Review of *Design, characterization, and first field deployment of a novel aircraft-based aerosol mass spectrometer combining the laser ablation and flash vaporization techniques* by *Hünig et al.*

Anonymous Reviewer

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1 Summary

In this work, Hünig et al. describe, for the first time, the design and characterization of ERICA. At the time of this review, ERICA is a unique instrument, but it does combine two well-known methods: (1) single-particle mass spectrometry using laser ablation to (partially) vaporize single particles and ionize their constituents, and (2) an AMS-style instrument that flash vaporizes the non-refractory component of aerosol using a hot tungsten filament and creates ions using electron impact. Method 1 will be referred to as ERICA-LAMS, and Method 2 will be referred to as ERICA-AMS, per the authors' designation. ERICA LAMS uses two time-of-flight mass spectrometers to analyze the positive and negative ions from a single particle; ERICA-AMS uses a compact-time-of-flight mass spectrometer to analyze positive ions. Both ERICA-LAMS and ERICA-AMS share a common aerosol focusing inlet (AFI), which is pressure-controlled and has been written about in a separate publications (Molleker et al., 2020). After exiting the AFI, the particles are sized by measuring the particle time-of-flight between two particle detection units (PDU1 and PDU2). Optical sizing was experiementally achieved for PSL between 80 nm and 5.145 μm Particles detection by PDU2 triggers a 266-nm quadrupled Nd:YAG ablation laser to fire (max repetition rate 8 hz^{-1}, $\sim$ 4 mJ/pulse). Particles that are not detected by PDU2 or are missed by the ablation laser are collected $\sim$55 cm from the exit the AFI, and $\sim$30.1 cm downstream from the ablation laser spot.

The authors give much attention in the paper to the particle beam diameter and the effective laser / vaporizer diameters. All are fitted parameters, which are fitted to a convolution of two functions–a top hat function for the effective laser / vaporizer width and a 2D Gaussian function for the particle beam width. In ERICA-LAMS, the particle beam width ranges from $\sim$30-40 μm for 335 nm

AN particles to ∼100-200 μm for all particles >400 nm to >500 μm for 103 nm PSL. For particles $\geq$ 208 nm, the particle beam diameters are smaller than the effective laser diameters in PDU1 and PDU2. For ERICA-AMS, particles with diameters > 91 nm have particle beams smaller than the effective diameter of the vaporizer, which, unlike the effective laser diameters, is similar to the physical dimensions of the vaporizer (3.8 mm).

The most userful meaasured parameters in the paper are the detection efficiency (DE) and the abation efficiency (AE). The former measures the number of particles detected by the PDUs compared to a separate measurement of particles counts by a CPC or OPC; the latter is the number of particles that has mass spectra divided by the number of particles that trigger PDU2. The DE analysis shows that, under ideal conditions (*e.g.,* idea beam position, which changes as a function of size), the DE for PSL is above 0.6 for particles $\geq$ 208 nm; however, for real-world particles the DE is generally lower across all sizes measured. Finally, the AE for real-world urban particles was presented. The AE has a maximum value of 0.52 @ 218 nm; however, the authors also found that the AE is a steep function of size, and hovers around 10-20% for particles below ∼ 200 nm and above ∼ 300 nm.

The paper finished with some example laboratory particles, as well as some example particles and science from the first aircraft deployment.

Overall, this paper is very well written and very well thought out. The scope of the paper also fits very well within the scope Aerosol Measurement Techniques. At the time of this review, ERICA is a completely unique instrument; thus, a detailed description and characterization paper is well-timed and necessary for future publications. This reviewer only has a few comments, which are outlined below.

2 General Comments

1. Section 3.1–It is unclear to this reviewer if the "razor blade" is integrated into the system like the "knife edge" in the PALMS instrument. If so, it is also unclear if ERICA uses the knife-edge to re-position the papers during flight, where they might have moved due to vibrations from the aircraft.

2. Section 3.3: It is unclear to the reviewer if the "effective laser radius" being much larger than the physical dimensions of the laser is supported by Mie theory (as was done for 108 nm particles). Is this true? Is this akin to a "scattering cross section?" If so, the authors should support that with some calculations in the supplemental. Otherwise, the authors risk comparing the physical beam diameters to a laser diameter that is fitted (as opposed to measured) and perhaps physically unrealistic.

3. Fig.10: I am slightly confused how it is possible that PDU2 can have higher values than PDU1. Can the authors comment on this?

4. Section 3.4: Because ERICA has both an optical DE for PDU2 and an AE,

it would be helpful for the authors to explicitly show a DE for ablation. This wold help the readers understand biases in ERICA number fractions etc.

5. Section 4: Towards the end of the paper, the authors compare ERICA-LAMS data to ERICA-AMS data on the same plot. This caused this reviewer to of biases between the measurements that should be addressed before having a combined interpretation of the LAMS and AMS results. The major bias, as understood by this reviewer from the figures in this paper, is that the "number fraction" will be highly dependent on the size and composition of the particles present. These should somehow be weighted accordingly–by internal DE curves or by normalizing to external quantitative measurements. No discussion of this correction is present in the current manuscript–this reviewer strongly suggests that the authors address that in this manuscript, as it will affect all future work from this instrument.

3 Minor Comments

1. P2L40: Since each paper should stand on its own–a brief description of the Dragoneas paper should be described here. That way the reader does not have to download a separate paper to fully understand your methods.
2. P3L24: "A large fraction" here is largely meaningless without some general numbers or statistics.
3. P3L34: Is the lens and geometry in ERICA the same as the lens in XU et al.?
4. P8L29: At what aerosol concentration (number and volume / mass), does ERICA-LAMS affect ~30% of the particles? This should be spelled out for the reader? I assume it could affect some areas of the Upper Troposphere.
5. P10L18: That the aerodynamic diameters of AN are similar to PSL suggest that they are spherical and of similar density. This not entirely surprising because AN is notoriously difficult to effloresce; however, the authors state that effective laser radius for AN do not match PSL because the AN are non-spherical. Can you reconcile these two statements?
6. P13L32: I'm not sure that "$w_{0,dia}$" is not the most meaningful measurement for overlap. Unlike the signal in PDU1 and PDU2, the intensity of the ablation laser will be essential to the interpretation of the mass spectra–especially for large or coated particles. Thus, a measure of the overlap between the particle beams and where the ablation laser is sufficiently powerful is indeed important to report.
7. P13L35: I don't think I saw any evidence that the 80 nm and 5145 nm particles were ablated and detected by the MCP. Is this true? If so,

perhaps a AE_{max} could be shown for PSL particles much like DE_{max} was?

8. P18L21: This reviewer is not an AMS expert–but, as written, it sounds like all RIEs are relative to the nitrate IE. So, why does nitrate have an RIE of 1.1?

9. P19L9: As written, it is unclear if it is most desirous to have a "small air beam sample" over no air beam sample.

10. P20L34: Can an estimate of the UT and LT altitude / altitude ranges be added to Fig. 17?

11. P22L25: It seems to this reviewer that different removal rates of EC and C_{total} suggests that the particles are not well mixed–because they would then be removed at the same rates.

12. P22L36: Are these EC particles from coagulation? They seem quite high to be primary particles.

13. P23L5: The authors often differentiate the EREICA-AMS data by say "the non-refractory components." This is misleading because ERICA-LAMS also measure the non-refractory components.

14. Figures: It is really hard, especially with the errors bars to differentiate the filled circles from the filled squares. Perhaps switch to filled and open squares?

15. Figure 10: Using 50% of the max is a bit strange in this plot–it results in PDU1 having larger D50s than PDU2, which is counter-intuitive given that PDU2 has better detection efficiencies.

16. Figure 11: Can you make the right side of this plot a log-scale (and also possibly the left?). It is hard to see if you're getting spectra for any particles below $\sim$ 120 nm or above $\sim 1\ \mu m$.

17. Figure 12: Why do you have a large Na^+ peak in your PAH spectra? Is your mass scale possibly off?

4 Technical Comments

- P1l11: What does "ERC" stand for?
- P1L15: Perhaps "*The same* aerosol sample can be sampled with both methods *simultaneously*?
- P1L20,25,26: The acronyms ADL, B-ToF-MS an C-ToF-MS are defined here, but are not used again in the abstract. The abstract should generally stand alone, and therefore these acronyms can be omitted, but need to be defined at their first use in the main section of the paper.

- P1L36: You probably can delete the comma after "anthropogenic-".
- P2L19: Perhaps use "e.g.," instead of "beside others by."
- P13L33: This reviewer is not sure "However" is the right word here–this statement does not seem to be related to the previous sentence.
- P16L10: It is hard to understand ion peak threshold as currently described. It might be easier to understand by splitting this statement up into two or more sentences.
- P19L19: You can probably delete "especially" in this line.
- P22L25: The statement "within the limitations of the applied method" is parenthetical and needs commas around it.

References

Molleker, S., Helleis, F., Klimach, T., Appel, O., Clemen, H.-C., Dragoneas, A., Gurk, C., Hünig, 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, Atmospheric Measurement Techniques, 13, 3651–3660, https://doi.org/10.5194/amt-13-3651-2020, URL `https://amt.copernicus.org/articles/13/3651/2020/`, 2020.