Reply to Referee comments by Anonymous Referee #2

We thank Anonymous Referee #2 for comments that have made an improvement of the manuscript possible. In response to the comments we have edited the manuscript in several parts to add comparison of our simulation results with measurements and to clarify some of the limitations of the study with the tools used.

General comments:
In this manuscript, Henriksson et al. used a global climate-aerosol model (ECHAM5-HAM) to study the spatial distributions and seasonal variations of different aerosols, including black carbon, organic carbon, sulfate, sea salt, and dust, in two major aerosol emitting Asian countries, India and China. The results are presented as aerosol mass concentrations (three size classes, PM1, PM2.5 and PM10) and aerosol optical depth (AOD) with four scenarios of emission inventories: the REAS prediction inventory (2006), the REAS prediction inventory without anthropogenic emissions (2006), the REAS REF scenario (2020), and -2%/year scenario in Asia based on the REAS prediction inventory (2020). This work obtained some important spatial and seasonal characteristics of aerosols in India and China from the modeling perspective, and could have important contributions to the field observation and the modeling community. Generally, the idea of this manuscript is good, and the contents of this manuscript are within the scope of Atmospheric Physics and Chemistry. However, several key points should be addressed before the manuscript is accepted.

Specific comments:
Studying the spatial and seasonal variations of aerosols in four different emission inventory scenarios is the major scope of this manuscript. Therefore, comparison among different emission scenarios is important and necessary. The authors only presented a brief description of these scenarios, and it is not very clearly in my opinion. I would suggest adding a comparative table showing the emissions of different species by sector, by country, and by scenarios.

A table is added to describe the different emission scenarios. For a sectorwise separation, the REAS reference should be consulted.

This work applied a "REAS prediction inventory" for the year 2006 as the current emission scenario. However, the original REAS inventory was developed for years 1980-2003. The authors should describe the differences between the prediction inventory in this work and the original REAS 2003 and elaborate the modifications made to the original inventory. In addition, the REAS inventory was reported to be unrealistically high for some species after 2000, for example, SO2 in China (Aikawa et al., 2010; Smith et al., 2011; Zhang et al., 2009). I am wondering whether the authors have considered other newly developed inventories such as INTEX-B (Zhang et al., 2009), GAINS-Asia (Klimont et al., 2009) or EDGAR4.1.

We have in the new version of the manuscript a more quantitative comparison of MODIS and simulation results, and the higher average AOD in China in our simulation with 2006 REAS emissions than in the MODIS results could well be caused by higher Chinese SO2 emissions in the REAS inventory than in reality. TÄYTYY KATSOA, MIHIN NOI VÄITTEET REAS:in LIIKASUURUUDESTA perustuu!

The authors did not describe several important emission sources in this work, including *volcanic emission, open biomass burning from forest and grassland, and open burning of agricultural waste. Specifically, the open burning of agricultural waste should be included because it is anthropogenic activity and contributes to a large fraction of BC and OC emissions. To my knowledge, the REAS inventory does not comprise the open burning of agricultural waste, either. I would like to know how the authors dealt with this issue.*

Anthropogenic emissions from open burning of biomass are unfortunately not included in the REAS inventory. A part of open biomass burning are treated in the model, namely forest fires. We have added a discussion on open biomass burning in Section 3. According to Venkataraman et al. (2006), for India the BC and OC emissions stand for about 25% of the total emissions and only for a negligible 1% of SO2 emissions. These contributions are well within uncertainty in estimates of total emissions. Still, taking the open biomass burning and their seasonal variations, especially crop waste burning during harvest seasons, into consideration would make the study more consistent, but unfortunately this was not possible with the tools used as is further explained in the answer to the next comment.

The manuscript mentioned that "seasonal variations in anthropogenic emissions are not considered in the REAS inventory" and the results shown are "purely due to meteorological conditions influencing aerosol processes and variations in natural aerosol emissions". However, the anthropogenic SO2, BC and OC emissions from both India and China have large seasonal variations. For example, the results of INTEX-B (base year 2006) showed that the ratios of monthly anthropogenic emissions of SO2, BC and OC in China between maxima and minima are 1.4, 2.1 and 2.8, respectively (Zhang et al., 2009). Open burning of agricultural waste has even bigger ratios since it is correlated with harvest seasons. Therefore, to model the seasonal cycles of aerosols in India and China, seasonal variations in anthropogenic emissions should be considered. Additionally, the interannual variations of anthropogenic aerosol emissions should also be considered for the simulations during 2005-2009. As the two most rapidly developing countries in the world, the anthropogenic emissions of India and China changed dramatically after 2000 (Klimont et al., 2009; Zhang et al., 2009).

We agree that there are seasonal variations in the emissions that are not taken into account either in the REAS inventory. This is a fundamental limitation of our study. We would also like to point out that estimates such as those made from the results of INTEX-B are also uncertain, with SO2 emission estimates being more reliable as based to a large part on known activity data but with residential BC and OC emission seasonalities based on an assumed relationship between provincial monthly mean temperatures and stove usage. Additionally, according to this kind of relationship, seasonal variations from Indian households should vary less because monthly mean temperatures vary less within a year. However, it seems clear that if the seasonality in emissions would be taken into account and total emissions would stay the same, then SO2, BC and OC concentrations would increase in the winter and decrease in the summer. Seasonal variations of emissions and the error caused from not taking them into account in the simulations are now discussed in Section 3.

In Section 4, the authors compared the modeling results with many previous studies. However, the comparisons were relatively qualitative, and few quantitative comparisons, either figures or tables, were presented. Based on the authors' discussion, it seems that the results from this work are consistent with essentially all the previous studies. However, MODIS AOD data cited in this work are mostly from 2000 to 2005 and have strong interannual variations, whereas this work presented the simulation results between 2006 and 2009 and did not take into account of either the seasonal or the *interannual variations of anthropogenic emissions. Thus, the concluded consistency is not reliable without any quantitative discussion.*

We have added Figure 13 and related discussion for a comparison of the simulation data and MODIS data from 2006-2009, including interannual variations.

To support and strengthen the findings of this work, I would recommend systemically comparing the modeling results in this study with AOD datasets (e.g., MODIS, MISR, OMI, AERONET) that are available online for the studying period from 2006 to 2009. Additionally, the modeling results of this work could be compared with other modeling studies. For example, the simulation results of GOCART model for the period of 2000-2007 is available online at the GIOVANNI website and it contains global monthly gridded AOD by five aerosol species, exactly the same as this work. It would be very interesting to see how the two modeling results are compared and what the advantages and disadvantages of the present model when comparing to the GOCART model.

We have extended the comparison with MODIS results with Figure 13, as mentioned in the reply to the previous comment. We have also added Figures 14 and 15 and related discussion to compare the simulation results with measurements from AERONET. GOCART model results are briefly discussed in the revised manuscript.

Finally, I am wondering how the authors determined the natural emissions (e.g., emissions from sea salt, dust, DMS, volcano, open biomass burning of forest and grassland) and meteorological conditions for the two future scenarios.

The emission schemes are described in Stier et al. (2005) and references therein and are now also presented in more detail in Section 2 of our manuscript.

Technical corrections:
I strongly recommend the authors to proofread the manuscript carefully after revising. There are quite a few editorial mistakes in the manuscript and some are listed below:

Page 4019, line 8∼9: Delete "Ramanathan et al. (2008)".

Deleted.

Page 4020, line 22: Add comma after "aerosol optical depth (AOD)".

Added.

Page 4021, line 25: Add period after "dust and sea salt".

Added.

Page 4022, line 12: Add comma after "In Sect. 4 of this article".

Added.

Page 4023, line19: "desulfurisation" should be "desulfurization" to be consistent with the usage of "desulfurization" in other parts of the manuscript.

Changed to "desulfurization".

Page 4024, line 19: Add comma after "emission reductions".

Added.

Page 4027, line 15: Add "(2003)" after "Streets et al.".

Added.

Page 4027, line 24: Add comma after "Moorthy et al. (2004)".

Added.

Page 4027, line 26～28: Please reorganize this sentence.

Reorganised into two sentences.

Page 4029, line 4: Remove "Meng et al. in".

Removed.

Page 4030, line 22: "In their modeling study on" change to "In Adhikary et al. (2007)'s modeling study on".

Changed.

Page 4030, line 23: "2005 Adhikary et al. (2007) obtain" change to "2005, they obtain".

Changed.

Page 4030, line 26: Add "(2007)" after "Adhikary et al.".

Added.

Page 4032, line 28: "strenght", typo?

Was typo, changed to "strength".

Page 4033, line 28: "Fig. 12" should be "Fig. 13".

Changed. Now "Fig. 16".

Page 4034, line 3: "Fig. 12" should be "Fig. 13".

Changed. Now "Fig. 16".

Page 4034, line 11: Add comma after "India and China".

Added.

I also have several comments on the figures and figure captions as listed below:

I do not think Fig. 1 was mentioned anywhere in the main text. Latitude and longitude should be indicated on this map.

Now mentioned in first paragraph of Section 4. Latitudes and longitudes added to the map.

It would be better to have the same scaling for the subfigures in Fig. 2 ~ Fig. 5. The authors used words "four-year average" in the captions of Fig. 2~Fig. 5, and "averaged over four years" in the captions of Fig. 8 ~ Fig. 13. However, each of them is referring to different time periods and should be clearly indicated.

The different scalings are actually a carefully weighted choice giving a larger scale of colors for each figure, as the concentrations of different species and their contributions to AOD are so different. Figures showing total AOD for India and China in both the simulation and MODIS results have the same scales to facilitate comparison. Captions related to four-year averages have been made consistent and the time periods for averaging indicated.

References:

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Stier, P., J. Feichter, S. Kinne, S. Kloster, E. Vignati, J. Wilson, L. Ganzeveld, I. Tegen, M. Werner, Y. Balkanski, M. Schulz, O. Boucher, A. Minikin, and A. Petzold: The aerosol-climate model ECHAM5-HAM. Atmos. Chem. Phys. 5:1125-1156, 2005.

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