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Interactive Comment

Interactive comment on "Measuring morphology and density of internally mixed black carbon with SP2 and VTDMA: new insight to absorption enhancement of black carbon in the atmosphere" by Y. X. Zhang et al.

Anonymous Referee #1

Received and published: 29 November 2015

1. Summary

The manuscript presents a method for inferring the effective density and particle morphology of atmospheric black carbon cores. Extensive optical modelling work is then developed to investigate the enhancement of light absorption as a function of the coating thickness and core density. This is an interesting idea and worth pursuing. The paper is well organized but it needs a grammar revision and rephrasing. Although the theoretical calculations are well explained, the characteristics and advantages of the measuring system (LII technique), compared to previous work, are poorly described. A

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comparison of the presented setup with more traditional techniques (DMA-APM) would have given more reliability to the absolute values showed here. The relative contribution of BC core density and morphology to absorption enhancement, compared to lensing effect is not stated. The application of the results presented here, especially the core density of black carbon, will be of major interest in radiative forcing modelling. However the manuscript needs rewriting, possible instrumental biases needs to be addressed and comparisons with more traditional technique are required. I do not recommend the publication of the manuscript under its current form.

2. Major comments

I suggest the author to read the paper of Petzold et al. of 2013 (DOI: 10.5194/acp-13-8365-2013) and consider a change in the nomenclature following Petzold's recommendations. In the methods and data section the author should mention the work of Gysel et al. (2011). In this work is explained that DMA-SP2 is a trustful technique to measure BC effective density. On the other side, Gysel et al. (2011) specifies that: "DMA-SP2 system can be used for fast effective density measurements of pure BC particles if an accurate calibration of the SP2 has been done using a APM". The author should specify which particle-selection approach have been used for the SP2 calibration. In this work two instruments based on different principles are compared. Comparability of optical diameter (SP2) and mobility diameter (DMA) of BC-free and BC-containing particles must be mentioned or provided in the supplementary. Especially because the scattering cross section measured by SP2 is a key factor in Dp calculation. My further concern is about the accuracy of your system compared to previous reference techniques used for effective density observation. I would have added an aerosol particle mass analyzer in parallel to SP2 and DMA2. Assuming that the thermo-denuder is 100% efficient in removing the non-refractory components, the effective density could be calculated from both SP2 and APM mass, giving more strength to your results. At the actual status, the accuracy of the technique is easily questionable because no comparison with a reference technique is presented. The features of the SP2 are not fully

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exploited, especially the coating thickness. There is a specific reason for this?

The efficiency of the thermal removal mechanism of non-refractory component is a critical point, but in this work is poorly investigated. Inefficiency in coating removal will positively bias the Cs and then the Dp, and possibly lead to an underestimation of the In-BC effective density, due to overestimation of In-BC core diameter determined with DMA2. Presence of BC-free particles after denuding would consistently affect the size distributions showed in figure 2. The SP2 is a useful tool to estimate the removal of non-refractory component and BC-free particulate after the heating stage. See attached figure. Coating removal may be estimated with LEO-fit approach.

The author assumes that during the coating evaporation the morphology of BC core does not change, this possibility is never mentioned. Did the author consider such process and what would be the effects on the final conclusions?

All the past works on absorption enhancement due to internal mixing of BC with other non-absorbing materials were focused on the lensing effect. The latter is never mentioned here, the reader has the impression that density and morphology of BC core lead the absorption enhancement. It would be interesting to specify the relative contribution of the density to absorption compared to the lensing effect. Figure 5 and 6 might be merged in a scatterplot composed by shell:core ratio as horizontal axes and enhancement as vertical axes. In this way it would be easier to visualize the enhancement due to lensing effect and the relative impact of density in one plot. Even if the coating is assumed to do not absorb light (imaginary part of refractive index = 0), the wavelength at which the absorption enhancement is estimated should be showed. I raise a provocative question. Here you just assume that the difference in dp/dc is due only to aging. It was observed in the past how emission source might change the optical properties of BC (Sandradevi et al., 2008). IS it possible that you are simply measuring two distinct BC types, like traffic and biomass burning emitted BC? Care must be used with climatology statements; I would avoid the term "long range transport" because the author is not able to justify the origin of the particles.

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3. Specific points

The grammatical, lexical and logical errors are ignored here, since a rewriting is needed.

P. 12030 L 5: please specify the manufacturer and possible customizations of the two main instruments.

P. 12030, L 23 - P 12031, L16: I strongly suggest improving the technical description of the SP2 functioning principles. The subchapter is definitely confused and confusing.

P. 12031 L 1: the vaporization or boiling temperature is usually expressed in K. Please add a reference. L 7-10: Were the standard aquadag particles been selected in function of their size (DMA) or mass (APM) during the calibration? Which kind of neutralizer have been used, corona discharger or Kr-source during calibration and atmospheric measurements? L15-16: You proved the validity of the LEO approach only for BC-free particle, where there is no evaporation in the laser beam. The author should show (in the supplementary) that LEO fit was working properly also for BC-containing particles.

P. 12033 L 11: I disagree with the author; in the paper of Cappa et al. (2012) BC density is never mentioned. L 8-20 Both Dp and Dc can be optically measured by the SP2 using the LEO fit. Despite the paper is focusing on aging and coating, a coating thickness derived with the SP2 is never showed. Why?

P. 12034 L 16-19. Unclear. I suggest showing in a table the observed values for each case.

P. 12035 L 9-10: in the SP2 community, what you call here "volume equivalent diameter" is commonly called "mass equivalent diameter".

P. 12036 L 13: I would avoid using symbols or specific abbreviations as subchapter title

L 20-21: In your system particles are selected by a first DMA, then measured by VT-DMA and SP2. SP2 does not select particles since is a destructive technique, at least

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for BC-containing particles.

Fig. 2 One is mass equivalent diameter (SP2) and the other is the electrical mobility diameter (DMA). This should be specified in the label. How the peaks of both SP2 and DMA were determined, which fit was used? Specify here or in the text.

Interactive comment on Atmos. Meas. Tech. Discuss., 8, 12025, 2015.

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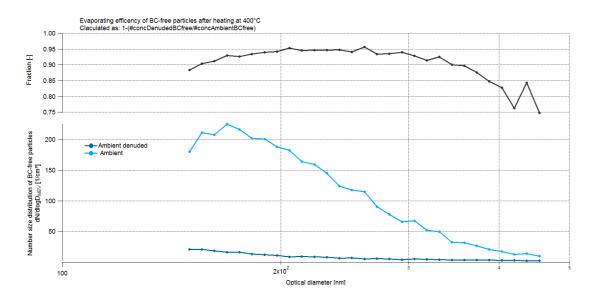


Fig. 1. Evaporation efficency of thermodenuder

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