

Interactive comment on “Laser ablation aerosol particle time-of-flight mass spectrometer (LAAPTOF): Performance, reference spectra and classification of atmospheric samples” by Xiaoli Shen et al.

Anonymous Referee #2

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It is my pleasure to provide a review for this manuscript. The paper describes a comprehensive set of test measurements conducted with a new single particle mass spectrometer, the LAAPTOF. The study provides a good overview over the instrument's core capabilities and performance. As there is limited information on the LAAPTOF in existing literature, this study is a much-needed reference for current and future LAAPTOF users, and for the aerosol mass spectrometry community in general. In terms of content, there is a wealth of information in this paper, with little to add or change; except some of the choices in the field data analysis should be explained better. In terms

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of organization and readability, there is room for improvement: The laboratory measurements presented in the paper were done with several different setups and sample preparation techniques, and, for interpretation, the samples are grouped according to the mixing state of the aerosol. Some of the measurements were size-selective whereas others were not, but for those that weren't, sizes are still reported. Currently, the information which aerosol type was generated how, sampled with what setup, and assigned to which group, is contained in the flow text of section 2.2, and in Table S1 and its footnotes. Both of these are difficult to refer back to quickly, and it is hard for the reader to keep it all straight. The presentation of experimental setups (Figure 1) is also a little confusing, as different laboratory setups are conflated with the field setup (making it look like one experimental setup even though it is not) and simple schematics are combined with a detailed functional sketch of the LAAPTOF instrument.

I recommend final publication after consideration of the following suggestions and comments:

1. The correlation method (a consistent name should be used for this method throughout the paper) should be explained better. Why were positive and negative spectra treated separately (10 and 7, respectively)? Aren't the positive-negative pairs intimately connected, coming from one particle? How were they re-combined to yield 13 particle types (Figure 10)? The caption of Figure S13 contains hints, but is extremely hard to understand. Also, referring to lines 371 – 372 (and the procedure description in the supplement): there are ways to determine the appropriate number of clusters aside from “experience of the operating scientist” – see for example Hinz et al. (1999). Is there a specific reason these were not used?

2. How was the particle size information obtained and handled? According Section 3.2, line 246, a d_m of 800 nm was chosen (presumably only for the samples passing through the DMA). In Table S1, this size only appears for PSL, and other sizes (d_{va}) are given for all samples. One of the footnotes mentions a Gaussian fitting, but this should be described more clearly in the “methods” section.

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3. Organization: There are three experiments here: 1. a rigorous determination of ODE for some aerosol types, 2. a comprehensive collection of reference spectra, 3. a field study with two different data analysis methods. For the sake of readability, the three should be treated separately as much as possible. Here are some suggestions to improve on clarity:

a) Move table S1 into the main manuscript. In the footnotes, make sure that the explanations for the generation methods are easy to spot – perhaps a list, rather than wrapped into a sentence.

b) The three (or four, including sampling from reservoir headspaces?) experimental setups should be defined in Section 2 (separately from the generation methods) and referred back to throughout the paper. Generation methods A, B1, B2, and S should also be defined in the text in Section 2, and referred back to.

c) My suggestion for Figure 1 would be to only show setup A, as there is no information in setups B and C that cannot be described easily in the text. Setup A should then be shown in more detail (flow rates, flow splitting between LAAPTOF inlet and CPC, etc.). This would make it easier for the reader to mentally separate the ODE experiments from the reference spectra experiments. Also, in the Figure, a box or some other designation should be put around the actual LAAPTOF instrument. One could also show setup B in a separate figure, with more detail on the particle generation (cyclones, APC chamber, CAST, etc.), but I believe it is not necessary, as there are descriptions and references in section 2.2.

d) Section 3.1. should be reorganized. Lines 167 – 174 in Section 3.1.1 should be part of Section 2 (Methods). Section 3.1.2 is currently a mix of discussion of ODE measurement results, interpretation of features in the reference spectra, and literature review. While the factors mentioned in lines 223 - 239 may influence ODE, their impact is not put into quantitative relation to the presented measurement results. The discussion would be more appropriately placed in the introduction (some of it, such as the

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40 mW laser setting, in Section 2.1). I would also argue that lines 211 – 221 could be discussed in the context of the reference spectra (3.2); at least, though, there should be several references to Section 3.2.

f) The data analysis methods should be presented in a separate subsection of Section 2. The sections headed “Procedures for ...” and “Seeking...” in the supplement (which could be very useful for other LAAPTOF users), should be proofread (informal expressions), designated (“Procedure 1”, “Procedure 2”, or something similar) and referred to in Section 2.

4. Presentation of Figures: Figure 2: Panel A is very important, but hard to read. Consider putting the y-axis on a linear scale and repeating the x-axis on the top of the graph. Figures 4-7: It is hard for the reader to go back between label shadings in the various panels and description of the shadings in the caption. A legend for the shading in or next to each panel would be much more convenient. What are the dva values in Figure 4 referring to exactly? Figures 4 C (+), and 7: It would be nice to have a few more peak labels (ions, not just m/z values). Figure 9: As much as possible, avoid extra abbreviations, they are not reader-friendly. For example, there is no reason that the axis tick label “SC” could not be “NaCl”. Figure 10: In the current layout, this Figure is too small.

Other comments:

5. lines 82-86: There is another recent paper describing LAAPTOF spectra by Wonaschuetz et al. (2017, Journal of Aerosol Science 113, 242 - 249)

6. line 124: Can provide a short description of what exactly is integrated in “stick integration”?

7. line 130-131, explanation of percentages: Is this quantity ever used in the paper? If not, this explanation could be omitted.

8. line 141: Consider “dissolved/suspended” (PSL spheres don’t dissolve)

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9. line 149: Can you add a very short description of the "different ways"?
10. line 161: How long was the LAAPTOF deployed?
11. line 173: In the samples generated by nebulizing salts, followed by size selection in the DMA, how was the possibility of multiply charged particles handled?
12. line 200: Can you provide a reference?
13. line 203: It seems that it would be easy to provide a quick comparison of Mie scattering efficiencies at the particle sizes/laser wavelength relevant to this measurement to validate the M-shape.
14. line 234: Was this controlled for in some way in the test measurements?
15. line 271: This is confusing. Which ratio is 1?
16. line 285: Can you provide a reference?

Minor corrections/typos:

17. line 296: "can be found".
18. line 305: "are weaker"
19. line 328: "panel A"
20. line 312: "are observed"
21. line 332 – 337: This is a very long sentence. Consider this construction: "In spectra (A), we also observed the following peaks:"
22. line 339: consider: "Most of the ... fragments of soil dust are similar to those of desert dust..."
23. line 348: Start the sentence with "The" and remove the "s" in "soil dust(s)" from Argentina".

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24. lines 454 – 456: "e.g." is in a strange place in the sentence. Perhaps: "...from different fragments: for example, m/z 26"
25. line 442: "In any case" seems to be a needless expression.
26. line 467: consider replacing "among the huge amount of ambient data" by "in the ambient aerosol"

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2017-338, 2017.

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