

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

Anonymous Referee #2

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The paper is the second of two related papers about the ECHAM/MADE model system. The ECHAM/MADE system is one of the few GCMs that predict both mass and number of the aerosol size distribution and that consider aerosol microphysics. The first paper describes the model and shows comparisons of simulations with observations of a multiannual simulation. This paper presents results from the same run but focuses on the contribution of individual processes on the life cycles of specific aerosol components and size modes, and on the particle number concentration. The process analysis gives new insights in the fate of aerosol in the atmosphere. It points out the importance of aerosol microphysics when simulating particle numbers, which are important for the radiative aerosol impacts and their effects on clouds and precipitation.

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General comments

The paper should be shortened and more focused. New results of the study should be emphasized and the title should be changed to be more attractive and meaningful, see suggestions below. I think the paper would be more interesting if section 3.1 including figures 1 to 4 that discuss aerosol mass were removed. Aerosol mass has already been treated in part I of the ECHAM/MADE papers, and the additional separation into Aitken and accumulation mode in part II is not very relevant as the contribution of the Aitken mode to the aerosol mass is insignificant for most aerosols. Furthermore, comparisons to observations are missing, and mass distributions have been extensively discussed in many previous papers. (Despite this recommendation some comments on section 3.1. can be found below.)

The aerosol size distributions are represented as two log-normal functions with constant sigma, i.e. the model is based on a two-moment modal, bulk microphysical scheme. This is a widely used technique for modelling global aerosol simulations, but still a coarse simplification of the complex aerosol size distributions. The limitations of such an approach for a process analysis should be discussed, and potential (dis-)advantages when comparing to an explicit (bin) scheme with higher number of bins should be considered. In addition, a comment on the effect of assuming only internal mixing is missing.

Aerosol microphysical processes are strongly non-linear and I doubt that the coarse mode can be simply neglected, especially in a processes study. Please give a broader discussion on the potential effects of not considering the coarse mode. The nucleation mode has also been neglected, and the authors relate the under-estimation of the Aitken mode particle size in case of strong nucleation to this short coming. One result of this study is that nucleation is the major particle number source, but at the same time it is this same process that is not well represented in the model. Please comment.

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The results presented here depend to a certain - but unknown - degree on their specific parameterization in the ECHAM/MADE system. The importance of individual processes is also determined by their representation in the model in MADE and depends on the boundary conditions provided by ECHAM. It would be interesting to know how robust the results are. Sensitivity studies with modified parameterisations or at least a discussion should be added showing that the authors are aware of this problem.

One more comment on the term 'aerosol dynamics': Although widely used in the community I find this term misleading because the processes the authors refer to do not have a strong relation to the dynamics (i.e., particle motion), but to the microphysics of aerosol. I would strongly recommend substituting 'dynamics' with '(micro-)physics'.

Specific comments

Title

The title should be more interesting and related to the new scientific results of the paper, something like 'The importance of individual aerosol microphysics processes for the global aerosol particle concentration'.

Abstract

The abstract should be modified. The residence times and burdens (why is NO₃ missing?) are not the main topic of the paper, and mass distributions have already been discussed in part I and elsewhere. Please change the abstract emphasizing the most interesting results of your paper (composition of Aitken and acc. mode, change with height, process analysis, size distribution, etc.). The abstract shortly summarizes the paper, but also serves to attract the interest of the reader!

Introduction, 3rd paragraph

'Most current global climate models include aerosols in the form of prescribed climatologies or treat aerosol mass only (e.g. Roeckner et al., 1996, 2003; Feichter et al., 1996; Lohmann et al., 1999a; Adams et al., 1999).'

Not true, there are several models, see AEROCOM

Introduction, 3rd paragraph / Section 2 Model description

What is the difference between the presented ECHAM4/MADE system and the ECHAM5/M7 system? Please give a very short idea.

Introduction, 3rd paragraph

'The new model system ECHAM4/MADE is able to calculate particle number concentration and the aerosol size-distribution explicitly in addition to the mass concentrations of the aerosol components sulfate (SO₄), ammonium (NH₄), nitrate (NO₃), black carbon (BC), organic matter (OM), sea salt, mineral dust and aerosol liquid water (H₂O).'

Not true, the presented model is a two-moment bulk microphysical scheme; hence the prediction of the size distribution is not explicit as it would be for a bin scheme.

Section 2.1 ECHAM: this section should be removed, referring to part I is enough.

Section 2.2 MADE

It is misleading to write three-moment scheme if in fact a two-moment scheme is applied. You use a two-moment scheme that has the capability to be run in a three-moment mode. In addition, it is not true that the particle size is 'explicitly calculated' as a bulk scheme with only 2 moments is used. Please correct throughout the paper.

Section 2.2 MADE

Another concern is that coarse aerosols have been neglected. The authors state: 'This is justified by the typically rather little interaction between fine and coarse particle size-ranges (Whitby, 1978)'. Coarse particles are mostly produced by mechanical processes, whereas fine particles are usually produced by combustion, condensation, or nucleation processes.

But this is not true for dust and sea salt which also include a fine aerosol fraction - which is simulated in this paper. In addition, coarse aerosols of these two species are transported inland where they interact with fine aerosol.

Furthermore, the time scale of transferring mass from fine to coarse mode by coagulation is typically in the order of weeks whereas the usual lifetime of fine particles is only in the order of days.

This might be true for intramodal coagulation, but does it also refer to intermodal coag.? I believe that the latter process is very fast and leads to efficient coagulation. Please comment.

Section 2.2 MADE

Do you consider aging of BC?

Section 3

The GCM has been used in a CTM mode, as the climate was not predicted by the GCM. This should be clearly said in the paper.

Section 3.1. (should be entirely removed, see above!)

Remove repetitions.

How about AEROCOM results?

Why do you normalize your vertical concentrations to STP conditions? Simply using a height-independent quantity like the mass mixing ratio would be more straight-forward.

Sulphate

What is the life time of SO₂? It determines the spatial source distribution of SO₄.

If you have strong volcanic S emissions in South-West America, why not in Indonesia?

Did GEIA not include those?

Please cite more recent literature on the sulphur cycle.

Please comment on the high SO₄ conc. at 100 hPa.

Dust

Again, I wonder what the effect of neglecting the coarse mode is.

Dust, 2nd paragraph

The concentration south of 75 S is very low for all components, not only for dust.

Sea salt, 4th line

‘the mid-latitudes of the southern hemisphere.’ But this is a well-known feature.

Why did you not discuss OM, NO₃, and NH₄? The latter two species are not well examined by models; this would be a new, interesting topic.

Section 3.2.

Please shorten text and remove repetitions. Rather comment on the difference between the Aitken and the acc. mode.

Section 3.2. , 1st paragraph

‘The average mass concentration of accumulation mode particles decreases from 4 956;g/m³ in the lowermost level to 73 ng/m³ (ambient concentrations, i.e. not STP) at 250 hPa.’

It would be interesting to know if the concentration decreases also when you normalize to STP conditions, as this decrease could be attributed to microphysical processes.

Section 3.2. , 1st paragraph

You did not consider the formation of secondary organic carbon, although this is an important source of particle numbers (and mass). Please comment!

Section 3.3.

Section should be shortened and re-ordered, repetitions removed. What is new in comparison to other studies? If nothing is new, it is enough just to refer to these studies.

Section 3.3

Please refer to the results of AEROCOM. Display the burdens and life times from other studies in a table. (Better write residence time than life time, see definition in Seinfeld Pandis book.)

Section 3.3. comparison to Tegen & Lacis paper.

I am not sure if the comparison is justified because of the non-linearity of the processes involved; see my comment on neglecting the coarse mode above. The life time of dust is extremely long, at least you should write 'residence time of dust in the acc. mode'.

Section 3.4.

Section should be shortened and re-ordered, repetitions removed (e.g. particles emitted at the surface above continents is always important). Comparison to observations is missing, these are only model results, how relevant/realistic are they? In addition, it seems to me that some of the results appeared already in part I.

Section 3.4.

'In contrast to most current climate models, ECHAM4/MADE calculates particle number concentration and particle size-distribution explicitly.' Not true, see comment above.

Section 3.4.

'This allows a more detailed view on the global aerosol characteristics than provided by previous studies, in which particle number concentration had to be calculated diagnostically using prescribed size distributions.' Not true, there are a few other models, see AEROCOM.

Section 3.4.

How about the maximum particle conc. at 100 hPa?

Section 3.5.

Why do you show 3 moments of the distributions in your figures? If you show them all, they should all be discussed. How relevant are these model results? Comparison to observations???

Section 4

Section should be shortened and re-ordered, repetitions removed. AEROCOM should be mentioned.

4.1. Particle number

The first paragraph would probably be easier to read if it was reordered: 1) sources, 2) sinks The whole section should be condensed, a summary of three paragraphs is not necessary.

4.1. Particle number 1st paragraph

Only in the model is SO₄ the only secondary aerosol, because sec. organic carbon is ignored.

4.1. Particle number 2nd paragraph

might be overestimated at all -> might be generally overestimated in the model ?
negligible small -> negligibly small

4.1. Particle number 3rd paragraph

How can intramodal coagulation in the accum. mode be a sink for the accum. model?
What happens to the particles from intramodal coagulation in the accum. mode, are they removed from the model? I thought this process would only increase the mode median size.

4.2. Sulfate

Again, my general concern: How much are these results dependent on the specific model parameterisations? Please add a comment in the paper. Also refer to the results of AEROCOM when discussing the sources and sinks of sulphate.

4.3. Black Carbon

This paragraph should focus on and interpret the differences and similarities between SO₄ and BC, and not just describe the results. Please rewrite

4.3. Black Carbon 1st paragraph

‘transfer of Aitken particles into the accumulation mode’

What exactly do you mean with ‘transfer’? Do you refer to ‘mode merging’ introduced in part I? Is it an artificial model process which has to be introduced because of the modal

representation of the particle sizes? Please add an explanation, possibly already in section 2.2.

4.4. Net production/-depletion

Why did you select SO₄?

4.4. Net production/-depletion 1st paragraph

How can the decreasing particle number conc. explain the decreased depletion with increasing height, but at the same time you find efficient coagulation even higher, just below region of nucleation, where the particle conc. is even smaller? Or is it due to sedimentation of particles from the nucleation region (but these have an extremely low fall velocity)? Please discuss and explain figure 11 together with figure 6 (more than just saying 'see also figure 6'). E.g., how can there be a maximum in acc. mode particle numbers at 100hPa in fig 6 if there is no strong source in fig 11?

Fig 11

Why do you not use STP units? I think this would be very useful for the comparison between figures 6 and 11. Maybe it would be less advantageous to show the decrease in particle number with height, but a comment in the text would be nice.

Sulfate

Again, what does 'transfer' mean?

Sulfate

Please add a discussion on the irregular pattern of fig 12, especially for the acc mode. Why is there production of sulphate mass above Antarctica and in the southern hemisphere tropics?

Conclusions

Again the description of the model is too positive and misleading (no explicit calculation of aerosol sizes etc.) Please do also mention the constraints of your mode.

Conclusions

The conclusions should be more focused and highlight only the new scientific findings of the study. They could be much more fascinating to read! The second paragraph could be replaced by just saying that the study corroborates previous studies. The higher importance of long range transport for the acc. mode is also well known (this is why this mode is called accumulation mode).

Conclusions

Also mineral dust and sea salt particles have an important contribution to the aerosol mass loading. What do you mean? These two components dominate the global aerosol mass.

Technical corrections

Throughout the paper

I am not sure if the use of the word 'nevertheless' is correct, I would rather expect in most of the cases where it is employed the word 'however' instead, but I am not a native speaker. ACP is a European journal, should the spelling not be European? e.g. Sulfate-> Sulphate ?

Section 3.2. , 1st paragraph

lowermost level -> lowermost model level

Section 3.3.

Rewrite equation 1: $\text{lifetimes}[\text{days}] = \text{burden}[\text{Tg}] / \text{SUM}(\text{sources} [\text{Tg/day}])$, i.e., change unit of sources and remove the factor 'number of days'

Section 3.4. 2nd paragraph

There are no significant sources of primary particles at the surface south of 30 S other than sea salt.

Figure 7

Please explain the blue and green lines in the figure caption.

Section 4

at the global scale or on the global scale? (throughout the paper)

Section 4

remove parenthesis

(Adams and Seinfeld, 2002) -> by Adams and Seinfeld, 2002

Section 4

Correct last sentence

Budgets of particle number concentrations as well as of the masses of sulfate (secondary aerosol) and black carbon (exemplary for primary aerosol) are investigated.

4.1. Particle number 1st paragraph

annual mean column changes -> annual mean, vertically integrated changes in particle number ?

4.1. Particle number 4th paragraph

(surface)-> especially at the surface

4.2. Sulfate

Title should be changed to Sulphate mass

4.2. Sulfate

1st sentence can be removed, repetition from section 4. Sulfate-> Sulphate ? ACP is a European journal, should the spelling not be European?

4.2. Sulfate 2nd sentence

further processes are analysed, not introduced

4.2. Sulfate 2nd paragraph

negligible small -> negligibly small

4.2. Sulfate 3rd paragraph

Remove the numbers for dry deposition in the text and just write one sentence saying

that dry deposition is the other sink process.

4.3. Black Carbon

Title should be changed to Black Carbon mass

4.3. Black Carbon 1st paragraph

Remove the numbers for dry deposition in the text and just write one sentence saying that the dry deposition is the other sink process.

Figure 10

Use the same range for vertical axes of the Aitken and acc. modes?

4.4. Net production/-depletion 1st sentence

Change to plural: distributions, change rates are analyzed

Sulfate

Subtitle should be changed to Sulphate mass

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