

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 3

1. To what extent does precipitation change in the transient climate simulation? What role does this play in the observed changes in aerosol lifetimes? What model results can be shown to separate the effects of precipitation changes over time, changes in the point of aerosol emission, and changes in aging processes?

We added a paragraph about the role of the precipitation for the aerosol lifetime as described in the reply to question 1 of reviewer 1.

Unfortunately, without further sensitivity studies it is not possible to untangle the different processes affecting the aerosol lifetime. However, the computational demand of this simulation setup does not allow to perform all relevant sensitivity studies. There-

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fore, we investigated effect of aerosol microphysical processes on aerosol lifetime with a range of shorter sensitivity studies under well constrained conditions (Stier et al., J. Clim., in press) that we cite in the manuscript.

2. At least a summary of how aerosol optical properties are calculated is required so that the reader can understand the associated results. It would fit well in Section 2, perhaps with 2.2 or as a separate sub-section.

As described in the response to question 2 of reviewer 1, we included an extended description to section 2.2 (p 3 c 1 | 36).

3. In several places, the authors say that residence times show non-negligible variations that must be accounted for. The largest change in aerosol lifetime shown is for BC, which is not quite a factor of two. The other species show much more modest (20-30%) changes in lifetime. While the nonlinear effects of mixing state are certainly interesting, what is negligible or not is open to debate. Most would agree that a factor of (almost) two for BC is important, but many would be willing to neglect a 20-30% effect for the other species given the much larger uncertainties in present-day burdens, past emissions, and future emissions. Statements about non-negligible effects should be softened or made quantitative (e.g. these effects are 20% for sulfate etc)

With the shortest (longest) residence time as reference, the aerosol residence times show variations of 40% (30%) for sulfate, of 60% (40%) for black carbon, and of 70% (40%) for particulate organic matter. Considering the cost (and necessary political will) associated with a comparable emission reduction, the phrase non-negligible does to our understanding not seem inappropriate. It is important to note that these variations in the residence time introduce uncertainty on top of other uncertainties - but are rarely taken into account. Additionally, we show in a quantitative “back of the envelope” estimate in the conclusions how this variations could bias simple projections of aerosol radiative effects when these variations are ignored.

4. For the anthropogenically relevant species SU, BC, and POM, their mass shifts from the Aitken modes to the radiatively important accumulation mode . This is an intriguing statement but receives no further explanation. Please explain or delete.

We have extended this paragraph to (p 7 c 1 l 15):

“For the anthropogenically relevant species SU, BC, and POM it is evident that under the higher polluted conditions their mass shifts from the Aitken modes to the internally mixed accumulation mode soluble. This mode is of particular importance for the aerosol radiative effects. On the one hand, hydrophilic particles in that size-range serve as cloud condensation nuclei and play therefore a key role for the indirect aerosol effects. On the other hand, it follows from Mie theory that particles in this size-range have the highest extinction efficiency for the visible wavelengths and therefore the strongest potential to contribute to the direct aerosol effects.”

5. The change in the co-SSA of the soluble accumulation mode is cited as a measure of absorption efficiency and the effect of mixing state on the properties of black carbon. However, the parameter is not well suited to this purpose. It says more about the relative amounts of scattering aerosol to BC emitted than the mixing state of BC per se. Also, it says nothing about the amount of absorption associated with BC in the insoluble modes. Absorption per unit mass of black carbon (as has been used in other studies) is much better. Normalizing to mass of BC accounts for changes in BC emissions and isolates the effect of mixing state.

Of course the co-single scattering albedo is not a measure of the mixing state on the radiative properties of black carbon. We checked the manuscript carefully and could not find the respective paragraph where we claim this. However, we used the phrase “absorption efficiency” somewhat imprecisely in the following paragraph.

“The increasing fraction of carbonaceous aerosols in the internally-mixed accumulation

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mode causes a more than threefold increase in its co-single scattering albedo, and therefore absorption efficiency, from 1860 to 2100.”

We removed this phrase so that this sentence now reads as (p 9 c 2 l 19):

“The increasing fraction of carbonaceous aerosols in the internally-mixed accumulation mode causes a more than threefold increase in its co-single scattering albedo from 1860 to 2100.”

We further changed the explanation of the CO-SSA to (p 7 c 2 l 17):

“ The CO-SSA, as a measure of the contribution of absorption to the total extinction, increases constantly from 0.02 in 1860 to 0.04 in 2020.”

As described in the reply to question 3b of the second reviewer, normalising the absorption by the mass of black carbon seems an unsuitable measure owing to the internal mixture of black carbon with other moderately absorbing species.

6. The fact that a significant fraction of the present-day fine mode is natural is an important point that deserves some elaboration. It is, if anything, a bit overdue that someone quantified this important bias in remote sensing studies. Please elaborate by saying what composes the natural one-third of the fine mode. One can deduce from the paper that only a small amount is fine mode sea-salt and dust (albeit from the 2020 results). What is the rest of the natural fine mode aerosol: DMS-derived sulfate? biogenic SOA?

We have added the following explanation for the source of the fine mode optical depth (p 7 c 2 l 52). Unfortunately, it is not possible exactly quantify the contribution from each source as this would require to repeatedly rerun the model with the individual sources deactivated.

“The natural fine mode optical depth is dominated by volcanic sulfate but shows also contributions from DMS derived sulfate, biogenic POM, as well as from sub-micron sea salt and mineral dust.”

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7. The paper talks about top-of-atmosphere forcings. Optionally, it would be nice to present the atmospheric absorption and/or surface forcings, which would be useful to those interested in hydrological impacts.

We agree that this would be a nice extension of the manuscript. However, owing to the limitations of the radiation diagnostics that we explain in our comment to question 4 of reviewer 1, these quantities have not been isolated in our simulation and are therefore not included in this manuscript.

Technical corrections:

“emissions of POM from secondary biogenic sources” This sentence is confusing. Does it mean that the authors treat secondary organic aerosol (SOA) by lumping it with the primary organic matter (POM)? If so, it would make more sense to call the model tracer simply organic matter (OM) with both primary and secondary contributions.

The formation and evolution of secondary organic aerosols is highly complex and associated with vast uncertainties. Thus, the HAM aerosol model in the current version does not explicitly deal with this processes. Instead, following the recommendation of the AeroCom aerosol model inter-comparison, the biogenic monoterpene emissions of Guenther et al. (1995), are scaled by the factor 0.15 to estimate the production of secondary organic aerosol from biogenic sources. This proxy of biogenic secondary organic aerosols is applied in the model as primary emissions.

“POM” is defined in the introduction of this study, following a widely used terminology, as “Particulate Organic Matter” not as “Primary Organic Matter”.

We added the following paragraph to the description of the emissions in Section 2.6 (p 4 c 2 | 19):

“ 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

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applied in HAM as primary aerosol source.”

“the inter-annual variability lies at $\sigma = 0.04$ ” : Presumably the standard deviation value is calculated taking data from all years into account. Therefore, it includes longterm trends caused by anthropogenic influences as well as natural climate variability. It s a bit confusing to call this inter-annual variability , which suggests natural climate variability. Call it something else for clarity.

We recalculated the inter-annual standard deviation according to the following new description in the manuscript (p 8 c 2 l 38):

“ The inter-annual variability is analysed in terms of the normalised inter-annual standard deviation $\sigma^{norm} = \sigma(E')/\bar{E}$, where E' is the inter-annual emission flux perturbation, calculated as difference between the annual-mean emission flux and its 20-year running mean, and \bar{E} is the integration-period mean emission flux.”

“In combination with the stagnation and even reversal of the increase of the solar irradiance after a bout 1930-1940, this [volcanic and anthropogenic aerosols] explains the well simulated small trend in global surface temperatures between 1950 and 1970” This very interesting trend in the 20th century temperature record is well simulated by the model but only mentioned in passing. Is this a new result? Have other models reproduced this feature as well? If so, it would be appropriate to cite them. If not, it seems like this result deserves more than a passing mention. What model features/inputs are necessary to give this good agreement with the observed temperature record?

We did not provide further details here as this topic has already been generally addressed in the IPCC (2001) assessment report (Chapter 8, McAvaney et al., 2001). However, therein the focus was on the relative contribution of greenhouse gases, natural forcings (stratospheric volcanoes and solar variability), and sulfate aerosols to the simulated surface temperature trend. Here we show that our simulation well reproduces the observed temperature trend considering the effects greenhouse gases,

natural forcings and all major global aerosol components. Apologies for the repetition, but to isolate the necessary model features to derive the observed temperature trend would require to numerously repeat this simulation and is beyond our current computational resources.

We added a reference to the respective chapter in the IPCC 2001 report (p 8 c 2 l 38):

“(see also discussion in McAvaney et al., 2001).”

“The projected increase in low-latitude carbonaceous aerosols cause an enhancement of local monsoon regimes.” See the general comment above about the focus of the paper. This result is mentioned only in passing in text and no figures are dedicated to illustrating it. Moreover, a separate paper (R2005) analyzes it in detail. To maintain the focus of the paper, I think it makes sense to delete this from the text and conclusions.

We agree that these dynamical processes are not the focus of this manuscript. However, they affect the sources and distribution of the investigated aerosol system. While the scope of this study does not allow to include all relevant connections, we believe that these cross-references allow the interested reader to obtain a better picture from the individual publications on this simulation.

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

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