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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Anthropogenic aerosol radiative forcing in Asia derived from regional models with atmospheric and aerosol data assimilation" *by* C. E. Chung et al.

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We thank this reviewer for a very careful review. Our plan to address the reviewer's comments is as follows.

With regard to three major comments:

1. Data uncertainties.

Answer: The accuracy of MODIS and AERONET data has been studied and discussed in many previous studies. We will summarize these studies and add relevant references to the text. Due to the high uncertainty associated with MODIS fine mode fraction over



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land, we have limited the assimilation process to include the MODIS AOD fine mode fraction over ocean only. Our assimilation approach also takes into account explicitly estimated uncertainty in the observations as well as uncertainties in the modeled fields.

2. Model simulation of aerosols. There are several issues. For example, the anthropogenic emission is fixed for the 4-year period, nitrate and secondary organic aerosols are not included, hygroscopic growth of organic aerosols are not considered. The uncertainty/error associated with these omissions should be addressed. Most of all, not much evaluations with measurements over the studied domain are given, especially in aerosol compositions.

Answer: The use of constant emissions for the study period is a source of uncertainty. For example, Asia SO2 emissions grew by $\sim$5% a year during the period we simulated. As the focus of our simulations was not to detect aerosol trends but rather to capture a more robust mean estimate of analysis we chose not to try to build in emission trends (as these would have to be interpolated from 2000 to 2006 emissions, which were only recently published). Furthermore since we were constraining aerosol mass through AOD and SSA assimilation, the influence of changes in emissions to the extent that they are reflected in observed AOD, should be reflected in our constrained aerosol distributions. We have studied and evaluated the impact of the interannual variability in meteorology and emissions on sulfur trends in Asia. (Z. Lu et al., "Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000", (2010) just submitted to ACPD). This work indicates that during the period 2000 to 2004 our constrained results did capture the observed trends in sulfur, with the meteorological variability contributing significantly to the variability in observed SO2 and sulfate.

We did not include nitrates and secondary organics, and this is stated more clearly in the text. In the case of nitrates it contributes less than 5% to the aerosol mass in Asia. We know that SOA can be an important source of aerosol mass. However the uncertainties in primary OC emissions as well as secondary production remain very large. We have compared our OC calculated values with OC and OM observations,



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and in terms of mass they are very consistent. We have included the hydroscopic growth based on OC, but do not include explicitly that portion due to SOA.

We have carefully compared our predicted fields with aerosol observations in Asia from a variety of sources including at ABC sites and through AERONET. We have published these results for PM2.5, PM10, BC, OC dust, sulfate previously (for example Adhikary et al., 2008). For this study we have also compared with the limited absorbing AAOD observation during this period. We have found that the calculated AAOD are reasonably captured, with the values having a tendency to underestimate the values by $\sim$25%. More recently we have compared our sulfur predictions to observations in East Asia as discussed in the Lu et al., 2010 paper in review in ACPD.

We will add further discussion to the text to reflect our evaluation (we may end up adding a new figure).

3. Forcing. The reported forcing values are for all sky, while the assimilated AOD, SSA, etc. are for clear sky only. This is certainly not consistent. At least the clear sky forcing value should be reported. Also, the forcing values are break down to "anthropogenic forcing" and "BC forcing"; is BC not a part of anthropogenic aerosol?"

Answer: We think that offering clear-sky forcing values will enhance the paper and so we will include these results in the manuscript. However, we are not sure if our all-sky forcing calculation is inconsistent.

As the reviewer pointed out, AOD, SSA, etc. are obtained for clear sky only. Specifically, MODIS AODs are computed over cloud free pixels (Engel-Cox et al. 2004). In our study, we calculated forcing averaged over a grid that is $0.45^\circ \times 0.4^\circ$ in the horizontal. Also, our forcing is monthly average. Over a grid box and a month, there are almost always enough cloudy free pixels to give AOD while over the same grid box and the same month monthly cloud exists. Similarly, at each AERONET site, up to 50 attenuation and 10 sky-radiance measurements are taken during a day [Kinne et al., 2003]. Cloud-contaminated data are removed. Again over a month, there are enough



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cloudy free measurements to give AODs while over the same month monthly cloud exists. Thus, it is a reasonable acceptable assumption to use these AODs and cloud observation to calculate all-sky forcing. Clear sky forcing, to our knowledge, is a forcing estimate assuming that there is no cloud at all everywhere at all times. Either all-sky estimates or clear-sky estimates make a comparable assumption.

BC is a part of anthropogenic aerosols. We provided BC forcing in addition, due to the growing importance of BC contribution to the aerosol radiative forcing.

With regard to minor comments:

We agree to most of minor comments and will revise the manuscript accordingly. However, we would handle the following comments differently.

p. 827, line 10: Can you estimate the effects of not considering anthropogenic emission changes on your results? Can you apply a growth factor for different years?

Answer: The annual emissions were not available for the 2001-2004 period studied in this paper. We agree that one possible source of error in the emissions used in the simulations stems from excluding the annual changes in anthropogenic emissions during the study period. Asian emissions have recently been updated for the year 2006. When compared to the 2000 TRACE-P emissions values used in this study, the BC and SO2 emissions grew by $\sim$5% a year during this period. Since we constrained aerosol mass through AOD and SSA assimilations, any influence due to changes in emissions that are reflected in the observed AOD, should be reflected in our constrained aerosol distributions. We have studied and evaluated the impact of the interannual variability in meteorology and emissions on sulfur trends in Asia (Z. Lu et al., "Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000", (2010) just submitted to ACPD) and this work indicates that during this time period that our constrained results did capture the observed trends in sulfur, with the meteorological variability contributing significantly to the observed variability in observed SO2 and sulfate.



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p. 831, line 8-9: Putting all dust and sea salt into coarse mode AOD can introduce large errors since fine mode dust and sea salt could contribute to 20-40% of dust and sea salt AOD. I don't understand why this artificial separation is necessary – you have find and coarse mode dust and sea salt simulated separately by the STEM model, why don't you just use them accordingly?

Answer: We choose not to separate out sea salt and dust into fine and coarse modes in the assimilation step because of the large uncertainty associated with resolving the dust and sea salt into size bins based on effective radii. To avoid propagating this uncertainty further in the assimilation method, sea salt and dust were adjusted using coarse mode/total AOD. We have studied the sensitivity of our final results on this assumption and find that the results are essentially unchanged.

p. 832, line 25: Dust and open biomass burning are not evident in Figure 4.

Answer: We have reworded the text to clarify this. "The anthropogenic contribution from fossil and biofuel combustion dominates over most of Asia as seen from the high anthropogenic and BC AOD values at locations around heavily populated and industrialized areas including Indo-Gangetic plain and East China. The open biomass burning is the major contributor to the high values of anthropogenic AOD seen over Southeast Asia. The largest impacts of dust and its outflow can be seen clearly in the high PM10 concentrations over the Middle East and Western China.

p. 833, line 14: Why do you use AERONET 2005 data but simulate 2001-2004 period? What is the period of "AERONET new" data? After 2005?

Answer: We have only used AERONET data on a monthly scale for the 4-year (2001-2004) study period. We have updated the text to clarify this. "The AERONET AOD data for 2001-2004 period used in this assimilation were downloaded from the AERONET website in the year 2005. However, the most recent data for the same 2001-2004 period (accessed in October 2009 from the AERONET website) have updated and expanded values and this newer AERONET AOD data are referred to as "AERONET-



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new" in Fig. 5".

p. 834, line 11-12: Show your own comparison with AERONET and OMI. Koch et al. 2009 is not relevant here.

Answer: The OMI data was not available for the 2001-2004 period studied in this paper as OMI was launched only in 2004. We have compared to the available AERONOT AAOD and this discussion has been added to the paper. In general our predicted values at 550 nm are biased low by $\sim$25%.

p. 835, line 2-4: The comparison with Textor et al. 2006 is not appropriate, as Textor et al 2006 showed global annual average vertical values, including land and ocean, but this work showed the profile at a specific location in the Indian Ocean during March 2006. There is no information to assess if the BC vertical profile in Figure 6 is "an improvement" over Textor et al., 2006. In addition, it is an common sense that the concentration in the boundary layer is much higher than in the free troposphere over the source regions, but the BL concentration can be lower than that aloft over transported regions such as over the Indian Ocean. Answer: We will download and look at AERONET data to investigate this issue. We'll update the manuscript accordingly.

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 821, 2010.