

We want to thank the referee for the very valuable comments. We appreciate the efforts and the time that the referee reserved to draft all the comments which helped us enhance the quality of our manuscript. We answered the comments as below.

Main comments answers.

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.

1.1R. *The ratio of Aitken to accumulation mode emissions actually varies per gridbox in the AeroCom data set. The Aitken/accumulation ratio is fixed per anthropogenic source sector, but the ratio can vary per gridbox due to varying contributions from different sectors. However, GAINS emissions are organized into much more detailed emission sources than AeroCom, with different particle number emissions and size distributions related to different fuels and technologies, as stated in the fourth paragraph of section 1. We understand that this point could be made clearer, thus we decided to explain this concept clearly in the second paragraph of section 2.3: "It should be noted that the ratio of Aitken to accumulation mode emissions can vary between grid cells in both AeroCom and GAINS. In AeroCom this variation is due to different mass-to-number conversion factors for different emission sectors, but in GAINS the size distributions are different also for different technologies and fuels within the emission sectors (e.g. different vehicle technologies, different domestic stove categories, diesel fuels with different sulfur contents, different coal types)".*

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.

1.2R. *Although we believe that adding further information related to the mass emissions may not be relevant in our research, we agree that a visual representation of the spatial distribution of the total mass concentrations could enhance the quality of our manuscript. We decided to add three plots as supplementary material, which include the total PM_{2.5} concentrations from AeroCom simulation, the total PM_{2.5} concentrations from GAINS simulation, and the ratio between GAINS PM_{2.5} and AeroCom PM_{2.5}.*

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.

1.3R. *We agree that the implementation of GAINS does not fully exploit all the information available related to the size distribution from the GAINS data set. Although it would be possible to let the ECHAM model read in the emission diameter for each grid-box alongside the mass emissions (as suggested by the referee), "the implementation of these settings would be quite laborious and beyond the scope of this study". We decided to specify this limitation more clearly by adding the previous sentence in the second paragraph of section 2.3.*

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 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.

2.1R. *Unfortunately we couldn't understand part of this comment. It is unclear to us how 2.5% of the mentioned 90% could represent about "one-quarter" of the total emissions (did the referee refer to 25% instead of 2.5%?). However,*

*we agree that the assumptions for the SO₂-to-(primary)particle conversion are important and certainly could affect the final results. We decided to make this clearer by mentioning this concept briefly in the third paragraph of section 3.2, where we discuss the model's sensitivity to these parameters and the particle nucleation in plumes. In more detail, we explained that our study "does not take into account any sensitivity analysis based on the primary sulfate emissions parameterization". We also added a reference to the study of Luo and Yu as suggested by the referee. Also, In our manuscript we referred to Lee et al., 2013 (<https://www.atmos-chem-phys.net/13/8879/2013/>) in the last paragraph of section 3.3, in which a thorough sensitivity analysis was performed to address these uncertainties, including the impact of different assumed median radii (see **3R**. answer to the main comment from Referee 1).*

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.

2.2R. *We agree that the implementation of seasonal cycle emissions in GAINS may be an important improvement in the future. We decided to specify this briefly in the last paragraph of section 4.*

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?

3R. *Even though the GAINS online model, with which the emissions of greenhouse gases and "traditional" air pollutants can be calculated, extends from 1990 to 2030 and beyond, the particle number emissions are currently available only for years 2010, 2020 and 2030. We will mention this concept briefly also in the revised version of the manuscript in the last paragraph of sector 2.2.*

Additionally, we decided to add the following notification in section 3.2, discussing Figure 4: "It should be noted that the emissions from different emission sources and observations are not all from the same years. However, even though the GAINS emissions are for year 2010 and AeroCom emissions for year 2000 (and observations for the years indicated in Table 2), the differences in the modeled concentrations with GAINS and AeroCom at most polluted sites, reaching factors of 2 and above, cannot be expected to originate from differences in emissions between 2000 and 2010."

Specific comments answers.

1. 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.

1R. *As replied to the specific comment from first referee in 11R, we now refer to Figure 3 in the text and leave the distinction between Figures 2 and 3, as they describe the emissions quite differently.*

2. 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 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?

2R. *There is difference in emissions in Africa between GAINS and AeroCom, but their origin is not presumably in agricultural waste burning. In Paasonen et al. (2013), agricultural waste burning is shown under “Agriculture” source sector and it doesn’t play a dominant role in GAINS emissions (Fig. 4 in Paasonen et al., 2013). The largest African particle number source sector in GAINS for year 2010 is road transportation, where the impact of high-sulfur fuel is an important factor. We would assume that this is the major difference between GAINS and AeroCom emissions in Africa.*

3. 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.

3R. *BVOC oxidation products participate in the growth below 3 nm, i.e. in the growth rate of the applied Kerminen-Kulmala equation. This is now explained more explicitly in the revised manuscript in the last paragraph of section 2.1.1: “The particle growth from nucleation size to the d_p of 3 nm was calculated according to Kerminen and Kulmala (2002), considering both sulfuric acid and organic vapour condensation”. Activation-type nucleation produces a total particle concentration of $\sim 1000\text{-}2000\text{ cm}^{-3}$ over large parts of the oceans (Makkonen et al., 2012, Supplementary Figure S1), which might be an overestimation. However, considering the focus of the current study on terrestrial Aitken and accumulation mode concentrations, we believe that the assumption is not affecting our analysis to meaningful extent.*

4. 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 sound 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 recalculate it just for land grid-boxes.

4R. *We understand that the caption needs to be modified. In Table 3 we modified the unit of the continental emissions from number(N) $\text{m}^{-1}\text{ s}^{-1}$ to annual total number (N yr^{-1}), and we specified that we included continental emissions only, with the expression “Annual total particle number (second and third columns) and global average ratios (fourth and fifth columns) of continental anthropogenic input emissions” in the caption. Also, we corrected the R_{tot} mean of accumulation mode.*

5. 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?

5R. *The M7 implementation in ECHAM5.5-HAM2 assumes that a monolayer of sulfate is required to transform insoluble Aitken mode particle to soluble mode. We have not changed this assumption. BVOCs oxidation products can condense on insoluble Aitken mode, but not transfer particles to soluble. In our implementation, there are no anthropogenic VOCs, although they could provide additional growth and mass in certain regions e.g. Nanjing.*

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

6R. *Yes they are at surface level. We decided to specify this in the caption.*

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

7R. *We agree that Figure 5 could be improved further. We decided to modify Figure 5 according to the referee's suggestions.*

Stylistic comments answers.

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”.

8R. *We appreciate the referee's effort to evaluate the stylistic form of our manuscript. We applied all the referee's suggestions to our manuscript.*