**Atmospheric
Chemistry
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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.

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Response to comments of reviewer 1

1. Precipitation is critical for the aerosol distribution and aging processes. It is not clear in the paper how the precipitation in a transient climate would affect the aerosol.

and

2. The description of aerosol optical properties (such as single scattering albedo) is not clear in the paper. Are they a function of aerosol mixing state?

Thanks for pointing that out. We extended the model description by the following paragraph (p 3 c 1 l 37):

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"Aerosol radiative properties as well as the sink processes dry deposition, sedimentation, and wet deposition are parameterised in dependence on the prognostic aerosol size distribution, composition, and mixing state and coupled to the ECHAM5 meteorology. The aerosol radiative properties are calculated in the framework of Mie theory. For each aerosol mode, effective refractive indices are calculated by volume averaging the refractive indices of all components, including aerosol water that is parameterised in dependence of ambient relative humidity. The effective complex refractive indices and the Mie size-parameters for each mode serve as input to look-up tables for the aerosol radiative properties, providing extinction cross-section, single-scattering albedo, and asymmetry parameter to the ECHAM5 radiation scheme. The aerosol wet deposition is parameterised in terms of the aerosol size distribution and mixing state via mode specific scavenging ratios, specifying in-cloud and interstitial aerosol fractions in the cloudy part of a grid box and in convective updrafts. The actual wet deposition is calculated from the resulting in-cloud aerosol content based on the precipitation formation and re-evaporation calculated by the ECHAM5 cloud scheme."

Thus, changes in the simulated precipitation do affect the global and regional aerosol distribution. The global annual mean precipitation increases throughout the simulation period by about 3%. Although the regional distribution of the precipitation changes is inhomogeneous, this change in precipitation is unlikely to explain the similated variations in the aerosol residence time. We added the following sentence for clarification (p 6 c 1 l 47):

"Changes in precipitation affect the aerosol residence time via the interactively calculated wet deposition rate. However, the general increase of the residence time from about 1950 onwards cannot be explained by the change in precipitation which actually increases by about 3% from 1860 to 2100 *(not shown)*."

I doubt the significance of aerosol forcing computed in the paper, because aerosol forcing also depends on surface reflectance.

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It is right that aerosol forcing depends on the surface reflectance. The spatial variations in surface reflectance are, of course, included in our analysis. However, following the forcing definition of the Intergovernmental Panel on Climate Change (Ramaswamy et al., 2001), forcing is calculated as the change in the net irradiance upon the introduction of an agent (here aerosols) with all other model components held fixed. Based on this concept, changes of the surface reflectance are considered as independent forcing. To remain consistent with the IPCC forcing definition, we minimised temporal variations by masking out areas with a change in surface reflectance larger than 0.03.

As the online coupling of the simulated aerosol cycles with the other components of the earth system model violates the strict definition of forcing as external perturbation, we instead use the term "aerosol radiative perturbation".

How do you consider the land use in the paper?

In traditional "IPCC style" coupled AOGCM transient climate simulations land use changes have generally not been considered. To our knowledge no published transient coupled atmosphere ocean climate simulation with interactive multi-component aerosols has considered the effect of land use changes. Although an interactive vegetation module has now become available in the MPI Earth System Model, the land properties in the presented simulations are based on their present day values as described in Roeckner et al. (2003). We will address the combined effect of land use and emission changes in future research.

For clarification, we added the following sentence to the description of ECHAM5 (p 3 c 1 l 5):

"In this setup, surface properties and vegetation cover, thus the surface albedo of snow free land surfaces are fixed at their present day values. The surface albedo is therefore a function of the snow and ice cover and of their albedo, parameterised in terms of the surface temperature."

I would recommend authors not mention the aerosol forcing numbers in the abstract, because the uncertainties in these numbers are too huge to be quantified.

It is true that aerosol forcing numbers, both from modelling and satellite retrievals, are generally associated with significant uncertainties. Nonetheless, the quantification of the model calculated radiative perturbation provides a measure of the strength of the direct aerosol effects and allows the comparison with other studies.

3. page 12786, near line 10. Why dust emission showed a significant increase while sea salt not? Are they both parameterized according to wind speed? If yes, is this difference because the wind speed decreases over the ocean while increases over the land in your simulation?

This is an interesting point, in particular because both mineral dust and sea salt emissions are parameterised according to wind speed.

To improve the description of the coupling of the emissions with the prognostic model parameters we extended the following paragraph in the model description to (p 3 c 2 l 4):

"Emissions of mineral dust are calculated online in dependence of the ECHAM5 10m wind speed, soil moisture, and snow cover (Tegen et al., 2002,2004). Preferential source areas and the vegetation cover are held at their present day values from Tegen et al. (2002). Freshly emitted dust is assumed insoluble. Sea salt emissions are parameterised in terms of the simulated 10m wind speed and sea-ice cover following Schulz et al. (2004). Emissions of DMS are calculated interactively from the simulated DMS seawater concentrations of the HAMOCC5 ocean biogeochemistry, applying the sea-air exchange formulation of Wannikhof (1992) that depends on the simulated 10m wind speed and sea surface temperature."

For sea salt, the ocean surfaces provide a large source area with identical emission characteristics (variations in the salinity are smaller than the uncertainty in the emis-

sion parameterisation and neglected). Contrary, most mineral dust is emitted from small scale preferential source areas (Tegen et al., 2002). Therefore, the mineral dust emissions are particularly sensitive to changes in the regional meteorology. For clarification we moved part of the explanation of the burden changes (Section 3.2) to the emission section (Section 3.1) and extended the respective paragraph (p 5 c 2 l 11):

"Mineral dust emissions, however, show a distinct inter-annual variability ($\sigma^{norm} = 0.08$) and an increase of about 10 % towards the end of the integration period. As most of the mineral dust emissions are confined to small scale preferential source areas (Tegen et al., 2002), they are particularly sensitive to changes in the regional meteorology. A more detailed analysis *(not shown)* reveals that the dominant changes in the dust emissions occur in the northern African source regions with a decrease in the central-north African source regions, including the Bodele Depression, and an increase in the Saharan north-western African source regions. The decrease in dust emissions in central-north Africa can be attributed to an increase in soil moisture and reduced surface windspeeds. Contrary, the increased emission in the north-western Saharan source regions are a consequence of increased surface wind speeds. These changes of the regional climatological conditions can partly be attributed to an alteration of the monsoon regimes owing to an increase in atmospheric absorption due to increased carbonaceous emissions from vegetation fires (see R2006). It has to be pointed out that the dust emissions are calculated assuming fixed preferential source areas and year 2000 vegetation cover. Therefore, the simulated century scale variability is likely to be a lower estimate."

4. page 12792, line 25, for technical reasons, areas with a change in surface albedo larger than 0.03 is masked out ? What are the technical reasons?

The calculation of aerosol forcing in GCMs is typically done by calling the radiation scheme twice, with and without aerosols, and taking the difference in the net irradiance. Unfortunately, GCM radiation schemes are computationally expensive. As this coupled Earth System Model simulation pushed the computational expense to the affordable

limit, we refrained from a double call of the radiation scheme and calculated the direct aerosol radiative perturbation from the diagnosed radiative fluxes. We extended the description of this calculation in the manuscript to (p 8 c 2 l 4):

"DARP is defined here as the deviation of the clear-sky net short wave radiation at the top of the atmosphere from the 1860-1870 mean. DARP is calculated as the change in the clear-sky net top-of-the-atmosphere solar radiation, corrected for variations in the top-of-the-atmosphere solar irradiance and for the associated change in the upward surface radiation. The correction for the change in the upward surface radiation from the stored model variables requires the assumption of constant surface albedos. Therefore, areas with a change in surface albedo larger than 0.03 are masked out. The masked areas constitute generally less than 14% of the earth's surface. "

If your forcing calculation doesn't include these areas, you should mention that your forcing value is only a near-global averaged value.

We added this to the description above. It seems worth mentioning that this method still provides a much better global sampling than satellite retrievals of aerosol radiative effects.

This again goes to the key point: how is the land use and surface processes modeled in your GCM?

See above.

5. If the aerosol tends to become more internally mixed in long term, then the overall aerosol single scattering albedo could increase also, because the internal mixing of soot (such as core/shell structure) has a larger single scattering albedo than external mixing (Ackerman and Toon, 1981, Applied Optics). Yet, your results are contradictory to this.

No, the internal mixing of soot with other aerosol components, such as sulfate, decreases the single scattering albedo of the aerosol population for all typical volume

fractions. This is also found by Ackerman & Toon, 1981 (page 3663): "It is obvious from Fig. 1 that for a given volume fraction of soot, the internal mixing produces lower values of the single-scattering albedo than does the external mixture of soot and sulfate particles with the same particle size distributions. For small soot particles in the external mixture the same is true for volume fractions of $< 30\%$." This has also be confirmed by other studies (Chylek et al, 1995; Chylek et al., 1996; Jacobson, 2001) and is in line with our findings.

We recently demonstrated the effect of internal mixing on the aerosol radiative properties with a subset of this modelling system, the ECHAM5-HAM aerosol climate model (Stier, P., J. H. Seinfeld, S. Kinne, J. Feichter, and O. Boucher, The Impact of Non-Absorbing Anthropogenic Aerosols on Atmospheric Absorption, submitted). We find a significant enhancement of absorption by internal mixing of black carbon with anthropogenic sulfate close to the source regions. Further away from the sources, enhanced microphysical aging of black carbon and subsequent enhanced wet removal cause generally a reduction in the atmospheric absorption. In the global annual mean these effects balance each other, resulting in a small (0.2 Wm^{-2}) reduction of atmospheric absorption by anthropogenic sulfate.

However, the setup of the fully coupled simulation in this manuscript does not allow to isolate these effects. Therefore, we refrained from addressing the topic in this manuscript and would like to refer the interested reader to the manuscript mentioned above.

You should have more explanation on how the aerosol forcing and aerosol properties are computed?

See above.

6. While this study primarily is a modeling study, I think there is still some validation that should be done easily. Otherwise, it is difficult to convince readers. I would like to see authors to present some comparisons between their AOD

values with those from satellite (such as AVHRR or MODIS) or ground-based observations (such as AERONET). The comparison can be done either in monthly or yearly basis for 10 or 5 years, either in regional or global averages. Without a good simulation of current aerosol state, the number and the significance of this paper is very doubtful.

We added a comparison of regional averages of the simulated aerosol optical depth around the year 2000 with AERONET sun-photometer measurements, and satellite retrievals from the MODIS, MISR, AVHRR, and TOMS instruments to the manuscript and extended the respective paragraph to section 3.5 (p 8 c 1 l 1):

"In Fig. 10 the simulated 1998-2002 mean aerosol optical depth is compared on a regional basis to five different measurement datasets: to a retrieval from the AERONET sun-photometer network (Holben et al., 2001) extrapolated and gridded on a 1° x 1° resolution (1998-2004 mean of available measurements, S. Kinne, personal communication, 2006), as well as to satellite retrievals from MODIS (2000-2004 mean, Tanre et al., 1997; Kaufman et al., 1997), MISR (2000-2004 mean, Martonchik et al., 2002; Martonchik et al., 2004), AVHRR (2001 mean, Ignatov and Stowe, 2002a,b), and TOMS (1996-2000 mean, Torres et al., 2002). For comparison, results from the Stier et al. (2005) year 2000 reference simulation with ECHAM5–HAM are provided, utilising the AeroCom emission inventory. At large, the regional distribution of the observed AOD is well captured in the MPI–ESM simulation. The global 1998-2002 mean AOD of 0.22 is somewhat larger than the global 2000-2004 mean AODs from MODIS (0.20) and MISR (0.21), that provide probably the highest quality datasets with global coverage. However, this good agreement on a global mean basis should be regarded with the caveat that the satellite retrievals exhibit non-negligible uncertainties and both the MODIS and MISR retrievals are likely to be biased high (Levy et al., 2005; Liu et al., 2004). On a regional basis, the overestimation of AODs is particularly pronounced in the Saharan dust outflow region but also in the main anthropogenic source regions, China, Europe, and the eastern United States. It is not possible to quantitatively attribute this overes-

timation to specific causes, but the comparison to the ECHAM5–HAM reference simulation (global annual mean AOD of 0.14) can give further insight. Higher emissions of mineral dust, of about 1200 Tg yr^{-1} around year 2000 in the free climate mode compared to emissions of about about 670 Tg yr^{-1} in the nudging mode applied in the reference simulation, contribute to the overestimation of AOD in the Saharan outflow region. These higher emissions are a consequence of a shift of the surface wind speed frequency distribution to higher wind speeds in the climate mode. See Timmreck and Schulz (2004) for more details. Stronger anthropogenic emissions in the NIES emission inventory compared to the AeroCom inventory contribute to the overestimation of the aerosol optical depths close to the main anthropogenic source regions compared to both the remote sensing data and the reference simulation."

We further extended the discussion of the AOD in the conclusions to (p 9 c 2 l 27):

"The simulated global annual mean AOD at 550 nm increases from 0.15 at pre-industrial times to 0.22 around the year 2000. The present day values are somewhat larger than satellite retrieved estimates of MODIS(2000-2004 global mean of 0.20) and MISR (2000-2004 global mean of 0.21). However, it has been shown that these satellite retrievals exhibit a positive bias on a local basis. The simulated values are also higher than a year 2000 reference simulation with ECHAM5–HAM that shows a better agreement with the remote sensing data on a regional basis. This overestimation can be attributed to stronger mineral dust emissions in the free climate mode compared to the nudged reference simulation as well as to stronger anthropogenic emissions in the NIES emission compilation compared to the AeroCom emission inventory applied in the reference simulation. From present day conditions to the pollution peak around 2020, AOD is projected to increase to 0.26."

7. Given there are large uncertainties in the results of this paper (as authors acknowledged in the end of the manuscript), there should be a table that lists all the key components (processes) relevant to the aerosol simulation and describes the treatment of those components. For instance, aerosol first indirect

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effect (considered), precipitation effect on aerosol (?), evolution of surface reflectance on aerosol forcing (?), . . .

We agree that a tabulated overview of all key components and their interactions would be desirable. However, owing to the explicit coupling of several complex models this list would be excessively long so that it could only be incompletely reproduced in this manuscript. Therefore, we believe that a detailed description of all included processes and their interactions, as given in the model description, serves the clarity of this study best.

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

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