



Interactive comment on “Development and characterization of a single particle laser ablation mass spectrometer (SPLAM) for organic aerosol studies” by F. Gaie-Levrel et al.

Anonymous Referee #2

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The paper describes the development and characterization of a single particle laser ablation mass spectrometer based on the combination of an aerodynamic lens system, an optical detection part and a linear ToF-MS. The design was made to characterize aerosol particles between about 100 nm and a few micrometer. Using PSL and SOA from the ozonolysis of an aromatic VOCs, the performance of the instruments is described, since a major goal of the experimental setup is the chemical characterization of organic aerosols. Since indeed the characterization of the organic aerosol fraction is still not sufficient to understand origin, fate and impact of these particles in the atmosphere, the motivation to improve the existing measurement techniques is obvious. In

my opinion also the instrumental aspects of the paper are properly described and this part of the paper is based on decent laboratory work that deserves publication in AMT. However, the conclusions of the paper in respect to the chemical/physicochemical discussion of SOA formation is highly questionable. Here ,the discussion has either to be more sophisticated in terms of experimental evidences or these parts should be abandoned from the manuscript.

Specific comments: Discussion on page 4186, 3rd paragraph, however, also part of conclusions and abstract:

The authors conclude from the differences observed in the mass spectra that “real chemical differences of the aerosol particles have been detected”. The line of argumentation to draw such a conclusions is very weak. Firstly, the authors themselves explain why differently sized aerosol particles might experience different energy uptake. Thus differences in the appearance of the individual mass spectra are to be expected. Obviously, mass spectra strongly rely on the energy supply to the individual steps (i.e. desorption/ionization). However, even more important is the question by which mechanism such a chemical differentiation of individual chemical species should proceed. In fact, the current understanding of SOA formation and gas-particle partitioning contain certain mechanisms for “enrichment processes”, i.e. to explain a higher relative contribution of individual species to different particles sizes (e.g. higher contribution of very low volatiles to smaller particles, higher relative contribution of intermediate/higher volatility compounds to larger OA particles). Nevertheless, these processes will always lead to internally mixed aerosol particles (with a slight but continuous change in composition with particles size) and not to externally mixed particles as indicated in the paper. SOA formation is an equilibrium process where constant evaporation/condensation of compounds will take place and therefore a differentiation is actually the complete opposite of what can be expected. The differences in the mass spectra shown in Fig. 10 are much more likely to be a result of a sensible desorption/ionization system, in which small changes (i.e. in respect to particle site and composition) heavily affect

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the ion formation process. Since the system works especially well for aromatic SOA, it is also likely that MALDI processes play an essential role in ion formation (meaning that the analyte composition itself strongly affects the mass spectra, however, not in a quantitative, easily predictable manner). Therefore, I would strongly suggest to take out the discussion on the differences in chemical composition of individual particles. Actually, the authors themselves state several times "... has to be investigated in the future". This should be done before such conclusions – which are not even explained mechanistically – are drawn from preliminary measurements with a newly developed system.

Minor comments:

page 4172 "... as function of their diameter..."

page 4178 "... sensibility..." ?? (better sensitivity ?)

page 4184 "... 4×10^3 to 1×10^3 ..." ????

page 4184 "As for DOP experiment, ..." The whole sentence is very difficult to understand. Please rephrase, perhaps by using several shorter sentences.

Interactive comment on Atmos. Meas. Tech. Discuss., 4, 4165, 2011.

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