

Thanks to the referee for his/her very helpful suggestions, which have allowed us to clarify and improve the manuscript. Below we addressed the reviewer comments, with the reviewer comments in italic and black, and our response in bold and blue. We have revised the manuscript accordingly.

General Comments

Aerosol effect on climate remains one of the largest uncertainties in projecting future climate change. This work uses the GEOS-Chem-APM model to investigate the aerosol direct radiative forcing and its uncertainties to a number of parameters. This work is relevant to the scope of Atmos. Chem. Phys. I have a few general comments and a number of specific comments that the authors need to address:

General Comments

1. There have been a lot of studies before to investigate aerosol direct radiative forcing. The authors need to make it clear in the introduction what are new in the study and how the methods and/or conclusions from this study differ from previous ones.

As the reviewer mentioned, there have a lot of studies previously to investigate aerosol direct radiative forcing (DRF). However, aerosol DRF still has a large uncertainty, and further investigations are needed. This study has several unique aspects. Firstly, we employ the GEOS-Chem-APM to examine aerosol direct forcing in our study. The GEOS-Chem is a global chemical transport model. It treats the comprehensive chemical reactions and produces the prognostic chemical species, which provides more reasonable chemical fields for aerosol production, compared to the prescribed chemical field as done by many global climate models. Secondly, we use a prognostic multi-type, multi-component, size-resolved aerosol microphysics model (APM). The model includes state-of-the-art nucleation schemes and condensation of low volatile secondary organic compounds from successive oxidation aging. Many previous modeling studies often assume particles are either externally mixed (particles of different components totally separated, i.e., zero mixing) or internally mixed (all aerosol components are completely mixed, i.e., 100% mixing). In the real atmosphere, particles exist between these two extreme cases and the degree of the mixing states of particles varies with time and location. The GEOS-Chem-APM is designed to explicitly predict the spatiotemporal variations in the degree of particle mixing which is important for their climatic impact through both direct and indirect effect. Thirdly, the model is driven by assimilated meteorology, which is presumably more reasonable than the model-produced meteorology.

We have added more descriptions in the introduction to emphasize what are new in this study.

2. There is some assumptions regarding the pre-industrial aerosols in this study, i.e., assume all biomass burning aerosols are from the anthropogenic source. Certainly this assumption is not correct. There were a lot of biomass burning emissions in the pre-industrial times (e.g., year 1850 or 1750), and in some regions (e.g., U.S.) the pre-

industrial emissions were higher than present emissions. The more robust way is to run simulations with pre-industrial emissions. From the flux difference between present-day and pre-industrial day simulations to derive the direct radiative forcing.

The emission inventories in GEOS-Chem model include sulfur emission and carbonaceous emission. Sulfur emissions are contributed from the different emission sources, including anthropogenic and natural sources. For anthropogenic carbonaceous emissions from fossil fuel and bio-fuel, there also have the corresponding emission inventories. But for emissions from biomass burning, the inventory does not separate the sources to anthropogenic or natural. In our study we can only simply assume that all biomass burning aerosols from anthropogenic aerosols in order to investigate the radiative forcing due to anthropogenic aerosols. We acknowledge that this assumption is not likely a good assumption. We will communicate with GEOS-Chem community to include new biomass burning emission inventories (separate anthropogenic and natural sources), and re-visit the forcing later.

3. Cloud fields (cloud fraction, cloud water) play an important role in the all-sky aerosol direct radiative effect and radiative forcing. It seems that the cloud effects in this study may be underestimated. The authors should add some discussions of cloud fields used in the model.

Agree. We included a short description on cloud fields in section 2.3.

Specific Comments

1. P195. There are a few places saying the AeroCom project aiming to “reduce” the uncertainties. I don’t think this is the achievable goal for AeroCom. The more realistic goal is to quantify the uncertainties.

We agree. Although the ultimate goal of such aerosol projects is to reduce the uncertainties, the more realistic goal currently is to quantify the uncertainties. We have modified the text accordingly.

2. P196. L12. About Stier et al. (2007). “the values of refractive index”. Is it for BC? The number “1.21 W m⁻²” and other numbers here, are they global annual means?

Yes, the values of refractive index mentioned here is for BC. The radiative forcing cited in the text is global annual mean.

3. P196 and 197, when you are discussing the uncertainties of aerosol direct forcing, you need to cite the Liu et al. (Uncertainties in global aerosol simulations: Assessment using three meteorological datasets. JGR, 112, D11212, doi:10.1029/2006JD008216, 2007). This work found that the uncertainties of aerosol direct forcing can be more than a factor of 2 with different sets of input assimilated meteorology (because of differences in cloud fields which are not usually assimilated).

Thanks for the suggestion. We cited this paper in the introduction.

4. P197, L18-20. I don't think the predicted aerosol in models is more accurate than the prescribed aerosol in global climate models. The prescribed aerosol in global climate models is usually taken from global aerosol-climate models or from global chemistry transport models. The reason that prescribed aerosol is used in global climate models is to save computer time since climate models need to run for decades to century long.

In the global climate models with the prescribed chemical species, aerosol produced via these prescribed chemicals including O₃, H₂O₂ etc., which are generally in time resolution of monthly mean. The full chemistry model like GEOS-Chem provides the prognostic chemical species, which allow us to predict more reasonable aerosol production compared with the models using prescribed chemicals.

In addition, for those modeling studies with the prescribed aerosol, aerosol size distribution is often assumed, e.g. log-normal distribution as done by many previous models. But in the real atmosphere, aerosol size distribution varies with time and location. The predicted aerosol model could predict a spatiotemporal aerosol size distribution, which significantly impact aerosol optical properties and their radiative forcing. The GEOS-Chem-APM is designed to capture the main particle properties (sizes, compositions, coating of primary particles by secondary species, etc.) important for radiative forcing. A number of computationally efficient schemes are used to keep the computational costs affordable.

5. P197, L21. That emission in GEOS-Chem is kept up to date does not necessarily mean that emissions is consistent with the real conditions and thus biases in emissions are reduced.

Agree. We modified the text accordingly.

6. P199, L28. The "refractive index" of all aerosol species? Please give the values you use in the model.

We added a new table (Table 1) and a short description in Section 2.2 to show the values of refractive index.

7. P202, Section 3.1. The emissions are not for 2006. Right?

The original emission is not for 2006. For example, sulfur emission inventory (from EDGAR) is for 2000. In GEOS-Chem, annual scale factors are used for anthropogenic emission, i.e. the factors will scale emissions for the year that we select (e.g. 2006)

8. P202, L15-16. Since your model considers aerosol microphysics, do you still need the e-folding time of 1.2 days for the aging of carbonaceous aerosol?

We can predict the coating on the particles based on our aerosol microphysics. This information is used in the model for the calculations of optical properties. We still use the e-folding time to convert hydrophobic to hydrophilic carbonaceous aerosols since their wet scavenging rates are different. If a scheme to calculate wet scavenging rates as a function of coated materials is available, then no more e-folding aging time is needed. This will be the subject of future research.

9. P202, L18-29. Please give the size bins for dust and sea salt used in the model, and also the total emissions for dust and seasalt.

We added a short description in Section 2.1.1 to give the information including size bins (for SP, sea salt and dust) and mode diameter (BC and POC) used in the model. We also included the total emissions of dust and sea salt to Table 3.

10. P203, L10. “SP”. Please define it when in the first time use it.

“SP” is defined in Section 2.1.

11. P203, L23. “The fraction of sulfate from anthropogenic aerosols”. Be explicit.

We modified the text to “The anthropogenic fraction of sulfate”.

12. P205. Section 3.2. I would suggest comparing modeled AAOD or SSA with AERONET data.

Thanks for the suggestion. We compared the modeled AAOD with AERONET observations. The comparison is shown in Fig. 4 (the old Fig. 4 is replaced). We also modified the text in Section 3.2.

13. P207, L1-5. Compare these numbers with other modeling studies (e.g., Liu et al., 2007) and satellite studies (e.g., Yu H, et al., A review of measurement-based assessment of aerosol direct radiative effect and forcing, Atmos. Chem. Phys., 6, 613-666, 2006).

We added a comparison between our model results with the suggested modeling studies and measurements-based assessment. See the text in Section 3.3.

14. P209 L8. Change “cloud radius” to “cloud effective radius”

Thanks for the suggestion. We modified the text accordingly.

15. P210, L25-27. What are the mode diameters you used for POC and BC in the control simulations? I think these information should be added in the section 2 (model description).

We added this information in Section 2.1.1.