

*Evaluating the influence of laser wavelength and detection stage geometry on optical detection efficiency in a single particle mass spectrometer*

Dear Anonymous Referee #2,

Many thanks for the your suggestion on how to improve the flow and readability of our manuscript . Our response to your comments is given below.

On behalf of the authors,

Nick Marsden

The response to the review is structured as follows: The original reviewer comments are given in black, followed by the author response in blue font colour. Changes that will be made to the manuscript are then indicated in red.

Authors reply to review comments by Anonymous Referee #2:

The scientific approach is sound and the contribution important. However, the paper is really not easy to read, it is indeed a technical paper, but I think the flow should be improved. After major reorganization, it surely can be accepted for publication.

We acknowledge that the paper is difficult to read and will make changes to improve the readability and flow of the manuscript.

Reference list. I found the introduction and the overall reference list very poor. Given the >20 years of SPMS, I would have hoped for a more sound reference review. Papers like Prather and Moffet (PNAS 2009) should be included. I would make a short review of ATOFMS-SPMS studies (San Diego, Birmingham, recent Canada-Ireland work, Germany) on detection efficiencies and particle matrix effect and so on. It would definitely help this paper.

We accept that more referencing is needed in the introduction.

Reference to the direct measurement of the scattering cross section with optical detection in SPMS (Prather and Moffet 2009) will be made (page 2 L53).

New paragraph to be added (page 3 L79) that gives an overview of the qualitative nature of SPMS and how variation of instrument function affect particle counting statistics.

New paragraph to be added at (page 3 L79) that gives a brief overview of recent ATOFMS publications as examples of the type of application for SPMS.

The paper needs some bullet points or a better flow. Page 4 line 110-120, page 6 line 140-170, section 3.3 for example.

Instrument acquisition modes (Page 4 L110-120) will be written as bullet points.

The detailed descriptions of the build of optical detection systems (Page 6 L148-170) will be moved to an appendix to improve the flow. A table will be added (Page 6 L148) that summarises the key differences between the optical detection systems.

Explanation of the parameters in section 3.3 requires a narrative that does not lend itself to bullet points.

Title to Section 3.3 to be changed to 'Instrument Performance Definitions'

Figure 2. Perhaps explain better the original setting and the modified settings, and the consequences?

Text at page 6 L140 will be changed to concentrate on the important differences consequences of the two detection systems, with a table for easy reference.

I am not sure I follow the result section, especially section 4.1 and 4.2. Perhaps a paragraph introducing the results and the sections?

A new introduction to the Results section will be written. The results will be organised into a new set of subsections.

4.1 Instrument Performance Measurements

4.2 Particle Beam Width Measurements

4.3 Derivation of the Transfer Function

4.4 Model predictions of the effective detection radius with selected wavelengths, collection angles and signal-to-noise conditions.

4.5 Ambient Measurements

Figure 11. Is it appropriate at the end of the discussion?

This figure is referred to as part of the discussion of possible solutions to the particle detection bias.

Figure 11 will be moved to the middle of the discussion section.

In summary, I am convinced of the new improvement of the instrument and the results are sound. I think they can be better presented, both in the introduction (state of the art of SPMS), and a better organization of the scattered results difficult to follow. Scientific context is sound and accepted, a better presentation of the results is needed.

The paper at this stage is very difficult to follow and is not the easiest read.