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Atmospheric Measurement Techniques Discussions

Interactive comment on "Sensitivity of the Single Particle Soot Photometer to different black carbon types" *by* M. Laborde et al.

Anonymous Referee #2

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The authors present a series of experimental results comparing the single particle soot photometer (SP2) response to a variety of black carbon (BC) particle types and present a new method for identifying the presence of "coatings" on BC particles. They compare the SP2 response to particles of known mass that were selected by an aerosol particle mass analyzer (APM) to examine its dependence on particle type and identify the most appropriate calibration material. They use their new coating identification method to determine if mass-selected BC particles are composed of purely BC material, as desired, or if they contain additional non-BC material that biases the APM-derived BC mass values. They conclude that BC emitted from a diesel source is the best candidate for SP2 calibration based on its lack of coatings and the importance of diesel emissions as a BC source.

The material presented in the paper is within the scope of AMT. It includes appropriate references to previous work on this topic and the methods and results sections are, for the most part, described in adequate detail. The writing is clear with some exceptions, some of which have been noted below, and would benefit from copy-editing to fix a few awkward phrases and grammatical problems. It builds on previous comparisons of APM-selected BC particles with the SP2 responses, in particular providing direct comparisons for diesel exhaust, wood smoke, and ambient particles in a different environment from that previously reported. It also includes a description of a new technique to identify coatings on BC particles. Together, these observations and methods represent sufficient new material to justify publication. There are, however, several areas that should be addressed in a revised version of the manuscript before it can be recommended for publication, described below.

General comments

The method and results from the new technique to identify coatings presented in the manuscript needs to be given greater weight in the abstract and introduction, given its importance for interpreting the results. In the current manuscript this new approach is not mentioned in the abstract, but would be of interest to the SP2 community. The introduction should also include some background material on previous attempts to identify coatings on BC particles.

The paper should also provide a more detailed description of the method, including information regarding the size range of BC-cores and total particles sizes over which coating information can be obtained, as well as much more details regarding the experimental uncertainties. They are occasionally referred to but not quantified, nor is there much description of how they were determined. This material is probably deserving of its own expanded section and should also include a comparison to the previously applied time-delay and optical estimates for identifying coatings. The section should also provide more information regarding details of the procedure, such as the specific number of scattering particles needed to determine the laser intensity, how frequently

this was done (are they the average of purely scattering particles for a few seconds before and after the BC particle detection, or something else). Ideally other SP2 users would be able to reproduce exactly the method used in this study from the information provided in the manuscript.

The paper stresses that errors are introduced in comparing the APM-derived BC mass to the SP2 response when additional non-BC material is present. Is there any way to correct for this material using the estimate of the coating provided by the optical data? For example, they could compare the SP2 versus APM response change when the analysis is restricted to particles for which there was no observable difference in the cross-sections.

The introduction should also mention potential variability of the SP2 response to ambient BC. Could be as simple as adding "in different locations featuring contributions by possibly different BC sources such as biomass burning, trash burning, etc." The paper states that diesel is likely the most important BC source in "most locations" but this should be clarified, since biomass burning emissions are estimated to contribute over half of the globally-emitted BC (Bond et al., 2004). Do they mean most urban locations? Or locations where vehicular emissions are predominant?

Specific comments (Pg-Line)

664-2: odd wording here, suggest either omitting "nowadays" or replacing with "now"

664-27: ". . .agrees with the one of. . ." suggest re-phrasing

665-2: suggest changing "curve" to "response" here and throughout the manuscript

665-10: "nowadays"

665-13: "earth" to "earth's"

665-15: "being" to "beings"

666-16: "that one" to "that"

666-25: should also add the CPMA as well as APM here, or refer to the method more generally (e.g., single particle mass analyzers, or something similar)

666-26: though thermodenuders are fairly common in the aerosol community a general reference to the technique is probably still needed here, or else could change to "Heating ambient particles to remove non-BC components using a thermodenuder may provide. . ."

667-23: suggest adding wavelength info somewhere here

668-19: "early in the laser" suggest re-wording (e.g., "determining the coated particle's optical diameter from the initially unperturbed scattering signal when the particle enters the laser beam"

668-24: the coating thickness estimate also depends on the assumed density of the rBC core since this determines the rBC volume/diameter

672-8-9: "within experimental uncertainty" As stated in the general comments, a description of how the experimental uncertainties were determined should be provided, along with an estimate of typical values for ambient particles. What is the critical difference in scattering cross-section needed to be observed to classify the particle as having a coating?

672-9: The approach described here is also similar to that described by Gao et al. (2007) and Schwarz et al. (2008) in that those investigators identified classified a BC particle as being coated if there was a decrease in the scattering signal prior to the onset of incandescence. The approach described in the manuscript is exploiting essentially the same feature in the scattering signal, but in a more quantified way.

668-25: The text describing the method used to identify thin coatings should be a separate sub-section within the methods section. Also, please give range of operating temperatures used with the thermodenuder in the study

669-3: Is the particle being used an example an ambient BC particle, from wood

smoke, or from diesel exhaust?

671-2: what is the physical meaning of the normalization factor? Also, brackets at end of “center” are sub-script.

671-27: there is an extra “/” at the start of the in-line equation

673-3: is this just the engine?

673-21: can any previous ambient sampling studies at the PSI facility be cited here to support this?

675-20: there could also be an absence of coatings if the organics and BC are externally mixed, though this is highly unlikely in this case. Suggest changing “consequently” to “consistent with”.

676-2: What is the potassium sulfate boiling point? Potassium chloride is also emitted and tends to be the majority of the inorganic composition, so may be worth including its boiling point as well. Text should clarify that these materials will be vaporized in the SP2 due to the very high boiling point temperatures of the BC.

676-5: What is the detection uncertainty? It would be useful to have additional lines on the Figure 3 indicated the region of non-significant differences between the scattering cross-sections of pure BC and the whole particle.

676-6: there are dramatically fewer BC particles detected after thermodenuding the wood smoke samples. What is the reason for this? Losses through the thermodenuder are probably not this large. How do the total numbers of BC-containing particles compare for the thermally denuded and non-treated samples? Are the differences in number due simply to different sampling times? If so perhaps it would be better to shade pixels by concentration rather than number. . ..

676-10: Does the Kondo et al. (2011) reference refer to BC-containing particles or total ambient particles? How much of the particle volume would have to remain to explain

the results in Figure 3e?

676-28: The Szidat paper describes measurements performed in Zurich, Switzerland so it is difficult to see how it supports the statement that BC mass is dominated by diesel exhaust in most locations. Emission estimates (e.g., Bond et al., 2004) suggest that over half of BC emitted globally arises from biomass burning of some type. Perhaps changing to most “urban” locations would be an improvement, but even then there are many cities where other BC sources are likely important (e.g., Mexico City; Yokelson et al. 2007). The Szidat reference should be cited to state that the ambient sample referred to in the manuscript is likely to be dominated by diesel emissions, or at least fossil fuel emissions, based on the carbon isotope analysis.

677-14: “An unbiased ambient calibration would likely. . .” In the absence of any estimate of the magnitude of the bias this statement isn’t particularly meaningful. If the bias were large enough the diesel calibration could potentially fall quite far from the ambient data on the other side of the 1:1 line. If the authors could provide an upper estimate of the bias it would help support the claim made here.

Figures

The aspect ratio of Figure 4 is quite horizontal and it would be easier to see the differences between the curves if it was made more square in the AMT version. Error bars should be described in the caption as standard deviations or appropriate parameter. It might also be useful to indicate an approximate range for ambient BC particle size/mass based on previous SP2 literature reports.

References

Bond et al., 2004, doi:10.1029/2006JD007315 Yokelson et al., 2007, http://www.atmos-chem-phys.net/7/5569/2007/

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