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

Hünig et al.

Replies to the comments by Anonymous Referee #1

General Reply:

We very gratefully acknowledge the detailed, diligent and careful review provided by Referee #1. This review significantly helped us to improve the manuscript.

The reviewer comments are written in this font style and color.

Our answers are written in this font style and color.

Changes to the revised version of the manuscript are printed in red.

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.

(Numbers of pages, lines and sections refer to the submitted manuscript for review)

- "vaporized" (P3L22) was corrected to "desorbed".

- Furthermore, the termini „LDI" (Laser Desorption and Ionization) for SPMS and „ TD-EI " (Thermal Desorption and Electron impact Ionization) for AMS were implemented to distinguish both methods in a clearer way.

- Correction of citation:
  The references Jimenez et al. (2003a), Jimenez et al. (2003b), and DeCarlo et al. (2004) refer to the definition of the vacuum aerodynamic diameter d_{va}. The reference Hinds (1999) was removed. The reference for sizing by means of a calibration in LAMS, Brands et al. (2011), was added. In total:

  "The time elapsing between the two light scattering signals is used to derive its vacuum aerodynamic diameter d_{va} (Hinds (1999), Jimenez et al. (2003b), Jimenez et al. (2003a), and DeCarlo et al. (2004)) by involving a calibration (see Sect. 3.2) and to determine the point in time the particle reaches the ablation spot of the ERICA-LAMS.

  was changed to

  "The time elapsing between the two light scattering signals is used to derive the particles vacuum aerodynamic diameter d_{va} (for definition see: Jimenez et al., 2003a, b; DeCarlo et al., 2004) by involving a calibration (Brands et al., 2011)"

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.

(Numbers of pages and lines refer to the submitted manuscript for review)

P2 L30-32:

"Also, since the repetition rate of high-power UV ablation lasers limits the number of particle detections per second, the addition of a thermal vaporization and electron impact ionization unit largely enhances the data yield for the particle analysis."

was changed to (Numbers of sections refer to the revised manuscript):

„ Since, beside other reasons (see Sect. 2.3), the temporal resolution of the ablation laser, limits the number of particles detected (e.g., Su et al., 2004). The addition of a TD-EI unit largely enhances the data yield for the particle analysis by complementary information."

P6 L13-14:

"This maximum repetition rate imposes a limit to the number of particles analyzed per time unit, which affects the spatial resolution for measurements from a fast flying aircraft."

was changed to:

"Beside other reasons, the maximum repetition rate of the ablation laser, particle losses in the ADL, the particle beam divergence, particle beam and laser beam alignment, the focusing width of the particle beam, the ionization efficiency of the particle components, and the sensitivity of the optical detection units limit the number of particles analyzed (Su et al., 2004; Zelenyuk and Imre, 2005; Brands et al., 2011; Marsden et al., 2016; Clemen et al., 2020), which affects the spatial resolution for measurements from a fast flying aircraft."

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.

(Numbers of pages, lines, sections, and figures refer to the manuscript submitted for review.)

Section 4 is not intended to be a comparison to highlight the differences of the ERICA-LAMS and the ERICA-AMS. Here, the possibility of obtaining complementary information and that this information can be merged is demonstrated. Therefore, not the differences are discussed here. In order to prevent the reader's expectation of a discussion on the differences, Fig. 17 was separated into 3 panels. See also our reply to RC2.

P20L37: To explain the high sulfate mass fraction value of 1 in 20 km altitude, following sentence was added: " Since no other species, such as nitrate or organics, were observed by the ERICA-AMS in significant amounts at this altitude, the convective and radiatively driven vertical transport within the Asian Monsoon Anticyclone (AMA; Ploeger et al., 2015) does not play as much of a role here anymore, as further below."

P21L24: We revised the following text passage and removed the misleading statement about the internal mixing state:

"The results can also be used to show that the aerosol composition between 10 km to 17 km differs from the aerosol composition above 17 km. For this, the mass fraction of sulfate (ERICA-AMS) and the number fraction of sulfate-containing single particle spectra (ERICA-LAMS) were examined. Below 17 km, the number fraction of sulfate-containing single particle spectra is stable around 0.6 and the mass fraction of the sulfate less than 0.2. This could be indicative for an internal mixing state of the measured aerosol particles, where the sulfate species within the single particles is assumed as predominantly refractory compound, since the mass fraction of the sulfate species is low compared to the number fraction

of sulfate-containing particles. The reason is that the ERICA-AMS only can measure non-refractory substances. Above 17 km, the composition is more complex. With increasing altitude, the sulfate mass fraction and the particle number fraction of sulfate-containing single particles increase up to 1. The change in mass fraction is strong compared to the number fraction of sulfate-containing single particles. Therefore, it can be assumed that the non-refractory content increases. Since the ERICA-LAMS is not able to detect pure (non-refractory) sulfuric acid, no distinct determination of the mixing state can be obtained. Here, an internal or an external mixing state but also a combination of both states can be present. In a conceivable internal mixing state, the non-refractory sulfuric acid has deposited on a particulate core, generating a coated particle or the sulfuric acid acts as a condensation nucleus for other substances. Additional pure sulfuric acid particles lead to an external mixing state."

Was changed to:

"The results can also be used to show that the aerosol composition and mixing state between 10 km to 17 km differ from those above 17 km. For this, the mass fraction of sulfate (ERICA-AMS) and the number fraction of sulfate-containing single particle spectra (ERICA-LAMS) were examined (Fig. 15). Below 17 km, the number fraction of sulfate-containing single particle spectra is stable around 0.6 and the mass fraction of sulfate in the non-refractory aerosol is less than 0.2. This indicates that many particles contain sulfate, but typically only in a small mass fraction (about 1/3 on average), because they are internally mixed with nitrate and organics. Above 17 km, with increasing altitude, the sulfate mass fraction and the particle number fraction of sulfate-containing single particles both increase up to 1. The observed change in the mass fraction is stronger, compared to the increase in the number fraction of sulfate-containing single particles. Since the two measurement methods provide not only different views on the aerosol, but also have different limitations, this observation must be interpreted with care. A possible interpretation for the increasing sulfate mass fraction could be that within the internally mixed aerosol of particles containing a refractory core, e.g. of meteoric dust, and a sulfuric acid coating (Murphy et al., 2014), the coating grows as a consequence of further condensation. However, since the ERICA-LAMS is not capable of measuring pure sulfuric acid particles (Murphy, 2007), it is also possible that partial external mixing of the internally mixed particles with sulfuric acid particles causes this observation."

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.

(Numbers of figures refer to the manuscript submitted for review.)

As the reviewer noticed, two different methods are used to determine the sulfate-containing (marker method) and the meteoric material -containing particle type (k-means). Both methods are briefly explained in the text. Since basically all "meteoric" particles are included in the "sulfate-containing" particles, the "meteoric" particles represent a subset of the sulfate-containing particles. Therefore, a summation of both particle number fractions is not meaningful. For better understanding and to avoid misinterpretation, Fig. 17 was divided into 3 panels and the description of the sulfate-containing single particles (measured by the ERICA-LAMS) was placed before the description of the mass fraction (measured by ERICA-AMS).

P20L38-40 was revised:

"To identify the sulfate-containing single particle spectra (ERICA-LAMS), the data set of the research flight of 04.08.2017 was filtered for single particle spectra that contained sulfate marker signals at m/z -96 (SO_4^-) or m/z -97 (HSO_4^-) or both markers."

Was changed to:

"To identify the sulfate-containing particle type, the ERICA-LAMS data set was filtered for single particle spectra that contained sulfate marker signals at m/z -96 (SO_4^-) or m/z -97 (HSO_4^-) or both markers. Since these sulfate marker signals are also found in the meteoric material containing particle spectra, by this approach, the "meteoric material containing particle type is a subtype of the sulfate-containing particle type."

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.

The paragraph was revised (see also reply to RC2)

"This indicates within the limitations of the applied methods that the composition of the sampled aerosol is well mixed within the particle boundary layer and in the free troposphere, although C_{total} changes. Thus, the EC particle number fraction cannot be used to define the particle boundary layer. In the ATAL, EC particles seem to play a minor role in the composition of the aerosol, while for the convective outflow levels the data suggest an increase in EC as result of detrainment."

was changed to:

"This indicates, within the limitations of the applied methods, that the EC particle type is well mixed within the boundary layer and in the free troposphere, although C_{total} changes. In the ATAL (> 16 km), EC particles seem to play a minor role in the composition of the aerosol, while for the convective outflow levels (< 16 km), the data suggest an increase of the EC particle number fraction as result of detrainment."

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).

The format was revised over the entire manuscript. The 'e.g.' was used to indicate that this reference is one example of many possible other references.

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.

(Numbers of sections refer to the manuscript submitted for review.)

Sections 2.3 and 2.4 were combined and the headers were simplified.

The instrument description is already kept to a minimum. The ERICA-LAMS is published here for the first time and we feel it should be explained in more detail. Some readers may not be very familiar with *both* techniques, as one reviewer actually indicated. And here we hope our description may be useful. The ERICA-AMS is an adopted Aerodyne AMS, but the actual settings such as vaporizer temperature, emission current, etc. are of interest for other AMS users. Although the information content regarding the ERICA-AMS has not been further reduced, the amount of text regarding the ERICA-AMS is now about half of the text regarding the ERICA-LAMS.

The major difference of the ERICA-AMS to the Aerodyne AMS, the use of the shutter unit instead of a chopper, was emphasized. Furthermore, it was highlighted that without a chopper, no size information can be obtained by the ERICA-AMS.

The difference was described in P7 L21-33 (Numbers of pages and lines refer to the manuscript submitted for review). However, the corresponding paragraph was revised.

"For quantitative aerosol composition measurements, the background signal, which originates from air molecules and residual vapor molecules inside the chamber, has to be considered and is subtracted from the aerosol sampling signal. For this purpose, in the commercial Aerodyne AMS (Canagaratna et al., 2007) the particle beam is periodically blocked by a chopper inside the low vacuum stage. By means of the chopper it is also possible to distinguish between different vacuum aerodynamic particle sizes, as the particle flight time duration between passing the (open) chopper and arriving at the vaporizer is size dependent. However, this flight time duration -and the corresponding flight distance between chopper and vaporizer- need to be long enough to achieve such size-resolved sampling. For ERICA-AMS the distance from the shutter to the vaporizer is very short. This would not be the case if we had placed a chopper directly behind the ball joint of the ADL. However, by periodically blocking the particle beam with a chopper at this position, the detection frequency of ERICA-LAMS would have been reduced accordingly. Thus, we decided to use a simple shutter device instead of the chopper. It consists of a C-shaped profile made of metal and is mounted on the shaft of a high-vacuum magnetically-coupled feed-through (Pfeiffer Vacuum GmbH, Germany). The shaft periodically rotates the C-profile by 90° into and back out of the particle beam axis. In this way, the particle stream to the vaporizer is blocked and permitted, respectively, for adjustable time periods."

Was changed to:

"For quantitative aerosol composition measurements, the background signal, which originates from air molecules and residual vapor molecules inside the chamber, has to be subtracted from the aerosol sampling signal. For this purpose, the SU is used to periodically block the particle beam. The SU consists of a C-shaped surface made of metal, which is mounted on the shaft of a high-vacuum magnetically-coupled feed-through (Pfeiffer Vacuum GmbH, Germany). The shaft periodically rotates the shutter by 90° into and back out of the particle beam path. In this way, the particle stream to the vaporizer is blocked

and permitted, respectively, for adjustable time periods. In the commercial Aerodyne AMS (Canagaratna et al., 2007), the particle beam is periodically blocked by a chopper inside the low vacuum stage. By means of the chopper it is possible to distinguish between different vacuum aerodynamic particle sizes, as the particle flight elapsed from its pass through the chopper until its arrival at the vaporizer is size-dependent. The distance between the chopper and the vaporizer and the corresponding flight time need to be long enough to achieve such size-resolved sampling. In the design of the ERICA-AMS, the distance from the shutter to the vaporizer is very short. This would not be the case, if a chopper was mounted directly behind the ball joint of the ADL. However, by periodically blocking the particle beam with a chopper at this position, the detection frequency of ERICA-LAMS would have been reduced accordingly. Thus, a simple shutter has been implemented and the particle size information can only be provided by the PDU of the ERICA-LAMS (see Sect. S4 in the supplement)."

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).

The particle time-of-flight calibration (particle size calibration) of the ERICA-LAMS was shifted to the supplement, since the approach with a polynomial fit is described in Brands et al. (2011).

The AMS IE and RIE sections were kept in the main part, since they are instrument specific and of interest for further publications. Also, the values differ from other AMSes. Thus, a presentation in the main part is reasonable.

(Numbers of figures refer to the manuscript submitted for review.)

Fig. 4 (example for the ALS position scan) was moved to the supplement, since the methodology of the measurement (including a figure) is described in Molleker et al. (2020). The basics of the methodology to determine the optical particle detection efficiency and the particle mass detection efficiency in our view should be better presented in the main text. Details of the complex determination procedure can be found in the supplement (Sect. S5, revised manuscript). It has to be emphasized that the ADL position scans are not only used for alignment, but also to determine the parameters for the particle and detection laser beam characteristics and, finally, the parameters DE_{max} and DE_{KTM}.

(Numbers of sections refer to the revised manuscript)

Following the reviewer's suggestion (for which we are quite grateful) Section 3 was restructured like this:

3 Instrument characterization

3.1 Characterization of the particle beam

3.1.1 Determination of efficiencies for optical particle detection and particle mass

3.1.2 Particle beam properties

3.2 ERICA-LAMS characterization

3.2.1 Detection and ablation laser beam widths

3.2.2 Optical particle detection efficiency

3.2.4 Hit rate

3.2.5 Single particle mass spectra

3.2.5.1 Single particle mass spectra from laboratory tests

3.2.5.2 Mass spectral resolution

3.3 ERICA-AMS characterization

3.3.1 Mass spectral resolution and data preparation

3.3.2 Particle mass detection efficiency

3.3.3 Ionization efficiency

3.3.4 Detection limit

3.3.5 Airbeam and water signal

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".

(Numbers of sections and equations refer to the revised manuscript)

The term ‚ablation efficiency (AE)' was replaced by the term ‚hit rate (HR)', since this is the more common term in the community and do not exclude other efficiencies as ionization and ion extraction efficiency. The definition is given by Eq. (5) and is the same as used by, e.g., Brands et al. (2011) (termed ablation efficiency), Su et al. (2004) (termed hit rate), and Gemayel et al. (2016) (termed hit rate).

The term ‚detection efficiency' varies within the SPMS literature: In Gemayel et al. (2016) this term is used as the overall detection efficiency: A product of the hit rate and the 'scattering efficiency (SE)'. The latter term is defined as the here used optical detection efficiency DE_{PDU} (Eq. (1)), related to one of the

particles detected at one of the detection lasers. In Marsden et al. (2016), the symbol E_{detect} is used. In Molleker et al. (2020), the term ‚detection efficiency' is used without an abbreviation, but with the same definition as in the manuscript here. In Brands et al. (2011), the detection efficiency of the ALABAMA refers to the number of particles detected at both detection units within a given time interval and whose sizes were successfully determined. In Clemen et al. (2020), the detection efficiency of the ALABAMA also refers to the number of particles detected at both detection units within a given time interval for the measurements at the optimal fixed position of the aerodynamic lens system, whereas the detection efficiency for the ADL scans, just like in this study, refers to the individual detection lasers. Finally, we kept the term ‚detection efficiency' (in the manuscript for clarification with the adjective ‚optical').

Furthermore, the detection efficiencies (optical detection efficiency measured at the PDUs DE_{PDU} and the particle mass detection efficiency measured at the ERICA-AMS vaporizer $DE_{vaporizer}$) are defined by Eq. (1) and in supplement Eq. (S16), respectively and also, the curve fit functions (Eq. (2), (S15), and (S17)). The combination to DE_{max} and DE_{KTM} is described in Sect. S5.6.

The term ‚collection efficiency' is not applicable for measurements with an optical device, since the particles are not „collected" literally. However, it is applicable for the ERICA-AMS, since at the vaporizer the particles get in a sense „collected". The definition of $DE_{vaporizer}$ (Eq. (S16)) is very similar to the definition of the ‚collection efficiency (CE)' used in the AMS community. However, to keep consistency and not to confuse the reader, we keep the term ‚detection efficiency' also for the ‚collection efficiency' of the ERICA-AMS. This is also one of the reasons, why we provide the equations, from which the terms become clearer. To consider the fact that $DE_{vaporizer}$ and CE are defined in the same way, the text paragraph (P12L1, submitted manuscript for review) has been adapted: „ Simultaneously to the measurements with AN particles at the detection units PDU1 and PDU2 of the ERICA-LAMS, the mean mass concentration of AN was measured with the ERICA-AMS, similar to the approach described in Liu et al. (2007). The efficiency with which particle mass concentrations were measured with the ERICA-AMS was determined. While this quantity is equivalent to the 'collection efficiency' (CE; e.g., Canagaratna et al., 2007; Matthew et al., 2008; Drewnick et al., 2015) in AMS measurements, we define it as 'particle mass detection efficiency' for consistency with the ERICA-LAMS discussion. "

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.

(Numbers of sections refer to the revised manuscript)

Readers may expect size distributions from the ERICA-AMS, because these commonly are provided by the Aerodyne AMS. As mentioned above, no size information can be obtained by the ERICA-AMS, due to the lack of a chopper. (The corresponding paragraph in Sect. 2.4 was revised in order to emphasize the difference to the commercial Aerodyne AMS.) Therefore, no size distribution can be shown for the ERICA-AMS.

The EC particle type (single particle data) is just an example that size information from the ERICA-LAMS is evaluable and is meant as "proof-of-concept". However, the original Fig. 19 and its discussion was shifted into the supplement, since the ability to provide size information is already shown in Fig. 11 (submitted manuscript for review), where the size dependency of the hit rate is shown for an ambient

measurement. The determination and evaluation of particle types other than EC and the evaluation of particle size distributions during this field campaign is beyond the scope of this manuscript and will be part of a forthcoming publication.

In the manuscript

"As an example that the ERICA-LAMS provides single particle size information, Fig. 19 shows the size distribution of EC-containing particles for the research flight on 04.08.2017 consisting of three modes. The first at the edge of the small particle sizes below 200 nm, the second between a particle size of around 300 nm and 1700 nm with a maximum particle number fraction of 0.08 at 800 nm, and the third between 1700 nm and 2600 nm with a maximum of 0.17."

was replaced by:

„An example for single particle information, which ERICA-LAMS is capable of delivering, is provided in Sect. S8 of the supplement. Due to the lack of a chopper, no particle size information can be determined by the ERICA-AMS."

5) Figures:

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

(Numbers of sections and figures refer to the manuscript submitted for review.)

Fig. 3 (and the corresponding Sect. 3.2) was moved to the supplement (see reply to major comment 2.3 above).

Fig. 4 was moved to the supplement, since such types of graphs are presented already in the literature. For example, Molleker et al. (2020) show a graph measured by the ERICA in the main part.

Fig. 7 and 8 were combined.

The combination of Fig. 8 and 9 is not meaningful. In case the reviewer meant Fig. 9 and 10: In order not to overload a graph and keep two different types of detection efficiencies separate, both figures were kept separately.

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.

Solid markers were changed to non-filled markers. In addition, the marker size was enlarged to make the error bars visible.

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?

(Numbers of equations and sections refer to the manuscript submitted for review.)

We changed the solid markers to non-filled markers to make the error bars visible. Determination of uncertainties is now described in the figure captions.

For one DE_{PDU} data point (see Eq. (4)), a single run of 30 seconds was performed (see Sect. 3.3.1). Since the measurement of $DE_{vaporizer}$ (see Eq. (S15) in the supplement) was simultaneously performed, the measuring time is the same as for the measurements of one DE_{PDU} data point.

More details on the applied methods can be found in Sect. 3.3 and in the supplement Sect. S2, S3, and S4.

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.

(Number of figures refer to the manuscript submitted for review.)

Fig. 12: The axes were changed to log scale and the labels were changed to „ion peak area in mV·sample“. The abscissas were changed to maximum m/z 250 (gold particle up to m/z 400). Note: BaA and gold particle spectra were swapped.

The spectra show single particle spectra, on which the ion marker threshold can be applied. Thus, a normalization is not appropriate.

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.

(Numbers of pages, lines, and sections refer to the manuscript submitted for review.)

- Section 3 of the manuscript was restructured as the reviewer suggested. By that, the instrument presentation was improved and the instrumental design should be much clearer now.
- The discussion on complementary results is part of Sect. 4. For better understanding, this section was revised.
- The instrument was designed initially for the mobile field deployment aboard the high-altitude research aircraft *Geophysica*. Here, valid for all (high-altitude) research aircraft, weight and space for the payload is limited. In addition, field deployments with research aircraft at high altitudes are rare, so as much information as possible (with as many instruments as possible) should be collected. Thus, a compact design is crucial for implementation on such aircraft and therefore a combination of two measurement methods into one apparatus a major advantage.

  „The final design of the compact instrument was implemented into an aircraft rack (Dragoneas et

al., 2022) of 60 cm x 74 cm x 140 cm (height x width x length) with a total weight of 200 kg. Such a compact and light-weight design is essential for aircraft implementation, especially aboard a high-altitude aircraft.“

was changed (and on request from Reviewer #3 shifted to Sect. 1):

„ Furthermore, the mechanical components of ERICA are designed to operate under the demanding conditions like thermal stress and vibrations aboard an aircraft. The final design of the compact instrument was implemented into an aircraft rack (Dragoneas et al., 2022) of 60 cm x 74 cm x 140 cm (height x width x length) with a total weight of 200 kg. In addition, field deployments with research aircraft at high altitudes are rare, so as much information as possible − with as many instruments as possible −should be collected. Thus, a compact design is crucial for implementation on such aircraft and therefore a combination of two measurement methods into one apparatus is a major advantage.“

– In the outlook (Sect. 5) on P24L33, a future mode for the ERICA is presented. This mode is only possible with a serial linkage of a LAMS and an AMS, like it is in ERICA. The paragraph was revised to highlight this unique feature as an advantage:

„For the same point in time, a data acquisition card is triggered and, similar to the procedure with a light scattering probe on the AMS (Cross et al., 2007; Freutel, 2012), the single particle mass spectrum is recorded. In this way it is possible to quantify the non-refractory components of a single particle. In addition, the size information of the measured single particle is obtained by means of the particle flight time between the two PDUs. Here, a future characterization of interest is the ablation laser’s effect to the particles that are only partly ablated and the residuals reach the vaporizer of the ERICA-AMS. For this purpose, a method has to be developed to ensure the linkage of the results to the very same particle. Such a procedure needs more implementations and further laboratory studies.“

was changed to

„For the same point in time, the data acquisition card is triggered and the single particle mass spectrum is recorded. For the ERICA this mode is called optically triggered AMS (OT-AMS) mode. With the method of the OT-AMS mode, it is possible to quantify the non-refractory components of single particles when the ablation laser is in idle mode. This method is similar to the procedure with a light scattering probe on the AMS (Cross et al., 2007; Freutel et al., 2013). In addition, the size information of the measured single particle is obtained by means of the particle flight time between the two PDUs. One possible future investigation by means of the OT-AMS mode is the ablation laser's effect on the particles that are only partly ablated and where the residuals reach the vaporizer of the ERICA-AMS. This investigation is only possible with the unique feature, the serial configuration of SMPS and AMS, as in the OT-AMS mode. A method has to be developed to ensure the linkage of the results to the very same particle. Such a procedure needs more implementations and further laboratory studies. “

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.

(Numbers of figures refer to the revised manuscript)

We don't understand which statements are regarded not to be valid and why particles smaller than 200 nm diameter should not be detected. The authors are aware, and this is also described in the manuscript or was measured by us, that the detection efficiency decreases significantly below 200 nm. However, the following arguments support that the detection efficiency for PSL particles of sizes 80nm and 108 nm is non-zero:

- Fig. S9 in the supplement shows the histograms of the PSL calibration measurements, which demonstrate the ability of the ERICA, to optically detect particles of sizes in a range between 80 nm to 5145 nm.
- Fig. S21 in the supplement shows the size distribution from a research flight during the second aircraft field campaign of StratoClim on 08.08.2017. Here, mass spectra from particles in a size range between 100 nm and 3700 nm were obtained.
- Fig. 8 was revised and shows the number of ablation laser shots and the number of recorded spectra now in log scale to highlight that ambient particles in the size rage of 80 nm to 4000 nm can be optically detected. Also, particles below 200 nm were ablated during this experiment. However, their hit rate and the numbers are low (HR: 2 to 11 %; 1 to 8 spectra).

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.

Done. Particle sizes are given now in ‚nm' (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.

"Within certain limitations this may become possible, if the data of other instruments are included in the analysis (e.g., in Froyd et al. (2019)).

was changed to:

„Within certain limitations this may become possible, if the data of other instruments are included in the analysis (e.g., Ault et al., 2009; Healy et al., 2012; Gunsch et al., 2018; Köllner et al., 2021)."

P3L25: Please cite the corresponding publications.

Since for ERICA the „large fraction“ is an assumption, we changed the text as follows:

„A large fraction of the particles is not ablated by laser pulses, either because the laser pulses miss the particles, or because the particles are too small for the optical detection. However, even most particles amenable for laser ablation, which pass through the ablation region, remain undestroyed, because the laser is firing at a limited maximum repetition rate of 8 pulses per second.“

was changed to (see also reply to RC2):

“It is assumed that a large fraction of the sampled particles will not generate a single particle spectrum. The major reasons for this effect are: First, the particles are not ablated, because the laser is firing at a limited maximum repetition rate of 8 pulses per second. During the idle time of the Nd:YAG laser, particles remain unablated, even if they are successfully detected by the units PDU1 and PDU2. This actually is by far the largest fraction of the sampled particles emerging from the ADL. If, for example, the ambient number density of particles with diameters above the optical detection limit is 100 cm^{-3}_{Std}, then, at most only 5.4 % (8 shots per second and sampling volumetric flow rate of 1.48 $cm^3\ s^{-1}$) of the detectable particles are hit by the laser. Second, the particles are too small for optical detection. Third, particles for which the calculation of the trigger failed continue their travel towards the ERICA-AMS vaporizer. Fourth, particles that primarily consist of materials that are transparent at a UV wavelength of 266 nm, such as pure sulfuric acid, are hard to ablate (Murphy et al., 2007). We selected a UV laser with 266 nm wavelength due to smaller dimensions of the laser and the fact, that chemical substances show less fragmentation compared to ablation with shorter wavelengths (Thomson et al., 1997). In general, however, it is also possible to implement excimer lasers operating at shorter wavelength to ablate pure sulfuric acid droplets. Also, pure sulfuric acid is detected by the ERICA-AMS. Thus, even most particles amenable for laser ablation, which pass through the ablation region, remain undestroyed. Another reason why a spectrum is not triggered over a signal threshold for recording is a low number of generated ions during the LDI process.“

P3L32: Please use 3.17 µm to keep consistency.

Done

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

(Numbers of sections refer to the manuscript submitted for review.)

The transmission efficiency of the deployed ADL as published by Xu et al. (2017) is mentioned two lines below. However, the term ‘transmission efficiency’ was not mentioned in the submitted manuscript.

“The detectable particle size range (d_{va}) of the ERICA-LAMS is between ~180 nm and 3170 nm (see Sect. 3.3.3). However, the signal-to-noise ratio of optical particle detection is sufficient for particle time-of-flight calibration between 80 nm and 5 µm (see Sect. 3.2). The detectable particle size range of the ERICA-AMS is assumed to be the same as published by Xu et al. (2017) for the deployed lens type.: ~120 nm to 3.5 µm.”

was changed to (Numbers of sections and figures refer to the revised manuscript; see also reply to RC2 and RC3):

"The particle size range within the 50 % cut-off in detection efficiency (d_{50}) of the ERICA-LAMS is between 180 nm and 3170 nm (see Sect. 3.2.2). The signal-to-noise ratio of optical particle detection is sufficient for particle time-of-flight calibration between 80 nm and 5000 nm (see Sect. S4 in the supplement). For the ERICA-AMS, the detectable particle size range is determined by the transmission and focusing properties of the aerodynamic lens. For the ADL used in our instrument, Xu et al. (2017), who used this lens in combination with an ACSM (Aerosol Chemical Speciation Monitor), determined a transmission range from ~120 nm to 3500 nm. We assume that the detectable particle size range of the ERICA-AMS matches this transmission range."

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

The difference was described in P7 L21-33 (Numbers of pages and lines refer to the manuscript submitted for review). For changes, see reply on major comment 2.2 (above).

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

Done

P4L26: Please change the dot in "$5 \cdot 10^2$ cm³ s^{-1} " to multiplication symbol "5×10^2 cm³ s^{-1}" and revise the others throughout the manuscript.

Done

P15L28: Fig 10 should be Fig 11.

Done

P16L11: Please give the reason for choosing these peaks.

„The ion peak area threshold is defined as the ion peak area at m/z, which are usually unoccupied (m/z 2 to m/z 6 for cations, m/z 2 to m/z 11 for anions), below which 99% of the baseline noise is present (Köllner et al., 2017)."

was changed to (see also reply to RC2):

„The ion peak area threshold is defined as the ion peak area at m/z, on which during ambient measurements typically no signals occur (m/z 2 to m/z 6 for cations, m/z 2 to m/z 11 for anions). To determine the ion peak area threshold, the normalized cumulative signal intensity distributions for each usually unoccupied m/z were made and the overall 99 % threshold was determined (Köllner et al., 2017). Below this ion peak area threshold, 99% of the baseline noise is present (Köllner et al., 2017). The result for cations and anions is an ion peak area threshold value of 7 mV·sample."

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

Done

P19L27: The left half of the bracket is missing.

Added left half of the bracket

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.

The paragraphs were re-arranged and revised. The transition was smoothed as follows:

"In this way, two particle types (in addition to other particle types not included in this publication) well described in the literature were found: A meteoric material containing (e.g., Schneider et al. (2021)) and an elemental carbon (EC) containing particle type (e.g., Pratt and Prather (2010)). In the following, we focus on the aerosol composition at high altitudes (> 10 km) considering particulate sulfate and the meteoric material containing particle type."

Was changed to

„With this approach, two particle types (in addition to other particle types not included in this publication) well described in the literature were found: A meteoric material containing (e.g., Schneider et al., 2021) and an elemental carbon (EC) containing particle type (e.g., Pratt and Prather, 2010). To identify the sulfate-containing particle type, the ERICA-LAMS data set was filtered for single particle spectra that contained sulfate marker signals at m/z -96 (SO_4^-) or m/z -97 (HSO_4^-) or both markers. In the following, first, we focus on the aerosol composition at high altitudes (> 10 km), considering particulate sulfate as well as the meteoric material containing particle type.

P20L31: Incorrect statement. Please revise.

„The sulfate particle type measured by the ERICA-AMS is a non-refractory species (Canagaratna et al., 2007) and consists mainly of pure sulfuric acid in the stratosphere (Murphy et al., 2014)."

was changed to

"Non-refractory sulfate (Canagaratna et al., 2007) measured by the ERICA-AMS consists mainly of pure sulfuric acid in the stratosphere (Murphy et al., 2014)."

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

„In 20 km altitude, the sulfate mass fraction is 1."

was changed to

„In 20 km altitude, the non-refractory aerosol sulfate mass fraction is 1.

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

We revised the text passage and added two references and highlighted our assumptions (changes see reply on major comment 1.3 above).

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.

(Number of figures refer to the manuscript submitted for review.)

The TMPs are now numbered from TMP1 to TMP3. TMP1 is a four-stage TMP with the numbered pumping stages PS1 to PS4.

The distances are provided in Fig. S3 in the supplement, since they are not further discussed

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.

After a bit of discussion, we decided to leave this figure as a not-to-scale-drawing, but at least we narrowed the laser beam.

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

Done

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

We prefer to leave the scaling as is. Scaling the left axis to 2.5, the details in presentation would get lost. Scaling the right axis to 0.25, the data points would be out of scale.

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.

Caption changed to „Exemplary stick mass spectra (m/z) of four laboratory generated single particles as measured by ERICA-LAMS.“

It is mentioned that the intensities are not normalized.

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

(Number of figures refer to the revised manuscript)

To clearly differentiate between maximum possible DE_{max} and DE_{KTM} during the aircraft campaign, we preferred to not merge the panels. However, the measurements at the ERICA-AMS were separated to a new figure (Fig. 12).

Fig.13: Please give the definition of the “sample number”.

(Number of figures refer to the manuscript submitted for review.)

In Fig. 13 the raw spectrum is depicted. The abscissa was changed from “sample number” to the (to a raw spectrum of a TOF-MS) more intuitive term “ion flight time”.

The sample number is the number of samples of the oscilloscope (Picoscope) during recording the single particle spectrum. The time resolution is set to 1.6 ns per sample. Thus, by multiplying the sample number by 1.6, the ion flight time (in ns) in the TOF-MS can be determined.

The caption was revised accordingly:

"Details of cation raw spectra (voltage output versus sample number of the digitizer, 1.6 ns per sample) of two ambient single particles at the airport of Kathmandu, Nepal. (a) Tin isotopic pattern (d_{va} = 277 nm). (b) Lead isotopic pattern (d_{va} = 311 nm)."

was changed to:

"Details of cation raw spectra (voltage output versus ion flight time in the B-ToF-MS) of two ambient single particles at the airport of Kathmandu, Nepal. (a) Tin isotopic pattern (d_{va} = 277 nm). (b) Lead isotopic pattern (d_{va} = 311 nm)."

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.

Panel (a) was removed, since no further discussion is presented in the text. The tags were changed to m/z values.

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