

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

Anonymous Referee #3

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General Comments:

This paper presents results from a long simulation with a model that couples atmospheric and ocean dynamics to aerosols and ocean biogeochemistry. The focus of the paper is on nonlinear responses of aerosol burdens and optical properties that result from mixing state effects. In fact, the main ideas of the paper are substantially similar to the author's in-press “Emissions-induced nonlinearities in the global aerosol system” paper. The relationship between this paper and the in-press paper should be clarified in the introduction: what more specific analysis is added to the basic idea of the earlier paper? Having an online aerosol module in a long climate simulation is rare and interesting. However, the focus of the paper on aerosol mixing state effects does not

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demand a long transient simulation. The points in the paper could be made just as well by running separate preindustrial, present-day, and future scenarios. I suggest that the title be changed to reflect this emphasis to something like “Nonlinear mixing state effects on aerosol properties: past, present, and future”.

It's true that passing reference is made to aerosol results that stem from the coupling to climate and ocean biogeochemistry (dust emissions changes and DMS cycle, for example). These are not analyzed in any detail, however, and separate papers have been written on each of them. I recommend that, for clarity, they be deleted from the paper to clarify the focus on mixing state effects. Since their results are only briefly mentioned, this could be done easily in the introduction so as to leave the main body of the paper focused on mixing state effects.

Overall, the paper is interesting and mostly very well written. I recommend it be published once the focus is clarified a bit and the following issues addressed.

Specific Comments:

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?

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.

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

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(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)

4) “For the anthropogenically relevant species SU, BC, and POM, Ë 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.

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.

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?

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.

Technical Corrections:

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“emissions of \ddot{E} 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 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 long-term 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.

“In combination with the stagnation and even reversal of the increase of the solar irradiance after about 1930-1940, this [volcanic and anthropogenic aerosols] explains the well simulated small trend in global surface temperatures between 1950 and 1970 \ddot{E} ” 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?

“The projected increase in low-latitude carbonaceous aerosols \ddot{E} cause an enhancement of local monsoon regimes \ddot{E} ”. 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.

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

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