**Atmospheric
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Discussions**

Interactive
Comment

Interactive comment on "The evolution of the global aerosol system in a transient climate simulation from 1860 to 2100" *by* P. Stier et al.

P. Stier et al.

Received and published: 28 May 2006

Response to comments of reviewer 2

1a. The emissions used here are poorly documented, in the case of the historical emissions and in the case of the future carbonaceous aerosol emissions, and poorly regarded in the case of the future SRES A1B. As a result, this study has primarily illustrative value of some possible impacts of aerosol microphysics on aerosol load and forcing. Throughout the text the study should be treated as such.

and

Show in either a table or figure, the fossil fuel, biofuel and biomass burning emissions for each component as a function of time.

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We acknowledge that the description of the emissions in the original manuscript should have been more detailed and have extended the paragraph as given below. The spatio-temporal evolution of the emissions, according to component and sources, however, was already documented (as suggested) in Fig.1 and Fig. 2.

We concretised the description of the emission inventory as follows.

The paragraph about the prescribed emissions that are held at their present day values throughout the simulation period has been moved from the HAM description to the description of the simulation setup (section 2.6) and extended to (p 4 c 2 l 12):

"We further periodically apply monthly mean emissions of biogenic terrestrial DMS and POM from secondary biogenic sources as well as SO_2 emissions from con-tinuously degassing volcanoes based on the year 2000 AeroCom aerosol model inter-comparison experiment (http://nansen.ipsl.jussieu.fr/AeroCom/) emission inven-tory (Dentener et al., 2006, available from ftp://ftp.ei.jrc.it/pub/Aerocom/). POM from secondary biogenic sources is therein estimated assuming an aerosol yield of 0.15 from the biogenic monoterpene emissions of Guenther et al. (1995) and applied in HAM as primary aerosol source. The emission size distributions of BC and POM follow the AeroCom recommendations (see Stier et al., 2005) as well as the assumption that 2.5 % of all SO_2 emissions are emitted in form of primary sulfate."

We further extended the description of the transient emissions to (p 4 c 2 l 27):

"Transient emission fluxes from 1860 to 2100 of SO_2 and BC from fossil fuel com-bustion, domestic fuel-wood consumption, agricultural waste burning, and forest fires are prescribed from a compilation by the Japanese National Institute for Environmental Studies (NIES, T. Nozawa et al., *pers. comm.*, 2004):

Historic SO_2 emissions are based on Lefohn et al. (1999) complemented by shipping emissions from the HYDE database (http://www.mnp.nl/hyde). BC biomass burning emissions are from the GEIA database (http://www.geiacenter.org) for the year 1987

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employing the methodology of Cooke et al. (1996). Pre-industrial biomass burning emissions are assumed to be 10% of the present day emissions (Andreae, 1991) and are scaled to present day conditions proportionaly to the population increase from the HYDE database. Present day emissions of BC from fossil-fuel (Cooke et al., 1999) as well as from biofuel and agricultural activities (Takemura et al., 2000) are scaled to pre-industrial values employing World Bank gross domestic product data for each country.

Future emissions from 2000 to 2100 are based on the SRES A1B scenario. Anthropogenic SO2 emissions are used as provided by SRES (Nakicenovic et al., 2000, available from http://sres.ciesin.org). For the carbonaceous aerosols, the present day fossil fuel emissions are scaled according to the individual source trends in the SRES data. BC biomass burning and biofuel emissions are extrapolated from the present day emissions based on the SRES A1B population scenario; BC emissions from agricultural activities are extrapolated proportionally to the cropland development in the SRES land use data.

For the total integration period we derived emissions of SO2 from vegetation fires and of POM from the BC emissions by assuming source specific emission ratios: SO2/BC = 1.28, POM/BC of 1.4 (fossil fuel), 5.6 (domestic and agricultural), and 11 (vegetation fires) (F. Dentener, *pers. comm.*).

It has to be pointed out that even for present day emission inventories, based on largely well determined fuel use data, significant uncertainties exist. These uncertainties are particularly large for the carbonaceous compounds so that present day inventories differ by as much as a factor of two for fossil fuel use (e.g. Schaap et al., 2004) and are even more uncertain for biomass burning emissions. These uncertainties propagate into past and future emission scenarios and further add to their uncertainties regarding population, technological, and legislative developments. For example, according to the used NIES SRES A1B emission inventory and the underlying original SRES A1B estimate (Nakicenovic et al., 2000) carbonaceous aerosol emissions are

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projected to increase significantly during the 21st century. However, the emission inventory by Streets et al. (2004), based on the same SRES A1B storyline, projects a decrease of the BC (POM) aerosol emissions from 1996 to 2030 of 11% (16%) and from 1996 to 2050 of 24% (18%) (see discussion in Streets et al., 2004)."

1b. Transient emission fluxes from 1860 to 2100 for the SRES A1B scenario ? Isn t the A1B scenario for future, not the past century? How are the past emissions derived? Provide more detail!

Apologies for the imprecise phrasing. See changes in the description of the emissions as described above. We further phrased the following sentence in the introduction more precisely (p 2 c 1 l 51):

"Future greenhouse gas concentrations as well as anthropogenic aerosol and aerosol-precursor emissions are prescribed based on the SRES A1B scenario."

1c. Compare carbonaceous aerosol emissions with those of Streets et al (2004) SRES A1b estimates. Compare historic carbonaceous emissions with Ito and Penner (2005). Compare past SO2 emissions with van Aardenne et al. (2001).

We have substantially extended the description and discussion of the emissions as described above and pointed out the uncertainties and differences in particular for future emission projections. A full inter-comparison of the different emission inventories would require a level of source specific detail that goes beyond the scope of this paper.

1d. I find the future increase (from present to 2050) in biomass burning especially difficult to understand, and this is crucial to the predicted aerosol evolution. Streets et al (2004) projected decreases in biomass burning during this period from their interpretation of the A1B scenario. Since open burning emissions don t depend on technology emission factor effects, there is clearly some important difference in methodologies that needs to be explained.

It has to be kept in mind that particularly biomass burning emission projections suffer

from the unpredictability of legislative developments as they do not only depend on slow economic and technological changes but often on political decisions such as land reforms or environmental awareness. The biomass burning emissions in the employed NIES emission inventory are scaled with the SRES A1B population scenario for wild-fires and biofuels and with the SRES land use date for agricultural emissions. Streets et al. (2004) combine projections of the IMAGE model for managed forests supplemented with estimates of wildfire emissions from mature forest projections of IPCC, assuming that changes in mature forests are proportional to wildfire emissions. Again, we believe that the extended description of the emissions as describe above provides the necessary detail for the reader. Additionally, we refer therein to the relevant discussion in Streets et al. (2004).

1e. Section 3.1 on emissions should perhaps be moved to the model setup, since it is not a result .

A large part of this section deals with the interactively calculated emissions of DMS, sea salt, and mineral dust. As it seems desirable to discuss the evolution of all emissions in one section and the results from the interactively calculated emission do not belong into the model description we believe that this position appears to be the best compromise.

The fixed dust source area might be justified based on Tegen et al (2004) which suggests a small, 10% anthropogenic component.

This is an interesting point, although the 10% anthropogenic contribution in Tegen et al. (2004) refers solely to the contribution of agricultural soils to the mineral dust emissions under present day climatological conditions. Tegen et al. (2004) perform a simulation with the same dust source and climate data from an A2 scenario simulation with the predecessor version of the AOGCM used in our simulation (ECHAM4-OPYC, Roeckner et al., 1999). Considering both changes in the climatic conditions and in vegetation cover they simulate a 24% increase of mineral dust emissions from 1970-1980 to 2040-2050 compared to an only 9% increase with fixed surface properties. Therefore, we

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think it is appropriate to warn the reader about the unaccounted effect of variations in vegetation cover.

Mention that historic eruptive volcanoes are not included among the interactive aerosols, according to Figure 1. Presumably they are treated as external climate forcing factors, but this is confusing.

Although this was already mentioned in the simulation setup and the discussion of the relevant Fig. 11, we have extended the description in the simulation setup to (p 4 c 1 l 52):

"From pre-industrial to present day conditions, optical depths of stratospheric aerosols from volcanic eruptions are prescribed annually in four latitude bands based on an updated dataset (http://www.giss.nasa.gov/data/strataer/) of Sato et al. (1993). Variations in solar irradiance are specified according to Solanki and Krivova (2003)."

Also mention here in the emissions section that biogenic POM source is assumed constant.

We have moved the respective section from the description of HAM to the simulation setup and have clarified this point as described above.

1f. How large is seasonal variability for biofuel? I am not aware of published estimates of biofuel emission seasonal variability, more discussion is needed.

For the year 2000, the standard deviation of the global monthly-mean emission flux normalised by its annual mean is 0.11. We included the Takemura et al. (2000) reference to the description of the biofuel emissions and refer the interested reader to this original description of the emission estimate.

2. Impacts of aerosol microphysics on lifetime and burden.

2a. Aerosol lifetime and burden are some complex mixture of aerosol microphysics and geographical shifts in aerosol emission. The importance of changes

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in the microillustrative micro- physical aging time is not apparent since in many cases the aging time changes in an opposite sense to the change in aerosol lifetime (e.g. while the aging time decreases, the lifetime increases). For species in which the aging time is much smaller than the lifetime (e.g. BC and POM), I suspect that the change in aging time is not so important. In other words, if the particle is not likely to encounter a removal process (rain) within 5-6 days then it doesn t matter whether it becomes soluble in 1 day or 2 days. The situation might be more interesting in some regions. Also the dust story may be more complex. But I remain unconvinced that microphysics has a large impact on aerosol lifetime. The only way I can think of to prove an effect would be to repeat the experiment without microphysics. Discussion should be added regarding timescale impacts.

We agree that it is not possible to isolate these specific effects from a fully coupled transient climate simulation as presented here. These issues can be better scrutinised in idealised sensitivity studies under well constrained emission changes. Therefore, we quantified the importance of microphysical aging on the aerosol lifetime in the manuscript "Emission Induced Non-linearities in the Global Aerosol System - Results From the ECHAM5-HAM Aerosol-Climate Model" (Stier et al., J. Clim., in print) that we reference in this manuscript. We further investigated the importance of the variations in the mixing state on the aerosol optical properties in a submitted manuscript (Stier, P., J. H. Seinfeld, S. Kinne, J. Feichter, and O. Boucher, The Impact of Non-Absorbing Anthropogenic Aerosols on Atmospheric Absorption, submitted). What we demonstrate with the presented simulation in this manuscript, however, is that the variations in the microphysical aging time in the sensitivity studies, under highly idealised emission changes, are indeed of practical relevance as they also occur under a realistic transient emission scenario.

2b. A shift in emissions to lower latitudes (e.g. BC in 20th century) does not always mean shift to drier emission regions. For example, Asian emissions are

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generally in relatively moist regions.

That is correct. Therefore, we generally do not draw conclusions solely on the emission regions but also mention for example the increasing importance of dry-season vegetation fire emissions. We concretised one ambiguous usage of this phrase to (p 6 c 2 l 7):

"For BC, τ decreases from 6.5 days in 1860 to around 5 days in 1960 and increases thereafter to to 8 days in 2100. The initial decrease in the residence-time is contradictory to the increasing importance of low latitude dry-season vegetation fire emissions (c.f. Fig. 2,3)."

2c. In some models the indirect effect impacts aerosol lifetime. Does this model? Are both 1st and 2nd indirect effects included?

This is an interesting question. Yes, as described in the model description in section 2.3, both the 1st and the 2nd indirect aerosol effect are included in this simulation. However, owing to the setup of this fully coupled simulation, clouds and their interactions with aerosols are not only affected by indirect aerosol effects but in general by changing climatic conditions under the influence of all anthropogenic and natural perturbations. Consequently, we can not isolate the impact of the indirect aerosol effects on the aerosol lifetime from this simulations. We are currently assessing the simulated indirect aerosol effects and their feedbacks with the aerosol system in well constrained sensitivity studies.

3. Impact of aerosol lifetime on radiative forcing.

3a. It would be interesting to see this model s prediction of the degree of non-linearity between aerosol emissions and load or optical depth or forcing. I suggest adding figures showing the ratio between the load (or tau or forcing) and the emission as a function of time in order to quantify the degree of non-linearity.

The ratio between the aerosol load and emissions is actually the definition of the at-

mospheric residence time. The temporal evolution of the residence time for all aerosol components was already included as Fig. 5 and discussed in Section 3.3 in the original manuscript.

We agree that an analysis of the linearity between the aerosol radiative perturbations and the emissions would be desirable. However, as aerosols occur generally as internal mixture, the only stringent way to extract the radiative perturbations of a specific aerosol component is as difference between two simulations in one of which the respective component is excluded. Unfortunately, the tremendous computational burden associated with this simulation does not allow this analysis. Thus, we are addressing this issue in accompanying sensitivity studies (e.g. Stier et al., J. Clim., in print).

3b. I am confused about the assumed optical and radiative properties. How are the internally mixed aerosol properties determined? How much is the increase in absorption due to increased BC emissions? Again, this could be demonstrated via the ratio between tau_absorption and emission as a function of time.

The description of the calculation of the radiative properties has been extended as described in the response to question 2 of reviewer 1.

At first view it seems that the BC absorption efficiency could be estimated from the ratio of the absorption optical depth and the BC emissions. However, as BC is not the only absorbing aerosol in the internally mixed aerosol modes, this measure turns out to be unsuitable: at the begin of the integration period most absorption is caused by mineral dust, resulting in a high ratios of AAOD to BC emissions. With increasing BC emissions throughout the integration period, this ratio decreases.

4a. Abstract. In accordance with comments above, I would place less emphasis on specific changes in optical thickness and radiative forcing. Provide more detailed information on the (percent) changes in lifetime, aging rate, mixing state. How non-linear is the relation between emissions and forcing?

As explained above, it is not straightforward to derive this (interesting) relationship from this coupled model setup and we have to refer the interested reader again to our sensitivity studies to appear in other publications.

4b. Section 2.6, How much do results for the 2 20th century realizations differ in their aerosol predictions?

Under similar climatic conditions and identical emissions, aerosols have a relatively similar atmospheric residence times. Thus, the differences between the two realisations in the global annual mean typically do not significantly exceed the inter-annual variability depicted in Fig. 3.

4c. Figure 1: In the BC figure, the dashing in the curves do not correspond to that shown in the key. Also, the yellow under SO2 is hard to see, I suggest another color, like the darker yellow-orange used in later figures.

We carefully checked all dashings in Figure 1 again and can confirm that they in fact do correspond to that shown in the key. We can only suspect that the reduction in size for the ACPD format caused some distortion for your printers resolution.

We replaced the yellow for SO2 by orange.

4d. Figure 2. The 2 reds at the top of the color scale look the same. Maybe make the top one lighter or purpler?

Again, this seems to be a printer specific problem. The reds appear very clear on our screens and printouts.

4e. The large predicted natural contribution to fine aerosol tau is interesting and pertinent. This could be pursued: how does the geographical distribution of this natural component compare with the satellite-based "anthropogenic" tau distributions?

In fact this is an interesting question that we will investigate in more detail in future

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work. However, for this comparison with satellite data a nudged simulation in which the model is forced to the observed meteorology seems more promising as it avoids sampling biases. Additionally, it has to be kept in mind that current satellite retrievals of the fine mode optical depth over land have significant uncertainties that question their usability (Anderson et al., J. Geophys. Res., 2005).

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 12775, 2005.

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