

General Comments

The authors present a comparison of the mass spectra obtained from the laser desorption ionisation of atmospherically relevant particle types using ns and fs laser systems in a LAAPTOF single particle mass spectrometer. Although a similar study of has been presented previously by Zawadowicz (2015), this manuscript dealt with a different instrument geometry, and therefore the study presented here has the potential to contribute to the understanding of laser-particle interactions in single-particle mass spectrometry.

However, the authors fail to emphasise the key difference in geometry between the LAAPTOF and the PALMS instrument in the previous study i.e the counter propagate vs orthogonal arrangement of the excimer laser with respect to the particle beam axis. This difference in geometry is likely to influence how the ablation process proceeds and will also influence the interpretation of the spectral characteristics for the reasons stated in the specific comments below.

In addition, the manuscript has some major deficiencies which need to be addressed. With the exception of the ion intensity comparison of the two systems, the key conclusions offered in the abstract in this manuscript are not supported in the data or discussion. Namely:

- There is no supporting data for the conclusions regarding the reproducibility of spectral patterns.
- The methods used for quantifying the fraction of spectra types is not provided.
- The conclusion that 'fs-laser ablation produce spectra with larger ion fragments and ion clusters, as well as clusters with oxygen, which does not render spectra interpretation more simple compared to ns-laser ablation' is only supported by a single hand-picked spectra of each particle type.
- There is no evidence or discussion of the claim in the abstract that quantification remains difficult due to the incomplete ablation of the particle.

Evidence of a rigorous statistical comparison of the mass spectral patterns is required.

Specific comments

P1, L23. Reproducibility is not specifically discussed in this manuscript.

P2, L64. Do you mean a solid particle or fixed target/substrate? Please clarify.

P3, L96. This is only applicable to positive Cl^+ ions. Cl^- ions are readily observed in negative in mode due to high electron affinity.

P3, L199 and P4, L1. Were the beam diameter and focal length/position measured or calculated?

P4, L3. Do you mean the excimer focal position? How do you know ionisation happens 3-4cm after the focal position with a counter propagate geometry? Can you comment on the depth of field of the focusing? Is it more likely that ionisation takes place before the focal position? Does the position of ionisation depend on the absorbing properties and ionisation threshold of the material with a counter propagate geometry?

P4, L129. What is the diameter of the focal point of the fs system?

P4, L141. Can the authors comment on the stability/reproducibility of the focal length, the depth of field and the effect on power density? How is the power density different at F1 than F2? Surely the densities are the same and the focal position has just shifted upstream in the particle beam. Why do the authors vary the focal length? The objectives and conclusions of this operation are not stated in the manuscript.

P5, L171 and throughout the manuscript. How were the mass spectra assigned to a spectra type?

P7, L243. Silicate particles predominantly come from mineral dust not sea spray. SiO_4 is an anion in crystalline orthosilicates and is not an appropriate description of silicate composition.

P7, L256. The work cited used **Silicon** substrate with organic and inorganic solutions and the claims regarding the fragmentation/ clusters were relating the compounds in solution not the silicon. Please be careful not to confuse clusters with fragments.

P10, L361. It should be pointed out that the conclusions are with respect to SPMS with a counter propagate geometry.

P10, L361. Do you mean reproducibility of average ion intensity or spectral pattern?

P10, L369. Cl^- was observed in negative ion spectra. See comment above.

Technical Corrections

P2, L92. GmbH not Gmbh.

P10, L372. Sentence structure/word order.

Figure1 Inset. It is not clear what the position label, defocus label and arrow are referring to.

Figure 2. It is not clear what the wavy line represents.

Figures 3-9. Please state the estimated power density or laser setting at which these spectra were acquired.

Figures 9 and 10. Please state how many particles were averaged and what the error bars represent. The error bars are difficult to see and the quality of the graphics are generally poor.