A pathway analysis of global aerosol processes
N.A.J. Schutgens & P. Stier

We thank the anonymous reviewer and Pete Colarco for carefully reading our manuscript and providing us with useful comments. Below you can find our responses to their questions/comments.

Answers to referee's comments

Anonymous referee nr 1

*Pg 15047, line 19-21. Include Adams and Seinfeld (2002) in JGR (TOMAS model) in list of global aerosol microphysics model references*

Adams and Seinfeld 2002 should have been in there but somehow was dropped. Thanks for spotting this.

*Pg 15051 final paragraph re: SOA. Based on this paragraph and not much mention of organic condensation elsewhere, is it only sulfuric acid condensation that is included in the pathway analysis? Organic condensation is known to dominate atmospheric nanoparticle growth (see for example, Riipinen et al. 2012, Nature Geoscience). Will this omission cause underestimation in the particle growth tendencies? I suggest discussing in more detail the impacts of organic condensation.*

This is a very good point. The standard version of ECHAM-HAM uses a simplified treatment of SOA where all organic volatiles condense at the time of emission. Consequently, particle growth through organic condensation is modeled but in an abstract way. In our budget this “condensation” process is lumped with other emission processes. There exists another version of ECHAM-HAM that uses a more sophisticated treatment of SOA (O’Donnel et al. 2011). We hope to publish results for this version, together with a comparison against another model (HadGEM-UKCA) in the future.

We have extended the paragraph with more explanation about our choice and its consequences. Note also that our paper initially stated that explicit SOA treatment is the standard model version, but it is not. This has been corrected.

*Pg 15052 line 7: "looses" should be "loses"*

Agreed, corrected.

*Pg 15052, line 25 and beyond. Might be worth mentioning what exactly the emitted sulfate size distribution is quantitatively (e.g. what fraction is distributed to what mode).*

Agreed, done.

*Pg 15056 and elsewhere. Make sure you are consistent with your spelling. "Ageing" is mostly used in the text but Fig. 10 uses "aging".*

Agreed, we only use ageing now.

*Pg 15059 line 13: I suggest giving the degree by degree resolution of T31L19 for those who might not be familiar with the ECHAM naming conventions.*

Agreed, done.

*Sect. 4.1 and Fig. 2: Any particular reason 1/m^3 is used as the unit instead of 1/cm^3? The latter is more universal for CCN, and would lower the magnitude of your numbers a bit in the legend.*

Other column densities and budgets in our paper use 1/m^3 or 1/m^2 so for consistency we decided to stay with meters instead of cm.

*Sect. 4.3 and Fig 4: Is it possible to include percentages of the total budget for the mass and number tendencies? The thickness of the arrows already gives a rough idea, but something quantitative would be useful.*

The figures are already quite crowded. Adding percentages might not be that interesting we think. The *logarithm* of the flux scales with the arrow's width, so most budgets would be close to zero anyway.

*Sect 4.4: Some issues here with Figure numbering. On line 14 of page 15063, I believe the correct reference is to Fig. 5, not 6. Similarly, the references to that figure in Sect 4.4.1 should be changed to Fig 5a, 5b, etc. There are some things listed as "Fig ??" which I believe should read Fig. 6 as it is presented in the list of figures. After that the figure numbering seems correct.*

Agreed, corrected.

*Fig 5, 8, 11... : in each of the 6 panel figures, there is a number in between the two columns in the last row (in Fig. 5, "45"). What is this? Explain or delete if a typo.*

Indeed, this seems to be caused by the ACPD latex style files. Running the same tex-file for ACP removes these numbers. We'll keep an eye on this.

*Sect 4.5: Can the correlation coefficient values for Fig 24 (and perhaps for some of the other regions) be reported? Yes the correlations look good, but best to be quantitative.*

This is a good idea. Correlations are now given in the figures and a new table lists them for different regions. The vast majority of these correlations is larger than or equal to 0.95 for any region and either sensitivity experiment. Text is added to Sect. 4.5 to explain the new information. The figures for East Asia have been replaced with those for North America which shows larger (PI-PD) deviations.

Comments by Pete Colarco

*The authors are to be commended for their general thoroughness, but the paper is challenging to read. The attentive reader will need their scorecard handy (Tables 1 & 2), but even so this is a paper that will need to be viewed on a large computer monitor to make the figures at all usable. I think the color schemes for the pie chart figures are difficult to read, and so difficult to interpret without the aid of the text. I would suggest perhaps a color scheme like those suggested at http://colorbrewer2.org for "qualitative" data as useful to people who like myself have a hard time distinguishing shading of various blues (e.g., Figure 5b).*

The referee points out an issue that we struggled with ourselves. In particular our colourtables are not very accommodating to color-blind researchers. Unfortunately, we

have not found a better way to represent our results. The suggested website is very illuminating in this respect (great resource, by the way!): only 2 different colour schemes are recommended for qualitative separation of data. Neither of which works for colour blind people and both of them accommodate at most 12 different classes. In contrast we are dealing with 33 different classes (see our Table 2), of which many are aggregates already.

We suppose the biggest problem is posed by the blue and green colour scales used for coagulation of resp nucleation and soluble Aitken particles. For these processes, a sequential colour scale was used and we tried Brewer scales to see if any improvement might be found. The difference between the Brewer scale and our original scale is however small and we could not decide which one is better. Finally, we considered using two colour scales instead of just one for e.g. coagulation with nucleation particles. One could use a blue scale for coagulation with soluble particles and another scale for coagulation with insoluble particles. This would allow us to add more differentiation. However, due to the use of many colours in our paper no secondary colour scale readily suggests itself (without causing conflicts or confusion in some other figure).

We also feel it is important to use a single consistent colour scheme throughout the paper. In light of the above, we suggest to stay with our original choice. Note that different shades of colour always refer to the same process but for different species or modes. Numbers of course refer to the relevant modes.

*1) Please clarify in the model setup description what year was run? Were there any important events in that year (volcanoes, wildfires)?*

The model was run for the year 2000. Standard AEROCOM emissions were used that contain no special events. Text of the paper has been updated.

*2) For the reduced spatial resolution run I am a little surprised at the consistency of the results. Although I understand the tuning of dust and sea salt emissions mentioned, my experience has been that clouds may be quite different across changes in horizontal resolution, with possibly large effects on in-cloud sulfate production, for example. Your baseline is already relatively coarse (_2x2 degree) so maybe your cloud fields aren't so different at the coarser resolution (my experience was in moving from 2x2.5 degree grids to 0.5x0.625, for example). Could you explain a few sentences more about the nudging? Are you imposing cloud fields, or are those solved from the imposed dynamics?*

The nudging is only applied to windfields (vorticity, divergence), temperature and specific humidity. Sea ice and SST are prescribed. Cloud water and number density are prognostic variables in our model. They develop freely, given atmospheric conditions. A possible explanation for the consistency of results across two different resolutions maybe that we consider only area-averages: while results maybe be very different per gridbox, they are similar over larger regions.

*3) Regarding the discussion of pre-industrial emissions (p. 15062, line 10), would preindustrial meteorology matter to the results?*

While aerosol will exert a feedback on the climate system and this will certainly affect meteorology which will in turn affect the aerosol, the effect of the aerosol feedback on itself tends to be smaller than natural variation in the meteorology anyway. We have shown this using an ensemble of model simulations with perturbed emissions (work in progress). See also Schwartz et al. JGR 2013.

*4) Section 4.4.1, page 15064 line 2 (and throughout section):reference to Figure 6 should be to Figure 5, and reference to Fig. ?? should be to Fig. 6 (also in Section 4.4, line 14).*

Agreed, corrected.

*5) page 15065, line 7: I think reference to figures 8d & e should be to Figure 8 c & d.*

Agreed, corrected.

*6) I'm a little confused about the presentation of the aging processes for the hydrophobic Aitken mode (Section 4.4.2, esp. lines17 - 20 and Figure 9). The text talks about coagulation with nucleation particles (which is process 15 on my scorecard) which wouldn't be a loss of particles in the Aitken mode, I realize (what is plotted in Figure 9) and condensation is called out in the lower troposphere. These things all make sense, but I can't tell what they have to do with Figure 9, where the processes illustrated there aren't seemingly discussed. Please clarify the intent here*

The explanation is that both processes coat hydrophobic particles with a sulfate layer, making them hydrophilic. In the model, this is done by transferring part of the hydrophobic mode to its hydrophilic counterpart (same model treatment is used for the Aitken, accumulation and coarse modes). This was discussed in Sect. 2.8. A line has been added in Sect 4.4.2 to clarify this further.

*7) There doesn't seem to be any reference or discussion of Figure 17.*

Actually there is but the order of the Figures and the explanation in 4.4.5 was different, possibly creating some confusion. This has been corrected.