Replies to the comments by Anonymous Referee #2:

First of all, we would like to thank Referee #2 for reviewing our manuscript and for her/his helpful comments to improve it. In the following we will comment on the individual points. The referee's comments are shown in black and our answers in blue. The referee's comments refer to the version of the manuscript submitted for review and our answers refer to the revised version of the manuscript.

Comments and Replies:

General comments:

This is a well-written and extremely thorough paper about improvements to the ALABAMA laser ablation aerosol mass spectrometer that greatly improve the detectable size range and the detection efficiency. Bravo

Specific minor comments:

1) The CPI is discussed first, so the CPI should be Figure 3 and the air diffusor and lens should be Figure 4.

   We agree that it is better to swap Fig. 3 and Fig. 4. It is now changed.

2) For the angle of the air diffuser, the text says 5.6 degrees, but the figure says 6 degrees. Maybe use the same value in both places.

   Thank you very much for the hint, we use now uniformly six degrees as indication for the opening angle.

   Changed:
   Sect. 3.1.1, pg:7, line: 152

3) Pg 8, lines 165-6: What does the acronym ODD stand for? Outer diameter diffuser, maybe? I think it would make more sense to mention the ID at the end of the diffuser which looks like 31.2 mm.

   ODD is really not the best choice in this case. ODD is actually the inner diameter of the air diffusor. Instead of ODD we now use the term maximum inner diameter of the air diffusor ($ID_{Di(max)}$) in the paper. $ID_{Di(max)}$ is 39 mm at its maximum extension directly before the 45° tapper.

   Changed:
   Sect. 3.1.1, pg:8, line: 167-169

4) Pg 9, line 182-186: What you are calling outer diameter lens (ODL) is not an outer diameter. It is the inner diameter of the lens. I think it would be better to use subscripts to distinguish between the ID of the lens and the ID of the orifice, rather than calling one an OD.

Yes indeed, this could lead to confusion for the reader, we now use subscripts for a clearer distinction between the inner diameter of the aerodynamic lens (ID_{Le}) and inner diameter of the orifices (ID_{Or}) or inner diameter of the first orifice ($ID_{Or(1)}$).
Changed:
Sect. 3.1.1, pg:8, line: 168-169
Sect. 3.1.2, pg:8, line: 189-193

5) Figure 7: I'm confused by the timing. The DIE trigger is shown 50 ns after the Q-switch out. Then it takes 50 + 5 ns for the voltages to turn on. Wouldn't the voltages be on 35 ns after the laser pulse, not 70 ns as indicated in the figure? Shouldn't the expected DIE (line 272) be 35 ns? What DIE did you determine from the high voltage signals? The 70 ns time is mentioned again on page 35, line 725.
We understand that our wording suggested that the HTS specific delay times (50 ns + 5 ns) are directly related to the time to switch on the ion extraction voltages. However, it must be mentioned that the box with the HTS´s inside is installed between the mass spectrometer housing and the high voltage modules and is connected to the mass spectrometer by high voltage cables, which means that a further delay of the switch-on times for the ion extraction voltages can be expected. The DIE was measured at the connections of the high voltage cables on the mass spectrometer housing by observing the triggered high voltage signals using an oscilloscope. At this point, a time delay of 140 ns ± 10 ns between the Q-Switch out signal and the high voltage signals was measured. Taking into account the time period of 70 ns between Q-Switch out and the laser pulse as determined by Brands et al. (2009), a DIE of about 70 ns can be expected for the measurements performed in this study.

The determination of DIE is now described in more detail in Sect. 3.2.2:
pg. 12, line 263-265
pg. 13, line 279-282

6) You should refer to Figure 10 in Section 4.1.3 about the INP measurements. There is currently no reference to Figure 10 in the text.

Thanks for the hint, the reference to Fig. 10 was added in Sect. 4.1.3 (pg. 15, line 329).

7) Pg 17, lines 353 – 361: Is this section about PSLs for which the size is known? If not, it is not clear how summing over large size ranges in the OPC will give you the correct concentration to compare with the number detected. Please clarify.

We agree that Sect. 4.2.1 "Definition of particle detection efficiency" was a bit misleading. Also for Referee #1 the background of the size selection in the OPC was not clearly visible from the text. Basically the summation over several size channels in the OPC was only performed for PSL particles with known size. First of all, a selection of size channels is not necessary as long as a DMA was used. With increasing size of the supermicron-particles it becomes more difficult to use our DMA in a suitable way. For monodisperse particles the applicable size range of the DMA can be extended, but not to all particle sizes we used. Therefore the large particle sizes were measured without DMA. Without DMA, however, we have the effect that in the OPC a second size mode was observed in the smallest size channels. We attribute this second mode to small droplets or substances dissolved in the PSL suspension. To avoid an influence of these unwanted small particles on our measurements, we have removed this mode by selecting the size channels in the OPC and corrected the particle concentration measured with the OPC accordingly. The use of the size selection in the OPC is now described in more detail in Sect. 4.2.1 (pg.17, line: 357-364).

8) Figure 12: I would add a legend with the results from the fit for sigma_p and r_DL.

The legend was added in Fig. 12.

9) Pg 21, lines 455 –6: I don't understand the source of this statement. The results in Kollner (2020) show DE at 2.5 hPa for lab experiments and DE at 3.2 hPa for field measurements which are actually higher, though the size is not indicated. Pg 22, lines 457-8: Please summarize the reasons for the much lower DE in Kollner (2020) vs Brands et al. (2011) for 200 to 600 nm particles. A PhD Thesis is not the easiest source to find this information.

Pg21: In Köllner (2020), Figure A.4 shows the experimental results of the detection efficiency against the lens pressure (laboratory measurements with PSL particles). It is shown that especially the measured particle sizes 1680 nm and 1890 nm show a decreasing detection efficiency for a lens pressure deviating from 2.5 hPa. The corresponding reference to Figure A.4 in Köllner (2020) was added in Sect. 4.3.1, pg 22, line 465.

Pg22: A possible cause for the lower measured detection efficiencies in Köllner (2020) compared to those of Brands et al. (2011) is clogging of the O-ring or non-optimal lens alignment. Furthermore, Köllner (2020) points out that possibly also the device modifications made to the ALABAMA in the meantime may have led to a reduction of the detection efficiency in the size range between 200 and 600 nm. For example, the results shown in Brands et al. (2011) were obtained using a diode-pumped continuous-wave laser with a wavelength of
$\lambda = 532$ nm. However, the exact cause of the different results could not be found out.
The summary of the points was included in Sect. 4.3.1, pg 22-23, line 466-475.

10) Figure 13 caption: I think you mean the detection efficiency is "relative to", not "related to"

Yes, we changed to "relative to".

11) Figure 14: What is the physical distance between the exit of the lens and the first detection laser? A particle beam width < 20 um seems really small. Is this consistent with your CFD calculations?

The particle beam widths shown in Fig.14 are the widths related to one sigma according to Eq. 4. The particle beam widths calculated with the aerosol lens calculator (Fig.S2) were originally given as widths in diameter and comprising 90% of all particles (Wang and McMurry, 2006). In order to compare the particle beam widths calculated with the aerosol lens calculator with our experimentally determined widths, the theoretical values are now converted into widths of one sigma. For the conversion of the theoretical values it was assumed that the particle beam diameters originally resulting from the aerosol lens calculator describe a 2-dimensional circular Gaussian distribution. A comparison of the experimentally determined particle beam widths with the values of the aerosol lens calculator shown in the Supplement Fig.S2 now shows a good agreement. In Fig.S2 an additional column with the experimentally determined values is now added and described in the text of Sect. S2. Furthermore, in the captions of Fig.14 and Fig.16 there is now the addition that the particle beam widths presented there refer to one sigma. In the caption of Fig.21, the particle beam width is given as diameter in four sigma.
Further additions:
Sect. 4.2.2, pg 19, line: 408
Sect. 4.2.2, Fig.12 in the legend
Sect. 4.2.2, pg 20, line: 419
Sect. 4.2.2, pg 21, line: 427
Sect. 4.3.3, pg 26, line: 552
Sect. 6, pg 40, line: 833

12) Figure 17: I understand why you ordered the x-axis in Figure 17b the way you did, but it makes it very hard to compare with 17a. I would use the same order for the x-axis in both 17a and 17b. You can draw the same conclusions in the text.

Ok, thanks for the hint, Fig. 17 was changed according to your suggestion.

13) pg 31, lines 646-7. What does the statement that “a voltage of 1100 was assumed” mean? Was the voltage not actually set to 1100V? Or you think the effective voltage at the electrode is different than the setting?

Actually, the voltage was set to 1100 V. However, small drifts in the high voltage supplies may lead to a slightly different real value. We changed to “A calculation of the particle deflection within the electric field after Eq. 1, using 1100 V for both electrodes, resulted in a shift of roughly 90 µm for 300 nm PSL particles” (see Sect. 4.5.3, pg 32, line 673-674).

14) Figure 19: Why are the curves for 308 nm, DIE(on) asymmetric?

The asymmetric curves of the fits shown in Fig. 19 result from the assumptions made in Sect. 4.4, which may lead to a slight distortion of the edges of the hit rate distributions. Furthermore, the measurements showed a slight offset between the beam center position of the second detection laser and the beam center position of the ablation laser, which also contributes to an asymmetry. In Sect. 4.5.3, which belongs to Fig. 19, an additional reference to the assumptions of Sect. 4.4 and the existing offset was added.

New (Sect. 4.5.3, pg 33, line 684-687):" *The asymmetric curves for 308 nm PSL particles using the DIE(on) setup result from the assumptions made in Sect. 4.4, which lead to a slight distortion of the edges of the hit rate distributions. Furthermore, the measurements revealed a slight offset between the beam center position of the second detection laser and the beam center position of the ablation laser, which also contributes to an asymmetry.*"

15) Pg 35, lines 703-4. It's not clear what "the offset between the two curves" is referring to. The difference between cation and anion curves, which anticorrelate? Or the difference between the DIE(on) and DIE(off+) curves which are slightly offset.

This refers to the offset between the DIE(on) and DIE(off+) curve. It is now described in Sect. 4.5.5, pg 35, line: 733-734 .

Supplement

16) Figures in the Supplement should be called S1, S2, etc. Same with the equations. And sections. This will make it less confusing when you refer to something in the main paper.

We agree. All sections, figures and equations in the supplement have been renamed accordingly.

17) Figure S2. The calculated particle beam diameters are orders of magnitude larger than the measured particle beam widths in Figure 14 in the paper. Do you have an explanation?

Motivated by your comment 11), there is now an additional column in the table (Fig. S2) with the experimentally determined particle beam widths, given in one sigma. All particle beam widths given in this study refer to the one sigma width of the particle beam unless otherwise stated. Comparisons of the experimentally determined particle beam widths with those calculated with the aerosol lens calculator are now directly comparable and show good agreement. In the caption of Fig. S2 the used width definition is now given, and further explanations in Sect. S2 are added (pg. 2-3, line: 28-37).

18) Figure S8 and S9: It is hard to compare these figures to Figure 21 in the main paper because the panels are in a different order. Please put the cations in Figure S8 and the anions in Figure S9. Also put the sum in the top panel and the number in the bottom panel.

Fig. S8 and Fig. S9 have been changed according to your suggestion. The text in Sect. S12 was adapted to the changed arrangement of Fig. S8 and Fig. S9 (see pg. 10-11, line: 177-187).

19) Pg 13, lines 203-4: Superscripts with m/z's seem like an odd notation for these ions. Normally, the superscript would be charge. Or, in the case of isotopes, the superscript would precede the symbol. Maybe put the sign and the m/z in parentheses instead.

We now use the label according to your suggestion with sign and m/z in parentheses (see Sect. S14, pg. 13, line: 215-216).

Technical corrections:

Pg 1, line 14: insert "than" before "500"
Added

Pg 1, line 19: "ALABMA" misspelled
Corrected

Pg 1, line 19-20: use "an" before "up" in two places
Done

Pg 2, line 41: use "an" before SPMS
Done

Pg 5, line 96: "a sufficiently light scattering signal" is missing the word "large"
Thanks, it was added

Pg 10, Figure 5 caption: should be "Schematic" not "Schematical"
Thanks, it is corrected

Pg 16, line 337: Please group the minus sign with the temperature value.
Done

Pg 21, line438: should be "a detectable" not "an detectable"
Changed

Pg 21, line 454: word missing between "this" and "not"
"is" was added

Figure 13 caption: I think you mean the detection efficiency is "relative to", not "related to"
Changed, see reply to comment 10

Pg 24, line 502: word missing between "likely" and "to"

Thanks, “due” was added

Pg 30, line 624: should be “a reduced” not “an reduced”
Changed

Pg 31, line 655: “new installed DIE result” should be “newly installed DIE results”
Done

Pg 33, line676: “mass spectra” should be “mass spectrum”
Corrected in Sect. 4.5.5, pg 33 line: 706, pg 34, line: 716, pg 35, line: 724

Figure 21 caption: “Dependence of mass spectral information on particle beam position. . .” is better English than “Mass spectral information in dependence of. . .”
Modified according to recommendation

Pg 34, line 692: “switched” instead of “dwitched” and “a reduced” instead of “an reduced”
Done

Pg 34, line 694: “significantly” instead of “significant”
Changed

Pg 40, line 838: Presumably, something should go here besides “TEXT”
Removed

Supplement

Pg 1, line 2: Presumably, something should go here besides “TEXT”
Removed

Pg 9, line 140: delete “can”
Removed

Pg 10, 161: “cumulated” is not commonly used in English. Maybe used “summed” instead.
Changed

Pg 13, line 195: “randomly” instead of “random”
Done

Pg 13, line 196: “spectrum” instead of “spectra”
Changed