Supporting information

Performance of an experimental high-repetition rate laser head (500 Hz) for HR LA-ICP-QMS imaging

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To test the analytical sensitivity of the 500 Hz max. repetition rate laser head in our modified LA instrument, we performed line scans on NIST SRM 612 reference glass with increasing repetition rates and beam sizes. 10, 20, 50, 100, 200, 300, 400 and 500 Hz repetition rates were used and 5, 10, 20 and 35 µm square shape laser beams. LA shot dosage of 20 was used and stage travel speed was automatically adjusted by LA software.

Larger beam sizes than 35 µm were not used as further increasing the beam size dramatically increases the amount of ablated aerosol in the cup of the ablation cell and in ARIS transfer line. This could potentially cause particle saturation and roll-off of the observed signal intensity (re-deposition on sample surface, deposition on cup walls and window, transfer tubing).

Following m/z$^+$ were recorded by ICP-MS: $^{7}Li^+$, $^{89}Y^+$, $^{139}La^+$, $^{232}Th^+$, $^{238}U^+$ and $^{254}UO^+$. Spectrum acquisition mode (6 repetitions) was used. U^+/Th^+ ratio was monitored to observe the effect of repetition rate on laser induced elemental fractionation, UO^+/U^+ was used to monitor rate of oxide formation.

Figure S1 presents signal intensities for measured elements vs. repetition for 10 µm square beam. We see linear increase of signal intensities with increasing repetition rates ($R^2 > 0.99$) for all nuclides measured, proving the possibility to use the full repetition rate range of the laser head, up to maximum of 500 Hz.

Signal intensities, elemental fractionation and oxide formation vs. repetition rates behave analogously in case of all other laser beam sizes we tested, proving that up to 35 µm square laser beam firing at 500 Hz no ill-effects (aerosol saturation, elemental fractionation, and oxide formation) could be observed.

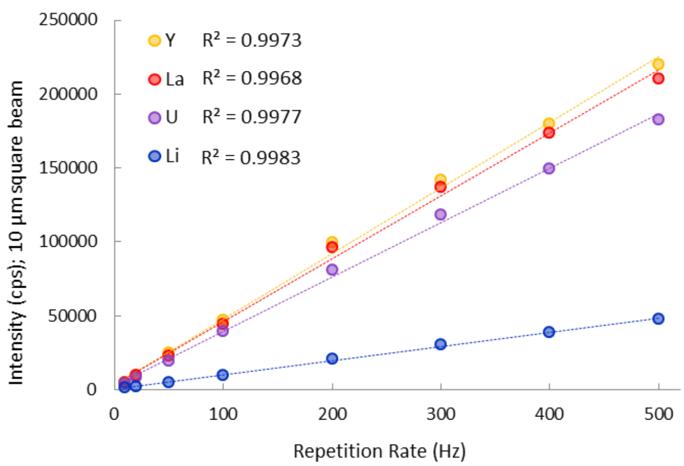

Figure S1: Elemental intensities vs. repetition rate (beam size, 10 µm [square mask]).

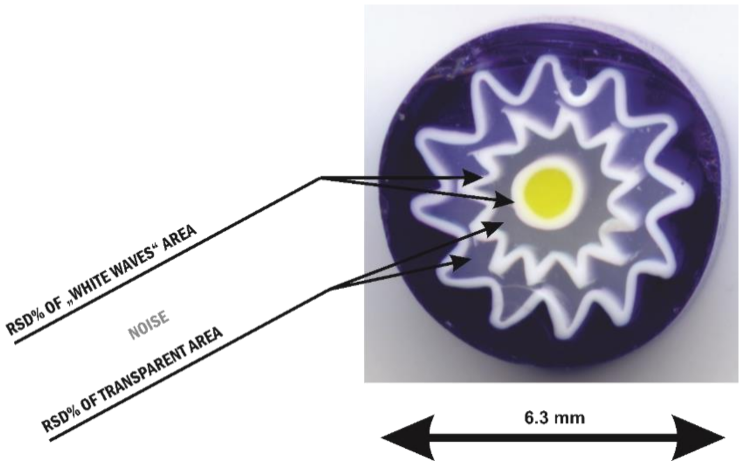

Figure S2: Murrina sample used in the LA-ICPMS imaging experiments with areas indicated where noise, defined by the RSDs in the elemental maps, was measured by ImageJ.

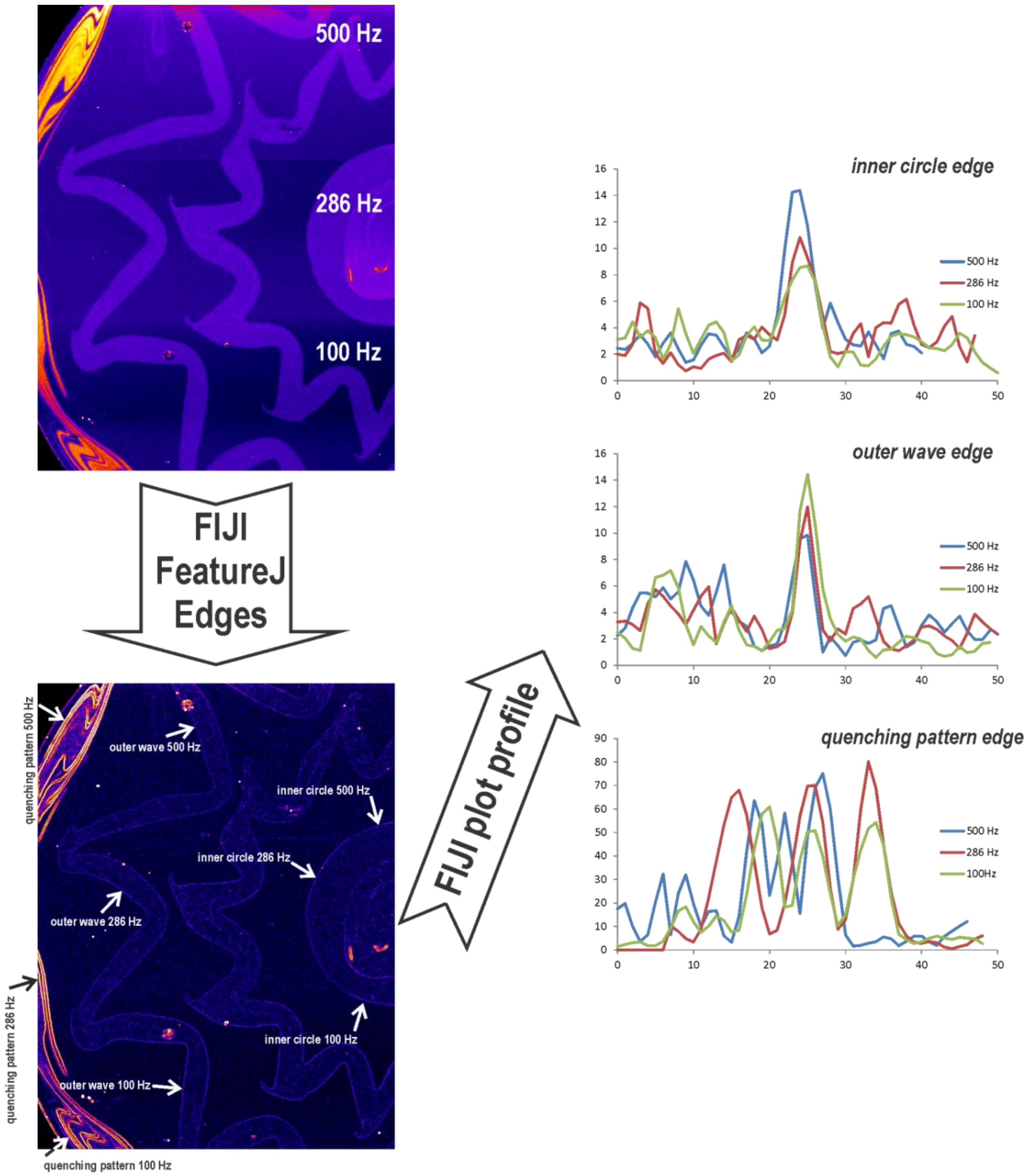

Figure S3: Image quality testing for three imaging conditions used (500, 286, 100 Hz) using FeatureJ Edges plugin in FIJI (ImageJ).

Image quality of three sub-areas (500, 286, 100 Hz) was tested by inspection of sharpness of the edges of sample's features. The original LA-ICP-MS image (top left) was converted to "edges" image by FIJI (ImageJ) by FeatureJ Edges plugin. The converted "edges" image (bottom left) is a sort of derivative of the original image. Plotting the profile of the edge on various locations of the edges image where distinct Ba patterns could be found (inner circle, outer wave, quenching pattern area), tests the width of the edge that is representing the sharpness of the original image. As it can be seen on the plotted profiles (right), there is not much difference in edge profiles for three imaging conditions, proving that in all cases images of equally high sharpness (quality) could be obtained.

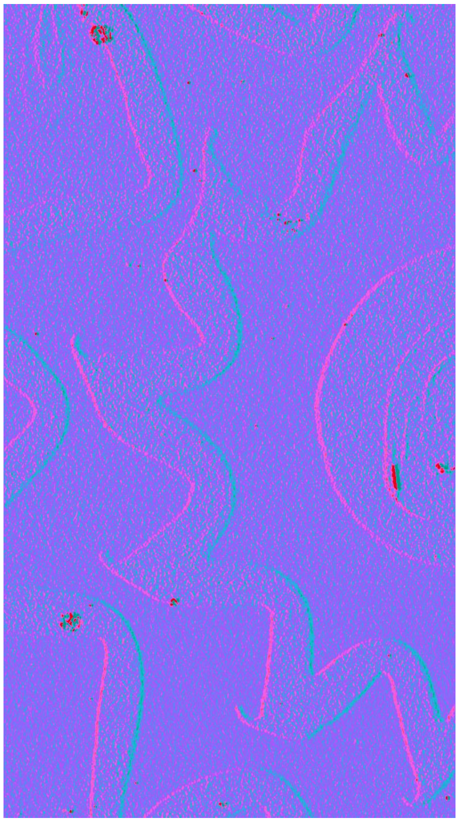

Figure S4: Image quality testing for three imaging conditions used (500, 286, 100 Hz) FeatureJ Derivatives plugin in FIJI (ImageJ).

Another approach for the image quality of three sub-areas (500, 286, 100 Hz) was tested by inspection of sharpness of the edges of sample's features. The original LA-ICP-MS image (top left Figure S3) was converted to "first derivative" image by FIJI (ImageJ) by FeatureJ Derivatives plugin. The edge blur from the derivatives of the sub-images, showing up gradients (red) and down-gradients (blue)shows no discernible differences in blur in the three subsections of the image.