Geosci. Model Dev. Discuss., 8, C4324–C4329, 2016
www.geosci-model-dev-discuss.net/8/C4324/2016/

Geoscientific Model Development Discussions

Interactive comment on "Coupling aerosol optics to the chemical transport model MATCH (v5.5.0) and aerosol dynamics module SALSA (v1)" *by* E. Andersson and M. Kahnert

E. Andersson and M. Kahnert

emma.andersson@chalmers.se

Received and published: 24 March 2016

Below you will find our answer (in black) together with the original comments (in blue). The references to pages and line numbers in this answers refers to the supplementary document (which also contains this answer).

Thank you for taking your time to review our paper. Your comments and feedback has helped us improving our paper. Below you will find answers to each of the comments we received, where the your comments are marked with blue and our answers below in black.

1 General comments

1. I had the general impression that the authors were crediting substantially the author's work, while a larger relevant literature exists. I recommend the authors to cite more independent work from other research scientists in the field, as it would strength the manuscript itself and the external perception.
   Since this manuscript is not a review paper, it was not our ambition to provide a complete list of references on black-carbon optics. Rather, this manuscript is a model-development paper, so it is both natural and necessary that we cite those publications that have been relevant to the validation and development of our aerosol-optics model. The present manuscript is, indeed, only the tip-of-the-iceberg of a long-term research project on black carbon, which involved a substantial amount of development work. The citations of our own work in the manuscript reflect this fact. For instance, we have performed exactly the kind of validation studies that the reviewer mentions under specific comments 1 (dependency of MAC and SSA on aggregate size, monomer size and number), and more (dependency of MAC and SSA on refractive index, which is even more important for MAC than the monomer size). However, we agree, of course, that it may be helpful for the reader if we include a more complete reference list, so we added additional citations (see specific comments 1 below).

2. Many of the plots are not easy to read, neither in the screen or in a printed version. I would recommend to make labels of the axis with a larger font size. I would also suggest to add a description of the plots in the caption, so a reader does not need to go back a forward in the text to find relevant details. The manuscript does not provide any code to perform calculations, which seems strongly suggested by the journal.

   We agree that the quality of the plots was in need of improvements. The plots have been revised along the reviewer's suggestions. We also added a section on

code-availability, see page 34 and section 5.

2 Specific comments

1. Page 10745 Please, state T-matrix estimated mass absorption coefficients (MAC) values and how close are to Bond and Bergstrom (2006) recommended values, in addition to SSA values.

   The choose of monomers radius of 25nm might be legitimate, nerveless I would recommend a statement addressing the variability in the monomers radius 15-25 nm. Different monomer radii exist due to differences in sources, which mainly depend on burning materials (i.e. Weztner et al., 2003, China et al., 2014., Chakrabarty et al., 2014)

   Optical properties of BC aggregates may vary depending on assumptions of BC monomers radius. One key manuscript addressing the issue at one wavelength is Liu et al., 2008, other papers based on observational driven constrains of BC particle aggregates may be worth to be cited, i.e., Scarnato et al., 2013, 2015. The choose of a Df=1.8 can be considered consistent with semi aged BC aggregates, i.e. China et al., 2015, China et al., 2014.

   A good fitting of observed values of MAC and SSA with T-matrix simulations reside on the choice of aggregate physical constrains (aggregate size, monomer size and number, other than fractal dimension).
   The MAC values of our aggregate model have been validated in detail in Kahnert and Devasthale (2011), which we have cited.

   We added, as suggested, that the monomer radius can vary between 10–25 nm with a citation of Bond and Bergstrom (2006) (see page 16 line number 12-13). However, we emphasise that our choice of a=25 nm has been validated in the AST-paper by Kahnert (2010), as explained in the text; it gives the best

agreement with measurements for the SSA. This is actually not surprising. It is much more surprising that in the literature authors often seem to tacitly assume that the best choice of the monomer radius a would be some mean value, $\int n(a)ada$, over a normalised monomer-size distribution $n(a)$. We cannot think of any good arguments to support this conjecture. Rather, the monomers are tiny compared to the wavelength, so if anything they should behave similarly to Rayleigh scatterers, for which the absorption cross section scales as a^3. Thus, in our own humble opinion a meaningful first-guess for an effective radius would be $a_{\text{eff}} = \int n(a)a^4da / \int n(a)a^3da$, the ratio of the fourth to the third moment of the size-distribution. I.e., one should weigh $n(a) \cdot a$ with the absorption cross section, which is proportional to a^3. If doing so, we would expect that the effective monomer radius that gives the most realistic estimates of the optical properties should lie close to the upper end of the size scale 10–25 nm, which is just what we assume.

We also added some of the citations suggested by the reviewer (China et al. (2014) at p. 10 l. 294, Chakrabarty et al. (2014) at p. 10 l. 293, Scarnato et al. (2013) and Scarnato et al. (2015) at p. 1 l. 320). However, we would like to point out that the manuscript by Liu et al. (2008), although highly cited, covered an enormous parameter space that far exceeds the range relevant for atmospheric black carbon (e.g. Df as low as 1.25 and as high as 3; as well as quite unrealistic refractive indices, 2+i and 1.75+0.5i). The AST-paper by Kahnert (2010) considered a considerably more constrained and realistic range of parameters and was much more relevant for the validation of our aerosol-optics model.

We agree that Df=1.8 is typical for rather fresh or "semi-aged" aggregates. Older aggregates may have higher fractal dimensions. However, exactly this question has been discussed in the ACP-paper by Kahnert and Devasthale (2011); this study showed that, despite being rather low, the value of Df=1.8 gives a better agreement of modelled and measured MAC values than Df-values that may be

more representative for aged black carbon.

2. The paper mainly focuses on the impact of various treatments of optical modules to chemical transport models. Secondly, the paper discusses how various optical treatments can impact estimates of backscattering coefficients and Angstrom exponent. The authors should consider, at least for a site, to make comparison with observational data (i.e., A comparison between space born lidar backscattering values with those predicted).

   In principle we very much agree with the reviewer that comparison with observations are extremely valuable. However, doing comparison with observations is an all-or-nothing business. It would not be very illuminating to simply show a plot of model results and observations. Chemical transport models are notoriously plagued by a large number of errors and biases, such as the emission estimates, deposition velocities, chemical reaction rates, meteorological input data (wind fields, precipitation rates, boundary-layer processes, etc), land use data, parameterisations of physical procesess, such as nucleation, condensation, coagulation, and water-uptake, to name just a few examples. All of these sources of error would effect a comparison of modelled optical properties with measurements. A meaningful comparison would not be possible without performing a comprehensive analysis of *all* involved sources of error. This would completely change the focus of the paper. Our intention with this paper is to educate the chemical-transport modelling community, and raise some awareness for the importance of aerosol optics modelling. This message would have become completely buried if we had set up this study as a CTM validation study.

3. Page 10756: Can you please provide an explanation for the statement: "Over the Mediterranean (Fig.6), the EXT and CGS model have almost identical AOD profiles in the green part of the spectrum. However, at longer wavelengths (not shown) EXT predicts substantially higher AOD values than CGS"

   This statement refers to a difference in the AOD at different wavelengths. In the paper, we only show AOD for the green spectrum, i.e. 500(EXT)/532(CGS) nm, but in order to explain certain behaviours, we had to look closer at some of the other wavelengths as well. In the revised edition, we have now added an appendix (Appendix E) where we include figures with optical properties for the other wavelengths. We also added in the text a reference to the appropriate figure that shows ADO at longer wavelengths; this should help to make this statement clearer.

4. Can you please provide an order of magnitude for the statement? "TOA net flux in EXT as compared to the CGS model. Note that the differences in SSA between EXT and CGS are fairly small, while the differences in g are rather large".

   The reviewer has a good point; this statement needs to be more precise and quantitative. We have changed the text accordingly and provided concrete numbers for the differences in the asymmetry parameter and SSA between the two models. See page 24 line number 703-706.

5. It might be useful to strength the perception of the paper, to add when possible in the abstract and conclusion a percentage (or order of magnitude) of the impact of different optical modules.

   An excellent point. We have taken some of the most important results from the new table 4 (which shows spatio-temporally averaged model differences) and cited them in the revised abstract and conclusions. See the abstract on page 2 line number 24-27 and the Conclusion at page 33 line number 823-827.

   Please also note the supplement to this comment:
   http://www.geosci-model-dev-discuss.net/8/C4324/2016/gmdd-8-C4324-2016-supplement.pdf

Interactive comment on Geosci. Model Dev. Discuss., 8, 10735, 2015.