Atmos. Chem. Phys. Discuss., 8, S9170–S9177, 2008
www.atmos-chem-phys-discuss.net/8/S9170/2008/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global error maps of aerosol optical properties: an error propagation analysis" *by* K. Tsigaridis et al.

K. Tsigaridis et al.

Received and published: 16 November 2008

We would like to thank the anonymous referee for his comments. Detailed answers to the explicit points mentioned follow:

1) From the title 'Global error maps of aerosol optical properties: an error propagation analysis' to the conclusions the impression is given that the investigated sensitivities would yield 'error' in an absolute sense. Limitations in chosen variations of the parameter space are not discussed sufficiently. However, it is clear that the analysis does not test the full range of parametric uncertainties (and it cannot be expected). This fact seems to be almost concealed by statements like 'It has to be noted that the error calculated by the present work is not the full uncertainty on the optical properties of a given model simulation on a given day, since it is based on monthly mean reference fields. Further, since it uses the output of a particular model (LMDz-INCA) as refer-

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ence, it is expected to contain a systematic bias characteristic to that model.' (page 16030). One example of those limitations is that previous work indicates that e.g. the choice of the mixing rules (concentric or volume weighed) has a considerably smaller effect on atmospheric absorption than the actual choice of the uncertain black carbon refractive index - that is not part of the sensitivity studies. Further, as all calculations are only performed at a wavelength of 550nm the analysis is completely ignoring the uncertainty in the infrared - that might be highly relevant for the larger natural species, in particular for mineral dust. As interesting some of the results are, above limitations should be clearly reflected in the manuscript and this should start with a title like 'Sensitivity of aerosol optical properties to variations in ..'.

The reviewer is right. By inserting the sentence he/she quoted, our aim was exactly that, to state the limitations of our work. We agree that additional discussion could be added. The suggestions from the reviewer regarding additional sources of error will be included in the abstract and in the discussion section. The wavelength dependence is of course a major issue, especially for the radiative forcing calculations that we had listed as 'future work'. Taking into account the other reviewers comments, we decided to include some illustrative radiative forcing calculations in the present paper, thus more wavelengths will be studied.

Concerning the title of the paper, we believe that it is already sufficiently vague and that it does not claim that we have solved the problem, but that we rather did 'an' error propagation analysis. We hope the reviewer agrees.

In addition, we will replace the words 'uncertainty' and 'error' in the text and define and use rather the wording 'partial uncertainty' and 'partial error', indicating that the errors we calculate are only related to a reasonable choice on variation in size, mixing and hygroscopic growth. The reviewer thankfully points to the general problem of how to compare error on different parameters. Although the present work was initially intended to provide a model error map for the optical depth to be used in an assimilation system, during the process of doing the computations we found it also interesting to state the

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consequences for single scattering albedo and asymmetry parameter variations. The paper has been carefully revised throughout to reflect this intention. We have tried to prevent this way the false impression that the general uncertainties with respect to forcing are just on AOD. We totally agree that refractive index of BC or dust and their spectral dependence is ultimately the larger source of uncertainty for forcing estimates.

2) The choice of the chosen measure of 'error', the standard deviation of AOD, SSA and g as derived from the parametric ensemble, does not allow for the direct comparison that the manuscript suggests: 'The AOD uncertainty range calculated maximizes to 70%, while for g and SSA the uncertainty reaches 18% and 28% respectively' in the conclusions and similar in the abstract seems to suggest that the uncertainty of AOD is larger than for SSA and implicitly for absorption. However, if the arguably more relevant co-single scattering albedo or absorption optical depth would have been used instead of SSA, these numbers - and conclusions - would be entirely different. It should be clear that those variations in SSA could be fully sufficient of switching the sign of the associated direct radiative forcing.

See our response to the reviewer comment above.

*3) This might or might not be a major issue but it is not clear to me how the radiative properties are actually derived from the three-dimensional fields. A simple vertical average over SSA or g would not make sense as it gives equal weight to every if so negligible AOD. The vertical averaging should normally been done with AOD (SSA) or AOD*SSA (g) weighting - but it is not explicitly mentioned that this has been done. If this has not been done, any resulting conclusions are of little value.*

The averaging was indeed done as the reviewer suggests, otherwise the result would be incorrect. We will explicitly mention this in the revised manuscript.

4) The global annual mean aerosol optical depth (550nm) in the reference case is about half of what current satellite or AERONET based estimates give. This indicates some systematic biases in the model that has not been addressed in this or other cited

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papers. It would be useful to provide the reader with some guidance, e.g. in form of a comparison with satellite retrieved AODs in Fig. 5.

A comparison with AERONET measurements will be included in the revised manuscript, also motivated by the comments of the other reviewers. The main reason for this underestimation is currently thought to be our aerosol water calculation. This is not expected to alter the relative error calculations much though (see also reviewer #2, point #4). However we will detail this further in the revision.

5) It seems that little effort has been made to put the manuscript into context of current literature. The overall number of references is very low and many sensitivity studies in the specific area of estimating aerosol model uncertainties (often covering other relevant parametric uncertainties) are not even mentioned (e.g. Spracklen et al. (2007) investigate the sensitivity to primary sources and cloud processing; Spracklen et al. (2006) the sensitivity to boundary layer nucleation; Pierce et al. (2007) the sensitivity to primary emissions and processes; Stier et al. (2007) the effect of internal mixing not statically on the radiative properties as done here but also considering the associated enhanced washout - and there are many others to mention).

We thank the reviewer for this reference list that we will include in the revised manuscript. Please note, however, that we have elaborated and documented just a 'partial uncertainty' which may justify that we have neglected some work on aerosol modeling uncertainties with respect to other parameters.

6) page 16029, line 14: Large particles scatter... This discussion makes more sense in the context of the Mie size parameter.

The reviewer is right on that point. We will add the phrase 'for a given wavelength' at the end of the sentence.

7) page 16029, line 23: 'Very few models assume that BC is always an inner core of soluble material (Lesins et al., 2002; Stier et al., 2005; Jacobson et al., 2007)' This is

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not true for some of the cited references - please be more precise.

The reviewer is right, the word 'always' should be removed from that sentence.

8) Introduction and Section 2.2: Some previous work of direct relevance does not seem to be considered. It seems worth pointing out that the sensitivity of aerosol radiative properties owing to uncertainties in the BC mixing state has been addressed in a number of relevant studies (Ackerman and Toon, 1981; Chylek et al., 1995; Jacobson, 2000; Stier et al., 2007) that are barely addressed. The topic of mixing rules beyond core/shell and simple volume weighted mixing is not touched and the limitations of the assumption of concentricity in the core-shell method has not been fully addressed. In fact, both volume weighted mixing and core-shell are unlikely to occur in nature - so what might me more realistic?

We disagree that an absorbing core in some transparent shell does not occur in the real atmosphere; we do agree though that the concentric sphere model is an oversimplification against a randomly-placed core. We also agree that the volume-weighted mixing has no direct physical meaning; this was selected on purpose, since this is the approach that most models use. Both of these issues are already addressed in the manuscript quantitatively (page 16035, lines 1-5). We will increment our discussion of this topic by refereeing to the work as suggested by the reviewer.

9) page 16031, line 5: The cited reference Kahnert et al. (2007) is missing.

We apologize for the omission and will add the reference to the revised version.

10) page 16033: Why is the asymmetry parameter not explicitly averaged over the log-normal distribution as this is normally done?

In order to go from the asymmetry parameter of a monodisperse aerosol population to the asymmetry parameter of a lognormally distributed aerosol population, the averaging should be weighted by the scattering of the monodisperse aerosol bins in the lognormal distribution, as described by d'Almeida et al. (1991); thus, Eq. 7 must be

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used.

11) Section 2.2: The chosen mixing scenarios are very specific - and it is unclear if they actually cover the uncertainty range or reflect reality. Recent measurements provide new insights into mixing (Schwarz et al. 2008) in terms of coating thickness that should be taken into account or at least clearly put in the context.

We thank the reviewer for bringing to our attention this very recent study (published less than a month before our actual submission); we will include it in the revised manuscript.

12) Section 2.2, page 16036, line 10: No details about the aerosol water uptake calculations, their evaluation and uncertainties are given. Thus, it is unclear how the actual limits in the uncertainty calculations are chosen.

We will include a description of how the model calculates the aerosol associated water mass in the revised version.

13) Section 2.3, page 16036, line 24: 'The chemical composition of aerosols together with their size and water content are the main parameters needed for the calculation of the aerosol optical properties.' This assumes that the refractive indices are known - which is not necessarily true (Bond and Bergstroem, 2006).

This is already stated in the manuscript: page 16031, lines 11-12 and page 16046, lines 1-2. We will rephrase these parts to make the reviewer's point clearer.

14) Section 2.3, page 16037, line 3: It is unfortunate that the 'reference' case does not refer to a detailed publication that would allow to understand the details of the chosen parameterisations.

A description of the aerosol module of the LMDz/INCA model will be included in the revised manuscript, also following the comment #7 of the anonymous reviewer #3.

15) Section 2.3, page 16037: The choice of the uncertainty ranges seem somewhat arbitrary and potentially to narrow: Any calculation of external vs. internal mixing of e.g.

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black carbon requires to make strong assumptions about the size distribution. In reality, mixing is typically part of growth processes (coagulation, condensation). Therefore, it can be expected that externally mixed particles are initially smaller than after mixing. It is unclear how the actual size distributions for the external / internal scenarios are chosen - and how the related uncertainties affect the results. Why is the spread in the radii so limited, given that you cite a much larger spread in AeroCom? Also the spread in the water uptake seems to be not very big (±50% by volume corresponds to a not so large change in radius, owing to the cubic dependency, uncertainty in growth factors can often be bigger). Realistic uncertainty ranges should be derived based on data or literature.

We agree that the choice is somewhat arbitrary. However, this also due to the observational data base which is hard to oversee. Note that the model results which we take as basis for the calculation introduces some realistic variance in the initial conditions of the partial uncertainty calculation. We will add a more detailed discussion on the choice of the ranges, based where possible on observations.

16) page 16038, line 13: The link to the data is wrong (not in the text).

We apologize for that; the link is now working.

17) page 16044, line 2: 'The annual mean uncertainty range reaches a maximum of about 58% above the Saharan desert,' It would be worth mentioning here that the actual uncertainty of AOD in this area is most likely dominated by the uncertainty in dust emissions itself.

This comment will be included in the revised manuscript.

18) Conclusions: The drawn conclusions are far too strong to my opinion, given the above discussed limitations in the set up of the study, most importantly the limited range of parametric uncertainties covered. Further it is not clearly described here what has and has not been done. It is stated that the calculations have been done for 'all

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possible mixtures' - while the actual mixing scenarios were quite specific and limited.

The phrase 'all possible aerosol mixtures' will be changed to 'all possible aerosol mixtures in the framework of the three mixing states studied here'.

19) It is stated that small absorbing aerosols and BC are the dominant absorbers - while this has not been explicitly analysed in the manuscript. As discussed above, the chosen measure of uncertainty might be misleading leaving the impression (to non experts) that absorption is better understood than AOD itself.

We will clarify this in order to state that small absorbing aerosols are the most sensitive ones to altering their absorbing capabilities with mixing state changes.

20) Figures: The size of the labels and color bars is too small, even for the expected increase in size with ACP formatting.

We will increase the size of the letters and color bars.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16027, 2008.