Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Improving the simulation of global aerosol with size-segregated anthropogenic number emissions" *by* Filippo Xausa et al.

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Review of "Improving the simulation of global aerosol with size-segregated anthropogenic number emissions"

This paper introduces the GAINS bin-resolved anthropogenic emission inventory to the ECHAM-HAM aerosol-climate model, and compares the results to a baseline simulation in which Aerocom emissions are used. Emissions in which particle numbers are resolved may be very useful in improving the climate models of the future. This paper is a first step in this process, and it emphasises the importance of more refined approaches that will hopefully follow it. The text is of good quality and the paper should

be published in ACP once the comments below (and those of the other reviewer) are addressed.

1 Major comments

1. The GAINS emissions come in 9 rather coarse size bins. If I understand correctly, at the moment these sizes are only used to determine whether to put the emissions into the Aitken or the accumulation mode. How the authors do this is a little unclear, as stated by reviewer 1, and should be clarified. Presumably, with size bins 3,10,20,30,50,70,100,200,400,1000, bins 2-6 inclusive are all assigned to the Aitken mode and bins 7-9 to the accumulation mode? The authors convert each AeroCom emitted component mass to number using the ECHAM-HAM density and size distributions with globally fixed mean diameters of 60 and 150nm. Then the component fractions for GAINS are straightforwardly derived from the ratio of the AeroCom components. The result is numbers of particles in each gridbox for each component and each mode using GAINS emissions (this is well explained in the text). Then the authors convert these numbers back into masses for each mode using the ECHAM-HAM size distribution, and put them back into the model. So the mass emissions can differ between GAINS and AeroCom, in line with the number concentrations from GAINS, and the ratio of Aitken and accumulation mode emissions can differ, but within each mode, the mode radii of the emissions is still fixed globally to the AeroCom values (30nm for the Aitken mode and 75nm for accumulation).

   So, if my interpretation is correct, Aitken mode emissions in GAINS all have the same mode diameter as the Aitken mode emissions in AeroCom, and likewise for the accumulation mode. It would be helpful to say this a bit more explicitly at line 334. However, the ratio of Aitken to accumulation mode emissions can vary per gridbox in GAINS, while they cannot in AeroCom - this is the key step forward

the authors have made, and it should also be made clearer with a couple of extra sentences.

Then, it would be very helpful to see what total mass in each aerosol mode the authors end up with in the GAINS model. Can they add the information about mass emissions to Table 3 (even though this would be easy to calculate by hand) and more importantly the mass concentrations to Table 4? And add subfigures to Figure 6 showing the spatial variation of the mass concentrations the model produces? I suspect that the authors will then also need to discuss whether these masses are reasonable in the text: I would speculate that in a few regions they will be unrealistically high, because the assumptions about the emission size distribution will probably give the particles too large a diameter within each mode. This would be interesting.

Furthermore, the authors should comment that (if I understand everything correctly) their approach (while perfectly reasonable) doesn't fully exploit all the information available in the GAINS size distribution, because the ECHAM model structure presumably doesn't allow the emission diameter to vary on a per-gridbox basis. In principle, if the implementation is similar to the models with which I am familiar, the authors could write some more code for ECHAM to read in the emission diameter for each grid-box alongside the mass emissions, and then adjust the mode diameters in each gridbox when the emissions are added to the existing particle concentrations to account for the diameters of the added particles. I appreciate that this might get quite complicated and must be beyond the scope of the current study.

2. The authors might do a sensitivity study simulation in which the 2.5% of primary sulphate is varied (or to use a more sophisticated scheme for "primary sulphate"). The existing treatment is pretty crude (the 2.5% number is highly uncertain and spatially very variable) and particulate sulphate is especially important for any paper concerning anthropogenic particulate number emissions. The AeroCom

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mass emission from Dentener et al (ACP 2006) for sulphate is about 90% of the total, so 2.5% of this is about one-quarter of the total emissions considered. See studies by Luo and Yu https://www.atmos-chem-phys.net/11/1949/2011/ or Stevens et al https://www.atmos-chem-phys.net/14/13661/2014/. At the very least this uncertainty should be discussed in the text.

Thinking of the conclusions, I imagine the treatment of primary sulphate would make more difference to the results than including nucleation of ELVOCs. Another possible improvement that could be mentioned as further work would be including the seasonal cycle of emissions (comparing to Maccity instead of Aerocom for example), as I suspect this would also make a big difference.

3. To add to the comments of the other reviewer concerning the quality of the comparison between GAINS and the ECHAM-HAM default, the Aerocom dataset is for 2000 and the GAINS set for 2010, though the authors mention the 2000 emissions are also available. In some areas the emissions must have changed quite a bit between 2000 and 2010. Do the authors have a quantitative indication of this from GAINS that they could discuss in the text?

2 Minor comments

Figures 2 and 3 could be reduced to one figure with three subplots (at the moment, Figure 3 is not mentioned in the text) and discussed in more detail.

There is a very large and widespread difference between GAINS and Aerocom in tropical Africa, also visible in Figure 3. The anthropogenic emissions in this area are presumably from agricultural waste (100nm diameter particles). These will be accumulation-mode in GAINS and Aitken-mode in Aerocom, if I understand correctly. This appears to be the case, from Figure 6. Therefore one would expect

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more of them overall in Aerocom than in GAINS, but the reverse is observed. Please could the authors discuss possible reasons for this in the text?

Line 218 I would imagine that such an activation nucleation scheme would lead to overprediction of aerosol formation over oceans. This is of secondary importance for this study (of non-shipping anthropogenic emissions), but the authors might wish to comment to this effect in the text. The authors might also specify whether or not BVOC oxidation products are able to grow particles to 3nm, or whether it is only sulphuric acid, as this has been done both ways in the literature.

Line 383, Table 3: Do the medians here include ocean grid-boxes, or are they just for land boxes? Please specify. The caption makes it sounds like they include the ocean. If this is the case the median anthropogenic emissions are presumably zero or very close to zero (all the emissions are natural over the ocean as shipping is not considered) and the median is not a helpful quantity. Please re-calculate it just for land grid-boxes.

Line 404: "sulfate condensation" presumably also condensation of BVOC oxidation products? Are there any anthropogenic VOCs in this version of the model? I assume not – perhaps the authors can comment on whether or not condensation of anthropogenic VOCs is likely to be important in (for example) Nanjing?

Figures 6-8 are these at the surface level? Please specify in the captions.

Figure 5: please make axis and legend labels larger. One legend for all subfigures would suffice.

3 Textual and stylistic comments

The paper is well written. A few things to correct I noticed on my way through:

- The sentence "In this work.." at line 30 is rather too long

- Line 35: "Special attention was paid to accumulation mode particles..."
- Line 71 "being" is not needed
- Line 146 ...with the M7...
- Line 371 ...the GAINS implementation
- Line 414 "a tendency to underestimate, especially for the locations with"
- Line 478 particle->particles
- Figure 4 caption: please replace with "Number of particles" or "Number".