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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

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1 General Remarks

In their manuscript "Global error maps of aerosol optical properties: an error propagation analysis" K. Tsigaridis et al. present studies with the LMDz-INCA model to investigate the sensitivity of aerosol radiative properties to variations in three model parameters: aerosol radius, representations of the aerosol mixing state and aerosol water uptake. "Error" in this context is expressed as the variability of simulations under the variation of the model parameters.

The quantification of model errors is a highly relevant issue in current aerosol and climate research well in the scope of ACP and the presented work is an interesting step in the right direction.

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Discussion Paper

However, the manuscript has some major and minor issues that to my opinion need to be addressed before publication.

2 Major Issues

- 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 reference, 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 550ñm 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 choice of the chosen measure of "error", the standard deviation of AOD, SSA

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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 conlusions 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.

- 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 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 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.

- 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

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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).

3 Specific Issues

- **page 16029, line 14**

  Larger particles scatter... This discussion makes more sense in the context of the Mie size parameter.

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

- **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?

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- **page 16031, line 5**

  The cited reference Kahnert et al. (2007) is missing.

- **page 16033**

  Why is the asymmetry parameter not explicitly averaged over the log-normal distribution as this is normally done?

- **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.

- **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.

- **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).

- **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.

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- **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. 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.

- **page 16038, line 13**

  The link to the data is wrong (not in the text).

- **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.

- **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

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

- **Figures**

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

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