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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Climatic effects of 1950–2050 changes in US anthropogenic aerosols – Part 1: Aerosol trends and radiative forcing" *by* E. M. Leibensperger et al.

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We thank Referee #2 for their valuable comments and suggestions. Their input has improved the manuscript.

Referee's comments are in plain text, our responses are boldface, and changes to the manuscript are italicized.

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

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.

We believe that the large changes in sources over this time period dominate the effects of meteorological variability and climate change on aerosol concentrations. We have added the following to Sect. 2.1:

Using the same meteorological year for the 1950-2050 simulation isolates the effects of emission changes. Climate change over that period might affect aerosol concentrations by ± 0.1 - 1μg m^{-3}, with even the sign of the effect uncertain (Jacob and Winner, 2009). Aerosol abundances are more sensitive to the large emission changes simulated here.

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.

We have added a table containing additional information on our aerosol distributions and direct radiative forcing calculations. Please see the table in the supplemental material to this response.

We have also added the following to Sect. 3, summarizing model performance:

Overall, the model shows mixed success in the simulation of 1990-2010 aerosol trends. We have an excellent simulation for sulfate, which accounts for 65% of anthropogenic aerosol mass and 78% of associated aerosol optical depth over the eastern US (Table 1). This gives us some confidence in the model representation of overall trends in aerosol radiative forcing, and we will quantify below the related error using the trend information in Fig. 3. In view of our underestimate of BC for the 1990-2000 period, we will focus our discussion of BC radiative forcing on present-day (2010) when the simulation shows no significant bias.

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 cloudalbedo 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.

We have added the following to Sect. 2.3:

Our use of monthly mean aerosol distributions archived from GEOS-Chem does not allow for feedbacks between aerosols and cloud properties. These feedbacks are likely very small relative to the changes of aerosol sources studied here. Using monthly mean aerosol distributions does not significantly bias our estimates of the direct radiative effect (Koch et al., 1999), but likely influences our calculation of the aerosol indirect effects due to the nonlinear relationship between aerosol amount and cloud droplet number (Jones et al., 2001).

The standard procedure for calculating aerosol direct forcing (e.g. IPCC AR4) does not include the change of cloud by aerosols. These semi-direct effects are considered in the companion study (Leiensperger et al., 2012) when simulating the climate response.

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?

The internal mixture consists of all sulfate, nitrate, OC, and BC. As in Chung and Seinfeld (2002), we assume the water uptake of internal particles is equal to the uptake of the externally mixed particles. Treating the aerosol particle as well-mixed increases the absorption cross-section of the particle. This is a common result (Chylek, et al., 1995; Jacobson, 2000) and has also been shown for

previous versions of this model (Chung and Seinfeld, 2002). Table 1 contains a comparison of the radiative forcing of internal vs external mixtures. We have made the following changes:

In Sect. 2.2:

As in Chung and Seinfeld (2002), we assume the water uptake of internally mixed particles is equal to the uptake of the externally mixed particles.

And in Sect. 4:

...assuming an internal aerosol mixture. The forcing increases by 10% for an external mixture due to lesser effect from BC absorption (Chylek et al., 1995; Jacobson, 2000).

As might be expected, an internal mixture has a slightly greater AOD, lower single scattering albedo, and is less forward scattering. We have added the following to Sect. 4:

When externally mixed, the radiative forcing is stronger, -2.2 W m^{-2}. The external mixture has 5% lower column integrated aerosol optical depth, 2% higher single scattering albedo, and 1% higher asymmetry parameter (forward scattering is increased) over the eastern US.

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.

We calculate externally mixed radiative forcing using this method. We have made

this clearer:

Radiative forcing is calculated by perturbing each aerosol component individually. The total anthropogenic aerosol radiative forcing is calculated by perturbing all aerosol components.

We have gone through the manuscript and ensured that the radiative forcing is understood to be anthropogenic (rather than pre-industrial) or US anthropogenic.

Additional information regarding external vs. internal mixtures is available in Table 1.

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.

BC is underestimated by a factor of 2 in 1990 and 2000 but by only 20% for 2010, i.e. present day. We focus on this latter period in our discussion of the significance of BC radiative forcing and now make this clearer. We have added the following to Sect. 3:

In view of our underestimate of BC for the 1990-2000 period, we will focus our discussion of BC radiative forcing on present-day (2010) when the simulation shows no significant bias.

And the Conclusions:

We have confidence in this result because of the ability of the model to reproduce observed BC concentrations in the US in 2010 (20% low bias).

5. Indirect forcing. The cloud droplet number concentration Nc 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

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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 Nc in the future.

Equation (1) relates the total number of moles of aerosol solute in a grid box to CDNC. The TOMAS simulations were conducted with a focus on sulfate and sea salt, but the relationship between the coefficients (A and B) and CDNC is general, and as shown by Chen et al. (2010) can be applied to global aerosol fields successfully. We do acknowledge though that A, B may change somewhat, and leave this to be addressed in a future study. We have added the following to Sect. 2.2 to make this clearer:

Since the relationship in Eq (1) relates total aerosol solute (in moles) to CDNC, the parameterization applies even if composition changes with the evolution of anthropogenic emissions.

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 SO2, does it mean there are too much ammonium in the model? The evaluation should be done more systematically considering all the processes.

Modeled values of ammonium concentration and ammonium deposition are in relatively good agreement with observations, suggesting ammonia is not the

issue. The cause of the nitrate discrepancies is unclear. We have added the following to Sect. 3:

The observed 2000-2009 decrease in nitrate, rapid decrease in sulfate, and near constant supply of ammonia seems at odds with current understanding of SNA thermodynamics. We did not explore this issue further, but it warrants future study.

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?

This work used the more recent emission inventory of Bond et al. (2007). We did not use the previous inventory in order to use a consistent dataset over the 1950-2000 period. The bias in BC is much reduced by 2010 and does not affect our conclusion that the radiative forcing of US anthropogenic BC is relatively small.

We have added the following to Sect. 3:

In view of our underestimate of BC for the 1990-2000 period, we will focus our discussion of BC radiative forcing on present-day (2010) when the simulation shows no significant bias.

And the Conclusions:

We have confidence in this result because of the ability of the model to reproduce observed BC concentrations in the US in 2010 (20% low bias).

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

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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?

Comparisons are made with "present-day" simulations and observations, which are typically close to the year 2000. LWP is somewhat constrained by the scale factor introduced in the autoconversion parameterization Eq. (3), so it is not too surprising that the values match despite differing N_c. Differences in TOA SW CF arise from the different spatial distributions of the N_c distributions. We have added the following to Sect. 5:

Chen et al. (2010b) report higher global mean N_c but similar cloud forcing, which may reflect differences in the geographical and vertical distributions of N_c.

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.

We have reordered the first 4 figures: Figs. 2a-2c are now Figs. 2, 4, 5; Fig. 3 remains Fig. 3. We feel the material presented in Figs. 5 and 6 are best presented together for comparison.

Added references:

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Jacob, D. J., and Winner, D. A.: Effect of climate change on air quality, Atmos Environ, 43, 51-63, 2009.

Jacobson, M. Z.: A physically-based treatment of elemental carbon optics: Implications for global direct forcing of aerosols, Geophys Res Lett, 27, 2, 217-220, 2000.

Jones, A., Roberts, D. L., Woodage, M. J., and Johnson, C. E.: Indirect sulphate aerosol forcing in a climate model with an interactive sulphy cycle, J Geophys Res, 106, D17, 20,293-20,310, 2001.

Koch, D., Jacob, D. J., Tegen, I., Rind, D., and Chin, M.: Tropospheric sulfur simulation and sulfate direct radiative forcing in the GISS GCM, J Geophys Res, 104, 23,799-23,822, 1999.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C15078/2012/acpd-11-C15078-2012-supplement.pdf

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