Does a heavy Fe-isotope composition of Akilia quartz-amphibole-pyroxene rocks necessitate a BIF origin?

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Running header; Do Fe-isotopes constrain Akilia protolith?

Abstract

The age and origin of the quartz-amphibole-pyroxene (qap) gneiss from the island of Akilia, southern West Greenland, has been the subject of intense debate since the light C-isotope composition of graphite inclusions in apatite was interpreted to indicate the presence of Earth's earliest biological activity. Although this claim for biogenic relicts has been vigorously challenged, the possibility that the rocks might represent some of the Earth's earliest water-lain sediments and, hence, a suitable repository for life, remains an open question. While some workers have suggested that the entire sequence represents an originally mafic-ultramafic igneous precursor subsequently modified by metasomatism, quartz injection, high-grade metamorphism and extreme ductile deformation, others maintain that at least a small part of the sequence retains geochemical characteristics indicative of a chemical sedimentary origin. Fractionated Fe isotopes with $\delta^{56}\text{Fe}$ values similar to those observed in Isua BIF reported from high-SiO₂ units of qap have been used to support a chemical sedimentary protolith for the qap unit. Here we present new Fe isotope data from all lithological variants in the quartz-amphibole-pyroxene gneiss on Akilia, including layers of undisputed ultramafic igneous origin. Since the latter require introduction of fractionated Fe into at least part of the qap unit, we argue that Fe isotopes must therefore be treated with considerable caution when used to infer BIF for part or all of the qap protolith.

Keywords: Archean, banded iron formation, biogenicity, isotope fractionation

Introduction

The south-west tip of the island of Akilia, which is located in the outer Godthåbsfjord, southern west Greenland, exposes mafic and ultramafic rocks enclosed by tonalitic Eoarchean Amitsoq gneisses (Fig. 1) and forms the type locality for the so-called Akilia Association enclaves (McGregor and Mason, 1977), which occur throughout the region. Hosted in the mafic and ultramafic rocks, a thin (<5 m thick), strongly deformed unit of relatively Fe-rich, quartz-amphibole-pyroxene (qap) gneiss (Figs. 1b,c, 2) has been the subject of an intense debate concerning its age, origin, and the possibility that it hosts biogenic relicts. Indeed, the scientific papers on this very small locality collectively serve as a textbook example of the difficulty to infer biogenicity in rocks of such great antiquity. The debate also illustrates how the need for proof of biogenicity can act as a key driver for analytical geochemical innovation.

Regardless of the sophistication of the analytical data, demonstration of a sedimentary protolith for all or part of the unit forms an essential prerequisite in any attempt to demonstrate early biological activity following the lines of previous claims (e.g. Mojzsis et al., 1996; McKeegan et al 2007), which have centered around the presence of isotopically light carbon as graphite inclusions in apatite, or indirectly by inference that deposition of banded iron formation (BIF), a primary marine chemical precipitate, most likely required biologically mediated oxidation (Towe, 1990). An initial proposal (Mojzsis et al., 1996) that the 5m-wide unit in its entirety represents BIF was based on a simple mineralogical comparison with undisputed BIF in the nearby Isua Greenstone Belt. Subsequently it has been shown that the unit is compositionally strongly heterogeneous (Fedo & Whitehouse, 2002) with a pervasive tectonic (Myers and Crowley, 2000; Whitehouse and Fedo, 2003) and polyphase metamorphic and metasomatic (Lepland and Whitehouse, 2010) overprint that obscures both the original nature of the rock and precludes determination of its exact

(>3.65 Ga) age (Fedó et al., 2006; Manning et al., 2006, *cf.* Whitehouse et al., 2009).

Additionally, Whitehouse et al. (2009) documented that the host ultramafic rocks, previously interpreted as an integral part of a volcano-sedimentary sequence together with the purported BIF (Nutman et al., 1997), contain olivine-rich lenses, which likely represent relict cumulates of dunite/peridotite. Such cumulates are common in deep-seated, layered intrusions and would not normally be associated with supracrustal rocks. However, the possibility remains that the cumulates could have formed in komatiite sills (Houle et al., 2009) or lava tubes (e.g. Gole et al., 2013) in proximity to the Earth's surface.

In view of the complexity of field, petrographical and petrological evidence that results from polyphase, high-grade metamorphism, the depositional context of the qap rocks cannot be uniquely constrained without additional geochemical and isotopic evidence. In this context, Dauphas et al. (2004, 2007) presented fractionated iron isotope data for qap rocks on Akilia, and proposed that these provided unequivocal evidence of a chemical sedimentary BIF protolith. The primary line of evidence used by Dauphas et al. (2004, 2007) was that qap rocks on Akilia are characterised by isotopically heavy iron (i.e. $0\text{‰} \leq \delta^{56}\text{Fe} \leq +1\text{‰}$) that is similar to that from undisputed >3.7 Ga quartz-magnetite BIF from the Isua Greenstone Belt, which crops out ~150 km northwest of Akilia on the edge of the Greenland Ice Cap (Fig. 1a). By contrast, the host ultramafic and tonalitic rocks were found to be 'typically' unfractionated ($\delta^{56}\text{Fe} \approx 0\text{‰}$), as expected for igneous protoliths. A secondary argument was made that the heavy Fe could not have been generated by metasomatic processes as described by Fedó and Whitehouse (2002) based on elevated Fe/Ti ratios in the fractionated qap rocks. The reported presence of mass independently fractionated (MIF) sulphur isotopes in an Akilia quartz-amphibole-pyroxene rock (Mojzsis et al., 2003) was further used by Dauphas et al. (2004, 2007) to support their BIF interpretation, although it has been shown that the key qap sample (G91-26 of Nutman

et al., 1997, also known as sample 92-197) does not contain MIF sulphur (Whitehouse et al., 2005).

The debate regarding the qap gneiss is further complicated by the relatively heterogenous nature of the 5m wide qap unit (Figs. 2a-e). Different publications have worked on various subsections of the unit, which have yielded mutually incompatible evidence. For example, of the three isotopically heavy Fe-bearing qap samples analysed by Dauphas et al. (2004), sample G91-26 (the sample with putative biogenic relicts, Mojzsis et al., 1996) is itself heterogenous, with a coarse-grained (<5 mm) quartz-rich part and a ca. 3 cm wide, finer-grained (<1 mm) part considered by some to have a likely BIF origin (Friend et al., 2002; Fig. 2c). A second sample (SM/GR/97/5) is also coarse grained (S. Moorbath, personal communication), in common with the bulk of G91-26 but in strong contrast to typically very fine-grained Isua BIF, and trace element studies of this particular sample show that it lacks the characteristic trace-element evidence for a BIF origin in the form of prominent positive La, Eu and Y anomalies (Bolhar et al., 2004). Insufficient details are given by Dauphas et al. (2004) to accurately characterise their third sample.

In order to investigate further the proposed link between isotopically heavy Fe and a possible chemical sedimentary (BIF) protolith, we have undertaken a detailed bulk-rock Fe isotope study on a wider range of rock types than those investigated by Dauphas et al. (2004). These include Fe-rich ultramafic veins and boudins (e.g. Fig. 2b), interpreted on the basis of trace-element geochemistry to represent either highly deformed, metasomatised relict igneous rocks (Fedo and Whitehouse, 2002) or the product of mafic and/or ultramafic rocks mixed with chemical sediment (Manning et al., 2006). Neither of these potential, clearly non-chemical sedimentary mafic/ultramafic protoliths would be expected to have a fractionated Fe-isotope signature.

Materials and methods

The samples selected for the Fe isotope investigation may broadly be categorised into (1) coarse and fine grained qap rocks, including a sample (AK12, Fig. 2e) which is lithologically equivalent to the finely banded portion of G91-26, the sample from which Mojzsis et al. (1996) reported isotopically light carbon; (2) ultramafic boudins and layers (Fig. 2b, d); (3) host ultramafic rocks (Fig. 2f) and amphibolites. Field relationships (including sampling linked to a detailed measured lithologic section), petrography, and bulk-rock geochemical data for samples with an AK- prefix were presented by Fedo and Whitehouse (2002).

Bulk-rock Fe isotope compositions were determined by solution multi-collector ICP-MS at the University of Hannover, following the anion exchange Fe purification method and analytical protocol described by Schoenberg and von Blanckenburg (2005) and Schoenberg et al. (2009). All samples were analysed in duplicate using a standard-sample bracketing approach and $\delta^{56}\text{Fe}$ values of samples are presented relative to the isotopically certified international standard reference material IRMM-014. Data are presented in Table 1.

Results

Bulk-rock Fe isotope data from these samples, together with the data of Dauphas et al. (2004), are plotted in Figure 3 against Fe/Ti, SiO₂, Mg# and total Fe. Quartz-bearing lithologies in the 5 m wide zone, regardless of whether they are fine grained like G91-26 or coarse grained, have $\delta^{56}\text{Fe}$ values that range from +0.17 ‰ to +1.1 ‰, with five of the six analysed samples in excess of +0.75 ‰. Fe/Ti ratios range from ca. 3,300 to 13,000, while SiO₂ content ranges from 60% to >90%. The low Mg# in this group is reflected by a marked increase in Fe content. Ultramafic layers, and layers segmented into structural

boudins made during intense deformation, span a similarly wide range of $\delta^{56}\text{Fe}$ from +0.23 ‰ to +0.96 ‰, with corresponding Fe/Ti ratios in the range 92 to 1,150, lower than those of the quartz-bearing rocks but still considerably higher than the bulk silicate Earth value of 45. Host ultramafic rocks and amphibolites are relatively unfractionated with $\delta^{56}\text{Fe}$ values slightly above zero and Fe/Ti ratios close to BSE, although it is worth noting that one of these samples (AK-02) has a $\delta^{56}\text{Fe}$ value of +0.14 ‰ that overlaps that of the least fractionated quartz-pyroxene rock (AK98 of Dauphas et al., 2004).

Discussion

Whereas the fractionated $\delta^{56}\text{Fe}$ values and high Fe/Ti ratios for qap agree with those analysed by Dauphas et al. (2004, 2007), the new data for mafic/ultramafic veins and boudins indicate elemental iron enrichment, expressed as high Fe/Ti and FeO_T values, and identify an unexpectedly wide range of $\delta^{56}\text{Fe}$. Such compositions are not compatible with an unmodified igneous origin and illustrate that isotopically heavy Fe cannot necessarily be used as a straightforward diagnostic signature of a BIF protolith (cf. Dauphas et al., 2004) in the absence of geological, textural, compositional, and isotopic evidence. Nonetheless, the presence of strongly fractionated Fe in such a diverse range of lithologies requires an explanation, particularly because most crustal rocks show only very small deviations in $\delta^{56}\text{Fe}$ from zero (e.g. Johnson and Beard, 2006). Figure 4 compares the obtained $\delta^{56}\text{Fe}$ values from the Akilia qap (this study and Dauphas et al., 2004, 2007) with a variety of BIF facies, as well as rocks from ore deposits and altered oceanic crust.

Magmatic processes appear incapable of generating the degree of fractionation observed in the Akilia samples. Even after extreme fractionation, very high- SiO_2 rocks generally reveal Fe isotope compositions similar to bulk earth (Beard and Johnson, 2004;

Schoenberg et al., 2006). Extreme degrees of crystal fractionation during the evolution of highly silicic magmas on Iceland have produced small fractionations < ca. +0.2 ‰ (Schuessler et al., 2009). In the isotopically heaviest high-SiO₂ granites, some of which contain $\delta^{56}\text{Fe}$ as high as +0.4 ‰, the fractionation was attributed to late-stage expulsion of isotopically light aqueous fluids (Poitrasson and Freydier, 2004) rather than magmatic processes. Heimann et al. (2008) also found $\delta^{56}\text{Fe}$ as high as +0.31 ‰ in high-SiO₂ granitoids and additionally demonstrated that the high- $\delta^{56}\text{Fe}$ resided in magnetite, implying loss of a low- $\delta^{56}\text{Fe}$ ferrous chloride fluid. None of these studies however, have revealed $\delta^{56}\text{Fe}$ values as high as those observed on Akilia. Thus, a primary magmatic origin for the isotopically variable but generally heavy Fe in the mafic/ultramafic veins and boudins appears implausible and any model for explaining the observed values can only build on Fe mobilisation by a fluid.

The simplest mechanism that can be envisaged is metasomatic transport of Fe from a reservoir in which fractionated Fe already exists, for example from BIF. Undisputed BIF preserving typical trace-element features of chemical sediments (Bolhar et al., 2004) is exposed at several localities in the Eoarchean of southern West Greenland, notably the extensive deposit in the Isua Greenstone Belt (IGB; Fedo et al., 2001), as well as smaller enclaves in the outer Godthåbsfjord close to Akilia (e.g. the Ugpik occurrence). Typically, BIF exhibits a range in $\delta^{56}\text{Fe}$, which is attributed to low-temperature, possibly biologically mediated, fractionation processes (Johnson and Beard, 2006). The IGB BIF is characterised by positive $\delta^{56}\text{Fe}$ < ca. +0.8 ‰ (Dauphas et al., 2004, 2007; Czaja et al., 2013). If this is a common feature of Eoarchean BIF in particular, such rocks could provide a fractionated source reservoir for metasomatic Fe. This fractionated Fe could have been mobilised and precipitated into the analysed rocks either during the 3.6 and/or the 2.7 Ga

metamorphic overprints and would therefore have no significance for deciphering the origin of the protoliths.

Dauphas et al. (2004) argued that observed Fe/Ti were incompatible with metasomatic alteration of mafic/ultramafic lithologies (Fedo and Whitehouse, 2002) on the basis that Ti should be immobile. However, in the Carajás region of Brazil, Neoarchean metasedimentary rocks include significant amounts of BIF from which Fe-ore is mined commercially on a large scale. Associated with these rocks are structurally controlled Fe-oxide Cu-Au deposits (e.g. at Igarapé Bahia) formed at ca. 2.57 Ga, which are characterised by, amongst other features, intense Fe metasomatism leading to grunerite, fayalite and/or magnetite/hematite formation, siderite alteration zones and silica dissolution (Tallarico et al., 2005). Importantly, in the context of the Akilia rocks, the siderite-rich mineralised zones at Igarapé Bahia exhibit similar extreme Fe/Ti ratios, ranging up to >5000 (Tallarico et al., 2005). Therefore, Fe/Ti ratios do not preclude metasomatic import of isotopically heavy Fe. Although no Fe isotopic studies of the Igarapé Bahia deposits presently exist, potentially isotopically fractionated high Fe/Ti lithologies may be generated that do not have a BIF protolith, though may actually derive their Fe from unrelated BIF.

If the origin of the observed Fe isotope compositions was more complex than simple redistribution of isotopically heavy Fe from BIF into metasomatised rocks, there is a significant hurdle to modelling this process. Akilia protolith(s) experienced extensive multiple Eoarchean and Neoarchean structural and metamorphic overprints (Whitehouse et al. 2009; Lepland and Whitehouse, 2011). Sulphides (pyrrhotite and chalcopyrite), which host some of the Fe in the qap unit, have Pb-isotopic signatures indicative of ca. 2.7 Ga (re-?) crystallisation (Whitehouse et al., 2005), metamorphic zircon is also predominantly ca. 2.7 Ga (Nutman et al., 1997, Fedo et al., 2006) with only rare >3.6 Ga

cores preserved (Whitehouse et al., 2009), while apatite records a Mesoproterozoic event (Sano et al., 1999; Whitehouse et al., 2009). The most recent U/Pb study of Akilia apatites documented a correlation between crystal size and apparent Proterozoic age caused by thermally activated volume diffusion of Pb (Chew et al., 2014). These events completely obscured the evidence for the likely protolith mineralogy of qap rocks and mafic/ultramafic lenses and veins, seriously limiting attempts at modelling, as Fe-solubility and isotope fractionation depend strongly on observed mineralogy and fluid composition.

Empirical studies of hydrothermally affected minerals and rocks clearly demonstrate that hydrothermal systems are capable of producing variable and highly fractionated $\delta^{56}\text{Fe}$ coupled with changes in bulk-rock chemistry (Fig. 4). For example, fractionation of Fe isotopes during hydrothermal remobilisation has been documented in sulphides from the Grasberg Cu-Au porphyry deposit of Irian Java (Graham et al., 2004), which record a total range in $\delta^{56}\text{Fe}$ from -2.1 to +1.1 ‰. Similarly large ranges in $\delta^{56}\text{Fe}$ have been described in a variety of Fe-bearing ore minerals from the Schwarzwald in southern Germany (Markl et al., 2006), with, for example, primary hematite ranging in $\delta^{56}\text{Fe}$ from -0.5 to +0.6 ‰ and secondary hematite alteration of siderite recording a $\delta^{56}\text{Fe}$ of < +0.9 ‰. Hydrothermally altered MORB has also been documented with variable $\delta^{56}\text{Fe}$ values < ca. +1.3 ‰ (Rouxel et al., 2003), with complementary negative signatures in other oceanic basement lithologies having $\delta^{56}\text{Fe}$ > -1.7 ‰.

Experimental mineral and solubility data and fractionation factors have been used to explain high- $\delta^{56}\text{Fe}$ values elsewhere. For geochemically unusual granitoids, Heimann et al. (2008) calculated that upon exsolution of ferrous chloride fluids, the $\delta^{56}\text{Fe}$ of magnetite but not those of Fe-silicates, increases alongside the bulk rock. This confirmed the possibility that in altered MORB, fluids might be capable of preferentially leaching light Fe (as

suggested by Rouxel et al., 2003). In carbonatites, Johnson et al. (2010) attributed high- $\delta^{56}\text{Fe}$ bulk rock values to interaction between low-Fe carbonates and Fe^{3+} -rich fluids. In view of the generally low $f\text{O}_2$ nature of serpentinisation, the preponderance of magnetite as a dominant Fe phase, and the possibility of a seafloor origin of the ultramafic rocks at Akilia (e.g. Nutman et al., 1997), we regard it as more likely that isotope fractionation between magnetite, Fe-silicates and a ferrous chloride fluid was involved in causing the elevated $\delta^{56}\text{Fe}$ values recorded in Akilia samples, which were most likely originally of ultramafic igneous origin (AK38 layer and boudins).

Finally, we reiterate that the polyphase nature of the metamorphic overprints on Akilia raises the possibility that the observed isotopic and geochemical features of the Akilia rocks are likely to result from a succession of non-isochemical processes rather than a single-event. By way of illustration, Fig. 3b investigates a schematic model in which early Fe-isotope fractionation or addition of fractionated Fe from an external source produced a sub-vertical data array in Fe-isotope vs. SiO_2 space, where low SiO_2 end-members are represented by the ultramafic boudins and veins. Subsequent silicification, could then have produced the quartz-rich lithologies by sub-horizontal displacement of this original alteration array to higher SiO_2 contents. We note that silicification of original volcanic rocks (Duchac and Hanor, 1990; de Vries et al., 2010) is common in the Archaean and may have arisen from interaction of Si-rich seawater (e.g. Thurston et al., 2012) with mafic volcanic protoliths (e.g. Baldwin et al., 2011), a model that already has been considered likely for qap rocks on Akilia (Fedo and Whitehouse, 2002). Furthermore, McGregor and Mason (1977) in their early study of rocks that comprise the Akilia Association distributed throughout the Godthåbsfjord region noted that “At many localities rocks of the Akilia Association with unusually high contents of quartz have been noted. Superficially they resemble banded iron formations, but [petrography and geochemistry] suggest that they are

rocks of varied parentage which have suffered intense silica metasomatism, often with quartz introduced along the layering of the parent rocks.”

Conclusions

Bulk-rock Fe-isotope data from the complete range of lithologies in the Akilia quartz-amphibole-pyroxene unit on Akilia confirm the elevated $\delta^{56}\text{Fe}$ signatures ($< \text{ca. } 1 \text{ ‰}$) in quartz-rich rocks previously noted by Dauphas et al. (2004, 2007). Such elevated $\delta^{56}\text{Fe}$ resembles that of early Archaean BIF from Isua and, on this basis alone, a chemical sedimentary origin for the Fe-isotope fractionation would be supported by the data.

Isotopically heavy Fe is also found in ultramafic boudins and layers within the qap unit, rocks which both sides of the Akilia protolith debate (Fedó and Whitehouse, 2002 cf. Manning et al., 2006) consider to have an igneous origin. Indeed, one of the highest $\delta^{56}\text{Fe}$ values, similar to that of the most quartz-rich lithologies, is found in a thin ultramafic layer. Regardless of whether these ultramafic rocks represent *in situ* strongly deformed igneous bodies (Fedó and Whitehouse, 2002; Whitehouse and Fedó, 2003; Whitehouse et al., 2009), or ultramafic relicts in a chemical sediment (Manning et al., 2006), their fractionated Fe cannot be an original igneous feature and must be introduced during alteration of the protolith(s) to the qap unit. Potential sources for this fractionated Fe include seafloor alteration (if the Akilia enclave represents an ocean floor), hydrothermal alteration, or remobilised, originally BIF/chemical sediment derived Fe. Accepting that fractionated Fe needs to be introduced into the ultramafic parts of the qap unit raises the possibility that the Fe isotope signature of all qap lithologies may be secondary and have no direct bearing on its protolith. These results also suggest that considerable caution needs to be used in the application of a fractionated Fe-isotopic composition to infer a chemical

sedimentary origin in rocks with a contentious and ambiguous protolith like those on Akilia.

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Disclosure statement

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Figure captions

Figure 1. (a) Location of Akilia within the Eoarchean gneiss complex of southern west Greenland. (b) Simplified geological sketch map (modified after Whitehouse et al., 2009) of the southwest tip of Akilia showing the thin qap gneiss outcrop within mafic and ultramafic gneisses of the Akilia Association enclave. Only pre-Qôrqt granite (2.55 Ga) units are shown. The asterisk marks the outcrop shown in Fig. 2a, while Fig. 2f was taken close to the end of the arrow indicating ultramafic rocks. (c) Aerial photograph of the Akilia enclave – asterisk corresponds to the same location highlighted in Fig. 1b. Due to the oblique angle of this photograph, the scale bar and north arrow are approximations and vary across the image. Pale brown weathering qap rocks are clearly visible. The prominent white unit (and other thinner white veins) is a Qôrqt granite sheet omitted from (b); darkening of the rocks surrounding qap represents the formation of hornblende due to extensive fluid activity (see Whitehouse et al., 2009 for discussion of the significance of this for geochronology on Akilia).

Figure 2. Geological features of Akilia. (a) Outcrop of qap at its widest point (ca. 5m), corresponding to the location marked by an asterisk in Figs. 1b and c. View looks approximately to the north west; dashed white lines mark outcrop boundary to hornblende to the right (east) and amphibolite to the left (west – note largely an artefact of relief in the photograph). (b) Ultramafic boudins (amph + px) in banded qap unit. (c) Finely banded layer (corresponding to sample AK33 of Fedo and Whitehouse (2002) within quartz-rich layers of qap unit; this sample is considered the in-situ equivalent to G91-26 and the mixed coarse/fine petrography of the latter is shown schematically by the dashed box. (d) Tectonically disrupted ultramafic layers (amph + px) which have been remobilised and injected by quartz perpendicular to foliation. (e) Dark, finely banded qap

unit from which sample AK12 (this study and Fedo and Whitehouse, 2002) was collected; adjacent parts of qap are exceeding quartz rich. (f) Massive olivine cumulate in ultramafic part of the enclave on Akilia (location noted in Fig. 1b).

Figure 3. Bulk-rock $\delta^{56}\text{Fe}$ values from Akilia samples from this study and Dauphas et al. (2004, 2007) plotted against (a) Fe/Ti, (b) SiO_2 wt. %, (c) Mg# and (d) total Fe wt. %. Data are grouped according to lithology following Fedo and Whitehouse (2002) as well as the “contact” rock group of Dauphas et al. (2007). Ultramafic layer AK38 and quartz-amphibole-pyroxene sample G91-26, which has been proposed as BIF (Mojzsis et al., 1996) are highlighted. Arrows in panel (b) show a possible two-stage process of initial Fe-isotope fractionation generating a range in $\delta^{56}\text{Fe}$ at relatively low SiO_2 (A) content followed by Si-enrichment (B)

Figure 4. Comparison of bulk-rock $\delta^{56}\text{Fe}$ values from Akilia samples (this study, Dauphas et al. 2004, 2007; symbols as in Figure 3) with Eoarchean (>3.7 Ga) BIF from the Isua Greenstone Belt (blue circles, data from Dauphas et al., 2004; 2007) and the range in compositions exhibited by different facies of BIF from the Kaapvaal Craton (Johnson et al., 2003; Frost et al., 2007). The range of $\delta^{56}\text{Fe}$ values observed in various hydrothermally altered environments are shown in the bottom panel, including mantle rocks (Beard and Johnson, 2004; Williams et al., 2004, 2005; Schoenberg et al. (2006), altered oceanic crust (Rouxel et al., 2003) and Black Forest ores (Schoenberg et al., 2006). The grey shaded vertical band represents the range of $\delta^{56}\text{Fe}$ observed in a wide range of common igneous rocks (Schoenberg et al. 2005, 2006).

- ultramafic boudins & layers (this study)
- quartz-amphibole-pyroxene rocks (this study)
- ◐ quartz-amphibole-pyroxene rocks (Dauphas et al., 2004, 2007)
- ▲ host ultramafics & amphibolites (this study)
- ▲ host ultramafics & amphibolites (Dauphas et al., 2004, 2007)
- ▽ contact rocks (Dauphas et al., 2007)
- ◆ granitoid gneisses (Dauphas et al., 2004)

Table 1. Bulk rock Fe-isotope data from Akilia samples

Sample	Lithology	Alias	$\delta^{56}\text{Fe}$ (‰)	*2 SE/SD	$\delta^{57}\text{Fe}$ (‰)	*2 SE/SD	SiO ₂ (wt. %)	Fe ₂ O ₃ (wt. %)	Fe/Ti	Mg #
AK10	boudin	run 1	0.798	0.035	1.162	0.074	50.1	23.6	1146	0.26
		run 2	0.819	0.042	1.232	0.066				
		average	0.808	0.030	1.197	0.098				
AK44	boudin	run 1	0.451	0.035	0.672	0.059	54.7	27.1	415	0.29
		run 2	0.458	0.033	0.676	0.071				
		average	0.455	0.010	0.674	0.005				
AK06	px-vein	run 1	0.241	0.033	0.318	0.060	51.0	10.9	165	0.49
		run 2	0.221	0.039	0.362	0.066				
		average	0.231	0.027	0.340	0.062				
AK38	"layer"	run 1	0.960	0.034	1.410	0.063	48.5	21.8	93	0.29
		run 2	0.968	0.034	1.401	0.062				
		average	0.964	0.011	1.406	0.014				
AK42	q-p	run 1	0.899	0.033	1.298	0.057	78.6	11.2	13008	0.22
		run 2	0.901	0.035	1.326	0.065				
		average	0.900	0.003	1.312	0.040				
AK12	q-p	run 1	1.059	0.040	1.582	0.063	63.5	26.7	7799	0.14
		run 2	1.069	0.039	1.572	0.055				
		average	1.064	0.014	1.577	0.015				
AK36	q-p	run 1	0.860	0.035	1.270	0.069	77.6	11.5	3340	0.22
		run 2	0.867	0.036	1.285	0.064				

		average	0.864	0.009	1.277	0.020				
AK05	hbl-ite	run 1	0.031	0.041	0.060	0.064	48.0	12.9	38	0.52
		run 2	0.058	0.038	0.083	0.064				
		average	0.045	0.037	0.072	0.031				
AK02	host-um	run 1	0.149	0.037	0.202	0.072	48.7	11.7	48	0.62
		run 2	0.129	0.039	0.180	0.069				
		average	0.139	0.029	0.191	0.031				
AK11	host-amph	run 1	0.071	0.040	0.136	0.071	47.1	12.5	24	0.35
		run 2	0.075	0.045	0.123	0.075				
		average	0.073	0.006	0.129	0.019				

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* uncertainties of single runs are given as 2 standard errors of the 20 measuring cycles; uncertainties of repeated measurements are given as 2 standard deviation. All $\delta^{56}\text{Fe}$ values are relative to the international Fe-isotope standard IRMM-014.