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.

Anonymous Referee #2

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The paper dedicated to the analysis by LA ICP-MS of atmospheric particles collected with a MOUDI cascade impactor fit well with the scope of the AMT journal. However, many questions arise. The study which intended to demonstrate the value of this combination (MOUDI / LA ICP-MS) not treated and discussed a lot of keypoints that deserved the proof of concept.

The main problem comes with the lack of a real reference material characterized for the different particle sizes tested here. The urban particulate matter of the SRM 2783 used as reference in this study has a median particle size of 3.2 μm and a size range of 2.5 μm and particles are collected on nuclepore polycarbonate membrane filters of 0.4 μm pore size. How it can be possible for the authors to compare its response by LA

ICPMS with what they obtained on their particulate samples of lower sizes, collected on PTFE filters? This is not discussed (influence of the particle size and the filter type on the laser ablation efficiency / influence of the laser beam mask size on the laser ablation efficiency and the mapping...)

The authors claimed that "The laser beam energy was sufficient to remove all of the particles during ablation, allowing elemental concentrations to be determined via a one-point calibration with the NIST standard" (P3L12). The authors have to prove that it is realistic. What's about deep-impacted particles into filters? As they mentioned in their previous paper published in Science of the Total Environment in 2008 ("A multi-element mapping approach for size-segregated atmospheric particles using laser ablation ICP-MS combined with image analysis") distribution of particulate matter in SRM 2783 is non-homogeneous... Why is it not taking into account here?

LA ICP-MS and "wet chemical" ICP-MS of several samples had been also compared for 5 samples which is informative but non-sufficient to understand the effect of the particulate size on the LA ICP-MS response.

Particulate collection is another keypoint of this particulate matter analysis. However, collection particle losses during MOUDI collection was not investigated (nozzle wall loss which is dependent on the size but also on the particle composition / clogging effects). Moreover, the effect of the rotation of the filter was studied by comparing the results got from two different cities, at different days and different climatic conditions...too much parameters varied to get really confident comparison.

Moreover, no validation data are given on this new measurement system. Validation steps of the concept should be considered (repeatability integrating the collection step).

Why no conclusion was drawn on the proof of concept...?

I suggest conducting the proof of concept on a very well characterized particulate sample (size/composition by other analytical tools), to study systematically each critical pa-

rameter and compare with the author's previous work and the literature (one important paper in this field is missed: Hsieh, Chen et al. "Elemental analysis of airborne particulate matter using an electrical low-pressure impactor and laser ablation/inductively coupled plasma mass spectrometry" J.Anal.At.Spectrom., 2011, 26, 1502).

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