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Interactive comment on "The aerosol-climate model ECHAM5-HAM" *by* P. Stier et al.

P. Stier et al.

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Response to the reviewers

We would like to thank the reviewers for their helpful comments that substantially improved the manuscript. We hope to have addressed all raised issues.

Response to comments of reviewer number 1

1. The major deficiency of the manuscript is that it does not discuss the sensitivity of the results to the chosen model parameters...

In general we agree that this manuscript does not discuss all sensitivities of the global results to the model parameters. The reasons for this are twofold. First, this manuscript is only the first of a series of papers on the ECHAM5-HAM aerosol model and many model sensitivities are being addressed in forthcom-

ing publications, such as a manuscript on the internal dynamics of the microphysical aerosol system submitted to Journal of Climate. Second, as many model parameters are non-linearly interdependent, changes in individual parameters do not necessarily reflect the uncertainty of the model to the uncertainty of the input parameters. A Monte-Carlo like approach appears desirable, however, it would be computationally too demanding in the global model. Nonetheless, an extensive effort was made to test the used parameterisations already in offline box-model studies in which cause and effect can be well constrained. In this context we want to emphasise that many of the used parameterisations have already been tested and published as cited and want to point out the extensive sensitivity studies with the microphysical core M7 described in the Vignati et al. (2004) paper. We carefully went through the paper again and clarified the unclear sections. As for example reviewer 2 has also objected section 2.3.2, we clarified the underlying assumptions and referred explicitly to the AEROCOM emission inventory on which most of the assumptions are based.

We apologise for an error found in the description of the emission size distributions for fossil fuels and wildfires. For fossil fuels it was erroneously stated that the number median emission radius r = 0.015 um and for wildfires r = 0.04 um. However, as the AEROCOM recommendation of r was based on a standard deviation of 1.8, we adapted that to fit the ECHAM5-HAM standard deviation of 1.59 for the respective modes. Therefore, although otherwise quoted in the manuscript, we correctly applied emission radii of r=0.03 um for fossil-fuel use emissions and r=0.075 um for wildfire emissions in the simulations. The manuscript has been corrected accordingly.

2. The description of the aerosol size distribution relies on the modal approach with a fixed standard deviation for each mode. Under many atmospheric con-

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ditions, this approach is, however, notably inferior to a modal approach with varying standard deviation...

First, we agree that more complex schemes have generally the potential to be superior to the used two-moment approach. In terms of accuracy, a high-resolution sectional model without a prescribed functional form is again potentially superior to a three moment modal approach. However, for the application in climate modelling, a balance in complexity among the simulated processes needs to be maintained. Based on these considerations, for the prognostic treatment of aerosol size-distribution and the mixing state, the chosen two-moment approach appears to be a good compromise between detail and computational efficiency for current global aerosol climate modelling. Second, while Zhang et al. (1999) show that the three moment modal approach is superior to simulate the aerosol evolution under certain conditions, they do not consider the cases of concurrent aerosol formation and emissions. To our knowledge, three moment modal aerosol schemes can have the tendency to unrealistically increase the standard deviation of the modes to account for these processes. Therefore, the superiority of three moment aerosol schemes under realistic atmospheric conditions is ambiguous.

3.The treatment of organic aerosol is not fully clear to me...

In fact, the description of the treatment of organic aerosols was not very detailed in the original manuscript. We clarified that in section 2.3.2 and added further references. Solubility in this context refers to the common classification as water-soluble organic carbon and is approximated in the model by the inclusion in the soluble modes.

4. The model distributes the sulfate produced in the aqueous phase only to accumulation and coarse modes...

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We agree that this is a limitation of the model version described in this manuscript. However, this issue can only be addressed if the fraction of Aitken mode particles in the water phase but also the cloud microphysics is well defined. This requires the application of an explicit size-dependent activation parameterisation as well as explicit treatment of the uptake of aerosols into the water phase by aerosol / hydrometeor collision processes. Further, it requires a cloud scheme with prognostic treatment of CDNC as not only the chemistry but also the in-cloud collision / coalescence processes affect the size-distribution or re-evaporated aerosol. These processes are included in the in the outlook mentioned refined model version including an explicit aerosol-cloud coupling and will be published in a future manuscript. For the current manuscript we added a sentence to section 2.4 clarifying the current in-cloud aerosol processing procedure and its limitations.

5. To which mode are the particles produced in a collision of an insoluble particle with a smaller soluble particle placed?

They are placed in the soluble mode corresponding to their own size class. We clarified this by changing on page 5570 line 6 in this case the particles are transferred to the soluble/mixed mode into in this case the particles are transferred to the corresponding soluble/mixed mode.

Is the intra-modal coagulation only omitted for the insoluble dust modes?

Yes, this is the case and based on two considerations. First, with the low coagulation rates and the low number concentrations in the accumulation and coarse modes the coagulation is of negligible efficiency. Second, by this we implicitly take into account the low expected sticking coefficient for two non-wetted dust particles. We reordered the sentence of line 11 on page 5570 to emphasise that this term is only omitted for the insoluble accumulation and coarse modes.

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6. One possible reason for the under prediction of the aerosol number in the lower boundary layer is the nucleation mechanism used.

This is a good point. We added the explanation an underestimation of nucleation to the explanation in line 6 of page 5580. The point is further discussed in the now included paragraph on the surface aerosol number size-distributions in section 2.4.

7. Titles and legends in some of the figures are very small and difficult to read.

We fully agree that the figures, titles and legends in the ACPD format appear too small. This is because the manuscript has been formatted for publication in ACP and the figures have been reduced in size for ACPD by the production office. In the ACP publication style the figures and captions should appear in a well readable size.

Technical corrections:

1. Throughout the text: replace "Hereby is x" with "Here x is"

Done.

2. Throughout the text: replace expressions of the form "after the condensation avail-able sulfate" with the form "sulfate available after the condensation"

Done.

3. replace relaxate with relax

Done.

4. To help the reader, the assumption of 4 soluble and 3 insoluble modes should be written out in the text with a brief motivation of this mode structure chosen.

Excellent point. We added a paragraph to section 2.2.

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5. Correct the parenthesis in the reference

Done

6. Replace "allowed to be condensated or nucleated" with "allowed to condense or nucleate"

Done.

7. The word tracer is printed twice

Removed.

8. Correct the parentheses in the reference

No error found.

9. The first two sentences of the section are confusing if presented here. They should be moved to section 2.7.5 where the omitted coagulation processes are explicitly discussed.

Good point. Actually, we consider these sentences as redundant and omitted them.

10. Replace "is condensed" with "condenses" and "are wet deposited" with "is wet deposited / is removed by wet deposition"

Done.

11. Replace evaluation with evaluation of

Done.

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Responses to comments of reviewer number 2

1. There are, of course, many assumptions made in choosing the model parameters. The paper often does a poor job of justifying them or referencing them...

Please refer to the response to the similar first remark of reviewer 1.

2. The processing of aerosol in clouds is not well described...

A clarifying comment has been added to section 2.4. We agree that the cloud processing in this model version is not as advanced as we would like it to be. An improved cloud-processing scheme is subject of ongoing research activities and will be described in future publications. (See response to the similar remark 4 of the first reviewer.)

As described, the SO2 oxidation appears not to be ever H2O2 limited.

The SO2 oxidation in the chemistry module of ECHAM5-HAM is certainly H2O2 limited. However, to leave more space for the discussion of the results, we still believe that a too detailed description of the details of the chemical mechanism would be beyond the scope of this paper. The interested reader is referred to the to the cited paper of Feichter et al. (1996) that describes the sulfur chemistry scheme in detail.

3. The treatment of the competition between nucleation and condensation gives cause for concern...

This is a very valid remark. Actually, the system involves the source of gas phase sulfuric acid as third parameter that should be solved instantaneously. However, the complexity of the simulated system sometimes requires the application of operator splitting. The treatment of the sulfuric acid formation before condensation, in turn before nucleation, does not necessarily constitute

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an underestimation of the nucleation as in reality a part of the produced sulfate will be depleted by condensation before threshold concentrations for the nucleation can be reached. Both behaviours could be observed in box-model sensitivity studies with the microphysical core M7 in which the timestep was varied from 1s over 100s to 1000s. However, the effects on the Aitken and accumulation modes were under most conditions negligible. A more advanced parameterisation, including the parameterisation of apparent nucleation rates including the competition between condensation and nucleation is subject of ongoing research activities. For this model version the current operator splitting order appears to be the most appropriate as it provides the best agreement with observations that generally show an absence of nucleation in presence of sufficient available surface area.

We have removed the second sentence of section 2.7.2 according to your suggestion.

4. My most serious criticism of the paper is the lack of evaluation of the new features of the model; namely the size distribution and number concentrations...

Excellent point that was planned to be addressed in a forthcoming publication. However, we now decided to extend the section 2.4 by a part on aerosol number size-distributions and their evaluation and added a paragraph in the discussion section. However, we want to point out that the acquisition of aerosol size-distribution measurements suitable for model evaluation is a demanding task, as they have to be either derived from long-term measurements with climatological relevance or (preferentially) performed in the simulated period. Unfortunately, for the year 2000 for which the simulations were performed and for which the emission data is representative, to our knowledge no suitable measurement compilation is available. So we decided to use as a first step a compilation of measurements published as European Aerosol Phenomenology

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(Putaud et al., 2003) that provided us with long-term median size-distributions representative for the period 1996-2001. In addition, we would like to point out the presented comparison of the simulated Angstroem parameter with satellite observations also provides an evaluation of aerosol size on the global scale. It has to be mentioned again that this manuscript is only the first paper describing the ECHAM5-HAM model. Further evaluations will be published in future manuscripts with increasing availability of observational data.

The number concentrations are evaluated against your own (as yet unpublished) CN concentrations

Unfortunately, a misunderstanding led to an incorrect citation of the measured CN concentrations. In fact, the measurements of the INCA measurement campaign have already been published in the Geophysical Research Letters (Minikin et al., 2003). We corrected that citation in the manuscript.

Technical corrections:

1. What is meant by long-term simulations?

We added the following explanation after the first usage of long-term. For space constraints we kept using long-term throughout the manuscript.

"long-term, i.e. depending on the model resolution centennial to millennial scale, transient climate simulations."

2. The term interactive approach is used several times. What do you mean by it?

Good point, as this term has not been used consistently and was therefore confusing. We removed the usage in the sentence Adam and Seinfeld (5554, line 11) so that it is used only to describe aerosol models in which different aerosol components are allowed to interact by microphysical processes. The

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second usage is for the description of the explicitly calculated processes, such as dry deposition or certain emissions. However, as these processes are also parameterised, explicit does not seem to be appropriate and we stick to the usage of interactive.

3. You say that the aerosol size distribution is predicted explicitly. This isn't strictly true

In fact this is not strictly true. We changed "predicted explicitly" into "prognostic parameters"

4. What is meant by soluble - in water? Non-solid? Miscible?

In this manuscript "soluble" refers to water-soluble. We added an explanation to the first occurrence of soluble.

5. What is relative composition? Relative to what?

Changed into composition

6. Physical assumption. Do you mean physically realistic?

Changed into realistic.

7. I don't understand the meaning of the sentence beginning...

We changed the beginning of the sentence into "The prognostic treatment of the aerosol size-distribution" to clarify this.

8. It sounds like the sedimentation speed is restricted to prevent numerical instability. Don't you mean that the timestep is restricted?

No, in fact the sedimentation velocity is restricted as sub time-stepping is undesirable for optimisation reasons. However, this criterion is only necessary to prevent spurious instabilities. For the considered aerosol size-range the sedi-

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mentation velocity is generally much lower than the vertical grid velocity (grid velocity dz/dt is around 100m/(15min)=11 cm s-1).

9. Its not clear what is meant by inter-modal coagulation being treated as operator-split. I don't see that this is described in the next section. What is meant here? It seems to be important.

The treatment of the inter-modal coagulation between insoluble and soluble modes as operator split refers to the fact that the inter-modal coagulation between insoluble and soluble particles is treated concurrent with the transfer of insoluble particles by absorption of sulfuric acid.

10. You talk about the dominating processes of coagulation being simulated. What are these?

The intention of this sentence was to highlight that the coagulation is limited to Brownian coagulation and that i.e. the intra modal coagulation of the insoluble dust modes is neglected. However, as these points are also described in the text, we consider these sentences as redundant and omitted them.

11. Sentence starting "The total condensable sulfate..." is unclear

We have the feeling that the numerical details of this procedure are beyond the scope of this manuscript but refer the interested reader to the Vignati et al. (2004) paper referenced in this section.

How do you justify this assumption?

This assumption has been derived in a series of sensitivity studies with the M7 box-model (see Vignati et al., 2004). Nonetheless, we acknowledge that it is of high uncertainty and pointed this out in the manuscript. We hope that future laboratory studies can help to define the amount of sulfate required to significantly increase the hygroscopicity of insoluble aerosol particles.

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What do you do about mixed sea salt/sulfate?

Sea salt is emitted a priori in the soluble modes and therefore does not undergo the transfer from the insoluble to the soluble modes. Nonetheless, sulfate is condensing also on the soluble modes, including sea salt.

12. I suggest to replot the number concentration as observable CN...

While the display of the number concentration as observable CN would have its advantages, the current separate display of the modes has the advantage to highlight the relative contribution of the different soluble and insoluble modes. Therefore, we decided to leave the plot unchanged. To a first order, observable CN can be estimated by eye as sum of the Aitken, accumulation, and coarse modes.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 5551, 2004.