Extreme isotopologue disequilibrium in molecular SIMS species during SHRIMP uranium-lead geochronology

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Isotopologue deficits of up to 200 ‰ are observed in UO_2^+ species during SIMS gechronological analyses using the SHRIMP II instrument. These are most easily identified by bombarding natural U-bearing minerals with an $^{18}O_2^-$ primary beam. The large anomalies are associated with repeat analyses down a single SIMS sputtering crater, analysis of high-uranium, radiation damaged zircon, and analysis of baddeleyite. Analysis of zircon under routine conditions yield UO_2^+ isotopologue anomalies generally within error of equilibrium. As the conditions under which the isotopologue anomalies are observed are also conditions in which the UO_x-based corrections for relative U vs Pb ionization efficiencies fail, the existence of these isotopologue anomalies suggest that failure of the various UO_x species to equilibrate with each other is the reason that none of them will successfully correct the U/Pb ratio. No simple isotopologue-based correction is apparent.

Isotopologue abundances for UO^+ and ThO^+ and their energy spectra are consistent with most or all molecular species being the product of atomic recombination when the primary beam impact energy is greater than 5.7 kV. This, in addition with the large UO_2^+ instrumentally generated isotopologue disequilibria, suggests any attempts to use SIMS to detect naturally occurring isotopologue deviations could be tricky.