Reply to Anonymous Referee #2

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We thank referee #2 for the encouraging and constructive comments. Below is our response to the review.

The authors present an interesting and well-written overview of recent improvements to the ECHAM-HAM global aerosol model. The sensitivity studies presented are well designed, and good effort has been put into comparisons with data.

As a general remark, however, the paper fails to give either a simple and concise overview of the actual effects of the recent improvements, for which it is too detailed and lacks a concise overview, or to provide the full set of details needed to thoroughly evaluate the combined effects of the updates. For the latter it does not seem to provide enough quantitative detail. There are two main uses for such a paper, which the authors that they are trying to achieve. One is to document the combined effects of changes presented elsewhere. For this, an extended model description would greatly ease the reading of the subsequent sections, as well as some extended discussions to place the sensitivity studies further in context (I note some points below, but as reviewer 1 has commented on this in great detail I will not repeat that). A second use is to serve as a baseline reference both for future studies with ECHAM-HAM2 and for intermodel studies such as the ongoing AeroCom Phase 2. For this use, a different set of details would be needed – especially regarding the radiative effects of the individual aerosol species. Details are given below. Adding this would greatly increase the impact of the paper.

Overall, however, I find the study highly relevant and believe it should proceed to ACP once some additional detail has been added.

In response to these comments, three major changes have been made to the manuscript:

1. The abstract of the paper has been re-written. The new version summarizes the main differences between aerosol climatology in HAM2 and HAM1, and points out the changes in model formulation that are responsible for these differences.
2. Section 2 (Model overview) has been extended substantially to provide a more detailed summary of the model. See also our response to the next comment and to reviewer 1.
3. For a more quantitative evaluation of the new model version, we added the following:
    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, a new figure comparing the simulated and observed vertical 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 is added 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 for the global domain;

g. In the last subsection before conclusions, two tables showing the global mean radiative forcing by anthropogentic aerosols.

P7550: Even though good references are given, an extended model overview of ECHAM5 (some general points only) and HAM would benefit the paper. What resolutions are used? E.g. if the resolution in the present work is higher than what most models in the AeroCom Phase 1 study (Schulz et al 2006) used, this may have an impact on the comparisons made with that work.

In the revised manuscript we have significantly extended Section 2 (Model overview) to include an overview of the main components of ECHAM5 and more 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) the calculation of aerosol optical properties and radiative effects, and 7) aerosol indirect effects.

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.

P7550: Sections 5.4 and 5.5 present a very interesting discussion of the simulated radiative properties of, and forcing from, aerosols. In the introduction, the authors state that "The four-band shortwave radiative transfer scheme in the atmospheric model has been extended with two additional bands (Cagnazzo et al., 2007)." Some additional detail on the radiative transfer scheme and how it has been updated from HAM1 would be relevant here.

Shortwave and longwave radiative transfer calculations follow the methods of Fouquart and Bonnel (1980) and Mlawer et al. (1997), respectively. The longwave scheme considers 16 spectral bands. The shortwave scheme has 4 or 6 bands depending on model version. The 6-band version adapted from the ECMWF model by (Cagnazzo et al., 2007) has the 250–690 nm interval sub-divided to better consider the absorption by water vapor at 440–690 nm. In addition, an ultra-violet band is added to consider the absorption by ozone, resulting in a total of 3 bands in the ultra-violet and visible range (185–250 nm, 250–440 nm and 440–690 nm).

Radiative properties of aerosols are dynamically computed in the model. 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).

The refractive indices of various aerosol compositions at λ=550 nm (shortwave) are listed in Table 3 of the revised manuscript. Corresponding quantities for the longwave bands are shown in Fig. 1 of Stier et al. (2007) as functions of wavenumber. The refractive indices of black carbon used in HAM2 are the updated values evaluated by Stier et al. (2007). HAM2 considers both the longwave and shortwave effects of aerosols, while HAM1 considers only the shortwave effects. Radiative effects of the nucleation mode particles are ignored due to their small sizes.

These are added to Section 2 of the revised manuscript.

References:

Fouquart, Y. and Bonnel, B. (1980): Computations of solar heating of the earths atmosphere: A new parameterization., Beitr. Phys. Atmos., 53, 35–62

Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A. (1997): Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the long- wave., J. Geophys. Res.,102, 16 663–16 682, 1997.

Cagnazzo, C., Manzini, E., Giorgetta, M. A., Forster, P. M. D. F., and Morcrette, J. J. (2007): Impact of an improved shortwave radiation scheme in the MAECHAM5 General Circulation Model, Atmospheric Chemistry and Physics, 7, 2503–2515, doi: 10.5194/acp-7-2503-2007, http://www.atmos-chem-phys.net/7/ 2503/2007/

Stier, P., Seinfeld, J. H., Kinne, S., and Boucher, O. (2007): Aerosol absorption and radiative forcing, Atmospheric Chemistry and Physics, 7, 5237–5261, doi:10.5194/acp-7-5237-2007, http:// www.atmos-chem-phys.net/7/5237/2007/

P7550: "… are prescribed according to the specifications of AeroCom.": Please give some details here, or at least a reference, for those not familiar with the AeroCom work.

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.

These are added to Section 2.2 (Model overview: emissions of aerosols and their precursors) of the revised manuscript.

P7552: "The responses of model results to formulation/configuration changes in the following sections are significant in magnitude, and are consistently seen in different diagnostics." This statement is not clear to me – clarify?

The sentence is removed.

P7553: "In the standard model configuration, these nucleation pathways are switched off." There are a number of statements like this in the paper (notably also on page 7561). While Table 1 (p.7585) gives a good textual overview over the features included, I kept feeling the need for an even clearer picture of what is included and not in the "default HAM2" – which I guess is what will mainly be used for future studies.

In the revised version, Section 4 (Model updates and their effects) is separated into two parts:

- The 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.

P7556: "(by changing model configuration via namelist)": Technical info beyond the level of the paper – remove?

Removed.

P7562: "Various other factors, including aerosol source, horizontal and vertical transport timescale and pathway, are also relevant in determining…" I would appreciate more quantitative detail here, especially on the vertical transport – either here or elsewhere in the paper. (See e.g. Schwarz et al 2010 – the final profiles of esp. BC aerosol after scavenging are highly relevant for the final modeled BC forcing.)

Schwarz et al. (2010) noticed a factor of five overestimate of the refractory black carbon concentrations in the 14-model mean of AeroCom Phase 1 in comparison to aircraft measurements obtained above the remote Pacific during the HIPPO campaign. They attributed the discrepancies to insufficient wet removal in the models. In our simulations, over the Pacific Ocean the strongest decreases in monthly mean BC concentration caused by the modified below-cloud scavenging are about 10 - 20% (relative difference), occurring in the lower troposphere in the middle and high latitudes. The temperature dependent in-cloud removal in mix-phase clouds has an opposite effect.

The impact of changing wet scavenging parameterization on aerosol mass concentration is shown and discussed in the revised manuscript. The statement cited by the reviewer is removed because in this study we did not carry out any simulation to analyze transport processes.

Reference:
Schwarz, J. P., J. R. Spackman, R. S. Gao, L. A. Watts, P. Stier, M. Schulz, S. M. Davis, S. C. Wofsy, and D. W. Fahey (2010): Global-scale black carbon profiles observed in the remote atmosphere and compared to models, Geophys. Res. Lett., 37, L18812, doi:10.1029/2010GL044372.

P7563: Radiative transport is at the heart of several of the sensitivity studies presented. What is the effect of the improved shortwave treatment in HAM2?

The original purpose of updating the shortwave radiation scheme was to improve model dynamics, specifically the ozone absorption and general circulation in the stratosphere. It resulted in a significant reduction of cold bias in the summer stratopause, an annual mean warming of 0.5 K in the middle troposphere, and a warming of 1 – 1.5 K in the upper troposphere. Close to the Earth's surface, the changes were mitigated by the imposed sea surface temperatures. The impact of the update is generally regarded as positive on the model climate (Cagnazzo et al, 2007). It would be interesting to analyze the impact of this update on the simulation of aerosol radiative effects as well. This has not been done because the old 4-band scheme is no longer available in the ECHAM-HAM2 code and would require considerable effort to bring back.

For our paper, it should be noted that

1) The aerosol radiative properties discussed in Section 5.4 (AOD, AAOD and Ångström parameter) are computed independently from the radiative transfer code, as diagnostic variables of the aerosol module.
2) In Section 5.5, results of the aerosol radiative effect in Fig. 20, in Table A2, and in Table 10 and 11 of the revised manuscript are obtained with different configurations of HAM2 which use the same radiation scheme.

The modifications in the shortwave scheme thus do not affect our analysis.

Reference:
Cagnazzo, C., Manzini, E., Giorgetta, M. A., Forster, P. M. D. F., and Morcrette, J. J. (2007): Impact of an improved shortwave radiation scheme in the MAECHAM5 General Circulation Model, Atmospheric Chemistry and Physics, 7, 2503–2515, doi: 10.5194/acp-7-2503-2007, http://www.atmos-chem-phys.net/7/ 2503/2007/

P7568: For the discussions of figures 15 and 16, some further quantification would be interesting – e.g. in the form of global or zonal means.

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.

P7569: Here I'm missing a concise table with the radiative properties of both the individual aerosol species and total anthtropogenic aerosols. E.g. what is the refractive index of BC now used, is POM treated as absorbing or purely refractive, what is the total single scattering albedos and absorption AOD,… Some of this is given in table A2 on page 7593, but an increased level of detail would greatly facilitate later intermodel comparisons.

BC refractive index is 1.85+0.71*i* at 550 nm in HAM2. POM is treated as slightly absorbing (refractive index is 1.53+0.0055*i* at 550 nm). A complete list of the refractive indices for all compositions, together with the references, are given in Table 3 of the revised manuscript.

A table is added that compares the (1) total AOD, (2) AOD of each individual species, (3) absorption AOD, (4) single scattering albedo in HAM1 and HAM2, over land, over the ocean, and for the global domain.

For the anthropogenic aerosols, see our reply to the next comment.

P7571: I find this very interesting section (5.5) to be rather brief – further details here would enhance the paper. What is the regional response of the TOA forcing in the various sensitivity tests? Can you say something about the effects of the changes in aerosol wet deposition on the total BC forcing, which is very sensitive to the aerosols' location relative to clouds (e.g. Zarzycki et al 2010, Samset et al 2011), or the effect of the change in water uptake on sulphate forcing? (I realize that treating individual species requires further sensitivity studies, but as you state that you participate in AeroCom Phase 2 these are perhaps already performed?)

The AeroCom Phase 2 intercomparison of direct aerosol forcing requires simulations that are nudged to the year 2006 meteorology and forced by the pre-industrial (PI, year 1850) and present-day (PD, year 2006) aerosol emissions. The present-day simulations are different from experiments in this study. We thus do not use the submitted data in this paper, but performed similar simulations following the Phase 1 specifications (year 2000 for PD, 1750 for PI).

A comprehensive evaluation following the review's list, especially that of the regional responses, would lead to a considerable extension of the manuscript. Since the manuscript is already long, we feel it would be better to present those results in a separate paper. In the revised manuscript, only two tables are added to present the global mean radiative forcing by anthropogentic aerosols. The first one follows the style of Table 5 in Schulz et al. (2006), which shows the anthropogenic AOD of all aerosol species, its contribution to present day total AOD, and the radiative forcing expressed in different flavors. The second table shows similar information, but for different aerosol species and sensitivity experiments. In both tables the AeroCom 1 results from Schulz et al. (2006) are included to put our numbers into perspective.

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

P7572: The Lohmann and Roeckner ref is given twice.
Corrected.

P7585: Spurious line break ("dy- namics") in upper left cell
Corrected.

P7593: Typo: larges -> largest
Corrected.

P7595: Fig 2, caption begins with "and zonal mean…".
Corrected.

P7601: This figure is central in seeing the combined effects of model improvements on the vertical aerosol profiles. However in the right column I find it hard to read off the areas of most significant change between the models, as the ratio will tend to become large when the concentration simulated by HAM1 becomes small. How does this picture look for (HAM2-HAM1) or (HAM2-HAM1)/(HAM2+HAM1)?

A column showing the (HAM2-HAM1) difference is added to the figure.

P7603: Fig 10: Some further degree of quantitative comparison between HAM2, HAM1 and the observations would be interesting here, in addition to the zonal profiles given. E.g. mean vertical profiles for the lat/lon regions with best coverage, or preferably a comparison close to aerosol source regions.

A figure is added that compares the simulated (HAM1, HAM2) and observed vertical files in selected regions.