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Atmospheric Measurement Techniques Discussions

Interactive comment on "Increase of the particle hit rate in laser single particle mass spectrometer by pulse delayed extraction technology" *by* Ying Chen et al.

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Dear Reviewer! Thanks a lot for your notes, comments and questions. We hope that they improve our understanding of the problems associated with processes in the single particle laser mass spectrometer. Please, find our answers/comments on your notes below:

1) Actually we don't observe a significant difference between mass spectral peaks in DE mode and DC mode, except their intensity and resolution. In our article we address other issues. i.e. the hit rate and the intensity of peaks in mass spectra. We have prepared the mass spectra acquired in DC and DE modes for comparison, for those

who are interested in this matter, and we will put them in the supplementary material. The plasma cloud after laser ionization is composed of ions, electrons and neutrals, and it is generally neutral. The plasma expands and disintegrates over time, and, at the same time, ion-neutral reactions and recombination processes between ions and electrons take place, so, the system is quite complicate. One can suppose, that if the effect of a strong electric field on the expansion of ions and electrons beyond 100 ns was significant, the signal in the case of DC extraction would be higher. On the contrary, we observe some increase in positive and negative ions signal in case of DE in comparison to the DC extraction mode. It can be assumed that the interaction of ions and neutrals makes a larger contribution than the recombination of ions with electrons. Presumably negative ions are formed by the capture of electrons by neutrals, while positive ions can be transformed into some cluster ions via interaction with neutrals. Thus, we can assume that the 100 ns delay can increase the interaction of charged particles with neutrals. It can also be assumed that the degree of ionization in the cloud is low. We upload three figures illustrating the comparison of mass spectra in DE and DC modes.

2) Thanks for the note about the language. We have corrected a number of errors by proof-reading.

3) We guess, the commercial X-ray neutralizer has two disadvantages, one is the price, it is more than 20,000 dollars if you want to buy a new one. The other is the X-ray lamp life time, which is only 8000 hours typically, and you need to change it frequently

4) About Nd:YAG – we agree, and we'll change the sentence in the article ("many instruments" instead of "most instruments")

5) Corrected to "hit rate"

6) We agree, it is better the section 3.1 should be transformed to 2.1. "Key factors affecting the efficiency" in the Instruments and Methods section.

7) The scatter can be related to the stochastic change in the surface charge of the particles. By the way, we have found a mistake in the Figure 4. Actually, it shows the hit rate per 1 second. In the manuscript, we described the whole time scale for this Figure as 30 minutes but actually it is 30 seconds. Because the hit rate was counted in every second, the particle detection rate is less than 10/second, so the result shows scattering according to the Poisson low. So we have change the minute into second in the Figure 4.

8) Unfortunately, it is hard to reproduce the experiment with the glass slide, as the instrument which was used for it is under construction now. But we don't suppose it will show any critical results, as the extraction pulse width is short (5 us), and the path of the particle moving with the velocity 100 m/s will be 0.5 mm. Hence, the effect of deflection will be $\sim$ two orders of magnitude lower than in case of the DE. Hence, the spot size on the glass slide is supposed to be the same as that obtained without the electrical field (Fig.5), and we'll not get any extra information. Note please, that in case of DE operation, this HV pulse does not affect on the trajectory of the particular sized particle, as it is applied after it is ionized. On the other hand, the HV pulse is unlikely to affect other particles, since the average particle counting frequency is less than 100 Hz. Actually, the used 266 nm laser has a max repetition rate of 100 Hz, that is why the maximal counting frequency is 100 Hz.

9) We would add some more details about a possible realization to pre-select particles by their mass/charge ratio in front of the ion source: " It could be done, for example, by using two pairs of deflecting plates before the ion source. By choosing the deflection voltage, only particles with a specific mass/charge ratio will pass this double deflector and can be ionized in the ion source" It is just an idea which would need a significant additional work for it realization

Please also note the supplement to this comment:
https://www.atmos-meas-tech-discuss.net/amt-2019-163/amt-2019-163-AC1-

supplement.pdf

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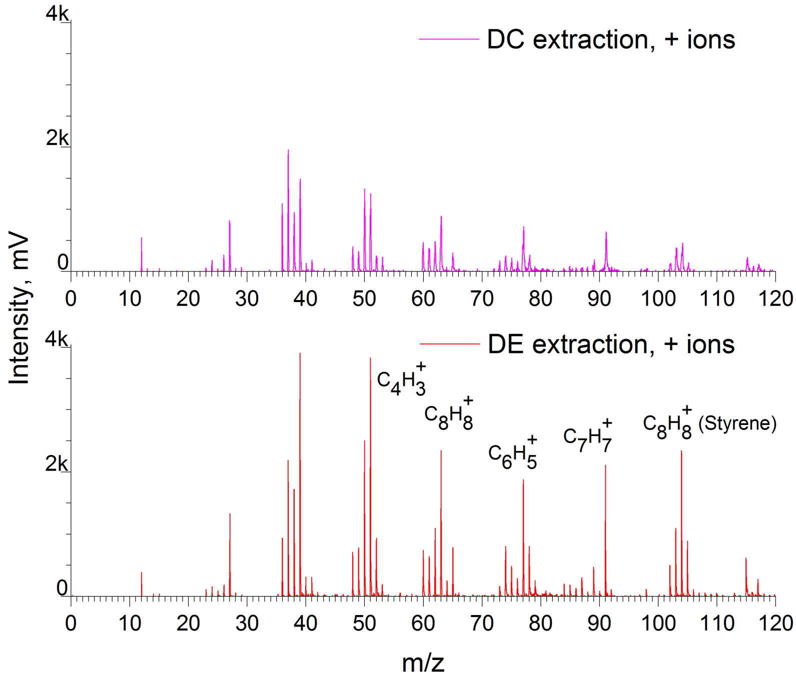

Fig. 1. Comparison of positive ions mass spectra obtained from PSL particles using SPAMS instrument with DC and DE extraction modes, in the mass range 0<m/z<120

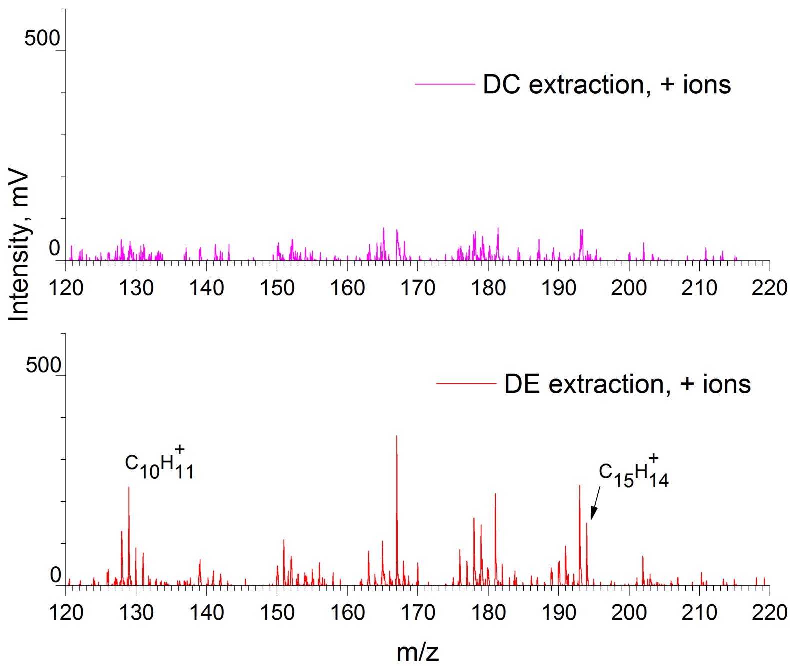

Fig. 2. Comparison of positive ions mass spectra obtained from PSL particles using SPAMS instrument with DC and DE extraction modes, in the mass range 120<m/z<220

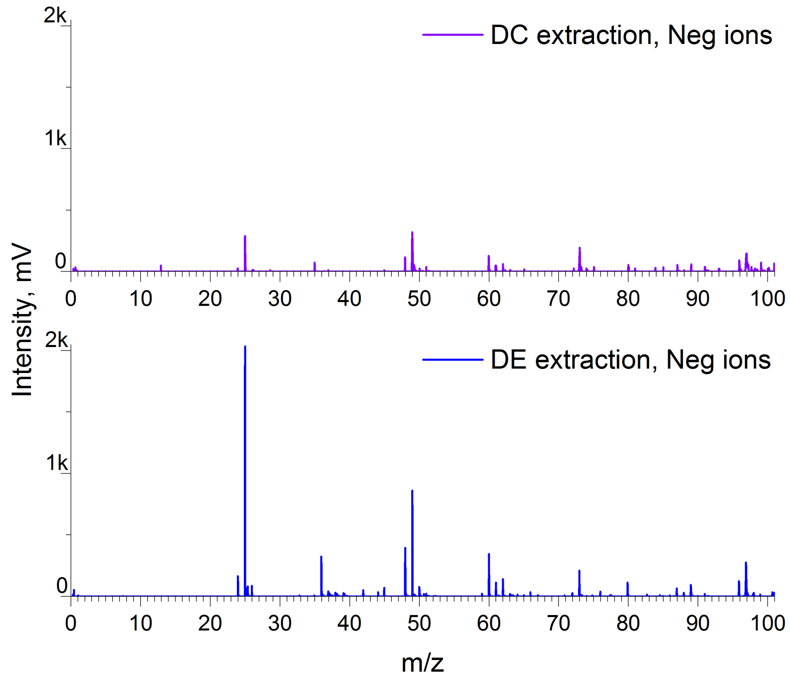

Fig. 3. Comparison of negative ions mass spectra obtained from PSL particles using SPAMS instrument with DC and DE extraction modes, in the mass range 0<m/z<120

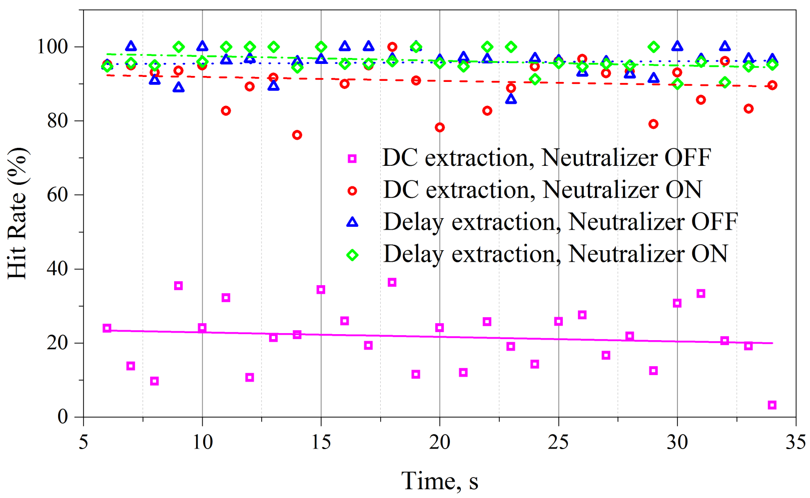

Fig. 4. Hit rate count for four cases: DC extraction& no neutralizer, DC extraction& neutralizer ON, Delay extraction & no neutralizer, and Delay extraction & neutralizer ON