Atmos. Chem. Phys. Discuss., 12, C10046–C10052, 2012
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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Effects of internal mixing and aggregate morphology on optical properties of black carbon using a discrete dipole approximation model" *by* B. Scarnato et al.

**Anonymous Referee #2**

Received and published: 3 December 2012

## 1 General

This paper presents DDA computations for black carbon aggregates internally mixed with NaCl. The morphological parameters, such as the fractal dimension, the monomer radius, and the mixing state are varied within realistic ranges, and the effects of particle morphology on optical parameters, such as AAE, SSA, MAC, the phase function, and the degree of linear polarisation are investigated. The study is timely and the methodology is state-of-the art. There are a few issues I would ask the authors to address before the paper can be accepted for publication in ACP.

## 2 Major comment

The optical properties are averaged over 64 target orientations only (see p. 26411). This is an extremely small number; I find it hard to believe that the authors have really attained convergence in the orientation averaging with such a small set of angles. This may work in some cases, such as for MAC, SSA, AAE, or for large wavelengths (small size parameters). But I would not expect this to be sufficient for computing differential scattering properties, such as the phase function or the degree of linear polarisation. For instance, Kahnert et al. (2012, see reference on p. 26420) performed various tests involving up to more than 20000 orientations. They found that one needs at the very least over 800 orientations to obtain converged orientation-averaged optical properties. The number of orientations used here is more than a factor of 10 smaller than that. This may limit the accuracy and thus the credibility of the results presented here.

I would ask the authors to comment on this point. Did you actually perform tests to check the convergence of the orientation-averaged results with respect to the number of discrete orientational angles? If yes, for what cases? If not, the proper way to correct this would be to perform such tests and, if necessary, to re-do the computations with an adequate number of orientation angles. The easier (but less satisfactory) solution would be to clearly comment in the text that the reliability of the computational results may be impaired by the fact that the number of orientational angles has not been validated and may be too low. It may also help if you state precisely how many discrete angles you used for each of the three Euler angles. Perhaps I misunderstood your statement, and the figure of 64 only refers to the first two Euler angles (which take most of the computation time), while you used a large number of discrete angles for the third rotation angle?



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## 3 Minor comments

- p. 26407, line 24: "[...] observed in the atmosphere ([...]; Bond and Bergstrom, 2006)." Are the observations discussed by Bond and Bergstrom really atmospheric field observations? My memory may let me down, but as far as I remember, most of the observations discussed in that paper were laboratory measurements.
- p. 26410, line 10-11: "geometrical cross section" should be "optical cross section".
- p. 26411, line 13: add "1=" in front of the normalisation integral.
- Same line: replace "$\theta \in [0, \theta]$" by "$\theta \in [0, \pi]$".
- Same page, lines 16-17: The authors assume a refractive index that does not change with wavelength, and they justify their choice with a reference to Moffet and Prather (2009). However, there are other authors, such as Chang and Charalampopoulos (Proc. R. Soc. Lond. A 430, 577-591, 1990), who observed a fairly strong spectral variation of both the real and imaginary part of the refractive index of BC, especially in the UV, but also in other spectral ranges. The refractive index value they observed at 550 nm is among the most realistic ones according to the discussion in Bond and Bergstrom (2006). I would like to ask the authors to discuss their choice in view of that study, and to state more clearly if their choice is mostly motivated by trying to keep things simple, or if they really believe that their assumption of a constant refractive index is most realistic (and if so, why?).
- p. 26415: There are several statements regarding the discussion of the phase function in Sect. 4.3 that should be made more precise:
  - lines 6-7: "Lacy BC (case F) exhibits a stronger forward scattering intensity than compact BC (case G)". This statement is a bit fuzzy, as the authors



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mix the discussion of the phase function with a discussion of the intensity. As the authors state themselves on p. 26411 (line 13), the phase function is a *normalised* quantity. Thus, it only tells us how much intensity is scattered in one direction *relative* to other directions. If we want to know the scattered intensity in absolute terms in any particular direction, such as the forward direction, then we need to consider the phase function multiplied with the total scattering cross section. So, to decide whether lacy BC gives more forward scattering than compact BC the authors need to consider not just their respective phase functions, but also their respective total scattering cross sections. The easiest way to improve the text would be to reformulate the sentence, e.g. "The phase function of lacy BC (case F) has a narrower and stronger forward scattering peak than that of compact BC (case G)"

This brings me back to my main comment (insufficient number of orientational angles), which raises the question how reliable these results for differential scattering properties really are. By showing S11 and S12 the authors may present more information than they can reliably compute with their methodology. In view of this, it may be better to simply discuss the asymmetry parameter, which is the first Legendre moment of the phase function. This quantity provides a measure for the partitioning between radiation scattered in the forward and backward hemispheres. Computation of the orientation-averaged asymmetry parameter may require a smaller number of orientational angles. This is, at least, my guess, but it should be checked.

- Lines 11-12: "[...] decreases the backscattered intensity". The same comment applies here; to investigate the backscattered intensity, you need to consider the backscattering cross section, not just the phase function. Please change the text accordingly, e.g. "reduces the phase function in the backscattering direction".
- line 23 (and, similarly, p. 26417, line 10): "depolarization features)". It is



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common to refer to the quantity (S11-S22)/(S11+S22) as the degree of linear depolarisation. Thus, it is the Mueller matrix element S22, not S12, that is related to depolarisation (see, e.g., the book by Mishchenko et al., 2002).Note that the element S12 connects the I and the Q component of the Stokes vector. Thus, the presence of this element can convert unpolarised incoming radiation into partially polarised radiation. It is therefore not just unconventional, but even a bit confusing to refer to negative polarisation as "depolarisation".

  - line 25: The term "rays" should be avoided, since it is strictly reserved to geometric optics, which is only valid for particles that are much larger than the wavelength of light. Also, the term "refraction" is mostly used in geometric optics. Perhaps one could say "[...] is due to interferences of partial waves originating from different spatial regions inside the particle". The authors may add a reference to the paper by J. Tyynelä, E. Zubko, G. Videen, and K. Muinonen, "Interrelating angular scattering characteristics to internal electric fields for wavelength-scale spherical particles", J. Quant. Spectrosc. Radiat. Transfer 106, 520-534, 2007.

- p. 26433: I wonder if the phase functions in Fig. 8 are properly normalised according to the normalisation condition on p. 26411, line 13. By a rough visual inspection, the phase function values vary in the range $10^{-4}$–$10^{-1}$. Integrating $(1/2)p(\Theta)\sin\Theta d\Theta$ over angles from 0 to $\pi$, this does not seem to add up to unity. (Note that the phase function has units 1/sr, so you have to integrate over solid angles in radians, not degrees.) Perhaps this figure shows the phase function multiplied by the total scattering cross section? What is the total scattering cross section for these particles anyway?



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## 4 Technical comments

- The abstract almost reads like a little introduction section. I think the authors should try to make it more succinct, while at the same time keeping it self-contained.
- The introduction contains a very good review of previous relevant work. I suggest some minor corrections and additions:
  - p. 26405, line 9: "Garnett, 1904" should be "Maxwell Garnett, 1904". Note that "Maxwell Garnett" is one name; "Maxwell" has nothing to do with the famous Scottish physicist who died in 1879.
  - Same page, line 26: "Liou and Yang" should be "Liou et al.". Also, the authors may want to add a reference to the paper by Worringen et al. here (A. Worringen, M. Ebert, T. Trautmann, S. Weinbruch, and G. Helas. Optical properties of internally mixed ammonium sulphate and soot particles - a study of individual aerosol particles and ambient aerosol populations. Appl. Opt. 47, 3835-3845, 2008.)
- The structuring of Sect. 2 is a bit strange. There is only one subsection 2.1. The usual convention is that there have to be at least two subsections, otherwise the sub-heading should be deleted. Also, within subsection 2.1, there is only one sub-subsection 2.1.1. The authors should either make 2.1.1 into 2.2, or omit all sub- and sub-subheadings.
- p. 26428: Curves E and S are very faint. Perhaps you could choose a different colour, or a different line style.



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## 5 Summary

In summary, the discussion paper needs some extra work before being publishable. Most of my comments are rather minor and straightforward to fix. The main point that needs clarification is the apparently very small number of discrete Euler angles in the computation of the orientation-averaged optical properties.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 26401, 2012.