EGUsphere, author comment AC2
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Reply on RC2

Antonis Dragoneas et al.

Author comment on "The realization of autonomous, aircraft-based, real-time aerosol mass spectrometry in the upper troposphere and lower stratosphere" by Antonis Dragoneas et al., EGUsphere, https://doi.org/10.5194/egusphere-2022-33-AC2, 2022

Dear Referee 2,

thank you for your overall positive feedback and your suggestions. We are addressing your comments below.

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 cm^3 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 cm^3, 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).

References

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