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

Anonymous Referee #1

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General comments:

In their paper “Single particle characterization using the soot particle aerosol mass spectrometer (SP-AMS)”, Lee et al. describe the first deployment of a soot particle aerosol mass spectrometer coupled with a light scattering probe, enabling single particle and ensemble analysis of both refractory black carbon and non-refractory species. They apply k-means clustering to the single particle mass spectra, and compare to the results of positive matrix factorization analysis of the ensemble data. The paper is well and clearly written and provides relevant information on the methods of single particle and ensemble analysis with the SP-AMS, as well as some interesting insights into particle mixing state. However, since the main focus of the paper is rather on a technical (method) development, it could be argued that this paper would be more suitable for

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AMT than for ACP. I therefore recommend publication either in AMT or as a “Technical Note” in ACP, subject to the following comments.

Specific comments:

1.) It is not made clear from the experimental section what data is acquired in LSSP mode and how this data is processed. Specifically, the statement on p. 15329, lines 24-25, is not correct: the computer is not triggered to save “the corresponding mass spectrum from the single particle”, it saves the whole PToF trace, from which afterwards the single particle mass spectrum is obtained. This is especially important as both PToF traces and integrated mass spectra are used later on (in Figs. 5 and 7, I think; this should be stated more clearly). Is the dva from LS or MS signal used in Fig. 7?

2.) The categorization of single particle mass spectra. First of all, “rotten” sounds rather colloquial to me to describe a category of mass spectra. The single particle data the authors describe rather seem to be “low in LS signal” or something similar. By looking at Fig. S1, it seems that the threshold during the data acquisition was set rather low, i.e. many saves were triggered by noise in the LS signal. So, instead of using rather arbitrary values (“signal < 0.2”), why not simply calculate a new threshold from the data in Fig. S1 using some kind of “baseline” region (e.g., dva < 40nm)? A similar approach could be used to determine a threshold for “null” mass spectra.

In relation to this, I am not quite sure about the comparison to the fraction of “null” particles found by Cross et al. and Liu et al. (p. 15336). While most of the reasoning seems valid, I am not sure on which evidence the authors base their assumption they had “a more sensitive light scattering detection system” than Liu et al. (lines 13-14). Furthermore, as said above, the threshold settings for data acquisition may have a large influence here; there may also be influences like different size distributions / chemical composition.

3.) As written, there seem to be some contradictions regarding the collection efficiencies: on p. 15332, the authors claim $CE = 1$ for NR-PM after comparison to an SMPS

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system. They do not base this CE on any theoretical reasoning (average chemical composition to justify this?). Later on (Section 3.3), it is observed that NR-PM components are biased low by 10-20% in laser off compared to laser on mode, which implies CE >1 for laser on mode. Then in several places (e.g., p. 15337), particle bounce is discussed, implying CE <1. Furthermore, when single particle data are analyzed, it becomes obvious that about half of the particles giving an LS signal do not give significant ion signal, i.e. CE <1. This needs some clarification.

4.) Concerning the statement on p. 15339, lines 22-25 (and related statements elsewhere): “In contrast, the clustering analysis clearly suggests that two major types of particles contributed to the PMF-HOA factor. The different size distributions of the two clusters also indicate two distinct particle types.” This claims there are two particle types, wholly unconnected, separated into two clusters. Is it not more likely that there is a gradual degree of mixing between HOA and rBC? This could be easily tested by plotting e.g. HOA vs rBC mass for particles from these two clusters. Moreover, in other places of the manuscript the authors seem to contradict themselves on this topic (e.g., on p. 15341, lines 17 and 21, they suggest that also the “pure” HOA particles contain a small rBC core, which rather seems an indication of gradual degree of mixing). This needs clarification. Furthermore, can the detection limit of rBC be somehow approximated? It is discussed on p. 15342, line 12, that rBC may be underestimated due to low ion counts. Could this be tested by comparing the average chemical composition derived from ensemble and single particle measurements?

5.) Similar to the discussion of distinct particle types vs gradual degree of mixing of rBC and HOA, it should be noted that the four particle cluster types of nitrate, sulfate, OOA cluster 1 and 2 likely represent four “extremes” of a large group of particles, all of the same type (internal mixture of sulfate, nitrate, OOA). This again should be easy to see in an according diagram (e.g. ternary plot).

p. 15327, line 15: Can the measurement location be described a bit more in detail? Is it adjacent to a road, or rather in a backyard? What’s the traffic density, inlet height?

p. 15332, line 15: provide model of SMPS instrument. What assumptions were used to infer mass from the SMPS measurements, and how well did it correlate exactly (slope?)

p. 15332, lines 20-24: Which m/z range was investigated in this study? I think the information on fullerenes could be easily improved by a simple back-of-the-envelope calculation: assuming the ratio found by Onasch et al. from the fullerenes to the smaller C_x ions, would a significant signal of fullerenes be expected at the observed signal of lower C_x fragments?

p. 15335, line 21: KNO_3 is not a good example of refractory species, since it is very easily thermally decomposed and detected very well with the AMS. Why should more acidic material be present in internally mixed rBC particles than in others? For very aged material, this does not seem very likely.

Technical corrections:

The authors use the different terms “LS-SP-AMS” and “SP-AMS” sometimes on purpose to distinguish between SP-AMS with and without light scattering module; but sometimes, both terms seem to be used interchangeably (e.g., p. 15342: In line 3, it is “SP-AMS” on purpose, because here we talk about an “usual” SP-AMS. But in line 7, again “SP-AMS” is used, where it is talked about the single particle data from this work. So, it should be more precisely “LS-SP-AMS” here.) More care should be taken in the whole manuscript to use the more appropriate of both terms in each situation.

Also, it might be confusing to the reader that on the one hand, there is the “LS-SP-AMS”, on the other hand, there is the “LSSP mode”. Maybe it could be shortened to “LS mode” to make the difference more clear? Especially since in one case SP means “soot particle”, in the other “single particle”.

p. 15324, line 18; Section 3.6; and p.15344, line 4: “low to mid-range aerodynamic diameter” could mean anything from a few nanometers to several μm . Please specify at least once the size range.

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p. 15325, line 13: “quantitative single particle detection”: do you really mean that the (optical) detection of the particles should be quantitative (i.e., the number concentration should be correct), or do you mean quantitative chemical composition analysis of the single particles which were detected (i.e., getting quantitative mass per particle information, i.e. do you want to stress the difference between AMS and ATOFMS single particle data)? Maybe reword to make more clear.

p. 15326, line 20: “which is able to quantify and characterize rBC and NR-PM”: sounds strange. How do you “characterize rBC”?

p. 15327, line 6: “SP-AMS ensemble measurement data”

p. 15327, lines 13 and 22: give a reference for “standard HR-ToF-AMS” and “V-mode”.

p. 15328, line 2: “the NR-PM that is internally mixed with rBC core”: strange wording. Maybe just delete “core”?

p. 15328, lines 18-19; and p. 15330, line 4: As reference, the URL should be provided where Squirrel / PIKA / Sparrow can be obtained.

p. 15330, line 18: “number of ions is the sum” should read “number of ions was calculated as the sum”

p. 15331, line 8: this should read “total ion signals in a single particle mass spectrum” more precisely

p. 15333, line 4: specify “SOA”

p. 15337, line 4: “Chi” should read “S”. Also, the same equation is given before (p. 15328). Maybe break up into a separate line there and simply refer to “Eq. (1)” later.

p. 15337, line 26-28: It is unclear what this sentence is supposed to say in the present form. If I understand correctly, what is meant is that clusterings of both laser on and laser off datasets were performed, and here only laser on clustering results are shown as an example. This should be made clearer here, especially since the results from

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both clusterings are compared later on.

p. 15338, line 3: should read “reduces” to not mix different tenses within one sentence.

p. 15338, line 11: “procedure” – do the authors mean “produce”?

p. 15339, line 9: rather “do not contain any inorganic species”

p. 15339, line 10: considering the associated uncertainty, “about 6%” seems more appropriate than “6.4%”

p. 15343, line 18: comma missing: “other, non-refractory”

Table 2: define “NA”. What does “measurable m/z 36 signal” mean? Any signal larger than zero? Signal above a certain threshold?

Figure 2: define “Org44”; it should be noted that these data are from ensemble measurements.

Figures 2 and 4: Is this local time? Please specify.

Figures 5, 7 and S9: The purple and grey lines are hard to distinguish.

Figure 6: b) how thin is a “thin” coating? Maybe would be better described as an “internal mixture of rBC and HOA”. c) should be “is classified” instead of “are”.

Figure 8: For clarity, it could be stated in the figure caption that these are results for the clustering of the “laser on” dataset. Also, what is the inset in panel (d)? Please clarify.

Figure S7: in which units are the signal intensities given?

Figure S9b: Label on the right y-axis is unclear. Should this read “. . .ratio or Total. . .”?

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

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