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IMPROVING THE SIMULATION OF GLOBAL AEROSOL WITH SIZE-SEGREGATED ANTHROPOGENIC NUMBER EMISSIONS

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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

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climate model were replaced with the recently-formulated number emissions from the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS)-model, where the emission number size distributions vary, for example, with respect to the fuel and technology. A special attention in our analysis was put on 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-

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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 being 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 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

105 estimated from the particle mass emissions by the ECHAM-HAM during the
106 initialization routine. In more detail, the estimation of number emissions
107 consists of a simplistic multiplication of the given AeroCom mass emissions
108 by a mass-to-number conversion factor. Each conversion factor that is
109 applied for building the log-normal distribution is calculated by assuming
110 that the mass emissions for each main source sector are distributed to
111 predefined modes according to predefined densities, geometric mean radii
112 and standard deviations, as described by Vignati et al., (2004) and Stier et
113 al., (2005). This simplistic mass-to-number conversion factor does not
114 represent the relationship between the particle mass and number size
115 distributions in a realistic way, because such framework does not take into
116 account the variation of emitted particle number size distributions from
117 different emitting sources. The AeroCom inventory includes anthropogenic
118 activities, from which the mass-to-number converted emissions are split into
119 half between the Aitken and accumulation modes, and finally converted into
120 log-normal modes. However, the recently-developed inventories allow for
121 global aerosol simulations with a more detailed aerosol emission size
122 distribution (Paasonen et al., 2016) with the GAINS emission scenario model
123 (Greenhouse gas – Air pollution INteractions and Synergies; Cofala et al.,
124 2009; Amann et al., 2011). GAINS data are organized into more detailed
125 anthropogenic sources than AeroCom, with different particle number
126 emissions and size distributions related to different fuels and technologies.

127 In this work, we first develop a novel module for anthropogenic particle
128 number emissions in Earth System Models. Our experiment, performed with
129 ECHAM-HAM, consists of replacing the mass-to-number converted
130 anthropogenic AeroCom aerosol emissions with number emissions from the
131 GAINS-model. This study has a dual target: first, it aims at improving the
132 ECHAM-HAM capability for estimating particle number concentrations, with a
133 special focus on accumulation mode particles, and second, it investigates the
134 feasibility of using the GAINS model for global climate modeling studies by
135 running the ECHAM-HAM with both AeroCom and GAINS data sets. We will
136 make a comparison between AeroCom and GAINS in terms of emissions,
137 modeled particle number concentrations and size distributions, as well as
138 modeled CCN number concentrations. Finally, we will compare the modeled
139 number size distributions with observations in different environments around
140 the world.

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2 Materials and methods

2.1 Model framework

2.1.1 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 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.

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). 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.

2.1.2 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 emissions for year 2010 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.

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212 2.2 Aerosol schemes

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

222 $$J = 1.7\,x\,10^{-6} s^{-1} * \left[H_2SO_4\right] \quad . \qquad (1)$$

223 The settings of our simulations included a specific module for SOA formation.
224 Here, we modeled the SOA formation with both kinetic condensation onto a
225 Fuchs-corrected surface area (CS) and partitioning according to a preexisting
226 organic mass (Riipinen et al., 2011; Jokinen et al., 2015). This SOA module
227 includes three biogenic volatile organic compound (BVOC) tracers: isoprene,
228 endocyclic monoterpenes and other monoterpenes, each having monthly
229 resolutions for emissions. We did not use any nucleation scheme for organic
230 vapors, because the simple activation-type nucleation, while not accurate for
231 individual sites, describes the nucleation in different environments
232 reasonably well (Paasonen et al., 2010). The particle growth from nucleation
233 size to the d_p of 3 nm was calculated according to Kerminen and Kulmala
234 (2002). BVOC emissions were implemented using the MEGAN2 (Guenther et
235 al., 2006) model. MEGAN2 estimates biogenic emissions for about 150
236 compounds from different ecosystems, paying a particular attention to
237 monoterpenes. This framework takes into account several factors that
238 influence BVOC emissions, including the leaf age, soil moisture and light
239 environment. MEGAN2 was run offline and its output data were used for the
240 ECHAM-HAM input initialization. More details can be found in Makkonen et al.
241 (2012).

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

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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.

2.3 Anthropogenic Emissions

2.3.1 AeroCom

The first simulation was performed with the 2000 anthropogenic AeroCom data set. 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. 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-core 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

$$N = M/m \quad , \qquad (2)$$

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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 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

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

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 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.

2.3.2 GAINS

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. Therefore, 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-only emission data without chemical speciation and vertical distribution (see Table 1). Thus, 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.

2.4 Comparison with observations

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. 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

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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 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

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very close to one (see R_{grid} in Table 3). 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 Rat_{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 Rat_{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 in a prevalent area of the global domain.

It is important to mention that the high differences between GAINS and AeroCom in terms of Aitken and accumulation mode emissions that are presented in Table 3 are partly caused by the different shares of BC, OC, and SO_4 in GAINS and AeroCom data sets. In the 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. The significant difference in accumulation mode emissions and concentrations results from non-existing accumulation emissions from fossil fuels and biofuels in the AeroCom data set.

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 of underestimating particle number concentrations in model simulations, especially for the locations having high observed particle number concentrations. Underestimation of the highest particle concentrations might be, at least partly, related to the spatial resolution of

418 ECHAM-HAM, due to which the typically high particle concentrations near
419 urban or industrial areas will be distributed evenly into a large model grid
420 cell (Stier et al., 2005). A comparison of the model results with the
421 observational data shows that the GAINS implementation significantly
422 improved the reproduction of observed concentrations in accumulation mode
423 (d_p > 100 nm), being closer to observations than AeroCom at all 11 sites. For
424 the Aitken mode (d_p = 10-100 nm), similar improvement was not reached, as
425 the observed concentrations were better reproduced with AeroCom than with
426 GAINS at 8 sites.

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

453 The above results suggest that in ECHAM-HAM, as well as probably in other
454 climate models, the current nucleation and growth schemes may need

455 further revisions. However, it is also likely that the anthropogenic emissions
456 of especially nucleation mode particles in GAINS are still severely
457 underestimated for many source sectors (Paasonen et al., 2016). This is
458 because many of the measurements, on which the GAINS emission factors
459 are based, are not sensitive to non-solid nucleation mode particles, such as
460 those formed via nucleation of sulfur or organic vapors immediately after the
461 combustion or at small downwind distances in plumes from different
462 combustion sources (Stevens and Pierce, 2013). In addition, the lower
463 modeled Aitken mode particle concentrations from GAINS emissions may, in
464 some parts of the global domain, be also related to possible overestimations
465 in the accumulation mode particle emissions in the GAINS model, which are
466 consequently affecting the formation and growth of smaller particles.
467 Nonetheless, all the model versus observation comparisons between the
468 simulations clearly represent a consistent challenge for climate models in
469 modeling ultra-fine particle number size distributions.

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

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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, a significant fraction of the global accumulation mode particle concentration was observed in India, China and south-east Asia (see Figure 6), and thus the increase in absolute CCN0.2 concentration due to anthropogenic emissions is largest 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

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 tangible 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.

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 of climatically active primary particles and simultaneous decreases in secondary ultrafine particle formation due to higher values of CS and coagulation sink.

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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, 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 first next step, the nucleation parameterizations and the sensitivity of the concentrations of sulfuric acid (the main precursor in the applied nucleation parameterization) to altered CS should be revised.

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, 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.

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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

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1011 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, 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.

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Table 3. 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.

Global emissions	**AeroCom** 10^9 **m^{-2} s^{-1}**	**GAINS** 10^9 **m^{-2} s^{-1}**	R_{tot} **mean**	R_{grid} **median**
Total	7.23	15.63	2.16	1.00
Accumulation	0.06	3.68	59.18	48.65
Aitken	7.17	11.96	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)

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1039 FIGURES
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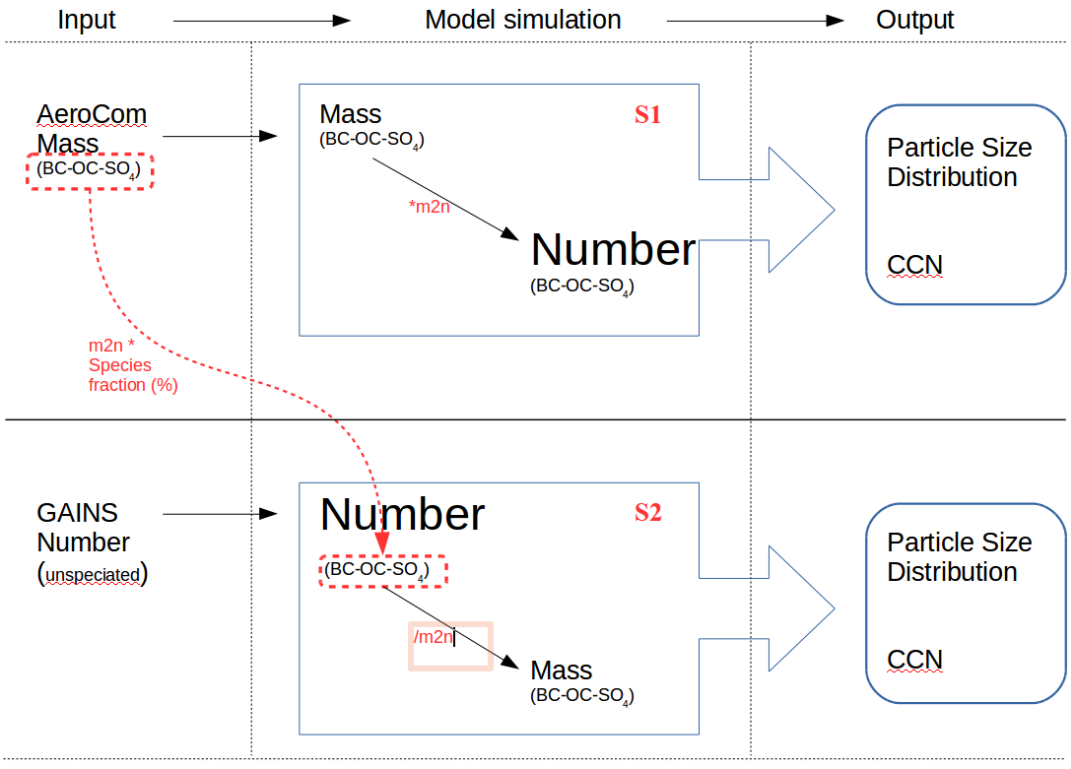

1042 Figure 1. Framework describing the off-line steps to implement GAINS mass and number
1043 anthropogenic emissions in the ECHAM-HAM. The AeroCom mass-to-number (m2n)
1044 conversion factors and the chemical species fractions (%) of AeroCom number emissions
1045 were used to speciate GAINS number emissions. A specific m2n factor was used for each
1046 species for either mass-to-number (*m2n) or number-to-mass (/m2n) conversion.
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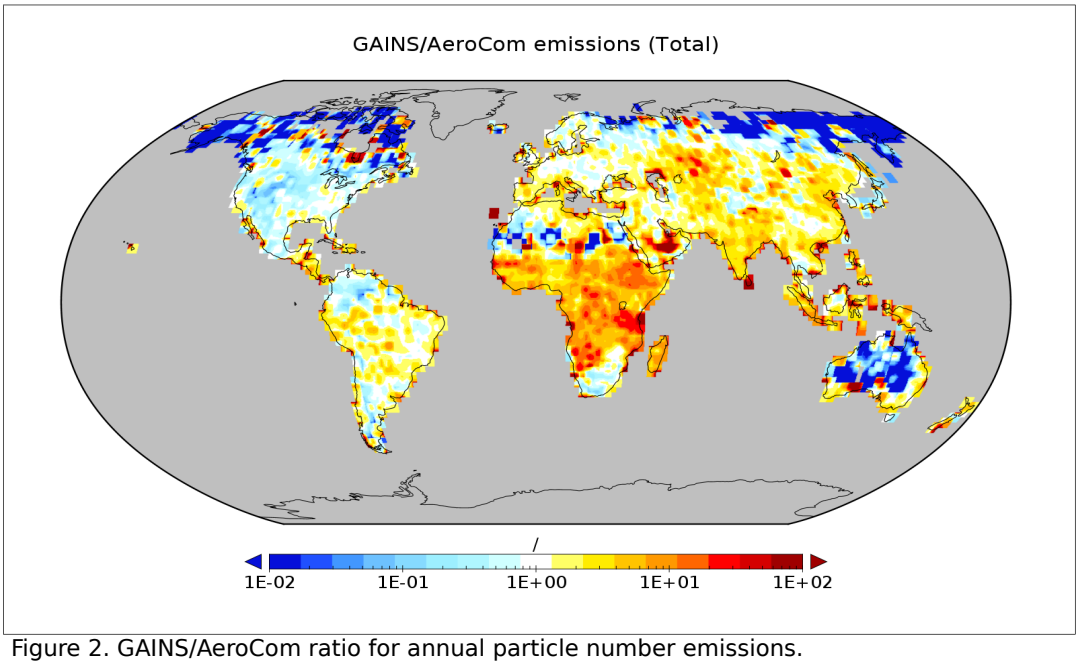

Figure 2. GAINS/AeroCom ratio for annual particle number emissions.

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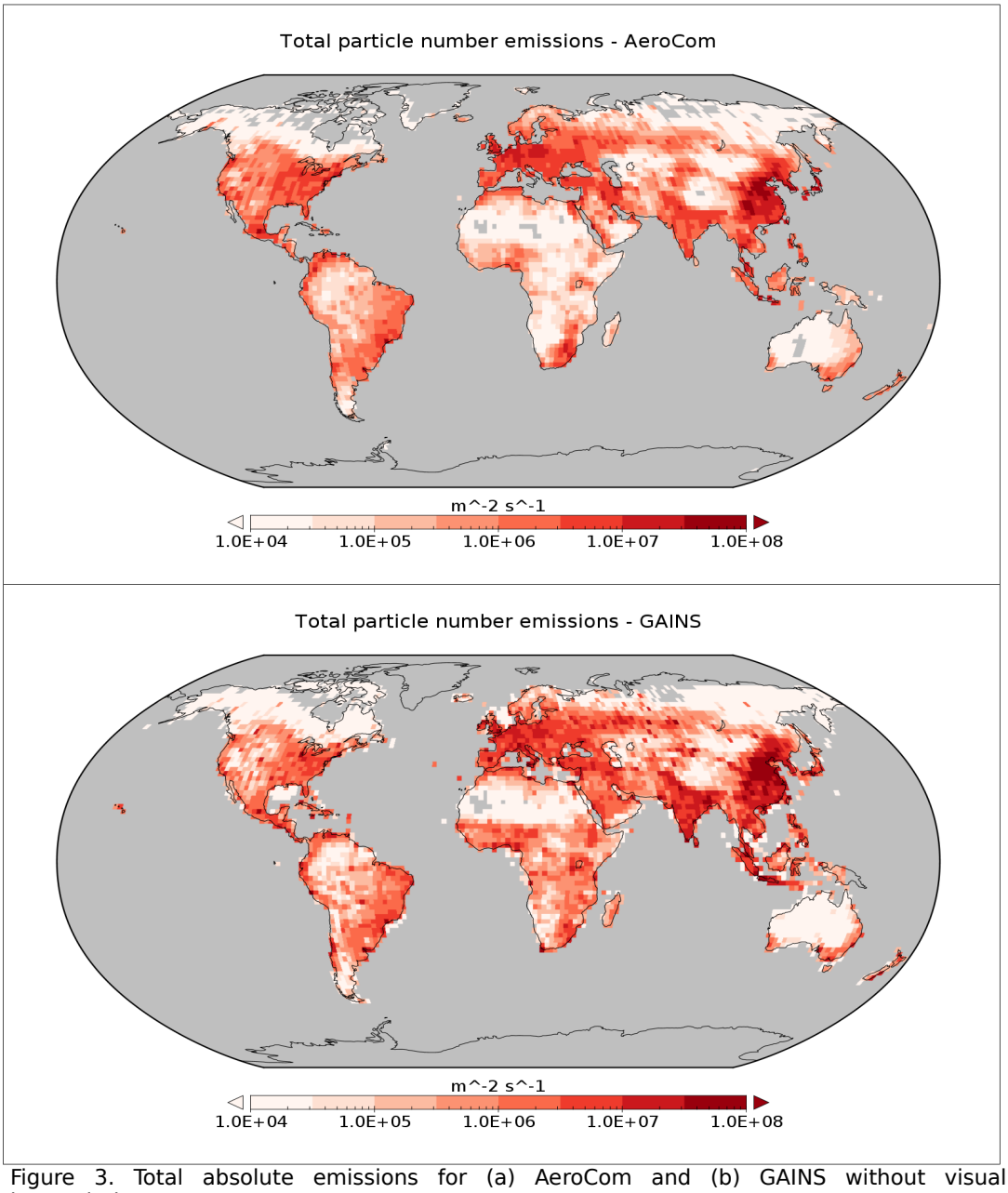

Figure 3. Total absolute emissions for (a) AeroCom and (b) GAINS without visual interpolation.

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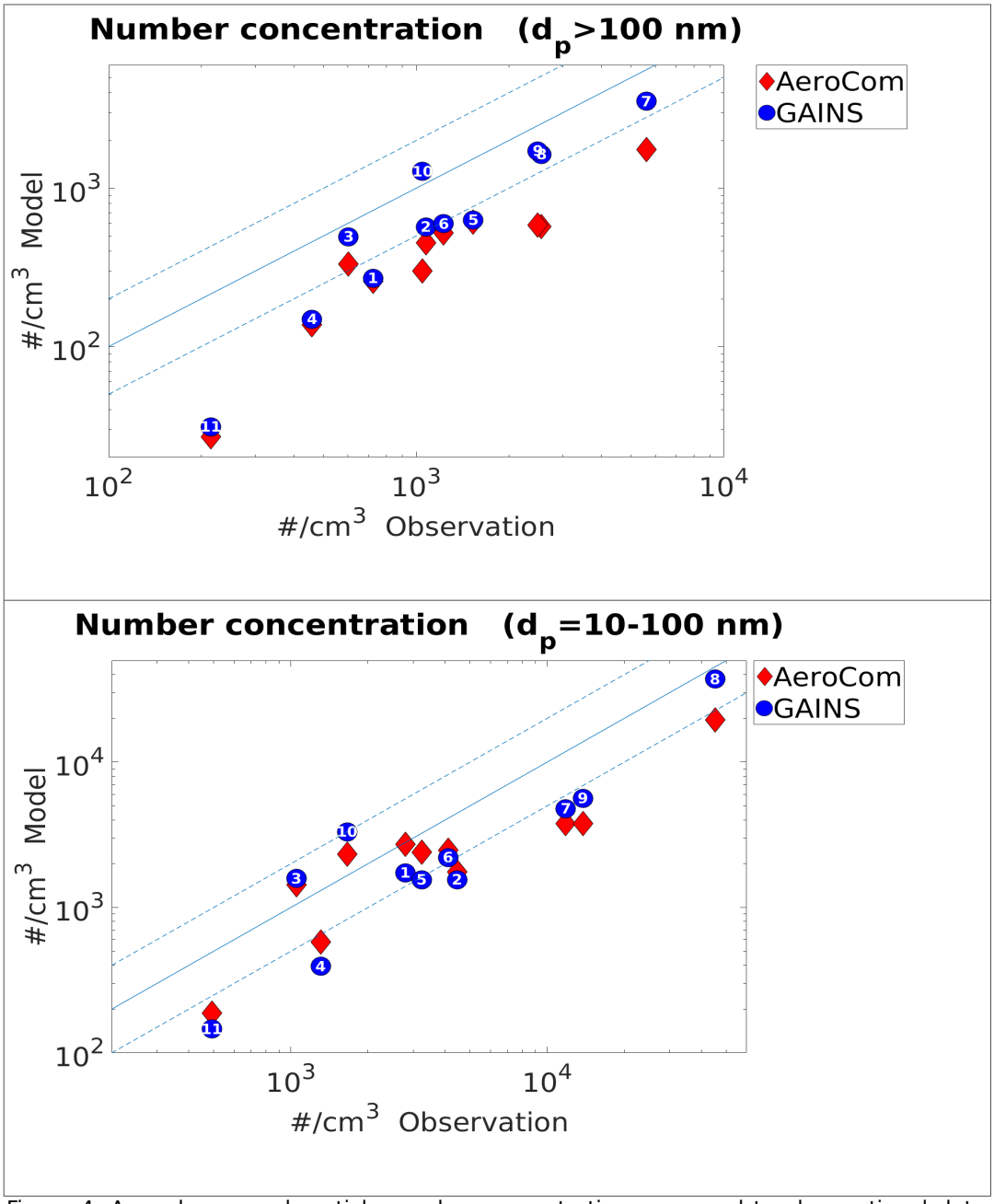

1071 Figure 4. Annual-averaged particle number concentration compared to observational data.
1072 Measurement sites: 1: Botsalano; 2: Cabauw 3: Hohenpeissenberg; 4: Hyytiälä; 5: K-Puszta;
1073 6: Melpitz; 7: Nanjing; 8: Po Valley; 9: Sao Paulo; 10: Tomsk FNV; 11: Värriö. Both plots
1074 include 1:1 and dashed 1:2, 2:1 lines.

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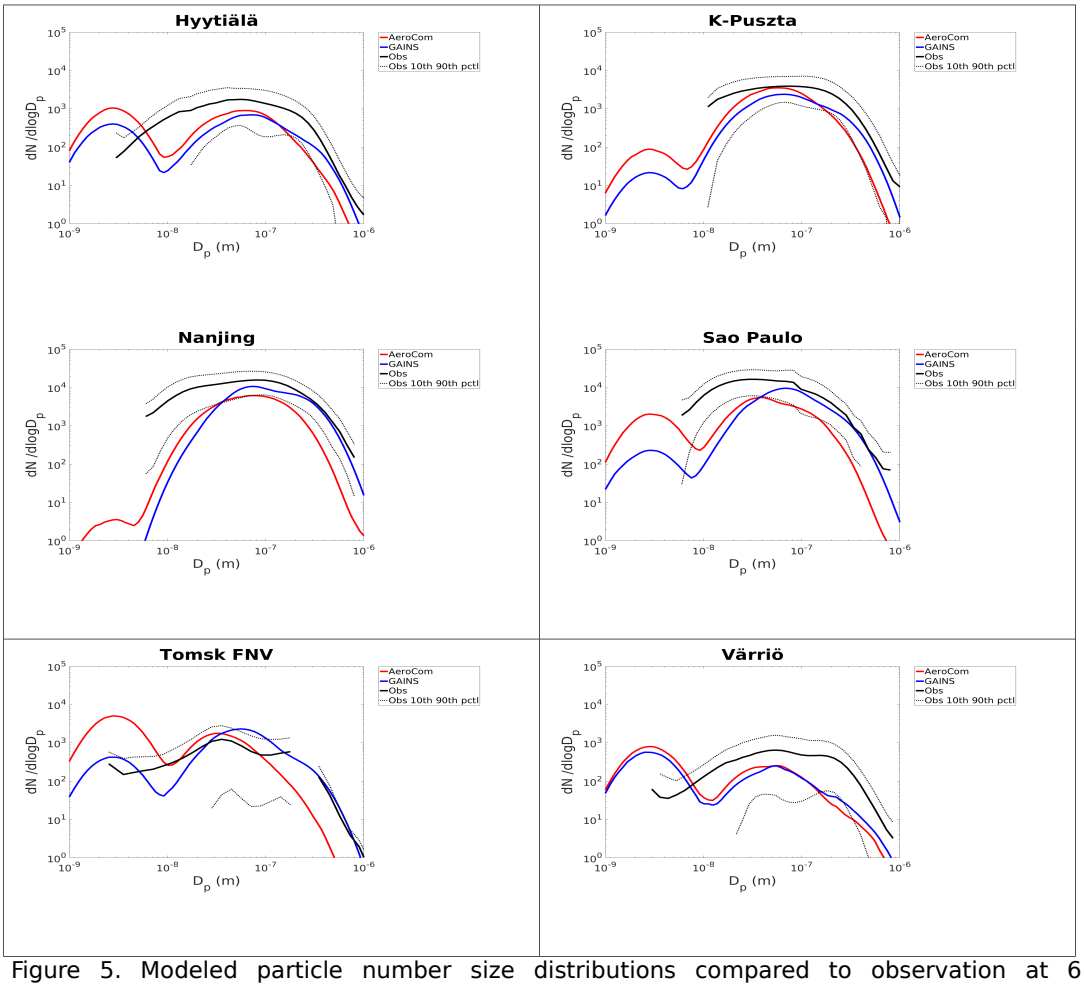

Figure 5. Modeled particle number size distributions compared to observation at 6 measurement sites.

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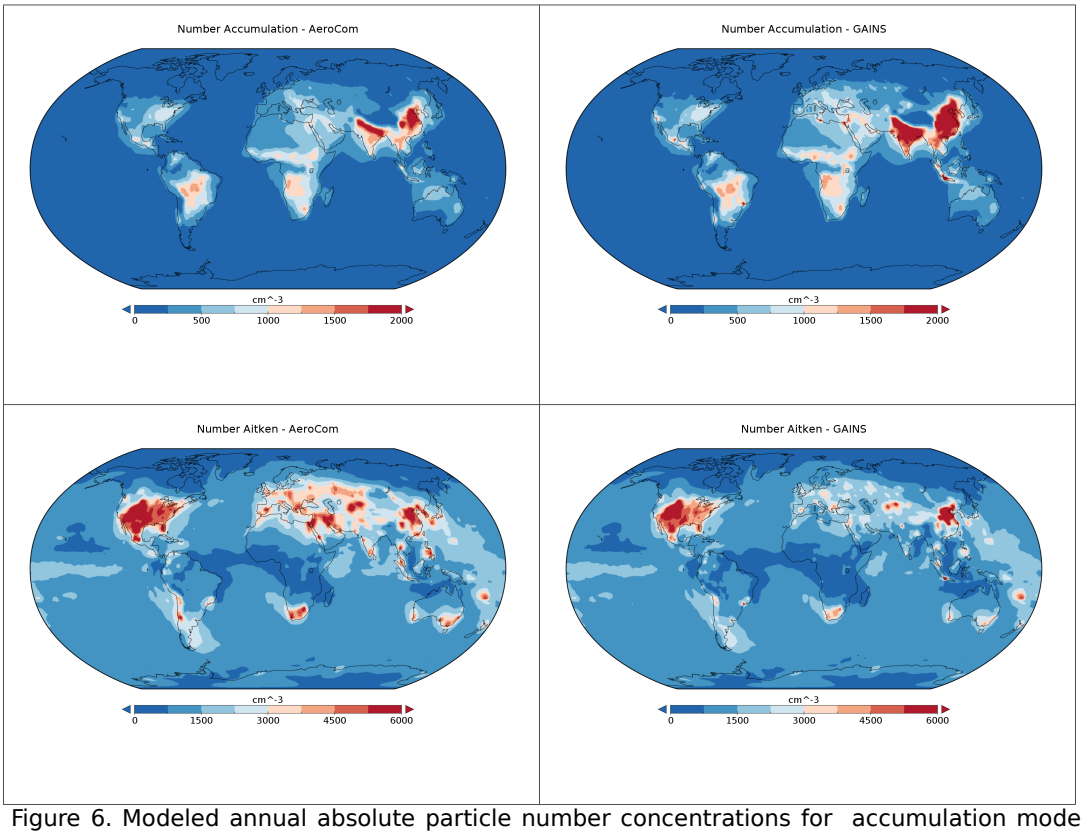

Figure 6. Modeled annual absolute particle number concentrations for accumulation mode (top) and Aitken mode (bottom).

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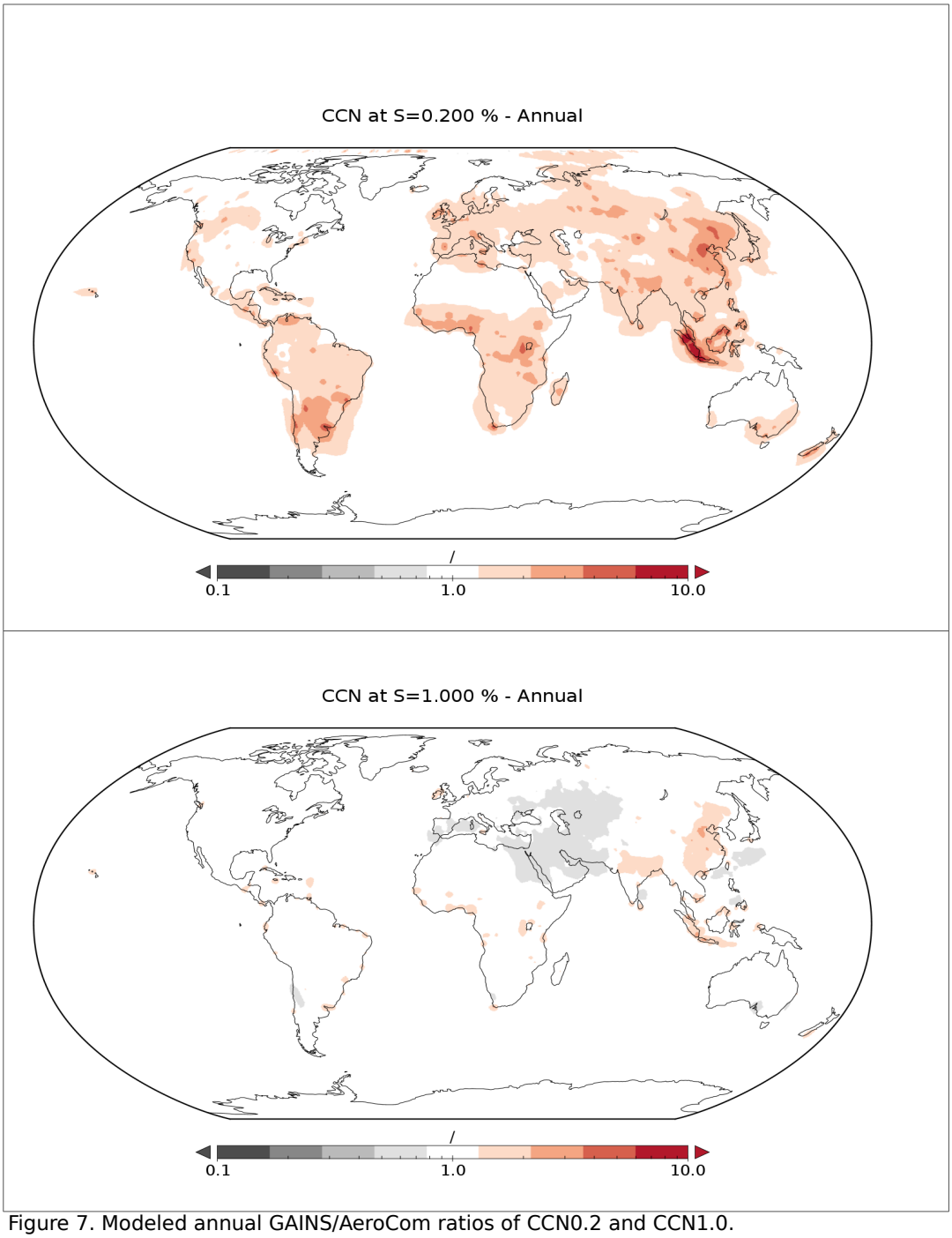

1101 Figure 7. Modeled annual GAINS/AeroCom ratios of CCN0.2 and CCN1.0.

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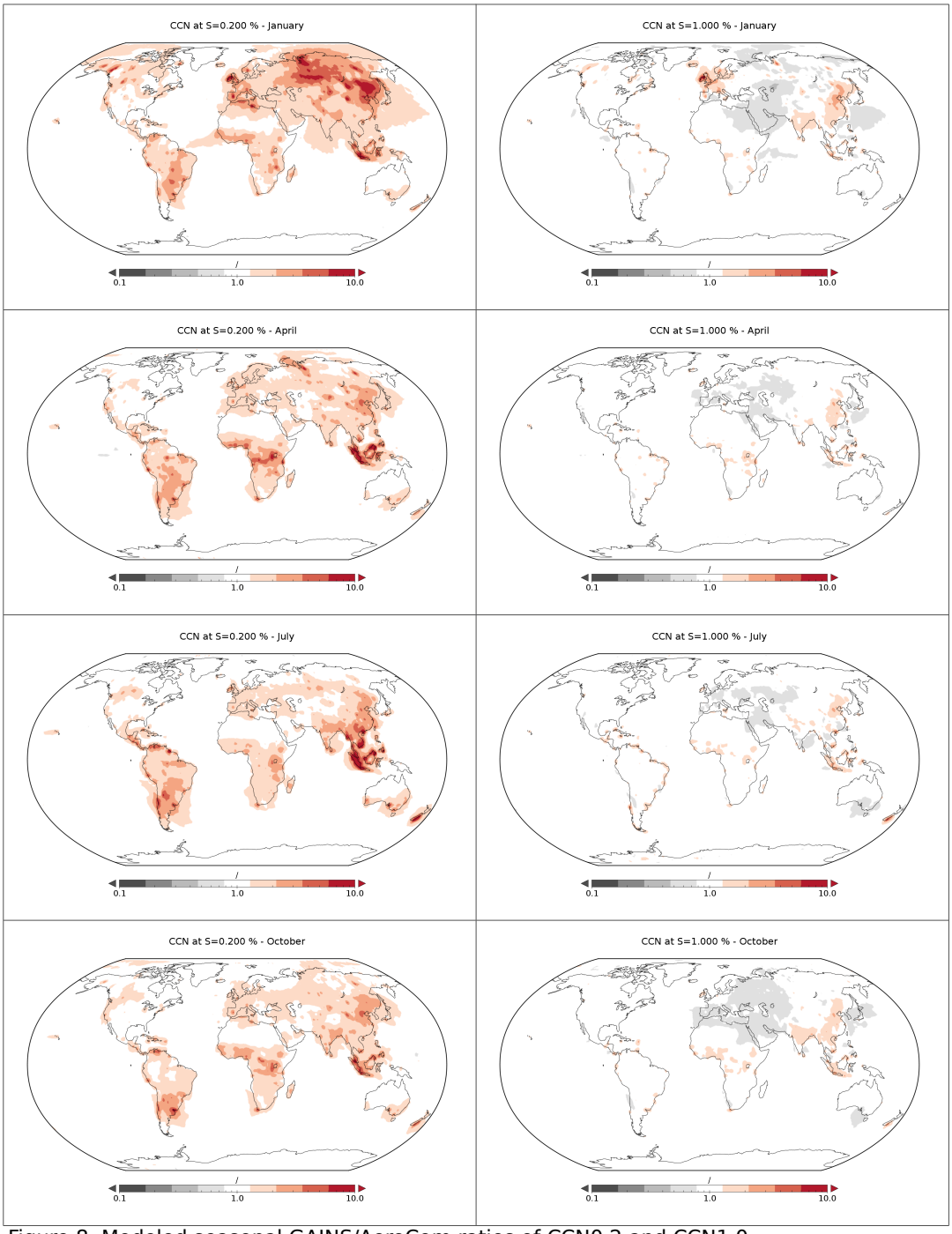

1102 Figure 8. Modeled seasonal GAINS/AeroCom ratios of CCN0.2 and CCN1.0.