

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

Anonymous Referee #2

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The manuscript discusses two model simulations calculating global aerosol distributions, one assuming pre-industrial and one present-day emissions. In both simulations meteorology and natural emissions are kept constant assuming present-day conditions. The study takes into account effects of changes in oxidant concentrations and in aerosol and aerosol precursor emissions. The aerosol model predicts mass mixing ratios of the chemical components assuming externally mixed aerosol. The model is superior to most other global models in considering secondary organic aerosol formation and the ammonium-nitrate-sulfate-water complex, in addition to the components dust, sea-salt and primary carbonaceous aerosol. The discussion focuses mainly on changes in the SOA distribution. According to this study the increase in SOA burden from pre-industrial to present-day is due to enhanced oxidant concentrations and

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higher primary particle concentration from anthropogenic sources. The increase in inorganic aerosol burden is smaller than expected from the changes in emissions indicating a non-linear response. Although these results are interesting and warrant publication in ACP, the aerosol model and the uptake of water on particles need better explanation. Even the relationship between changes in emission and resulting burden and AOD should be discussed more systematically.

Detailed Comments

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.

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.

3. p 5590 In 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.

4. p5593 In 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?

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).

6. p 5594 In 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.

7. p 5595 In 14-27: A comparison between observed and calculated aerosol mass of species which size spectrum is resolved as for sea-salt and dust, is only meaningful

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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.

8. p 5596 In 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 Veeffkind. 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.

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

Croft, B. et al. (2005): Black carbon ageing in the Canadian Centre for Climate modelling and analysis atmospheric general circulation model, *Atmos. Chem. Phys.*; 7 (5) ; 1931-1949

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Kotchenruther, R.A., and P.V. Hobbs, 1998: Humidification factors of aerosols from biomass burning in Brazil. *J. Geophys. Res.*, 103, 32081-32089.

Schulz, M., 2004: AeroCom Aerosol model intercomparison home page. <http://nansen.ipsl.jussieu.fr/AEROCOM/>.

Stier et al., (2006): The evolution of the global aerosol system in a transient climate simulation from 1860 to 2100, *Atmos. Chem. Phys.*, 6, 3059-3076

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