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Interactive Comment

Interactive comment on "Single particle characterization using the soot particle aerosol mass spectrometer (SP-AMS)" *by* A. K. Y. Lee et al.

Anonymous Referee #2

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Lee et al., describe results obtained with the Aerodyne Soot-Particle Aerosol Mass Spectrometer equipped with a light scattering module to enable single particle characterization of both refractory and non-refractory particle constituents (for particles larger than the optical detection limit of the instrument). The manuscript focuses on four days of continuous sampling in downtown Toronto, from which the authors demonstrate the capability of the LS-SP-AMS through single particle k-means cluster analysis and bulk ensemble positive matrix factorization (PMF) approaches. The work is an important contribution to the field especially in the context of improving our understanding of the mixing state of ambient particulate matter (including combustion-derived rBC) within urban environments. The manuscript is well written and the insights drawn from the work warrant publication in ACP. The following comments should be addressed by the

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authors prior to final publication.

General Comments

Grey area terminology:

- Throughout the manuscript the authors refer to ‘low to mid-range aerodynamic diameter’ this range should be explicitly defined. The first mention of an actual mid-range diameter is not found until P15337 L12.

- ‘HOA-rich’ : how is this classified?

- ‘Nearly external mixing’ : I believe the authors wish to convey the fact that they measure nearly chemically pure, distinct, particle populations within the ensemble. For example, a chemically pure rBC-only particle type or HOA+rBC particle type or pure HOA particle type. The authors should carefully consider what they mean by ‘nearly external mixing’ and discuss external and internal mixing from the ensemble and single particle perspectives. Within any given urban environment there will be a gradient of mixing and the exact nature of this gradient is dependent on many factors. The authors should make a concerted effort to more clearly define what they interpret as externally vs internally mixed particle populations in the context of the real-world gradients of particle chemistry encountered in an urban environment like downtown Toronto.

- In paragraph 2 of P15325 the authors introduce the term refractory BC (rBC) in connection with the SP2-defined measurement technique. Prior to (and following) this the authors use ‘BC’. I think it would be helpful to the general reader to include a sentence describing the difference between rBC and BC. The authors should make a point not to switch back and forth between using these two terms throughout the manuscript, if possible.

- The authors make an effort to differentiate the SP-AMS from the LS-SP-AMS and this ends up causing more confusion than clarity. I recommend that they use LS-SP-AMS throughout the manuscript and describe the menu-switching modes of operation to

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differentiate single particle from ensemble modes of operation. The introduction of the LSSP mode (which is not defined, when introduced on P15328, L6) further confuses the acronym-overloaded of the text. Consider removing this, while still clearly defining the single particle mode of operation.

- P15331, L23: Authors should specify the # of clusters that can be combined with the CAP program – not just 'a few'.

Specific comments:

P15324, L20-21: The general reader may not be familiar with 'the conventional interpretation' of PMF results and as such, this language should be avoided or more concretely defined.

P15325, L1: Do the authors mean to highlight the varying morphology of the 'coating' on the BC or the particle as a whole.

P15326, L18: Should read 'flash vaporize at $\sim$ 600 C'

P15327, Section 2: Should include a more comprehensive description of the sampling location and sampling inlet configuration relative to near-by source of pollution. Specifically, proximity to major roadways and COA sources (such as food trucks, carts, etc.) would help place the results in a more relevant context. If possible, inclusion of a map of the area surround the sampling site may be particularly useful for the reader. This inclusion would also allow the authors to draw finer lines of connectivity between their results and important sources of particulate matter within busy, active, urban environments. They should also make a point of identify the days of week across which sampling occurred (weekday vs weekend) – and the difference that makes to likely emission sources in the area.

P15327, L22: V-mode should be describe. This is a meaningless term to readers not familiar with the AMS. Define mass resolving power for your instrument, during this campaign.

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P15327, L23: It would be helpful to add a sentence describing the motivation behind running the instrument in laser-on vs laser-off mode throughout your sampling campaign. To the unfamiliar reader, this just looks like you are cutting your duty cycle on sampling rBC down by 50%.

P15328, L10: Consider adding a header here: Calibration

P15328, L22-28: I would strongly recommend removing the equation for effective density from within the text and writing the formula out with label: Eqn. 1. In fact the authors should define effective density in this section. Once organized as Eqn 1, the authors can more easily refer back to this equation later in the manuscript (P15337 L4-5). - In connection with the discussion of effective density – as the authors point out – highly non-spherical particles (i.e. soot agglomerates, dominated by rBC) will appear at lower dva values than typical accumulation mode particles. Given the importance of this type of particle to results discussed in this manuscript, the authors should briefly discuss the influence that highly non-spherical shape has on the particle beam width as it transits the time-of-flight chamber of the LS-SP-AMS / intersecting different regions (intensities) of the 405 nm scattering laser and 1064 SP laser.

P15329, L10: Consider revising sentence: 'overlaps perpendicularly with the particle beam $\sim$ 0.265 m downstream of the chopper and $\sim$ 0.130 m upstream of the tungsten vaporizer as shown in Fig. 1.

P15329, L15: The authors should define what they mean by 50% cut-off diameter.

P15329, L19: should read: 'may have a lower cut-off dva than…'

P15330, L2: Consider adding a sentence describing the width of the chopper slit used in the LS-SP-AMS (0.5, 1, 2%) and how this width would impact the ability to accurately size (LS_dva) and predict arrival times at the vaporizer.

P15330, L7: Avoid use of term 'rotten'

P15330, L9: Is it possible to relate the >/= 6 ions/spectra threshold to an estimate of

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mass-based (fg/particle) detection limit?

P15330, L13: It would be informative to include the Gaussian distribution figure in the supplemental. Why did the authors choose the 3 sigma boundary as their classification of prompt vaporization-ionization? What is the physical explanation for an 'early' ion pulse arrival time?

P15330, L25-27: The authors reference Fig S1 as evidence of clear separation of the prompt single particles from all other particle types. Although it is clear that their 3 sigma classification for prompt particles has identified the vast majority of real- LS events from noise events – it is not clear that from the graph that early or delayed particle vaporization types are effectively identified. Rather than plotting the MS ion scatter plot vs LS_dva – the authors could consider plotting and differentiating the early, prompt, and delayed particle types vs MS_dva, whilst leaving out the 25% of LS events that were noise. This, combined with the Gaussian distribution of arrival times may provide a more comprehensive separation of the prompt-vaporization particle types from all other non-noise particle events.

- It is very interesting to see the prominence of single particle events that appear in S1b that result from having the SP laser ON – particles comprised of rBC, rBC+HOA. This begins to show the potential for determining a size and chemistry-dependent collection efficiency for the LS-SP-AMS sampling across a matrix of particle types encountered in an urban environment. Did the authors complete any size-dependent CE assessments (whether from the perspective of non-spherical rBC-dominant particle types or larger, accumulation mode particles with higher SO4 content)?

P15331, L1: The authors should explain their justification for using only the prompt particles in their cluster analysis. Did the early and delayed particle types show consistently lower total ion counts for the same LS_dva when compared to prompt?

P15332, L12-16: Graph or correlation coefficient should explicitly stated comparing LS-SP-AMS mass vs SMPS. SMPS assumptions about particle shape and density should

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be given.

P15332, L25: Should read 'average ensemble size distributions..'

Section 3.1:: Opportunities to link the observations of particle chemistry back to details about the sampling site and potential sources – this would follow from a more thorough description of the sampling site back in section 2.1.

P15334:: The authors introduce Figure 4 at this point in the paper, but only describe the PMF portions of the analysis. The next mention of Figure 4 comes in the very last section of the paper (3.7) – connecting the cluster analysis results (also displayed in Figure 4). Splitting the descriptions of Figure 4's overlapping content is confusing and the authors should re-consider reorganizing this content so that the resultant figure is more coherent with the text or vice versa. One thing that draws the reader's eye is the presence of the three red rectangles in the figure – which are highlighting areas of agreement and disagreement within the data. These are not mentioned until P15342, L23.

P15339, L16: How does the rBC-rich cluster's C1/C3 ratio of 0.53 impact the quantification of total BC mass concentration, given that the frag table assumes a C1/C3 ratio of 0.61? Related to this, it is not clear why the authors choose to use the C1/C3 ratio obtained from regal black instead of that acquired from the HR analysis of the ambient rBC (which is used in the case of the C4/C3 and C5/C3 ions as described in Table S2).

P15341, L14: The authors should take care in explicitly stating that the rBC-rich particles they measure have a 'thin HOA coating'. As the radiative impacts of this particle type vary dramatically based on the nature of the associated HOA on the surface of the BC, it may not be appropriate to classify this as a thinly HOA-coated rBC particle – based on the LS-SP-AMS measurements alone. It is certainly clear that the authors have a particle population that is comprised of rBC+HOA components, but how they are distributed within the particle itself, remains ambiguous with the current instrumentation. Similarly, classifying HOA coatings as 'thick' may also be inappropriate.

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Comments on Figures

Obviously, the authors put a tremendous amount of effort into this analysis and as a result of the complexity of the dataset and diversity of analysis approaches applied, have generated many figures. I worry that the sheer number of figures obscure the main take home points of the manuscript. Of the 8 figures within the manuscript body itself, Figures 7 and 8 stand out as important demonstration of the power of the technique for single particle characterization. Given the information in Figure 7, Figure 5 becomes somewhat redundant and the color scheme used in Figure 5 leads to some confusion (red, blue, green – signifying metrics that are not sulfate, nitrate, organic). The same comments applies to the colors used in Figure S9. The data displayed in Figures 2a and 4 could also be re-combined into a single figure, along with appropriate reorganization of text within the manuscript itself. Figures S2 and S10 seem unnecessary. Their content could be effectively conveyed with text alone.

If Figure 5 is kept in the manuscript, the authors should plot their empirical data that comprises the NH4NO3 d50 line. Likewise, this data could be included in Figure S9, if it remains in the final manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 15323, 2014.

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