Atmos. Chem. Phys. Discuss., 6, S3617–S3623, 2006
www.atmos-chem-phys-discuss.net/6/S3617/2006/

Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Change in global aerosol composition since preindustrial times" *by* K. Tsigaridis et al.

K. Tsigaridis et al.

Received and published: 6 October 2006

We would like to thank the reviewer for his comments, since his recommendations contribute to clarify specific aspects of the manuscript. His/hers consideration have been taken into account for the revised version of the paper. We provide below the answers to the specific comments he/she made.

1. Abstract: I would avoid expressions like "This is the first study...". In fact there are numerous model simulations studying the impact on chemical composition of the atmosphere due to changing emissions between pre-industrial and present-day. For instance, within the EU funded network ACCENT a model comparison including 10 global models was organized (Gauss et al., 2006). In the ACCENT exercise simulations were performed using pre-industrial as well as present-day emissions. Four of the ten models are going even further than Tsigaridis et al. by including stratospheric

chemistry and six models by performing coupled climate-chemistry simulations. Numerous simulations of aerosol distributions have already been performed for present-day and pre-industrial emissions. For instance: 16 modeler groups performed such simulations within the Global Aerosol Model Intercomparison (AeroCom) prescribing year 2000 meteorology (Schulz, 2004, Kinne et al., 2005). Stier et al. (2006) presented the evolution of aerosol parameters in a changing climate from 1860 to 2100 taking into account interactions between aerosol, radiation and clouds.

Reply: As also requested by the first reviewer, we had removed this statement that was meant to reinforce the fact that our model treats all major aerosol components including SOA and nitrate and not that it is unique with regard to pre-industrial study. The AEROCOM exercise was already taken into account in the paper and comparisons are made (see for instance in Table 5). On the contrary, comparison with Gauss et al. (2006) is not considered as appropriate for the present paper since Gauss et al. (2006) focus on changes of the ozone distribution and the resulting radiative forcing since pre-industrial period. From the 10 models in that study, only one includes aerosols, and another has only sulfate. Moreover, Stier et al. (2006) take into account the aerosol impact on cloud microphysics and focus on the impact of aerosols on climate due to changes in aerosol properties and mixing state. However, they do not consider secondary organic aerosol in their simulations. Although no comparison with this work was made, it is now referenced in the text. Note that we also included a paragraph at the end of section 3.2 which compares the preindustrial and present-day burdens of various aerosol components with Liao and Seinfeld (2005) (pointed out to us by the first reviewer) that is the closest to our study already published work.

2. p 5590 ln 2-4: If I got the idea of the model, uptake of water is only calculated for inorganic aerosol components but not for sea-salt or POA and SOA although for wet removal of these components the conversion from hydrophobic to hydrophilic aerosol is calculated. If so, please, discuss the error introduced by this assumption. However, even if organics may not be very soluble, Kotchenreuther and Hobbs (1998) report that

at low relative humidity more water is often associated with the organic fraction than with the inorganic one.

Reply: It is true that the Aerosol Associated Water (AAW) is underestimated by the model over areas with low relative humidity and significant contribution of organics to the particulate phase. This will be improved in future simulations when appropriate parameterizations will become available. However, AAW does not affect any other model results since it is calculated only as a diagnostic variable. In addition, the effect of aerosol size increase by water uptake on the extinction coefficient and thus on the Aerosol Optical Depth (AOD) calculations is taken into account for all hydrophilic aerosol components, including carbonaceous ones based on the polynomial fit by Veefkind (1999) for sulfate aerosols as also described in Tsigaridis et al. (2005). This is now clarified in the model description section 2.1.

3. p 5590 ln 18-22: For the increase in SO2 emissions my calculations reach a factor of 3.4 (30 Tg pre-ind. / 103 Tg present-day) rather than a factor of 30. Non-linear relationship between source strength and atmospheric burden is an important issue and warrants maybe a separate table including all relevant species, gaseous and particulate, and a discussion about the causes of this non-linearity.

Reply: The increase of anthropogenic SO2 emissions is indeed a factor of 30, since, as shown in Table 1, these SO2 emissions increase from 2.4 Tg/y to 73 Tg/y. However, the referee is right that we better compare with the sulfur emissions both as SO2 and as dimethylsulfide since both will form sulfate aerosols. Therefore we modified this part of the discussion in order to make clear that we now refer to the reactive sulfur taken into account in the model to be emitted into the atmosphere. We also added a comment in section 2.2 to reinforce the importance of non-linearities in the earth system.

4. p5593 ln 6-7: Table 2 includes the size range for each mode of sea-salt and dust. Does that mean that the mean particle diameter varies within this range and that the model includes some aerosol physics? Does the sink processes of sea-salt and dust

depend on particle size?

Reply: As the reviewer points out, some elemental particle physics is taken into account in the model for the dry deposition parameterization, since sedimentation dominates for larger particle sizes, but is not so important for smaller ones (Table 4). Wet removal though is assumed to be independent of the size but depends on its hydrophobic or hydrophilic character of the particles. Two sentences have been added at the end of section 2.4.

5. Chapter 3.1 Oxidant fields: Please, compare not only pre-industrial oxidant concentrations to observations but also present-day values. How do the changes calculated in this study compare to other model estimates? We expect that changes in temperature, humidity, and UV radiation intensity due to climate warming climate change could affect ozone significantly. What would be the effect of neglecting the observed increase in atmospheric temperature and humidity on oxidant concentrations and nucleation and condensation of semi-volatile species? (see e.g. Gauss et al., 2006). Please, discuss. What is the reason that the discussion of changes in the aerosol constituents is in this chapter rather than in the chapter 3.2? Chapter 3.2 and 3.4 Aerosol burden and composition: Please, compare the changes between pre-industrial and present-day conditions to other estimates (e.g. AeroCom).

Reply: This paper's aim is to study the aerosol composition in the atmosphere. Although gas-phase chemistry is of major importance in this task, a validation of the gas-phase mechanism in the model is by far beyond the scope of the present manuscript, at least for the present-day atmosphere, since several papers using the same CTM and gas phase chemistry (like for instance Houweling et al., 1998 and Lelieveld and Dentener, 2000) have been already published and provide a comprehensive evaluation of the model performances. More references are now provided in the model description. For the preindustrial case, the oxidant fields are evaluated within the limitations introduced by the scarce ice-core measurements available and the uncertainties in the early years O3 records.

Further, we do agree with the reviewer that changing the meteorological parameters will affect ozone, both in the troposphere and in the stratosphere. Such impacts have been studied and presented in earlier publication by Tsigaridis et al. (2005) for the present day. However, because our model is an off line model with regard to meteorology and climate, the impact of changing temperature, atmospheric water cycle and UV radiation since preindustrial times can not be computed. Such calculation requires a General Circulation Model coupled with chemistry.

Only sensitivity simulations can be performed with a CTM. Indeed, we have performed such calculations by changing the air temperature (2oC increase in the boundary layer, 1 oC in the free troposphere and no change in the lower stratosphere) and found almost no remarkable changes in O3 concentrations. However, we decided not to include such a preliminary sensitivity study in the paper.

The discussion of aerosol constituents from the end of section 3.1 has now moved to section 3.2 where it belongs, indeed. That section (3.2) also presents the comparison of modeled aerosol burdens between present-day and preindustrial conditions (our model and other models, including AEROCOM, detailed table 5; comparison and table already in the earlier version of the paper).

For the section 3.4 such a comparison cannot be made, since this is the first study that focuses on the SOA chemical composition itself.

6. p 5594 ln 23: The aging time in respect to oxidation of one day until BC becomes hydrophilic seems quite low. For instance Croft et al., (2005) report an aging time of less than one day when including the physical aging of condensation and coagulation, and the chemical aging of oxidation, and they found the contribution of oxidation to the BC ageing process to be small.

Reply: This point has been addressed in our reply to the point 7 of the reviewer #1.

7. p 5595 ln 14-27: A comparison between observed and calculated aerosol mass of

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

species which size spectrum is resolved as for sea-salt and dust, is only meaningful when the lower cut-off of the measurements and the model data is about the same. Maybe this explains some of the differences between observations and simulations.

Reply: We agree with the reviewer on the importance for the model evaluation of the cut-off size of the experimental and the model data. However, in general this will reduce all coarse aerosols in the model, while the overestimation exists only at low aerosol sea-salt and dust loads. Furthermore, to correct for the appropriate cut-off, information for each set of measurements is needed, which unfortunately is not available for most datasets.

8. p 5596 ln 22-25: The extinction coefficients are corrected for the uptake of water due to ambient humidity assuming the same properties for inorganic and organic components, based on a polynomial fit by Veefkind. Why the growth rate is not used for inorganic aerosols as calculated with EQSAM? The same growth rate is used for organics and inorganics. Please give an estimate of the error introduced by this assumption.

Reply: EQSAM calculates the AAW for inorganic aerosols only; we thus used the approach by Veefkind (1999) that corresponds to a more “real” aerosol mixture. This growth is used for both inorganic and other hydrophilic aerosols as an upper limit, which will lead to an overestimation of AOD. If we assume that aerosol size of non-inorganic aerosols does not increase with relative humidity, as a lower limit, we calculate about 20% less AOD as a global average. This is now mentioned in the section 3.5.

9. p 5604 ln 16-17: When condensation of SOA on primary particles is calculated, what are the assumptions for this parameterization (particle surface, etc.)?

Reply: As requested also by the reviewer #1 and Henze and Griffin’s interactive comment, we significantly extended the model description to address all the requested questions.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5585, 2006.