

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. It is unclear how the authors convert from the sectional emission size distributions in GAINS to the modal scheme of the model. (e.g. Do you fit the GAINS size distribution to lognormal modes? If that is the case how do the median radii from GAINS compare with the assumptions in ECHAM-HAM? Or do you take the total particle number in the defined Aitken and accumulation modes and emit them using the same median radii as in the previous assumption?)

1R. *We took the total particle number in the M7-defined Aitken (10-100 nm) and accumulation (100-1000 nm) modes and emit them using the same median radii as the ECHAM-HAM assumes for the AeroCom emissions data set. We understand that this concept was not expressed clearly enough in the manuscript. We decided to modify the second paragraph of section 2.3 by adding a few lines to clarify this step: “The conversion of GAINS emissions from sectional to modal size distribution is performed by taking the total particle number in the defined Aitken and accumulation modes and emitting them with the same median radii as for the ECHAM-HAM default assumptions (Stier et al., 2005)”.*

2. I think the authors should be careful when claiming this as a comparison between number concentration in AeroCom and GAINS. Different models convert aerosol mass to number differently. For instance, a different assumed count median radius for organic aerosol would result in a different number concentration. Really, the comparison is between aerosol number in GAINS and the default assumptions in ECHAM-HAM.

2R. *We understand that the expression “comparison between AeroCom and GAINS in terms of emissions” in the last paragraph of section 1 could be misleading. We redrafted it by stating that our work “will include a comparison between the novel GAINS implementation and the default implementation of AeroCom in ECHAM-HAM”. Also, we want to clarify that the main goal of our work is to improve the model’s capability for estimating particle number concentrations, and assess the feasibility of using GAINS in a global climate model, as stated in section 1. In order to achieve this, the AeroCom data set was used in the simulation to represent a point of reference for the simulation with GAINS dataset. We applied ECHAM-HAM mass-to-number default assumptions in GAINS implementation in order to respect the balance of mass/number ratio in the ECHAM-HAM model as it is done for AeroCom emissions in ECHAM-HAM by default (with ECHAM-HAM default mean radii assumptions).*

3. Considering points 1 and 2, it is unclear to me if the differences in the simulations result from different total aerosol number concentrations or different emission sizes (or both). For instance, POM from biomass burning and biofuel is often emitted with a larger count median radius. How much of the

observed differences in aerosol number between the 2 simulations could be accounted for by changing the assumed emissions count median radius and standard deviation in the ECHAM-HAM model (and thereby changing number)? Or is the regional variability in size distributions from various technologies in GAINS that is important?

3R. *The simple answer to the question is that the differences result from differences in both total number emissions and their size distributions. It should be noted that, as described in the manuscript in section 1, the GAINS and AeroCom number emission are derived in a very different way. While the AeroCom number emissions are derived by converting the mass emissions to aerosol number emissions by applying one log-normal mode, in GAINS many of the number emission factors, especially in traffic and domestic combustion sectors, are directly based on the literature or number emission databases, and the GAINS size distributions often include more than one mode. Because there is no bi- (or tri-) modal emission size distributions in AeroCom, changing the assumed emission count mean diameter and standard deviation does not lead to same number emissions than GAINS: in GAINS the mode with smaller mean diameter typically dominates the number emissions, but the mode with larger diameter often dominates or at least contributes significantly to the mass emission, even in size range with diameter below 300 nm (which is often calculated before doing the mass to number conversion). Thus, adjusting the AeroCom single mode measures to correspond to the dominant number mode in GAINS would not lead to the same number emissions than GAINS. The regional variability in size distributions for different fuels and technologies also certainly adds to the differences between the simulations.*

As a whole, the impacts of the different factors causing the differences in simulated results are very difficult to determine. These impacts should be analyzed with the emission size distributions directly and perhaps by modifying gradually either AeroCom or GAINS emissions or both. While this is an important analysis to be made in near future, we find that it is not in the scope of this manuscript, where we want to present the first implementation of the first version of GAINS particle number emissions in global circulation models. In relation with observed differences accounted for by changing the assumed median radius and deviation we referred to Lee et al., 2013, in section 3.3, which describes the model's CCN sensitivity to emission diameters.

4. The comparison to the observation sites is not very quantitative. A linear regression (or something similar) could provide quantifiable metrics to compare the 2 simulations.

4R. *We agree that this comparison needs to be improved to deliver the message more clearly. We decided to add a numerical comparison in terms of relative bias to indicate how the modeled concentrations deviate from the observed value. This analysis shows quantitatively that the GAINS simulations are closer to the observations than AeroCom simulations.*

We added the following to the methods in section 2.5: "In addition to visual comparison between the modeled and observed concentrations, we calculated

the relative bias as $\exp\left(\left|\log\left(\frac{\text{model}}{\text{observation}}\right)\right|\right)$. This relative bias returns the factor,

larger than 1, with which the model under or over predicts the observation."

In the results, first paragraph of section 3.2, we added: "The average relative bias for the accumulation mode concentrations with GAINS emissions was 2.37

and with AeroCom emissions 3.51. The average relative bias for the Aitken mode concentrations with GAINS emissions was 2.25 and with AeroCom emissions 2.12.”

Specific Comments Answers

1. The title is perhaps misleading, as the GAINS model only improves accumulation mode number concentration.

1R. *We understand that the word “improving” in the title may sound too strong, however we also believe that our work represents a step forward in modeling global aerosol. We decided to replace the title with “Advancing global aerosol simulations with size-segregated anthropogenic particle number emissions”.*

2. Lines 91-96: What is meant by “input”?

2R. *A model can elaborate data provided as an external file, which represents part of the model “input” We specified that by using the expression “model-input” in section 1.*

3. Lines 206-209: What is the relationship between the GAINS inventory and the ECLIPSE inventory?

3R. *We clarified this issue by reformulating the end of section 2.2: “In this study, we applied the gridded emissions for year 2010 (Paasonen et al., 2016), in which the activity measures and emission abatement technology shares are based on the ‘ECLIPSE version 5’ dataset (Klimont et al., 2016) developed within the EU FP7 ECLIPSE project (Stohl et al., 2015). The gridded dataset...”.*

4. Sections 2.1 and 2.2: Given the importance of modeled representation of size-resolved aerosol to this paper, I think the discussion of aerosol schemes should be its own section. Section 2.1 starts with discussing aerosol representation and Section 2.2 ends discussing shipping and biogenic emissions.

4R. *We understand that the aerosol schemes section could be drafted as its own section. We think that the settings of the ECHAM and the aerosol schemes are strongly connected, however we agree that all section 2 could be rearranged differently. We decided to change the structure of the “Material and methods” section 2 as below:*

2.1 The ECHAM5.5-HAM2 climate model

2.1.1 Aerosol microphysics

2.1.2 Natural emissions

2.1.3 Anthropogenic emissions

2.2 Emission scenario model GAINS

2.3 GAINS implementation in M7

2.4 Simulation setup

2.5 Comparison with observation

5. Lines 305-308: Are the emission sectors in GAINS different from those in AeroCom? How does this impact the comparison between the simulations?

5R. *Although both AeroCom and GAINS datasets represent anthropogenic emissions, the sector-wise information differs in the two datasets. With the exception of shipping, we assume in our analysis that both datasets represent total anthropogenic aerosol emissions. While the division into different sectors may have only minor direct impacts on the simulation results, the main part of the differences in the two simulations could be explained by the detailed sub-sectoral information in GAINS as opposed to AeroCom. Since the aerosol fields simulated by ECHAM-HAM do not retain any source-specific information, we can not quantify any sector-specific impact from our simulations.*

6. Lines 324-334: I was a little confused by this section. Doesn't the GAINS (or ECLIPSE) inventory have mass concentrations?

6R. *GAINS does provide mass emissions of BC, OC and PM1, also with ECLIPSE scenarios. However, there are some issues due to which we decided not to use these for the first implementation of GAINS particle number emissions.*

Firstly, applying GAINS mass emission factors would require replacing both the aerosol mass emissions and SO₂ gas phase emissions, which are in ECHAM-HAM default emissions now taken from AeroCom. This would lead to more detailed studies of the impact of different emission vectors on the modeled concentrations and size distributions: what are the impacts of differences in mass emissions, what is the impact of differences in SO₂ gas phase emissions and what is the impact of different number emissions and their size distributions. We have planned to continue the implementation of GAINS emissions by next replacing the mass emissions with the GAINS/ECLIPSE emissions, but since the emission data sets are not entirely consistent in the way they are constructed, we find that this is not within the scope of this first manuscript.

*Secondly, the GAINS PM1 emissions are not as frequently updated as BC and OC emissions. This is because PM1 is a measure which is not strictly tied to the legislation (like PM2.5), nor to the traditionally recognized health/climate impacts of the aerosols (like BC and OC). Thus, the calculation of sulfate aerosol emissions, based on subtracting BC and OC mass from PM1 is not necessarily reasonable, when BC, OC and PM1 emissions are not totally consistent. On the other hand, estimating sulfate from PM2.5, BC and OC is also not reasonable, because in PM2.5 already several other compounds (ash, unburned parts of the hard fuels etc.) have major contribution to the mass. It is possible to replace the AeroCom SO₂ emissions with those from GAINS, and use the same 2.5 % share for sulfate aerosol (see answer **2.1.R** to the main comment from Referee 2), but this leads to drastic expansion of the needed analysis, as stated above.*

We still agree with the referee that this issue should be better expressed in the manuscript, and we will add a text to the manuscript at the end of section 2.2, stating that implementing GAINS/ECLIPSE mass emissions is not straight forward and that we are planning to make this implementation in the next stage of our project.

7. Are "Rtot" and "Rattot" different (perhaps a typo? I cant find the definition)?

7R. *"Rattot is a typo. It is now corrected.*

8. Is Rgrid weighted by surface area of the gridcell?

8R. Yes it is. It is now specified in the caption of Table 3.

9. Lines 399-400: I thought the composition between the 2 simulations was held fixed (though as in Main point 5 I found this unclear)?

9R. According to ECHAM-HAM assumptions, the mass-to-number conversion of Aitken mode particle of AeroCom is performed by the model by using defined shares of BC, OC and SO₄, while the accumulation mode emissions include SO₄ particles only. In the GAINS particle number data set, there is no information about particle composition. It can be speculated that the actual composition in GAINS is different than what it is assumed for AeroCom. However, the shares of BC, OC and SO₄ components were held fixed at the moment of implementing the GAINS data set into the ECHAM-HAM, so that it was consistent with AeroCom. We understand that mentioning the role of the data sets composition may be irrelevant or even misleading while discussing the data sets comparison from Table 3. We decided to redraft the last paragraph of section 3.1. In more detail, we reduced the length of the paragraph and we specified the main difference between the two data sets in terms of Aitken mode and accumulation mode particle emissions: "It should be noted that in the ECHAM-HAM assumptions made for the AeroCom emissions, fossil fuel and biofuel emissions are implemented in Aitken mode only. In more detail, all BC emissions from AeroCom are implemented in the M7 module as insoluble Aitken mode particles, which are converted to soluble particles after sulfate condensation. In GAINS, the particles estimated to contain BC are distributed in diameter ranges around 100 nm. The difference between the diameters of emissions from fossil fuel and biofuel combustion is the major reason behind the differences in accumulation mode emissions and concentrations."

10. Lines 544-545: Is it possible to calculate CCN0.2 from the same measurements sites used to compare in Figure 4 in order to provide a more quantitative comparison?

Or a comparison to the number of particles with diameters greater than 60 nm (as a proxy for CCN)?

10R. We were not able to retrieve such information from most measurement sites, nor it was possible to retrieve enough data points from the data. However, we also think that N100 is close to CCN at 0.2% supersaturation (see Kerminen et al., 2012 in manuscript references).

11. Was Figure 3 discussed anywhere in the main text?

11R. We corrected the manuscript by adding the reference of Figure 3 in section 3.1.