

Replies to comments of Referee 1:

Suggested additions:

1)

Referee comment: *For both the single particle and thermal desorption mass spectrometers, please list some dimensions and voltages. The energy of the ions is especially important since it affects detection at the MCP. The electric field across the ion source is also important for understanding the ionization processes. Simply calling it an Ionwerks spectrometer is not sufficient.*

Answer: Regarding the physical characteristics of the mass spectrometers, the ERICA-LAMS bipolar mass spectrometer measures 250 x 667 x 90 mm; the longest dimension (667 mm) is equally distributed to the positive and negative ion-flight chambers, wherein the ions follow a V-shaped trajectory. The dimensions of the ERICA-AMS mass spectrometer (including the ionization chamber) are 300 x 290 x 130 mm. Also, detailed information about the ERICA-LAMS dimensions and voltages are given on p. 190 (Fig. 90, Table 19) in the PhD thesis of Hünig (2021). Similarly, the respective information for the ERICA-AMS is given on p. 191 (Fig. 91, Table 20) of the same thesis.

The ion impact energy for the ERICA-AMS is approximately 2800 eV and, noteworthy, the detection is strongly influenced by the electron avalanche, which is determined by the voltage across the MCP stack (about 2000 V). The ion source itself is designed to primarily be field-free. The geometry of the ion extraction field is similar to those of other commercial Aerodyne AMS instruments (Drewnick et al., 2005), but its exact dimensions are unknown to us. Nevertheless, the ionization process is rather dependent on the acceleration and energy of the incident electrons, which is 70 eV, as mentioned in Line 135.

Changes: The dimensions of the ERICA-LAMS and the ERICA-AMS have been added to Section 1, together with the description given above. We have also added a reference to the PhD thesis of Hünig (2021), wherein the voltage settings are given in detail.

2)

Referee comment: *In a bipolar mass spectrometer, one or both detectors must be floating at high voltage. Please describe in detail how the signals are coupled to ground and what preamplifiers are used.*

Answer: Both units (for anions and cations) are identical and decoupled from ground using one high voltage capacitor (100 pF, 7 kV) each. The preamplifiers are manufactured by Tofwerk AG, they are AC-coupled, their bandwidth is 3 kHz to 1.8 GHz and their gain is 21 db.

Changes: This information has been added to the text (Line 130).

3)

Referee comment: *Describe the spot size for the ionization laser. A 10 mJ pulse has very different implications depending on how tightly it is focused.*

Answer: The ablation laser characteristics are discussed in Section 3.2.1 of Hünig et al. (2022). The spot diameter is approximately 250 μm .

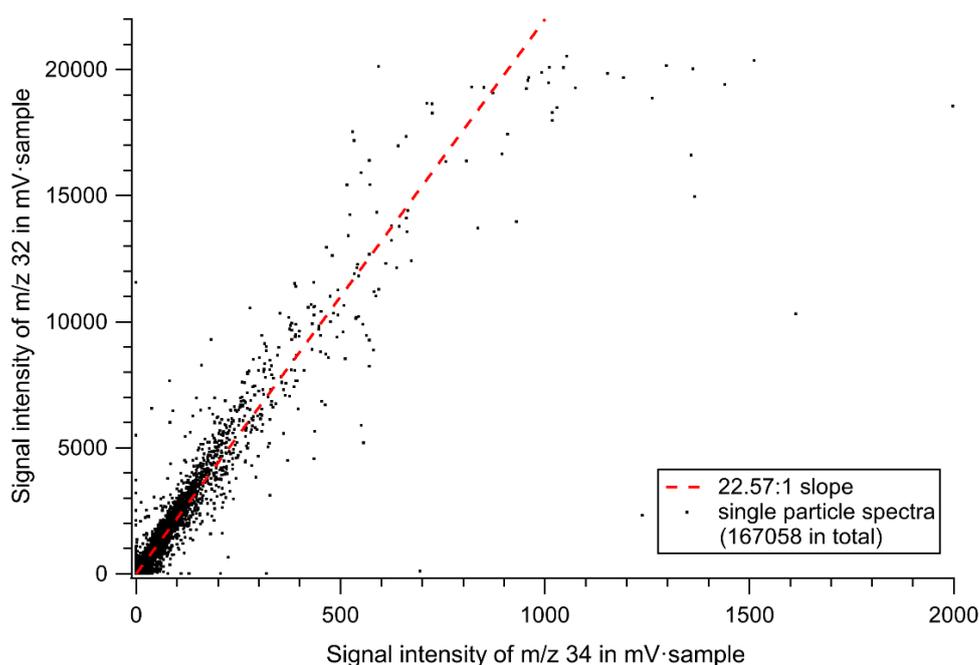
Changes: This piece of information has been added to the text (Line 122).

4)

Referee comment: *What fraction of laser shots in the single particle instrument result in spectra? Is this a function of particle size?*

Answer: The fraction of laser shots which results in single-particle spectra is discussed in Section 3.2.3 of Hünig et al. (2022) and the size-dependence of it is illustrated in Fig. 8 of the same publication. This fraction, also referred to as “hit rate”, can exceed 50% for particle sizes around 250 nm, while it typically ranged from 10% to 30% for the particles collected during the aircraft deployments of the instrument.

Changes: A relevant sentence regarding the hit rate, together with a reference to Hünig et al. (2022), have been added to Section 4.3.



5)

Referee comment: *It might be helpful to show an isotope ratio plot as a diagnostic of the linearity of the single particle spectrometer. 41K versus 39K, 54 and 56Fe, or 32 and 34S would be possibilities. There are other contributions to the peaks but a scatter plot will show a locus of points along the isotope ratio.*

Answer: To demonstrate the linearity of the ERICA-LAMS, we have added a plot of m/z 32 versus m/z 34 to the supplement, as per your suggestion, which is shown above. The dashed red line indicates the ratio of the naturally occurring abundance of the two isotopes of sulfur.

Changes: A sentence regarding the linearity of the instrument with a reference to this figure has been added to the text (Line 665). The figure has been added to the Supplement.

6)

Referee comment: *What detection limits were achieved for the thermal desorption (AMS) spectrometer?*

Answer: The ERICA-AMS detection limits are discussed in detail in Section 3.3.4 of Hünig et al. (2022). More specifically, Table 1 of this paper, gives the detection limits for chloride, ammonium, nitrate, organics, and sulfate.

Changes: Since this information is provided by the companion paper by Hünig et al. (2022) and extends beyond the scope of the work presented here, we have made no changes in this case.

Technical comments:

a)

Referee comment: *Line 61: The strong statement about quantitation is not true. SPMS instruments can be quantitative both for types of particles and components within particles:*

Cornwell et al., Direct Online Mass Spectrometry Measurements of Ice Nucleating Particles at a California Coastal Site, JGR, 2019.

Cziczo, et al., Ablation, Flux, and Atmospheric Implications of Meteors Inferred from Stratospheric Aerosol, Science 291, 1772 (2001);

Froyd et al., A new method to quantify mineral dust and other aerosol species from aircraft platforms using single-particle mass spectrometry, Atmos. Meas. Tech., 12, 6209–6239, 2019

Qin et al., Comparison of Two Methods for Obtaining Quantitative Mass Concentrations from Aerosol Time-of-Flight Mass Spectrometry Measurements, Anal. Chem. 2006, 78, 6169-6178

Answer: Although a strong statement may not be apt in this case, we still think that the basic idea behind that statement is true: SPMS methods are generally incapable of providing quantitative aerosol concentrations without applying scaling provided by independent external measurements.

We believe that the cited publications under your comment support this statement. In some of these publications, the authors even explicitly state that SPMS is not capable of providing quantitative information. In some of these papers, as well as in a large number of other SPMS publications, fractional contributions of individual aerosol types to the total aerosol are determined, often as a function of particle size. However, as stated in some of the cited papers, this approach is limited by the fact that particle detection and ablation / ionization can depend on the type of particles, due to their different particle shapes and optical properties. Such effects even limit these semi-quantitative capabilities of SPMS, i.e. the determination of fractional contributions of different particle types.

Quantitative determination of components within particles, i.e. the measurement of concentrations of chemical components within the particles or within the aerosol, is further limited due to the generally particle-size-dependent and often particle-type-dependent detection of particles and the LDI ionization process that produces absolute and relative ion signal intensities, which can strongly depend on the particle matrix. Also, this is stated in several of the publications, cited by the reviewer. The presented approaches to calculate absolute concentrations of different particle types, all rely on the use of external size distribution measurements, combined with assumptions about the composition of individual particle types. Using this approach, indeed, some quantitative information can be indirectly extracted from the SPMS measurements.

Changes: To make clearer that SPMS cannot deliver quantitative measurements by itself, while it can contribute (together with other measurements) to the calculation of quantitative data, we re-worded the statement (Line 60): *“Most importantly, a number of studies have shown that the use of simultaneous vaporization and ionization, as it happens with LDI, results in strongly matrix-dependent ion signals, which constitutes the main reason why SPMS does not intrinsically provide quantitative measurements (Reilly et al., 2000; Allen et al., 2000)”*.

b)

Referee comment: *Line 72: It is worthwhile to mention that a bipolar SPMS has been flown at lower altitudes: Pratt et al., Anal. Chem. 2009, 81, 1792–1800, Development and Characterization of an Aircraft Aerosol Time-of-Flight Mass Spectrometer*

Changes: We have added the following sentence to the text (Line 75): *“However, it is worth mentioning that at least one bipolar SMPS instrument has been demonstrated to operate aboard an aircraft, albeit at much lower altitudes (Pratt et al., 2009)”*.

c)

Referee comment: *Line 105: Please mention the model of the PMT.*

Answer: The model of both PMTs used in the instrument is H1021-210, manufactured by Hamamatsu Photonics K.K. Japan. These products are PMT modules featuring an embedded high-voltage supply; their gains are externally controlled by two separate low-voltage signals.

Changes: This information has been added to the text (Line 108).

d)

Referee comment: *Line 131. Having the motor outside is potentially much cleaner than the motor inside the vacuum. Is the organic background in the AMS region lower than in a stock Aerodyne AMS?*

Answer: Indeed, the reason of using a magnetically-coupled shaft is to substantially reduce contamination, especially due to the proximity of the shutter to the ionization region of the ERICA-AMS. As a side note, it is worth mentioning that the organic background of any AMS instrument deployed for aircraft measurements is largely depended on the pumping time elapsed prior to the onset of measurements. This is due to the fact that all aircraft instruments are typically powered down and only turned on a few hours before each take-off. Different pumping times yield different vacuum levels, which in turn, result in different organic backgrounds. As also discussed above (answer 6), the detection limits of the ERICA-AMS for the StratoClim measurements are given in Section 3.3.4, in the companion paper by Hünig et al. (2022). The value for organics was 500 ng/m³ for measurement cycles of 10 s, which corresponds to approximately 290 ng/m³ for a 30-second average. Although better values have been reported for Aerodyne AMS instruments deployed for aircraft measurements, i.e. 70 to 140 ng/m³ (Schmale et al., 2010), and 110 ng/m³ (Schulz et al., 2018), a direct comparison cannot be easily made, as the pre-flight pumping times may differ.

Changes: The sentence *“The shutter is driven by an external servo motor via a magnetically-coupled feedthrough rated for ultra-high vacuum”* has been changed to: *“Due to the proximity of the shutter the ionization region of the ERICA-AMS and to avoid contamination, a servo motor has been installed outside the vacuum chamber and connected to the shutter via a magnetically-coupled feedthrough rated for ultra-high vacuum”* (Line 139). We haven't given any reference to the organic background due to the reasons discussed in the answer above.

e)

Referee comment: *Line 145. An AMS has previously been flown on a stratospheric balloon. Although not built by Aerodyne, it was an AMS: it included an aerodynamic lens, vaporizer, shutter for the particle beam, and a mass spectrometer. Schreiner et al., A mass spectrometer system for analysis of polar stratospheric aerosols, Review of Scientific Instruments 73, 446 (2002); <https://doi.org/10.1126/science.290.5497.1756>.*

Changes: We have added the following statement to the Introduction (Line 81, not in Section 1): “In addition, instruments employing the TD-EI technique have been used for high-altitude balloon-borne measurements (Voigt et al., 2000)”

f)

Referee comment: *Line 178: I found the discussion of changing pressure confusing – the instrument is in a pressure vessel, only talked about later. Readers will think the electronics are exposed to changing pressure.*

Answer: That sentence refers to the instrument compartments (bays) of the aircraft. To avoid confusion, we have slightly rephrased that sentence.

Changes: Sentenced rephrased to: “Moreover, the technical specifications and limitations of the aircraft were taken into consideration; such parameters are the lack of pressurized instrument compartments, the absence of an instrument operator on board, the electrical supply limitations, and the high speed of the platform” (Line 188).

g)

Referee comment: *Circa line 220: Given the radiative cooling, it is surprising that the instrument couldn't just stay fairly indefinitely in low power mode with a modest external fan blowing on the pressure chamber rather than a dedicated air conditioning unit. Radiative cooling with a 10 to 20C temperature difference is hundreds of watts.*

Answer: We can confirm that your assumption on radiative cooling holds true; as an example, for a temperature difference of 20 K (i.e. pressure vessel temperature = 40 °C, cowling surface temperature = 20 °C) and an emissivity of 0.8 for painted aluminium, the radiative cooling is approximately 470 W. However, as stated in Line 362, the power consumption of the instrument in Low-Power Mode is about 600 W while the ambient temperature on ground was even exceeding 30 °C during pre-flight operations. The ambient temperature can be seen in Fig. 7. For these reasons, the use of an external air-conditioning unit has been deemed necessary.

Changes: No changes have been made in this case.

h)

Referee comment: *Line 665: I am convinced the gold is from contamination; you don't need to mention space debris.*

Changes: We have removed the statement regarding space debris (Line 701).

i)

Referee comment: *Section 4.4 first paragraph: You can probably reduce the number of times the same references are cited in successive sentences.*

Changes: The double citation has been removed from the first paragraph of Section 4.4.

j)

Referee comment: *Figure 4: I think Figure 4 belongs in supplemental.*

Answer: Regarding your comment on Fig. 4, we suggest keeping it in the main manuscript rather than moving it to the supplement. The main reason is that it illustrates all units (sub-systems) which are described in the text, while it clearly shows the signals exchanged between them, making easier for the reader to understand the operation of the entire system.

Changes: No changes have been made in this case.

k)

Referee comment: *What supplemental material is there is appropriate and appreciated. One could also consider a digital file of the spectra shown in the figures in the manuscript.*

Changes: The files with the spectral data shown in all relevant figures are available online.

Replies to comments of Referee 2:

a)

Referee comment: *It would be nice to see more descriptions of how the SP and TD/EI mass spectrometry systems work together to provide a more complete dataset than is currently possible. Do the data sets from ERICA-LAMS and ERICA-AMS strongly educate one another in the context of UT/LS deployments? Such a demonstration would highlight the (already fair) statement that the two aerosol MS approaches are complementary. Perhaps some of this material is published in other papers on this instrument, but herein lies the confusion that can ensue without a good overview, in what is truly an instrument development paper. How does the output of this instrument and its unique configuration/construction/engineering/implementation advance science? It would be entirely germane to discuss the complementarity of the LAMS and AMS subsystems in the context of the UT/LS studies that have been indicated in the paper already. There is a short mention of this concept toward the end of the paper, but it is perhaps the most important advancement that this instrument affords from a data collection perspective (of course the high-altitude capabilities are clear).*

Answer: It is true that the major driving force for the design and development of the ERICA have been the limitations and requirements imposed by the available platform for stratospheric research. For a comprehensive chemical composition analysis of aerosols in the UT/LS, the development of a compact and versatile instrument was imperative. Furthermore, this unique combination of an LDI and a TD-EI mass spectrometer has introduced new aerosol research possibilities, which are currently subject to investigation. Most notably, one of these advances is the recent development of an optically-triggered single-particle AMS mode, as discussed in Section 5.

Addressing the overall scientific contribution of the instrument so far, we would like to highlight that the deployment of the ERICA in the UT/LS during the StratoClim project has already yielded a quite detailed elucidation of the chemical composition of the Asian Tropopause Aerosol Layer (ATAL) during the StratoClim project (Höpfner et al., 2019; Appel et al., 2022). Here, the data sets of the quantitative measurements from the ERICA-AMS demonstrated that this layer mainly consists of nitrate particles, with organics being the second most important component. This would not have easily been shown

by the ERICA-LAMS alone, considering that the single particle measurements do not very well reflect particle number densities. These measurement results may be indicative of the existence of a ‘layer’ within the UT/LS of the Asian Monsoon Anticyclone, i.e. by showing an increased frequency of detected nitrate lines in particles, albeit not in a quantitative way. During its operation in the UT/LS, the ERICA-AMS provided crucial quantitative values expressed in micrograms per cubic meter (Appel et al., 2022).

Secondly, from AIDA laboratory measurements, it was found that the ATAL ammonium nitrate particles can only be crystalline / solid, if traces of sulfate are present in the particle (Höpfner et al., 2019). Pure nitrate containing particles did not crystallize inside AIDA chamber at the ATAL temperatures. In the same paper, the ERICA-LAMS individual particle mass spectra showed that each nitrate particle that was collected in-situ, within the ATAL, indeed contained sulfate lines. Furthermore, the ATAL in-situ measurements of the GLORIA instrument of KIT, which were performed simultaneously with those of the ERICA, were indicative of solid nitrate particles and can be directly connected to the AIDA laboratory findings. Using this combination of GLORIA, ERICA-LAMS, and AIDA measurements, not only the particle chemical composition, but also the particle phase can be identified. This would not have been possible with the ERICA-AMS alone this.

Thirdly, the ERICA-LAMS typically detects particles with sizes above 100 nm, while the ERICA-AMS particle size range extends down to 60 nm. This way, young particles resulting from post-nucleation growth can be found by the ERICA. Moreover, the ERICA-AMS is only capable of analysing non-refractory components like sulfate, nitrate, organics and particulate ammonia, while the ERICA-LAMS provides additional information on components like mineral dust, metals, soot, and other refractory materials.

In conclusion, the combination of the two particle mass spectrometry methods, when sampling identical air parcels, substantially extends the information in terms of size range and chemical composition information. For the same reason, two separate mass spectrometers, one based on LDI and the other on TD-EI, have been deployed in both ground and aircraft missions, as discussed in Lines 67-69. Moreover, the UT/LS findings discussed above are based on in-situ measurements exclusively performed with the M-55 Geophysica aircraft, which payload limitations would not have allowed the deployment of two separate mass spectrometers.

We believe that the aforementioned facts demonstrate the advantages of the ERICA when compared and contrasted to other aircraft-ready instruments and combinations thereof. More details on the complementary nature of the common application of both methods are described in the companion paper of Hünig et al. (2022).

Changes: We believe these topics are adequately discussed in the companion paper of Hünig et al. (2022), as well as in Höpfner et al. (2019) and Appel et al. (2022). In addition, we have added the following text to the Summary (circa Line 739): *“This comprehensive set of data was the result of the simultaneous and complementary operation of the ERICA-LAMS and the ERICA-AMS, which otherwise, would have only been possible with the deployment of two separate instruments on board”*.

b)

Referee comment: *What fraction of the particles that enter the inlet trigger an ERICA-LAMS spectrum, compared to the particle mass that is detected by the ERICA-AMS part of the instrument during these high altitude flights? Is there a notable improvement in the quality of data obtained by this instrument above and beyond that afforded by other aircraft-ready instruments? The easy answer is an*

unqualified “yes” – but the answer remains to be demonstrated. There could certainly be devils in the details, and it would be helpful to evaluate the limitations of the instrument in the UT/LS environment.

Answer: This topic is discussed in detail in Section 2.5 of the companion paper of Hünig et al. (2022). Therein, it is estimated that for particle concentrations of 100 per cm³ and for an overestimated hit rate of 100%, only 5.4% of the sampled particles result in LAMS spectra due to the maximum laser pulse repetition frequency. In this case, only a small fraction of the sampled particles is excluded from the ERICA-AMS quantitative analysis. At the lower extreme, i.e. in remote environments like the Arctic with particle concentrations as low as 5 per cm³, a significant fraction of the sampled particles can be ablated by the ERICA-LAMS, significantly influencing the measurement on the ERICA-AMS. Only in this occasion, the deployment of two separate instruments may be advantageous. However, the particle concentrations experienced during the deployment of the instrument in the UT/LS were substantially higher.

To answer your question, a precise calculation of this fraction is indeed very difficult to be made. This has been one of the main reasons that have led to the recent development of a new measurement technique, as mentioned above and also discussed in Section 5. In this new mode of operation, the ERICA-LAMS measurements are only performed during the half-cycle, in which the shutter of the ERICA-AMS is closed to allow for background measurements. In the other half-cycle, the shutter is open and the ablation laser is inactive ensuring that the ERICA-AMS measurements are totally unaffected by the ERICA-LAMS.

Changes: We suggest making no changes to the text, considering that these topics have been covered in the companion paper by Hünig et al. (2022).

Replies to comments of Referee 3:

a)

Referee comment: *Calculated particle transmission efficiency is shown in Figure 3, but were there any laboratory tests of transmission efficiency performed? It would be difficult to test the aircraft inlet without a wind tunnel, but what about the instrument inlet? It might have been published elsewhere, but it is important enough to merit description here as well.*

Answer: The particle transmission of the employed constant pressure inlet (CPI) system was experimentally studied in Molleker et al. (2020), as mentioned in Line 324. This study covers a wide range of inlet pressures from sea level down to 65 hPa. For the aerosol sampling line, as discussed in Section 3.2, we have calculated its transmission efficiency based on the study from von Der Weiden et al. (2009), using the same software tool. Therefore, we are confident that the tube transmission losses presented in Fig. 3 are realistic. The only missing contribution to particle losses is the aspiration efficiency of the aerosol inlet intake, which indeed, would require wind tunnel tests.

Changes: No changes have been made, as this topic is covered in detail by Molleker et al. (2020) and this reference is given in the text.

b)

Referee comment: *Similarly, it is stated that 60 nm is considered to be the limit of detection for ERICALAMS, and it would be good to see some data from the optical transmission tests that were used to arrive at this number.*

Answer: We would like to clarify that 60 nm is the lower particle size limit of the ERICA-AMS. The size range of the detected and ablated particles of the ERICA-LAMS is discussed in Section 4.3 (i.e. 92 nm to 4186 nm). Detailed information about the optical measurements is given in Section 3.2 of the companion paper by Hünig et al. (2022), and in Molleker et al. (2020).

Changes: To avoid confusion, we have rephrased the sentence in Line 312 to “*At the lower end, where a size of 60 nm has been found to be the detection limit of the ERICA-AMS, the tube transport losses are below 5%*”.

c)

Referee comment: *The manuscript covers a lot of ground on best practices of ERICA deployment on aircraft, but it would be useful to also describe the calibration protocols and frequency during these aircraft campaigns.*

Answer: Regarding the calibration procedures, the optical components of the ERICA-LAMS were aligned, adjusted and characterized before and after each field campaign. The ERICA-AMS is typically calibrated every 2-3 flights by performing a single-ion-signal measurement and a total ionization efficiency calibration. Moreover, thanks to the telemetry system of the instrument, continuous monitoring of all its critical parameters was carried out by ground operators during each flight, regardless of its autonomous operation.

Changes: We have added this information to Section 4.1 (circa Line 581).

d)

Referee comment: *For Figure 8, the distribution of particle sizes sampled, it would be particularly useful to know the inlet and optical system transmission curves to assess how representative of the actual aerosol population this size distribution is.*

Answer: There are four factors contributing to the overall size distribution of the particles which ultimately yield spectra. These are: (a) the sampling line transmission, (b) the transmission of the constant pressure inlet (CPI), (c) the optical detection efficiency, and (d) the so-called hit rate (i.e. ratio of recorded spectra over the number of ablation laser shots). From the factors, the prevalent and most size-dependent one is the hit rate (factor d), for which detailed information is shown in Fig. 8 in the companion paper by Hünig et al. (2022).

The remaining factors mostly affect the size ranges above one micrometre or below 100 nm; this assumption is confirmed by the sampling line transmission (factor a) data shown Fig. 3, as well as by the CPI transmission measurements (determined by factors b and c), which are given in Fig. 8 of Molleker et al. (2020).

Changes: The first paragraph of this answer has been added to Section 4.3 (circa Line 635).

e)

Referee comment: *What other instruments were co-deployed with ERICA during the two field campaigns described? It would be very useful to see an intercomparison with a co-located size distribution measurement, such as a UHSAS, to quantify any sampling biases and losses.*

Answer: Regarding your question on the instruments deployed together with the ERICA during the StratoClim project, M-55 Geophysica was equipped with a comprehensive set of gas-phase, aerosol and remote-sensing instruments in all flights. A list of relevant publications is given in a special issue of ACP/AMT (Stratoclim). Unfortunately, a project overview paper remains to be published.

Addressing your specific question on particle size distribution, a UHSAS, specially modified for high-altitude measurements, indeed operated during those flights (Mahnke et al., 2021). Nevertheless, we believe that a comparative study between the UHSAS measurements and those of the optical detection system of the ERICA-LAMS extends beyond the scope of this paper. However, they such comparison definitely be included in a future study. Furthermore, it is worth underlining that the optical detection system of the ERICA-LAMS measures the vacuum aerodynamic diameter (d_{va}). This quantity differs from the optical diameter (d_{opt}) measured by the UHSAS, which calculation also depends on assumptions about the refractive indices of the sampled particles.

Changes: No changes have been made in this case.

Additional changes

1. The reference to the preprint from Hünig et al. (2021) has been replaced by a reference to the published companion paper (Hünig et al., 2022).
2. Figure S2 has been added to the Supplement. As a result, the numbering of the figures in the Supplement has been changed.

References

- Allen, J. O., Fergenson, D. P., Gard, E. E., Hughes, L. S., Morrical, B. D., Kleeman, M. J., Gross, D. S., Gälli, M. E., Prather, K. A., and Cass, G. R.: Particle Detection Efficiencies of Aerosol Time of Flight Mass Spectrometers under Ambient Sampling Conditions, *Environmental Science & Technology*, 34, 211-217, 10.1021/es9904179, 2000.
- Appel, O., Köllner, F., Dragoneas, A., Hünig, A., Molleker, S., Schlager, H., Mahnke, C., Weigel, R., Port, M., Schulz, C., Drewnick, F., Vogel, B., Stroh, F., and Borrmann, S.: Chemical analysis of the Asian Tropopause Aerosol Layer (ATAL) with emphasis on secondary aerosol particles using aircraft based in situ aerosol mass spectrometry, *Atmos. Chem. Phys. Discuss.*, 2022, 1-37 [preprint], 10.5194/acp-2022-92, 2022.
- Drewnick, F., Hings, S. S., DeCarlo, P., Jayne, J. T., Gonin, M., Fuhrer, K., Weimer, S., Jimenez, J. L., Demerjian, K. L., Borrmann, S., and Worsnop, D. R.: A New Time-of-Flight Aerosol Mass Spectrometer (TOF-AMS)—Instrument Description and First Field Deployment, *Aerosol Science and Technology*, 39, 637-658, 10.1080/02786820500182040, 2005.
- Höpfner, M., Ungermann, J., Borrmann, S., Wagner, R., Spang, R., Riese, M., Stiller, G., Appel, O., Batenburg, A. M., Bucci, S., Cairo, F., Dragoneas, A., Friedl-Vallon, F., Hunig, A., Johansson, S., Krasauskas, L., Legras, B., Leisner, T., Mahnke, C., Mohler, O., Molleker, S., Muller, R., Neubert, T., Orphal, J., Preusse, P., Rex, M., Saathoff, H., Stroh, F., Weigel, R., and Wohltmann, I.: Ammonium nitrate particles formed in upper troposphere from ground ammonia sources during Asian monsoons, *Nat Geosci*, 12, 608-+, 10.1038/s41561-019-0385-8, 2019.
- Hünig, A.: Development, characterization, and first field deployments of a novel aerosol mass spectrometer combining laser ablation and flash vaporization techniques for aircraft application at high altitudes, *FB 09 Chemie, Pharmazie u. Geowissenschaft, Johannes Gutenberg-Universität Mainz, Mainz*, 10.25358/openscience-5554, 2021.
- Hünig, A., Appel, O., Dragoneas, A., Molleker, S., Clemen, H. C., Helleis, F., Klimach, T., Köllner, F., Böttger, T., Drewnick, F., Schneider, J., and Borrmann, S.: Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques, *Atmos. Meas. Tech. Discuss.*, 2021, 1-43 [preprint], 10.5194/amt-2021-271, 2021.

- Hünig, A., Appel, O., Dragoneas, A., Molleker, S., Clemen, H. C., Helleis, F., Klimach, T., Köllner, F., Böttger, T., Drewnick, F., Schneider, J., and Borrmann, S.: Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques, *Atmos. Meas. Tech.*, 15, 2889-2921, 10.5194/amt-15-2889-2022, 2022.
- Mahnke, C., Weigel, R., Cairo, F., Vernier, J. P., Afchine, A., Krämer, M., Mitev, V., Matthey, R., Viciani, S., D'Amato, F., Ploeger, F., Deshler, T., and Borrmann, S.: The Asian tropopause aerosol layer within the 2017 monsoon anticyclone: microphysical properties derived from aircraft-borne in situ measurements, *Atmos. Chem. Phys.*, 21, 15259-15282, 10.5194/acp-21-15259-2021, 2021.
- Molleker, S., Helleis, F., Klimach, T., Appel, O., Clemen, H. C., Dragoneas, A., Gurk, C., Hunig, A., Köllner, F., Rubach, F., Schulz, C., Schneider, J., and Borrmann, S.: Application of an O-ring pinch device as a constant-pressure inlet (CPI) for airborne sampling, *Atmos Meas Tech*, 13, 3651-3660, 10.5194/amt-13-3651-2020, 2020.
- Pratt, K. A., Mayer, J. E., Holecek, J. C., Moffet, R. C., Sanchez, R. O., Rebotier, T. P., Furutani, H., Gonin, M., Fuhrer, K., Su, Y., Guazzotti, S., and Prather, K. A.: Development and Characterization of an Aircraft Aerosol Time-of-Flight Mass Spectrometer, *Analytical Chemistry*, 81, 1792-1800, 10.1021/ac801942r, 2009.
- Reilly, P. T. A., Lazar, A. C., Gieray, R. A., Whitten, W. B., and Ramsey, J. M.: The Elucidation of Charge-Transfer-Induced Matrix Effects in Environmental Aerosols Via Real-Time Aerosol Mass Spectral Analysis of Individual Airborne Particles, *Aerosol Science and Technology*, 33, 135-152, 10.1080/027868200410895, 2000.
- Schmale, J., Schneider, J., Jurkat, T., Voigt, C., Kalesse, H., Rautenhaus, M., Lichtenstern, M., Schlager, H., Ancellet, G., Arnold, F., Gerding, M., Mattis, I., Wendisch, M., and Borrmann, S.: Aerosol layers from the 2008 eruptions of Mount Okmok and Mount Kasatochi: In situ upper troposphere and lower stratosphere measurements of sulfate and organics over Europe, *Journal of Geophysical Research: Atmospheres*, 115, 10.1029/2009JD013628, 2010.
- Schulz, C., Schneider, J., Amorim Holanda, B., Appel, O., Costa, A., de Sá, S. S., Dreiling, V., Fütterer, D., Jurkat-Witschas, T., Klimach, T., Knote, C., Krämer, M., Martin, S. T., Mertes, S., Pöhlker, M. L., Sauer, D., Voigt, C., Walser, A., Weinzierl, B., Ziereis, H., Zöger, M., Andreae, M. O., Artaxo, P., Machado, L. A. T., Pöschl, U., Wendisch, M., and Borrmann, S.: Aircraft-based observations of isoprene-epoxydiol-derived secondary organic aerosol (IEPOX-SOA) in the tropical upper troposphere over the Amazon region, *Atmos. Chem. Phys.*, 18, 14979-15001, 10.5194/acp-18-14979-2018, 2018.
- StratoClim stratospheric and upper tropospheric processes for better climate predictions (ACP/AMT inter-journal SI): https://acp.copernicus.org/articles/special_issue1012.html, last
- Voigt, C., Schreiner, J., Kohlmann, A., Zink, P., Mauersberger, K., Larsen, N., Deshler, T., Kröger, C., Rosen, J., Adriani, A., Cairo, F., Donfrancesco, G. D., Viterbini, M., Ovarlez, J., Ovarlez, H., David, C., and Dörnbrack, A.: Nitric Acid Trihydrate (NAT) in Polar Stratospheric Clouds, *Science*, 290, 1756-1758, doi:10.1126/science.290.5497.1756, 2000.
- von der Weiden, S. L., Drewnick, F., and Borrmann, S.: Particle Loss Calculator - a new software tool for the assessment of the performance of aerosol inlet systems, *Atmos Meas Tech*, 2, 479-494, DOI 10.5194/amt-2-479-2009, 2009.