Responds to the Referees

Referee #2

Review of Du et al., The design and characterization of a high-performance single-particle aerosol mass spectrometer (HP-SPAMS)

This manuscript is, as the title implies, a technical description of a single particle mass spectrometer. This is a commercial instrument, but the description is primarily technical and does not feel too much like an advertisement. It falls reasonably into the type of paper that describes the performance of a commercial instrument.

The technical specifications are in some respects quite impressive. This instrument is, for example, significantly better than the ATOFMS sold some years ago in the US by TSI, Inc. The writing is good and the references are balanced. The manuscript should be publishable with minor revisions.

1) One general change that would benefit the manuscript is to do fewer comparisons to the previous SPAMS instrument developed by the same group and emphasize absolute performance more. The authors are justifiably proud of their improvements, but the eventual paper should be presented for a general audience of all users of single particle mass spectrometers, not just those who own an older instrument developed/sold by the same authors. An example of something to de-emphasize or delete: “the number of lead-containing particles is 145 times higher than that detected by SPAMS” (abstract). An example of something to emphasize more: the comparison in Figure 4 of the detection efficiency of various kinds of particles, presumable due to spherical or non-spherical shapes. This is a very useful comparison between types of particles. These are just examples: there are probably dozens of places in the manuscript where the comparisons to the older SPAMS could be reduced. A few are OK, but only a few.

Answer: Thank you for your suggestion. We have also noticed this problem and have added comparative descriptions with ATOFMS, LAAPTOF, as well as reduced some comparative descriptions with older instruments, in order to better demonstrate the absolute performance of the instruments. In detail, we have made the following changes to the manuscript:

(1) Revise the abstract section "For the analysis of individual particles, the HP-SPAMS's improved resolution helps distinguish between most organic fragment ions and metal ions and facilitates the analysis of complex aerosol particles" to " For the analysis of individual particles, HP-SPAMS achieves an average mass spectral resolution of 2500 at m/z 208, which helps distinguish between most organic fragment ions and metal ions and facilitates the analysis of complex aerosol particles."

Delete " for example, HP-SPAMS can completely differentiate the isotopes of lead elements and the number of lead-containing particles is 145 times higher than that detected by SPAMS. " In the abstract.

(2) Delete " The repetition frequency of the laser was increased 5 times compared to 20 Hz in SPAMS, which could reduce the "busy time" of the laser and improve the temporal resolution. " in Section 2.2.

(3) Add "This is due to the fact that for irregular particles there is a force perpendicular to the

direction of gas motion, while for spherical particles this force is zero, resulting in greater dispersion of non-spherical particles after passing through the lens system. (Liu et al., 1995)"in section 3.1.

(4) Delete"… and this correlation was significantly higher than that of SPAMS results." in section 3.1.

(5) Delete" … indicating that HP-SPAMS improved the total number of detected particles by 47.8 times compared to SPAMS."

(6) Added description of resolution compared to other instruments (LAAPTOF and A-ATOFMS). Change "It can also be seen from Figure 8 that the resolution of HP-SPAMS using exponential pulse delay extraction is, on average, 2-3 times better than that of the DC-extraction SPAMS, and the average resolution is up to 2500 (Full Width at Half Maximum) at m/z 208. This resolution is better than that of instruments of the same type such as LAAPTOF (600) and A-ATOFMS (1072)." to" It can also be seen from Figure 8 that the resolution of HP-SPAMS using exponential pulse delay extraction is, on average, 2-3 times better than that of the DC-extraction SPAMS, and the average resolution is up to 2500 (Full Width at Half Maximum) at m/z 208. This resolution is better than that of instruments of the same type such as LAAPTOF (600) and A-ATOFMS (1072)" in Section 3.2.

(7) Delete" This meant a difference of about 145 times between each other." and" In addition, the 7955 lead-containing particles detected by HP-SPAMS could be further analyzed in time series for changes in the concentration, which is not possible using SPAMS at low concentrations. Thus, the improved instrument performance of HP-SPAMS relative to SPAMS could better characterize particles, especially in low-concentration environments" in Section 3.2.

2) One important technical detail that is missing is a list of the spot sizes of the detection and ionization lasers. One cannot interpret the performance of the aerodynamic lens without knowing how big of a target is provided by the detection lasers beams. And one needs the ionization laser spot size to know the fluence available to ionize particles.

Answer: Thank you for your suggestion, we have added the spot size in Section 2.1 and Section 2.2. " Two beams of continuous-wave Nd:YAG (532 nm) laser (500 mW, LaserWave, LWGL532 nm-500 mW) spaced orthogonally 6 cm apart were focused by a plano-convex lens (PLCX-25.4-51.5-C-532, CVI Laser Optics) into a 300 μm-sized spot at the first focus of the ellipsoidal mirror, respectively. Scattered light is produced as the particles pass through the laser beam and is focused by the ellipsoidal mirror to the photomultiplier tube (PMT, H10721-110, Hamamatsu) for detection. " and " HP-SPAMS uses a diode-pumped Nd: YAG 266 nm (100 Hz, 9 ns pulse width, Centurion Plus, Quantel) that is focused by a UV fused silica plano-convex lens (f=175 mm, SPX026, Newport Corporation) into a 300 μm square uniform spot at the center of the ion source. "

3) I found the discussion of the aerodynamic concentrator incorporated into the aerodynamic focusing inlet to be confusing. Maybe because it is incorporated into the inlet, it was never clear to me what the baseline was for its performance. In the abstract it says that it "enables concentration… but a factor of 3-5 times". Compared to what? And then in section 3.1 I was not clear in the discussion of detection efficiency what the maximum detection efficiency should be. Should it be 100%? Or perhaps the maximum should be 300 to 500%, because the best possible

performance would be 100%, but the concentrator would then multiply that by 3 to 5? I think a clear definition of how the baseline is defined would solve these. By the way, incorporating the aerosol concentrator into the inlet instead of having a separate concentrator upstream looks like a clever idea with several advantages.

Answer: Thank you very much for your suggestion, we have made the following changes to this:

（1） Change "enables concentration… but a factor of 3-5 times" to "The combination of an aerodynamic particle concentrator (APC) system and a wide range of aerodynamic lenses (ADLs) enables the concentration of particles in the 100-5000 nm range. Using APC increases the instrument inlet flow by a factor of 3-5".

（2） We have changed "detection efficiency" to "scattering efficiency". The scattering efficiency of particle is defined as the ratio of the response pulse count of the photomultiplier tube to the total number of injected particles, in which the total number of injected particles can be obtained from the particle concentration (from CPC) and the injection flow rate. Therefore, the scattering efficiency is the result obtained after normalizing the inlet flow and the maximum scattering efficiency value is 100%.

4) It would be interesting to see a few spectra on a logarithmic scale, perhaps in supplemental material, to see the dynamic range from the high range/low range digitization and whether or not that induces any artifacts.

Answer: Thanks to your suggestion, we have shown the example data in logarithmic scale in the supplemental material Figure S6. It is worth noting that since there is a zero value in the original intensity, therefore, the intensity is overall biased by 0. 1 mV when plotting this graph. From Fig. S6(a) and Fig. S6(b), it can be seen that the maximum data acquisition range can reach 20V (K+), and the smallest signal can be acquired to 0.004V. Fig. S6(c) shows that this is a standard EC containing particle, and from Fig. S(d), which is displayed in logarithmic scale, it can be seen that this EC particle still contains a very small amount of OC.

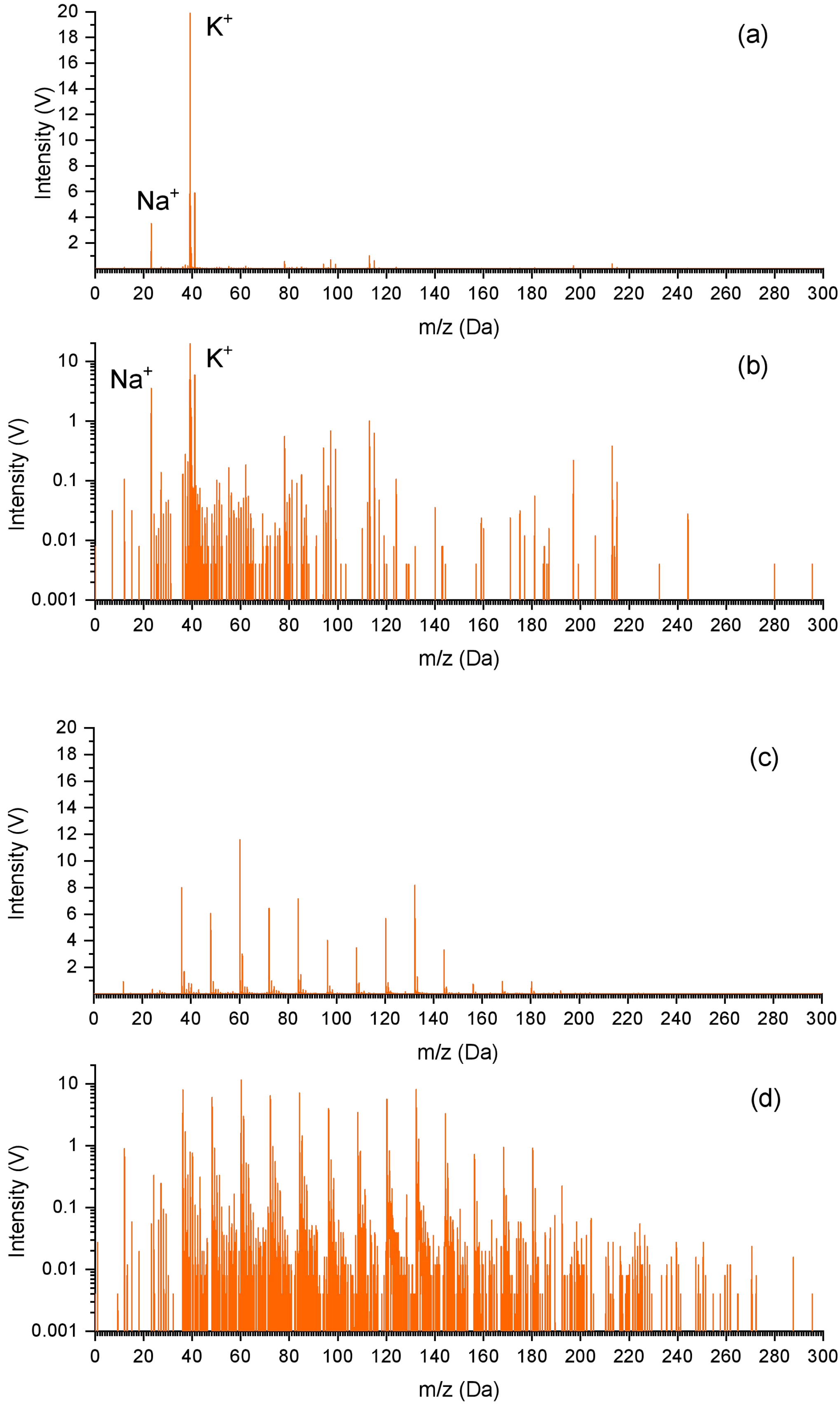

Figure S6: Example particles of mass spectra in linear scale and logarithmic scale (a)&b Na-K containing particles; (c)&(d) EC containing particle.

5) Near line 104, the pressure would be helpful as well as the orifice diameters. I think the symbol/line labels in Figure 5 may be incorrect.

Answer: Thank you for your suggestions:

(1) We have added "The aerodynamic inlet pressure is in the range of 2.0 ± 0.2 Torr and the exit nozzle size is 3 mm." in Section 2.1.

(2) we have exchanged the "Hit rate of HP-SPAMS (PSLs)" with "Hit rate of SPAMS(PSLs)" in legend.