Responses to Referee's Comments

We appreciate careful reading and lots of valuable comments. We wrote referee's comments in black, our responses to comments in blue and italics, and the revised manuscript in green.

Anonymous Referee #1:

This manuscript studied the sensitivity of SSA calculations to the physical properties of absorbing aerosols, namely Black Carbon (BC), Brown Carbon (BrC), and dust in the GEOS-Chem 3-D global chemical transport model using a post-processing tool for the aerosol optical properties (FlexAOD). Sensitivity studies were performed to study the influences of the physical parameters of BC, mixing state, dust size distributions, and the presence of BrC on the calculation of SSA. However, it seems that the authors only assessed the sensitivity of one small component of the model (FlexAOD) to selections of some parameters for a fraction of the filtered AERONET sites. Assessing the impact of most of these parameters has been widely done in the literature (e.g. Boucher and Anderson, 1995; Sokolik and Toon, 1996; Liao and Seinfeld, 1998; Haywood et al., 1997; Myhre et al., 2002). It seems that the present work does not add new information. It does not provide a systematic estimation of the uncertainties in these parameters, does not reduce the uncertainties at a global scale by using the AERONET measurements, and does not reduce the uncertainty in the simulated SSA by combining the model and observation estimates (e.g., Bayesian). Thus, the present manuscript lacks the global significance and the broad scientific significances that can improve the simulations of SSA. I do not recommend publishing the present manuscript in the journal of ACP. Some detailed comments are below:

➔ *Thank you so much for careful reading and valuable comments. The main purpose of this study is to investigate the effects of different physical assumptions on SSA by comparing model results against observations including surface $PM_{2.5}$, AOD, and SSA. We set up several sensitivity cases, and every sensitivity case was evaluated against observed SSA at different wavelengths. These heavy evaluations differ from previous studies focusing on calculated sensitivities. Although we used the single radiative effect calculation method (FlexAOD), this method has been widely used in global modeling studies and should not be importantly different from the method of other models using Mie algorithm.*

Line 12, Page 6: The physical process is not clearly described. How does the model consider the conversion of hydrophobic to hydrophilic BC (or OA)? First, the authors did not explain how they compute the optical properties for hydrophobic and hydrophilic BC (or OA), which is particularly important in their simulations of internal mixture. Second, if the authors adopted a fixed aging rate for BC (or OA), it can affect the simulated concentrations of BC (or OA).

➡️ *Hydrophobic components of BC and OA become hydrophilic with an e-folding timescale of 1.15 days. Hydrophilic OA constitutes shell and hydrophobic OA is treated as the core for the internal mixing calculation. BC is assumed as core regardless of its hygroscopicity. We added the text as follows:*

Hydrophobic components of BC and OA converted to hydrophilic with an e-folding time of 1.15 days.

Hydrophilic OA constitutes shell and hydrophobic OA is treated as core for the internal mixing calculation. BC is assumed as the core regardless of its hygroscopicity. Other aerosols including inorganic, dust, and sea salt constituted the shell. A more detailed description of the aerosol optical property calculation with different mixing states can be found in Section 2 of Curci et al. (2015).

Line 20, Page 7: Please specify the temporal resolution of GFED data used in this study. If the authors used the annual mean data, it should affect the simulated seasonality of BC and BrC.

➡️ *We used the monthly mean data. We added the text as follows:*

The Global Fire Emission Database version 4 (GFEDv4) inventory (Giglio et al., 2013) was used for biomass burning emissions at a 0.25° x 0.25° spatial resolution and monthly temporal resolution.

Line 9, Page 8: Regarding the homogeneous internal mixing, it is unclear how the authors treat this mixing case in their simulations (Maxwell Garnett or volume mixing?).

Line 15, Page 8. As aerosol optical property calculation is important for the present paper, the authors can give more information.

➡️ *We added the text as follows.*

In case of internal mixing, two widely used internal mixing representations were used. Homogeneous internal mixing assumed that all aerosols were well mixed, and the core-shell internal mixing assumed that an insoluble well-mixed core was coated by a concentric well-mixed soluble shell. In both assumptions, the refractive index was calculated as the volume-weighted average of the components. Calculating optical properties approximated the integrals for the Mie efficiencies by dividing the size range into 100 geometrically spaced bins, and then calculated the wet volume (hygroscopic growth is considered) concentration of each species in the well-mixed particle, or the well-mixed core and the shell, in each size bin from the sum of all log-normal modes.

In case of homogeneous internal mixing, the Mie efficiencies (extinction, absorption, and scattering) were calculated in each bin for a monodisperse aerosol of radius r using the

Mishchenko et al. (1999) code. Then extinction, absorption, and scattering coefficients were calculated summed over the whole size distribution.

In case of core-shell internal mixing, the same averaging procedure was applied separately to the core and the shell. The Mie efficiencies are calculated in each bin for a monodisperse aerosol of radius r and the calculated core-to-shell volume ratio using the Toon and Ackerman (1981) code for stratified spheres. Hydrophilic OA constitutes shell and hydrophobic OA is treated as the core for the internal mixing calculation. BC is assumed as core regardless of its hygroscopicity. Other aerosols including inorganic, dust, and sea salt constituted the shell. A more detailed description of the aerosol optical property calculation with different mixing states can be found in Section 2 of Curci et al. (2015).

Line 25, Page 8. It is unclear how the authors changed the size distribution of aerosols in the model. First, it lacks a description of the method on how GEOS-Chem treats the size distributions in the model. Does it use a bulk scheme, or a sectoral scheme? If I understood it correctly, the model used by the authors uses a bulk method. When the authors change the size distributions in FlexAOD, did the authors also make the same changes in the GEOSChem transport model? This is important, because the size distributions also affect the lifetimes of BC and BrC in the transport. Please make it clear and justify the method.

➔ *We agreed with the reviewer's comment. However, we thought the deposition change by the change of size distributions of $PM_{2.5}$ aerosols were not significant. Koch et al. (2009) conducted sensitivity test by changing BC effective radius from 0.06 μm to 0.1 μm, which resulted in 3% change of global BC burden (from 0.36 to 0.35 mg m^{-2}).*

Line 25, Page 8. The authors can consider showing three maps for the site locations of the AMS, SPARTAN and AERONET networks. Even it is helpful to zoom in one or two regions to show the relationship (overlap) of these networks.

➔ *We added three maps in the supplementary figures.*

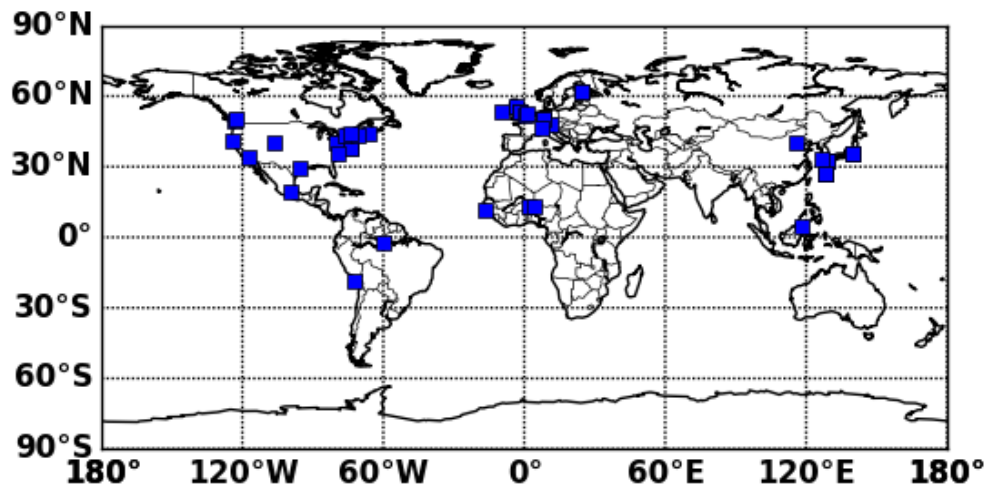



Figure S3. Global distribution of AMS sites used in this study.

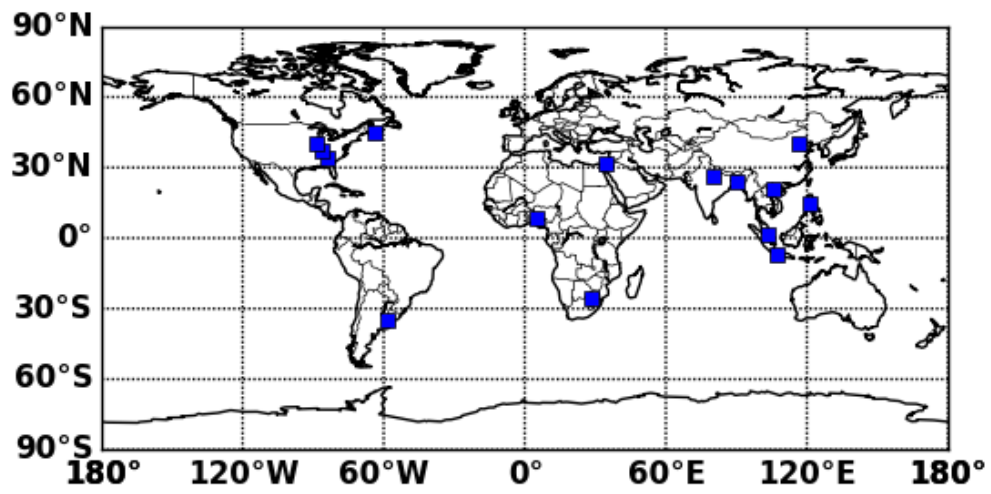



Figure S4. Global distribution of SPARTAN sites used in this study.

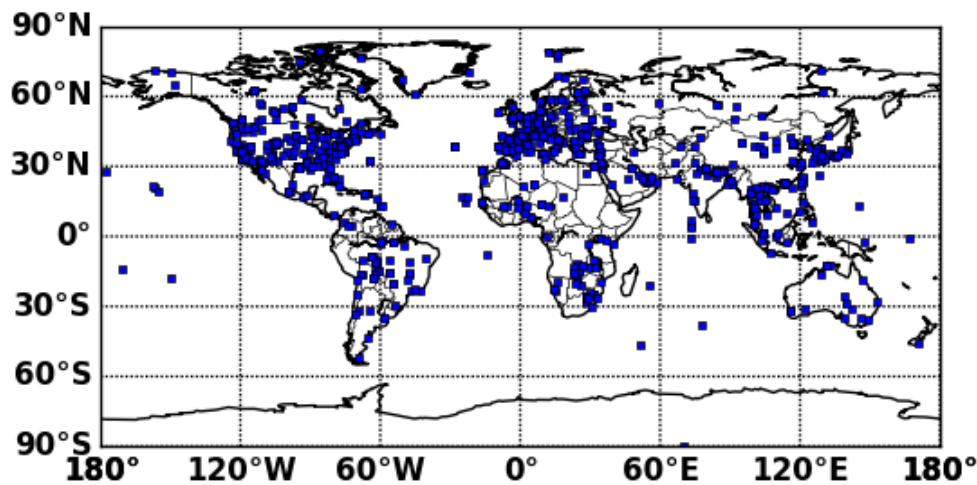



Figure S5. Global distribution of AERONET sites used in this study.

Line 20, Page 9: Please change "X" to "×".

➔ *We changed it.*

Line 19, Page 13. The authors need to cite a reference to support this statement.

➔ *We added the references as follows.*

The spatial resolution of the model (2° x 2.5°) was too coarse to capture local sources on the island (Li et al., 2016; Chen et al., 2009).

Line 6, Page 14: The authors should confirm that the two studies are using the same observational network before doing such a comparison.

➔ *We removed this part and changed the text as follows.*

Simulated PM$_{2.5}$ concentrations were generally in good agreement with the observations (R = 0.76) with a slight low bias observed for the mean concentrations (-24 %). However, the model significantly overestimates or underestimates individual aerosol concentrations in some observation sites. We screened out some poor representation sites based on

criteria, which was described in the next section.

Line 10-20, Page 14: It is not clear how the authors "selected simulate results at AERONET sites". There are two possibilities: first, they had run N simulations, but used only a fraction of these simulations to compare with the AOD and SSA at all AERONET sites; second, they had run N simulations, but compared all these simulations with the AOD and SSA at a fraction of AERONET sites. Please make it clear.

➔ *The second reflects our intention. We changed the text as follows:*

The model appeared to appropriately capture the spatial and temporal (monthly) variability of the observed PM2.5 described above. This may result from the combination of the nitrate overestimation and the OA underestimation. To ensure the reliability of the model for the AOD and the SSA evaluation in this section, we screened out AERONET sites which did not satisfy the following criteria:

Line 5-17, Page 15 and Figure 5: Regarding the AERONET data, it is unclear if the authors are using the Level 1 or Level 2 data. It is unclear if the authors are comparing the daily, monthly or the yearly mean SSA between model and measurements. It is also unclear if the authors are comparing the SSA for exactly the same days between model and measurements or not. Please make them clear.

➔ *We used Level 2 the monthly mean AERONET product. We compared our monthly mean model results against the AERONET. We think the evaluation results will not be significantly changed (within 0.01 for mean SSA) even if we use daily AERONET product, based on the study by Schutgens et al., (2016). We added the text as follows.*

Here we used monthly mean AERONET and model outputs for 2008 – 2010. We noted that temporal averaging without collocation could cause significant errors up to 0.05 for SSA in specific region and time (Schutgens et al., 2016; Schutgens et al., 2017). However, mean temporal sampling error caused by using monthly averaged SSA is less than 0.01 (Figure 16 in Schutgens et al., 2016), which is less than the mean model error in this section (~0.05).

Line 10, Page 15: "the model underestimated the observed absorption (1-SSA) by 50 %". According to this sentence, it seems to say that absorption is equal to (1-SSA). Thus, the authors neglected the fact that the absorption depends on the mass of BC as well as the SSA. It can be rephrased.

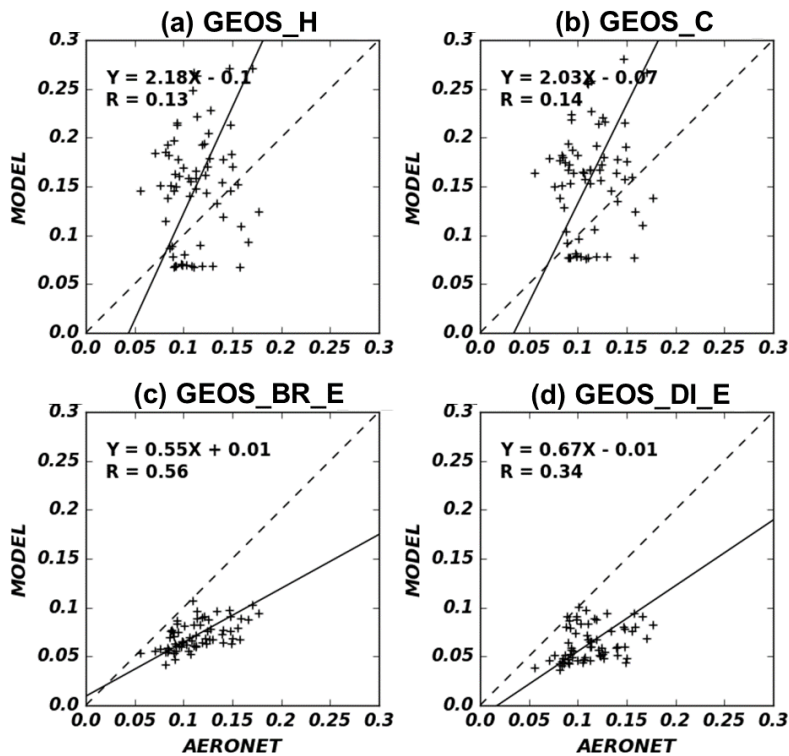
➔ *Thank you for the comment. We agreed with the comment and changed the text as follows:*

The SSA of the model (0.949) was higher than that of the AERONET (0.897) by 0.052. In other words, the model underestimated the observed single scattering co-albedo (1-SSA) by 50 %.

Line 3, Page 16: The SSA should be sensitive to the imaginary part of the dust refractive index (Liao and Seinfeld, 1998), which is not considered.

➡ *We agreed with the reviewer's comment that different dust imaginary refractive index can affect SSA. However, maximum imaginary refractive index of dust is ~0.0055 (Curci et al., 2015) which is similar to the maximum imaginary refractive index of inorganic aerosols (~0.006, Curci et al., 2015), and significantly lower than BC (~ 0.79, Bond and Bergstrom, 2006) or BrC (0.27, Alexander et al., 2008). Therefore, we think it is not the main driving factor for SSA calculation.*

Line 20, Page 16: Better to show (1-SSA) than SSAs, since absorption is proportional to (1-SSA).

➡ *We plotted the figure as follows for (1-SSA). We agreed with the reviewer's comment but we think SSA is better than (1-SSA) for the discussion since readers are more familiar with SSA, and regression values between SSA and (1-SSA) comparisons showed same values for the slope and correlation.*

References: Boucher O., and T. L. Anderson, 1995: GCM assessment of the sensitivity of direct climate forcing by anthropogenic sulfate aerosols to aerosol size and chemistry. J. Geophys. Res., 100, 26 117–26 134. Liao H, Seinfeld J H. Radiative forcing by mineral dust aerosols: sensitivity to key variables. Journal of Geophysical Research: Atmospheres, 1998, 103(D24): 31637-31645. Haywood J M, Roberts D L, Slingo A, et al. General circulation model calculations of the direct radiative forcing by anthropogenic sulfate and fossil-fuel soot aerosol. Journal of Climate, 1997, 10(7): 1562-1577. Myhre G, Stordal F, Berglen T F, et al. Uncertainties in the radiative forcing due to sulfate aerosols. Journal of the atmospheric sciences, 2004, 61(5): 485-498. Sokolik I N, Toon O B. Direct radiative forcing by anthropogenic airborne mineral aerosols. Nature, 1996, 381(6584): 681-683.