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Atmospheric Measurement Techniques Discussions

Interactive comment on "Laser Ablation ICP-MS of Size-Segregated Atmospheric Particles Collected with a MOUDI Cascade Impactor: A Proof of Concept" *by* Marin S. Robinson et al.

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The paper investigates " Laser Ablation ICP-MS of Size-Segregated Atmospheric Particles Collected with a MOUDI Cascade Impactor: A Proof of Concept" The topic of the manuscript is very interesting and not yet investigated in literature to the best of my knowledge. The manuscript is concise and well written and conclusion adequately supported by experimental data. I suggest publication in Atmospheric Measurement techniques journal pending minor revision as noted: The developed method is quite promising for analysis of elemental composition of size-segregated atmospheric particles collected on filters. The authors compared this method with the "wet chemical" ICP-MS. However, a comparison with other techniques from the literature to validate

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their method is missing. For example, how this technique proofs useful compared with some instruments aimed for online analysis of the elemental composition in single atmospheric particles, such as A-TOF-MS, for instance. A-TOF-MS is also based on laser desorption technique.

Authors' response: Based on the concentration levels found in the filters, validation with similar imaging techniques like XRF, PIXE, etc. is questionable as LA ICP-MS is the most sensitive technique for elemental microanalysis. However, by comparing the elemental concentration in size-segregated particles with a sensitive bulk analytical technique like ICP-MS, after digestion of the filter, an indirect comparison can be made as explained in section 2.4 and highlighted in Table 3. As such we feel that the LA ICP-MS imaging method used yields accurate and precise data. An instrument like A-TOF-MS is meant for the determination of single particle size and analysis of individual particles and refractory materials such as sodium chloride, elemental carbon and mineral dust constituents. As such specific classes of particles are identified based on fingerprinting or combining peaks but true elemental composition analysis as achieved with our approach is not possible. Although we restricted ourselves to a limited suite of elements, in theory most elements of the periodic table can be measured routinely.

Furthermore, the authors should better explain what are really the advantages and disadvantages of their method. Therefore, a discussion about atmospheric implications of this method should improve the quality of the manuscript and make it more interesting for the readers. I suggest including a section "Atmospheric implications".

Authors' response: In this proof of concept paper, we have demonstrated the usefulness of LA ICP-MS as a tool for analysing the elemental composition of size-segregated atmospheric particles collected on filter-based media and have addressed these issues in the Conclusions section. Previous problems associated with LA ICP-MS were addressed: (1) MOUDI rotation sampling overcomes the lack of uniformity in particle deposition, creating a sample highly suitable for LA-ICP-MS 2D mapping, 2) the 2D mapping mode yields results which show a high degree of accuracy when

larger areas are ablated and superior detection limits, and 3) quantification problems due to non-matrix matched standards are circumvented by ablating through the filter or obliterating the particles on the filters, warranting the reliable use of one-point calibrating on NIST SRM 2783. Together, these improvements allowed for an efficient and sensitive measurement of elemental composition. Compositional graphs of particles such as those shown in Fig. 4 for Ljubljana and Martinska, will be useful to the atmospheric community by allowing comparison of elemental profiles of particulate collected at diverse sites (e.g., urban industrial centres to remote background locations). Such profiles can be compared over days, months, or even years; short-term and long-term compositional changes can be used to monitor atmospheric changes such as a new pollution source, the impacts of pollution remediation, and the effects of climate change.

How was chosen the NIST standard? I this the best option for this kind of study? On which scientific basis were chosen the 9 elements in this study? On which basis the authors decided to cut the filters? How do we know for sure that the elements are homogenously distributed on the filters? I did not understand why some filters are ablated individually and some spot by spot? How was this decided?

Authors' response: We used NIST SRM 2783 as an absolute calibration standard as this seems to be the only "reliable" elements standard available for particulate matter although we are aware of the heterogeneity of the NIST standard on the microscale level as the certificate declares that a sampling area of 1 cm2 is deemed necessary for reaching the certified uncertainty. In the current manuscript we routinely analysed 1 cm2 to comply with these requirements. We have chosen the most important trace elements based on inhalation risks associated with particle-bound metals as explained in the introduction. Filters were not cut but we measured half-filters although measurement of areas of 1 cm2 would suffice for accurate analytical results. Since the laser samples a statistically significant portion of the filter we can be confident that heterogeneity issues are circumvented. Using the MOUDI with and without rotation, gener-

ating homogeneously distributed particles and “particle spots”, respectively, we show two approaches used for field collection of size-segregated particles and the limitations of both approaches, i.e. higher sensitivity and noisier data in the case of “particle spots” and lower sensitivity and better reproducibility in the case of homogeneously distributed particles.

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