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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 #3**

Received and published: 25 July 2014

The manuscript “Single particle characterization using the soot particle aerosol mass spectrometer (SP-AMS)” by Lee and coworkers reports the first deployment of an Aerodyne aerosol mass spectrometer equipped with both an infrared intracavity laser for black carbon measurement and a single particle light-scattering probe. This combination was used during a four day period in downtown Toronto. The collected data was then analyzed using standard AMS methods, positive matrix factorization as well as cluster analysis using the k-means algorithm.

General comments:

The manuscript is well structured and written, although in a few places the language could use a revision regarding sentence structure, grammar (tenses) and punctuation.

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In some parts the use of abbreviations and “AMS” specific terms could be reduced to improve readability for readers less familiar with the Aerodyne AMS.

The manuscript has some major shortcomings that should be addressed prior to publication.

- In large parts it deals with the techniques and methods applied to obtain and analyze the data. The results of the mathematical methods are compared regarding the methods used; the atmospheric relevant results and their implications, however, are only shortly mentioned and not really discussed.

- It is not comprehensible nor explained, why the authors use the k-means algorithm instead of its successor. K-means cluster analysis has some major shortcomings that have been addressed long ago by the c-means (Fuzzy) algorithm. K-means requires each data point to belong to one (and only one) cluster, a condition which is difficult to justify for atmospheric aerosols with a history of constant mixing and exchange.

- In general both algorithms (k-means as well as c-means) do require equal distribution of data points into all clusters and thus are prone to oversee small clusters. This issue could be addressed by in addition using a different cluster method for comparison, for example hierarchical clustering or a neuronal network like ART-2A.

- I recommend that the authors reconsider the main focus of the manuscript. If they want to keep the current focus, the methodical part should be enhanced, and the manuscript should be moved to AMT. To merit publication in ACP, the authors have to shift the focus towards a more scientific (non-methodical) discussion and atmospheric relevance.

Specific comments:

P15324, L21: What is “the conventional interpretation of the PMF results”?

P15324, L25-26: “Processes such as . . . are the primary sources of ambient BC.” This suggests that there are other sources for atmospheric black carbon particles. Which

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sources would that be?

P15325, L13: “Real-time, quantitative single particle. . . ”: the comma is unnecessary.

P15326, L1: “In contrast, the Aerodyne . . . separates the particle vaporization and ionization steps . . .”: “In contrast” should be removed because in this context it is wrong. For example, the SPLAT II (Zelenyuk et al., 2009) also uses a two-step vaporization and ionization process.

P15326, L3-5: (Jayne et al., 2003) should be cited as a reference for the Q-AMS (when mentioning different versions of the AMS).

P15327, L1: “Clustering analysis” should be changed to “Cluster analysis” or simply “Clustering”.

P15327, L9ff: More details on the measurement location would be favorable, e.g., was it close to a major street, restaurants, etc.

P15327, L23: “The IR laser . . . was switched on and off during data acquisition.” This sentence sounds very trivial.

P15327, L23-26: Please correct the grammar: “. . . the SP-AMS was operated . . . ., whereas the instrument . . . detects both . . .”

P15328, L14: “Regal 400R Pigment” should be “Regal 400R Pigment black”.

P15329, L10: “. . . of a particle sizing chamber . . .” should be “. . . of the particle sizing chamber . . .”.

P15329, L15: What was the lower cut-off diameter for the “Regal black” particles?

P15330, L5-6: “The weak scattering trigger events (i.e., . . .).” Where do these values come from? What is the unit of the scattering signal?

P15330, L7: Please replace “rotten” with another expression, for example “useless”. Or just paraphrase it.

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P15330, L8-9: How is a “strong” scattering signal defined? Why six ions? And how are the ions calculated? By integrating over the mass spectrum divided by the (measured) single ion?

P15330 L10-12: “. . . could accurately predict . . . the particle is categorized . . .” Correct the grammar. Why “Specifically. . .”?

P15330, L19: Was there a reason for removing potassium? If so, where did the high background come from? For sure, it did not come from the gas phase. Did you use the high resolution data do check if  $m/z$  39 really was potassium, and not for example  $C_3H_3^+$ ?

P15331, L1-8: Again, c-means should be favored over k-means, especially for a atmospheric data where a distinct separation of different populations is very unlikely.

P15331, L17: “A Clustering Analysis Panel was developed . . .”: This is “IGOR slang”. A reader not familiar with Wavemetrics IGOR Pro will not understand, what a panel is (in this context), or why it is worth mentioning. Please rephrase.

P15332, L7-8: Why is the “collection efficiency” for uncoated Regal Black particles lower than for coated? Besides, I find the term “collection efficiency” not really suitable when used with the intracavity laser vaporizer. Wouldn’t “incandescence efficiency” be a better term?

P15332, L21-24: Why are these observations by Onasch et al. mentioned, if the authors did not see anything alike (as mentioned before)?

P 15333, L1: Why is that? Are there only local sources for rBC, or are there any sinks? If so, what could these local sources be? Could it be that particles containing aged rBC are simply not vaporized by the laser (e.g., low black carbon content, high organic and inorganic content, low absorption at 1064 nm)?

P15334, L6, and Figure S4: The residual (of the PMF analysis) is quite high, especially in the first two days and at the end (10% and higher). Any explanation?

P15334, L28: If there was significant particle bounce, why did the authors assume a CE=1 for the tungsten vaporizer?

P15335, L28: 75% in “laser on” mode, in “laser off” mode, or total?

P15336, L13-15: How do the authors know that their light scattering system was more sensitive than the one used by Liu et al. (2012)?

P15336, L22-24: “. . . the single particle size distributions measured . . . are generally consistent with the ensemble PToF data (Fig. 2c).” This cannot be seen in this Figure.

P15337, L20-21: How should the laser vaporizer have bounce effects?

P15337, L26ff: It would be helpful to mention why the twelve-cluster solution was selected.

P15340, L17: What does low-range Dva, what mid-range Dva stand for?

P15344, L8-9: As stated by the authors before, it could well be that they were not able to detect rBC in larger particles. Thus this statement should be modified.

Table 2: What are the uncertainties and errors of these values? Is a difference of 6.4% between “laser on” and “laser off” HOA significant at all?

Figure 2: Please hyphenate “laser on” and “laser off”, or use it with quotes (throughout the manuscript).

Figure S1: The sharp cut-off for “laser on” prompt particles should be explained either in the diagram or in the caption.

Figure S2: Why did the authors select 12 clusters for their analysis? Is there a relation with the cluster distance? What is the purpose of this plot?

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

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