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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 anonymous reviewer 2:

I thank the reviewer for his helpful comments. Below follows my response, along with a description of the changes I intend to make to the revised manuscript.

1. *The author cuts the size range of aggregate up to Ns=350. It looks correct to cut the size range Rv around 0.17 as long as extinction efficiency in Figure 2 is referred. However, the extinction efficiency is calculated based on "spherical*

particle model" not aggregate. Therefore judging the size range from the Figure 2 is incorrect.

The discussion of Figure 2 was merely meant to yield a first rough estimate of the size range of interest. However, I do agree with the reviewer that it will be interesting — and safer — to extent the range of N_s. Therefore, I performed additional calculations up to N_s=1000, which I will include in the revised version of the manuscript.

2. *As shown in the paper by Liu and Mishchenko JGR doi:10.1029/2004JD005649 Figure 6, when we consider aggregates, the extinction efficiency does not largely decrease (like sphere) for increased number of composing particles (Ns). They are rather expected to have flat Cext for further increase of the Ns (see Figure 6 of them).*

If I understand this comment correctly, then it largely agrees with the point I am trying to make. Figure 6 in the paper cited by the referee shows C_{abs}/m, where m denotes the mass. In the terminology of the discussion paper, this is equivalent to C_{abs}/R_V^3. Thus, the observation that C_{abs}/m becomes independent of N_s is equivalent to stating that C_{abs} scales as R_V^3. The calculations in the paper by Liu and Mishchenko extend up to $N_s = 400$. The extended calculations I did for the revised manuscript confirm that the scaling $C_{\mathsf{abs}} \propto R_V^3$ and $C_{\mathsf{sca}} \propto R_V^3$ hold over the entire range up to N_s=1000. I will add a reference to the paper by Liu and Mishchenko in the revised manuscript.

3. *Moreover, researchers working on soot have shown that there are several thousands (not hundreds) of composing particles seen in the soot from the electron tomography (e.g., L.H. vanPoppel et al. GRL doi:10.1029/2005GL024461, 2005). From the Cext of aggregates shown in Liu and Mishchenko (2004) and study by van- Poppel et al. 2005, it is important to consider aggregates composed of thousands (not only up to Ns=350). If this kind of extended calculation is com-*

putationally difficult, the author must denote that the paper based on the limited calculation and extended calculation to check the influence of increased number of composing particles required as future works.

Again, the range of aggregate sizes has now been extended up to N_s=1000. I have now fitted the optical properties over the range up to N_s=1000. The fitting constants for C_{abs} and C_{sca} have changed by less than 1 % as compared to the fitting exercise limited to the range up to N_s=350. Thus the main benefit of extending the range of N_s up to 1000 monomers is to confirm that the scaling relations $C_{\mathsf{abs}} \propto R_V^3$ and $C_{\mathsf{sca}} \propto R_V^3$ hold over a large size range.

At the same time, one must not forget that the main purpose of the present manuscript is to make a coupling between aerosol optical modelling and chemical transport/aerosol dynamic computations. Here one must be very careful with the size range one assumes. In a *modal* aerosol dynamic model, I see no problem with including such large aggregates. However, in *sectional* models, which become more and more popular, one would grossly overestimate the optical depth if one did not cut off the size distribution at a reasonable maximum size. Thus, since we use a sectional representation of the size distribution in the MATCH model, I did stick to the cut-off size of R_V=170 nm in the coupled AOP/CTM calculations shown in Figs. 9 and 10.

4. *Another thing to care is that the results of aggregates in Figure 9, 10 and 11 are obtained only for a single set of parameters. These limited-parameter-range results cannot be considered as "representative" of the general LAC properties. They may reflect part of the general property "by chance" but we cannot guarantee it by seeing limited range of results. Future systematic study to reduce biases of the AOP and radiative forcing are required after this research paper.*

   I completely agree that this study will, by far, not be the last word on LAC aggregates. Note that the point raised by the referee is very clearly stated in the manuscript (p. 25464, lines 13-28).

I do think that changing a, D_f, and k_0 within *reasonable* ranges will not remove the discrepancy between modelled and measured MAC (see my response to the other two reviewers). More studies of the sensitivity to a variation in the refractive index m would be very helpful. It is also possible that the computed MAC results are too low, because the assumed mass density ρ has been to large. I believe that the experimentalists will have the final word. More detailed measurements of m, ρ, and MAC will be needed.

Apart from assumptions about the physcial properties of aggregates, I do think that the main limitation of Figs. 9–11 is that they have been obtained for one instant in time, at one particular location, and at only two wavelengths. General conclusions about the significance of LAC morphology to climate forcing could only be drawn from broadband studies within a global model, integrated over something on the order of a decadal time scale. This is clearly beyond the level of ambition of the present paper.