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Extreme Hf isotope composition of metamorphic zircon from an Eoarchean Fe-rich lithology, Akilia, SW Greenland – clues to protolith, process or both?

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A thin (5m) layer of an Fe-rich quartz-amphibole-pyroxene (*qap*) lithology on the island of Akilia, south-west Greenland, was claimed to host Earth's earliest (>3.8 Ga) biogenic remnants in the form of isotopically light graphite. Subsequent to this claim, a wide range of studies were conducted in order to address the true age of the unit, the nature of its protolith and, hence, suitability to host biogenicity, as well as the nature of the putative $d^{13}C$ chemofossil. Previous geochronology has yielded ages consistent with known metamorphic events in the region at 3.65 Ga and 2.7 Ga. The present study analysed metamorphic zircon from *qap* for their Lu-Hf systematics, along with complementary whole-rock analyses in both *qap* and selected ultramafic host rocks. Whole-rock *qap* is considerably more radiogenic than surrounding ultramafic rocks ($\varepsilon_{Hf}(2.7) \approx +1$), extrapolating back to $\varepsilon_{Hf}(2.7)$ from +39 to +103, their evolution trends converging to mantle-like values at ~3.6 Ga. Zircon hosted in *qap* show a wide range in $\varepsilon_{Hf}(2.7)$ from +38 to +141 (< +231 by SIMS), while a single core has $\varepsilon_{Hf}(3.6) \approx +19$. Given the exceedingly low Lu/Hf in zircon, the elevated $\varepsilon_{Hf}(2.7)$ values are considered to reflect re-equilibration with variably fractionated and heterogeneous Hf reservoirs, including radiogenic whole rock host, a potentially extreme Hf isotope composition expected from apatite, and relatively unradiogenic older (3.6 Ga) zircon. The elevated $\varepsilon_{Hf}(3.6)$ of the analysed core possibly records similar processes occurring already in the Eoarchean. In terms of constraining protolith, a sub-unit of *qap* that geochemically most closely resembles the host ultramafic rocks also has radiogenic Hf at 3.6 Ga ($\varepsilon_{Hf}(2.7) = +18$), suggesting that a high Lu/Hf protolith already existed at this time. Since Fe-rich sediments can have high Lu/Hf ratios (e.g. BIF at Isua ranges in Lu/Hf between ~1 and 3), perhaps there is a vestige of such lithologies in Akilia. Attributing this anomalous reservoir instead to extreme metasomatism of an ultramafic protolith is consistent with other field and geochemical evidence from the entire *qap* unit, which clearly remains an unsuitable target for any ancient biogenicity search.