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**Interactive
comment**

Interactive comment on "Key factors affecting single scattering albedo calculation: Implications for aerosol climate forcing" *by* Duseong S. Jo et al.

Anonymous Referee #3

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This paper addresses an important topic: which factors impact the optical absorption of modelled aerosol and how does it affect aerosol forcing calculations.

Unfortunately, the paper is very limited in scope. The analysis is performed for a single model and just few factors while many others are not discussed. There is no coherent argument presented that the factors discussed in this study are the most important ones. E.g. wavelength dependence of imaginary refractive indices and wet growth are completely ignored. While the authors present this paper as a sensitivity study it feels more like a small tuning study (and perhaps should be published as a technical paper).

However, the authors make use of a very interesting dataset that comprises both species concentrations and optical measurements. This dataset offers the possibil-

ity of testing modelled optical properties as a function of chemical mixing state. The authors have done this to some extent but I feel so much more is possible.

Authors are also commended for linking changes in optical properties to aerosol radiative effects, although I would like to have seen an analysis of not just Direct Radiative Effect but also Present Day - PreIndustrial forcing.

The absence of a proper description of the evaluation methodology for optical properties means I cannot judge whether it is appropriate or not. However, what information is presented suggests that there may be significant sampling errors in their comparison of modelled and observed SSA.

On the whole, the paper provides too little substance and may have used improper methodology and I suggest rejection. I encourage the authors to consider expanding their analysis of GOES-Chem model evaluation using species concentrations and optical properties simultaneously for a future publication.

Specific comments

One paper that deserves mention is Goto et al GRL 2011 as it seems to apply a very similar methodology. Klingmueller et al GM 2014 studies different mixing approximations and their impact on optical properties. Two other papers that deserve more attention are Schulz et al ACP 2006 and Myhre et al ACP 2013. Both describe variations in optical properties across AEROCOM models and are directly relevant to the present work. Myhre et al is quoted in the current paper but in a different context.

Abstract p 2, l 24: please state values of DRE (before & after) as 'increased' is ambiguous in this context.

p 3, l 4: Iaconno et al is a strange reference here as its main topic is long-lived greenhouse gases

p 3, l 5: "most contributes to the uncertainty of the aerosol DRF calculations". Please provide references.

p 3, l 9: "For example, a decrease in the SSA from 0.9 to 0.8" I would not call this a small change in SSA, given that it normally varies between 0.7 - 1.

p 3, l 10: the sign change highly depends on surface albedo which should be mentioned here

p 4, l 5: "which can cause a wide range of calculated aerosol SSAs" I agree with the authors but it's important to point out there are differences due to regional variations (unavoidable) and differences due to measurement uncertainties (in principle this allows improvement).

p 4, l 21: "variable for the mass median" âĂŤ> "variable in the mass median"

p 5, l 1: "internally mixed BC aerosol" I believe this depends on the assumed internal mixing. The authors assume either complete mixing or a solid core- liquid shell morphology. See also Klingmueller et al GMD 2014, see also Brunamonti et al GRL 2015, Liu et al Nature GeoSci 2017

p 6, l 9: "2° x 2.5° for this study" This is then the model resolution at which you preformed your simulations? Please state so in the text.

p 6, Sect 2: Please briefly describe the aerosol model: how many tracers do you use for aerosol? Do you use a modal scheme or a bin scheme. Is the modal scheme one or two-moments? How many modes/bins? To what extent is internal mixing explicitly calculated?

p 6, l 23: It's not clear if this BrC calculation is off-line or on-line? Do you use the same GEOS-Chem simulaton but add a correction to the fields to describe BrC or is BrC an additional tracer? Or do you simply assume part of organic aerosol is absorbing in FlexAOD?

p 8, l 6: Does FlexAOD in principle use the same method as GEOS-Chem to calculate aerosol optical properties? I assume that GESO-Chem calculates optical properties (for radiative forcing) and wonder how the two codes relate.

p 8, l 14: Even though a reference is provided to Curci et al 2015, please provide more details on the main assumptions. We are talking about essential assumptions in your study so please state them explicitly. E.g. homogeneous internal mixing includes all aerosol are mixed together (some models allow external mixtures of internal mixtures, e.g. coated BC and coated dust)? Also discuss how these assumptions relate to the GES-Chem simulation of aerosol. E.g. a full external mixture and a full internal mixture will show different wet deposition behaviours. I suppose some of your FlexAOD modelling may be inconsistent with GEOS-Chem modelling?

p 8, l 23: "updates to the method of Martin et al." Does Martin et al describe GEOS-CHem or FlexAOD?

p 9, l 15: "which makes it difficult to choose the appropriate BC radius in the Mie calculation" So how did you resolve this problem?

p 10, l 15: As far as I understand you do not really consider different morphologies (apart from core-shell) as all optical properties are calculated with Mie theory. Please state this clearly.

p 10, l 19: Please define BB, BBR and BBHR.

p 11, l 6: "We also implemented a new dust size distribution in the model as suggested" Implemented in FlexAOD or in GEOS-Chem? If in GEOS-Chem then please move this to Sect 2. If in FlexAOD, then wouldn't this imply an inconsitency between GEOS-Chem and FlexAOD?

p 12, l 13: "was degraded" Degraded compared to what? Earlier studies? Maybe AMS? In that case, say something like "Compared to AMS, the model performs worse with respect to SPARTAN".

p 13, l 7: "nitrate overestimation" See also recent AEROCOM comparison of nitrate simulation (Bian et al ACP 2017) which shows $\sim$ 10x difference amongst models

p 13, Sect 4.1: Fig 2 & 3 show evaluation of individual species. Can something be

said about how well the model simulates chemical mixing? If it overestimates sulfate when it underestimates BC, chemical mixing states are very poorly represented. But if it overestimates both, SSA may be quite accurate.

p 14, l 5: "The performance of the model was comparable to that of global PM2.5 estimates constrained by using satellite AOD observations" This sentence is not that clear, can you rephrase? van Donkelaar's PM2.5 evaluation yielded similar results to yours but they adjusted PM2.5 based on AOD?

p 14, Sect 4.2: Please describe how you compare model data with observations. I believe you only have monthly model data so you need to construct monthly AERONET data somehow. According to the caption of Fig 5, monthly AERONET averages were only calculated when there were more than 10 days of observations with AOT > 0.4? In that case monthly model data and monthly observations are based on very different temporal sampling. This will lead to large errors in the comparison, see Schutgens et al ACP 2016 (on temporal collocation) and Schutgens et al ACP 2017. Note that such errors need not be random but can be biases, especially because observations use AOT>0.4 but monthly model data do not. Please discuss.

p 14, l 17: can something be said about AOD errors as agreement with AMS becomes better and better? Or conversely, can you imagine a plot of AOD error vs AMS error? I wonder if AOT errors are (relatively) smaller or larger than AMS errors. Smaller AOT errors suggest balancing errors, larger errors suggest incorrect assumption on morphology, mixing state etc. I realise this is very hand-wavey but can the authors say something about this?

p 15, l 1: "From this result, we concluded that the aerosol optical property calculation in FlexAOD was properly performed". I am not sure what the authors mean by this. Clearly there are still errors. There is no proof that your calculation is bug-free. I admit it's a sanity test of your data. But the best you can conclude is that there are no major errors in either your GES_Chem calculation, your FLexAOD calculation or your

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observations.

p 15, l 8: In addition to Dubovik et al. 2002, see also: E. Andrews at al ACP 2017

p 15, l 19: "The model showed an improved result especially for the SSA at 440 nm". For 870nm, I see that R decreases a little bit. There is no info on RMS errors. I suggest removing 'especially' and explicitly describe the results for 870nm.

p 15, l 21: "we concluded that the model simulation capability of the AOD and SSA was improved when we selected AERONET sites where the model showed good results against the AMS and SPARTAN network" I think this conclusion is too strong, as there is no improvement for 870nm.

p 15: You have a very interesting dataset of both species concentrations and optical measurements. It is worthwhile to dig deeper into this dataset. How do the aerosol change when you filter the AERONET sites in Fig 5? It appears filtered AERONET sites have less absorbing aerosol. Do they also have less BC? Does SSA agreement improve when chemical mixing is better modelled? You filter by requiring species concentrations to be within 2x of observations but that does not necessarily mean mixing is better. You can also adress questions like: are dust-heavy cases better modelled than BC-heavy cases, etc.

p 15, Sect 4.2: One thing I miss in this Section is the realization that you are talking about fundamentally different observations: optical column measurements and surface measurements. This creates uncertainty whenever you interprete SSA model results and needs to be acknowledged.

p 15, discussion of Fig 5: The most obvious reason why SSA might agree at 870nm but is overestimated at 440nm is of course wavelength dependency of imaginary refractive index. However, the authors do not really discuss this. Note that due to the temporal sampling issue I discussed earlier, I'm not convinced that the agreement at 870nm is undisputed.

p 16, Sect 5: There is a difference between a sensitivity study (as the Section's title inidcates) and a parameter estimation (as the first paragraph indicates). Please decide which one you want to do and adjust the text accordingly.

p 16, l 18: I still don't understand how BrC 'works'. GEOS-Chem simulates organic carbon but assumes zero imaginary refractive index? So in FlexAOD you now assume that part of the organics are actually absorbing? Please describe this earlier in the model or FlexAOD Sections.

p 17, l 5: "this remaining gap could be reduced when we consider strongly absorbing BrC" Many reasons can be used to explain this. Why do the authors ignore e.g. wavelength dependence of BC? Your dataset of both sepcies conentrations and optical properties can give you hints as to what is the problem?

p 17, l 11: The authors never really discuss how they define the radius of BC. Clearly the Mie sphere is a bad approximation for the strands of black carbon observed in nature.

p 17, l 17: Why is BC only contributor? I cannot find information on the wavelength dependence of BrC absorption in the paper.

p 18, Sect 6: It's a pity the authors do not provide a similar analysis for direct aerosol forcing (DRE Present Day - PreIndustrial). I surmise in that case, dust would not be the main cause of sensitivity but perhaps BrC.

p 19, Sect 7: I suggest the authors start the Conclusions with a brief recap of their study. It also is better to prevent use of abbreviations like GEOS_BR_DI_E, OP_E, BB_E which will only be obvious to people who have read the full paper.

p 19, l 18, Finding 1: Actually what is critical to SSA calculation is the Mass Absorption Coefficient (MAC), which is the ratio of AAOD by column load (and so includes density). This MAC is still uncertain, see Boucher et al PNAS 2016

p 20, l 24: "resulting in a decrease of the positive SSA bias at 440 nm generally found

in global models." The authors have only shown this to be the case for one model (GEOS-Chem)

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Discussion paper