**Atmospheric
Chemistry
and Physics
Discussions**

# *Interactive comment on* "Modelling the optical and radiative properties of freshly emitted light absorbing carbon within an atmospheric chemical transport model" *by* M. Kahnert

**Dr Kahnert**

michael.kahnert@smhi.se

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**Response to review comments by M. Kocifaj:**

I thank the reviewer for his helpful input. The comments 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, as well as a few additional comments.

1. *The author evaluates the radiative forcing using the Eq. (7). Is the diffuse radia-*

*tion explicitly incorporated into $F_\lambda$?*

Yes, it is. I added the definition of the radiative net flux to the text preceding Eq. (9) in the discussion paper [which was Eq. (7) in the first manuscript version]:

"...we compute the spectral radiative net flux $F_\lambda = F_\lambda^s + F_\lambda^- - F_\lambda^+$. Here $F_\lambda^s$ denotes the direct solar flux, i.e. that part of the incoming flux that survives extinction, $F_\lambda^-$ represents the diffuse downwelling flux, and $F_\lambda^+$ is the upwelling flux."

2. *The simpler version of the code by Xu & Gustafson enables to calculate the optical properties of a particle in a fixed orientation. If so, the formulation at page 7, line 8, column 2 needs to be corrected.*

I replaced this sentence with

"A simpler version of the same code, which computes the optical properties for discrete orientational angles and performs the angular integration numerically, proved to be sufficiently fast and stable for our task."

3. *As for the representativeness of the results: how these can change with particle's morphology? Is the chosen morphology typical for LAC?*

The fractal dimension and prefactor are indeed typical for LAC. They conform both with measurements and with diffusion-limited cluster aggregation simulations (see CM Sorensen and GC Roberts, "The prefactor of fractal aggregates", Journal of Colloid and Interface Science 186, 447-452 (1997)). The monomer radius $a$ may vary between 10-25 nm. However, as pointed out by Bond and Bergstrom (2006), MAC is insensitive to a variation in $a$ for all values of $a < 40$ nm (i.e. diameter smaller than 80 nm).

Physical properties of LAC aggregates are briefly discussed just below Eq. (2). In this context two additional aspect are interesting to mention:

(a) Liu and Mishchenko (2008) Investigated the optical properties of soot aggregates as a function of fractal dimension. Inspection of Fig. 2 in their

paper shows that, e.g., the absorption cross section at $m$=1.75+0.5i, $a$=25 nm varies for an aggregate of fixed size by less than $\pm$2% as we allow the fractal dimension $D_f$=1.8 to vary by $\pm$0.2, which is quite a generous variation for freshly emitted soot. Note that fractal dimensions $D_f >$2 are typical for aged soot aggregates.

(b) Equation (1) defines only a class of fractal geometries. There exist infinitely many fractal geometries to each pentupel $(a, N_s, R_g, k_0, D_f)$. However, the optical cross sections are well-known to be rather insensitive to such geometric variations. I did some tests to verify this, but I had not mentioned them in the original manuscript. In response to the point raised by the reviewer, I added the following text on p. 25455:

"Incidentally, we verified that the optical cross sections are sufficiently constrained by specifying the values of $a$, $N_s$, $R_g$, $D_f$, and $k_0$. For each of the aggregates up to $N_s$=100 monomers, we repeated computations for five different geometries having the same values of these parameters. The results for the optical cross sections varied by less than 1 %."

4. *The range of size parameter in Figs. 6-7 doesn't fit with size classes [0.02,0.1)* $\mu$m, [0.1,1.0) $\mu$m, [1.0,2.5) $\mu$m and [2.5,10.0) $\mu$m. Note that x=0.15 corresponds to the particle diameter about 0.02 $\mu$m, so the particles with x<0.15 become smaller than a monomer.*

There may be a slight misunderstanding here with regard to Figs. 6-7. The x-axis of these plots shows the volume-equivalent *radius* $R_V$ (in micrometers), not the *size parameter x* (unitless). I added "...as a function of volume-equivalent radius $R_V$..." to the figure caption to preclude any misinterpretations.

Concerning the size classes and the range of Rv: The reviewer is right in that the range of relevant sizes of LAC aggregates does not coincide with the size class boundaries in the MATCH model. Fortunately, this does not pose any problems,

not even technical ones. The MATCH model tells us the *total* amount of LAC in each of the lowest two size classes. When integrating the optical properties of externally mixed LAC, I simply assume that the LAC in the second size class does not extend all the way up to $R_V$=500 nm, but only up to $R_V$=170 nm. So I simply limit the range of the size-integral. To make this more explicit, I added on p. 25453, line 15: "In the calculation of size-averaged optical properties, LAC is assumed to extend into the second size class only up to a size of $R_V$=170 nm."

5. *The cubical fit (Eq. 3) is well-founded for cross sections of strongly absorbing particles (refers e.g. to well-known book of Bohren & Huffman). Is there some reason for using such a fit for asymmetry parameter too?*

This is an important point that I tried to make more clear in the discussion paper.

I agree with the reviewer that the *absorption* cross section of small particles is proportional to its volume/mass, i.e. to $R_V^3$ (see, e.g., Eq. (7.4) in the book by Mishchenko, Travis, and Lacis (2002), or Section 6.31 in van de Hulst's book (1957)). The *scattering* cross section, on the other hand, is proportional to $R_V^6$ (Eq. (7.3) in Mishchenko et al., or Section 6.31 in van de Hulst). However, making the ansatz $C_{\text{sca}} = c_6 R_V^6$ fails for the aggregate results (I checked it!). The reason is simply that the largest aggregates considered in this study are no longer within the Rayleigh regime. Thus, except for the absorption cross section, there is no deeper physical reason for the third-order polynomial fitting approach. The Taylor approximation is a purely mathematical tool which only serves one purpose, which is outlined on p. 25452, last line in Section 2:

"Thus the strategy is to compute AOP for a selected number of sizes, followed by a suitable interpolation. Size-integration of AOP can then be achieved by integrating the interpolation functions weighted by the size distribution."

To make the idea of this approach more explicit, I added the following text to Sec. 4.1.

"Note that for small homogeneous spheres the optical cross sections behave asymptotically as

$$C_{abs} \xrightarrow[\substack{R_V << \lambda \\ |m|R_V << \lambda}]{} \frac{3\pi^2}{\lambda} \mathrm{Im}\left(\frac{m^2-1}{m^2+2}\right) R_V^3, \qquad (7)$$

$$C_{sca} \xrightarrow[\substack{R_V << \lambda \\ |m|R_V << \lambda}]{} \frac{128\pi^5}{3\lambda^4} \left|\frac{m^2-1}{m^2+2}\right|^2 R_V^6 \qquad (8)$$

(van de Hulst, 1957). For $C_{abs}$, this yields an asymptotic value of $c_3^{abs}$=47.22 $\mu m^{-1}$ for small spheres, which is somewhat smaller than the result obtained for the aggregate model. However, the absorption cross section of homogeneous spheres in Fig. 6 cannot accurately be fitted over the entire size range by the simple ansatz $C_{abs} = c_3^{abs} R_V^3$. The absorption cross sections of the larger homogeneous spheres are consierably lower than those predicted by Eq. (7). The physical reason for this is that the electromagnetic field is unable to penetrate into the centre of larger, highly absorbing spheres. This means that only the mass near the surface of the larger particles contributes to absorption. Hence, the increase of $C_{abs}$ with $R_V$ is weaker than the third power. By contrast, the geometry of fluffy aggregates is such that most of the mass interacts with the electromagnetic field, which is why the relation $C_{abs} \propto R_V^3$ holds over the entire size range considered here.

The asymptotic form for the scattering cross section given in Eq. (8) is known as the Rayleigh approximation, which is also referred to as the dipole approximation. It is based on the assumption that all electrons in the particle oscillate in phase in response to the external electromagnetic forcing, just like a dipole. This only holds for particles considerably smaller than the wavelength. Recall that the radius of gyration $R_g$ of fractal aggregates with $R_V$=170 nm is around

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550 nm, which is comparable to the wavelength. Thus it cannot be assumed that all monomers in the aggregate oscillate in phase, as they would in Rayleigh scattering. For this reason, Eq. (8) is not valid over the size range considered here. Thus, it is clear that the polynomial fitting ansatz made here is physically motivated only for $C_{\mathrm{abs}}$. For the other parameters, it is a purely mathematical approximation. The merit of this fitting approach is that the coupling of AOP and CTM computations is greatly facilitated. For instance, the output of an aerosol dynamic model with a log-normal or sectional size distribution can be easily employed in conjunction with Eq. (3) for computing size-averaged AOP, as the moments of such size distributions can be computed analytically."

Discussing this point is greatly facilitated by focusing on $C_{\mathrm{abs}}$ rather than $C_{\mathrm{ext}}$. For this reason, I replaced $C_{\mathrm{ext}}$ in Figs. 6 and 7 by $C_{\mathrm{abs}}$.

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