Atmos. Chem. Phys. Discuss., 15, C12732–C12737, 2016
www.atmos-chem-phys-discuss.net/15/C12732/2016/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantification of black carbon mixing state from traffic: implications for aerosol optical properties" *by* M. D. Willis et al.

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Received and published: 24 February 2016

Response to Anonymous Referee # 1

We thank Referee #1 for their thoughtful reading of this manuscript, and for their important questions about the SP-AMS technique. Our responses to specific comments and the corresponding changes to the manuscript are detailed below.

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General comments

1. A potentially major technical issue relates to the fact that in the SP-AMS, it is not assured that the particle will completely vaporise. If the particle beam is wider than the laser beam (which given that soot particles are non-spherical, is a distinct possibility), then particles may pass through the "tails" of the laser beam, which may mean that the particles absorb sufficient energy to vaporise the coating of the particle but not the core. This would occur if the peak temperature reached was between the boiling points of the coating of the core, which given this covers a temperature range of thousands of degrees, this is a distinct possibility. Furthermore, a report of this behaviour in diesel emission particles was presented at the most recent AMS users' meeting: http://cires1.colorado.edu/jimenezgroup/ UsrMtgs/UsersMtg16/JDASPAMSfocusing.pdf. In this paper, the reported population of particles that contained little or no rBC could be attributed to this incomplete vaporisation occurring. It could also give rise to the PMF result as well. The authors should see if they can discount this as a possibility, or failing this, add this possibility in as a caveat. In the worst case that the observation of the "HOA rich" population turns out to be erroneous, what effect would this have on the paper?

Authors' response: The incomplete evaporation of rBC-containing particles in the "tails" of the laser vaporiser is indeed a distinct possibility in our measurements, and is now more explicitly addressed in this paper. As the reviewer states, incomplete vaporisation can take place when the particle beam is wider than the laser beam and this may be the case in our measurements (as mentioned in the Methods section, beam width probe measurements used to estimate the collection efficiency at the roadside site suggest this is the case). We agree that this could certainly have an effect on the mf_{rBC} measured by the SP-AMS; however, we believe we can discount the possibility that the HOA-rich particle class is an artefact caused by incomplete vaporisation because HOA-rich and rBC-rich particle classes have different size distributions in single particle data. If HOA-rich particles arose exclusively from incomplete vaporisation

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in the edges of the laser, resulting in underestimated mf_{rBC}, we would then expect HOA-rich and rBC-rich particles to have the same size distributions (i.e., the particle-time-of-flight would be the same whether the aerosol was completely or incompletely vaporised). In contrast, we observe that HOA-rich and rBC-rich particle classes have distinct size distributions, supporting the conclusions that their mass spectra arise from different particle types. The importance of the size distributions for interpretation of our results has been highlighted in the revised version of the paper (Section 3.1).

Though we can discount that the HOA-rich population is erroneous, we agree that it is very important to discuss the effect that incomplete vaporisation could have on the mf_{rBC} values we report. To better address these uncertainties a more detailed description of uncertainties in SP-AMS measurement of mf_{rBC} has been added to Section 2.1 as follows: "...two additional uncertainties in SP-AMS measurements may affect calculation of mf_{rBC}. First, there are uncertainties in the recommended RIE for organic species evaporating from rBC in the SP-AMS of up to $\sim$50% (Lee et al., 2015; Willis et al., 2014), which could cause an overestimation in the mass of coating material and a corresponding underestimation in mf_{rBC}. Second, it is possible for rBC-containing particles to pass through the edges of the laser vaporizer, thus producing a heating effect sufficient to evaporate some fraction of the coating materials but not evaporate the rBC itself. This effect may also lead to an underestimation in mf_{rBC}. SP-AMS CE and quantification are discussed in further detail in Section 1 of the Supplement." A reference to the cited AMS Users' Meeting report has been added to Section 1 of the Supplement, along with a more detailed discussion of incomplete vaporisation. An explicit reference to Section 2.1 and uncertainties in SP-AMS mf_{rBC} has also been added to Section 3.1 to make clear to the reader that these uncertainties must be considered.

2. Generally speaking, there is perhaps too much of a tendency to put things in the supplementary material. While this would be considered usual practice for a journal

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with a strict word or page limit, I feel that certain sections of the supplement would be better featured in the main article as they contain information very pertinent to the paper's conclusions. Personally, I would consider that sections 2, 3, 4 and 7 may be suitable for the main article.
Authors' response: Sections 2, 3, 4 and 7 of the Supplement have been added to the main paper, and Figures 1, 2 and 3 have been modified to include data from both the roadside and non-roadside sites. The discussion and figure relating to particle coating thickness estimation have been added to the main text of the paper.

Specific comments

1. Little detail on the PASS-3 operation is presented here. Why was the 405 nm channel used? How was it calibrated? Was any attempt to correct the scattering channel for truncation made?
Authors' response: Omission of this information was an oversight on our part and a new section has been added to the Methods describing the PASS-3, as follows:

"A photoacoustic soot spectrometer (PASS-3, Droplet Measurement Techniques, Boulder, CO) was used to measure aerosol absorption (b_{abs}) and scattering (b_{abs}) coefficients (Mm^{-1}) at 405 and 781 nm. A 532 nm laser is not installed in this particular unit. The PASS determines aerosol absorption (Mm^{-1}) in a cavity which acts as an acoustic resonator. The absorption of incoming radiation heats the particles, which in turn heat the surrounding air in the cavity (Arnott et al., 1999). The aerosol-laden air thus expands, resulting in a pressure disturbance. By modulating the laser power at the resonance frequency of the cavity, the pressure disturbance is amplified and the resulting acoustic wave is measured using a microphone. Light scattering at both wavelengths is concurrently measured using reciprocal nephelometry (Moosmüller et al., 2009; Flowers et al., 2010; Chan et al., 2011). Signals were not corrected for

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truncation; however, it should be noted that total particulate loading is relatively low at this site and saturation was not observed in scattering signals. The instrument was calibrated using a propane soot generator (miniCAST, 6203A, Jing); since a 532 nm laser was not present in this unit an NO_2 calibration was not possible. PASS measurements of the bulk single scattering albedo at 405 nm (selected due to superior signal-to-noise ratio for scattering relative to the 781 nm channel) are used here only to illustrate differences in optical properties in vehicle plumes with varying composition."

2. Regarding the use of PMF, I would request that the authors include the graphs from the rejected solutions as well in the supplement, so as to justify their choice of solution. **Authors' response:** The relevant plots for the 2 – factor, 3 – factor and 5 – factor solutions have been added to the Supplement.

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