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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulating aerosol microphysics with the ECHAM/MADE GCM – Part II: Results from a first multiannual integration" *by* A. Lauer and J. Hendricks

A. Lauer and J. Hendricks

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Reply to anonymous referee #2

We wish to thank referee #2 for the detailed comments and the in-depth review of our article, which enabled us to significantly improve our paper.

General comments:

1. - We are sorry that the referee thinks that our paper is much too long. In our opinion, the section about aerosol mass distribution is essential to provide a complete and consistent picture of the aerosol properties simulated and presented in this paper. This is necessary to go into further details on the simulated composition, size-distribution,

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and in particular the impact of aerosol microphysics for the simulation of submicrometer particle mass and number. We agree with the referee, that the basic findings on the mass distribution are not new. Nevertheless, we think this section provides the basis for a better understanding of the results presented in the following sections. Thus, we don't think it's a good idea to remove the section on aerosol mass distribution just to reduce the length of the paper. However, we shortened all paragraphs of section 3.1 as much as possible. - We added a short discussion and a figure of the primary aerosol compound black carbon to section 4.4 to extend the presentation of the new results of this work. - The title has been extended: "Results from a first multiannual integration of the submicrometer aerosol" - For comparisons with observations, we refer to section 3 of the first part of this study (Atmos. Chem. Phys., 5, 3251-3276, SRef-ID: 1680-7324/acp/2005-5-3251), which intensively compares model results to observations covering all relevant aerosol properties: mass concentration of various aerosol compounds, particle number concentration, aerosol size-distribution. In our opinion, any repetition of these comparisons from part I would unnecessarily lengthen part II.

2. We agree with the referee that the modal approach may cause uncertainties in representing the particle size-distribution. We therefore included the following statement in the conclusion section:

"The representation of the aerosol size-distribution by lognormal modes with constant width is a widely used technique applied in global aerosol simulations. Nevertheless, this approach implies simplifications of complex aerosol size-distributions. It may cause uncertainties, for instance, in the case of fresh aerosol generated by nucleation or emissions which can perturb the size-distribution by well pronounced bursts. Inaccuracies may also occur in representing aerosol size-distributions perturbed by cloud processing. Nevertheless, the appropriateness of lognormal modes to represent observed aerosol size-distributions under many different atmospheric conditions has been documented in many publications (e.g., Whitby, 1978). Furthermore, severe perturba-

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tions of the aerosol size-distribution as discussed above frequently occur on scales smaller than those of current GCM. Hence we think that representing the aerosol size-distribution by a more detailed approach as, for instance, a sectional model would not fundamentally change the main findings of this study."

A discussion of the effects of assuming internal mixtures only has already been provided in the first part of this study (Atmos. Chem. Phys., 5, 3251-3276, SRef-ID: 1680-7324/acp/2005-5-3251, Section 2.2.1).

3. According to the literature, the coarse mode mostly has little effect on the submicrometer size-range for both, particle number concentration and particle mass concentration. We do not claim that this is true for all cases found in nature (in particular for the source regions of sea salt and dust), but we think we won't lose too much accuracy due to neglect of the coarse mode when investigating average properties of submicrometer particles dominated by other components than sea salt and desert dust. To give some more details on the omission of the coarse mode in the manuscript, we added the following sentences to the model description of MADE (section 2.2):

"A calculation of Binkowski and Roselle (2003) for a typical average continental aerosol size-distribution showed, that the Aitken mode loses only about 0.1%/hr of particle number concentration and about 0.02%/hr of particle mass to the coarse mode due to intermodal coagulation. The calculated loss rates of the accumulation mode to the coarse mode are even lower (0.002%/hr and 0.0008%/hr for number and mass concentration, respectively). Thus, about 4 weeks would be needed to reduce Aitken mode particle number concentration to 50% due to intermodal coagulation with coarse particles, which is substantially longer than the typical residence times of Aitken mode particles. Hence, the coarse mode can be omitted without losing much accuracy when focusing on average properties of submicrometer particles under typical continental conditions. Nevertheless, the concentrations of sea salt and dust in the accumulation mode could be overestimated by the model due the neglect of intermodal coagulation with coarse particles which can show comparatively large concentrations close to their

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sources. However, this overestimation is limited by the short residence time of the coarse particles due to efficient sedimentation."

We agree with the referee that the budget of Aitken particle number concentration is dominated by a process (nucleation) which is uncertain in the model. The effect occurs since the global mean Aitken mode number concentration is dominated by very large concentrations of very fine particles near the tropopause. However, these very fine particles have a negligible contribution to Aitken mode mass concentration and do not affect the particle number and mass concentrations in the boundary layer. Hence this is a special feature of the altitudes near the tropopause which impacts the global budget of Aitken mode particle but which does neither affect the accumulation mode nor the Aitken mode particle properties at lower altitudes.

In order to point out the importance of nucleation for the simulation in general we included several statements in the manuscript (end of section 3.1, sulfate; section 4.1, 2nd paragraph).

4. We absolutely agree with the referee, that our results depend on the specific parameterizations and boundary conditions used. Actually, this is true for every model study. Since ECHAM/MADE is a climate model producing its own meteorology, several years have to be simulated and averaged to minimize the influence of the interannual variability on the results. Varying numerous kinds of parameterizations would require a huge ensemble of such multiannual simulations to obtain significant results. Due to the high computational resources required by a climate model, such ensembles are currently beyond the capacity of any super computer. The only way to deal with this issue in a climate model is to implement every parameterization as reasonable as possible and to compare the final results with observations. This is exactly what we have done so far (part I). Since we have no information on how individual parameterizations and boundary conditions change our results, we could just make a guess which wouldn't add any substantial information to the paper. However, we think this is actually the case and a true limitation in every GCM model study.

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5. We agree with the referee and replaced "aerosol dynamics" with "aerosol microphysics" throughout the text.

Specific comments:

Title: This is the second part of two closely related papers. In order to stay consistent with the title of part I, which cannot be changed any more, we do not want to change the common first part of the title. In addition, we think the title should be related to the whole content of the paper. However, in accordance with the referee comments of referee #1, we changed the second part of the title to: "Results from a first multiannual integration of the submicrometer aerosol".

Abstract: We omitted the budget of NO3 in the abstract because of it's high uncertainty. HNO3 required to calculate the gas/aerosol partitioning is currently implemented in form of a prescribed climatology only. However, budget and average residence time of NO3 can be found in table 1. Again, in our opinion, the section about aerosol mass distribution is an essential part of the paper to provide a complete and consistent picture of the aerosol properties simulated. As we think the abstract should provide a short summary of the whole paper, we did not remove this part of the abstract. As recommended by the referee, we emphasized the results on the role of aerosol microphysics by extending the corresponding part of the abstract.

Introduction:

1. We agree with the reviewer that it is a good idea to mention current work on global aerosol models performed within AeroCom. We added two further examples of GCMs including aerosol microphysics and we included the following paragraph in the introduction: "Due to the high relevance to climate research, the representation of aerosols is currently also subject to improvement in some other general circulation models. Different approaches and techniques are applied regarding the representation of the aerosol size-distribution (modal with fixed standard deviation or bin scheme), the number of modes or bins, the aerosol components considered and the number of aerosol micro-

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physical processes included. The progress achieved so far is well documented under the framework of the AeroCom Aerosol Model Intercomparison Initiative (Textor et al., 2006). Model results and further information can also be found on the AeroCom web page http://nansen.ipsl.jussieu.fr/AEROCOM/."

2. The differences between MADE and M7 are already discussed in the first part. We do not think these have to be repeated again. In Part I we wrote: "The major differences between HAM and MADE can be characterized as follows: HAM considers seven log-normally distributed modes, each representing a specific aerosol composition in a fixed size-range. In contrast, MADE considers a trimodal log-normal size-distribution and assumes a perfect internal mixture of the different aerosol compounds. The log-normal modes predicted by MADE are not fixed to prescribed size-ranges as in the case of HAM. The computer capacities saved by MADE due to the smaller number of modes is spend to simulate a larger number of aerosol compounds. While MADE predicts the full SO4/NO3/NH4/H2O system, HAM currently neglects nitrate (NO3) and considers a prescribed degree of SO4 neutralization by ammonium (NH4)."

3. Please see response to referee comment on section 2.2, 1.

Section 2.1: As suggested by the referee, we shortened the model description of ECHAM4 in section 2.1 as much as possible leaving only the most fundamental references.

Section 2.2:

1. As the standard deviation is kept constant in this MADE version and we neglected the coarse mode, the aerosol size-distribution is described by 2 (instead of 3) modes with 2 moments each. This results in 4 prognostic moments used to simulate the submicrometer aerosol size-distribution. Due to the constant standard deviation and the assumption of log-normally distributed modes, the geometric mean diameter of each mode can be calculated from the simulated total volume (calculated from the mass concentrations of the individual compounds) and particle number concentration.

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Thus, we do not understand exactly, what the referee means by stating that it is not true that particle size is calculated explicitly. Nevertheless, we removed all "explicitly" throughout the text.

2.+3. Again, according to literature, the impact of intermodal coagulation with the coarse mode on an average continental aerosol size-distribution is only weak. Please see response to general comments, 3.

4. Yes, as discussed in the first part (section 2.1 and 2.2.2), we consider aging of BC and POM. The conversion of hydrophobic into hydrophilic BC/POM is parameterized as an exponential decay using an e-folding time of 24 h.

Section 3:

The GCM has neither been used in a CTM mode nor nudged to reanalysis. All meteorological fields have been calculated in a self-consistent way by ECHAM itself.

Section 3.1:

- For discussion on the potential removal of this section, see response to "General Comments, 1.". - We integrated several statements on the AeroCom results throughout Section 3. - We decided to use STP conditions since this is a common way to express particle number concentrations in studies focusing on in-situ aerosol measurements.

Sulfate:

- The average lifetime of SO2 is about 2 days. - We used the volcanic SO2 emissions from Spiro et al. (1992). These include contineous emissions only, explosive volcanoes are not included. According to these emission data, there are volcanic SO2 emissions in Indonesia, but these are spread over a larger area and thus weaker than the single emission point in southwest America. For details please see the original work of Spiro et al., J. Geophys. Res. (1992). - We removed the two oldest references on the sulfur cycle "Feichter et al. (1996)" and "Feichter et al. (1997)" and added the more recent references "Berglen et al. (2004)" and "Koch et al. (2006)" instead. - In fact, the

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high SO4 concentrations in figure 1 are not at 100 hPa, but even at a higher altitude (maximum at the model top). SO4 in the stratosphere results from COS oxidation, which is followed by condensation and nucleation of H2SO4. Due to the large altitude, the small SO4 concentrations in the stratosphere are strongly amplified by converting to STP conditions shown in figure 1. Stratospheric SO4 does not contribute significantly to the modeled atmospheric sulfate burden.

Dust:

1. See response to "General Comments, 3."

2. We agree with the referee and reformulated the sentence: "As for all other aerosol compounds simulated, the mineral dust concentration south of 75S is very low. The calculated dust concentration does not exceed 0.01 micro-g/m3 at all heights."

Sea salt:

The referee is absolutely right, so the model is able to reproduce a well known feature of the global sea salt distribution.

Section 3.1 (general comment on selection of species):

We included discussions on OM and NH4 in the subsections focusing on BC and sulfate, respectively. We do not discuss NO3, since its contribution to the global aerosol budget is rather small.

Section 3.2:

1. We removed repetitions from the text and included a comment on the major differences between the composition of Aitken and accumulation mode.

2. Concentrations in section 3.2 and figure 5 have been converted to STP conditions.

3. The referee is right, we do not consider the formation of secondary organic aerosol explicitly. However, production of secondary organic aerosols is considered implicitly

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in a simplified manner assuming 99% of emitted mass attributed to secondary organic compounds condensing on pre-existing particles and 1% generating aged nucleation particles. This approach implicitly accounts for both, condensation and nucleation. For details we refer to section 2.2.2 (condensation) of the first part of our study.

Section 3.3:

1. Following the referee comment, section 3.3 has been shortened and repetitions have been removed.

2. We replaced every "lifetime" with "residence time" and displayed the results from all other model studies in a new table as suggested by the referee. The AeroCom results have been taken into account.

3. We added "in the accumulation mode" to "residence time of mineral dust".

Section 3.4:

1. Again, for comparisons with observations, we refer to section 3 of the first part of this study, which intensively compares model results to observations covering all relevant aerosol properties. Any repetition of these comparisons from part I would only lengthen part II unnecessarily. Part I showed vertically integrated total (sum of both submicrometer modes) mass concentrations only. In contrast, part II shows size resolved geographical distributions and vertical cross sections.

2. Please see response to referee comment on the introduction, 1. and on section 2.2, 1.

3. We agree with the referee and thus added the word "most" before "previous studies" in the sentence cited by the referee. For the reference of AeroCom, see response to referee comment on introduction, 1.

4. As for sulfate, the maximum (accumulation mode) number concentration is not located at 100 hPa, but at an even higher altitude. The COS oxidation in the stratosphere

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finally produces H2SO4. Because of the low particle surface area concentration at this altitude, some H2SO4 can nucleate forming new particles. In the model simulation, some of these particles can grow into the size-region of the accumulation mode, increasing the low number concentration in the accumulation mode at this altitude. However, the high number concentrations shown by figure 4 result in particular from the conversion to STP conditions at this high altitude (about 10 hPa). These are not relevant to the particle number concentrations simulated and analyzed in this study.

Section 3.5:

In many atmospheric processes not only the number concentration but also the surface and volume concentration are driving parameters. Therefore number, surface and volume size-distributions are presented in the figure. The discussion in the text focuses mainly on the number concentration since surface and volume concentration are simply derived from the number size-distribution. Changes in each of the size-distributions are related to the same processes. Thus any discussion beyond the integrated properties (total surface / volume) mentioned in the text would provide no new insights.

Section 4.1:

1. The first paragraph has been reordered as recommended, sources first followed by the sinks. The short summary at the end of section 4.1 has been removed.

2. The referee is right, SO4 is currently the only secondary aerosol in our model. Production of secondary organic aerosols is only considered implicitly in a highly simplified manner assuming 99% of emitted secondary organic carbon mass condensing on pre-existing particles and 1% generating aged nucleation particles. For details we refer to section 2.2.2 (condensation) of the first part of our study.

3. We changed the formulations according to the recommendations of the referee.

4. Intramodal coagulation reduces particle number concentration in the mode, whereas total particle mass concentration in the mode remains constant. This is not only true

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for the accumulation mode, but for intramodel coagulation in general. Thus, intramodal coagulation is a sink process for the particle number concentration. Since particle number concentration decreases in the mode and total particle mass in the mode is constant, the modal mean diameter increases. The details of intramodal coagulation in the model are documented in part 1 of this study (Lauer et al., 2005; section 2.2.3).

Section 4.2:

Again, we agree with the referee, that our results depend on the specific parameterizations used - as in every modeling study. Due to the high computational resources required by a climate model, we cannot do an ensemble of sensitivity studies varying all parameters because this is far beyond the capacity of any super computer. To at least partially circumvent this issue, we compare our results to numerous other model studies performed with other parameterizations and model configurations. This allows an estimation of the uncertainties associated with different modeling approaches and parameterizations applied. As recommended by the referee, we added references to AeroCom on the sources and sinks of sulfate.

Section 4.3:

1. We added more interpretation and a reference to the AeroCom results.

2. The term "transfer" has been replaced by "growth" (as used in figs. 8-10). This process refers to the transformation of particles from the Aitken into the accumulation mode due to continued growth. It is technically realized as described in part 1 of this study (Lauer et al. 2005; section 2.2.3).

Section 4.4:

1. We selected sulfate, because in terms of global burden, SO4 is the dominant submicrometer compound in our model simulation. In accordance with the comments of referee #4, we added a short discussion and a figure of the primary aerosol compound black carbon to section 4.4 to show the differences to the secondary aerosol compound

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sulfate.

2. As shown by figure 6, the particle number concentration in the Aitken mode decreases from the surface up to heights around 600 hPa, but then increases with height again due to particle formation due to nucleation. We now see, that our formulation was not precise and indeed was leading to wrong conclusions. Thus we reformulated this paragraph as follows: "Above this layer, intra- and intermodal coagulation effectively remove particles due to the high number concentrations resulting in a strong net depletion of particle number concentration in the Aitken mode. With increasing height this net depletion decreases as particle number concentration in the Aitken mode decreases up to altitudes around 600 hPa. Above 600 hPa, in particular in the upper troposphere and tropopause region, very effective formation of new Aitken mode particles by nucleation occurs. Just below this region of strong net production in the upper troposphere, newly formed Aitken particles are effectively removed by coagulation leading to a net depletion in the altitude range of about 300-400 hPa."

Regarding the high particle number concentration (STP conditions) in figure 6, please see response to comment on section 3.4, 4.

Figure 11: We decided not to use STP conditions in figure 11 to allow a comparison of different altitudes in terms of absolute contribution to the sources and sinks. The relevance of different geographical regions and altitudes for the change of the total budgets discussed in section 4.1-4.3 can be judged more easily as when STP conditions were used. However, the referee is right that this strategy complicates a comparison with figure 6. But since we think section 4.4 is more closely related to section 4.1-4.3 than to section 3, we rather chose ambient concentrations.

Sulfate:

1. Please see response to comment on section 4.3, 2.

2. The sulfate production over Antarctica is mainly caused by in-cloud oxidation of SO2

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during summer. The cloud formation in this area is driven by the strong meridional circulation (Rossby cell). SO2 is likely produced from DMS emitted from the ocean, in particular during Antarctic summer. We added a corresponding discussion to the manuscript. We think that the sulfate production in the middle troposphere over the tropics is not a significant feature and is therefore not discussed.

Conclusions:

1. We added a discussion on the limits of our model approach to the conclusions section (see also "General Comment, 2."). For a more detailed evaluation of the quality of our model, we refer to part 1 of this study (Lauer et al., 2005).

2. We would like to keep the 2nd paragraph of the conclusions since it discusses major features of the global distribution of aerosol number concentration in the different size modes. This information is one of the innovative features of our study.

3. The whole paper focuses on submicrometer aerosol, so do the conclusions. To be more precise, we added "of submicrometer particles" to this sentence.

Technical corrections:

In our opinion, it shouldn't matter whether British English or American English is used since no recommendation is given by the editorial office.

Section 3.2: Done.

Section 3.3: Done.

Section 3.4: We inserted the sentence to the 2nd paragraph as recommended by the referee.

Figure 7: Done.

Section 4:

1. We changed every occurrence of "at the global scale" to "on the global scale".

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2. Done.

3. Done.

Section 4.1:

1. As recommended by the referee, we changed "annual mean column changes" to "annual mean, vertically integrated changes in particle number".

2. Done.

Section 4.2:

1. We added "mass" to the title of section 4.2.

2. We removed the first sentence of section 4.2 as recommended. Again, in our opinion, it shouldn't matter whether British English or American English is used since no recommendation is given by the editorial office.

3. The second (now first) sentence of section 4.2 has been rewritten.

4. Done.

5. As stated by the referee, aerosol mass and in particular sulfate has been investigated by many previous model studies. Thus, we think it is an important information how the splitting between the two major sinks, dry and wet deposition, of ECHAM/MADE compares to other studies and should not be removed.

Section 4.3:

1. Done.

2. See above for sulfate mass.

Figure 10: Both vertical axis have been set to the same range.

Section 4.4:

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1. Done.

2. "mass" has been added to the subtitle.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 7519, 2006.