

Reviewer Comments on ACP-2011-412, “Climatic effects of 1950–2050 changes in US anthropogenic aerosols – Part 1: Aerosol trends and radiative forcing” by E. M. Leibensperger et al.

This paper calculates the trends of anthropogenic aerosols of sulfate-nitrate-ammonium from 1950 to 2050 and their direct and indirect radiative forcing over the U.S. The aerosols are first simulated using a CTM (GEOS-Chem) with fixed meteorological conditions (2001) as well as fixed natural and biomass burning emissions (so called “climatology”) but time-varying anthropogenic emission from fuel combustions. The resulting aerosol fields are then used by a GCM (GISS) to calculate the aerosol direct and indirect radiative forcing with decadal time resolution. Two major conclusions are (1) US anthropogenic emissions has already been reduced significantly such that at 2010 62% of the reduction estimated at 2050 level has already been achieved, therefore aerosol will be a weaker climate forcer and reducing it would cause further warming, and (2) BC forcing is rather weak so reduction of BC does not provide a “win” for reducing global warming.

There are certainly some interesting points in this paper and it is well written. But I feel that the major conclusions are based on a quite weak foundation, because (1) the model is not able to reproduce the atmospheric trends of anthropogenic aerosols other than sulfate over the US, and (2) the BC concentration was severely underestimated by the model. Efforts should be invested into building a stronger foundation for this study. In addition, the approach is mainly off-line without resolving the interactions between aerosol and clouds. My comments on the deficiencies of this paper are listed below.

1. Simulating aerosol trends. Simulating aerosols using time-fixed meteorological fields but changing emissions is valuable to separate the changes due to emissions from that due to meteorology. However it is not sufficient to present the actual aerosol trends, unless you can claim that meteorology doesn’t matter for aerosol loading. Except sulfate, the model does not reproduce the surface concentration trends of other aerosol species, i.e., ammonium, nitrate, BC, and OC. Because of the theme of this paper (and the companion paper) is to investigate the climate effects of the CHANGES of US anthropogenic aerosols based on the model results, the conclusions from this work would be problematic based on the incorrect trends. More importantly, some key radiative and microphysical properties for aerosol and clouds in calculating aerosol direct and interact radiative forcing (e.g., AOD, single scattering albedo, phase function or asymmetry factor, cloud droplet number concentrations and size, LWP) are not discussed or evaluated at all.

2. Aerosol and radiation/cloud systems are not interactive. I wonder why using two different models, GEOS-Chem for aerosols with the GEOS-4 meteorology for 2001 (2000 simulation is a spin-up), and GISS for clouds and radiation with the GISS GCM meteorology. These two met fields can be quite different, particularly in clouds, therefore the results could be inconsistent and the resulting conclusions could have large errors from these decoupled systems. There are no interactions between cloud and aerosols; no matter how much effects aerosols have on clouds (changing cloud

albedo and lifetime), clouds has no influence on aerosols. It seems that the change of clouds by aerosols does not even considered in the direct forcing calculations.

3. Mixing states. Two mixing states are tested when calculating the aerosol direct radiative forcing, one is 100% external mixing and one is 100% internal mixing of sulfate, nitrate, BC, and OC. It is stated that there are some differences in resulting direct forcing, i.e., total forcing with external mixture is 10% more than that with internal mixture, but I am not convinced that the difference is due to the decrease of BC absorption in the external mixture. I have several questions regarding the mixtures: How different are the AOD, single scattering albedo, and asymmetry factors are between the two mixing state assumptions? Why should BC absorption change from volume-weighted refractive indices of internal mixture? What is the hygroscopic growth of the internal mixture? Are the internal mixtures for anthropogenic sulfate, nitrate, BC, and OC, or they include those species from natural sources as well?

4. Direct forcing. The forcing calculation for the externally mixed aerosols should be done in the same method as for the internally mixed aerosols, i.e., total forcing should not be additive of each components separated, but should be calculated as a whole. The forcing from each component should be calculated as the difference between the total forcing and the forcing with that particular component excluded. Also, it is a common practice that the “forcing” refers the difference between the conditions of present day and pre-industrial time. This study does not consider the “pre-industrial” situation, which is fine but should be clarified.

5. BC forcing. It has been mentioned that BC forcing is rather weak compared to other aerosols. However, considering the model severely underestimating BC, than the positive forcing from BC would be much stronger.

5. Indirect forcing. The cloud droplet number concentration N_c is calculated based on the parameterization of sulfate and sea salt concentrations, which implies that other aerosol types don't matter. This approach has a serious problem as the sulfate has been decreasing faster than other anthropogenic aerosols, and will continue so in the future. Therefore, the parameterization (equation 1) developed from the conditions in the 90's or early 2000's when sulfate was the dominant anthropogenic aerosol is not suitable for predicting the N_c in the future.

6. Model evaluation. The model simulated sulfate, nitrate, ammonium, BC, and OC are evaluated against observed surface concentrations and wet deposition. I see several problems on the descriptions of those evaluations.

Example #1: Nitrate (page 24099, line 5-16). The authors attributed the model underestimation of nitrate wet deposition from 1980 to 2000 to emissions being too low. But at the sametime the model overestimates the nitrate concentrations; would that mean the emission being too high? Why deposition and concentrations are not consistent? If the overestimation of nitrate is because of more ammonium available to form nitrate due to the decreasing of SO_2 , does it mean there are too much ammonium in the model? The evaluation should be done more systematically considering all the processes.

Example #2: BC and OC (page 24099, line 17-the end of the paragraph). BC is severely underestimated by the model by more than a factor of 2 before 2000. The authors described that the previous GEOS-Chem studies did not show such severe bias because the observation-based emission was used. Then why not use the better emissions to produce more realistic concentrations and trends in this study, as conclusions of which would critically depend on the amount and change of aerosols?

Example #3: Comparisons of cloud properties with Chen et al. 2010b (Table 1). What year is this comparison based on? How do you explain that, with the same GCM and cloud parameterization, the Nc is much higher but TOA SW CF is lower in Chen et al. than those from this study but LWP values are similar?

7. The flow of text and figures does not go together. The contents in the later figures are discussed in the text before those in the earlier figures (e.g., Figure 2a, Figure 3, Figure 2b..., Figure 5 top, Figure 6 top, Figure 5 bottom, Figure 6 bottom...). Either the figures or the text should be rearranged to have the same flow.