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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Quantitative determination of carbonaceous particle mixing state in Paris using single particle mass spectrometer and aerosol mass spectrometer measurements" *by* R. M. Healy et al.

Anonymous Referee #1

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General comments:

Overall, this is a strong paper that describes a significant effort in data analysis. The authors integrate and compare data across platforms in new ways, and set a precedent for what can be done with data from a variety of aerosol mass spectrometry experiments. The authors have treated their data carefully and have provided a thoroughly documented report of their work. The results are surprisingly good, in that the single-particle mass spectrometry data, often presented as non- or only semi-quantitative, proves to be comparable to that of the AMS, which is often portrayed as fully quantita-

tive, while providing more subtlety in the analysis. The paper is strong and should be published, but in its current form, the paper is too long and I believe that the balance is not correct, between descriptive results and method evaluation.

Specific comments:

1. I believe that a shortcoming of this paper is that it is presented as an exhaustive report rather than as a presentation of a scientific story. It could be dramatically improved if the authors were to tailor it to one of the two major themes which are present in the research: 1) the result that is relevant to the air quality in Paris, that most PM1 sampled there is transported to the city, or 2) the result that is relevant to the quantitative analysis of atmospheric aerosol data using mass spectrometry. Right now, it seems that both themes are emphasized, which has the effect that neither one stands out.

2. The paper should include some general statements about which instruments need to be co-located to do an analysis such as the one presented, and to discuss the aspects of the method that can be replicated or exported to other studies.

3. A discussion of how generally applicable the authors think these results are, would be welcome; do they think these good correlations between ATOFMS and AMS data will be observed everywhere, or are there special conditions in Paris which facilitated them?

4. In general, I feel that the method developed should be presented within the results section of the paper, as it is a new application of a data analysis method. It requires significant discussion, and seems out of place in methodology.

5. p. 10350, line 26: It is not clear if the particles sampled by the ATOFMS were dried as were the particles sampled by the other instruments. This should be mentioned here and, if they weren't, the implications of this should be discussed in the results.

6. p. 10352, lines 10 – 12: This statement suggests that there were no metal/inorganic particle types that made up significant classes. This result should be compared to

other urban areas that have been studied. Is this typical or novel for Paris during this measurement?

7. p. 10353, lines 14 – 19: The authors should also use naturally occurring isotope patterns as evidence for the fact that isobaric interferences are not likely a problem, when possible.

8. p. 10354, line 2: Was the relative peak area really obtained by normalizing each peak to the sum of all ions in both the positive and negative spectra? Most examples in the literature show normalization to total peak area of the same polarity of the ion under investigation.

9. p. 10355, line 1: There needs to be a description of how the mass concentration of ATOFMS classes is obtained.

10. p. 10356, line 14: The number and duration of the back trajectories is ambiguously described: is it 72 hour back trajectories or 70 2-hour back trajectories?

11. Section 3.1: This section should be shortened significantly (50 % or so). Much of the general description of the various classes can be provided in a table, or the text can be moved to the Supplementary section. The information that should be retained in the text of the main manuscript is the interpretation of the clusters. For the example of the K-OA cluster, this would be p. 10358, lines 13 – the end of the paragraph (starting with "The strong diurnal. . .").

12. Section 3.2 is written from the perspective that the ATOFMS is the only instrument contributing to uncertainty. The uncertainties and assumptions inherent in the other instruments' quantitative results should be mentioned and discussed.

13. Section 3.3 is, it seems to me, the heart of the paper, but it is buried a bit too deep and is too wordy. This goes back to my Specific Comment #1 – this should be elevated to the main story, or it should be relegated to Supplementary with the Paris story being the central theme of the paper. A significant editing of the text, and relegating some of

the details to Supplementary Information, would strengthen this section significantly.

14. Figure 5, K timeline: is the explanation for the big discrepancy between the PILS and the ATOFMS at the peak concentration that the ATOFMS peak area was saturated or mis-calibrated? Either of these could be occurring if the K levels in particles are especially high. This could easily be found by looking at the spectra during this time period.

15. Figure 10, temporal trends of local and regional/continental sources: This figure would be enhanced by the addition of a third pane, showing the total.

16. Figure 11, average composition of local versus regional/continental sources: The language in the caption ("local" and "regional/continental") is not consistent with the labels in the figure ("local" and "transported"). They should be the same.

17. Figure 12, estimated relative mass concentrations of local and regional/continental emissions: This caption should state that this figure is based on ATOFMS data.

18. Supplementary Information, Figure S3: It appears that the ATOFMS data is scaled significantly based on particle number. However, the ATOFMS is sampling through an aerodynamic lens, so this is surprising. Was the instrument not working well during the data collection?

Technical corrections:

1. p. 10348, line 5: would read better as "Particulate matter is known to impact air quality. . ."

2. p. 10348, line 13: remove "upon" to make it read better.

3. p. 10352, line 26 and p. 10353, line 4: The approach taken in this paper is described in the first instance as "an adaptation" and in the second instance as a "different approach" – these should be made consistent.

4. p. 10363, line 6: ". . .are estimated to be composed of 62% EC. . ." needs to specify

whether this is by mass, by number, or something else.

5. p. 10364, lines 14 and 16: these two lines refer, respectively, to EC cores and BC cores. These should be consistent, or the distinction the authors are making should be clarified.

6. p. 10365, lines 19 – 20: the phrase “which is higher than that of the HR-ToF-AMS.” Needs to be added after “. . .ATOFMS (150 nm)” to clarify the meaning.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 10345, 2013.