Review of "Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques"

General comments

In this study, the authors present a novel mass spectrometer ERICA (ERC Instrument for Chemical composition of Aerosols), which combines two ionization techniques, i.e., laser ablation and the flash vaporization with electron impact ionization. Given the complementary strengths of the techniques, ERICA allows for in-situ and real time measurements of size and chemical composition of the aerosol particles, provides qualitatively information of almost all the particulate components and the quantitative information of the non-refractory components. The authors have done comprehensive laboratory and ground-based field measurements to characterise this instrument and tried to demonstrate its improved chemical characterization capability. As shown in the manuscript, such a hybrid instrument with compact and light-weight design is good for aircraft measurement. This study would be quite useful for atmospheric science research, especially in the mass spectrometry community. However, the presentation is not very well structured and not clear enough in current version, which needs to be improved. In addition, the authors should do more literature research on single particle mass spectrometry (SPMS) and aerosol mass spectrometer (AMS) to make correct statements. Therefore, I recommend it to be published after major revisions.

Major Comments:

1. Several confusion/wrong statements on these two complementary techniques need to be revised.

1) Please note that the SPMS uses laser for desorption and ionization, while AMS uses vaporization followed by electron impact ionization. "vaporized" (P3L22) needs to change to "desorbed". Please distinguish these two ionization techniques in a clearer way throughout the manuscript. In addition, SPMS and AMS use different way to determine particle size d_{va}. The authors miscited some references in section 2.1, Page (P) 3 Line (L) 20. Please correct.

2) Limited repetition rate of ablation laser is only one of the reasons for the low detections, but not the main one. There are several other influencing factors on the low detection efficiency and detailed discussions on such topic. Please refer to and cite the corresponding SPMS publications, e.g., from the most related instrument ALABAMA, and revise accordingly, e.g., P2 L30-32 & P6 L13-14.

3) The authors should be very cautious when compare ERICA-LAMS with ERICA-AMS.

For example, in section 4 the authors compare the number fraction of sulfate containing particle with the mass fraction of sulfate and discuss the difference (P21 L23-30 & Fig. 17). However, the reasons for the difference are not convincing. Please reconsider the explanations.

In Fig. 17 the sum of the number fractions of meteoric and sulfate containing particles are larger than 1 at higher altitude. This is confusing and needs more explanation. Apparently, the methods to obtain these two particle types are not the same: the meteoric type is based on k-means clustering, while the sulfate containing particle type is very likely based on the maker peaks' intensities (please describe). Consider modifying Fig. 17 or add detailed descriptions in the figure caption.

The discussion on total mass concentration (measured by ERICA-AMS) and EC-containing particles (ERICA-LAMS) cannot come to the conclusion that "the sampled aerosol is well mixed within the particle boundary layer and in the free troposphere", also cannot show the complementary strength. Please reshape the statements.

2. Presentation quality needs to be improved.

1) Citation formats: Please pay attention to the formats between Author et al. (year) and (Author 1 et al., year; Author 2 et al., year; Author 3 et al., year; ...) and use them properly. Please revise the citation format throughout the manuscript and keep consistency.

E.g., P1 L35 "(See for example Fuzzi et al. (2015))" should be changed to (Fuzzi et al., 2015); P2 L10: Change "(e.g., in Froyd et al. (2019))" to (Froyd et al., 2019).

2) Section 2 Instrument description: I would suggest refining the descriptions of ERICA-LAMS and EIRCA-AMS modules, since most of them have been well described in SMPS and AMS papers. Please emphasize the difference, e.g., the shutter unit (SU) needs more descriptions. Consider combining 2.3 and 2.4. Pleas simplify the headers.

3) Section 3 Instrument characterization: This section is very important and with comprehensive information, but the key points are buried. It would be very hard for the readers to follow since the LAMS and AMS information is mixed in an unclear way. I would highly suggest rewriting this section by considering the following points.

Please separate the characterization of LAMS and AMS first and then discuss complementary features, and also revise the corresponding figures. Besides, move some detailed descriptions, regarding e.g., calibration (e.g., particle size cal in LAMS; AMS IE and RIE cal), instrument alignment (e.g., ADL position scan), in the supporting information, since they are very well described in other publications or user's manual. An example of restructuring: 3.1 Particle beam characterization; 3.2 ERICA-LAMS characterization (Laser beam; Optical detection efficiency; Hit rate; LAMS mass spectra); 3.3 EIRCA-AMS characterization (Collection efficiency; Detection limit; AMS mass spectra; Mass concentration), and 3.4 Overall performance comparison (sensitivity, size, spectra, etc).

Please keep the terminology same as the ones commonly used in SPMS and AMS communities, respectively, e.g., use “hit rate” instead of “ablation efficiency”; use “collection efficiency” instead of “detection efficiency”.

4) Section 4: The authors only describe the sizes of EC-containing particles in the last paragraph in this section, which is not strong enough. Please give the information of chemical resolved size distributions obtained by both ERICA-LAMS and ERICA-AMS, and add more discussion accordingly.

5) Figures:

Consider moving some to the SI, e.g., Fig. 3 and 4, and combing some, e.g., Fig 7 and 8, Fig 8 and 9.

In Fig. 5, 6, and 7, the solid squares, diamonds and circles with the same colour are not easy to distinguish. Please modify them in a clearer way.

Error bars: Since this sentence “The error bars are in some cases smaller than the symbol” is shown in most of the figure captions (Fig. 3, 5, 6, 7, 8, 9, 10, 11, 16, 17, and 18), I suggest that put the corresponding values in SI. The following question is that in the laboratory how many repeated experiments have been done to generate one data point?

Mass spectra: The x and y scales, as well as the axis labels, are inconsistent among all the spectra. E.g., for y axis, in Fig. 12 it is “signal intensity in a.u.” in linear mode, while in Fig. 15 and 18, it is “ion peak area in mV. sample” in log mode. Please try to keep consistency. Please normalize the spectra to the total ion intensity and keep the same scales (both x and y) for consistency.

3. The advantages of this hybrid instrument are not very well demonstrated, not only due to the poor manuscript structure, but also lacking discussion on complementary results. Please try to improve. Besides, in addition to the compact size, are there any other big advantages of using such a hybrid instrument compared to deploying SPMS and AMS instruments in parallel? Please state the differences.

4. For the current configuration of the LAMS module, it is hard to believe that PSL particles with smaller size < 200 nm can be detected. Several statements on the PSL 80 nm and 108 nm with the corresponding data shown in the Figures 3, 5, 6, 7, 9, 10, and 11 are not valid. Please consider modifying or removing accordingly.

Minor Comments:

P1L23-25: Please change 3170 nm to 3.17 µm or change 3.5 µm to 3500 nm to keep consistency and revise throughout the manuscript.

P2 L10: Change "(e.g., in Froyd et al. (2019))" to (Froyd et al., 2019; Author 2 et al., year...), and please add more corresponding references. Lots of quantification work has been done by using ATOFMS and other reference instruments like OPC, AMS, and so on.

P3L25: Please cite the corresponding publications.

P3L32: Please use 3.17 µm to keep consistency.

P3L32-33: It would be more helpful to mention the transmission efficiency of the ADL instead.

P3L36-38: Please describe the difference between shutter and chopper.

P4L25: The full name of LAAPTOF should be "Laser Ablation Aerosol Particle Time-Of-Fight mass spectrometer" rather than "…spectrometry".

P4L26: Please change the dot in "$5 \cdot 10^2$ cm^3 s^{-1} " to multiplication symbol "5×10^2 cm^3 s^{-1} " and revise the others throughout the manuscript.

P15L28: Fig 10 should be Fig 11.

P16L11: Please give the reason for choosing these peaks.

P16L34: Please assign the peak at m/z 228.

P19L27: The left half of the bracket is missing.

P20L28-30: The reader would expect the following focus on meteoric and EC containing types rather than particulate sulfate, which is a compound. Please reshape this sentence to make the transition smoothly.

P20L31: Incorrect statement. Please revise.

P20L35: Please clarify that when only considering the non-refractory species, the sulfate mass fraction is 1 at 20 km.

P21L32-35: Please add references to support the assumption.

Fig.1: Please add TMP 1 to 4 in the figure or point out their positions. Please add the distances between LD1, LD2, ablation spot, shutter unit, vaporizer, etc.

Fig.2: Consider rescale some sizes/distances. E.g., the distance between convex lens and the quartz window (10 mm) should be twice the size of the ablation laser beam (5 mm). This can be easily done.

Fig.3 caption: (b) is not clear, please reshape the sentence; (3) is confusing, please rewrite.

Fig.6: Please use the same scales for the left and right Y-axes.

Fig.12: Please clarify that whether the stick spectra are for individual particles or the averaged ones? If averaged, please give the total number of the spectra for averaging. Please normalize the spectra, e.g., to the total ion intensity, and keep the same scales (both x and y) for consistency. E.g., m/z can be fixed from 0 up to 250 amu. for each spectrum. This can be applied to the special case of gold particles too, only need to illustrate the Au_2^+ additionally.

Fig.9 and 10: Please combine them. Please remove the AN measured by AMS and put it in a separate figure.

Fig.13: Please give the definition of the "sample number".

Fig.14: (a) It is hard to see the signal difference between shutter open and closed. Please consider a better way to demonstrate. (b) The calculated difference does not agree with the left spectrum. E.g., the bars are apparently not at the same positions between two plots; the most intensive peak m/z 28^+ (labelled N_2^+) is even a bit higher than the corresponding one in (a), as well as the m/z 32+, 40+, etc. The labels of N_2 and O_2 are confusing, since the peaks also contain the organic and sulfate fragments, respectively. Please modify them with a clearer way.