The authors present a manuscript that describes the investigation of enhanced resonance ionisation as a means of improving the detection of metals in aerosol particles with single-particle mass spectrometry (SPMS). This is a very worthwhile objective, as such a technique could improve the online study of the distribution and sources of important metal species in the atmosphere. The authors focus on the resonance enhancement of Iron using laser desorption ionisation (LDI) at 248nm, which coincides with a major absorption line of Fe, which is an element of relevance in the study of micronutrients to the oceans and anthropogenic aerosol pollution. The physical basis for the techniques is sound and the work represents a substantial investment in resources to achieve a demonstrable improvement in detection of Fe along with Zn and Mn.

With the acceptation of some minor improvements in the reporting of the results, the technical merits of the manuscript is good. However there are some major concerns about the atmospheric relevance of the field study presented. Whilst this field data supports the technical development, it does not offer insight into the atmospheric implications of the presence of Fe in the environment in which it was measured. As this is a key requirement of this journal, the authors are requested to provide this discussion or consider submitting to a technical journal such as AMT.

Minor Comments:

L51 Most of the iron in mineral dust is in the form Fe2O3 (hematite) which is also insoluble. A distinction between soluble and insoluble Fe should be made as it affects bioavailiblity which is a major justification for carrying out this work.

L59 Replace ‘were’ with ‘have been’.

L61 Replace/remove ‘herein’, as it implies it is a feature if this study.

L63 Not all SPMS is bipolar, e.g. PALMS.

L64 Replace ‘typical ionization products’ with ‘typical observed ions’

L65 It would be useful to be more specific about which metals have been detected. Provide some references.

L68 The matrix effect will influence all compounds, not just the minor ones. The description of the matrix effect should be developed further.

L75 The description of LDI and REMPI are a little conflated. It would be easier to follow if they were described in separate paragraphs. It should also be clear when talking about the techniques generally, or specifically to SPMS. The reference Gunzer *et al* (2019) is a review paper not specific to SPMS for example.

L87 The conclusions of the present study and those of Schade et al (2019) should be clearly separated.

Experimental Section An introductory paragraph at the start of this section would be useful to set the scene for the experimental approached that follow. A better description of the instrument geometry should be provided. The authors reference an early technical paper for the Hexin instrument (Li *et al* 2011) but it is unclear what other modification have been made (if any) that could influence the instrument performance. Are any prior publication made with this instrument platform (e.g. Schade et al 2019)?

Results and Discussion A short introductory paragraph at the start of this section would also be helpful. The subsection titles should have some equivalence e.g. 3.2 Resonance enhancement of trace metals.

In the lab experiments, why were different OPO wavelengths used for the soot (250.0nm) and test dust (242.2nm)?

Figure1 What is the right hand axis of panel (B)?

L152 'remarkably width'

L182-186 Seems to be introductory material. Could this be moved to the introduction?

L190 The NIST reference material is described as 'well characterised'. A summary of this characterisation should be given e.g. from NIST certificate of analysis https://www-s.nist.gov/srmors/certificates/1649b.pdf

L200 'prove that not only the sum signals of the metals are higher in resonant case, but also more individual particles reveal their signatures.' This sentence is unclear and should be re-written with reference to limit of detection.

L213 Data that describes this instability should be provided.

Figure 4 Caption 'The two lasers fired alternately on 1500 particle each.'

Conclusion Some quantitative estimation of the enhancement achieved should be given. The trade-off between metal enhancement and lower particle detection rates should be highlighted.

References

Gunzer, F., Krüger, S., and Grotemeyer, J.: Photoionization and photofragmentation in mass spectrometry with visible and UV lasers, Mass Spectrom. Rev., 38, 202–217, doi:10.1002/mas.21579, 2019.

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Li, L., Huang, Z., Dong, J., Li, M., Gao, W., Nian, H., Fu, Z., Zhang, G., Bi, X., Cheng, P., and Zhou, Z.: Real time bipolar time-of-flight mass spectrometer for analyzing single aerosol particles, Int. J. Mass Spectrom., 303, 118–124, doi:10.1016/j.ijms.2011.01.017, 2011.