Origin and Significance of Archean Quartzose Rocks at Akilia, Greenland

Fedo and Whitehouse (1) presented data to support their hypothesis that granulite grade quartz-magnetite rocks on the island of Akilia, southwest Greenland—which are conformably interposed with pyroxenite and hornblendite units that date from before 3830 million years ago (Ma) (2–4)—had a metasomatic ultramafic igneous origin. They offered that interpretation despite the fact that a key sample in earlier studies of this area, Akilia quartzite G91-26, which has been investigated for carbon isotope evidence for early life (5) and described in previous studies as a metamorphosed marine sediment (2–9), is a 70 to >90% quartz unit containing magnetite (2, 5, 6). Fedo and Whitehouse (1) made no apparent distinction between the petrogenesis of a structurally coherent quartzite/magnetite unit and that of cross-cutting pyroxenite veins, nor did they explain the origin and nature of the remarkable metasomatic fluid that could have precipitated quartz-magnetite mineral assemblages hosting isotopically light carbon while removing Cr, Ni, Mg and platinoid (Pt, Ir) elements (7) typical of the mafic and ultramafic rocks from which they argue the quartzite was derived. We show here that all the data presented by Fedo and Whitehouse (1) are actually consistent with a primary sedimentary origin for the quartz-magnetite rocks on Akilia. Indeed, a more complete evaluation of geochemical evidence from previous studies (2, 5–10) demonstrates that the invasive pyroxenite veins are of no significance to the origin of the quartzite unit on Akilia and that these silica-saturated rocks could not be derived from metasomatism of ultramafic mineral assemblages.

Fedo and Whitehouse (1) compared rare earth element (REE) patterns from quartz-rich lithologies with patterns from pyroxenite (AK 38), adjacent ultramafic rocks (AK 2 to 5), and pyroxenite vein-rich samples (AK 9, 10, 31, 36, 38, 44, and 46) on a chondrite-normalized plot. This geochemical relation, however, constitutes
Table 1. Shale-normalized Ce and Pr anomalies and chondrite-normalized Eu anomalies for variscan supracrustal rocks in Akilia, compared with values for Isua BIF. n.d., no data.

<table>
<thead>
<tr>
<th>Sample</th>
<th>(Ce/Ce)SN*</th>
<th>(Pr/Pr*)SN*</th>
<th>(Eu/Eu*)SN*</th>
</tr>
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<tbody>
<tr>
<td>AK02</td>
<td>0.80</td>
<td>0.99</td>
<td>1.36</td>
</tr>
<tr>
<td>AK03</td>
<td>0.83</td>
<td>0.97</td>
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</tr>
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<td>AK04</td>
<td>0.94</td>
<td>0.99</td>
<td>2.72</td>
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<tr>
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</tr>
<tr>
<td>AK09</td>
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<td>0.98</td>
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<td>1.00</td>
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</tr>
<tr>
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<td>1.06</td>
<td>1.30</td>
</tr>
<tr>
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<td>1.02</td>
<td>1.08</td>
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<td>1.03</td>
<td>1.30</td>
</tr>
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<td>Avg. pyroxenite</td>
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<td>1.03</td>
<td>2.05</td>
</tr>
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<td>AK12</td>
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<td>1.03</td>
<td>3.45</td>
</tr>
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<td>1.08</td>
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<td>2.77</td>
</tr>
<tr>
<td>avg. G91-26</td>
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<td>n.d.</td>
<td>n.d.</td>
</tr>
<tr>
<td>avg. Isua QM/BIF</td>
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<td>n.d.</td>
<td>n.d.</td>
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<tr>
<td>avg. Isua MgBIF</td>
<td>0.70</td>
<td>n.d.</td>
<td>n.d.</td>
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</table>

*[(Ce/Ce)SN] = Ce(Ce)*/LaN / (NdN)*. 1/(Pr/Pr*)SN = Pr/(Pr)* / CeN / (Nd)*. 1/(Eu/Eu*)SN = Eu/CeN / Sm/CeN / (Nd)*.
High ductile strain and repeated high-grade metamorphism (3, 4) on Akilia mitigate against straightforward interpretation of the rocks and require careful sample selection (5). The problem of veining by secondary material in the siliceous unit, and of subsequent metamorphism and deformation of those two lithologies together, was already fully understood before the recent work of Fedo and Whitehouse; Nutman et al. [p. 2477 in (6), for example, noted that the “unit interpreted as a chert/BIF is traversed and locally extensively infiltrated by quartz and pyroxene ... veins. Such secondary material can form during diagenesis and then be metamorphosed, or might have developed during high grade metamorphism. This ... silica penetration is superimposed on top of the layering in the unit interpreted here as a BIF. ... G91-26 comes from one of the most quartz-poor layers in the best preserved BIF.” Thus, the material that Fedo and Whitehouse analyzed [figures 2 and 3 in (7)] was from precisely those layers that were metamorphosed and recrystallized (Fig. 1D) and that needed to be specifically avoided if one were seeking the characteristics of the sedimentary protolith. G91-26, the proposed metamorphosed chemical sediment studied in earlier work (6), came from a finer-grained, magnetite-layered part of the unit (Fig. 1D, top) clearly older than some cross-cutting veins and very different in composition. Fedo and Whitehouse have not produced data on samples similar to G91-26, which appears to fall between samples AK 32 and AK 34 in their section. Therefore, Fedo and Whitehouse did not analyze any material equivalent to sample G91-26, the proposed metamorphosed chemical sediment (6).

We agree with Fedo and Whitehouse that some pyroxene layers and boudins are highly modified parts of the stratigraphic package containing introduced silica (6, 7); such samples yield rare earth element (REE) patterns for clinopyroxenes (3, 7, 12) diluted with quartz (Fig. 2A). A fatal flaw in the claim by Fedo and Whitehouse that the siliceous rocks were produced by metasomatism of ultramafic rocks, however, is that the clinopyroxene-like REE patterns cannot be simply derived from the komatitites and the Komatiopsida (2, A and B). There is no easy way to elevate a 5-times-chondrite komatite pattern to the 10- to 13-times-chondrite start point depicted [figure 3A in (5)]. In contrast, the REE pattern for G91-26 (Fig. 2B) resembles REE patterns for Isua supracrustal belt BIF, which dates from 3800 to 3700 Ma (13).

Commonly, less metamorphosed and deformed komatitite-basalt sequences from <3500 Ma show hydrothermal alteration, with interspersed siliceous horizons and discordant veins, considered to have several different origins. Most represent chemical sediments, but some are formed by silification of flow-top breccias and elastic sediments, whilst others are entirely vein material (14–16). This is a geological context analogous to that seen on Akilia; thus, we conclude that the Akilia rocks represent variably altered mafic

Fig. 1. (A) Sketch map of southwestern Akilia island reproduced from Fedo and Whitehouse (7). (B) Geological map of southwestern Akilia depicting the full extent of the supracrustal unit across the headland and its contact with the bordering quartzofeldspathic gneisses. All mappable siliceous units are shown and sample sites for relevant geochemical studies are indicated. (C) Photograph of the debated siliceous unit, showing the location of sample G91-26 in relation to the section discussed by Fedo and Whitehouse (7). Unit is a rusty layer that preserves the fine-grained, magnetite-layered remnants and could equate to sample AK 33 of Fedo and Whitehouse (7), for which no data were presented. (D) Detailed photograph of a sliced portion of re-collected G91-26 (originally collected by CRLF, APN, and P. Kinny) demonstrating the clear distinction between the remnant, finely banded, magnetite-layered portion (top) and the later, coarse-grained, remobilized and recrystallised, quartz-rich, pyroxene-bearing portion (bottom).

Fig. 2. (A) Chondrite-normalized (17) REE plot of amphibolite (VM90/13) and ultramafic rock (G91-27) from either side of the siliceous unit [after (4)]. The REE pattern for G91-26 (bold red) is distinct from the adjacent meta-igneous units. REE patterns for clinopyroxene and amphibole from an Archaean granulite facies assemblage (3, 12) are shown. The fields of REE data (7) for the pyroxene layer and boudins (green-shaded field) and quartz-pyroxene rocks (blue-shaded field) are given for comparison. (B) Chondrite-normalized (17) REE plot of the average BIF values from Isua (13) and two high-grade BIF samples (3). The REE pattern for G91-26 (bold red) is similar to the Isua BIF patterns, although at lower overall abundance, which suggests less contamination by terrigenous sediments.
TECHNICAL COMMENTS

Response: Our recent paper (1) pertaining to metasomatized ultramafic rocks on Akilia has stimulated lengthy comments from workers who favor a BIF origin for the lithology and who have authored or coauthored papers (2–5) making claims about traces of life from a 5 m thick sequence of quartz-pyroxene rock on the island. A serious problem with these previous papers is that the putative evidence for life comes from a single sample, G91-26, for which, until publication of the Friend et al. comment, there has never been a specified field photograph, photomicrograph, or description revealing its details. Surprisingly, figure 1C of Friend et al. suggests that this critical sample was collected from an out-of-place boulder.

Nutman et al. (3) summarized the entire evidence supporting a BIF origin for G91-26 as follows: “[R]ocks have been interpreted as BIF on the basis of their magnetite layering and by comparison with other units such as in the Isua supracrustal belt and meet all the criteria of James (1954) [i.e. (6)] for BIF,” without specifying the comparison with Isua or clearly disclosing that the rocks possess a penetrative tectonic foliation (layering). We measured and sampled the entire ~5 m of the lithology partly because of its descriptions in previous studies: (i) “[O]ne metachert-BIF unit is broken up neither by boudinage, nor minor shearing, nor grossly disrupted by silica mobility. It forms a parallel sided, ~5 m thick unit with 70-80% quartz between . . . amphibolite . . . and ultramafic rocks” (3). (ii) “The oldest known sediment . . . is a layer ~3 m thick of BIF within a body of amphibolite . . .” (3). (iii) “A 40 m long section of BIF on Akilia has escaped boudinage, shearing, or disruption of the unit by silica mobility and forms a parallel-sided, ~5 m thick outcrop with up to 70% SiO2 locally” (5). Although we were aware that the BIF interpretation was based on analysis of sample G91-26, in every published paper (2–5) the entire ~5 m thick unit has been interpreted as unqualified BIF, in contradiction of both the general caution regarding secondary processes now expressed by Friend et al. and the current, unsubstantiated claims of “secondary pyroxene veins” of Mojszis and Harrison.

Regrettably, neither of these comments resolves the considerable ambiguity about the composition of sample G91-26. Previous studies (2, 3), as well as the current comment by Friend et al., have maintained that this sample comes from the most quartz poor layer (35% quartz, 45% pyroxene, 15% amphibole, and 5% magnetite), yet Mojszis and Harrison now claim that exactly the same sample “is a 70 to 90% quartz unit containing magnetite.” Apparently, they no longer accept their own earlier descriptions that the original sample contained 60% mafic minerals. In our detailed petrographic examination of 19 thin sections representing every lithologic variant, we have not recognized a “quartz-magnetite” rock anywhere in the ~5 m thickness of this unit. It forms a unique lithology observed only by Mojszis and Harrison, and as such requires detailed locality, field, lithological, and petrographic description. Our sample AK 33—a sample equivalent of G91-26—comes from a 7 cm thick band and comprises 36% quartz, 46% pyroxene, 9% amphibole, 6% magnetite, and 3% garnet and pyrite, a composition compatible with a metasomatized and metamorphosed ultramafic protolith.

Despite repeated inferences in previous studies (2–5) and the current comments that the Akilia quartz-pyroxene rock formed by sedimentary and biological processes, critical evidence for chemical sedimentation is lacking. Suggested comparison with classical BIF (2, 3) is misleading and inappropriate because secondary minerals are abundant, fine banding has a structural rather than a depositional origin, and the iron is hosted in non-sedimentary phases. Additionally, Mojszis and Harrison have misused the term “conformably”: In such repeatedly ductilely deformed, transposed, and recrystallized rocks, no field relationship can demonstrate depositional conformity; thus, there is no discernible temporal connection between the quartz-pyroxene rocks and the enclosing mafic-ultramafic assemblage.

The suggestion by Mojszis and Harrison that our metasomatic interpretation is “incompatible with all previous . . . studies” is wrong. The original study of Akilia association rocks (7) explicitly states: “At many localities rocks . . . with unusually high contents of quartz have been noted. Superficially they resemble banded ironstones, but . . . they are rocks of varied parentage . . . which have suffered intense silica metasomatism, often with quartz introduced along the layering of the parent rocks.” There is no compelling geologic requirement that vein quartz should be intruded along all geologic contacts, as Friend et al. apparently suppose. Widespread metasomatism of mafic and ultramafic rocks in the Isua greenstone belt is well documented (8, 9), including another sample that was also reported as BIF by Mojszis et al. (2) but that now has been demonstrated to be a mafic igneous rock with secondary carbonate veins (10). Associated graphite in this rock, interpreted as biologic in origin (2), was produced abiotically by siderite decomposition (10).

Our geologic map [figure 1B in (1)] is entirely germane to the scale of our sampling and independently dovetails with a published larger scale map [figure 3 in (11)]. A comparison between our map and the low-altitude, slightly oblique air photograph labeled with geologic contacts presented in an earlier study by Mojszis and Harrison [figure 4 in (4)] shows that the inferred fold closure of the quartz-pyroxene rock alluded to by Friend et al. does not occur. Near this inferred closure, the rocks are crosscut by a strongly sheared and intruded zone [at break in contact in figure 4 of (9)] that imposes near vertical foliation in rocks directly adjacent to the cross-cutting Qorqut granite pegmatite. Quartz-pyroxene rocks are exposed east of the sheared and intruded zone with no evidence of a fold closure [figure 1B in (1)]. Repeated failure in previous studies (2–5) to recognize high-strain foliation, boudinage,
quartz infiltration, cross-cutting ductile shear zones, and intrafolial folding where the rocks are 100% exposed—indeed, to specifically declare the absence of some of these features (3, 5)—seriously undermines any comments pertaining to geologic field relationships and interpretations.

Constraining the age of the quartz-pyroxene rock using U-Pb zircon geochronology of nearby granitoid gneisses (3, 4) infers a sedimentary relationship with the intervening mafic-ultramafic igneous rocks for which evidence is completely lacking. Furthermore, evidence for intrusion of the granitoids into the mafic-ultramafic package remains unconvincing because of the highly tectonized nature of the contact zone (11) and continued ambiguity over geochronological interpretations.

Without justification, Mojzsis and Harrison reject structural, mineralogic, and geochemical data indicating that sample AK 38, a thick pyroxenite band not invaded by metamorphic quartz, is integral to the lithology (1), preferring instead to consider it unrelated to other quartz-pyroxene rocks. By extension, pyroxenite layers that show pinch-and-swell structure [figure 2D in (1)] and pyroxenite boudins [figure 2B in (1)] must also be considered unrelated. We clearly demonstrated that thin banding is mineralogically similar to, and generated from, the boudinage of thicker pyroxenite bands [figure 2, B to D in (1)]. Following the argument put forth by Mojzsis and Harrison, the pyroxenite band represented by AK 38 has an ultramafic igneous origin, yet the remaining pyroxene-banded samples, including cm-thick pyroxenite boudins and mm-wide pyroxenite bands, are BIF. In the absence of evidence to the contrary, the most reasonable interpretation of boudins and thin pyroxenite bands, now encased in quartz, is that they have the same mafic-ultramafic protolith as the directly adjacent, thicker pyroxene bands.

We used REE profiles from quartz-pyroxene rocks to demonstrate compositional relatedness and to test proposed comparisons (3) with Isua BIF, not to determine protolith (1). We reiterate: All patterns of the quartz-pyroxene rock share important characteristics [conceivable light REE (La-Nd), Gd∕Nd∕LuN > 1] with sample AK 38, an undisputed ultramafic rock, and are distinct from Isua BIF (conceivable light REE, Gd∕Nd∕LuN < 1). Mojzsis and Harrison claim that whereas “pyroxene-rich” samples from the quartz-pyroxene unit have “conceivable light [ REE patterns,” three “quartz-saturated” samples (AK 12, 41, 42) have “patterns that parallel average Isua BIF,” a patently false statement that is unsupported by their figure 1A [or our figure 3A in (1)]. Sample AK 43 (strike-equivalent to AK 12), contains 60% pyroxene and only 32% quartz, and so is incorrectly labeled by Mojzsis and Harrison as “quartz-saturated”; samples AK 41 and 42 only have 0.2 and 0.1% magnetite, respectively, which requires that their REE patterns are derived from associated mafic minerals that Mojzsis and Harrison claim are “secondary veinings.” Indeed, a major problem with the data analysis in both comments is that every sample except AK 38 apparently contains an abundance of “secondary” minerals, so how can whole rock geochemistry be used to show a BIF protolith? Ignoring this, Mojzsis and Harrison suggest that Gd∕Nd∕LuN = 0.56, calculated from their analysis of G91-26, agrees with values for quartz-magnetite BIF from Isua. The characteristic positive La anomaly of these sediments, however, is revealed only by the combined criteria of (Ce/Ce*)SN = 0.56, calculated from their analysis of G91-26, agrees with values for quartz-magnetite BIF from Isua. The characteristic positive La anomaly of marine sediments, however, is revealed only by the combined criteria of (Ce/Ce*)SN < 1 and (Pr/Pr*)SN ≥ 1 (12, 13). Available Isua data (14) preclude such an analysis because they lack Pr, as do the data of Mojzsis and Harrison (figure 1A in their comment), which also contain an apparent analytical artefact in the form of a prominent, geochemically unreasonable Nd anomaly. The REE data presented for G91-26 in the Friend et al. comment, however, yield estimated (Ce/Ce*)SN = 1.04 and (Pr/Pr*)SN = 0.94 (15), both close enough to unity to dismiss the presence of a sediment-dissociative positive La anomaly.

Mojzsis and Harrison note that in a Cr/Th-versus-Th/Sc plot, most of our quartz-pyroxene samples (as well as G91-26) plot in the very broad field defined by Isua BIF. However, they fail to recognize that this field also encompasses Early and Late Archean basalt, Early and Late Archean andesite, Early Archean graywacke, and Early Archean cratonic shale (16), and therefore does not uniquely constrain a BIF protolith. In Cr/Th-versus-Th/Sc and TiO2-versus-P2O5 plots, cm-wide pyroxenite boudin samples (AK 09, 10, 44) that have an undisputed ultramafic igneous protolith lie in the field of Isua BIF, just as do the other quartz-pyroxene rocks. We are not surprised by their broad compositional range, considering that metasomatic quartz (and likely carbonate) has dilute the original ultramafic composition by more than 90% in some samples.

We categorically reject the claims in these comments that our field, mineralogical, and geochemical data support a sedimentary origin for any of the quartz-pyroxene rocks on Akilia. Our interpretation is that they formed via repeated deformation, metasomatism, and metamorphism of an ultramafic igneous protolith (1) is framed on well-documented examples and processes from the Early Archean in Greenland (7–11), as well as examples from other locations representing different parts of the geologic record.

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References and Notes
15. Because no data tables were provided for the new geochemical analyses of G91-26, we estimated these values from figure 2B of Friend et al. 
17. We thank B. Kamber, A. Lepland, M. van Zulzen, and G. Young for helpful discussions and for commenting on an early version of this response.
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Editor's Summary

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