

Fast High Resolution 2D and 3D Laser Ablation–Inductively Coupled Plasma–Mass Spectrometry Imaging via Deconvolution Approaches and Ablation Cell Design

Stijn J.M. Van Malderen¹, Johannes T. van Elteren² & Frank Vanhaecke¹

¹ Department of Analytical Chemistry, Ghent University, Krijgslaan 281 - S12, B-9000 Ghent, Belgium

² Analytical Chemistry Laboratory, National Institute of Chemistry, Hajdrihova 19, SI-1000 Ljubljana, Slovenia

Corresponding author: Frank.Vanhaecke@UGent.be

Laser Ablation–Inductively Coupled Plasma–Mass Spectrometry (LA-ICP-MS) is a powerful tool to visualize the spatial distribution of nuclides on a μm -level over a solid sample in a wide variety of application fields. Practically all LA-ICP-MS imaging studies focusing on high lateral resolution produce an elemental map by ablating the material under investigation according to a grid of distinct positions, and relating the isolated response of each ablation position to a corresponding pixel. The lateral resolution achievable with this approach is limited by the beam waist dimensions of the focused laser beam, which is either restricted by the diffraction limit in the far field or the sensitivity available for the targeted nuclides. The speed of data acquisition and sensitivity are often limited by the dispersion of the aerosol. This fundamental study pursues a combined strategy of ablation cell development and (pre- and post-) acquisition methodology to overcome these limitations, thus enhancing the sensitivity, lateral resolution and throughput capabilities of LA-ICP-MS beyond state-of-the-art¹. The laser ablation cell and transport assembly developed² in-house are characterized by a fluid dynamic design favourable to aerosol transport, enabling discrete pulse responses, resolved via signal deconvolution, at laser pulse repetition rates up to 200-300 Hz. Overlapping ablation positions inside a 2D or 3D sampling grid results in an image or volume in which the nuclide distribution is artificially convolved with the point spread function (PSF) of the LA system. Image deconvolution was applied to reconstruct the nuclide distribution such that features below the physical size of the laser beam become resolved. The PSF is derived from atomic force microscopy topography maps of single pulse craters, allowing the approach to account for the laser beam shape and aberrations, and the laser-solid interaction, which in turn enhances the lateral resolution of the reconstructed volume.

(1) Giesen, C.; Wang, H. A.; Schapiro, D.; Zivanovic, N.; Jacobs, A.; Hattendorf, B.; Schuffler, P. J.; Grolimund, D.; Buhmann, J. M.; Brandt, S.; Varga, Z.; Wild, P. J.; Gunther, D.; Bodenmiller, B. *Nature methods* **2014**, *11*, 417-422.

(2) Van Malderen, S. J. M.; van Elteren, J. T.; Vanhaecke, F. *J. Anal. At. Spectrom.* **2015**, *30*, 119-125.