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Interactive comment on “Simplifying the calculation of light scattering properties for black carbon fractal aggregates” by A. J. A. Smith and R. G. Grainger

J. C. Corbin

joel.corbin@env.ethz.ch

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Smith and Grainger present a concise and clear paper arguing that scattering by black carbon fractal aggregates (BCFAs) cannot be accurately represented by an “equivalent” sphere. In our opinion, the paper has performed meaningful calculations using atmospherically representative input parameters. We would be interested in results or discussion addressing these additional issues relevant to atmospheric BCFAs:

1. A single value was used for the spherule radius a (25 nm). For atmospheric BCFAs, the value of a may vary considerably between different combustion sources.

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- The current value of 25 nm would be representative of wood-combustion aerosol (Gwaze et al. 2006; Zelenay et al., 2011). Since diesel soot typically has smaller spherules – about 7.5–20 nm in radius, with more-modern engines having smaller a (Burtcher, 2005) – it would be interesting to see results or discussion on the impact of smaller a .
2. What was the motivation for choosing D_f as low as 1.6? Although this low value would reflect a DLCA-formed aggregate (Diffusion-Limited Cluster-Cluster Aggregation) of highly-polydisperse spherules (Eggersdorfer et al., 2012), to our knowledge natural soot aerosols have always been observed to consist of near-monodisperse spherules so that $D_f = 1.8$ (Sorensen, 2011). This would be significant to Figs. 7 and 8, since the errors there are largest for the lowest D_f . The authors will probably agree that $D_f = 1.8$ is most relevant, as they have already focussed on this case in general.
 3. As the authors have noted, the fractal dimension of soot particles may increase to 2.3 (Bambha et al., 2013) or higher (Zhang et al., 2008) following coating. The value of 2.3 is already outside the range of the D_f studied, presumably because the clustering algorithm does not represent subsequent restructuring. But since the coating-induced increase in D_f was prescribed during cluster formation and not afterwards, even the $D_f = 2.2$ aggregates may not have the same structure as restructured DLCA soot with $D_f = 2.2$. In particular, the anisotropy (Heinson et al. 2010; Eggersdorfer and Pratsinis, 2013) and fine structure (Mitchell et al., 2003) of restructured aggregates are not constrained by D_f alone. So the higher D_f values of restructured BCFAs may not be precisely captured by tuning the clustering algorithm to produce higher D_f values. This theoretical expectation is confirmed by a comparison of microscopy images (Bambha et al., 2013; Zhang et al., 2008) with the authors' modelled BCFAs (shown in their Fig. 1). The modelled BCFAs appear to have a different, less-compact structure than the real restructured BCFAs. This would imply that the real restructured BCFAs may have

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smaller deviations from Mie theory than calculated in the manuscript. Would it be possible for the authors to somehow synthesize aggregates that are more spherical to provide an upper bound? We realize that this may be quite challenging without a quantitative restructuring model, but if the clustering algorithm is not representing the restructuring process, a manually-synthesized BCFA would not necessarily be less physical than the current high- D_f BCFA.

One final comment. Since the asymmetry parameter g was quite small for the smallest clusters (Fig. 5c, bottom row), perhaps it would be better in Fig. 7c to plot the absolute error in g instead of the relative error. The actual deviation here appears to be relatively small.

We agree with Smith and Grainger's conclusion that light-scattering by BCFA's cannot be accurately modelled as equivalent spheres in general, but we wonder whether this conclusion could be reversed under certain conditions: could the errors in an equivalent-sphere treatment in an atmospherically-relevant parameter space ($D_f > 1.8$ and $n_s < 500$) be small enough to be acceptable in at least some modelling applications?

- Joel C. Corbin and D. Neubauer

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