REFEREE 1

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 $\exp\left(\left|\log\left(\frac{model}{observation}\right)\right|\right)$ *. 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 SO_2 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 SO_2 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 SO_2 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 SO_4, while the accumulation mode emissions include SO_4 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 SO_4 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.*

REFEREE 2

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 PM2.5 concentrations from AeroCom simulation, the total PM2.5 concentrations from GAINS simulation, and the ratio between GAINS PM2.5 and AeroCom PM2.5.*

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 SO_2-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 ~1000-2000* 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 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 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)* m^{-1} 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.*

ADVANCING GLOBAL AEROSOL SIMULATIONS WITH SIZE-SEGREGATED ANTHROPOGENIC PARTICLE NUMBER EMISSIONS

FILIPPO XAUSA[1], PAULI PAASONEN[1,5], RISTO MAKKONEN[1], MIKHAIL ARSHINOV[2], AIJUN DING[3], HUGO DENIER VAN DER GON[4], VELI-MATTI KERMINEN[1], MARKKU KULMALA[1]

[1] *Division of Atmospheric Sciences, Department of Physics, University of Helsinki.*

[2] *Institute of Atmospheric Optics, SB RAS, 634055, Tomsk, Russia.*

[3] *Joint International Research Laboratory of Atmospheric and Earth System Sciences, School of Atmospheric Sciences, Nanjing University, Nanjing 210023, China.*

[4] *TNO, Department of Climate, Air and Sustainability, Utrecht, the Netherlands.*

[5] *International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria*

Keywords: AEROSOL, NUMBER SIZE DISTRIBUTION, GAINS, GLOBAL CLIMATE MODEL

ABSTRACT

Climate models are important tools that are used for generating climate change projections, in which aerosol-climate interactions are one of the main sources of uncertainties. In order to quantify aerosol-radiation and aerosol-cloud interactions, detailed input of anthropogenic aerosol number emissions is necessary. However, the anthropogenic aerosol number emissions are usually converted from the corresponding mass emissions in precompiled emission inventories through a very simplistic method depending uniquely on chemical composition, particle size and density, which are defined for a few very wide main source sectors. In this work, the anthropogenic particle number emissions converted from the AeroCom mass in the ECHAM-HAM

climate model were replaced with the recently-formulated number emissions from the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS)-model. In the GAINS model the emission number size distributions vary, for example, with respect to the fuel and technology. Special attention was paid to accumulation mode particles (particle diameter d_p > 100 nm) because of (i) their capability of acting as cloud condensation nuclei (CCN), thus forming cloud droplets and affecting Earth's radiation budget, and (ii) their dominant role in forming the coagulation sink and thus limiting the concentration of sub-100 nanometers particles. In addition, the estimates of anthropogenic CCN formation, and thus the forcing from aerosol-climate interactions are expected to be affected. Analysis of global particle number concentrations and size distributions reveal that GAINS implementation increases CCN concentration compared with AeroCom, with regional enhancement factors reaching values as high as 10. A comparison between modeled and observed concentrations shows that the increase in number concentration for accumulation mode particle agrees well with measurements, but it leads to a consistent underestimation of both nucleation mode and Aitken mode (d_p < 100 nm) particle number concentrations. This suggests that revisions are needed in the new particle formation and growth schemes currently applied in global modeling frameworks.

1 Introduction

In recent years, the link between anthropogenic aerosol particle and climate change has been a subject of several studies (e.g. Baker et al., 2015; Zhang et al., 2016). Anthropogenic aerosol particles play an important role in the global climate system via aerosol-radiation and aerosol-cloud interactions by scattering and absorbing solar radiation and by acting as cloud condensation or ice nuclei, thereby changing many cloud properties (Boucher et al., 2013). The global and regional radiative effects of aerosol particles depend on the spatial and temporal distribution of the aerosol number size distribution and chemical composition (Lohmann and Feichter, 2005; Schulz et al., 2006; Forster et al., 2007; Stier et al., 2007).

While anthropogenic primary emissions introduce cloud condensation nuclei (CCN) directly into the atmosphere, a significant fraction of the global CCN population is likely be formed through condensation of organic and other low-

volatility vapors onto ultra-fine particles (particle diameter d_p < 100 nm) in the atmosphere (Spracklen et al., 2008; Merikanto et al., 2009; Kerminen et al., 2012; Paasonen et al., 2013). Aerosol particles and their precursor vapors are emitted from both biogenic and anthropogenic sources, in addition to which they may also result from interactions between biogenic and anthropogenic emissions (Spracklen et al., 2011; Shilling et al., 2013). The increasing number concentration of accumulation mode particles decreases the formation and growth of smaller particles by increasing the sink for condensing vapor molecules, termed the condensation sink (CS, Kulmala et al., 2001), and by increasing the coagulation sink for small freshly-formed particles. Hence, the number concentration of accumulation mode particles from primary emissions affects secondary aerosol formation. The effects of these physical processes on future aerosol climate forcing requires application of detailed aerosol microphysical schemes in global climate models. Furthermore, the global uncertainty in CCN is highly sensitive to the assumed emission size distribution (Lee et al., 2013).

The global aerosol climate model ECHAM-HAM (Stier et al., 2005; Zhang et al., 2012) is a useful tool that aims at increasing our understanding of aerosol-climate interactions. Past simulations performed with the ECHAM-HAM include an extensive analysis of particle nucleation (Makkonen et al., 2009, 2014; Kazil et al., 2010), aerosol properties (Roelofs et al., 2010), and emission data set implementation (Zhang et al., 2012). Although the ECHAM-HAM has a detailed microphysics module for describing the aerosol size distribution (Vignati et al., 2004), previous studies have not included an exhaustive module for the model-input particle number size distribution. Also in other climate models, the mass-only aerosol input is a commonly applied setting (Jones et al., 2007; Shindell et al., 2007). The main reason behind this resides in the structure of the input data rather than in the models themselves.

One of the input emission inventories that has been widely used in ECHAM-HAM simulations, as well as in other Earth System Models (Pozzoli et al., 2011; Makkonen et al., 2009, 2012; Tonttila et al., 2015), is the Aerosol Inter Comparison data set, AeroCom (Dentener et al., 2006), developed for the purpose of conducting improved simulations of aerosol-climate interactions (Samset et al., 2014). However, the AeroCom emission inventory does not include a specific framework for particle number emissions. Hence, the input particle number emissions used in the simulations with AeroCom are

estimated from the particle mass emissions by the ECHAM-HAM during the initialization routine. In more detail, the estimation of number emissions consists of a simplistic multiplication of the given AeroCom mass emissions by a mass-to-number conversion factor. Each conversion factor that is applied for building the log-normal distribution is calculated by assuming that the mass emissions for each main source sector are distributed to predefined modes according to predefined densities, geometric mean radii and standard deviations, as described by Vignati et al., (2004) and Stier et al., (2005). This simplistic mass-to-number conversion factor does not represent the relationship between the particle mass and number size distributions in a realistic way, because such framework does not take into account the variation of emitted particle number size distributions from different emitting sources. The AeroCom inventory includes anthropogenic activities, from which the mass-to-number converted emissions are split into half between the Aitken and accumulation modes, and finally converted into log-normal modes. However, the recently-developed inventories allow for global aerosol simulations with a more detailed aerosol emission size distribution (Paasonen et al., 2016) with the GAINS emission scenario model (Greenhouse gas – Air pollution INteractions and Synergies; Cofala et al., 2009; Amann et al., 2011). GAINS data are organized into more detailed anthropogenic sources than AeroCom, with different particle number emissions and size distributions related to different fuels and technologies.

In this work, we first develop a novel module for anthropogenic particle number emissions in Earth System Models. Our experiment, performed with ECHAM-HAM, consists of replacing the mass-to-number converted anthropogenic AeroCom aerosol emissions with number emissions from the GAINS-model. In more detail, the implementation of GAINS data set is performed by using ECHAM-HAM default assumptions for AeroCom data set implementation. This study has a dual target: first, it aims at improving the ECHAM-HAM capability for estimating particle number concentrations, with a special focus on accumulation mode particles, and second, it investigates the feasibility of using the GAINS model for global climate modeling studies by running the ECHAM-HAM with both AeroCom and GAINS data sets. We present a comparison between the novel GAINS implementation and the default implementation of AeroCom in ECHAM-HAM, including modeled particle number concentrations and size distributions, as well as modeled CCN number concentrations. Finally, we compare the modeled number size distributions with observations in different environments around the world.

4

2 Materials and methods

2.1 The ECHAM5.5-HAM2 climate model

We used the global aerosol climate model ECHAM5.5-HAM2 (Stier et al., 2005; Zhang et al., 2012) with the M7 microphysics module (Vignati et al., 2004). The M7 describes the aerosol number size distribution with seven log-normal modes, in which the Aitken, accumulation and coarse modes are present in both the soluble and insoluble phases, while the nucleation mode is present only as the soluble mode. The compounds modeled in our simulations are black carbon (BC), organic carbon (OC), sulfate (SO_4), dust and sea salt. The emission module used in ECHAM-HAM reads data for anthropogenic, biogenic, wildfire, volcanic, agricultural emissions, secondary organic aerosols (SOA) and shipping sources. In our experiments, we modified only the part of the ECHAM-HAM source code that handles the anthropogenic emissions. The model has a horizontal gaussian grid (192×96) with a grid box size of ~200×200 km at the equator, and a vertical resolution of 31 hybrid sigma layers.

2.1.1 Aerosol microphysics

The version of ECHAM-HAM used in this work includes nucleation, condensation and coagulation modules. Previous studies have shown that the implementation of an activation-type nucleation improves particle number concentration estimations in modeling (Spracklen et al., 2010; Makkonen et al., 2012). In our experiment, we coupled a binary sulphuric acid-water nucleation scheme (Vehkamäki et al., 2002) with an activation-nucleation scheme described by Paasonen et al., (2010, Eq. 10), in which the nucleation rate (J) is a function of the activation coefficient and sulphuric acid concentration, expressed as

$$J = 1.7\,x\,10^{-6}\,s^{-1} * \left[H_2SO_4\right] \quad . \qquad (1)$$

The settings of our simulations included a specific module for SOA formation. Here, we modeled the SOA formation with both kinetic condensation onto a Fuchs-corrected surface area (CS) and partitioning according to a preexisting organic mass (Riipinen et al., 2011; Jokinen et al., 2015). This SOA module

includes three biogenic volatile organic compound (BVOC) tracers: isoprene, endocyclic monoterpenes and other monoterpenes, each having monthly resolutions for emissions. We did not use any nucleation scheme for organic vapors, because the simple activation-type nucleation, while not accurate for individual sites, describes the nucleation in different environments reasonably well (Paasonen et al., 2010). 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. More details can be found in Makkonen et al. (2012).

2.1.2 Natural emissions

BVOC emissions were implemented using the MEGAN2 (Guenther et al., 2006) model. MEGAN2 estimates biogenic emissions for about 150 compounds from different ecosystems, paying a particular attention to monoterpenes. This framework takes into account several factors that influence BVOC emissions, including the leaf age, soil moisture and light environment. MEGAN2 was run offline and its output data were used for the ECHAM-HAM input initialization.

All non-anthropogenic emissions, such as volcanic emissions, dimethyl-sulfide (DMS, Kloster et al., 2006) emitted by the sea and dust, were taken from AeroCom in both simulations. All emission data, excluding SOA precursors, DMS emissions and wildfire, were input as annual-averages. As a result, the seasonality in concentrations of anthropogenic compounds is mostly due to the nudged meteorology.

2.1.3 Anthropogenic emissions

The first simulation was performed with the ECHAM-HAM default implementation of anthropogenic emissions from AeroCom data set for year 2000. The AeroCom emissions taken by the ECHAM-HAM are provided by mass as kg m^{-2} s^{-1} with a chemical differentiation that includes BC, OC and SO_4, and a bi-level vertical distribution (2-zL) that consists of two surface layers: a lower level below 100 meters above the sea level for emissions from transportation and domestic combustion, and a higher level for industrial activities whose emissions reach altitudes higher than 100 meters.

6

While BC does not require preprocessing during the simulation, input emissions of OC and SO_4 undergo a further conversion during the initialization routine: OC mass is converted into primary organic matter (POM) mass with a multiplying factor 1.4 (Turpin et al., 2000; Kupiainen and Klimont, 2007), and emissions containing sulfur (S) are input as both sulfur dioxide (SO_2) and SO_4. The primary SO_4 particle fraction is estimated as 2.5% of gaseous SO_2, as described by Dentener et al. (2006). The masses of BC and POM are uniquely treated as Aitken mode particles (d_p = 10-100 nm). The mass of SO_4 is divided between the Aitken mode, accumulation mode (d_p = 100-1000 nm) and coarse mode (d_p > 1 μm) through a rough estimation: the lower-surface-level SO_4 is split equally between the Aitken mode and accumulation mode, whereas the higher-surface-level SO_4 is split equally between the accumulation mode and coarse mode. The mass is then converted by the model into a particle number size distribution. The mass-to-number flux factors, expressed as *m2n* in Figure 1, are embedded in the emission-reading routine. The number of particles is calculated through the generic function

$$N = M/m \quad , \tag{2}$$

where M is the mass of given emissions and m is the average mass estimated for a single particle. The particle mass m in Eq. (2) is extended in the model according to the Hatch-Choate conversion equations (Hinds, 1982), in which the density, count median radius and standard deviation are predefined for each chemical compound and size mode, as described by Stier et al. (2005). The emission count median radius is fixed at 30 nm and 75 nm for the Aitken mode and accumulation mode, respectively, and the standard deviation is set to 1.59 for all the modes except the coarse mode for which it is 2.0. The species density is set to 1841 $kg\ m^{-3}$ for SO_4 (input in the model as H_2SO_4) and 2000 $kg\ m^{-3}$ for BC and OC. Altogether, these parameters differentiate the species according to their chemistry and solubility. The number flux conversion is therefore expressed as

$$N = \frac{M}{\frac{4}{3} \cdot \pi \cdot \rho_i \cdot \left(\mathrm{cmr}_{jk} \cdot \mathrm{cmr}\ 2\ \mathrm{ram}_{jk}\right)^3} \quad , \tag{3}$$

where ρ is the density of a determined chemical compound i, and the expression in brackets is the mean radius of a particle with certain solubility j

7

and size mode *k.* The quantity cmr is the predefined count median radius as it is expressed in the model code, while cmr2ram is a conversion factor that multiplies cmr in order to estimate the radius of average mass. The cmr2ram factor depends uniquely on the standard deviation of the log-normal particle number distribution.

2.2 Emission scenario model GAINS

The GAINS (Greenhouse gas – Air pollution Interactions and Synergies) model is an integrated assessment model developed at IIASA (International Institute for Applied Systems Analysis) in Laxenburg, Austria (Amann et al, 2011). In order to calculate the emissions related to specific anthropogenic source sectors, it combines the information of the annual level of the anthropogenic activities, amounts of different fuels consumed for combustion activities, shares of different emission abatement technologies, and emission factors for different activity-fuel-technology-combinations.

The GAINS scenarios include information on the annual activity levels and shares of emission control technologies for nearly 170 regions, being countries or parts or groups of countries, in five-year intervals from 1990 to 2050. The activity levels are based on national and international statistics, latter available from International Energy Agency (IEA), Organisation for Economic Co-operation and Development (OECD), United Nations (UN) and Food and Agriculture Organization of the United Nations (FAO) and Eurostat, and the shares of control technologies are derived from national and international information on the related legislation, discussion with national experts and scientific publications. The emission factors for all combinations of source sectors, fuels and technologies are determined from the scientific publications or measurement databases. For detailed description of sources and methods to derive underlying particulate matter emissions see Klimont et al. (2016).

The particle number emission factors with the related number size distributions were recently implemented to GAINS (Paasonen et al., 2016). This implementation allowed for detailed assessment of particle number emissions with more than 1000 measures controlling emissions in each of the close to 170 regions, and in internally consistent manner with emissions of other air pollutants and greenhouse gases. The GAINS particle number

emissions are known to be subject to uncertainties, especially in terms of nucleation mode emissions, but the major particle number sources, such as road transport and residential combustion, are reasonably well represented down to the control technology level. The determination of emission factors for particle number emissions and particle size distributions is based on the European particle number emission inventory developed by TNO (Denier van der Gon et al., 2009, 2010).

In this study, we applied the gridded particle number 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 and their brief characterization is freely available from the IIASA website:

http://www.iiasa.ac.at/web/home/research/researchPrograms/air/PN.html.

2.3 GAINS implementation in M7

In the second simulation, the sub-module that converts the input mass to the number flux described in Eqs. (2-3) was switched off and we implemented the recently-developed 2010 GAINS anthropogenic emissions (Paasonen et al., 2016; see also section 2.1.2). The emission sectors considered for our experiment included the energy production, flares, industrial combustion and processes, transportation, waste combustion and domestic/commercial combustion. A detailed description of the sectors and emission factors is presented in Paasonen et al. (2016).

The number size distribution data provided by GAINS are organized into nine size bins with a geometric diameter ranging from 3 nm to 1000 nm. However, in this study we implemented the GAINS data for the Aitken mode and accumulation mode only (d_p = 10-1000 nm), so that the particle number implementation was consistent with the AeroCom simulation which lacked the nucleation mode conversion factor in the source code aerosol module. 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). It should be noted that the ratio of Aitken to accumulation mode emissions can vary between

9

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). This choice of implementation does not fully exploit all the information available in the GAINS size distribution, because the default ECHAM-HAM emission module does not allow the emission diameter to vary on a per-gridbox basis. Although it would be possible to upgrade the ECHAM-HAM in this sense, it would be quite laborious and beyond the scope of our study. 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).

In the GAINS simulation we kept the AeroCom data for the gas phase sulfur and coarse SO_4 in order to identify the global impact of GAINS implementation on submicron particles. Furthermore, we used the same bi-level 2-zL scheme as for the SO_4 vertical distribution in AeroCom: emissions from the transportation, agriculture fires, waste combustion and domestic combustion were put into the lower level (<100 m a.s.l.), whereas the energy, flares, industry and power plant sectors of GAINS were implemented into the higher level (>100 m a.s.l.).

GAINS provides the number emission data without chemical speciation and vertical distribution (see Table 1), and separately mass emissions of particle mass, particulate OC and BC, as well as gaseous pollutants, including SO_2. However, distributing the different compounds between the different number sizes bins is non-trivial task which requires, in order to be properly completed, elaboration of the proper GAINS model, not only the implementation. For this reason, we decided to use the default ECHAM-HAM particle composition from AeroCom in this study and leave the implementation of GAINS chemical composition for future studies. We followed a series of steps in order to partition the GAINS raw data into BC, POM and SO_4 in a consistent format for the model. Table 1 and Figure 1 visually illustrate the implementation framework. In more detail, we (I) off-

line converted AeroCom mass into number using ECHAM-HAM factors, (II) estimated the chemical species fraction among the respective Aitken mode and accumulation mode in AeroCom numbers, (III) applied such fractions to the total Aitken mode and accumulation mode particle numbers in the GAINS to have the correspondent BC, OC and SO_4 repartition, and finally, IV) used the mass-to-number factors used in (I) to estimate the speciated GAINS mass.

Shipping emissions are embedded in the AeroCom data set, but not included in GAINS. In our experiment, we masked out the AeroCom shipping emissions with a land-sea mask produced by applying Climate Data Operator (CDO) to the AeroCom. Hence, shipping emissions were not taken into consideration.

2.4 Simulation setup

Our experiment consisted of two one-year simulations, using identical model settings but different data set for anthropogenic sources: AeroCom and GAINS (see Sect. 2.3). The experiment run was set to start indicatively on October 1, 2009 and end on December 31, 2010 with a three-month spin-up period and one-hour time resolution for the output. The modeled data for our analysis were collected from January 1, 2010 to December 31, 2010. The model was nudged against 2010 ECMWF ERA-Interim (Berrisford et al., 2011) observed meteorology data in order to reduce noise in model estimations and to increase the statistical significance of the eventual anthropogenic aerosol perturbation signal (Kooperman et al., 2012).

2.5 Comparison with observation

Our study focused on particle number concentration and size distributions along with CCN concentrations at the supersaturations of 0.2% (CCN0.2) and 1.0% (CCN1.0). We compared the modeled particle number concentrations and size distributions against observations collected from 11 sites around the world. A detailed description of the observation data is illustrated in Table 2. The modeled data extracted from all sites were averaged over the year and plotted against observations to investigate the overall model performance. In addition to visual comparison between the modeled and observed concentrations, we calculated the relative bias as

$$\exp\left(\left|\log\left(\frac{model}{observation}\right)\right|\right) \quad , \qquad (4)$$

This relative bias returns the factor, larger than 1, with which the model under or over predicts the observation.

The particle number concentration and mean particle radius of the whole output data were used for plotting the number distributions from 6 of the 11 original sites, which were chosen to represent areas with a strong presence of anthropogenic emissions (Nanjing, Sao Paulo and Tomsk) as well as areas dominated by biogenic emissions (Hyytiälä, K-Puszta and Värriö). In both annual-average and number distribution comparisons, the modeled layer closest to Earth's surface was chosen for analysis. Modeled CCN concentrations were studied by comparing simulations with AeroCom emissions against those from GAINS emissions for both CCN0.2 and CCN1.0. CCN concentrations were extracted and averaged from the lowest three model layers in order to reduce background noise in mapping the global concentrations. Due to the coarse grid size and inhomogeneous sources around measurement sites, the evaluation against observations is not expected to yield one-to-one validation of aerosol concentrations (Schutgens et al., 2016).

3 Results and discussion

Here we show the comparison between AeroCom and GAINS implementation before (emissions, section 3.1) and after (atmospheric concentrations, sections 3.2 and 3.3) running the ECHAM-HAM model. Our experiment was performed with the same model settings in both simulations and it was nudged against meteorology data. As a result, our analysis focused merely on the differences between the particle number emissions of the two data sets and their different effects on modeled particle concentrations. In the following sections, we will first show the difference between AeroCom and GAINS in terms of input emissions, after which we will compare the model-simulated particle number concentrations and size distributions with observational data. Finally, we will assess the effect of the GAINS implementation on global CCN concentrations.

3.1 Differences in particle number emissions

In this section, we present a preliminary assessment of input emissions to illustrate the main differences between the two gridded data sets before starting the simulation. Table 3 shows global emissions and their ratios between GAINS and AeroCom for the whole domain. When the emissions were globally averaged (R_{tot}), GAINS showed higher total number emissions by a factor of 2.2. However, when looking at individual grid cells, the total particle number emission ratios between Aerocom and GAINS had a large spatial variability (Figure 2), even though the median value of this ratio was very close to one (see R_{grid} in Table 3). Figure 3 shows the spatial distribution of both emissions data sets. Globally, the Aitken to accumulation mode particle emission ratio was about two orders of magnitude in AeroCom emissions, while being less than a factor four in GAINS emission. The averaged emission ratios demonstrate that accumulation mode emissions play a critical role in the GAINS implementation, with both R_{tot} and R_{grid} ratios increasing dramatically compared with AeroCom. The averaged Aitken mode particle emissions from GAINS did not show a similar increase, and the R_{grid} median value was even lower than that in the AeroCom emissions. The R_{tot} and R_{grid} ratios of Aitken mode emissions were 1.7 and 0.7, respectively. This difference shows that the Aitken mode particle emissions are quantitatively higher in GAINS than in AeroCom when their geographical distribution differences are not taken into account. However, when the data sets were compared by confronting each grid cell one by one, AeroCom emissions were higher than GAINS emissions in a prevalent area of the global domain.

It should be noted that in the ECHAM-HAM assumptions made for the AeroCom emissions, fossil fuel and biofuel emissions are implemented in the 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 into particle size bins at around 100 nm (Paasonen et al., 2016). 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.

The differences in Aitken and accumulation mode emissions between GAINS and AeroCom implementations originate from three main differences

13

between the emission data bases. Firstly, the GAINS emission factors, especially in traffic and residential combustion sectors, are directly based on literature or databases of particle number emissions, whereas in AeroCom the number emissions are converted from mass emissions. This causes differences in the relative shares of different source sectors in the emission size distributions. Secondly, the original emission size distributions in GAINS contains from one to three different modes, whereas in AeroCom the emissions are represented with only one mode. In many GAINS sources, e.g. road transport, the mode with a larger mean emission diameter contributes significantly to the emission of particles with d_P > 100 nm, even though the total number emission is clearly dominated by a mode with a smaller mean diameter. Finally, as stated earlier, the GAINS emission size distributions are different for different technologies and fuels, in diesel powered road transport also for different fuel sulfur contents. This increases the regional variability of the emissions.

3.2 Simulated particle number concentrations and size distributions

Here we present the core of our analysis, which includes an assessment of the modeled particle number concentrations against observations. Figure 4 shows the annual-averaged modeled particle concentration in comparison with observations from eleven sites. Overall, both emission data sets showed a tendency to underestimate particle number concentrations in model simulations, especially for the locations with high observed particle number concentrations. The underestimation of the highest particle concentrations might be, at least partly, related to the spatial resolution of ECHAM-HAM, due to which the typically high particle concentrations near urban or industrial areas will be distributed evenly into a large model grid cell (Stier et al., 2005). A comparison of the model results with the observational data shows that the GAINS implementation significantly improved the reproduction of observed concentrations in accumulation mode (d_p > 100 nm), being closer to observations than AeroCom at all 11 sites. For the Aitken mode (d_p = 10-100 nm), similar improvement was not reached, as the observed concentrations were better reproduced with AeroCom than with GAINS at 8 sites. The average relative bias described in Eq. (4) 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

were 2.25 and 2.12 with GAINS and AeroCom emissions, respectively. 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.

Figure 5 shows the modeled particle number size distributions against observations at 6 measurement sites. The size distributions modeled with the GAINS emissions agreed relatively well with the measurements for the accumulation mode, whereas the nucleation and Aitken modes were underestimated in simulations with both emission data sets. GAINS underestimated the Aitken mode particle concentrations more heavily than AeroCom, by a factor of two to three in Hyytiälä, Värriö and Kpuszta, suggesting that the higher condensation sink associated with higher accumulation mode particle emissions in GAINS had a significant impact on modeled ultra-fine particle number concentrations. In addition, Hyytiälä and Värriö are regions in which BVOC emissions and clean air are the key influencing factors for new particle formation and particle growth (Ruuskanen et al., 2007; Corrigan et al., 2013; Liao et al., 2014). This was reflected in the model results: particle number size distributions in Hyytiälä and Värriö were quite similar between the two simulations based on different anthropogenic emission data sets. Contrary to this, Nanjing, Sao Paulo and Tomsk are areas with strong influences by anthropogenic emissions, so that in comparison with AeroCom, the simulations with GAINS emissions produced higher accumulation mode and Aitken mode particle number concentrations as well as better agreements with the observations in these regions. Nevertheless, the model was not able to reach the observed ultra-fine particle concentration in either simulation in most areas, and the higher CS in GAINS significantly reduced particle number concentrations of the smallest particles in most regions. Some areas showed a dramatic reduction in simulated ultra-fine particle number concentrations e.g. in Nanjing the whole modeled nucleation mode was wiped out when using the GAINS emissions.

The above results suggest that in ECHAM-HAM, as well as probably in other climate models, the current nucleation and growth schemes may need further revisions. However, it is also likely that the anthropogenic emissions

15

of especially nucleation mode particles in GAINS are still severely underestimated for many source sectors (Paasonen et al., 2016). This is because many of the measurements, on which the GAINS emission factors are based, are not sensitive to non-solid nucleation mode particles, such as those formed via nucleation of sulfur or organic vapors immediately after the combustion or at small downwind distances in plumes from different combustion sources (Stevens and Pierce, 2013). It should also be noted that our study does not include any sensitivity analysis based on the primary sulfate emissions parameterization (Luo and Yu, 2011). In addition, the lower modeled Aitken mode particle concentrations from GAINS emissions may, in some parts of the global domain, be also related to possible overestimations in the accumulation mode particle emissions in the GAINS model, which are consequently affecting the formation and growth of smaller particles. Nonetheless, all the model versus observation comparisons between the simulations clearly represent a consistent challenge for climate models in modeling ultra-fine particle number size distributions.

Figure 6 shows absolute annual-average particle concentrations for the accumulation mode and Aitken mode with both AeroCom and GAINS emissions. While the regional distributions had similar patterns in both simulations, there were evident differences when looking at the two size modes. Accumulation mode particle concentrations were higher for the simulation with the GAINS emission in most regions, which is consistent with the input emissions assessment. The differences were particularly evident over the developing areas where anthropogenic activities represent the main source of atmospheric particles, especially in South America, central Africa, India, China and south-east Asia. As observed in Figure 5, the high accumulation mode particle number concentrations in the simulation with the GAINS emission has a critical effect on Aitken mode particle concentrations at most sites. A peculiar pattern is observed in China where the dominant presence of anthropogenic sources from GAINS led the model to predict high concentrations of ultra-fine particles. The decrease in GAINS-derived Aitken mode particle number concentrations in areas where emissions were actually higher than the AeroCom emission implies that Aitken mode particles had been removed, or their secondary production was hindered, by the prominent increase of the CS caused by a higher number of emitted accumulation mode particles.

16

3.3 Concentrations and sources of CCN

This section presents the impact of particle emission data on atmospheric CCN concentrations on annual and seasonal perspectives. It is important to note that the applied anthropogenic number emissions did not have a seasonal variation, so the seasonal differences are entirely due to the variation of other emissions, and mainly to the strong temperature dependence of biogenic SOA formation affecting the CCN concentration (Paasonen et al., 2013). Our results showed clear differences in the simulated CCN concentrations between the two primary emission data sets, and these differences depended strongly on the considered supersaturation (Figure 7 and 8).

At the 0.2% supersaturation, the CCN concentrations were higher with the GAINS emissions compared with the AeroCom emissions in practically all the regions and during all seasons (Figure 8). The annual-average CCN0.2 concentration ratio between the GAINS and Aerocom was two to three in most areas, with peaks of four to ten in south America, central Africa and east Asia (Figure 7). However, relatively high accumulation mode particle concentrations were observed in India, China and south-east Asia (see Figure 6), and also an increase in absolute CCN0.2 concentration due to anthropogenic emissions was observed in eastern China and south-east Asia. Our analysis of the seasonality revealed that the difference between GAINS and AeroCom simulations in terms of CCN0.2 concentrations was the largest during the cold season in January, with boreal and arctic regions showing an increment of GAINS/AeroCom CCN0.2 ratio up to a factor of seven to ten. The southern hemisphere also displayed notable differences in both South America and South-East Asia, with GAINS/AeroCom CCN0.2 ratios of three to ten during the warmest season.

At the supersaturation of 1.0%, a significant fraction of Aitken mode particles is capable of acting as CCN. Opposite to the CCN0.2 concentrations, the simulated CCN1.0 concentrations with the GAINS emissions were lower than with AeroCom emissions, with a GAINS/AeroCom ratio between 0.5 and 1 in most regions (Figure 7). Our seasonality analysis showed that the simulation with the GAINS data set produced higher CCN1.0 concentrations than AeroCom in Europe, India and East Asia during the winter. However, such ratio was equal to one or below in most regions, except eastern Asia, during the warmer seasons. The substantially lower CCN1.0 concentrations with

17

GAINS emissions arise from the relatively similar Aitken mode number emissions between GAINS and AeroCom, but significantly larger CS from GAINS, causing a decrease in secondary ultrafine particle formation. However, in China and South-East Asia, the annual CCN1.0 concentration from GAINS was higher than from AeroCom by at least a factor of two, suggesting that these regions may play a key role in contributing for the global anthropogenic emissions and increment of CCN.

It is important to remark that the substantial differences in CCN concentrations illustrated above are linked to the implementation of different data sets, and therefore the modeled estimations might be affected by uncertainties of the GAINS model as well. Furthermore, it may be questioned whether the ECHAM-HAM is actually able to estimate CCN concentrations with GAINS better than with AeroCom. This goes beyond the fundamental goal of this study, which is to address the feasibility of using GAINS emissions in global climate modeling. However, the modeled GAINS accumulation mode particle number concentrations agree with observation significantly better than AeroCom. This, based on the sensitivity analysis by Lee et al. (2013), suggests that the GAINS implementation is likely to estimate CCN concentrations better than AeroCom. In any case, further studies are needed to address the contribution of the GAINS model in improving modeled CCN concentration. Furthermore, it would be beneficial to investigate how the applied nucleation scheme, combined with the GAINS anthropogenic emissions, affects the estimation of CCN concentration to better identify the driving forces behind the uncertainties of modeling particle number size distributions with the global climate models.

4 Conclusions

The outcome of our experiment shows that the most significant differences between the GAINS and AeroCom emissions data sets are (i) the particle size distribution in the Aitken mode and accumulation mode, and (ii) the geographical distribution of the particle number emissions over the global domain. The accumulation mode particle emissions from GAINS are significantly higher than AeroCom, by factors from 10 to 1000, thus potentially resulting in dramatic increases in climatically active primary particles and simultaneous decreases in secondary ultrafine particle formation due to higher values of CS and coagulation sink.

18

In comparison to AeroCom emissions, GAINS emissions produced much higher accumulation mode particle concentrations, but the consequently higher CS and coagulation sink led to lower Aitken mode concentrations with GAINS emissions than with AeroCom emissions. In comparison to observation data at eleven measurement sites, the modeled annual-averaged concentrations with GAINS emissions performed better than with AeroCom emissions, in terms of bringing the modeled accumulation mode particle concentrations closer to observation at all eleven sites, and Aitken mode particle concentrations closer to observation at three sites. However, a higher underestimation was observed in the simulation with GAINS emissions for particles with d_p < 30 nm.

The underestimation of d_p < 30 nm particle concentrations in the simulation with GAINS emissions highlighted the sensitivity of nucleation mode and Aitken mode particle concentrations to CS and coagulation sink. This underestimation is presumably partly caused by underestimations in emissions of non-solid nucleation/Aitken mode particles in the GAINS model (Paasonen et al., 2016). As a next step, the modules for nucleation and subsequent growth and the sensitivity of the concentrations of sulfuric acid (the main precursor in the applied nucleation parameterization) to altered CS should be revisited.

It is important to note that the simulations performed in this study did not implement an up-to-date secondary organic aerosols (ELVOCS) nucleation scheme, nor a seasonal cycle of anthropogenic emissions, which may represent a further step to reduce the gap between the modeled and observed concentrations. Finally, given the high spatial variability of global emissions, more observation data and the establishment of new measurement stations in varying environments are urgently needed to better evaluate the model results.

Acknowledgements

This project was funded by the MAJ JA TOR NESSLING grant n. 201600369 and the Academy of Finland Center of Excellence (FCoE) grant n. 307331.

Particle number size distributions at Melpitz were provided by Wolfram Birmili, Kay Weinhold, André Sonntag, Birgit Wehner, Thomas Tuch, and

Alfred Wiedensohler (Leibniz Institute for Tropospheric Research, Leipzig, Germany).

Particle number size distributions at Hohenpeissenberg were provided by Harald Flentje and Björn Briel (German Weather Service, Hohenpeissenberg, Germany). Both measurements were supported by the German Federal Environment Ministry (BMU) grant UFOPLAN 370343200, project duration 2008-2010. Both data sets can be publicly accessed through the German Ultrafine Aerosol Network (GUAN) at https://doi.org/10.5072/guan.

Particle number size distributions at Botsalano were provided by Ville Vakkari and Lauri Laakso (Finnish Meteorological Institute, Helsinki, Finland).

Particle number size distributions at Sao Paulo were provided by John Backman (Finnish Meteorological Institute, Helsinki, Finland).

Particle number size distributions at San Pietro Capofiume (Po Valley) were provided by Ari Laaksonen (Finnish Meteorological Institute, Helsinki, Finland).

We thank Chris Heyes and Zbigniew Klimont from the Air Quality and Greenhouse Gases program at IIASA, and Kaarle Kupiainen from IIASA and Finnish Environment Institute (SYKE) for their help and communication.

The EU FP7 BACCHUS project (grant n. 603445) and the Nordic Center of Excellence eSTICC (Nordforsk grant n. 57001) are acknowledged for financial support.

REFERENCES

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F. and Winiwarter, W.: Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, Environ. Model. Softw., 26(2), 1489–1501, 2011.

Backman, J., Rizzo, L. V., Hakala, J., Nieminen, T., Manninen, H. E., Morais, F., Aalto, P. P., Siivola, E., Carbone, S., Hillamo, R., Artaxo, P., Virkkula, A., Petäjä, T., and Kulmala, M.: On the diurnal cycle of urban aerosols, black carbon and the occurrence of new particle formation events in springtime São Paulo, Brazil, Atmos. Chem. Phys., 12, 11733-11751, doi:10.5194/acp-12-11733-2012, 2012.

Baker, L. H., Collins, W. J., Olivié, D. J. L., Cherian, R., Hodnebrog, Ø., Myhre, G., and Quaas, J.: Climate responses to anthropogenic emissions of short-lived climate pollutants, Atmos. Chem. Phys., 15, 8201-8216, doi:10.5194/acp-15-8201-2015, 2015.

Berrisford, P., Kållberg, P., Kobayashi, S., Dee, D., Uppala, S., Simmons, A. J., Poli, P. and Sato, H.: Atmospheric conservation properties in ERA-Interim. Quarterly Journal of the Royal Meteorological Society 137:1381-1399, 2011.

Birmili, W., Weinhold, K., Rasch, F., Sonntag, A., Sun, J., Merkel, M., Wiedensohler, A., Bastian, S., Schladitz, A., Löschau, G., Cyrys, J., Pitz, M., Gu, J., Kusch, T., Flentje, H., Quass, U., Kaminski, H., Kuhlbusch, T. A. J., Meinhardt, F., Schwerin, A., Bath, O., Ries, L., Gerwig, H., Wirtz, K., and Fiebig, M.: Long-term observations of tropospheric particle number size distributions and equivalent black carbon mass concentrations in the German Ultrafine Aerosol Network (GUAN), Earth Syst. Sci. Data, 8, 355-382, doi:10.5194/essd-8-355-2016, 2016.

Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., Rasch, P., Satheesh, S.K., Sherwood, S., Stevens, B. and Zhang, X.Y.: Clouds and aerosols. In Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. T.F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Doschung, A. Nauels, Y. Xia, V. Bex, and P.M. Midgley, Eds. Cambridge University Press, 571-657, doi:10.1017/CBO9781107415324.016, 2013.

Corrigan, A. L., Russell, L. M., Takahama, S., Äijälä, M., Ehn, M., Junninen, H., Rinne, J., Petäjä, T., Kulmala, M., Vogel, A. L., Hoffmann, T., Ebben, C. J., Geiger, F. M., Chhabra, P., Seinfeld, J. H., Worsnop, D. R., Song, W., Auld, J., and Williams, J.: Biogenic and biomass burning organic aerosol in a boreal forest at Hyytiälä, Finland, during HUMPPA-COPEC 2010, Atmos. Chem. Phys., 13, 12233-12256, doi:10.5194/acp-13-12233-2013, 2013.

Dal Maso M., Sogacheva L., Anisimov M. P., Arshinov M., Baklanov A., Belan B., Khodzher T. V., Obolkin V. A., Staroverova A., Vlasov A., Zagaynov V. A., Lushnikov A., Lyubovtseva Y. S., Riipinen I., Kerminen V.-M. and Kulmala M.: Aerosol particle formation events at two Siberian stations inside the boreal forest. Boreal Env. Res. 13, 81-92, 2008.

Denier van der Gon, H., Visschedijk, A., Johansson, C., Hedberg Larsson, E., Harrison, R. M., and Beddows, D.: Size-resolved Pan European Anthropogenic Particle Number Inventory, EUCAARI Deliverable 141, 2009.

Denier van der Gon, H., Visschedijk, A., Johansson, C., Ntziachristos, L., and Harrison, R. M.: Size-resolved Pan-European Anthropogenic Particle Number Inventory, paper presented at International Aerosol conference (oral), 29 August 3 September 2010, Helsinki, 2010.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J.-P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6, 4321-4344, doi:10.5194/acp-6-4321-2006, 2006.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D.W., Haywood, J., Lean, J., Lowe, D.C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., Van Dorland, R. and Miller, H.L.: *Changes in Atmospheric Constituents and in Radiative Forcing Chapter 2*. United Kingdom: Cambridge University Press, 2007.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, doi:10.5194/acp-6- 3181-2006, 2006.

Gultepe, I. Isaac, G.A.: Scale Effects on Averaging of Cloud Droplet and Aerosol Number Concentrations: Observations and Models. J. Climate 12:1268-1279, 1999.

Hamed, A., Joutsensaari, J., Mikkonen, S., Sogacheva, L., Dal Maso, M., Kulmala, M., Cavalli, F., Fuzzi, S., Facchini, M. C., Decesari, S., Mircea, M., Lehtinen, K. E. J., and Laaksonen, A.: Nucleation and growth of new particles in Po Valley, Italy, Atmos. Chem. Phys., 7, 355-376, doi:10.5194/acp-7-355-2007, 2007.

Hari P., Kulmala M., Pohja T., Lahti T., Siivola E., Palva L., Aalto P., Hämeri K., Vesala T., Luoma S. and Pulliainen E.: Air pollution in Eastern Lapland: challenge for an environmental measurement station. Silva Fennica 28: 29–39, 1994.

Hari, P. & Kulmala, M. Station for Measuring Ecosystem-Atmosphere Relations (SMEAR II). Boreal Env. Res., 10, 315–322, 2005.

Herrmann, E., Ding, A. J., Kerminen, V.-M., Petäjä, T., Yang, X. Q., Sun, J. N., Qi, X. M., Manninen, H., Hakala, J., Nieminen, T., Aalto, P. P., Kulmala, M., and Fu, C. B.: Aerosols and nucleation in eastern China: first insights from the new SORPES-NJU station, Atmos. Chem. Phys., 14, 2169-2183, doi:10.5194/acp-14-2169-2014, 2014.

Hinds, W. C. (1982) Aerosol Technology, p. 85. Wiley, New York.

IPCC, Climate Change: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp, doi:10.1017/CBO9781107415324, 2013.

Jones, A., Haywood, J. M. and Boucher, O.: Aerosol forcing, climate response and climate sensitivity in the Hadley Centre climate model, J. Geophys. Res., 112, D20211, doi:10.1029/2007JD008688, 2007.

Jokinen, T., Berndt, T., Makkonen, R., Kerminen, V., Junninen, H., Paasonen, P., Stratmann, F., Herrmann, H., Guenther, A.B., Worsnop, D.R., Kulmala, M., Ehn, M., Sipilä, M.: Production of extremely low volatile organic compounds from biogenic emissions: Measured yields and atmospheric implications, Proceedings of the National Academy of Sciences 112:7123-7128, 2015.

Kazil, J., Stier, P., Zhang, K., Quaas, J., Kinne, S., O'Donnell, D., Rast, S., Esch, M., Ferrachat, S., Lohmann, U., and Feichter, J.: Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 10, 10733-10752, doi:10.5194/acp-10-10733-2010, 2010.

Kerminen, V-M., Kulmala, M.: Analytical formulae connecting the “real” and the “apparent” nucleation rate and the nuclei number concentration for atmospheric nucleation events. J Aerosol Sci 33(4):609–622, 2002.

Kerminen, V.-M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi, E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala, M., and Petäjä, T.: Cloud condensation nuclei production associated with atmospheric nucleation: a synthesis based on existing literature and new results, Atmos. Chem. Phys., 12, 12037-12059, doi:10.5194/acp-12-12037-2012, 2012.

Kinne, S., Schulz, M., Textor, C., Guibert, S., Balkanski, Y., Bauer, S. E., Berntsen, T., Berglen, T. F., Boucher, O., Chin, M., Collins, W., Dentener, F., Diehl, T., Easter, R., Feichter, J., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Herzog, M., Horowitz, L., Isaksen, I., Iversen, T., Kirkevåg, A., Kloster, S., Koch, D., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Lesins, G., Liu, X., Lohmann, U., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, O., Stier, P., Takemura, T., and Tie, X.: An AeroCom initial assessment – optical properties in aerosol component modules of global models, Atmos. Chem. Phys., 6, 1815-1834, doi:10.5194/acp-6-1815-2006, 2006.

Kiss, G., Varga, B., Galambos, I., & Ganszky, I. Characterization of water-soluble organic matter isolated from atmospheric fine aerosol. J. Geophys. Res., 107, 8339–8347, doi:10.1029/2001JD000603, 2002.

Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J. and Schöpp, W.: Global anthropogenic emissions of particulate matter including black carbon, Atmospheric Chem. Phys. Discuss., 2016, 1–72, doi:10.5194/acp-2016-880, 2016.

Kloster, S., Feichter, J., Maier-Reimer, E., Six, K. D., Stier, P., and Wetzel, P.: DMS cycle in the marine ocean-atmosphere system – a global model study, Biogeosciences, 3, 29-51, doi:10.5194/bg-3-29-2006, 2006.

Kooperman, G. J., Pritchard, M. S., Ghan, S. J., Wang, M., Somerville, R. C. J., and Russell, L. M.: Constraining the influence of natural variability to improve estimates of global aerosol indirect effects in a nudged version of the Community Atmosphere Model 5, J. Geophys. Res., 117, D23204, doi:10.1029/2012JD018588, 2012.

Kulmala, M., Dal Maso, M., Mäkelä, J., Pirjola, L., Väkevä, M., Aalto, P., Miikkulainen, P., Hämeri, K., and O'dowd, C.: On the formation, growth and composition of nucleation mode particles. Tellus B, 53(4). doi:http://dx.doi.org/10.3402/tellusb.v53i4.16622, 2001.

Kupiainen, K. and Klimont, Z.: Primary emissions of fine carbonaceous particles in Europe. Atmospheric Environment 41:2156 – 2170, 2007.

Laakso, L., Laakso, H., Aalto, P. P., Keronen, P., Petäjä, T., Nieminen, T., Pohja, T., Siivola, E., Kulmala, M., Kgabi, N., Molefe, M., Mabaso, D., Phalatse, D., Pienaar, K., and Kerminen, V.-M.: Basic characteristics of atmospheric particles, trace gases and meteorology in a relatively clean Southern African Savannah environment, Atmos. Chem. Phys., 8, 4823-4839, doi:10.5194/acp-8-4823-2008, 2008.

Lee, L. A., Pringle, K. J., Reddington, C. L., Mann, G. W., Stier, P., Spracklen, D. V., Pierce, J. R., and Carslaw, K. S.: The magnitude and causes of uncertainty in global model simulations of cloud condensation nuclei, Atmos. Chem. Phys., 13, 8879-8914, doi:10.5194/acp-13-8879-2013, 2013.

Liao, L., Kerminen, V.-M., Boy, M., Kulmala, M., and Dal Maso, M.: Temperature influence on the natural aerosol budget over boreal forests, Atmos. Chem. Phys., 14, 8295-8308, doi:10.5194/acp-14-8295-2014, 2014.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5, 715-737, doi:10.5194/acp-5-715-2005, 2005.

Luo, G. and Yu, F.: Sensitivity of global cloud condensation nuclei concentrations to primary sulfate emission parameterizations, Atmos. Chem. Phys., 11, 1949-1959, https://doi.org/10.5194/acp-11-1949-2011, 2011.

Makkonen, R., Asmi, A., Korhonen, H., Kokkola, H., Järvenoja, S., Räisänen, P., Lehtinen, K. E. J., Laaksonen, A., Kerminen, V.-M., Järvinen, H., Lohmann, U., Bennartz, R., Feichter, J., and Kulmala, M.: Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model, Atmos. Chem. Phys., 9, 1747-1766, doi:10.5194/acp-9-1747-2009, 2009.

Makkonen, R., Asmi, A., Kerminen, V.-M., Boy, M., Arneth, A., Guenther, A., and Kulmala, M.: BVOC-aerosol-climate interactions in the global aerosol-climate model ECHAM5.5-HAM2, Atmos. Chem. Phys., 12, 10077-10096, doi:10.5194/acp-12-10077-2012, 2012.

Makkonen, R., Seland, Ø., Kirkevåg, A., Iversen, T., and Kristjánsson, J. E.: Evaluation of aerosol number concentrations in NorESM with improved nucleation parameterization, Atmos. Chem. Phys., 14, 5127-5152, doi:10.5194/acp-14-5127-2014, 2014.

Merikanto, J., Spracklen, D. V., Mann, G. W., Pickering, S. J., and Carslaw, K. S.: Impact of nucleation on global CCN, Atmos. Chem. Phys., 9, 8601-8616, doi:10.5194/acp-9-8601-2009, 2009.

Paasonen, P., Asmi, A, Petaja, T, Kajos, M.K, Aijala, M, Junninen, H, Holst, T, Abbatt, J.P.D, Arneth, A, Birmili, W, van der Gon, H.D, Hamed, A, Hoffer, A, Laakso, L, Laaksonen, A, Richard Leaitch, W, Plass-Dulmer, C, Pryor, S.C, Raisanen, P, Swietlicki, E, Wiedensohler, A, Worsnop, D.R, Kerminen, V Kulmala, M.: Warming-induced increase in aerosol number concentration likely to moderate climate change. Nature Geoscience 6:438-442, 2013.

Paasonen, P., Kupiainen, K., Klimont, Z., Visschedijk, A., Denier van der Gon, H. A. C., and Amann, M.: Continental anthropogenic primary particle number emissions, Atmos. Chem. Phys., 16, 6823-6840, doi:10.5194/acp-16-6823-2016, 2016.

Pozzoli, L., Janssens-Maenhout, G., Diehl, T., Bey, I., Schultz, M. G., Feichter, J., Vignati, E., and Dentener, F.: Re-analysis of tropospheric sulfate aerosol and ozone for the period 1980–2005 using the aerosol-chemistry-climate model ECHAM5-HAMMOZ, Atmos. Chem. Phys., 11, 9563-9594, doi:10.5194/acp-11-9563-2011, 2011.

Riipinen, I., Pierce, J. R., Yli-Juuti, T., Nieminen, T., Häkkinen, S., Ehn, M., Junninen, H., Lehtipalo, K., Petäjä, T., Slowik, J., Chang, R., Shantz, N. C., Abbatt, J., Leaitch, W. R., Kerminen, V.-M., Worsnop, D. R., Pandis, S. N., Donahue, N. M., and Kulmala, M.: Organic condensation: a vital link connecting aerosol formation to cloud condensation nuclei (CCN) concentrations, Atmos. Chem. Phys., 11, 3865-3878, doi:10.5194/acp-11-3865-2011, 2011.

Roelofs, G.-J., ten Brink, H., Kiendler-Scharr, A., de Leeuw, G., Mensah, A., Minikin, A., and Otjes, R.: Evaluation of simulated aerosol properties with the aerosol-climate model ECHAM5-HAM using observations from the IMPACT field campaign, Atmos. Chem. Phys., 10, 7709-7722, doi:10.5194/acp-10-7709-2010, 2010.

Ruuskanen, T. M., Kaasik, M., Aalto, P. P., Hõrrak, U., Vana, M., Mårtensson, M., Yoon, Y. J., Keronen, P., Mordas, G., Ceburnis, D., Nilsson, E. D., O'Dowd, C., Noppel, M., Alliksaar, T., Ivask, J., Sofiev, M., Prank, M., and Kulmala, M.: Concentrations and fluxes of aerosol particles during the LAPBIAT measurement campaign at Värriö field station, Atmos. Chem. Phys., 7, 3683-3700, doi:10.5194/acp-7-3683-2007, 2007.

Samset, B. H., Myhre, G., Herber, A., Kondo, Y., Li, S.-M., Moteki, N., Koike, M., Oshima, N., Schwarz, J. P., Balkanski, Y., Bauer, S. E., Bellouin, N., Berntsen, T. K., Bian, H., Chin, M., Diehl, T., Easter, R. C., Ghan, S. J., Iversen, T., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Penner, J. E., Schulz, M., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., and Zhang, K.: Modelled black carbon radiative forcing and atmospheric lifetime in AeroCom Phase II constrained by aircraft observations, Atmos. Chem. Phys., 14, 12465-12477, doi:10.5194/acp-14-12465-2014, 2014.

Schulz, M., Textor, C., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Dentener, F., Guibert, S., Isaksen, I. S. A., Iversen, T., Koch, D., Kirkevåg, A., Liu, X., Montanaro, V., Myhre, G., Penner, J. E., Pitari, G., Reddy, S., Seland, Ø., Stier, P., and Takemura, T.: Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations, Atmos. Chem. Phys., 6, 5225-5246, doi:10.5194/acp-6-5225-2006, 2006.

Schurgers, G., Arneth, A., Holzinger, R., and Goldstein, A. H.: Process-based modelling of biogenic monoterpene emissions combining production and release from storage, Atmos. Chem. Phys., 9, 3409–3423, doi:10.5194/acp-9-3409-2009, 2009.

Schutgens, N. A. J., Gryspeerdt, E., Weigum, N., Tsyro, S., Goto, D., Schulz, M., and Stier, P.: Will a perfect model agree with perfect observations? The impact of spatial sampling, Atmos. Chem. Phys., 16, 6335-6353, doi:10.5194/acp-16-6335-2016, 2016.

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley and Sons, 1998.

Shilling, J. E., Zaveri, R. A., Fast, J. D., Kleinman, L., Alexander, M. L., Canagaratna, M. R., Fortner, E., Hubbe, J. M., Jayne, J. T., Sedlacek, A., Setyan, A., Springston, S., Worsnop, D. R., and Zhang, Q.: Enhanced SOA formation from mixed anthropogenic and biogenic emissions during the CARES campaign, Atmos. Chem. Phys., 13, 2091-2113, doi:10.5194/acp-13-2091-2013, 2013.

Shindell, D. T., Faluvegi, G., Bauer, S. E., Koch, D. M., Unger, N., Menon, S., Miller, R. L., Schmidt, G. A. and Streets, D. G.: Climate response to projected changes in short-lived species under an A1B scenario from 2000–2050 in the GISS climate model, J. Geophys. Res., 112, D20103, doi:10.1029/2007JD008753, 2007.

Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V.-M., Mann, G. W., and Sihto, S.-L.: The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales, Atmos. Chem. Phys., 6, 5631-5648, doi:10.5194/acp-6-5631-2006, 2006.

Spracklen, D. V., Karslav, K. S., Kerminen, V-M., Sihto, S-L., Riipinen, I., Merikanto, J., Mann, G. W., Chipperfield, M. P., Wiedensohler, A., Birmili, W. and Lihavainen, H.: Contribution of particle formation to global cloud condensation nuclei concentrations, Geophys. Res. Lett., 35, L06808, doi:10.1029/2007GL033038.

Spracklen, D. V., Carslaw, K. S., Merikanto, J., Mann, G. W., Reddington, C. L., Pickering, S., Ogren, J. A., Andrews, E., Baltensperger, U., Weingartner, E., Boy, M., Kulmala, M., Laakso, L., Lihavainen, H., Kivekäs, N., Komppula, M., Mihalopoulos, N., Kouvarakis, G., Jennings, S. G., O'Dowd, C., Birmili, W., Wiedensohler, A., Weller, R., Gras, J., Laj, P., Sellegri, K., Bonn, B., Krejci, R., Laaksonen, A., Hamed, A., Minikin, A., Harrison, R. M., Talbot, R., and Sun, J.: Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation, Atmos. Chem. Phys., 10, 4775-4793, doi:10.5194/acp-10-4775-2010, 2010.

Spracklen, D. V., Jimenez, J. L., Carslaw, K. S., Worsnop, D. R., Evans, M. J., Mann, G. W., Zhang, Q., Canagaratna, M. R., Allan, J., Coe, H., McFiggans, G., Rap, A., and Forster, P.: Aerosol mass spectrometer constraint on the global secondary organic aerosol budget, Atmos. Chem. Phys., 11, 12109-12136, doi:10.5194/acp-11-12109-2011, 2011.

Stevens, R. G. and Pierce, J. R.: A parameterization of sub-grid particle formation in sulfur-rich plumes for global- and regional-scale models, Atmos. Chem. Phys., 13, 12117-12133, doi:10.5194/acp-13-12117-2013, 2013.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125-1156, doi:10.5194/acp-5-1125-2005, 2005.

Stier, P., Seinfeld, J. H., Kinne, S., and Boucher, O.: Aerosol absorption and radiative forcing, Atmos. Chem. Phys., 7, 5237-5261, doi:10.5194/acp-7-5237-2007, 2007.

Stohl, A., Aamaas, B., Amann, M., Baker, L. H., Bellouin, N., Berntsen, T. K., Boucher, O., Cherian, R., Collins, W., Daskalakis, N., Dusinska, M., Eckhardt, S., Fuglestvedt, J. S., Harju, M., Heyes, C., Hodnebrog, Ø., Hao, J., Im, U., Kanakidou, M., Klimont, Z., Kupiainen, K., Law, K. S., Lund, M. T., Maas, R., MacIntosh, C. R., Myhre, G., Myriokefalitakis, S., Olivié, D., Quaas, J., Quennehen, B., Raut, J.-C., Rumbold, S. T., Samset, B. H., Schulz, M., Seland, Ø., Shine, K. P., Skeie, R. B., Wang, S., Yttri, K. E., and Zhu, T.: Evaluating the climate and air quality impacts of short-lived pollutants, Atmos. Chem. Phys., 15, 10529-10566, doi:10.5194/acp-15-10529-2015, 2015.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S.,

Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, I., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777-1813, doi:10.5194/acp-6-1777-2006, 2006.

Tonttila, J., Järvinen, H., and Räisänen, P.: Explicit representation of subgrid variability in cloud microphysics yields weaker aerosol indirect effect in the ECHAM5-HAM2 climate model, Atmos. Chem. Phys., 15, 703-714, doi:10.5194/acp-15-703-2015, 2015.

Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., Balkanski, Y., Bauer, S. E., Bellouin, N., Benedetti, A., Bergman, T., Berntsen, T. K., Beukes, J. P., Bian, H., Carslaw, K. S., Chin, M., Curci, G., Diehl, T., Easter, R. C., Ghan, S. J., Gong, S. L., Hodzic, A., Hoyle, C. R., Iversen, T., Jathar, S., Jimenez, J. L., Kaiser, J. W., Kirkevåg, A., Koch, D., Kokkola, H., Lee, Y. H., Lin, G., Liu, X., Luo, G., Ma, X., Mann, G. W., Mihalopoulos, N., Morcrette, J.-J., Müller, J.-F., Myhre, G., Myriokefalitakis, S., Ng, N. L., O'Donnell, D., Penner, J. E., Pozzoli, L., Pringle, K. J., Russell, L. M., Schulz, M., Sciare, J., Seland, Ø., Shindell, D. T., Sillman, S., Skeie, R. B., Spracklen, D., Stavrakou, T., Steenrod, S. D., Takemura, T., Tiitta, P., Tilmes, S., Tost, H., van Noije, T., van Zyl, P. G., von Salzen, K., Yu, F., Wang, Z., Wang, Z., Zaveri, R. A., Zhang, H., Zhang, K., Zhang, Q., and Zhang, X.: The AeroCom evaluation and intercomparison of organic aerosol in global models, Atmos. Chem. Phys., 14, 10845-10895, doi:10.5194/acp-14-10845-2014, 2014.

Turpin, B.J., Saxena, P., Andrews, E.: Measuring and simulating particulate organics in the atmosphere: problems and prospects. Atmospheric Environment 34:2983-3013, 2000.

van Ulden, A. and Wieringa, J.: Atmospheric boundary layer re- search at Cabauw, Bound.-Lay. Meteorol., 78, 39–69, 1996.

Vignati, E., Wilson, J. and Stier, P.: M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models, J. Geophys. Res., 109, D22202, doi:10.1029/2003JD004485, 2004.

Zhang, K., O'Donnell, D., Kazil, J., Stier, P., Kinne, S., Lohmann, U., Ferrachat, S., Croft, B., Quaas, J., Wan, H., Rast, S., and Feichter, J.: The global aerosol-climate model ECHAM-HAM, version 2: sensitivity to improvements in process representations, Atmos. Chem. Phys., 12, 8911-8949, doi:10.5194/acp-12-8911-2012, 2012.

Zhang, S., Wang, M., Ghan, S. J., Ding, A., Wang, H., Zhang, K., Neubauer, D., Lohmann, U., Ferrachat, S., Takeamura, T., Gettelman, A., Morrison, H., Lee, Y., Shindell, D. T., Partridge, D. G., Stier, P., Kipling, Z., and Fu, C.: On the characteristics of aerosol indirect effect based on dynamic regimes in global climate models, Atmos. Chem. Phys., 16, 2765-2783, doi:10.5194/acp-16-2765-2016, 2016.

27

TABLES

Table 1. Input data provided from AeroCom inventory and GAINS model for submicron particle emissions. The data is sorted according to its original structure in terms of mass, number, chemical species differentiation (BC, OC and SO_4), bi-level vertical distribution (2-zL) and base year. (✓) and (✗) indicate whether the data set contains a certain information or not, respectively.

Data	M	N	Species	2-zL	Year
AeroCom	✓	✗	✓	✓	2000
GAINS	✗	✓	✗	✗	2010

28

Table 2. Description of measurement sites for model versus observation evaluation.

Station	Lon	Lat	m. a. s. l.	Years	Reference
Botsalano, South Africa	25.8 ° E	25.5 ° S	1424	07/2006-08/2007	Laakso et al., 2008.
Cabauw, Netherlands	4.9 ° E	52.0 ° N	60	04/2008-03/2009	van Ulden and Wieringa, 1996.
Hohenpeissenberg, Germany	11.0 ° E	47.8 ° N	980	06/2007-11/2008	Birmili et al., 2016.
Hyytiälä, Finland	24.3 ° E	61.9 ° N	180	01/2009-12/2010	Hari and Kulmala, 2005.
K-Puszta, Hungary	19.6 ° E	47.0 ° N	125	03/2007-03/2009	Kiss et al., 2002.
Melpitz, Germany	12.9 ° E	51.5 ° N	84	01/2007-12/2008	Birmili et al., 2016.
Nanjing, China	118.9 ° E	32.1 ° N	40	12/2011-12/2014	Herrmann et al., 2014.
Po Valley, Italy	11.6 ° E	44.7 ° N	11	09/2004-09/2006	Hamed et al., 2007.
Sao Paulo, Brazil	46.7 ° W	23.5 ° S	760	10/2010-09/2011	Backman et al., 2012.
Tomsk FNV, Russia	84.1 ° E	56.4 ° N	80	01/2012-12/2013	Dal Maso et al., 2008.
Värriö, Finland	29.6 ° E	67.8 ° N	400	01/2009-12/2011	Hari et al., 1994.

29

Table 3. Annual total particle number (second and third columns) and global average ratios (fourth and fifth columns) of input emissions computed for the whole domain. R_{tot} ratios are calculated by firstly averaging the emissions among the whole domain for each data set, and secondly divide GAINS by AeroCom. This method aims at studying absolute differences in the global emissions with no regard to geographical distribution differences. In R_{grid} we firstly divide the data sets to keep the information of data sets differences for each grid cell, and secondly compute the median of gridded ratios. R_{grid} is weighted by surface area of the grid cell.

Global emissions	**AeroCom 10^{25} yr^{-1}**	**GAINS 10^{25} yr^{-1}**	**R_{tot} mean**	**R_{grid} median**
Total	3.42	7.39	2.16	1.00
Accumulation	0.028	1.74	62.14	48.65
Aitken	3.39	5.66	1.67	0.71

Table 4. Modeled global annually-averaged concentrations of total particle, CCN0.2 and CCN1,0 with AeroCom and GAINS data sets (second and third columns). Continental and (global) average ratios of total particle and CCN concentrations were calculated as in Table 3.

Global concentrations	**AeroCom 10^{12} m^{-3}**	**GAINS 10^{12} m^{-3}**	**R_{tot} mean**	**R_{grid} median**
Total	37.08	33.98	0.83 (0.91)	0.96 (0.99)
CCN0.2	1.65	2.47	1.69 (1.49)	1.16 (1.04)
CCN1.0	7.04	6.77	0.96 (0.96)	0.99 (0.98)

30

FIGURES

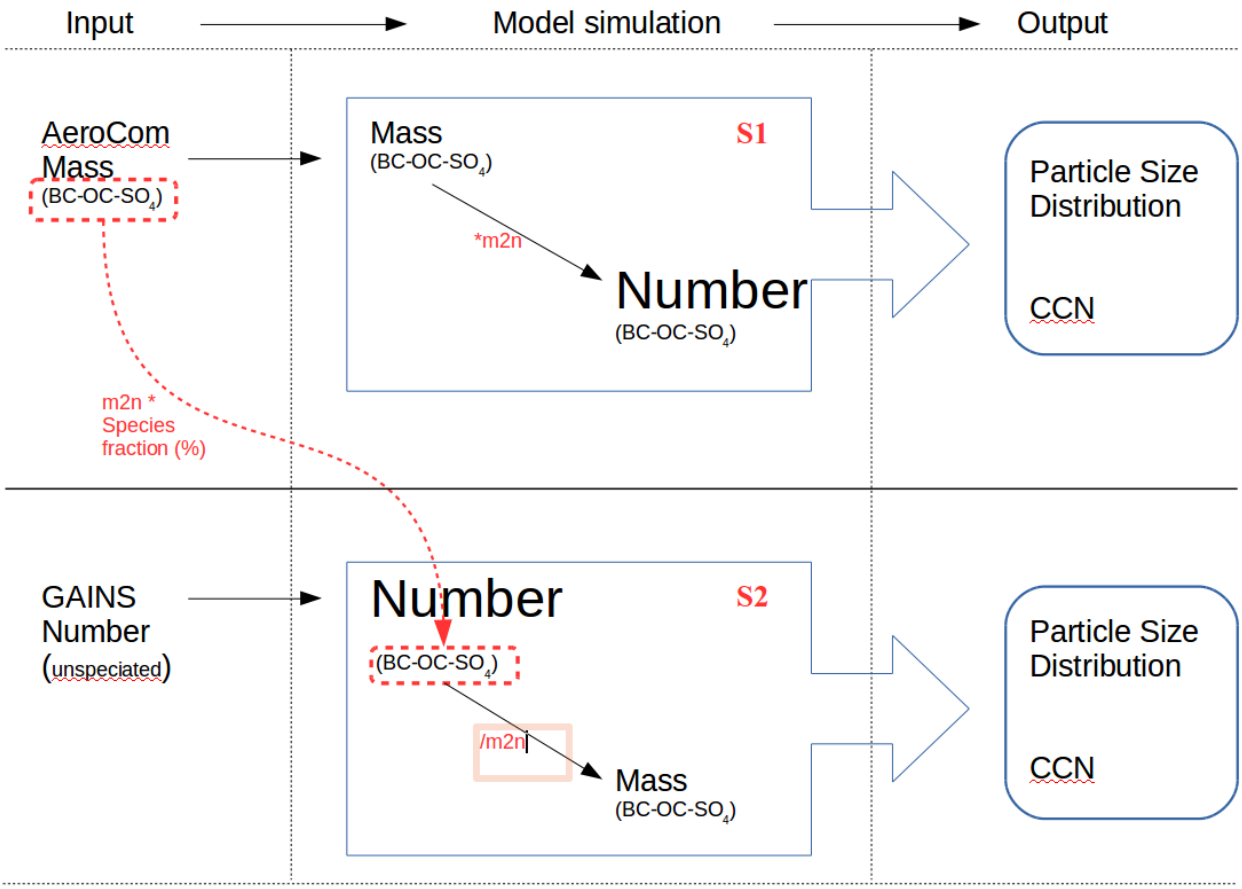


Figure 1. Framework describing the off-line steps to implement GAINS mass and number anthropogenic emissions in the ECHAM-HAM. The AeroCom mass-to-number (m2n) conversion factors and the chemical species fractions (%) of AeroCom number emissions were used to speciate GAINS number emissions. A specific m2n factor was used for each species for either mass-to-number (*m2n) or number-to-mass (/m2n) conversion.

31

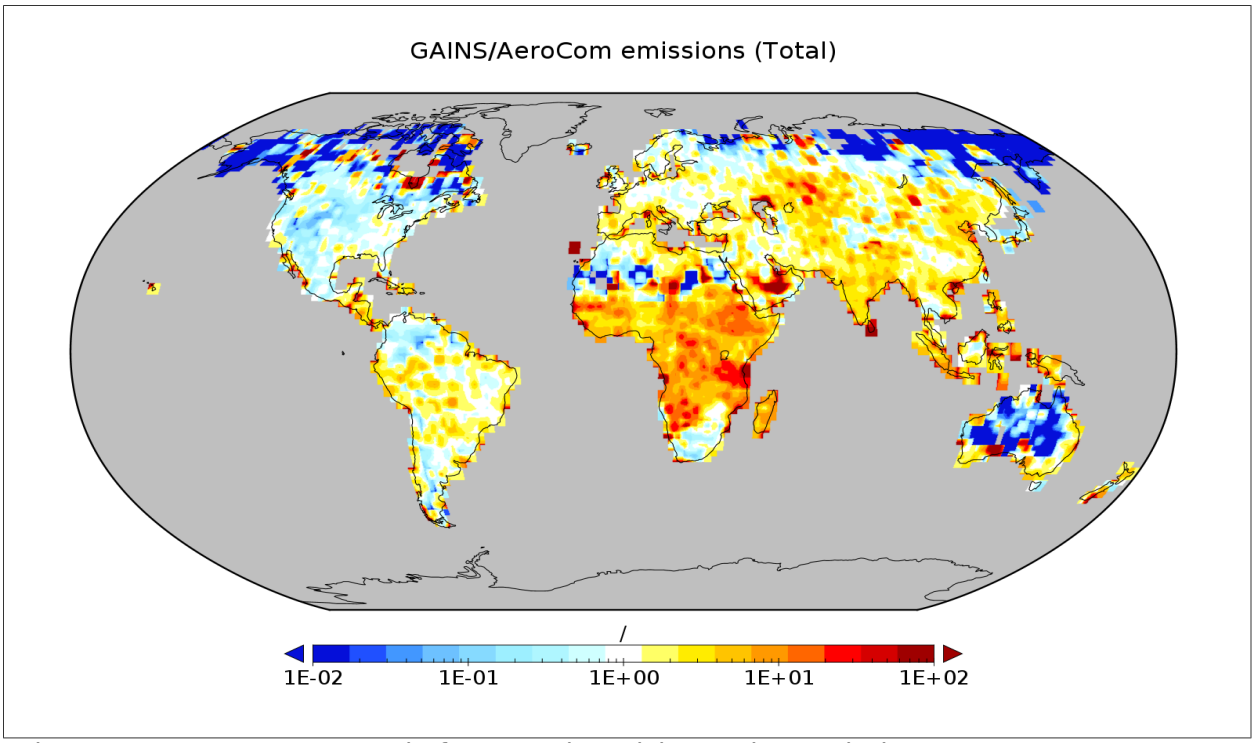


Figure 2. GAINS/AeroCom ratio for annual particle number emissions.

32

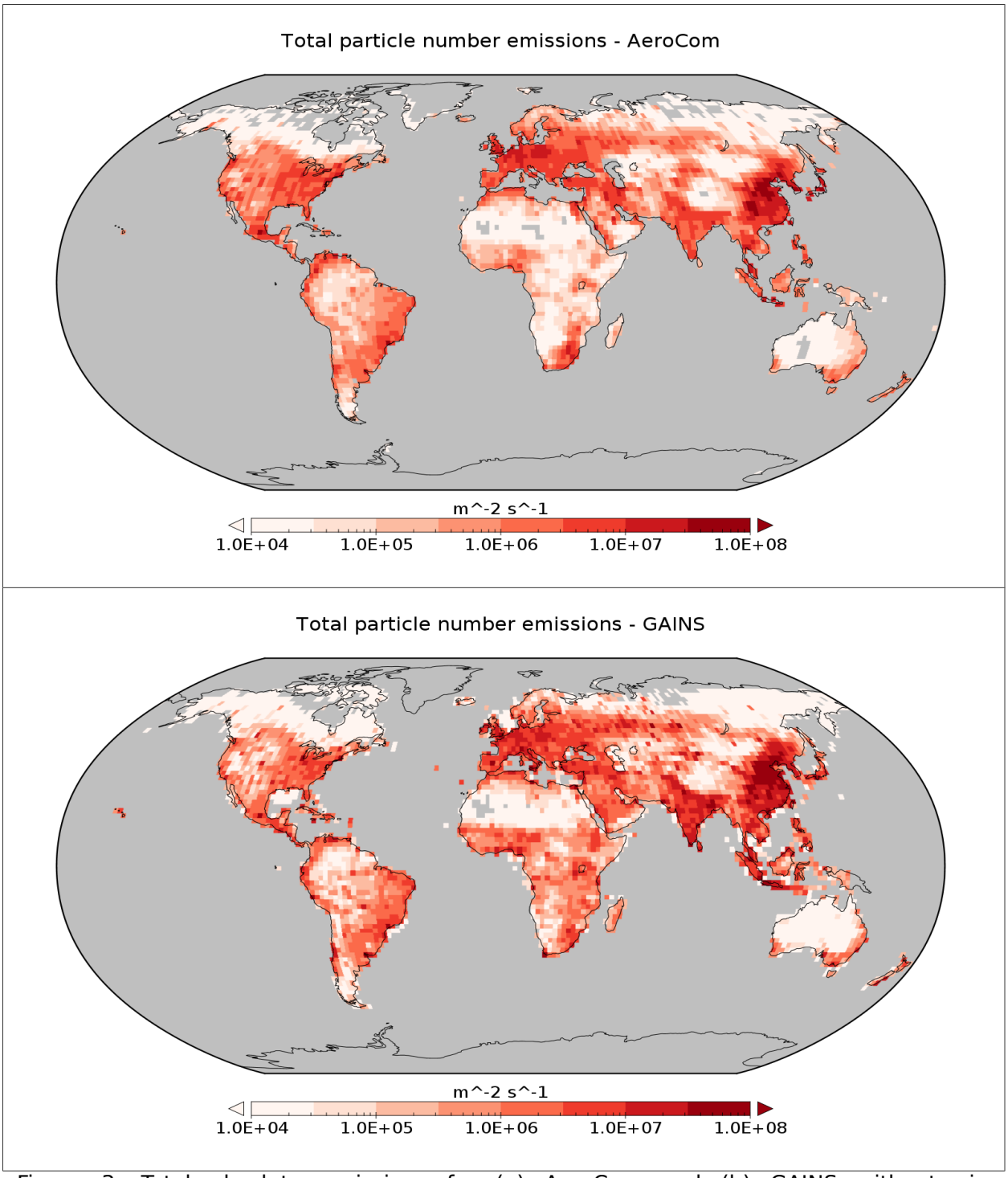


Figure 3. Total absolute emissions for (a) AeroCom and (b) GAINS without visual interpolation.

33

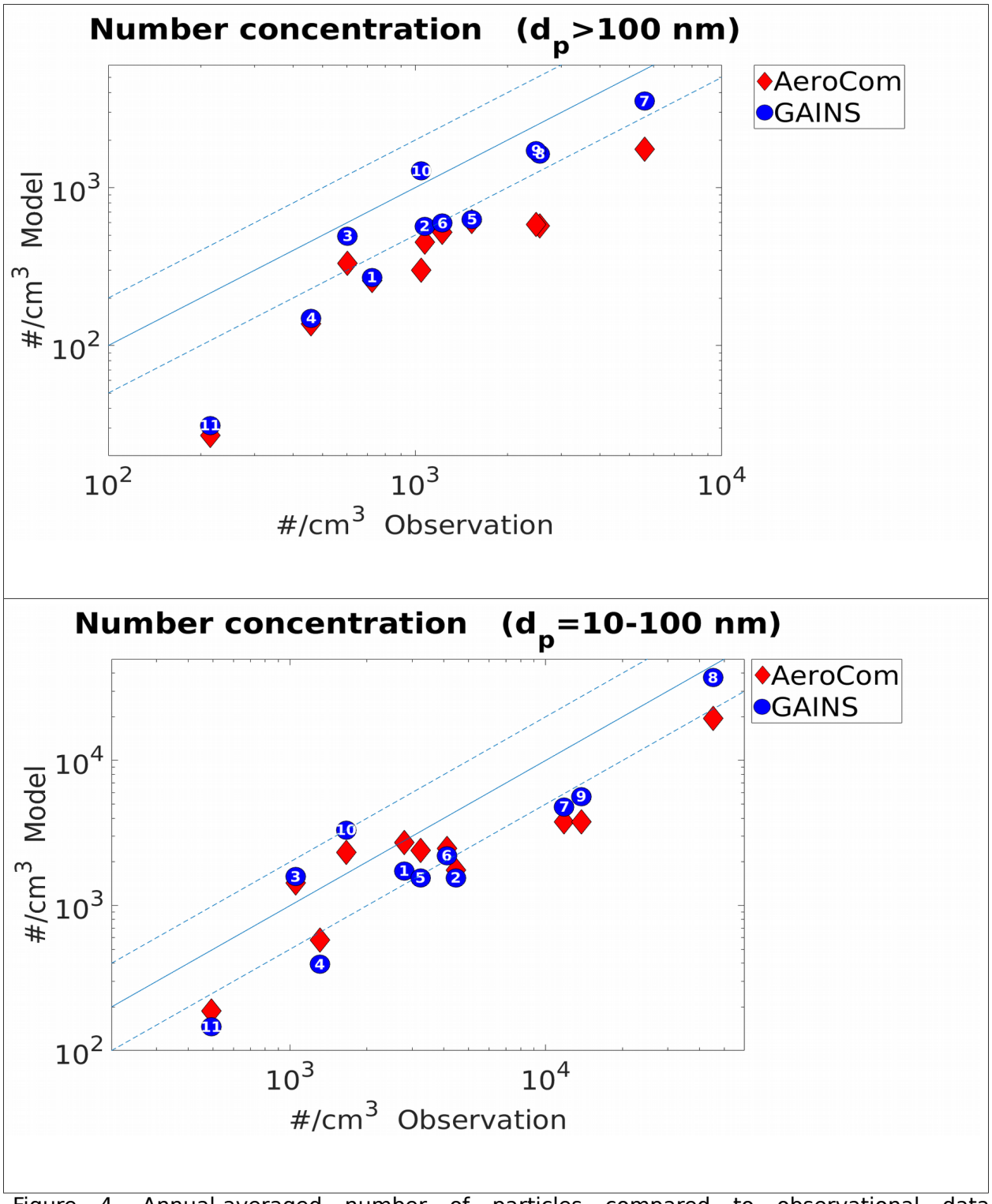


Figure 4. Annual-averaged number of particles compared to observational data. Measurement sites: 1: Botsalano; 2: Cabauw 3: Hohenpeissenberg; 4: Hyytiälä; 5: K-Puszta; 6: Melpitz; 7: Nanjing; 8: Po Valley; 9: Sao Paulo; 10: Tomsk FNV; 11: Värriö. Both plots include 1:1 and dashed 1:2, 2:1 lines.

34

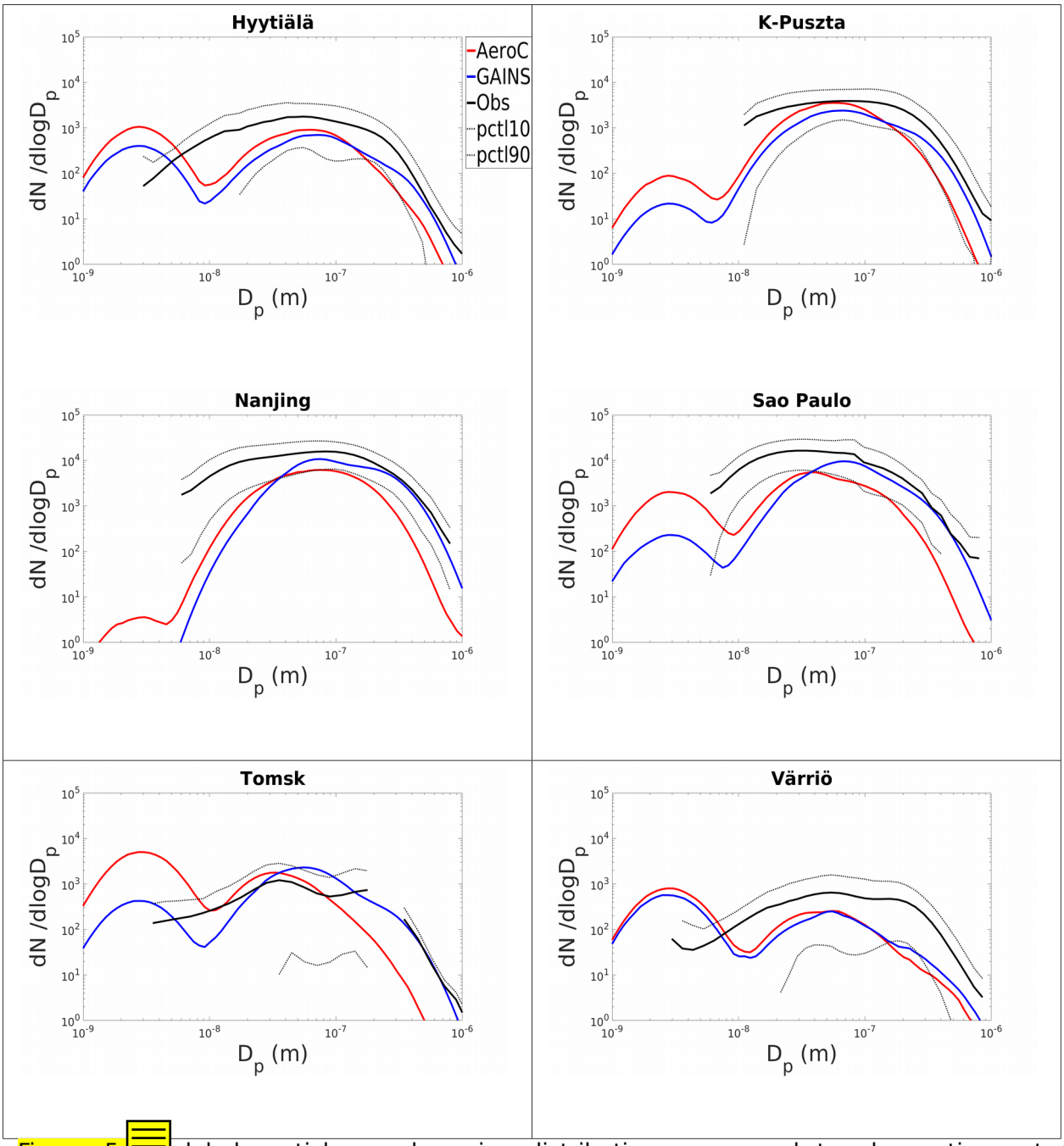


Figure 5. Modeled particle number size distributions compared to observations at 6 measurement sites.

35

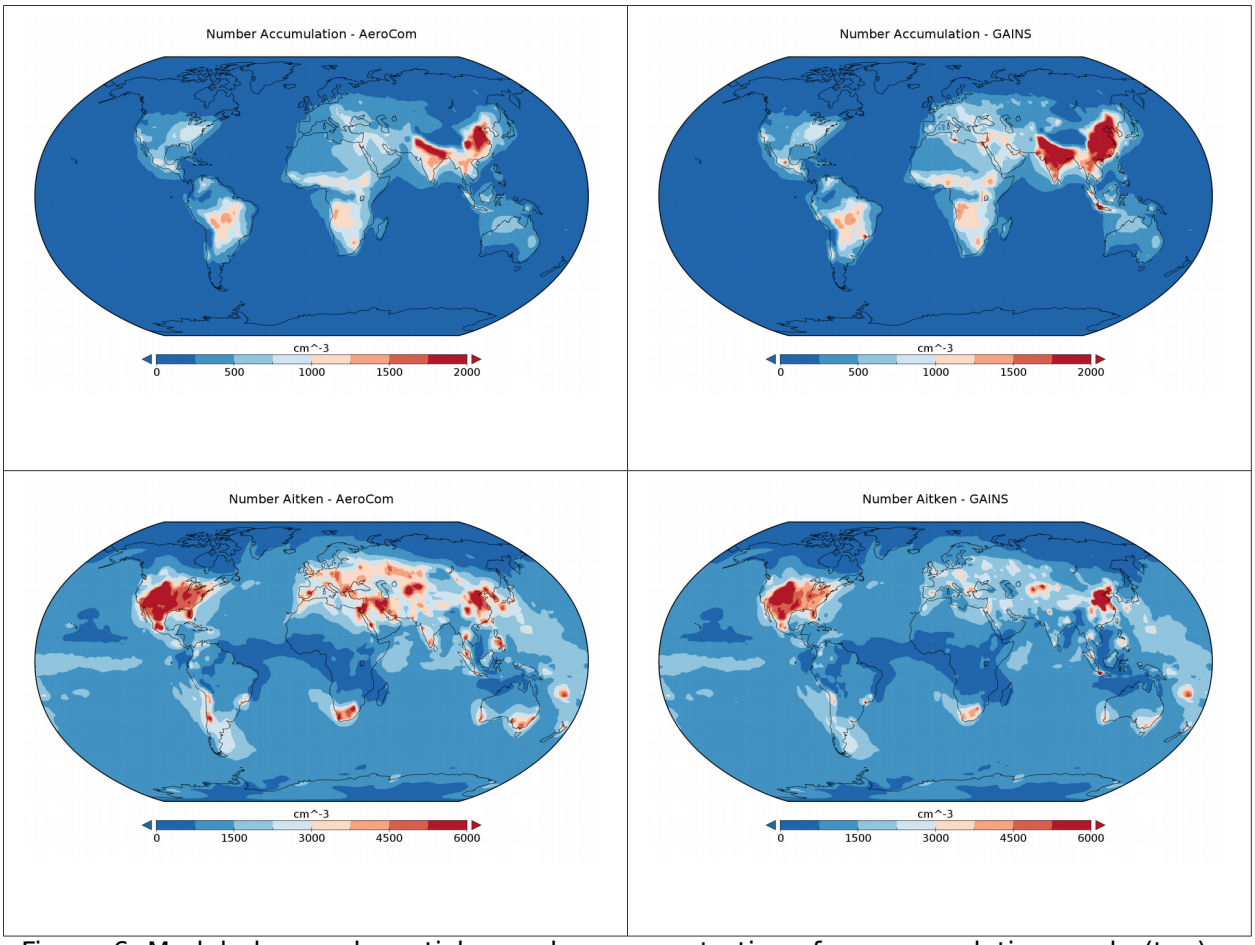


Figure 6. Modeled annual particle number concentrations for accumulation mode (top) and Aitken mode (bottom), at surface level.

36

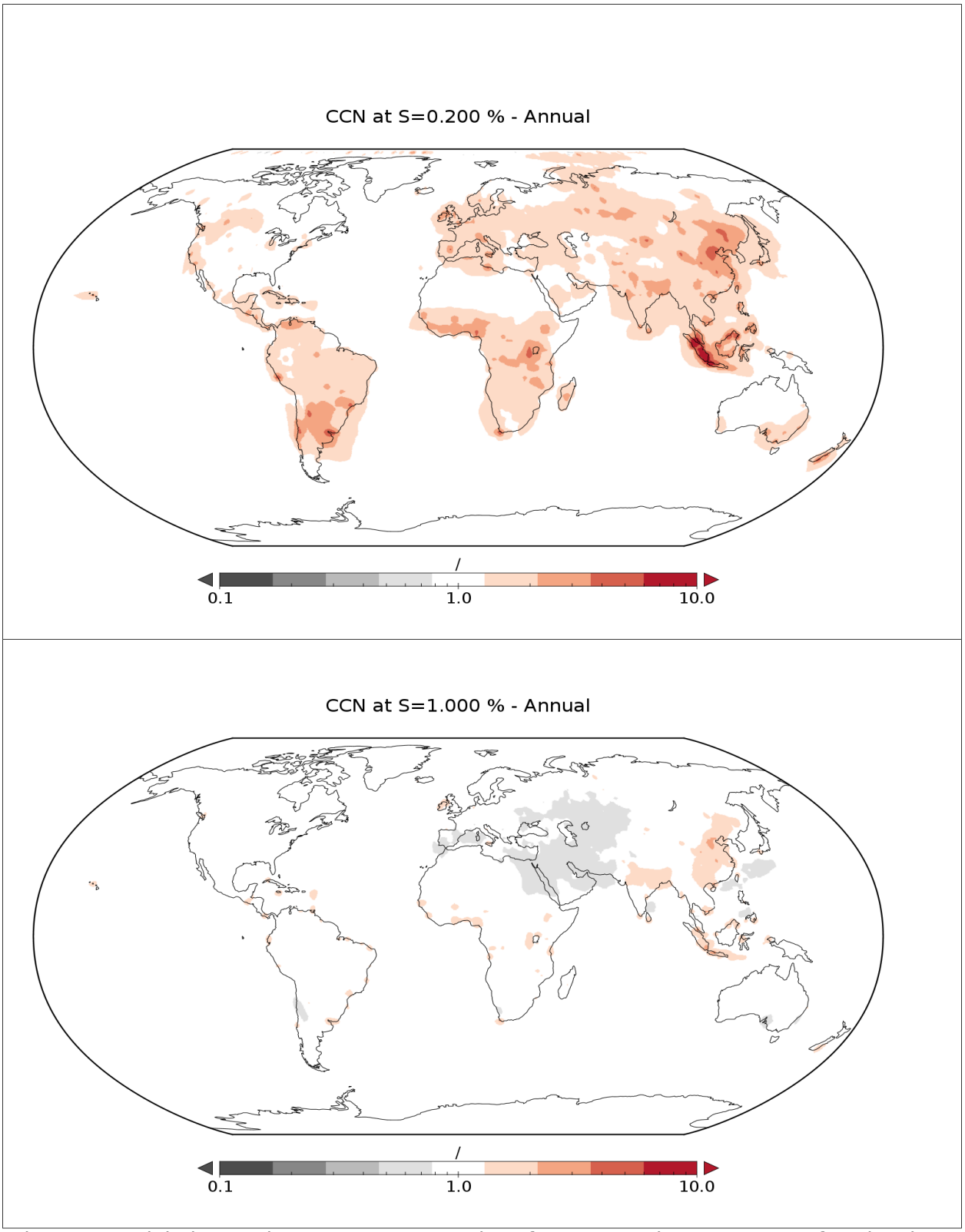
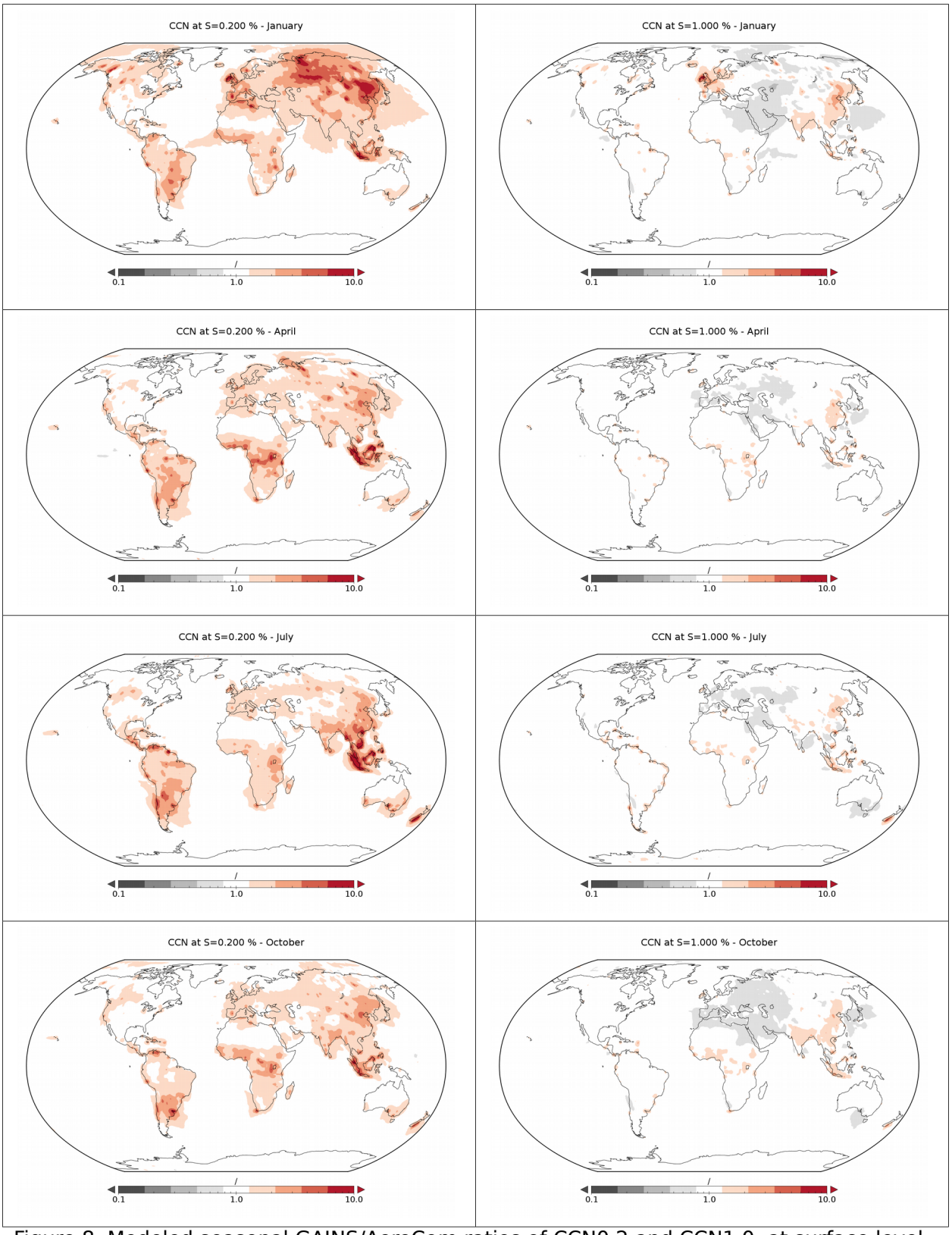


Figure 7. Modeled annual GAINS/AeroCom ratios of CCN0.2 and CCN1.0, at surface level.

37



1176 Figure 8. Modeled seasonal GAINS/AeroCom ratios of CCN0.2 and CCN1.0, at surface level.

38