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Evaluating the Accuracy, Precision and Resolution Limits of LA-ICP-MS Measurements of Multiple Me/Ca Ratios in Carbonates with Matrix-matched Calibration Standards

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As one of the primary climate archives, carbonates in different geological settings can record multiple facets of the climate system with their isotopic and elemental compositions. Geochemical measurements of carbonates at high spatial resolution have the potential to generate climate records at annual, seasonal or even higher temporal resolution, yet such measurements are often costly and limited in analytical precision. The accuracy of *in-situ* measurements of trace elements in carbonates can be additionally complicated by the availability of homogeneous and matrix-matched calibration standards. Here we report a systematic evaluation of the accuracy, precision and resolution limits of LA-ICP-MS measurements of multiple element-to-calcium (Me/Ca) ratios in a series of commercially available carbonate standards that are made with homogenized nano-pellets of natural coral, foraminifera, clam, speleothem and limestone materials. The measurements were performed on an ESL-NWR 213nm laser ablation system coupled to a Perkin Elmer NexIon 5000 ICP-MS instrument. We find that the analytical uncertainties of most elements of interest can be predicted by Poisson statistics with a laser ablation energy of ~3 J/cm^2 and repetition rate of 5 Hz. While the analytical uncertainties of single laser spots can be relatively high (~20% for 5 μm spots, ~1% for 100 μm spots), integrating over multiple replicates of a spot and averaging multiple spots from the same standard can significantly improve reproducibility. We also find it necessary to measure isotopes of higher abundance for different elements (e.g. ^{24}Mg, ^{88}Sr, ^{138}Ba) when laser spots are smaller than 10 μm to maintain reasonably accurate calibrations, while lower abundance isotopes (e.g. ^{25}Mg, ^{86}Sr, ^{137}Ba) are sufficient for larger spots. Given the range of Me/Ca ratios in different standards, relatively accurate calibrations can be established for Mg/Ca, Sr/Ca and Ba/Ca at 5 μm resolution, for Li/Ca, Na/Ca, K/Ca at 10 μm resolution, for B/Ca, P/Ca, Mn/Ca, Fe/Ca, Nd/Ca, U/Ca at 15 μm, and for Cd/Ca at 20 μm and above. Our matrix-matched calibration method offers the potential to better characterize the elemental heterogeneities of natural carbonates and generate sub-annual resolution paleoclimate records with a relatively low-cost LA-ICP-MS setup.