

Silicon isotopes in ancient zircons

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Isotopically heavy oxygen in Hadean zircons implies the existence of material reworked by low-temperature fluid-driven weathering as far back as 4.3 Ga [1]. Here, we apply this concept to the silicon isotope composition of zircons. As with O isotopes, mantle melting and magmatic differentiation both have a limited effect on Si isotopes [2], while chemical weathering generates larger Si isotope fractionation, whereby secondary minerals are much lighter than protolith [3]. The key difference between O and Si is that Si isotopes in bulk sediments are more robust during weathering; only material that has experienced significant chemical denudation shows resolvable Si isotope offsets from primary igneous compositions [4]. Hence, zircons crystallising from ‘S-type’ melts should only be resolvably light if the fusing sediment had experienced extensive, or multiple cycles, of weathering.

We performed experimental quantification of Si isotope fractionation between zircon and quartz, as well as solution MC-ICPMS analyses of natural zircon-quartz pairs and ion-probe analysis of individual ancient (>3.2 Ga) zircons. Experiments show that zircon is isotopically light relative to quartz, $\Delta^{30}\text{Si}_{\text{zircon-quartz}} \approx -0.3$ to -1.0‰ , at temperatures relevant to dry and wet granitic melts. Natural zircon-quartz pairs confirm this sense and magnitude of fractionation; also, we see resolvable differences between mantle-derived and crustal (I-type) zircons, which is most likely a result of the different temperatures and mineral assemblages of these two settings.

We see evidence for isotopically light ($\delta^{30}\text{Si} < -1.0\text{‰}$) zircons only after 4.0 Ga, which implies the presence of weathered material in the melt source. Our preliminary ion microprobe data show that, in the ancient zircon record, light $\delta^{30}\text{Si}$ signatures appear to lag heavy $\delta^{18}\text{O}$, which is perhaps evidence for the development of sediments that have experienced more extensive chemical denudation later in the Hadean and Eoarchean.

[1] Mojzsis et al (2001) *Nature* **409** 178-181; [2] Savage et al (2014) *Lithos* **190-191** 500-519; [3] Opfergelt et al (2012) *Chem. Geol.* **326-327** 113-122; [4] Savage et al (2013) *GCA* **109** 384-399; [5] Shahar et al (2009) *EPSL* **288** 228-234