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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Numerically exact computation of the optical properties of light absorbing carbon aggregates for wavelength of 200 nm – 12.2 µm" *by* M. Kahnert

Dr Kahnert

michael.kahnert@smhi.se

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I thank the reviewers for their encouraging evaluation and for their helpful input. The comments by M. Kocifaj refer to the first version of the manuscript before it was published on the web. Below I list a description of the changes I had implemented prior to publication as a discussion paper. Also, I list an important change I intend to make in the final version of the paper.

1. *Page 1–2: This is an interesting study on radiative effects by irregularly shaped LAC particles. It has been clearly shown that ignoring the realistic morphologies can lead to errors in estimated radiative forcing. The author does a good job of*

explaining how the broadband optics of strongly absorbing LAC and HSA can differ. The numerical simulations seem to be logical and straightforward.
My comments are below:
Page 3: The author calculated AOPs for 5 different geometries. Is this statistically acceptable? Were the differences between broadband optical properties of LAC and HSA also evaluated for each of 5 individual geometries? If so, do the results behave similar to those for averaged case?
A sample of 5 different geometries would certainly not allow us to compute statistically reliable averages. But I think that considering larger particle ensembles would not be feasible, since numerically exact electromagnetic scattering computations for fractal aggregates are extremely time consuming. Performing broadband computations for an ensemble of geometries that is large enough for statistical averaging is completely beyond the reach of our current computational abilities. Fortunately, this is not a serious limitation. The idea of the scaling relation given in Eq. (1) is to define a class of geometries that have well constrained optical properties. It is truly remarkable and very fortunate that this really works, as can be seen in Fig. 2. Only the backscattering cross section C_{bak} displays a certain variation among different geometries with the same fractal parameters. It is mainly for the sake of C_{bak} that I introduced the approach of identifying "typical" geometries. (Note, however, that the backscattering cross section does not enter into the scalar radiative transfer equation, so it is not needed in radiative forcing computation. It is a quantity that enters in computing lidar backscattering coefficients, so it is mainly interesting for active remote sensing, not for climate modelling.)

To answer the reviewer's second question: No, broadband optics of LAC and HSA were not evaluated for each of the 5 individual geometries separately, because broadband optical properties were not computed for each of the 5 geometries. Broadband optical properties were only computed for one "typical" geometry for each size and wavelength. This took several weeks of wall-clock time on a paral-

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lel machine. Repeating these computations for 5 different geometries would have required several months of wall-clock time. This enormous computational effort and budget costs would have been in no relation to the benefits. As can be seen in Fig. 2 the "typical" geometries yield a smooth dependency of C_{bak} on size. Although the selection of these "typical" geometries is based on computed averages that are, as the reviewer correctly points out, based on a statistically fairly small sample, I do believe that this approach is the best possible compromise I could achieve with an acceptable cost-benefit ratio. It is certainly an improvement as compared to most previous studies that were based on one randomly selected geometry per size only.

I very much agree with the reviewer that these points were not made sufficiently clear in the pre-discussion manuscript. In the discussion paper, I therefore rewrote the entire paragraph as follows:
"We generate our model geometries by use of a random cluster generation algorithm, which was developed by Mackowski (1994). The idea of the scaling relation given in Eq. (1) is to define a class of geometries that have well constrained optical properties. The total absorption and scattering cross sections display indeed very little variation among different geometries that obey the same scaling relation. However, computational results reported by Kahnert (2010a, 2010b) indicate that differential scattering properties, such as the backscattering cross section C_{bak}, may not be sufficiently well constrained by specifying $\{a,\, N_{\mathrm{s}},\, D_{\mathrm{f}},\, k_0,\, R_{\mathrm{g}}\}$. This can give rise to numerical artifacts in the form of fluctuations of C_{bak} as a function of aggregate size. Although C_{bak} does not enter into the computation of the radiative forcing of aerosols, we shall test a pragmatic and computationally inexpensive approach for alleviating the fluctuation problem associated with C_{bak}. In principle one could repeat computations for an ensemble of different geometries and compute ensemble-averaged AOPs. However, for the broadband calculations we intent to perform, such an approach would be prohibitively time consuming. We therefore try to identify, for each size, a "typical"

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geometry that gives a value of C_{bak} close to the mean. To this end, we select a single wavelength of λ=533.2 nm, and compute the AOPs as a function of size for five different geometries at each discrete size. Although a sample of only five geometries is not expected to give a highly accurate estimate of the mean value of C_{bak}, this approach proves to be sufficiently robust for reducing the fluctuations of C_{bak} as a function of size, while keeping the computational costs within reasonable limits. Figure 2 shows computational results represented by circles for the absorption cross section C_{abs} (upper left), the scattering cross section C_{sca} (upper right), the asymmetry parameter scaled by the scattering cross section $g \times C_{\mathrm{sca}}$ (lower left), and the backscattering cross section C_{bak} (lower right). The AOPs are presented as functions of the volume-equivalent radius R_{V}. The quantities that are important for radiative forcing computations are C_{abs}, C_{sca}, and $g \times C_{\mathrm{sca}}$. The five different geometries yield almost identical results for these optical properties. By contrast, C_{bak} displays some variation (note the logarithmic scale!). We compute at each particle size R_{V} an average $\langle C_{\mathrm{bak}} \rangle$. Next we select for each particle size R_{V} that geometry that yields a value of C_{bak} closest to $\langle C_{\mathrm{bak}} \rangle$. The computed AOPs obtained for these "typical" geometries are indicated by the green pluses in Fig. 2. Comparison with the red fitting curves shows that these geometries indeed yield a smooth size dependence for all AOPs, thus significantly reducing the fluctuations of C_{bak} as a function of R_{V}. These "typical" geometries are the ones we employ for the broadband calculations. So in the broadband computations, we only employ one geometry for each size R_{V}. This approach reduces numerical artifacts sufficiently well without increasing the required computation time."

2. *Page 7: It is not clear to me how the location-specific microphysical parameters of aerosol populations were taken into account. It seems that fixed Rv0 and sigma were considered in Eq. (4).*
   Yes, the parameters characterising the lognormal distribution were fixed. Again,

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the explanations given in the pre-discussion manuscript may have been somewhat unclear. The point of Eq. (4) and Fig. 4 was not to discuss the asymmetry parameter and phase function in the context of a geographic variation, but to understand the single scattering properties of aggregates as compared to homogeneous spheres. The asymmetry parameter is the first Legendre moment of the phase function. Radiative transfer modellers are well familiar with this quantity and understand how it describes the partitioning between forward and backward scattered radiation. However, others may find it intuitively more appealing to inspect the corresponding phase function, which is the normalised differential scattering cross section. I therefore try to explain here the differences in asymmetry parameters by inspecting the corresponding phase functions. This involves a technical problem. Whereas the asymmetry parameter is a function of size and wavelength and can be represented in a contour plot, the phase function is a function of scattering angle, size, and wavelength. Since I cannot show a four-dimensional plot, I need to get rid of one of the three independent variables. I chose to do that by integrating over a fixed size distribution. Certainly, the size distribution of LAC emissions depends on the source and thus varies with location. For instance, a shift of the size distribution to larger sizes will narrow the diffraction peak. Since this is well known, I did not feel compelled to discuss such issues here. Rather, I make a *relative* comparison of the phase function of aggregates and homogeneous spheres *both* averaged over the *same* size distribution. Considering different size distributions would effect the phase function of aggregates and spheres in the same way (e.g. narrowing of the diffraction peak), so this would not change any of the conclusions about the differential scattering behaviour of aggregates *in comparison to* spheres.

To preclude misinterpretations, I included the following explanations in the discussion paper:
“The asymmetry parameter is the first Legendre moment of the phase function. This quantity may be less intuitively appealing than the phase function, which rep-

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resents the normalised angular distribution of the scattered radiation. Thus the misrepresentation of the asymmetry parameter by the HSA may become clearer if we compare the phase functions computed with the two different models. The phase function depends on λ, R_V, and on the scattering angle Θ. To visualise this four-dimensional functional relation in a contour plot, we need to eliminate one of the independent variables. We choose to eliminate the dependence on R_V by integrating over a fixed log-normal size distribution

$$n(R_V) = \frac{1}{R_V \sigma \sqrt{2\pi}} \exp\left[-\frac{\ln^2(R_V/R_V^0)}{2\sigma^2}\right], \quad (1)$$

where we assume R_V^0=52 nm and σ=0.42, which are obtained by fitting measurements of diesel soot emissions (Färnlund et al., 2001)."

3. *Page 9: Although the author declared that the real-time calculations are possible, it is not stated what is the CPU needed to obtain the overall picture such as in Fig. 9.*
This is a good point that I tried to make clearer in the discussion paper. We must not mix up the computation time required for performing the AOP computations, and that needed for running chemical transport or climate computations. The AOP computations and the size-averaging for each size mode are actually performed off-line, as is stated in Sect. 2.2 and in Sect. 3.2.

The picture in Fig. 9 was obtained by reading in a pre-computed look-up table of AOPs tailored to the size distribution model employed in MATCH, which is based on eight different size classes (four internally and four externally mixed ones). The MATCH model needs to average the AOPs from the look-up table over the eight size classes in each grid cell, where the averaging is done by weighing the AOPs with the number densities in each size class computed in MATCH. For the internally mixed size classes, one also needs to compute (on-line) an effective refractive index based on the chemical composition of the mode in each grid cell

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and time step. The computation time required for these operations is negligible compared to the time required for computing transport, chemical transformation, deposition, and (depending on the model version) aerosol dynamic processes. So the CTM does not really "care" whether the AOPs have been pre-computed by use of the homogeneous sphere approximation or by use of morphologically more sophisticated models.

On the other hand, the time to generate the AOP look-up table can be substantial, and although these computations are performed off-line, there do exist limitations to how sophisticated aerosol optical models we can afford to employ. To avoid confusion of this issue with the CPU time requirements for running a CTM, and to address the reviewer's question, I had added in the discussion paper (first paragraph of Sect. 2.2) the following sentence:
"Since the AOP look-up table is computed off-line, the CPU time requirements for computing size- and composition-averaged AOP within MATCH based on the MATCH aerosol field and based on the look-up results is negligible compared to the computational efforts involved in computing chemical transformation, transport, and deposition."
Further, I had changed the beginning of the last paragraph of the conclusion section as follows:
"On-line computations of ensemble-averaged AOPs in a CTM or an Earth-system model based on an off-line computed look-up table requires only little computation time in comparison to that needed for computing chemical and meteorological processes. However, the computational effort to create an AOP look-up table in the first place can be quite substantial, since electromagnetic scattering computations for such complex particles as fractal aggregates tend to be very time consuming."

There is one additional important change I intend to make in the final version of the paper. I want to add an extra subsection to section 2 about comparison of modelling

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results with measurements. In the reference given on p. 11902, lines 7–9, a study has been presented that was dedicated to comparing modelled optical properties of LAC aggregates with measurements. Further, the sensitivity of computed optical properties to the physical parameters of the LAC aggregates had been investigated. These are important results, which form a basis for the present study. However, the discussion of these results is still somewhat obscure and unclear, since it is spread out over several places in the present version of the paper. I therefore plan to add a dedicated subsection in which these sensitivity studies and comparisons of modelling results and measurements are described and discussed in detail.

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