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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.

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Reply to Comments from Referee #2

The authors greatly appreciate the constructive comments from the reviewer. The reviewer found the study timely and the methodology state of the art, but asked us to resolve a few issues before accepting the paper for publication in ACP. The reviewer distinguished between one main issue and other minor issues that are (were) straightforward to fix. Our response to each comment is discussed below. We revised our manuscript in a number of locations in response to the reviewer's comments. The specifics are listed below.

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?

Response: The comment regarding the number of random target orientations (originally 64 in our paper) was also raised by another reviewer.

We re-computed all optical properties including scattering phase function and degree of linear polarization for 1000 and 3000 target orientations. We found that 1000 and 3000 orientations converged on the same solutions. There were small differences between solutions obtained with 64 and 1000 target orientations, but none of our conclusions have changed. We revised the paper and now report optical properties for 1000 orientations, and we include the results of this sensitivity analysis, as follows.

We modified line 188-193 in the revised paper with the following sentence: "Reported optical properties are the average values computed for 1000 random target orientations (Kahnert et al., 2012) and 9 multiple scattering planes that satisfy the condition |m|kd< 0.3. Optical properties were also computed for 64 (according to Shen et al. (2008, 2009)) and 3000 orientations.

As noted below, we found that 64 target orientations were sufficient to converge on the optical properties (i.e., MAC and SSA) of bare and internally mixed compact aggregates, but a larger number of orientations was required to attain converged orientation averaged values of the MAC of internally mixed lacy aggregates. Predicted optical properties using 1000 target orientations are consistent with results using 3000 orientations."

Results for 64 and 100 orientations are discussed in the revised paper at line 260. "Absorption amplification factors computed as MAC(λ)mixture/MAC(λ)bare at a $\lambda = 550$ nm are listed in Table 2. We also computed absorption amplification factors averaged over the 200-1000 nm range, which were 1.0 ± 0.0 for lacy BC aggregates partially included in NaCl (case FIS), 1.7 ± 0.1 for lacy BC aggregates fully included in NaCl (case FI), 1.4 ± 0.1 for compact BC with an inclusion of NaCl (case GIS), and 2.2 ± 0.2 for compact BC aggregates fully immersed in NaCl (case GI).

The amplification factors stated above correspond to optical properties averaged over 1000 target orientations. Amplification factors computed using 3000 target orientations were the same. Amplification factors computed using 64 target orientations were the same as those stated above for compact BC aggregates (cases GIS and GI), but were

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slightly higher for lacy BC aggregates: 1.1 ± 0.0 for case FIS and 2.0 ± 0.1 for case FI. This illustrated the significance of using a sufficient number of target orientations to converge on predicted optical properties using DDSCAT."

We find that the spectral features of S11 and -S12/S11 are maintained at 466 and 733 nm, using either 64 or 1000 orientations, but the magnitude may vary in the backscattering range between 100 and 180 degrees.

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.

At page 48 of Bond and Bergstrom 2006 MAC values are discussed for freshly generated particles and compared with literature values reported in ambient air.

Response: Agreed. We changed "observed in the atmosphere" to "consistent with freshly generated soot particles"

- p. 26410, line 10-11: "geometrical cross section" should be "optical cross section". Response: Agreed and fixed.

- p. 26411, line 13: add "1=" in front of the normalisation integral. Response: We modified the sentence in the following way at line 181 on page 7

"the scattering phase function (S11(θ)) as the intensity (F) at θ relative to the normalized integral of the scattered intensity at all angles (F(θ) / int(0,π) F(θ)sin(θ)dθ)."

- Same line: replace "Theta = [0, theta]" by " [0, pi]".

Response: Agreed and fixed.

- 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?).

Response: We agree with the reviewer that the Chang and Charalampopoulos data do show some spectral variation. The reviewer is also correct that, since our study is focused on the effect of NaCl and BC mixing, we use constant index for simplicity, as a first order of approximation.

To clarify this point, we modified the sentence on line 184 at page 7 in the following way: “To focus on the effects of aggregation and mixing on optical properties of BC, refractive indices of BC and NaCl were treated as constant in the spectral range between 200 and 1000 nm (Moffet and Prather, 2009), where mBC =1.95−0.79i and mNaCl = 1.5. We assume the density of BC BC =1.8 g/cm3 (Bond and Bergstrom, 2006).”

- 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 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.

Response: The sentence has been modified in the revised manuscript as suggested: “The phase function of lacy BC (case F) has a narrower and stronger forward scattering peak than that of compact BC (case G)"

We presented S11 and -S12/S11 for 64 and 1000 orientations, and we underline that the discussion and interpretation of the results do not change.

– 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".

Response: Agreed and revised as requested.

– line 23 (and, similarly, p. 26417, line 10): "depolarization features)". It is 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".

Response: Agreed and we revised the paper eliminating the text “(depolarization features)”

– 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.

Response: Agreed and we revised as suggest on page 11 and line 303

“This complex structure is characteristic of Mie scattering and due to interferences of partial waves originating from different spatial regions inside the particle (i.e. different scattering angles) (Tyynelae et al., 2007).”

- p. 26433: I wonder if the phase functions in Fig. 8 are properly normalized 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(_) sin_d_ over angles from 0 to _, 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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Response:

We clarified the sentence in the following way at page 7:

“the scattering phase function (S11(θ)) as the intensity (F) at θ relative to the normalized integral of the scattered intensity at all angles (F(θ) / int(0, π) F(θ)sin(θ)dθ). The scattering angle, θ âĹĹ[0,π], is defined as the angle between the incidence and scattering directions.”

- 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.

Response: Agreed. We modified the abstract to be more succinct and focused on results

- 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.

Response: Agreed and corrected.

– 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.)

Response: Agreed. The Liou et al. reference was fixed and we added a reference to Worringen et al.

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- 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.

Response: Agreed We changed subsection 2.1.1 to 2.2 and added the subsection 2.3 in the methodology, which was preciously section 3.

- p. 26428: Curves E and S are very faint. Perhaps you could choose a different colour, or a different line style.

Response: Agreed and fixed.

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