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Atmospheric Measurement Techniques Discussions

Interactive comment on "Evaluating the influence of laser wavelength and detection stage geometry on optical detection efficiency in a single particle mass spectrometer" *by* Nicholas Marsden et al.

Anonymous Referee #1

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General comments The authors describe the introduction of a custom detection laser system to improve the detection efficiency of a single particle mass spectrometer LAAP-TOF. A numerical model was developed to predict particle detection efficiencies in dependence on various parameters and the results were compared to a data set of ambient measurements during a field campaign. The results of measurements suggest that particle hit efficiency directly correlates to scattering cross section of the particle depending on particle size, particle refractive index and wavelength of incident light.

The manuscript is not very easy to read because the authors use their chosen symbols instead of a written explanations. They expect that readers also have these sym-

bols in mind all time. E.g. L 353: "...particle beam profile would theoretically give E600nmDetect= 0.53..." -> "...particle beam profile would theoretically give an optical detection efficiency of 0.53..."

Some statements are inconsistent: 1. L 427: "Our initial laboratory studies with Detection System A showed that a low detection efficiency in the size range 300 − 800nm produced a sampling efficiency that was insufficient for the targeted application. However, the hit-rate with this set-up was exceptionally high (close to 1)..." but "A collimated beam maximises the active area of detection whilst minimising the stray light divergence at the light collection optics, but comes at the expense of slightly reduced hit-rates compared to a focussed beam system." (L 476) -> A more precise discussion is necessary and data should be shown, e.g. for "...slightly reduced hit-rates compared to a focussed beam system." (L 478) 2. Is it really an advantage to have higher detection efficiency for spherical particles with diameters between 500 and 800 nm instead of lower efficiency but a wider detection range? 3. What does it help that "The new laser system resulted in an order of magnitude improvement in sensitivity to spherical particles..."? But, in real world applications most of the particles are not spherical! 4. The authors should take into account the laser power density in the focal point in theirs discussions.

-> The presented calculations and data have to be revised and completed.

Specific comments: L 39: "...as well as the the probing of internal mixing state..." -> "...as well as the probing of internal mixing state..."

L 151: "...create a beam with a D4σ (second moment width) focal point diameter of 51.2μm..." and Figure 2, caption: "...a 405nm diode based system with a focussed beam (D4σ of 50μm)..." – Which value of focus diameter is correct?

L 207 and L 210: What is meant with "sampling efficiency of the LAAP-TOF"? The manuscript contains discussions about various efficiencies, but it has to be clearly defined here. Probably, the "hit rate", i.e. the ionization efficiency, is meant, but for

which operation mode?

L 209: Please use SI-units: “cc” -> “cm^3”

L 212: Please show the parameters for this calculation, e.g. the inlet flow?

L 247: “3.4 Modelling the optical detection geometry” -> “3.4 Modelling the optical detection geometry for collimated detection laser beams”

L260: “The general relationship between particle beam width σp and R in Eq. (2) was used to quantify EDetect.” -> “The general relationship between particle beam width σp and R in Eq. (2) was used to quantify EDetect for a certain particle diameter d.”

L 281: “σ = 0.10mm” -> “σd = 0.10mm”

Figure caption Figure 4 – L 270/274/276/281: Please be consistent using terms, e.g. “σd” <-> “σdetect” <-> ”σDetect“

Figure 7: is not good visible and too small.

L 345: “The resulting in particle density profile” -> “The resulting particle density profile”

L 348/349: “σ = 0.13mm” -> “σp = 0.13mm” – Where are the values of σp determined, at 1/e2? This has to be mentioned.

L 349: “detection stage 1(120mm from the lens exit), and σ = 0.26mm at detection stage 2 (240mm. . .” compared to Figure caption of Figure 7: “. . .rear detection stages at a distance of 115mm and 230mm from. . .” which are the correct values?

Figure caption Figure 7 – L 349: Please be consistent using terms, e.g. “detection stage 1” = “front detection stage”

L 359: What is the value of “σd” for this calculation?

L 364/368: “Figure8” -> “Figure 8”

L 374: “cut-off than a the narrow angles” -> “cut-off than the narrow angles”

L 378: “Figure 8B” -> “Figure 8 B”

L 270 and Appendix <-> L 367 and Figure 8: What is the correct name for R? – “active radius of detection” or “effective detection radius” or “active detection radius”

Figure 8: - font sizes of the 4 parts are different. - The indications for the 4 parts (A, B, C, D) are missed. - The indications for the 4 parts (A, B, C, D) should be consistent: NOT “a, b, c, d” in Figure caption - description of Y-axis: “active detection radius” or “active beam diameter”? - “signal-to noise” -> “signal-to-noise” - What is the meaning of System X2 and X10 in Figure 8 C? This should be explained in the text in more detail.

L 349: The authors should use “aerodynamic lens” instead of “lens” if the inlet system is meant and “optical lens” if focusing a laser beam is meant (-> see e.g. L 386) for more clearness.

Figure 8: - D is not discussed in the text. Please add an appropriate discussion. - Why do the authors use a laser power of 300 mW for modelling R at 532 nm? The laser power is originally 1 W. Did they measure the value of 300 mW at the exit of the collimation path?

L 396 to L 400: It is surprising that 250.000 particles are clustered in only three classes. -> Add the clustering conditions of the algorithm. -> Show the patterns of the three classes. -> Add the abundances of the classes as pie chart or table.

Figure 9, - X-axis: “dat” -> “date” - How was the “clustered particle number concentrations” (L 400) calculated? - What is the time resolution of this drawing? - Y-axis: “LAAP-Tof” -> “LAAP-TOF”

Figure 10: - What is the bin width of LAAP-TOF size distributions? - In the drawing “A” and “B” is used in Figure caption “a” and “b”. Please correct accordingly. - “RI” is not used in the text. Please use “refractive index”

L 416: “a” -> “at”

L 427 to 429: “Our initial laboratory studies with Detection System A showed that a low detection efficiency in the size range 300 − 800nm produced a sampling efficiency that was insufficient for the targeted application.” The authors should show their data for this statement.

L 440 to 442: “The particle size distribution of the target application must be considered when choosing the output power and focussing characteristics of the detection laser system if using light collection optics with a narrow collection angle.” This is hard to realize for ambient and field measurements because at the beginning of such measurements the size distribution is not known and it can vary during a measurement period. It would be helpful if the authors give a hint how to overcome this problem.

L 445: “the low Eion of some of the silicate mineral dust” – Do the authors have data for this statement? If so, the authors should show it. Usually, mineral particles have high ionization efficiency and mass spectra can be detected down to particle diameters of 200 nm. Therefore, Arizona dust particles are often used for laboratory test measurements.

L 446: “Size dependent detection bias may also influence the sampling efficiency” - Do the authors have data for this statement, e.g. size-resolved offline measurements with an impactor? Otherwise, the statement is speculative and should be deleted.

L 448: “Comparison with the LAAP-TOF size distribution with the APS suggests that the size distributions of the silicate and secondary classes represent the tail of an accumulation mode with a center below 300nm (Figure 10 A?).” – It is not very good visible. Maybe a logarithmic Y-axis gives better evidence. The value of “300 nm” is not shown.

L 503: “evaluation of the effective of particle size and morphology on the overall sampling efficiency”?

L 506: “ESizedMS could be improved by reducing the length of the particle flight path by

shortening the analyser housing.” - The authors should propose possible instrumental improvements.

References: References are sometimes incomplete and have to be revised substantially. The format should match the rules of the journal! e.g.: - Murphy, D. M.: Guest Editor : Albert Viggiano THE DESIGN OF SINGLE PARTICLE LASER MASS SPECTROMETERS, pp. 150–165, doi:10.1002/mas, 2007 -> doi:10.1002/mas.20113 and/or Mass Spectrometry Reviews, 2007, 26, 150– 165. -> reference of “guest editor” is unusual.

- Jonsson, H. H., Wilson, H. C., and Brock, R. G.: Performance of a focused cavity aerosol spectrometer for measurements in the stratosphere of particle size in the 0.06-2.0 um diamater range., American Meteorological Society, 1995. -> H.H. Jonsson, J.C. Wilson, C.A. Brock, R.G. Knollenberg, T.R. Newton, J.E. Dye, D. Baumgardner, S. Borrmann, G.V. Ferry, R. Pueschel, Dave C. Woods, and Mike C. Pitts: Performance of a Focused Cavity Aerosol Spectrometer for Measurements in the Stratosphere of Particle Size in the 0.06–2.0-μm-Diameter Range. Journal of Atmospheric and Oceanic Technology, 1995, 12:1, 115-129.

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