Atmos. Chem. Phys. Discuss., 7, S3426–S3439, 2007
www.atmos-chem-phys-discuss.net/7/S3426/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on “Aerosol absorption and radiative forcing” *by* P. Stier et al.

P. Stier et al.

Received and published: 25 July 2007

We would like to thank the reviewer for the helpful comments that substantially improved the manuscript. We very much appreciated the detailed remarks and hope to have addressed all raised issues. All page numbers refer to the revised manuscript in the submitted form.

1. The main shortcoming of the present study is that other studies have examined all the individual sensitivities examined in the present study to different degrees (e.g., refractive indices, mixing rules, surface albedo, absorbing inclusions within clouds), but not in the same paper. Whereas the present paper offers a new evaluation of the issues (and possibly more sensitivity tests), it is not clear what new scientific insight is gained, aside from different estimates. The authors should motivate better the reason why it is important to provide additional estimates of these parameters.

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We are fully aware that individual aspects of the present study have been previously addressed in the literature and give representative studies of each aspect credit in the introduction (now extended as given below). Yet, we do not see this as shortcoming of our study, that is aimed as “comprehensive examination of aerosol absorption with a focus on evaluating the sensitivity of the global distribution of aerosol absorption to key uncertainties in the process representation”.

Previous studies of individual issues relevant to aerosol absorption, with vast differences in emissions, process representations, and model complexities, did generally not allow to compare or assess the importance of individual effects. In addition, very few of the studies evaluated the simulated aerosol absorption. Here, we quantify and evaluate the effect of the model sensitivities with respect to measurements (AERONET retrieved absorption optical depth) and provide the community with quantitative information on the importance of the relevant processes for aerosol absorption and the resulting radiative forcing.

We have further motivated our study in the introduction:

We added the citations Haywood and Shine (1995), Myhre et al. (2003,2004) as examples for an investigation of the effect of the surface albedo on the aerosol radiative forcing (page 2, col 2, par 4).

We extended the motivation section in the introduction (page 3, col 1, par 3):

“There exist a number of studies in which the global distribution of aerosol absorption has been computed, generally as part of an overall analysis of aerosol radiative forcing. As noted, the absorption portion is likely the most uncertain component of direct aerosol radiative forcing. Only few studies evaluate the simulated aerosol absorption. Other studies focus on individual processes relevant for aerosol absorption, such as refractive index mixing rules (e.g. Ackerman and Toon, 1981; Chylek et al., 1995,2000; Jacobson 2000). The objective of the present study is to carry out a comprehensive, measurement constrained examination of aerosol absorption. A focus is on evaluat-

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ing the sensitivity of the global distribution of aerosol absorption to key uncertainties in the process representation. These include aerosol absorbing component refractive indices, surface albedo, refractive indices mixing rules, and the absorption of aerosol inclusions in cloud droplets and ice crystals. Predictions of present day aerosol absorption optical depths and the sensitivity to the process representations are evaluated and quantified with observations from the global AERONET sun-photometer network."

2. P. 7178. "an extensive evaluation of this base model can be found in" The model appears to have been evaluated only at coarse global resolution and not at high resolution against field data. If this is correct, the authors should state so and remove the term extensive since global-scale evaluations are useful only for evaluating mean properties, not so much instantaneous, location-specific properties.

The ECHAM5-HAM model has been evaluated at a global resolution of T63L31, i.e. horizontally about 1.8x1.8 degrees - among the highest in the AeroCom aerosol model inter-comparison. We believe that this evaluation at the same resolution that we actually applied for transient climate simulations (Roeckner et al. 2006, Stier et al., 2006) is probably more representative than an evaluation of a short simulation with high resolution. Considering that the evolution of the aerosol system (as many other components of global models) is inevitably to some degree sub-grid scale, any evaluation of global results is obviously challenging. Nonetheless, we believe that an evaluation of the performance in a statistical sense (mean, median, percentiles) is a key benchmark for all global atmospheric models.

We refer with the cited sentence to the evaluation in Stier et al. (2005) in that the base model is evaluated against: surface mass measurements from the EMEP, IMPROVE, Prospero & Savoie, and GAW measurement networks/datasets, measurements of vertical aerosol number concentration profiles from the INCA-NH, INCA-SH, and UFA-EXPORT campaigns, measured aerosol size-distributions at numerous surface measurement sites from Putaud et al. (2003), aerosol optical depth from the AVHRR,

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MISR, MODIS, and TOMS satellite instruments as well the AERONET sunphotometer network, the Angstrom parameter from MODIS, and the absorption optical depth retrieved from AERONET. As this is arguably one of the more detailed published evaluations of a global aerosol model, we consider it more an issue of semantic whether or not one calls this evaluation "extensive".

P. 7178. "Water vapour, cloud liquid water, cloud ice, and trace components are transported in grid-point space"

The following questions refer to the general circulation model ECHAM5 (Roeckner et al., 2003, 2005) that is hosting the aerosol model HAM for the present study. While we agree that a detailed description of the host model is desirable, we think we have to meet a balance in detail in the model description (as similar detail could be of interest for any aspect of the complex model) and would like to refer the interested reader to the numerous references given in this section.

What is the implication of transporting cloud liquid water and cloud ice across course global grid cells? This would seem to indicate that clouds are spread across entire coarse grid cells, which is not realistic, and that numerical diffusion (which will occur with any transport scheme) will spread such clouds further.

In ECHAM5, cloud liquid water and ice are defined as prognostic sugridscale variables for the cloudy fraction of each grid box. For the global transport, they are converted to grid-box mean averages transported employing the flux form semi-Lagrangian transport scheme by Lin and Rood (1996), that is significantly less diffusive than the previously used semi-Lagrangian scheme. Details of this treatment can be found in the cited Roeckner et al (2003) and Lohmann and Roeckner (1996) references.

The authors then state, "Cloud cover is predicted with a prognositic-statistical scheme total water." Does this mean that, on top of the cloud water transported, more cloud water is produced?

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This complete sentence reads: “ Cloud cover is predicted with a prognostic-statistical scheme solving equations for the distribution moments of total water (Tompkins, 2002).” The fractional cloud cover of each grid box is prognosed from a prognostic representation of the subgrid scale variability of the total water via the distribution moments of its probability density function. This scheme and details of its implementation in ECHAM5 is complex. As this issue seems to be only of limited relevance for the impact of aerosol absorption and radiative forcing, we would like to refer the interested reader to the cited references.

The authors need to clarify the following: (1) How exactly are convective clouds versus stratus clouds treated

The treatment is very similar to the the one in most global circulation models. We realize that the separation of stratiform and convective cloud schemes was not fully clear in the model description and clarified this by adding “stratiform” to the following sentence (page 3, col 2, par 3):

“ ECHAM5 contains a bulk microphysical stratiform cloud scheme (Lohmann and Roeckner, 1996) with prognostic equations for cloud liquid water and ice.”

For clarification we further added the following description to this paragraph (page 3, col 2, par 3):

“Cloud water detrainment from convective updrafts is used as source term in the stratiform cloud water equations.”

(2) Are convective clouds subgrid scale or grid scale, and how many convective clouds can form in a grid cell during a time step

In the underlying (widely used) Tiedtke (1989) convection scheme, convective clouds are sub-grid scale, based on steady-state equations for mass, heat, moisture, cloud water, and momentum for an ensemble of updrafts and downdrafts. The equations are formulated for grid box mean mass fluxes and do not explicitly define the number of

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convective clouds or updrafts.

We have extended the description of the convective scheme in the manuscript to (page 3, col 2, par 3):

"Convective clouds and convective transport are based on the mass-flux scheme of Tiedtke (1989) with modifications by Nordeng (1994), based on steady-state equations for mass, heat, moisture, cloud water, and momentum for an ensemble of updrafts and downdrafts."

(3) Are clouds formed then dissipated each time step (e.g., are they equilibrium clouds or do they grow and evolve and travel each time step)

Clouds water and ice are prognostic variables, thus travel each time step as described above.

(4) Are clouds bulk (modal) or size-resolved (discrete size bins)

In the used model version, a bulk cloud microphysical scheme is employed with cloud water and ice as prognostic variables. Alternatively, also a version with prognostic cloud droplet and ice crystal number concentrations is available that has been coupled with ECHAM5-HAM in Lohmann et al. (2007). We additionally pointed out the usage of a bulk scheme for this study by adding "bulk" to the following sentence (page 3, col 2, par 3):

" ECHAM5 contains a bulk microphysical stratiform cloud scheme (Lohmann and Roeckner, 1996) with prognostic equations for cloud liquid water and ice."

One remark: this comment reads like modal schemes are equivalent to bulk schemes and size-resolved schemes equivalent to bin schemes. To clarify: "Bulk" commonly refers to schemes without representation of size that solely simulate the global distribution of the mass of a certain component (aerosol or water). Thus any information of the particle size or its distribution has to be prescribed. In contrast, both modal and bin-schemes prognostically represent the size-distribution of a population, of course

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with very different approaches.

(5) How do the bulk or size-resolved clouds interact with solar radiation?

The SW cloud radiative properties are explained in detail in Section 2.2.3 for both standard ECHAM5 and the extended version used in the CL- studies.

We extended the description of the ECHAM5 SW radiation scheme (page 3, col 2, par 3):

“The short-wave radiation scheme of Fouquart and Bonnel (1980) is employed with 4 spectral bands, one for the visible and ultra-violet, and three for the near-infrared. It uses the Eddington approximation for the integration over the zenith and azimuth angles and the delta-Eddington approximation for the reflectivity of a layer.”

(6) How is cloud fractio calculated?

See explanation above and for more details the cited Tompkins (2002) and Roeckner et al. (2003) references.

For balance, the authors should point out possible sources of error/uncertainty if the clouds are not subgrid scale (e.g., if one convective cloud forms per grid cell or if the clouds are treated with bulk microphysics rather than size-segregated microphysics).

Not applicable.

P. 7179. ’The microphysical aerosol module HAM predicts the evolution of an ensemble of seven interacting internally- and externally-mixed log-normal aerosol modes. Zhang et al. (Aer. Sci. Technol. 31, 487, 1999) found that with appropriate numerical algorithms and size resolution, a sectional representation can predict more accurate chemical composition and size distribution than a modal representation. The authors should mention that the lognormal assumption is a source of error in the ECHAM-HAM model.

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The simulation of aerosol processes on the global scale is complex. Thus, the majority of global aerosol models employ bulk aerosol schemes, without any prognostic representation of the size distribution. Only recently, size resolved multicomponent microphysical aerosol modules suitable for long term climate simulations become available. However, their application for global scale application remains the exception, as evident from the AeroCom aerosol model inter-comparison (see Textor et al., 2005). The application of computationally efficient modal aerosol microphysical schemes helps to maintain a balance in complexity between the numerous processes in general circulation models (other processes, such as cloud microphysics, are typically represented higher parameterized in global climate models) and provides the basis for long-term climate simulations.

So while we are fully agree that the modal representation in ECHAM5-HAM is a model, a simplified representation of the real aerosol system, as to some degree is the sectional representation, it can be seen as both: an advancement over the prevalent current state of the art in aerosol climate modeling or as source of error as compared to more complex representations (that are generally not yet computationally affordable for transient climate studies). We would like to leave this judgment to the reader.

P. 7179. "The microphysical core M7 calculates coagulation among the modes." This needs to be explained, since modes do not coagulate, particles of individual size coagulate. Please explain the treatment and mention that it is a potential source of error.

We have replaced this somewhat imprecise statement by (page 4, col 1, par 3)

"The microphysical core M7 (Vignati et al., 2004) calculates coagulation of aerosol particles, condensation of gas-phase sulfuric acid on the aerosol surface, binary nucleation of sulfate, and water uptake."

and would like to refer the interested reader to the Vignati et al. (2004) reference for more details.

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P. 7179. "using pre-calculated monthy mean oxidant fields." The use of precalculated fields would indicate that the model cannot calculate the climate response completely. This should be mentioned as another potential source of uncertainty.

We agree that the usage of pre-calculated monthly mean oxidant fields is a limitation for climate response studies. Therefore, we discussed this source of error in detail in the application of the model in a transient climate simulation (Stier et al., 2006). This simplification can be justified by the results of Pham et al. (2005) who investigated the effect of changes in the oxidation fields from 2000 to 2100 on the global sulfate distribution based on the SRES A2 scenario. They showed that the effect on the global mean sulfate burden is less than 1% and on regional surface concentrations about 5%.

For the present study, the meteorology is nudged to the year 2000 ECMWF ERA-40 reanalysis. Thus, no climate response is considered or analysed in this study. For future studies of this issue, a fully coupled aerosol-chemistry climate model ECHAM5-HAMMOZ (an extension of ECHAM5-HAM by the MOZART chemistry scheme) is available (Pozzoli et al., submitted).

P. 7179. How are lognormal mode radii determined? Do they vary in time or are they fixed? Can chemicals dissolve from the gas phase into different modes, changing radii? Do they do so competeitively (e.g., do several modes compete for the same gas). Same question for condensation. Does chemistry occur within modes?

The lognormal mode radii are determined by the governing microphysical processes of coagulation and condensation, as well as by cloud processing and size-dependent sink processes. The seven modes are grouped into four geometrical size-classes with fixed mode boundaries but varying mode radii. A repartitioning algorithm is applied to transfer particles consistently among the modes. Currently, sulfuric acid is the only condensible vapor included, no dissolution of other chemicals into the mode or chemistry

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within the modes is considered.

Given the already relatively detailed model description of the manuscript, we would like to refer the interested reader to the cited references, in particular Vignati et al. (2004) for details on the aerosol microphysical scheme M7 and Stier et al. (2005) for its implementation in ECHAM5-HAM.

P. 7180. How have clouds been evaluated against data (e.g., cloud fraction, cloud liquid/ice, cloud optical depth, precipitation). Wet deposition depends on precipitation, for example. What evaluation of precipitation rates has been performed?

The introduction of the aerosol module HAM has not modified the parameterizations of the base model ECHAM5. In fact, with the aerosol radiative effect deactivated, the ECHAM5-HAM model produces bit-identical results to ECHAM5. Therefore, the extensive evaluation of ECHAM5 also applies for this study: Roeckner et al., 2006, Wild and Roeckner 2006, and in particular the “Evaluation of the hydrological cycle in the ECHAM5 model” (Hagemann et al., 2006). See also the special section “Climate Models at the Max Planck Institute for Meteorology” in Journal of Climate, (2006).

Specifically, precipitation has been evaluated in Hagemann et al. (2006) for a serious of simulations following the AMIP protocol (the standard benchmark for climate simulations) with the global precipitation climatologies from the Global Precipitation Climatology Project (GPCP; Huffman et al.1997) and the Climate Prediction Center (CPC) Merged Analysis of Precipitation (CMAP; Xie and Arkin 1997).

The main effect of the introduced effect of absorbing aerosols on the cloud radiative properties (Sections 2.2.3 and 4.5) is adding a perturbation of the cloud single scattering albedo (the cloud optical depths are very similar), all other aspects of the cloud scheme are unchanged. Thus, the evaluation of the base model also applies for this sensitivity study.

P. 7181. The authors should mention specifically which previous studies have

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tested which mixing rules on a global scale.

For this part of the manuscript, the model description, we chose to cite a number of relevant references (Garnet 1904,1906; Bruggeman, 1935; Aspnes, 1982; Chylek et al., 1995,2000; Lesins, 2002) that deal with the mixing rules itself and their evaluation with explicit radiation models.

We had already listed a number of studies dealing with internal mixing rules in the introduction:

“The internal mixing of a population of black carbon with other aerosol components enhances the co-single scattering albedo of the population, and therefore the absorption optical depth, i.e. the column integrated extinction owing to absorption (Ackerman and Toon, 1981; Chylek et al., 1995; Jacobson, 2000; Stier et al. 2006c)”.

and further motivated our simulation setup with

“The internal mixing of a population of black carbon with other aerosol components enhances the co-single scattering albedo of the population, and therefore the absorption optical depth, i.e. the column integrated extinction owing to absorption (Ackerman and Toon, 1981; Chylek et al., 1995; Jacobson 2000; Stier et al., 2006c)”

As the differences in the other model representations or assumptions do not allow for a direct comparison of the results we do not feel that more details would serve the readability of the manuscript.

Does the model treat the effects of absorption on snow or sea ice reflectivity?

The model treats the absorption of solar radiation on snow and ice. However, it does not include the (uncertain) effect of black carbon deposits on the snow albedo. We would like to refer the interested reader to the recent study by Flanner et al., JGR, (2007) estimating a radiative forcing of this effect of 0.007–0.13 W m^{-2}.

P. 7190. ’For all simulations, the AERONET evaluation of AAOD shows generally

good agreement. The agreement should be quantified numerically with statistics.

We have given this issue some thought and concluded for the original manuscript, as given in the result section, that " The incomparability of different stations and the consideration of a highly variable number of measurements at each site do not seem to allow deriving a global measure of the overall model performance."

Instead, we chose to include the scatterplots of simulated versus retrieved daily mean AAOD in the Appendix and the Supplementary Online Material, that give an exact representation of every single measurement/simulation contributing to the plots of the local monthly mean values in Fig. 3–6.

P. 7190. How is absorption irradiance changes (W/m2) separated out from total irradiance changes?

The separation of the irradiance change owing to absorption from the total irradiance change owing to absorption and scattering is tricky and rarely done. Therefore, we specifically designed the sensitivity study TRANS to isolate the effect of absorption through setting the imaginary part of all aerosol refractive indices to $n_i = 10^{-9}$, i.e. making all aerosol virtually non-absorbing. The difference between TRANS and the other performed studies allows to quantify the effect of absorption in the respective studies.

The instantaneous atmospheric aerosol radiative forcing (aerosol absorption) is calculated as the difference of instantaneous TOA and surface forcings. The details of the forcing calculations are described in Section 2.2.5.

P. 7192. The minimum and maximum surface albedos in Figure 2 (0.18 and 0.36) seem to be extreme variations. Why not use realistic variations in surface albedo?

Indeed the variations of the surface albedos in the Figures seem to be extreme,

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and this is partly the point of this sensitivity study. While the differences seem unrealistically large, the fields have been derived from actual submissions to the AeroCom aerosol model inter-comparison and therefore represent the spread in currently used assumptions for aerosol radiative forcing estimates. We demonstrate through this study that the spread in basic model assumptions alone can explain a nonnegligible part of the spread in the resulting aerosol radiative forcing estimates. This is a key motivation for the proposed AeroCom Prescribed model/satellite intercomparison experiment that aims to isolate the contribution of basic model assumptions through prescribing identical aerosol radiative properties for all models/retrievals (http://wiki.esipfed.org/index.php/AeroCom_Prescribed).

P. 7197. From this evaluation we conclude...provide the best representation" The authors should qualify this result to say that it is specific to the model used and its assumptions. Other models or changes in treatments of physical processes in the present model may yield different results.

This is a good point that we already partly addressed in the complete original sentence in that we say that the "BB-M and the similar BRUG simulation seem to provide the best representation", therefore already specifically refer to our results:

" From this evaluation we conclude that the BB-M and the similar BRUG simulation seem to provide the best representation of the AERONET retrieved AAOD – for the black carbon emissions used in all simulations (Bond et al., 2004)."

To avoid any ambiguity we now specifically state "– for the black carbon emissions used in all ECHAM5-HAM simulations (Bond et al., 2004)" (page 11, col 1, par 1)

P. 7199 "As previous studies indicated, the simulated effect of absorption by aerosol inclusions embedded in cloud droplets and ice crystals on the global radiation budget is small." The authors have not referred to any previous studies examining the global effect of embedded inclusions. The authors should specify which studies are being referred to.

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We did actually refer to previous studies examining the global effect of embedded inclusions in the following section of the introduction (page 2, col 2, par 3):

"It has been estimated that absorption from embedded aerosols could be a non-negligible contributor to short-wave cloud absorption, potentially reaching up to 1–3 $\mathrm{W\,m^{-2}}$ (Chylek et al.,1996) in the global annual-mean. In their lower estimate Chylek et al. (1996) have found relatively small global impact, in agreement with other studies (Chuang et al., 2002; Liu et al., 2002); however, as pointed out in Chylek et al. (1996), the underlying assumptions about the BC abundance in their work and the work of Liu et al. (2002) might not be applicable for highly polluted areas."

We now additionally added the Chuang et al. (2002) and Liu et al. (2002) citations to the quoted sentence so that it reads now as (page 11, col 2, par 2):

"As previous studies indicated (Chuang et al., 2002; Liu et al., 2002), the simulated effect of absorption by aerosol inclusions embedded in cloud droplets and ice crystals on the global radiation budget is small."

Figure 7. The figures are too small to see anything useful. I suggest reduce the number of figures significantly and increase their size to illustrate a specific point.

We fully agree that the figures appear to small in the formatting of ACPD. We have prepared them for a full page in the ACP format and tried to convince the EGU production to spread the Figure over a full page in the ACPD print version. Unfortunately, this was unsuccessful. The figure is significantly larger in the revised version in ACP formatting.

Figure 8. This figure seems not to be so useful. I would suggest removing it.

We believe that this figure is valuable for the interpretation and understanding of the spatial distribution of the forcing differences given in Figure 9.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 7171, 2007.