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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis and quantification of the diversities of aerosol life cycles within AeroCom" *by* C. Textor et al.

C. Textor et al.

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Dear referees,

Thank you very much for your comments. Please find detailed answers below.

Best regards,

Christiane

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Referee #1

Page 8334, line 7: briefly provide the reason for this.

We are reluctant to do so, because we do not have a definite explanation. We give however three hypothesis in section 9.1

Page 8336, line 15: 'AER=total aerosol' mention here that it concerns the dry aerosol and provide also abbreviations for H2O and for the wet aerosol that have to be used also in the caption of Figure 9. ok

Page 8342, last line: An additional problem is that the participating models are not fully independent one from the other, as also shown in the description tables and discussed later in the analysis of the results.

Very important statement. We added a sentence already in line 16: An additional problem is that the participating models are not fully independent from each other, as shown in the tables with the model descriptions, and discussed later in the analysis of the results.

Page 8344, equation 3, numerator : replace 'result' by 'individual model result' done, also added in eq. 1

Page 8344, line 13: close parenthesis ok

Page 8345, line 24: ECMWF ok

Page 8346, line 9: 'emissions' do you mean 'size of emitted particles'? no: This is especially the case in ARQM, where the emitted SS mass is more than one order of magnitude larger than in the other models.

Page 8347, lines 20- 21: This means that the total chemical production of the aerosol component is better constraint than the individual pathways. yes, this is what I mean.

Page 8350, equation (6): crosses should not be exponents Equations (4) and (7) take care of the italics. ok

Page 8352, line 17, 75% should be 76% to agree with Table 10. ok

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Page 8353, line 6, ‘the sum’ add ‘of’ ok

Page 8354, lines 15 & 16 : ‘larger’ do you want to say ‘smaller’? Sentence deleted.

Page 8354, line 22: ‘due’ add ‘to’ ok

Page 8354, line 27: ‘weak correlations’ positive? reanalysed and changed to

We do not find clear correlations between the precipitation rates and the contributions of wet deposition to the total removal, nor between the precipitation rates and the wet deposition rate coefficients.

Page 8356, line 18: ‘the demand’ which one? Explain better.

The description of the particle size distributions depends on the type of the scheme (bulk, modal, or spectral, see section 2.2 and Table 2) and on the number of prognostic variables (see Table 4). The attribution of aerosol mass to three size ranges (diameters of d<1m, 1m <d<2.5m, and 2.5m <d) was compiled within the AeroCom exercise. The modelers distributed the mass in different ways on these size ranges, based on their representation of aerosol sizes. For spectral schemes, the bins within the intervals were simply summed up. In this case the results can be somewhat misleading for schemes with only a few size classes, if these are situated close to the interval boundaries. For modal schemes some participants have used the mass median diameter for classification of the size, and others have more accurately integrated over the distribution within the interval boundaries. In addition, the specific contributions of the accumulation, Aitken and nucleation modes to the fine fraction are not resolved within the AeroCom diagnostics. Furthermore, the results do not document the differences regarding the largest simulated particle sizes, which is especially important for ‘natural’ aerosol as mentioned in section 4. Despite these reservations the analysis of AeroCom data provides for the first time an overview of the actual size distributions of dry particles in different models and allows for a general view of the diversity of particle sizes in current

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aerosol modules. For simplification, we focus here on the split of 'fine' (d<1m) and coarse (d>1m) mode particles. In Figure 8a, we show the mass fractions of particles in the fine mode, and Figure 8b shows the corresponding model diversities, the numbers are given in Table 10. We concentrate on mass fractions rather than on total burdens in order to remove the effects of contrasting burdens.

Page 8359, reference to Figure 9: Figure 9 is not correct, (Figure 8 is shown twice) figure corrected and modify caption to fit the aerosol component abbreviations listed in the page 8336.

All figures updated, colors changed removing all potential red/green confusions. Text changed

Page 8360, 2nd paragraph: Please rephrase this paragraph. It is not clear what the authors want to point out here, what is the significance of this no correlation?

Water uptake should increase with increasing local relative humidity for an aerosol of given composition. We compared the simulated optical-depth-weighted relative humidities and found a range of grid cell averages between 55% and 77%. There was no relationship between these relative humidities and aerosol water masses for the ensemble of AeroCom models (not shown). However, several models use a local, sub-grid scale relative humidity instead of the grid cell average. The various methods to obtain the local relative humidity represent additional sources for the model diversity (see Table 9).

Page 8360, line 13: particles. ok

Pgae 8363, lines 3-5, explanation of fig 11a: use this explanation to improve the figre caption. ok

Page 8363, line 12: simulated precipitation and nudging of ECWMF data as mentioned in page 8346, line 2 could be also reason for the discussed deviations.

I agree. Text modified Assuming that the differences in precipitation resulting from

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slightly different nudging constants in these two models are small, we can mainly attribute the deviations of their vertical dispersivities to differences in the parameterizations of aerosol processes.

Page 8364, line 11: The authors could refer to the recent review paper on organic aerosol published in ACP this year that provides some estimates on the contribution of the secondary organic aerosol to the organic aerosol mass in the free troposphere - Kanakidou et al. SRef-ID: 1680-7324/acp/2005-5-1053.

paper cited in section 4 (sources)

Page 8364, lines 15-16: What is the origin of the differences, the wet deposition or the vertical dispersive? Obviously the one could cause the other. Here the authors mention that the vertical dispersive is at the origin of the differences in the wet deposition whereas in the conclusion there is in a more general statement that leaves the question open (page 8368, lines 17-20). Some rephrasing here might help the reader.

We did not mean that dispersivity is in general the reason for the differences in wet deposition, but mentioned it as one of the potential reasons in three models: There are several reasons for the differences of the simulated vertical distributions of BC and POM within a given model. The formation of secondary POM from organic precursor gases within the atmosphere, if included in the model, increases the amount of POM at greater heights. At the same time, the greater solubility of this species leads to an increased wet scavenging rate, thus reducing high-altitude-POM. Finally, divergent spatial distributions of the two species resulting from transport influence their deposition rate coefficients, which in turn affect the spatial distributions. Weaker vertical dispersivity of BC than of POM could explain the faster wet deposition rate coefficients of this species in three models (LOA, LSCE, MATCH). However, not all models with weaker vertical dispersivity for BC show faster wet removal rate coefficients for this species.

We feel that our text is quite clear and do not think that rephrasing would help here.

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Page 8365, line 15: As I translate Figure 6a, the model diversity of the wet deposition rate coefficients for sulfate is slightly lower than for BC and POM.

The reviewer is completely right. The text is based on an earlier version of model submissions and analysis, when different results were included in the calculation of the diversity. I apologize for this inconsistency. Text corrected.

Page 8366: Mass fractions at the poles depend also on the source location. Is it possible that model differences in source for instance boreal forest emissions of BC contribute to the diversity between models at the top of the long rage transport mechanism?

Perfectly possible, text changed to: We assume here that the differences in the simulated spatial distributions of the sources have no influence on our analysis. This assumption is justified as our analysis on the AeroCom experiment with unified sources demonstrates Textor et al. 2005.

Page 8368: line 4: residence times: could you give range deduced from AEROCOM results? Unfortunately the aerocom data do not allow for the calculation of such ranges because the source and sink fluxes are only given as column data.

Page 8369, line 4: remove ‘are’ ok

Page 8396: Table 3: ‘using a factor’ provide it if constant. the factor is given in Griffin et al., text changed to included in POM, terpene + NVOC from GEIA database of, then adjusted to SOA emi according to Griffin et al., 1999

Page 8405: Table 9: for the LSCE model information on RH max is missing. added

Page 8406: Caption of Table 10. Close parenthesis. closed

Last line of Table 10 in page 8407: ‘aerosol’ ok

Figure 1 - caption mentions ‘199% and 176%’ whereas on figure 1b the numbers 625 and 550 are shown. corrected

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Figure 6: similar problem as for Fig. 1, numbers in caption do not fit with numbers in the figures. corrected

Figure 7: caption line 2 ‘global annual convective add ‘wet deposition’ changed

Figure 9 (see comment above). changed

Figure 10: Please clarify if it is dry aerosol (AER) or wet aerosol. clarified, it is dry aerosol. We also added a sentence in the methodology - section 3: AER denotes total dry aerosol mass, i.e., the sum of the five aerosol species included in this study. Total aerosol is examined here because many observations refer to bulk properties, such as mass and volume measurements, or aerosol optical depth. The terms ‘ambient’ or ‘wet’ aerosol’ describe AER+H2O.

Figure 11: rephrase caption as explained in the text ok and also in fig 12

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Referee #3

Pp 8333, line 19: The burdens of dry masses decreases from largest to smallest: ... Rewrite: The burdens of dry masses in decreasing order are:... changed

Pp 8334, line1: Here the term diversities is already being used where it is defined later in the text, or least explained why they use diversity rather then uncertainty. Could you not use in the beginning simply “differences” and define already in the introduction what you mean with/the motivation to use diversities?

Abstract changed: Simulation results of global aerosol models have been assembled in the framework of the AeroCom intercomparison exercise. In this paper, we analyze the life cycles of dust, sea salt, sulfate, black carbon and particulate organic matter as simulated by sixteen global aerosol models. The differences among the results (model diversities) for sources and sinks, burdens, particle sizes, water uptakes, and spatial dispersals have been established. These diversities have large consequences for the

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calculated radiative forcing and the aerosol concentrations at the surface. Processes and parameters are identified which deserve further research.

We prefer not to change the structure of the current text, because the term 'diversity' can be used as a synonym for 'difference', or 'scatter' in common speech, without the specific definition we use to quantify model diversity. We firstly describe the models and then the method we apply for the analysis including the introduction of our definition of the model diversities.

Line 8/9: similar for a given species: what is meant with that? changed to 'for the individual species'

Line 16: "acid rain and toxic chemicals"; Change to "acid rain and contamination with toxic chemicals such as.. ", could you give an example of which toxics you refer to?

We are referring to 'contamination with toxic chemicals such as heavy metals.' The sentence has however been shortened to 'Atmospheric aerosols play a key role in many important environmental issues including climate change, stratospheric ozone depletion and tropospheric air pollution.' in order to straighten the text.

Pp 8335, line 1-3: The sentence is quite vague, embarks? multi-angle strategy? Suggest to change it to "AeroCom aims at evaluating the performance of global aerosol models by intercomparisons and by comparison with observations"

The approach of AeroCom is twofold: The performance of global aerosol models is evaluated by intercomparisons with each other and by comparisons observations of aerosol properties and processes.

Line 8: it would be better to already indicate about what kind of diagnostics are involved here.

Within AeroCom the diagnostics have been greatly extended and allow now for the analysis of aerosol life cycles in the different models, because information about sources and sinks, particle sizes, aerosol water, and others are included.

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Line 11: The statement about “constraining” raises a lot of questions. How are the models constrained with observation data? From what I inferred from the model descriptions are the models mostly run as they are, without forcing them, except maybe of the meteorology in the nudged model simulations. But are there also constraints imposed on the aerosol processes? If so, then this should be described.

We clarified the text: All global aerosol models taking part in this study have been carefully validated when the model authors compared them to various high-quality observational data sets. These included in-situ measurements of aerosol concentration, size distribution, and chemical composition, lidar measurements of the vertical distribution of the aerosol extinction coefficient, sun photometer measurements of the aerosol optical depth and column size distribution, and satellite measurements of the spatial distribution of the aerosol optical depth. Please refer to the literature about the models cited in table 2.

In addition, we hope the sentence ‘The simulations have been performed with the models in their usual configuration (AeroCom experiment A).’ some lines below excludes further misunderstandings.

Line 27-28: the sentence should be reformulated: It is not clear what is meant with “on all processes except for one, which is under investigation”

The quantification of model diversities facilitates identifying weak components where research is needed in order to improve our understanding of global atmospheric aerosol. It is not the objective of this work to judge the different ways of modeling the aerosol life cycle. This would necessitate investigating individual processes (e.g., water uptake) or concepts (e.g., representation of the size distributions). As these are strongly interrelated, several full sets of sensitivity simulations with strong constraints on all components except for the one under investigation should be performed. However, such studies are not feasible in the context of a volunteer based model intercomparison such as AeroCom, and because of the large differences of the participating

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models.

Pp 8338, line 13: “to investigate the global models” I guess you should chance that to “to investigate the host models”. Instead of using host models, an alternative could be “driver models” very good idea! ‘host’ changed to ‘driver’ in entire document

Line 21: Not all the readers might be familiar with the term nudging and consequently this should be explained in a little more detail and/or including a reference that describes the technique.

Explication added: The application of nudging techniques to GCMs allows them to closely represent observed weather patterns. Nudging signifies a Newtonian relaxation technique when an additional term is added to the model’s equations at each time-step to force them to a specific weather period (Jeuken et al., 1996).

Page 8340, line 5: turbulent dry deposition. Throughout the document you use this term but within the different contexts it gets confusing. Why don’t you simply write it down as “wet and dry deposition and sedimentation”. I assume that you used the term turbulent dry deposition since in the model the split is made between sedimentation and the dry deposition due to turbulent transport, Brownian diffusion and impaction where in the real world measuring a dry deposition flux would include all four terms (and resuspension/re-bouncing). If this is indeed the motivation to use this definition you could include a statement: “Note that hereafter dry deposition refers to surface removal due to turbulent transport, Brownian diffusion and impaction excluding the contribution by sedimentation which is generally considered separately in the model analysis”

Changed throughout the paper; sentence changed to: Furthermore, the aerosol modules describe the sources of aerosols and their removal processes. Most models distinguish between three removal pathways: wet deposition, dry deposition, and sedimentation. Note that hereafter dry deposition refers to surface removal due to turbulent transport, Brownian diffusion and impaction, excluding the contribution by sedimenta-

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tion which is generally considered separately in the model analysis.

Pp 8342, line12: See previous comment: Could you move this statement forward? See comment above

Pp 8347, line 8: “SO2 stems from emission datasets”; weird sentence. Rephrase to something like “The SO2 emissions are prescribed based on anthropogenic emission inventories (e.g., EDGAR, refer ˇ E) whereas DMS emissions are calculated online from global oceanic DMS concentrations fields and sea-air transfer coefficients as a function of windspeed” done!

Pp 8348, equation 4 and some other equations; the fonts are not consistent (normal versus italics) Ok

Pp 8350, line 1, see previous comment; wet and dry deposition and sedimentation Ok

Pp 8355, line 8; (table 8), remove parenthesis changed throughout the paper

Line 17-18: This sentence is not clear to me: It is meant to express that you would expect a different order in species with respect to the relative contribution by convective rain due to for example their solubilities. So what you mean with a consistent sequence? wording changed to: Ěmodels do not agree on the order in species along which the contribution of convective rain increases.

Pp 8356, line 4; In the next Section, we examine... wording changed

Pp 8358, line 1: “we do not explore in this paper” Will there then be another paper on this, one that is already in preparation? There is a paper on radiative forcing in preparation, but it is not sure that size effects will be discussed. Therefore it is not cited here.

Line 27-30: “This can be due to concerns about the particle size data in AeroCom..” How does the concern explain the lack of correlation between dry deposition coefficients and the mass fractions? We explained this in the paragraph above.

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Coming back to one of the previous points: What do you with particle size data? Are these some of the data used to constrain the models? The models have not been 'constrained' in any way the reviewer understood this term. With 'constrained' we meant 'validated' and he understood some constrains of the AeroCom experiment. We tried to clarify this in the introduction and with the following sentence: This can be due to the concerns about the particle size data available for analysis within AeroCom discussed aboveĚ.

Pp 8362, line 11-12: The statement about dry deposition being underestimated above suggests that dry deposition also occurs higher up in the PBL, which is not the case. Dry deposition is the removal at the earth surface where models to describe this process also include the turbulent transport from the reference height in the surface layer (Ÿ 10% of PBL depth) to the surface where the removal occurs. I guess you want to express that there is a possible overestimation of surface removal (dry deposition) and an underestimation of the downward turbulent transport in the PBL to compensate for the efficient surface removal. In the previous sentence you express somehow that this problem is related to the representation of turbulent transport in the models, which is correct with the models failing to reproduce observed turbulence features. But so, dry deposition is more then turbulent transport. The dry deposition of accumulation mode particles is controlled by the Brownian diffusion, being the limiting parameter. It is also interesting to think about having a problem in the opposite direction, associated with the operator splitting problem. There could be potential too fast upward transport of the emitted species if you include for example the emission flux as the lower boundary condition in your vertical flux calculations explaining the possible underestimation of surface layer concentrations. Anyhow, an important issue is addressed here about the role of surface exchanges in controlling surface layer concentrations which are used for the model evaluation. This stresses the importance of further tackling this problem in follow-up studies where a main limitation is imposed by the lack of direct aerosol flux observations. This is something that could be stressed in the conclusions/recommendations for further research.

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Interactive Comment

changed to: Firstly, the different parameterizations of dry deposition and their temporal and vertical integrations might play a role in causing this minimum. The dry aerosol removal at the Earth surface also includes the turbulent transport from a reference height in the surface layer (˜10% of planetary boundary layer (PBL) depth) to the surface where deposition occurs. Overestimation of surface removal together with underestimation of downward turbulent transport in the PBL, which would compensate for the efficient surface removal, could cause a minimum close to the surface. In contrast, there could also be potential too fast upward transport of the emitted species explaining the possible underestimation of surface layer concentrations.

remark added to the conclusions: In some models, a minimum of the aerosol concentration is simulated in the lowest model layer. As potential reasons for this minimum we propose the parameterizations of surface exchange processes by turbulent transport, dry deposition, as well as the use of operator splitting methods to solve the advection-diffusion-processes equation. Further research is needed to clarify the reasons for the simulated minimum of aerosol concentrations at the surface, especially, because they are compared to observational data for model validation.

Pp 8368, line 14: Rephrase to; “Surface and boundary layer turbulence and surface cover properties largely control aerosol dry deposition” changed

Pp 8369, line 1: If you talk about residence times should it not be longest and shortest instead of greatest/least changed

Pp 8370, line 5-7: “disagreement on the particle sizes, and possibly also to the application of operator splitting technique...” Coming back to the previous comment; has it been discussed in the AeroCom community how to address this problem? It would be an option to include here a suggestion how to proceed to tackle this problem, e.g., using a selection of the models that give very contrasting results in terms of the vertical dispersion in case studies changing the sequence of the calculation of surface exchanges, vertical mixing, wet deposition, etc. but also comparing simulated boundary

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layer aerosol profiles with field observations (when available).

sentence added: Further analysis of vertical aerosol profiles and surface concentrations including a comparison of models to data is provided in Guibert et al. 2005???.

Pp 8372, line 3: remove the statement “(and this is not trivial)” okĚ

Line 10-15; Closing statement suggestion: “Several processes and parameters, which are particularly relevant for aerosol radiative forcing calculations, with high diversities are: - Masses.. - ..dispersal Consequently the improved representation of these processes and parameters in large-scale aerosol models deserves a high priority to reduce the uncertainty of the..

done

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