The authors present the design and development of a mass spectrometry system for comprehensive measurement of aerosol composition, in which two commonly used techniques, single particle mass spectrometry (SPMS) and aerosol mass spectrometry (AMS) are combined in a single tandem instrument. The manuscript represents a substantial body of work that required considerable expertise in instrument design including differential pumped vacuum systems, optical particle detection and time-of-flight mass spectrometry (TOFMS). A substantial amount of data is presented to evaluate the instrument design. The subject matter is very suitable for this journal but some important issues need to be addressed in the content if this manuscript is to be used as an instrument characterisation reference for future publications.

Major Comments

Both instrument use TOFMS as an analyser. This should be introduced and the benefits explained. They both also use aerodynamic lens inlet. The main difference is with the ionisation techniques employed to achieve the desired measurement. The pros and cons to each technique and the consequences on the data should be developed in the introduction. Both techniques are hard ionisation that causes intense fragmentation that has to be dealt with in the data analysis. In the case of laser desorption ionisation (SPMS), this renders the measurements inherently non-quantitative for molecular ion species. The thermal desorption ionisation method used in the AMS method is only quantitative with careful calibration. The authors present some details of the mass calibration in terms or the relative ionisation efficiencies (RIE) of nitrate, sulphate, and ammonium using the same method used for the Aerodyne AMS family of instruments. This is where my first major concern with the work arises.

In various places throughout the document the authors state the ERICA-AMS is ‘similar’ in design to the Aerodyne AMS, but the similarity is not described nor are the differences. In fact, no detailed description of the vaporiser, ioniser and ion extraction optics is given. The Thermal Desorption ionisation technique (TDI) is not well understood and Quantitative nature of the Aerodyne AMS instrument is underpinned by a large body of publications and method development (See Jimenez 2016 and references therein). If the authors wish to convey these characteristics onto their instrument, they need demonstrate equivalence in the design, particularly regarding the geometry of the ionisation source and the incident particle beam.

This leads to the second point of major concern with this manuscript regarding the measurement/calculation the particle beam width. The method description is extremely difficult to follow in the current version of the document and it is impossible to get any sense of the error in the calculation. This needs to be addressed. The authors use a method in which the particle beam is tracked across optical detection system which is kept static, in a very similar method to that presented in Marsden 2016 (not cited here) with the LAAPTOF single particle mass spectrometer, an instrument with many common features to the ERICA LAMS. The results are quite different regarding the ratio of particle beam and detection laser beam width compared to the LAAPTOF. This may be due to a superior quality aerodynamic lens, but the result should be discussed with respect to LAAPTOF and other instrument design as this is an important factor in instrument design.

Finally, I have concerns about the dynamic range of the ion detection system in ERICA LAMS. The A/D has only 8bits if vertical dynamic range which equates to 3 orders of magnitude within spectrum signal. This is insufficient in the reviewers experience and will either produce excessive saturation of intense ion signals or the complete loss of minor signals depending on the gain setting. Can the authors comment on this in section 3.5.2?

Minor Comments

- Take care to make accurate definitions upfront in the introduction, and then stick to those definition throughout the document.
- Please check the correct use of commas throughout the document and avoid excessive paragraph length.
- The writing style changes part way through the document which is rather odd.

Introduction

Page 1 ln 35 Chemical composition measurements can provide…

Ln39 Comma after 'in situ' not required

Page 2, Ln 1 Define the 'pulsed laser technique' as 'single particle mass spectrometry (SPMS)'

Page2, Line 5 the correct term is 'Thermal Desorption (TD)' and should be used throughout the document.

Page2, Ln8 This sentence is a little muddled. Maybe replace 'previous' with 'former'?

Page2, Ln10 Froyd et al. (2019) demonstrates a method for quantifying particle classes, not absolute mass concentrations of specific ions. There is an important distinction.

Page2, Ln 11 Consider starting a new paragraph

Page2, Ln 30 Perhaps introduce the term 'tandem measurement'

Page2, Ln31 Replace 'repetition rate' with the term 'temporal resolution'

Page2, Ln37 'Tandem Instrument'?

Instrument Description

- I brief principal of operation required before getting into the detail. Both techniques are sampling to same particle beam with the ERICA AMS at the end of the particle path. The LDI is requires optical detection to size particles and trigger the pulsed laser part way along the path.

Page3, Ln12 More effort should be made to describe Fig1.

Page3, Ln12. Define LAMS and AMS in the introduction or consider changing to Laser desorption ionisation (LDI) and Thermal desorption Ionisation (TDI) therefor highlight the actual distinction between the two techniques.

Page3, Ln14 Why is a constant pressure inlet required? Should this have already been introduced as part of the challenges of aircraft measurement?

Page 3, Ln23 The term 'ion extraction' instead of acceleration would be more appropriate

Page3, Ln25 Some particles are partially vaporised. What happens to particle fragment and partly ablated material?

Page3, Ln28	Un-ablated particles do not pass through the B-TOF-MS section because they are not extracted.
Page3, Ln31	use ‘extracted’ instead of ‘injected.
Page3, Ln31	C-TOF-MS has not been properly introduced.
Page3, Ln31,	You have to be more specific than ‘Detectable particle size’ as that would appear to conflict the next sentence. Do you mean you get composition measurement from that size range?
Page3, Ln33	Xu 2017 describes the ACSM – please state that. Is it valid to assume the detectable particle size range is the same as the ACSM? This requires some discussion.
Page 3, Ln39	Consider putting the final paragraph of this section as part of the introduction.
Page4, Ln 30	Are the vacuum pressures measured or calculated? A schematic of the vacuum system would be helpful.
Page5, Ln15	How is the vacuum seal achieved on a movable assembly?
Page 5, Ln20	How do you know that the system collects 75% of the scattered light. Has this been modelled or measured?
Page6, Ln10	What shape beam profile is produced by the pulsed laser system. Is there variation in the power density with respect to position on the particle beam axis?
Page6, Ln29	8bits the effective dynamic range including the noise? This equates to around 3 orders of magnitude.
Page6, Ln30	The positive and negative ion signals are measured by separate detection systems. Whilst having different gain on each channel is beneficial, it does not actually increase the dynamic range of the A/D, nor the dynamic range within the spectra. This is misleading.
Section 2.5	The writing style changes to prose, which is rather odd.
Page 8, Ln1	Replace ‘serial configuration’ with ‘tandem configuration’
Section 2.6	Is the data for 5% reduction in particle mass on the AMS with LAMS switched on actually presented in this paper? Where?
Section 3.1	The detection laser beam waist (250um) is much smaller than particle beam, but much larger that the particle diameters. Particles can encounter very different laser fluence depending on their trajectory through the Gaussian profile, therefore the effective irradiance encountered cannot be calculated by diciding the laser power by the beam area. See Marsden et al 2018.

References

Jose L. Jimenez, Manjula R. Canagaratna, Frank Drewnick, James D. Allan, M. Rami Alfarra, Ann M. Middlebrook, Jay G. Slowik, Qi Zhang, Hugh Coe, John T. Jayne & Douglas R. Worsnop (2016) Comment on “The effects of molecular weight and thermal decomposition on the sensitivity of a thermal desorption aerosol mass spectrometer”, Aerosol Science and Technology, 50:9, i-xv, DOI: 10.1080/02786826.2016.1205728

Marsden, N., Flynn, M. J., Taylor, J. W., Allan, J. D., and Coe, H.: Evaluating the influence of laser wavelength and detection stage geometry on optical detection efficiency in a single-particle mass spectrometer, Atmos. Meas. Tech., 9, 6051–6068, https://doi.org/10.5194/amt-9-6051-2016, 2016.