Reply to Anonymous Referee #1

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We thank the referee for his constructive comments that helped to improve the manuscript. Our reply to the review is listed below.

Impacts of different parameterization changes in HAM2 on aerosol- and climate-related results from ECHAM are analysed. The basic approach in the paper is useful given large uncertainties that still exist for the representation of aerosol effects in climate models.

Unfortunately, the discussion of some features of HAM2 in the paper is confusing. Several parameterizations are emphasized as important model improvements in the abstract and elsewhere. After reading descriptions of these parameterizations and analyzing model results the reader is informed that these parameterizations are not actually used in HAM2. This includes the cluster and kinetic nucleation schemes in the boundary layer (p. 7553) and various changes for aerosol wet deposition (p. 7561).

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Model components that are not used in HAM2, or which are still under development, can be better described in separate publications with a specific focus on certain model shortcomings and strategies for improvement. This would permit a more complete and convincing analysis of these parameterizations.

We agree that the description of new model features can be structurally improved. In the revised manuscript, the old Section 4 ("Model updates and their effects") is separated into two parts:

- Standard configuration of HAM2, and
- Alternative configurations.

The first category contains the major part of the old Section 4, while the second one includes Section 4.6 of the discussion paper and contents from Section 4.1 about boundary layer nucleation, as well as explanations why these schemes are switched off in the standard model.

We prefer to keep the second bullet above in the paper even though these components are not part of the standard model configuration, because i) they describe parameterizations that are of importance for scientific process studies (see, e.g., Makkonen et al., 2009, Kazil et al. 2010), and ii) we feel that such information can help to provide the model users with a clear idea about the basis for future research.

Reference:

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

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

Finally, a different activation scheme is apparently used in HAM2 instead of the scheme which is described in the paper (Lin and Leaitch, p. 7563). No further information about this aspect of the model is provided although an important purpose of HAM2 is to provide improved modelling capabilities for climate.

It seems that wording in the last paragraph of Section 4.7 in the discussion paper has caused some misunderstanding. The default activation scheme in HAM2 is indeed that of Lin and Leaitch (1997). The implementation and evaluation of a Köhler-theory-based scheme will be presented in a separate paper by Stier et al. (In preparation). During the revision we have re-written the paragraph and moved it to the end (outlook part) of the paper.

"Furthermore, the aerosol activation scheme of Lin and Leaitch (1997), currently used in HAM2, is highly simplified in terms of particle size and composition effects. Stier et al. have recently implemented the Köhler theory based scheme of Abdul-Razzak and Ghan (2000). The evaluation will be presented in a separate paper."

References:

Lin, H. and Leaitch, W. R. (1997): Development of an in-cloud aerosol activation parameterization for climate modeling. In: WMO Workshop on Measurement of Cloud Properties for Forecasts of Weather, Air Quality and Climate, pp. 328–355, Geneva, Switzerland, World Meteorology Organization

Abdul-Razzak, H. and Ghan, S. J. (2000): A parameterization of aerosol activation - 2. multiple aerosol types, J. Geophys. Res., 105, 6837–6844, doi:10.1029/1999JD901161

On the other hand, it would be useful to include additional results for particulate organic matter in the paper. An interesting aspect of HAM2 is the replacement of the highly idealized treatment of organics in HAM1 by parameterizations for chemical and microphysical processes. Does this modification improve the agreement between simulated and observed concentrations for organic matter?

Following Heald et al. (2011) we compared the vertical profiles of total POM (POA + SOA) mass concentration from HAM1 and HAM2, sampled in the month-of-year and location of 17 field campaigns. Consistent with Fig. 3 in the discussion paper (annual and zonal mean cross-sections), increases in POM concentration are seen in HAM2 in the lower troposphere. Despite such increases, the model still generally underestimates organic mass compared to the observations in Fig. 3 of Heald et al., 2011. Similar results have been obtained earlier by O'Donnell et al. (2011) for surface POM concentrations in the United States and Europe. Such underestimation is also seen in the GEOS-Chem model evaluated by Heald et al. (2011, again see Fig. 3 therein), a model that has an explicit treatment for the semi-volatile SOA.

<u>Reference:</u>

Heald, C. L., Coe, H., Jimenez, J. L., Weber, R. J., Bahreini, R., Middlebrook, A. M., Russell, L. M., Jolleys, M., Fu, T.-M., Al- lan, J. D., Bower, K. N., Capes, G., Crosier, J., Morgan, W. T., Robinson, N. H., Williams, P. I., Cubison, M. J., DeCarlo, P. F., and Dunlea, E. J.: Exploring the vertical profile of atmospheric organic aerosol: comparing 17 aircraft field campaigns with a global model, Atmos. Chem. Phys., 11, 12 673–12 696, 2011.

The discussion of model results in the paper generally lacks quantitative information. The analysis of impacts of parameterizations on model results is limited to a description of simulated concentration patterns etc. It is often difficult to decide whether a new parameterization leads to actual improvements in model results from the results that are shown in the paper.

In order to present a more quantitative evaluation of the model, we added to the revised manuscript

a. A figure showing zonal mean cross-sections of aerosol water content in various sensitivity experiments, together with the global mean values, total AOD and total water AOD;
b. Global mean values and the RMS differences in Fig. 6 of the discussion paper;
c. For Figs. 10 and 11 of the discussion paper, a new figure comparing the simulated and observed vertical CN files in selected regions;
d. For Figs. 12 and 13, a table comparing the simulated and observed aerosol number concentrations at the sites.
e. For Figs. 15 and 16, the observed and simulated meridional distributions of AOD and AAOD, and a new figure showing Taylor diagrams that compare the observed and simulated AOD, Ångström parameter and AAOD in different regions;
f. For Fig. 19, a table that compares AOD of each individual species, the total AOD, absorption AOD and single scattering albedo in HAM1 and HAM2, over land, over the ocean, and over the global domain;
g. In the last subsection before conclusions, two tables showing the global mean radiative forcing by anthropogenic aerosols.

As explained in more detail in the following, the model overview is lacking detail. This is in contrast to the expectation of a synthesis paper according to title and abstract. Individual - sometimes developmental - components of HAM2 have been described in various other papers and so a sufficiently complete and detailed summary needs to be provided in this paper.

In the revised manuscript we have extended Section 2 (Model overview) substantially to include an overview of the main components of ECHAM5 and detailed summaries of various aspects of the HAM module. The latter includes 1) basic assumptions of aerosol composition and size distribution, 2) emissions of aerosols and their precursors, 3) sulfur chemistry, 4) aerosol microphysics, 5) removal processes, 6) calculation of aerosol optical properties and radiative effects, and 7) aerosol indirect effects.

How is HAM implemented in ECHAM? What is the horizontal, vertical, and temporal resolution of the model? The model time step is important for the results (e.g., p. 7555, l. 5). How are aerosol tracers affected by convection and other mixing processes in ECHAM?

In the revised manuscript, three paragraphs are added at the beginning of Section 2 (Model overview) to introduce the ECHAM5 model, describe its resolution, and summarize its connection to HAM.

It is clarified that the large-scale, convective, and turbulent transport of aerosols and their precursors are handled in the same way as other passive tracers in the host model (e.g., water vapor and hydrometeors). The large-scale tracer transport is represented by a flux-form semi-Langrangian algorithm of Lin and Rood (1996) with piecewise parabolic sub-grid distribution. Cumulus convection and convective tracer transport are parameterized with the mass-flux scheme of Tiedtke (1989), with further modifications by Nordeng (1994). Turbulent transport of momentum, heat, moisture and passive tracers is described by the eddy diffusivity theory (Louis, 1979), with the vertical diffusion coefficient computed as functions of atmospheric stability and the turbulent kinetic energy (Brinkop and Roeckner, 1995).

The tropospheric version of ECHAM5 is most often used at T63 resolution (approximately 2 degrees latitude × 2 degrees longitude grid spacing), with 31 vertical levels up to 10 hPa and a default time step of 12 min. This is also the configuration used in the paper. Most of the AeroCom Phase I models (Textor et al., 2006) used resolutions between 1.1×1.1 to 5×4, except ULAQ which was 22.5×10. The T63 resolution we are using in this study is similar to the GOCART (2.5×2.0, Chin et al., 2000), MATCH (1.9×1.9, Barth et al., 2000), MOZGN (1.9×1.9, Tie et al., 2001), UMI (2.5×2, Liu and Penner, 2002), and PNNL (2.5×2.0, Easter et al., 2004) models. These are mentioned in the revised manuscript.

Basic assumptions about aerosol chemical composition and size in HAM need to be clarified at the beginning of section 2. It is mentioned that particles are internally and externally mixed but this is not explained. What exactly are the mixing assumptions that are made for different types of aerosol? How many tracers are advected in the model and what is the numerical approach for tracer transport? Perhaps Fig. 1 should be included here and used for the description of the basic approach? According to Fig. 1, aerosol species can be soluble or insoluble. This needs to be explained. How are these defined and what are the relationships between these?

The aerosol module HAM predicts the evolution of an aerosol ensemble of five compositions: sulfate (SU), black carbon (BC), particulate organic matter (POM), sea salt (SS), and mineral dust (DU). The size distribution of this aerosol population is described by seven log-normal modes with prescribed variance. A schematic is shown in Fig. 1 in the revised manuscript. The detailed mathematical formulation can be found in Stier et al. (2005, Eqn. (1) therein). Different compositions within a mode are assumed as internally mixed, meaning that each particle may consist of multiple compositions. Aerosols of different modes are externally mixed, meaning that they co-exist in the atmosphere as independent particles. Four of the modes contain at least one soluble composition, thus the particles can take up water. These are referred to as soluble modes. The other three modes consist of insoluble species only. Through the aging processes, insoluble particles can become soluble (explained in Section 2.4 of the revised manuscript).

The HAM module predicts the particle number concentration of each mode, as well as the mass concentration of the compositions present in that mode. Not counting the secondary organic aerosol

(SOA), there are 25 aerosol tracers (18 aerosol mass plus 7 aerosol number) in the model that are affected by large-scale, convective and turbulent transport. The 3 precursor gases of sulfate, namely SO2, dimethyl sulfide and sulfuric acid gas, are also transported. In HAM1 SOA was considered to have the same properties as POA (primary organic aerosol). They are denoted collectively by OA in the revised Fig. 1, implying there is no additional tracer for SOA. In HAM2, different SOA species are distinguished according to their sources (anthropogenic, isoprene-derived and terpene-derived). The actual number of SOA-related tracers (including precursors and semi- and non-volatile secondary organics) depends on the lumping assumption. In this study we have 13 transported tracers in the SOA module.

The SOA module introduces 13 or 21 new tracers to the model that need to be transported, depending on whether the xylene-, toluene- and benzene-derived SOA is lumped together. This include 5 precursor gases (isoprene, monoterpenes, xylene, toluene and benzene), 4 condensable organics from the oxidation of isoprene and monoterpenes (i.e., 2 precursors × 2 products each, before gas-aerosol partitioning), and 1 (lumped) or 3 (not lumped) anthropogenic SOA mass concentrations in each of the mode that contains organic matter (cf. revised Fig. 1)

These contents have been added to the revised manuscript.

It would be beneficial to include a reference to Table 1 in the description of sea salt and dust parameterizations and other parameterizations on page 7550 because this table includes information that is relevant to the description of parameterizations in this section.

Reference to Table 1 has been added at various places in the “model overview” section.

P. 7551, l. 17: How are mixing state and size of the particles accounted for in the radiation calculations? Are separate radiation calculations performed for each size mode in the aerosol scheme? References?

The aerosol radiative effects are calculated as follows: From the chemical composition (including water content) and particle size, the Mie-scattering size parameter and volume-averaged refractive indices are derived for each aerosol mode assuming internal mixing of different chemical compositions. They are passed on to a look-up table that provides the extinction cross-section ϕ, single scattering albedo ω and asymmetry parameter γ. The look-up table is established using the Mie theory assuming 24 spectral bands for shortwave and 16 bands for longwave. The ϕ, ω, γ parameters are then re-mapped to the bands of the ECHAM radiation scheme. For each band, the ϕ, ω, γ parameters of different modes are synthesized into a single triplet for the radiative transfer calculation, assuming external mixing of the modes (Stier et al., 2005).

This is explained in Section 2.6 of the revised manuscript.

How do aerosols affect microphysical and macrophysical properties of clouds?

In the earlier model ECHAM5-HAM1 aerosols did not directly affect clouds because the one-moment cloud microphysics scheme therein (Lohmann and Roeckner, 1996) assumed fixed cloud droplet number concentration. In HAM2, the aerosol activation and ice nucleation parameterizations in the double-moment scheme of Lohmann et al. (2007) provide links between the simulated aerosol population and concentrations of cloud droplet and ice crystal. This version is thus able to simulate the impact of aerosols on cloud microphysics (droplet number and size) and macrophysics (liquid water path). This is clarified in Section 2 of the revised manuscript, with more details discussed in section 4.1.6.

What emissions are used for primary particles and what are the size and hygroscopic properties of the emitted particles?

Emissions of sulfur dioxide (SO2) and particulate sulfate, black carbon and primary organic aerosols (POA) are prescribed following the year 2000 specifications of AeroCom (Dentener et al., 2006): Non-eruptive volcanic SO2 emissions are taken from Andres and Kasgnoc (1998). Locations of eruptive emissions are from Halmer et al. (2002). The total strength of volcanic SO2 emission follows the recommendation of the GEIA inventory (http://www.geiacenter.org). Anthropogenically modified sources of SO2, BC and POA include wild-land fire, biofuel emissions and fossil-fuel emissions. Wild-land fire emissions are based on the Global Fire Emission Database inventory (van der Werf et al., 2004). Biofuel and fossil fuel emissions of BC and POA are prescribed according to the Speciated Particulate Emissions Wizard inventory (Bond et al., 2004). Biofuel and fossil fuel emissions of SO2 (including off-road, road transport, domestic, international shipping, industrial, and power plant emissions) are based on Cofala et al. (2005) and EDGAR (Olivier et al., 2005). The injection heights follow Table 1 in Dentener et al. (2006).

Primary aerosol emissions are distributed to different aerosol modes according to the emission type and the assumed soluble fraction. For sulfur emissions except DMS, 2.5% of the emission is assumed to be in the form of primary sulfate aerosols. For POA, 65% of the biomass burning and biogenic emissions are assumed to be soluble. *Table 2* in the *revised* manuscript summarizes the partitioning mass fraction of the primary aerosol emissions among different modes in ECHAM-HAM.

This is explained in Section 2.2 (Model overview: missions of aerosols and their precursors) of the *revised* manuscript.

P. 7552, l. 18: "The responses...are significant...and are consistently seen..." This statement is out of context. Such a statement should be made after the discussion of the actual results, if appropriate.

The sentence is removed.

P. 7554, l. 6: An important assumption for the parameterization of gas-to-particle conversion in HAM1 and HAM2 is the operator splitting between condensation and nucleation. This approach will work well if either condensation or nucleation rates are low. However, given that nucleation is so non-linear and that the time scales are often quite short for condensation and nucleation compared to the model time step, it is difficult to see how this approach can provide accurate results when nucleation and condensation occur at the same time. Although numerical errors may not be apparent for simulated aerosol mass, production rates for aerosol number concentrations are much more sensitive to

numerical errors. How confident are the authors that results presented for HAM2 in Fig. 2 indeed provide evidence for model improvements, as claimed? Given the use of operator splitting techniques in HAM1 and in HAM2, this is questionable. Have the authors considered to use shorter time steps to investigate numerical errors of the operator splitting methods?

It is true that operator splitting for nucleation can lead to numerical error. Our confidence in the HAM2 results comes from two sources:

1) Using box model calculations, Kokkola et al. (2009), compared the HAM2 time integration scheme to the original HAM1 scheme, the implicit (backward) Euler scheme, and to results from a reference numerical solver VODE (Brown et al., 1989). The new time integration scheme performed as good or better than the Euler backward scheme and significantly better than the HAM1 numerical scheme. Time steps up to 1800 s were considered in their comparison, which is longer than the time step in the ECHAM-HAM2 simulations. These tests support the use of the new time integration scheme.

2) With the full ECHAM-HAM model, we have performed numerical convergence test for the sulfuric acid gas equation using sub-stepping with up to 256 sub-steps per each step of model physics, and with various time integration schemes. A reference solution is established. It is found that the simulated H2SO4 gas concentration and aerosol nucleation rate are much more sensitive to the splitting technique applied between production and condensation than that between condensation and nucleation. With respect to the reference solution, the relative error in annual mean H2SO4 gas burden is reduced by a factor of more than 10 from the HAM1 numerics to HAM2. The error in total nucleation rate is reduced by a factor of about 5. Reduction of error can be seen at most of the model grid points though the factor is not homogeneously distributed. This indicates that from a numerical point of view, the solution procedure in HAM2 better represents the H2SO4 processes.

A summary of the results from the convergence test is added to the manuscript.

<u>References:</u>

Kokkola, H., Hommel, R., Kazil, J., Niemeier, U., Partanen, A.-I., Feichter, J., and Timmreck, C. (2009): Aerosol microphysics modules in the framework of the ECHAM5 climate model – intercomparison under stratospheric conditions, Geoscientific Model Development, 2, 97–112, doi:10.5194/gmd-2-97-2009, http://www. geosci-model-dev.net/2/97/2009/

Brown, P. N., Byrne, G. D., and Hindmarsh, A. C.: VODE, A Variable Coefficient ODE Solver, SIAM J. Sci. Stat. Comput., 10, 1038–1051, 1989.

Why are no results shown for nucleation mode number concentrations in the lower troposphere in Fig. 2, where aerosol number is important for CCN concentrations? Also, there are numerical methods available that do not require operator splitting. Has this been considered?

Nucleation mode number concentrations are shown in the upper row of Fig. 2. We did not zoom into the lower troposphere because in this region the differences between HAM1 and HAM2 are much smaller - both in absolute and in relative terms - than in the upper troposphere.

Currently there is research going on in the ECHAM-HAM community to implement and evaluate additional nucleation parameterization schemes in the planetary boundary layer. In this context, the numerical methods used for the H2SO4 processes will possibly be refined. Time integration schemes with adaptive step size and dynamical error control, such as those used by Herzog et al. (2004) and Zaveri et al. (2008) will be considered.

References:

Herzog M., D. K. Weisenstein, J. E. Penner (2004): A Dynamic Aerosol Module for Global Chemical Transport Models: Model Description. J. Geophys. Res., Vol. 109, D18202, doi:10.1029/2003JD004405

Zaveri R. A., R. C. Easter, Jr, J. D. Fast, and L. K. Peters (2008): "Model for Simulating Aerosol Interactions and Chemistry (MOSAIC)." Journal of Geophysical Research. D. (Atmospheres) 113(D13):Art. No. D13204. doi:10.1029/2007JD008782.

P. 7556: The partitioning of mass between the gas- and particle-phase for organic matter in HAM2 is not clear. Does this involve a numerical solution of the condensation equation, similar to condensation of sulphuric acid? What accommodation coefficients are used?

An oxidation product is considered either as volatile or semi-volatile, depending on the precursor. Organic matter formed from anthropogenic sources is assumed as non-volatile following Ng et al. (2007). Its mass is allocated to the aerosol phase immediately on formation. The oxidation products of isoprene and terpene are considered to be semi-volatile. The mass is partitioned between the gas and aerosol phases based on an equilibrium scheme, namely the two-product model of Odum et al. (1996), which assumes the mass of the gas phase, G, and that of the aerosol phases, A, are related by

$$A = Kp*M_0*G$$

where M_0 is the total SOA-absorbing mass in the aerosol phase, while Kp is an empirical parameter (cf. Eqn. (3) and Table 1 in O'Donnell et al., 2011).

Unlike the condensation of sulfuric acid gas, there is no differential equation involved in the gas-aerosol partitioning for SOA.

References:

Ng, N. L., Chhabra, P. S., Chan, A. W. H., Surratt, J. D., Kroll, J. H., Kwan, A. J., McCabe, D. C., Wennberg, P. O., Sorooshian, A., Murphy, S. M., Dalleska, N. F., Flagan, R. C., and Seinfeld, J. H. (2007): Effect of NOx level on secondary organic aerosol (SOA) formation from the photooxidation of terpenes, Atmospheric Chem- istry and Physics, 7, 5159–5174, doi:10.5194/acp-7-5159-2007, http://www.atmos-chem-phys.net/7/5159/2007/.

Odum, J. R., T. Hoffman, T., Bowman, F., Collins, D., Flagan, R. C., and Seinfeld, J. H. (1996): Gas/Particle Partitioning and Secondary Organic Aerosol Yields, Environ. Sci. Technol., 30, 2580–2585, doi:10.1021/es950943+

O'Donnell, D., Tsigaridis, K., and Feichter, J. (2011): Estimating the direct and indirect effects of secondary organic aerosols using ECHAM5-HAM, Atmospheric Chemistry and Physics, 11, 8635–8659, doi:10.5194/acp-11-8635-2011, http://www. atmos-chem-phys.net/11/8635/2011/

P. 7556, l. 22: What is the reason for the large difference in SOA yield between AeroCom and HAM2?

The yield of 15% used in AeroCom is realistic at lab temperatures (typically 25°C), while our model has a temperature-dependent SOA yield from monoterpenes (Saathoff, 2009), which predicts decreasing SOA formation with increasing temperature. Since the majority (over 75%) of the monoterpene mass is emitted at high temperatures in the tropics, the SOA yield drops below the figure used for AeroCom. The fact that we get a SOA production similar to the AeroCom specification despite a considerably lower yield reflects the dominance of isoprene emissions, which were not included in AeroCom.

This is explained in the revised manuscript.

Reference:
Saathoff, H., Neumann, K.-H., Möhler, O., Jonsson, Å. M., Kallquist, M.,Kiendler-Scharr, A., Mentel, Th. F., Tillmann, R. and Schurath, U. (2009): Temperature dependence of yields of secondary organic aerosolss from the ozonolysis of a-pinene and limonene, Atmos. Chem. Phys. 9, 1551-1577, doi:10.5194/acp-9-1511-2009

P. 7557, l. 25: This approach has been shown to produce large errors for the aerosol water content at low relative humidities (Kreidenweis et al., Environ. Res. Lett., 2008). How does this affect comparisons with the approach in HAM1 and the accuracy of radiation calculations in HAM2? It seems that these parameterization biases may contribute to the reduction in aerosol water content (P. 7558, l. 5)?

Results in the work of Kreidenweis et al. (2008), who applied simplifying assumptions to the theory developed by Petters and Kreidenweis (2007), have been widely interpreted as meaning that predictions using the k-Köhler theory are inherently and severely low-biased at low RH at least for certain species, especially NaCl. In our model the simplifications of Kreidenweis et al. (2008) are *not* used. Our implementation uses the full k-Köhler theory. Eqn. (11) in Petters and Kreidenweis (2007) is solved numerically for the growth factor for each soluble mode, taking into account ambient temperature and relative humidity, the mode's number median particle radius, and the k value. Our approach still underpredicts the water uptake of pure NaCl when compared to results from the Aerosol Inorganic Model (AIM, Wexler and Clegg (2002), the reference model used in Kreidenweis et al., 2008), but not as severely as indicated in their paper. Furthermore, relative humidity in the near-surface layers over the ocean (where most sea salt is found) is generally in the higher range where the k-Köhler theory and AIM are in good agreement. For these reasons, we believe that underestimation of the water uptake of certain species by the k-Köhler theory at low relative humidity has very limited effects on our model results. This is clarified in the revised manuscript.

For the purposes of radiation calculations, it is the growth in particle radius (not volume) that is important. For example, for a 100nm dry diameter particle of pure NaCl at 298.15K and 60% relative humidity, the difference between wet particle radius calculated by AIM model 3 (as used in Kreidenweis et al. (2008)) and our approach is 13%. In practice, sea salt in the model is always mixed with other compounds, which mitigates the underestimate that is less severe for other species in the model than for NaCl.

As for the decrease of aerosol water from HAM1 to HAM2, our investigation reveals that the cause is largely the different RH ceilings used for the accumulation and coarse mode particles (100% in ZSR and 95% for k-Köhler). This is demonstrated by a newly added figure that shows two sensitivity experiments performed using HAM2 but with the water uptake scheme reverted to ZSR. Despite an increase in dry aerosol burden compared to HAM1, which leads to an increase in aerosol water content from 75 Tg (HAM1, ZSR method, RH ceiling=100%) to 100 Tg (HAM2 with ZSR method, RH ceiling=100%), the change of RH ceiling results in a considerable reduction in aerosol water (~50 Tg) that more than compensates the dry burden effect. Replacement of the ZSR method by the k-Köhler theory (using the same RH ceiling), in contrast, has only marginal effect on the results (56 Tg vs 51 Tg). These results are consistent with the work of Adams et al. (2001) who found the use of higher maximum RH cutoff resulted in stronger water uptake and higher estimated direct forcing in the model of the Goddard Institute for Space Studies, especially when relative humidity is higher than 95%. These results are added to the revised manuscript.

<u>Reference:</u>

Petters, M. D. and Kreidenweis, S. M. (2007): A single parameter representation of hygroscopic aerosol growth and cloud condensation nucleus activity, Atmos. Chem. Phys. 7, 1961-1971, doi:10.5194/acp-7-1961-2007

Kreidenweis, S. M., Petters, M. D. and DeMott, P. J. (2008): Single-parameter estimates of aerosol water content, Environ. Res. Lett. 3, 035002, doi:10.1088/1748-9326/3/3/035002

Adams, P. J., J. H. Seinfeld, D. Koch, L. Mickley, and D. Jacob (2001), General circulation model assessment of direct radiative forcing by the sulfate-nitrate-ammonium-water inorganic aerosol system, J. Geophys. Res., 106(D1), 1097–1111, doi:10.1029/2000JD900512.

Wexler, A. S. and Clegg, S. L. (2002): Atmospheric aerosol models for systems including the ions H^+, NH^+, Na^+, $SO2^-$, $NO3^-$, Cl^-, Br^-, and H_2O, J. Geophys. Res., 107, 4207

Also, a figure should be included here for illustration of the differences, perhaps for zonal mean aerosol water.

A figure is added to the manuscript showing the annual and zonal mean cross-section of aerosol water content in (1) standard HAM1, (2) standard HAM2, (3) HAM2 but with the ZSR scheme and 100% RH ceiling for the water uptake of the accumulation mode and coarse mode particles, and (4) HAM2 but with the ZSR scheme and 95% RH ceiling. The corresponding global mean aerosol water contents, as well as the absolute and relative contributions of aerosol water to the aerosol optical depth, are also given in the figure.

P. 7559, l. 24: It appears that dust emissions increase by a factor of 4 for only a few grid points. Increases for concentrations and aerosol optical depth seem to indicate weaker increases in Asian dust emissions than a factor 4?

The largest relative differences in dust emission mass flux are somewhat misleading because they occur at grid points where the fluxes are small. In the revised manuscript we have replaced the panel of relative difference in dust emission by that of the absolute difference.

P. 7560, l. 20: How is R used for mixed-phase clouds in the model? Is the same value of R used for the liquid and frozen portions of the cloud condensate or are there differences for in-cloud scavenging of aerosol through warm- and ice-phase processes in mixed-phase clouds?

Yes, the same value is used for the liquid and ice phase portions in mixed-phase clouds. This is clarified in the revised manuscript.

The coupling between HAM2 and the cloud microphysics scheme in ECHAM needs to be explained in more detail.

In both HAM1 and HAM2, cloud microphysics can lead to removal of aerosols from the air. The loss of aerosol particles due to their role as cloud condensation nuclei (or ice nuclei) is parameterized via a scavenging coefficient, which denotes the fraction of the available aerosols in the cloudy part of a grid box that is embedded in the cloud droplets and ice crystals. Different values of the scavenging coefficient are prescribed for different aerosol modes and cloud types (cf. Table 3 in Stier et al., 2005). Because interstitial and cloud-born aerosols are not distinguished in the model, the particles embedded in cloud water/ice are considered as removed from the atmosphere only when the condensates are converted into precipitation. Below-cloud scavenging describes the removal of aerosols due to collection by rain or snow. The removal rate depends on the precipitation rate and area, as well as the collection efficiency (Seinfeld and Pandis, 1998; Stier et al., 2005). Different collection efficiencies are assumed for rain and snow.

In HAM2 the impact of aerosols on cloud microphysics is implemented as follows: Aerosol activation in warm clouds is described by the semi-empirical scheme of Lin and Leaitch (1997). Autoconversion of cloud droplets to rain is parameterized as in Khairoutdinov and Kogan (2000). Homogeneous ice nucleation in cirrus clouds is assumed to happen at air temperatures below -38°C when supercooled solution droplets freeze. The production rate of ice crystals is computed following Kärcher and Lohmann (2003). Heterogenous nucleation happens in the model when dust exists and the air temperature lies between -38°C and 0°C. Internally mixed dust and black carbon aerosols are assumed to act as immersion nuclei while only externally mixed dust particles act as contact nuclei. Contact freezing by black carbon is not considered as it is quite uncertain (Lohmann and Hoose, 2009).

These connections between aerosols and cloud microphysics are described in Section 2.5 (Model overview: removal processes) and Section 4.1.6 (Model updates and their effects: Cloud microphysics and aerosol activation) of the revised manuscript.

P. 7561, l. 6: What is the difference in global mean aerosol optical depth in Fig. 6a?

Global mean and RMS of the differences are added to all panels in the figure.

P. 7561, l. 26: The title of this section is misleading because effects of cloud microphysical processes on aerosols were already considered in the previous section.

The title is changed to “Cloud microphysics and aerosol activation”.

P. 7562, l. 28, Fig. 7: Observations are available for cloud radiative forcings and should be included in this figure. Without the observations, it is not clear whether the model changes lead to improved model results or not. Some of the changes do not appear to be beneficial for the simulated cloud radiative forcing considering climate biases in ECHAM. On the other hand, it is not clear why results are included for the cloud radiative forcing in the paper because aerosols typically have relatively small effects on overall cloud radiative forcings compared to other, often very uncertain, processes. For instance, it may be possible to tune the autoconversion efficiency to produce much better agreement in results in Fig. 7. Hence, it would be better to remove or replace this figure.

The figure is removed.

P. 7563, l. 10: It is not clear why the absolute autoconversion rate should be considerably smaller? Since the mean precipitation rate is similar this would imply that the accretion rate is much greater? On the other hand, increases in cloud water path likely point at a reduced *efficiency* of autoconversion. Perhaps it would be possible to include more quantitative information, e.g. a cloud water budget?

Our original statement was not correct. A cloud water budget analysis has been performed, in which it is found that for example for the liquid phase, the global mean net rain production rate (autoconversion + accretion – evaporation) does not show a significant change, while the net water-to-rain conversion efficiency, defined as the net rain production rate divided by liquid water path (LWP), decreases in the two-moment cloud scheme because of significant increase in LWP. Because the in-cloud scavenging is directly related to this conversion efficiency in the model, weaker in-cloud scavenging is obtained with the two-moment cloud scheme, and consequently the longer aerosol lifetime in Table 6 of the discussion paper.

The corresponding subsection in the manuscript is revised.

Furthermore, an increase in cloud water path may lead to increased in-cloud oxidation of sulphur species. Is there any evidence for increased in-cloud production rates?

The sulfate production attributed to aqueous oxidation does not show significant change (42.533 Tg(S)/yr in HAM2, 42.507 Tg(S)/yr with the old cloud scheme). So far we have not performed detailed analysis of sulfur chemistry in the simulations discussed in the paper.

P. 7564, Table 5: The dry deposition rates are considerably lower in HAM than in the AeroCom models. Add an explanation and a brief description of the dry deposition scheme in HAM.

Turbulent dry deposition of aerosols is considered for all modes except for the nucleation mode, following Ganzeveld et al. (1998). The deposition flux is computed as the product of tracer concentration, air density and deposition velocity, first calculated on each of the surface types considered by the ECHAM5 model (snow/ice, bare soil, vegetation, wet skin, open water, and sea ice), and subsequently summed up using the fractional surface area. Deposition velocity is obtained from the aerodynamic resistance and surface resistance, which in turn depend on particle size and density, properties of the Earth's surface, and characteristics of atmospheric turbulence.

Gravitational sedimentation of a single aerosol particle is described by the Stokes theory (Seinfeld and Pandis, 1998). The correction of Slinn and Slinn (1980) is applied to get the sedimentation velocity of a log-normal mode. To avoid violation of the Courant-Friedrich-Lewy stability criterion, the sedimentation velocity is limited to $V_s <= \Delta z/\Delta t$ where Δz and Δt are the model layer thickness and time step, respectively. In the model, sedimentation is considered only for the larger particles, i.e., the accumulation and coarse modes.

The descriptions are included in Section 2.5 (Model overview: removal processes) of the revised manuscript.

Regarding the lower deposition rates in comparison to the AeroCom multi-model mean, we would like to bring the readers' attention to the following references:

1) Textor et al. (2006) analyzed the annually and globally averaged effective sink rate coefficients in the AeroCom Phase 1 models (see Section 7 therein), where large diversities are seen in the results and in the way the dry deposition processes are parameterized.
2) Zhang et al. (2010) compared the global mean aerosol mass budget in ECHAM-HAM1 with those in GAMIL-LIAM and CAM3-LIAM. The latter two are the same aerosol module connected to different climate models. Both HAM and LIAM use dynamically computed turbulent dry deposition velocity, but with different parameterization schemes. Sedimentation calculations are based on the same theory, but with different numerical treatments. For all aerosol types except dust, the three models feature similar deposition rates (see Tables 5 and 6 therein). There are no systematic differences between ECHAM-HAM and the two -LIAM models. In fact in many cases, the difference in dry/sedimentation deposition rate between ECHAM-HAM and GAMIL-LIAM is smaller than between either of them and CAM3-LIAM. This suggests that differences in model meteorology also place an important role in the deposition processes.

To provide an explanation for the weaker (than the AeroCom mean) dry deposition in HAM, one would need to carry out a comprehensive intercomparison of the results and the schemes used in these models. Having not done such analysis, we would not speculate in the manuscript.

<u>References:</u>

Ganzeveld, L., Lelieveld, J., and Roelofs, G.-J. (1998): A dry deposition parameterization for sulfur oxides in a chemistry and general circulation model., Atmos. Env., 103, 5679–5694., doi: 10.1029/97JD03077

Seinfeld, J. H. and Pandis, S. N. (1998): Atmospheric Chemistry and Physics: From Air Pollution to Climate Change., J. Wiley, New York

Slinn, S. A. and Slinn, W. G. N. (1980): Predictions for particle deposition on natural waters., Atmospheric Environment, 14, 1013–1026

Textor, C., M. Schulz, S. Guibert, S. Kinne, Y. Balkanski, S. Bauer, T. Berntsen, T. Berglen, O. Boucher, M. Chin, F. Dentener, T. Diehl, R. Easter, H. Feichter, D. Fillmore, S. Ghan, P. Ginoux, S. Gong, A. Grini, J. Hendricks, L. Horowitz, P. Huang, I. Isaksen, T. Iversen, S. Kloster, D. Koch, A. Kirkevåg, J.E. Kristjansson, M. Krol, A. Lauer, J.F. Lamarque, X. Liu, V. Montanaro, G. Myhre, J. Penner, G. Pitari, S. Reddy, Ø. Seland, P. Stier, T. Takemura, and X. Tie (2006): Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777-1813

Zhang, K., Wan, H., Wang, B., Zhang, M., Feichter, J., and Liu, X. (2010): Tropospheric aerosol size distributions simulated by three online global aerosol models using the M7 microphysics module, Atmos. Chem. Phys., 10, 6409-6434, doi:10.5194/acp-10-6409-2010

P. 7565, Fig. A1: More quantitative information needs to be included, e.g. mean concentrations for all sites. There is no further discussion and so it is not clear why this figure is shown in the paper.

The figure is removed from the manuscript since it is very similar to the HAM1 results shown by Fig. 3 in Stier et al. (2005). The following sentence is added at the end of Section 5.1:

“A comparison of the simu- lated monthly mean surface mass concentrations against the EMEP (European Monitoring and Evaluation Programme), IMPROVE (Interagency Monitoring of Protected Visual En- vironments) and the University of Miami networks further confirms that the surface concentrations are similar to those in HAM1 (not shown).”

P. 7567, l. 6: The authors state that the aerosol number concentration is very well simulated but no evidence is shown. An integration over the size distributions would likely reveal large differences for the aerosol number concentration. Furthermore, the double logarithmic scale that is used in the figure is problematic. For instance, the total number and maximum of the size distribution are underestimated by about a factor of 10 for Ispra for DJF in Fig. 12.

It is true that the logarithmic scale and linear scale plots can give different impressions visually. The double-logarithmic scale used in the figure is consistent with the mathematical formulation (i.e., the modal method) used in our model for representing particle size distribution. It also helps to illustrate whether the model can, or cannot, capture the characteristic shapes of the observed distribution over wide ranges of particle size (10^{-2} – 10^{0} micron) and number concentration (10^{1} – 10^{5} cm^{-3}). Such plots have been used frequently in observational studies and in previous modeling studies (see, e.g., Fig. 4 in Herzog et al., 2004; Fig. 20-22 in Liu et al., 2005; Figs. 3-4 in Elleman and Covert, 2010; Figs. 1 and 3 in Mann et al., 2012).

To make the differences between model versions more visible in the revised manuscript, we have changed the aspect ratio of the panels in Figs. 12 and 13. In addition, a table is added that compares the total aerosol number concentrations (integrated over the size distributions) at each site, so as to provide a more quantitative comparison between the observations and simulations.

References:

Herzog M., D. K. Weisenstein, J. E. Penner (2004): A Dynamic Aerosol Module for Global Chemical Transport Models: Model Description. J. Geophys. Res., Vol. 109, D18202, doi:10.1029/2003JD004405

Liu, X., J. E. Penner, and M. Herzog (2005), Global modeling of aerosol dynamics: Model description, evaluation, and interactions between sulfate and nonsulfate aerosols, J. Geophys. Res., 110, D18206, doi:10.1029/2004JD005674.

Mann, G. W., Carslaw, K. S., Ridley, D. A., Spracklen, D. V., Pringle, K. J., Merikanto, J., Korhonen, H., Schwarz, J. P., Lee, L. A., Manktelow, P. T., Woodhouse, M. T., Schmidt, A., Breider, T. J., Emmerson, K. M., Reddington, C. L., Chipperfield, M. P., and Pickering, S. J.: Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model, Atmos. Chem. Phys., 12, 4449-4476, doi:10.5194/acp-12-4449-2012, 2012.

Elleman, R. A., and D. S. Covert (2010), Aerosol size distribution modeling with the Community Multiscale Air Quality modeling system in the Pacific Northwest: 3. Size distribution of particles emitted into a mesoscale model, J. Geophys. Res., 115, D03204, doi:10.1029/2009JD012401.

It should also be mentioned that differences between HAM1 and HAM2 are small compared to differences between model results and observations.

The statement is true in terms of the characteristic shape of the size distribution. It is mentioned in the revised manuscript.

P. 7567, l. 17: What sites exactly are considered to be affected by heavy pollution here?

The sites at which the simulated aerosol size distribution are evaluated in Figs. 12 and 13 of the discussion paper are mostly background stations representing relatively clean cases (except for Ispra, which is influenced by pollution outflow from cities). The "polluted regions" in line 18, p 7567 refers to the Northern Hemisphere middle latitudes (120-150°E, 30-60°N and 240-270°E, 30-60°N), i.e., row 3 in Fig. 10 of the previous subsection. This is clarified in the revised manuscript.

P. 7567, l. 25: Underestimates for aerosol number concentration in polluted regions and overestimates in more remote locations are to be expected because it is unlikely that the model fully resolves the horizontal scales of aerosol plumes. Insufficient horizontal resolution likely explains this kind of concentration biases. Note the similarities between simulated size distributions at different sites.

We thank the reviewer for pointing out this point that was not mentioned in the discussion paper. It is included in the revised manuscript.

P. 7568, l. 10: Another factor that might affect this comparison is the location of the observations in the vertical. What is the height of the observations and corresponding model results in the figure? Perhaps the vertical resolution is too low in the model to sufficiently resolve the rather shallow layer of sea salt aerosol above the surface of the ocean?

The observational data used here are ship cruise measurements obtained on board at about 10-20 m above the sea level. The lowest computational level of the model is about 30 m above the surface (Roeckner et al., 2006). So far HAM has not yet been run at higher vertical resolutions in the boundary layer. It is not clear how the simulated aerosol distributions would change, especially because some aspects of the climatology of the host model (e.g., surface wind speed, turbulent transport) have been found sensitive to vertical resolution.

Reference
Roeckner, E., Brokopf, R., Esch, M., Giorgetta, M. A., Hagemann, S., Kornblueh, L., Manzini, E., Schlese, U., and Schulzweida, U. (2006): Sensitivity of Simulated Climate to Horizontal and Vertical Resolution in the ECHAM5 Atmosphere Model., Journal of Cli- mate, 19, 3771–3791

Simulated concentrations for the coarse mode seem to be systematically lower than the observed concentrations. Is this consistent with other results? According to these results (i.e. unrealistically low concentration and large particle sizes in HAM), one would expect the model to severely underestimate the aerosol optical depth over the ocean. However, the opposite is the case (Fig. 15), which indicates an inconsistency either in the model or in the way model results are compared with observations.

The two modes showed in the figure are Aitken mode and accumulation mode. The simulated accumulation mode particle concentrations are systematically lower than observation. One possible reason is that the model underestimates the accumulation mode aerosol number concentration associated with marine organic aerosols, which have smaller water activity than sea salt aerosols.

The Heintzenberg et al. (2000) data set does not include concentrations of the coarse mode particles that are an important contributor to the aerosol optical depth (AOD) over the ocean. The underestimate of accumulation mode concentrations in Fig. 14 does not contradict the positive bias of AOD over the ocean in Fig. 15 of the discussion paper.

Reference
Heintzenberg, J., Covert, D. C., and van Dingenen, R. (2000): Size distribution and chemical composition of marine aerosols: a compilation and review, Tellus, 52B, 1104–1122, doi:10.1034/ j.1600-0889.2000.00136.x

P. 7568, l. 24: These comparisons are very qualitative. At a minimum it will be necessary to include global mean values for a more quantitative assessment of model accuracy. An even better approach would be the additional use of Taylor diagrams.
P. 7569, l. 18: Again, more quantitative information needs to be provided in addition to merely describing differences in patterns in Fig. 16.

Figures showing zonal averages of the observed and simulated AOD and AAOD are added to the manuscript. In addition, Taylor diagrams of AOD, Ångström parameter and AAOD are added to compare simulations with observations in different regions.

P. 7571, l. 8: How do these results compare to other estimates in the literature? If there are differences, what may be causing these differences and is there any indication that HAM2 produces better results than HAM1?

The term “aerosol radiative forcing” used in Section 5.5 should better be called “aerosol radiative effect” because what we really mean is the impact of *all* aerosols present in the atmosphere. The aerosol forcing defined by the IPCC, in contrast, refers to the impact of *anthropogenic* aerosols derived from certain kinds of *PD – PI* (present day minus pre-industrial) simulations. This is clarified in the revised manuscript. In the literature most modeling studies have been reporting the *forcing*, while results of the *total aerosol effect* defined in our sense are not commonly seen.

In the revised manuscript we included two tables that present the radiative forcing of anthropogenic aerosols derived from a series of *PD – PI* simulations carried out according to the suggestion of reviewer # 2. Results from the AeroCom Phase 1 intercomparison (Schulz et al., 2006) are included for comparison.

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

Furthermore, no information is provided about the accuracy of the simulated relative humidity, which plays an important role for radiative forcings and aerosol optical depth.

The evaluation of simulated relative humidity on a global scale is not a trivial issue considering the coverage and accuracy of the available observations.

From model evaluations performed by the ECHAM developers and users, we are not yet aware of concerns about severe systematic biases in relative humidity in the model.

That being said, we do agree with the reviewer that relative humidity has an important impact on AOD because it directly affects water uptake, especially in the relatively wet regions. A further investigation of the RH bias in the model would be valuable. Our current results have revealed positive biases of AOD over the oceans in comparison to the MODIS data (Fig. 17 of the discussion paper). It would be useful to check how this is related to relative humidity in ECHAM.

P. 7572, l. 3: No evidence is provided in the paper to support this statement.

The statement replaced by “The incorporation of a two-moment stratiform cloud microphysics scheme allows aerosols to directly affect cloud microphysics through cloud droplet activation and ice nucleation”.

P. 7572, l. 14: Correct the reference.

Corrected.