numbers were used in an attempt to include all sensitivity cases in the calculation. For the numbers comparing two initial size-distribution assumptions, the cases with sub-grid coagulation of biomass burning aerosol and the cases without were both used in the calculation. For the numbers discussing the addition of sub-grid coagulation of biomass burning aerosol, all initial size distribution cases were used in the calculation. This approach made things unnecessarily complicated, and the numbers have been replaced with simpler calculations that should be more intuitive for the reader. For the numbers comparing two initial size distributions, the case without sub-grid coagulation of biomass burning aerosol is now used alone. For the numbers discussing the addition of sub-grid coagulation of biomass burning aerosol, only the default initial size distribution case is now used (D_{pm0} being 100 nm and σ_0 being 2).

These changes are in section 3.3 where the text is now, "Biomass burning contributes nearly three times more to N80 when decreasing the modal width (a 43% increase in global N80 from adding biomass burning with the decreased width versus a 14.5% increase from the original width) without sub-grid coagulation of biomass burning aerosol, as seen in panels c to e in the four figures (4, 9, S9, and S16). This increase in N80 leads to a 104% increase in the magnitude of the globally, annually averaged AIE due to biomass burning, from -76 mW m^{-2} to -29 mW m^{-2}. Biomass burning contributes only about one-third as much N80 when increasing the median diameter (a 4.8% increase in global N80 from adding biomass burning with the larger median diameter verses a 14.5% increase from the original median diameter) without sub-grid coagulation of biomass burning aerosol, as seen in panels c to a in the same four figures. This decrease in N80 leads to a 62% decrease in the magnitude of the globally, annually averaged AIE due to biomass burning, from -76 mW m^{-2} to -29 mW m^{-2}. Sub-grid coagulation similarly decreases the biomass-burning contribution to N80 from 14.5% to 9.2% when assuming the original initial size distribution case (D_{pm0} =100 nm and σ_0 = 2). Hence, sub-grid coagulation decreases the biomass-burning AIE by 43% globally (-76 mW m^{-2} to -43 mW m^{-2})".

12. Page 21, Lines 21-25:
Can the authors add some statistics for more quantitative discussions of this paragraph?

To make this discussion quantitative, the bullet point in question now reads "In the current model set-up, it is assumed that smoke plumes do not overlap. Overlapping of smoke plumes would lead to a higher initial number concentration and slower dilutions and therefore more sub-grid coagulation. The impact on N80 when the smoke plumes are allowed to completely overlap (i.e., all fires in the gridbox form one "superplume") is shown in Fig. S18. Without sub-grid coagulation of biomass burning aerosol, biomass burning increases N80 by 10.4% (globally and annually averaged). With sub-grid coagulation of biomass burning aerosol as it is generally presented in this paper (no overlapping plumes), the increase in global N80 due to biomass burning is decreased by about a quarter to 8.0%. When sub-grid coagulation of biomass burning aerosol includes total overlap of the smoke plumes, the increase in global N80 due to biomass burning is further decreased to only 0.5%. This strong sensitivity of our results to plume overlap highlights that the degree of plume overlap likely needs to be understood".

13. Section 3.4:
In addition to the points raised by the authors, I suggest to add the following two points to this section.

Firstly, the simulations made by the authors are for year 2010 only. Biomass burning emissions and meteorological conditions have large year-to-year variability. Please add some discussions on the features of biomass burning emission in 2010 (compared with other years) and on their potential impacts on the estimation of sub-grid coagulation importance.

This is a very good point. We have added the following caveat to Section 3.4: "We only simulated emissions and meteorology for 2010. As there is significant interannual variability in biomass burning emissions as well as the meteorological inputs to the Sakamoto et al. (2016) parameterization, we expect that our results are at least somewhat sensitive to the choice of year (O'Dell et al., 2019)".

We did not do a detailed comparison of emissions between years. Given the range of factors that contribute to the impact of biomass burning on particle concentrations, radiative forcing, and the Sakamoto et al. (2016) parameterization, it would be very challenging to speculate about how the results for 2010 may specifically be different from other years.

Secondly, the uncertainty ranges of DRE and AIE in this study (e.g. Figure 6) were estimated from sensitivity simulations with changing single parameter at one time (e.g., median size of emissions, sigma of emissions, mixing state, subcoag timescale, biomass burning emission data). However, in the real atmospheric conditions, multiple parameters change simultaneously.

Therefore, the uncertainty ranges of DRE and AIE in the real atmosphere might be larger than those estimated with single parameter change (conducted in this study). The authors can add discussions on the potential importance of this effect.

We have made this addition to Section 3.4: "Because sensitivity simulations varied only one parameter at a time, uncertainty ranges of DRE and AIE in the real atmosphere may be larger than those estimated in this study. Uncertainty in other factors -- such as emissions, clouds and their susceptibility, and brown carbon -- also affect DRE and AIE uncertainty ranges".

14. Page 23, Line 26:
the estimated DRE –> the estimated DRE (increases cooling)

We have made this change.

15. Page 24, Line 2:
Sakamoto et al. (2017) –> Sakamoto et al. (2016)

We have made this change.