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# On the true antiquity of Eoarchean chemofossils – assessing the claim for Earth’s oldest biogenic graphite in the Saglek Block of Labrador

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## Abstract

A recent claim to have found traces of Earth’s earliest life (>3.95 Ga) through isotopically light-carbon in graphite-bearing metapelites from the Saglek Block of northern Labrador, Canada is re-evaluated applying rigorous geological and geochronological criteria. The establishment of these criteria in previous evaluations of early life claims from southern West Greenland and northern Canada is reviewed in order to provide a backdrop to discussion of the Saglek claim. In particular, we emphasise the importance of the scale of lithological continuity in determining the veracity of such claims, which are considerably easier to demonstrate from large, relatively less tectonised supracrustal remnants like the Isua Greenstone Belt than they are from smaller, isolated enclaves of the kind found on Akilia or the highly tectonised and imbricated unit that is found in the Saglek Block. Unambiguous field relationships between ca. 3.9 Ga tonalitic gneiss and the graphite-bearing metasediments have not been demonstrated in the literature that the Saglek claim relies upon, and earlier U-Pb-Hf isotopic studies on zircon from metasediment at one of the localities used in the claim indicate a Mesoarchean to Neoarchean time of deposition. We conclude that, irrespective of the validity of the light carbon evidence, field relationships and geochronological evidence fail to demonstrate an age of >3.95 Ga for the first traces of life.

## **Introduction**

Nearly four decades have passed since Schidlowski et al. (1979) first proposed that a light carbon isotope signature in metasedimentary rocks, as found in the 3.7 Ga Isua Greenstone Belt (IGB) of southwest Greenland, might be used to indicate the presence of life in this early part of the Earth's history. Isotopic evidence was relied upon because the host rocks have been metamorphosed and deformed to a degree that precludes preservation of unambiguous biomorphic features, such as those seen in the remarkable ca. 3.4 Ga algal mats at Strelley Pool, Western Australia (Allwood et al., 2007). Since Schidlowski et al.'s seminal work, a number of claims have been made for low  $\delta^{13}\text{C}$  indicating life in the Eoarchean (Mojzsis et al., 1996; Rosing, 1999; Tashiro et al., 2017). Such cases, often hailed at the time of publication as evidence for 'the Earth's oldest life', attract great publicity and debate. However, a light carbon-isotope 'chemofossil' signature is just one part of the body of evidence required to establish a credible claim for Eoarchean life; it needs to be accompanied by unambiguous geological observations and geochronological constraints. This contribution restates the essential criteria that must accompany any 'chemofossil'-based claim. The application of these criteria in well-documented cases from the Isua Greenstone Belt (Rosing, 1999) and Akilia (Mojzsis et al., 1996) in southwest Greenland is presented, and the most recent claim (Tashiro et al., 2017), from the Eoarchean gneisses of the Saglek Block of northern Labrador, is reassessed.

## **Criteria for confident recognition of early biogenicity**

A number of key criteria, required for the credibility of claims for early life, were proposed in the review by Whitehouse and Fedo (2007). These criteria include, along with the carbon isotopic evidence that most Eoarchean claims rely upon, the suitability of host rocks as possible environments in which life might have developed, and temporal constraints provided either directly by the isotopic dating of the host rock or through field relationships with well-dated lithologies. Table 1 updates and expands the summary presented in Whitehouse and Fedo (2007), including notable Paleoarchean localities from which early life has been claimed, as well as the subsequently described Eoarchean localities of the Nuvvuagittuq Greenstone Belt (NGB) and the Saglek Block of northeast Canada. It is self-evident that the carbon isotopic evidence only becomes relevant when the suitability and antiquity of the host rocks is reliably established, with due consideration to geological context.

A clear distinction can be made between relatively robust evidence for early life in the Paleoarchean and less substantial claims for the Eoarchean. In large part, this distinction reflects lower degrees of metamorphism and deformation that have affected the Paleoarchean examples (from Barberton and the Pilbara), in which sedimentary environments are unambiguous and hosts of the claimed biogenicity can be reliably dated using intercalated felsic volcanic horizons. In these cases, applying the geological constraints is, in principle, no more difficult than it would be in a much younger sequence of rocks, and these studies have largely focused on the veracity of the biomorphic evidence, principally in the preservation of

trace fossils. Since the Paleoproterozoic examples of biogenicity are well-established and relatively uncontroversial, these will not be discussed further.

### **The importance of scale: lessons from Isua, Akilia and Nuvvuagittuq**

In any discussion of biogenicity in the early Earth, the IGB (Appel et al., 1998; Fedo et al., 2001) is of a critical importance, because its arcuate exposure preserves the most laterally extensive (ca. 35 km long, 1–5 km wide) package of metasedimentary lithologies known from the Eoarchean. Despite the fact that, in common with other Eoarchean examples listed in Table 1, the IGB has undergone locally intense ductile deformation and metamorphism to at least amphibolite facies conditions (with extensive metasomatic alteration in places that has, for example, transformed ultramafic rocks into metacarbonates: see Rose et al., 1996), it remains a relatively intact tectono-stratigraphic package across its entire exposure and is thus amenable to both the recognition of sedimentary protoliths as plausible early-life repositories (e.g. banded iron formations and fine-grained siliciclastics) and to unambiguous dating. As such, the IGB is the benchmark against which other Eoarchean claims for biogenicity may be evaluated.

The IGB comprises at least two groups of similar supracrustal rocks of demonstrably different age: a suite older than 3790 Ma, based on the dating of cross-cutting granitoids; and a younger suite containing a ca. 3710 Ma layer of felsic volcanics (Nutman et al., 1997a). The latter includes a metamorphosed ‘turbidite’ with well-preserved sedimentary structures, within which isotopically light graphite has been described by Rosing (1999). The turbidite unit is interbedded with mafic volcanics (the so-called Garbenschiefer Formation) and these have been combined on a Sm-Nd regression line to yield a direct date of  $3.78 \pm 0.08$  Ma. While the biogenicity of the turbidite-hosted light carbon has not been challenged, Pb-isotopic evidence from the host rocks for unusual, redox-controlled Th/U fractionation behaviour, which could be consistent with oxidative photosynthesis by cyanobacteria (Rosing and Frei 2004) remains equivocal (see Fedo et al., 2006, p. 864 and Fig. 12). In addition to the isotopically light C isotope evidence from the meta-turbidite, additional ‘chemofossil’ evidence in the IGB may be represented by the huge volume of >3.7 Ga banded iron formation (ca. 2 billion tonnes), since microbial oxidation of  $\text{Fe}^{2+}$  to  $\text{Fe}^{3+}$  is a plausible mechanism for formation of this enigmatic lithology (Konhauser et al., 2002; Towe, 1991; also see discussion in Whitehouse and Fedo, 2007).

As noted in Table 1, the IGB has been claimed on several occasions to host trace fossils. In the earliest study, spherical objects were interpreted as single or colonial cells and even assigned a taxonomic name, *Isuasphaera isua sp.* (Pflug and Jaeschke-Boyer, 1979). These objects were, however, shown to be fluid inclusions generated during recrystallization of chert (Bridgwater et al., 1981) or microcavities caused by carbonate dissolution (Roedder, 1981), with their organic content ascribed to post-glacial endogenic contamination (Appel et al., 2003; cf. Pflug, 2001). A recent report of preserved stromatolite-like features in a low-strain part of the IGB (Nutman et al., 2017) has also been robustly countered by Allwood et al. (2018), who provide evidence that the ‘stromatolites’ are merely structures produced during

ductile deformation. Thus, even in metasedimentary rocks as relatively well-preserved as those found in the IGB, the controversial nature of biogenic attribution demonstrates the difficulty of identifying anything other than chemofossils in such ancient rocks.

Elsewhere in the Eoarchean Itsaq Gneiss Complex (Nutman et al., 1996) that hosts the IGB, similar supracrustal lithologies occur as small enclaves within polyphase tonalite-trondhjemite-granodiorite (TTG) gneisses and are collectively referred to as the Akilia Association (McGregor and Mason, 1977), after the eponymous island south of Nuuk which exposes one of the larger examples (<30m wide and ~1km in total length when unfolded; Whitehouse et al., 2009). Indeed, the IGB is generally considered to be the largest (by far) Akilia Association enclave, likely representing the lithological precursor prior to tectonic dismemberment and intercalation with, and/or intrusion by, multiple generations of TTG gneisses. This process has effectively erased the field relationships that have been used to distinguish two suites of different age in the IGB. Consequently, any single enclave can only be assigned a specific age if it can be dated directly or is intruded by datable magmatic bodies with unambiguous ages.

Stepping down in scale from a coherent unit the size of the IGB to smaller, isolated enclaves in a TTG dominated terrane exacerbates the problems that bear on the identification and dating of any putative biogenic remnants such enclaves might contain. Regardless of whether crustal growth occurred in the Eoarchean via a mechanism akin to modern day plate-tectonics or an alternative mechanism (e.g. Kamber, 2015; Shirey et al., 2008), it is likely that sedimentary deposits of varying age (as seen in the IGB for example) would have been brought into both intrusive and tectonic juxtaposition with a succession of igneous rocks over time scales of tens to hundreds of millions of years. It thus cannot and should not be tacitly assumed that all supracrustal enclaves in a high-strain gneissic terrane are contemporaneous and predate the oldest magmatic precursors to the TTG gneisses. As demonstrated on Akilia (Whitehouse et al., 2009), the determination of the precise age of any given enclave requires either direct dating of the unit itself (or, in the case of sediments, estimation of a maximum age of deposition from detrital zircon) and/or an unambiguous cross-cutting relationship with dated igneous bodies (dykes or plutons). Whilst it is beyond the scope of the present paper to elaborate on the long-running debate about early life on Akilia, for which the reader is referred to Whitehouse et al. (2009, 2015, and references therein), it does provide arguably the most detailed and extensive investigation of a claim for Eoarchean biogenicity and, as such, demonstrates the problems associated with a reduction in scale of the host rocks and/or increasing tectonic/igneous dismemberment.

Firstly, recognition of supracrustal protoliths is complicated by the general lack of low-strain regions preserved in small enclaves. For example, spectacularly well-preserved pillow structures found in the Garbenschiefer unit of the IGB clearly identifies these as mafic volcanics that were extruded underwater. In contrast, mafic and ultramafic rocks on Akilia, interpreted as a metavolcanic basement to a Fe-rich quartzitic unit that was equated with banded iron formation (BIF; Manning et al., 2006; Mojzsis et al., 1996, Nutman et al., 1997b) have been alternatively interpreted as parts of a layered mafic intrusion, based on the presence of olivine-rich layers that potentially represent dunite or peridotite cumulates (Whitehouse et

al., 2009). Similarly, the part of the Akilia enclave claimed to host biogenic remnants, a <5 m wide discontinuous layer over an (unfolded) length of ~400 m of a quartz-amphibole-pyroxene gneiss (the *qap* unit of Whitehouse et al., 2015) bears little resemblance to the quartz-magnetite BIF of the IGB, despite early claims to the contrary (Mojzsis et al., 1996), and has alternatively been interpreted as the product of silification of an ultramafic protolith (Fedo and Whitehouse, 2002; Whitehouse et al., 2015; *cf.* Manning et al., 2006).

Secondly, assigning an age either to the *qap* unit or the associated mafic-ultramafic enclave has encountered difficulties due to the demonstrably tectonised contacts (Whitehouse and Fedo, 2003) that were previously claimed to be cross-cutting (Nutman et al., 1997). Even if a cross-cutting relationship was preserved, there is the additional complication of assigning true magmatic ages to thin granitic veins, which may represent relatively small melt fractions containing largely inherited zircon that have been extracted from older gneisses (Whitehouse and Kamber, 2005).

Thirdly, attempts to directly date the *qap* unit provide a good example of how this approach is rarely straightforward in ancient, polyphase metamorphic terranes where isotopic chronometers are susceptible to partial or complete resetting. Whereas the Akilia *qap* unit contains zircon, these predominantly record a ca. 2.7 Ga high-grade metamorphic event (Fedo et al., 2006; Nutman et al., 1997; Whitehouse et al., 2009) with only two ca. 3.6 Ga cores reported, the latter also probably metamorphic in origin given their contemporaneity with well-documented high-grade events in the Itsaq Gneiss Complex (Griffin et al., 1980; Nutman et al., 2000). Apatite, the mineral actually claimed to host the biogenic remnants (Mojzsis et al., 1996) records a late Paleoproterozoic age (Chew et al., 2014; Whitehouse et al., 2009). A Sm-Nd isochron derived from ‘metagabbros’, a component of the Akilia Association enclave on Akilia, combined with a single sample from the *qap* unit, yields an age of  $3677 \pm 36$  Ma (Whitehouse et al. 2009), which could represent either a protolith age or granulite-facies metamorphic disturbance (e.g. Whitehouse, 1988) of Sm-Nd isotope systematics.

Another proposed candidate for evidence of Eoarchean life occurring in a supracrustal association that is neither as extensive nor as well preserved as the Isua Greenstone Belt occurs in the Nuvvuagittuq Greenstone Belt (NGB) of northern Canada (O’Neil et al., 2007). The NGB is a folded belt, ca. 0.5 km wide and over 4 km long (unfolded), comprised mostly of metamafic rocks (so-called “faux amphibolite” interpreted as altered pyroclastic mafic volcanics), within which thin bands of metaBIF and metachert occur in the western limb of a macrofold. Evidence of life, as claimed by Dodd et al. (2017), is in the form of haematitic microtubes and filaments in Fe-rich metasediments, comparable to those formed by bacteria near modern seafloor hydrothermal vents. Age constraints are provided by field relationships, with minimum ages from cross-cutting ca. 3.77 and 3.75 Ga trondhjemites and ca. 3.65 Ga metamorphism (Cates and Mojzsis, 2009; Darling et al., 2013), along with a maximum age of deposition from detrital zircon in quartzite (ca. 3.78 Ga; Cates et al., 2013). Regardless of the long-running debate about the possibility of a 4.3 Ga age of formation for some components of the NGB (O’Neil et al., 2012 *cf.* Guitreau et al., 2013), it appears to preserve evidence of biogenicity that is of similar age to that from the IGB (Rosing, 1999). As such, the IGB and NGB localities represent the best examples of early life claims constrained by field

relationships with dated lithologies, whereas the Akilia locality is considered unsubstantiated (Table 1).

### **The Labrador claim**

The Saglek Block of northern Labrador, Canada, is the locality most recently added to the corpus of claims for early terrestrial life. Tashiro et al. (2017) presented carbon isotopic evidence from small metapelitic enclaves in predominantly Eoarchean TTG gneisses, which they claimed to have ages in excess of 3.95 Ga, thus representing evidence for traces of Earth's earliest terrestrial life. Tashiro et al.'s (2017) claim exceeds the age of examples from the IGB and NGB by up to 200 million years, to a time when the rate of meteorite bombardment was considerably greater (Bottke and Norman, 2017), with a concomitantly increased "impact frustration" to the establishment of terrestrial life (Chyba, 1993; Maher and Stevenson, 1988; Sleep et al., 1989). In the Saglek claim neither the carbon isotopic data, nor the sedimentary character of the protolith to the host rocks, are in question. However, Tashiro et al.'s (2017) assignment of a >3.95 Ga age, which is based on field mapping and geochronology presented in recent publications by the same group (Komiya et al., 2015; 2017; Shimojo et al., 2016), conflicts with previous observations from the Saglek Block published over several decades. The rest of this current paper therefore examines this new claim in detail, in order to test whether the rocks of the Saglek region may be truly claimed to host traces of Earth's earliest known life.

### *Geology and geochronology of the Saglek Block*

In the northern Labrador Peninsula, Archean gneisses of the Nain Province extend for approximately 500 km along the coast, constituting the westernmost part of the North Atlantic Craton (NAC). The gneisses have been subdivided into the southern Hopedale and northern Saglek blocks, with the latter being the focus of recent geological studies (Komiya et al. 2015, 2017; Kusiak et al., 2018; Morino et al., 2017; Sałacińska et al., 2018; Shimojo et al., 2016; Tashiro et al., 2017; Vezinet et al., 2018). The Saglek Block is composed of Eoarchean to Neoproterozoic TTG gneisses tectonically intercalated with lesser amounts of supracrustal rocks and syn-tectonic granitoids, assembled during amphibolite to granulite facies metamorphism in the Neoproterozoic (Bridgwater et al., 1975; Krogh and Kamo, 2006; Kusiak et al., 2018; Ryan and Martineau 2012). Archean gneissic crust was significantly reworked along the western margin of the Nain Province during the 1.8-1.7 Ga Torngat orogeny (van Kranendonk and Helmstaedt, 1990). Most geochronological work on the Saglek Block has been focussed around Saglek Bay (Fig. 1), where TTG gneisses are considered by most authors to be dominated by the Uivak I Gneiss, with magmatic protoliths of Eoarchean age (3.8-3.6 Ga; Kusiak et al., 2018; Sałacińska et al., 2018; Schiøtte et al. 1989). Tashiro et al.'s (2017) claim for >3.95 Ga traces of early life rely on studies by the same group that obtained a wider spread of ages for the Uivak I Gneiss (3890-3610 Ma; Komiya et al., 2017), and that proposed the presence of an older component of 'Iqaluk' tonalitic gneiss (>3.9 Ga; Shimojo et al.,

2016). The presence of a pre-3.8 Ga igneous component in the TTG gneisses has been demonstrated by zircon dating in other studies, including a ca. 3.9 Ga ‘Nanok Gneiss’ proposed by Collerson (1983) but unconfirmed in later studies on the same locality (Komiya et al., 2017; Sałacińska et al., 2018), and a ca. 3.86 Ga grey felsic orthogneiss on the shore opposite Nulliak Island (Vezinet et al. 2018). However, only Shimojo et al. (2016) claim the existence of a protolith older than 3.9 Ga. Since these age estimates are critical for the claim of >3.95 Ga life by Tashiro et al. (2017), they will be discussed in more detail in the next section.

In the field, the Uivak I Gneiss is generally described as fine-grained grey orthogneiss that, in the absence of zircon geochronology, is not readily distinguishable from younger orthogneisses of similar composition and texture, due to the intensity of later deformation and metamorphism (Collerson et al., 1982; Komiya et al., 2015; Krogh and Kamo, 2006). Some of these ‘grey’ gneisses have Eoarchean protoliths (e.g. Krogh and Kamo, 2006; Komiya et al., 2017; Sałacińska et al., 2018); however, widespread felsic magmatism also occurred between 3.3 and 3.0 Ga, as well as during two high-grade metamorphic events at ca. 2.7 Ga and 2.5 Ga that produced the gneissic structure (Bridgewater and Schiøtte, 1991; Krogh and Kamo, 2006; Komiya et al., 2017; Kusiak et al., 2018; Schiøtte et al., 1990, 1992). In addition, an earlier ca. 3.6 Ga high-grade metamorphic event affecting the Uivak I Gneiss has been recognised (Sałacińska et al., 2018).

These TTG gneisses are tectonically intercalated with ‘supracrustal’ assemblages that include clastic and chemical metasediments along with mafic to intermediate metavolcanics (Fig. 1). Associated ultramafic gneisses may be metamorphosed cumulates (Komiya et al., 2015, Morino et al., 2017). These assemblages have been divided into two groups; an older, pre-Uivak I Nulliak assemblage, and a younger, Meso- to Neoproterozoic Upernavik assemblage (Bridgewater and Schiøtte, 1991). These are not readily distinguishable in the field, since pre-tectonic geological relationships with the host TTG gneisses have been largely obliterated by ductile deformation. The importance of distinguishing pre-Uivak I lithologies by intrusive field relationships was recognised by Komiya et al. (2015), who state “... it is important to find supracrustal rocks, cut by granitoid intrusion with well-determined Eoarchean ages”. However, they provide no evidence of Uivak Gneiss intruding metasedimentary-mafic-ultramafic associations, with photographs only showing tectonised contacts with mafic gneisses or intrusions of “young granite” (*sic* Komiya et al., 2015); as is also the case in subsequent publications from the same group (Komiya et al., 2017; Shimojo et al., 2016; Tashiro et al., 2017).

Another field test, and one that has been more widely adopted in the Saglek area than intrusive relationships with host TTG gneisses, is the presence or absence of the metamorphosed equivalents of mafic ‘Saglek’ dykes, originally described by Bridgewater et al. (1975) as a texturally distinctive generation of plagioclase-phyric to megaphyric metadykes, emplaced into the older Uivak TTG gneisses and Nulliak assemblage but pre-dating the formation of the Upernavik supracrustals and younger granitoids (Bridgewater and Collerson, 1977). In the latest version of the 1:100,000 Saglek Fjord – Hebron Fjord Area map sheet (Ryan and Martineau, 2012), the definition of Saglek dykes is broadened to include



aphyric mafic and ultramafic varieties, and the supracrustal assemblages are further distinguished by the presence of ‘chemical precipitate’ metasediments, especially ironstones and metacherts, that are present in the Nulliak but absent in the Upernavik supracrustals. However, the map authors emphasize that the distinction is problematic, and that the criteria are not used consistently. Several large sections of supracrustal rocks mapped as ‘Upernavik’ in survey reports on which the map is partially based (Ryan et al., 1983; 1984) have been changed by Ryan and Martineau (2012) to ‘Nulliak’ (presumably on the presence of ironstones, although this is not explicitly stated). Conversely, supracrustals near Hebron that are cut by “metamorphosed porphyritic basic dykes and layered porphyritic sills that resemble the Saglek dykes” (Ryan and Martineau, 2012) were mapped as Upernavik, not Nulliak, supracrustals due the absence of associated ironstones. The authors of the map also acknowledge that, at least in the case of the ‘Upernavik supracrustals’, zircon dating has demonstrated that these cannot belong to a single volcano-stratigraphic sequence, as demonstrated by Bridgwater et al. (1990). Thus, it is recognised by multiple authors that the ‘Saglek dyke’ test is not a reliable means of distinguishing pre and post-Uivak I supracrustal enclaves.

Dating of detrital zircon in metasediments that belong to supracrustal assemblages provides better constraints on the timing of pre- and post-Uivak I packages. A quartzite from 2 km south of St John’s Harbour, mapped by Ryan and Martineau (2012) as part of the Nulliak assemblage (Fig. 1B), yielded concordant ages between ca. 3850 and 3700 Ma, along with discordant, younger ages that were interpreted as having been affected by ca. 2.7 Ga metamorphism (Nutman et al., 1989). Samples of metasediment mapped as Upernavik supracrustals from Torr Bay and the northeast entrance to St. John’s Harbour (Fig. 1A) yielded concordant ages of 3.65 - 3.2 Ga and 3.35 - 2.95 Ga, respectively (Schjøtte et al., 1992). The authors of these studies interpret the youngest grains in their samples as providing maximum ages of deposition for the sedimentary protoliths; taken together, they indicate the varied provenance of clastic sediments in different supracrustal enclaves within the Uivak I gneiss.

#### *Age constraints from field relationships*

Graphite-bearing metasediments with light carbon analysed by Tashiro et al. (2017) were collected from localities around St. John’s Harbour, Big Island and Shuldham Island (Fig. 1). Lithologies at these localities were previously mapped as part of the Upernavik assemblage (Fig. 1A, B; Ryan and Martineau, 2012; Schjøtte et al., 1992). The only sample locality in Tashiro et al. (2017) that was previously mapped as belonging to the Eoarchean Nulliak supracrustal assemblage (Ryan and Martineau, 2012) is at Pangertok Inlet, but samples from there contain no graphite. Tashiro et al. (2017) rely on revised maps and field interpretations presented in Komiya et al. (2015, 2017) and Shimojo et al. (2016) for their correlations (Fig. 1C, D). The former two papers reassigned many occurrences of Upernavik supracrustals to the Nulliak assemblage, based on claims of observed intrusions of Uivak felsic orthogneisses and Saglek metadykes into the assemblages.

Assessing the supporting evidence for this re-assignment first requires a comment on the reliability of using mafic metadykes to differentiate between the two supracrustal assemblages. This approach was questioned over 40 years ago (Glikson, 1977 *cf.* Bridgewater and Collerson, 1977), but little has been done since to verify such a division in the Saglek Block. Even though they have been regarded (e.g. by Komiya et al., 2015) as equivalents of the ca. 3.5-3.3 Ga Ameralik dykes of southern west Greenland (Nutman et al., 2004), the Saglek metadykes have never been dated; furthermore, most metadykes assigned by later authors (such as Komiya et al., 2015) to the Saglek ‘swarm’ do not possess the distinctively porphyritic texture described by Bridgewater et al. (1975). Non-porphyritic relicts of metabasic dykes and discontinuous amphibolites are abundant in both TTG and supracrustal gneisses of the Saglek Block (e.g. Krogh and Kamo, 2006). By extension, some later authors (e.g. Komiya et al., 2015; Ryan and Martineau, 2012) assume that all pre-metamorphic relicts of mafic dykes are ‘Saglek’ dykes, and that supracrustal gneisses intruded by them are consequently part of the Nulliak assemblage. However, the authors that first established this distinction recognised later (Bridgewater and Collerson, 1977) that there are not only multiple generations of mafic metadykes, but also that the Upernavik assemblage itself, as previously defined, is likely to encompass various tectonic enclaves of supracrustal rocks of different ages and origins (Bridgewater and Schiøtte, 1991). Indeed, ca. 2.8 Ga ages from a “mafic enclave” in tonalitic gneiss at Tigigakyuk Inlet (Komiya et al., 2017) were interpreted as dating a relict metadyke, in the same study that uncritically assumes other mafic metadykes to be Saglek dykes. The presence of undated mafic metadykes clearly cannot be considered as a reliable basis for distinguishing the Nulliak assemblage from younger supracrustal packages.

The photographic evidence in Komiya et al. (2015, 2017) and Shimojo et al. (2016) for the intrusion of felsic or mafic magmatic protoliths into ‘supracrustal’ rocks is only presented for mafic and ultramafic gneisses that are assumed to be contemporaneous with graphite-bearing metapelites. In none of these studies do the authors demonstrate, either through field relationships or geochronological data, that granitoid ‘dykes’ are related to Uivak I gneisses (as opposed to the syn- and post-tectonic granites that abound in the area), or that discontinuous layers or sheared lenses of mafic gneiss represent metamorphosed Saglek dykes. In Shimojo et al. (2016), gneisses yielding zircon with ages older than 3.8 Ga were derived from a single outcrop near St. John’s Harbour (Fig. 1), a few metres across and isolated from surrounding outcrops by vegetation (as shown in the field photo and sketch from Shimojo et al. (2016) and confirmed by the authors of this review during a visit in 2017 – see Fig. 2).

Because zircon dating of this single outcrop (Shimojo et al., 2016) is the sole geochronological basis for the claim of >3.95 Ga life by Tashiro et al. (2017), the relationships between rock types within the outcrop deserve special attention. The outcrop (Fig. 2) is a 12 by 4 m exposure of felsic and mafic orthogneiss that has been scrubbed clean of lichen to reveal its structure. Shimojo et al. (2016) identify seven magmatic generations (Fig 2a), affected by ductile deformation (foliation and folding). Samples of banded grey gneiss (‘third generation’, LAA 995, see Fig. 2a) and late cross-cutting granitic orthogneiss (‘seventh generation’, LAA 994, see Fig. 2a) were dated by Shimojo et al. (2016), yielding

comparable distributions of age data that led these authors to assume that metamorphism occurred at ca. 3.87 Ga, and that the tonalitic protolith to the banded grey gneiss intruded at ca. 3.9 Ga, for which reason it is given a new name, the Iqaluk Gneiss. Re-examination of these age data will be undertaken below; here, it suffices to point out that Komiya et al. (2017), Shimojo et al. (2016), and Tashiro et al. (2017) extrapolate this result to surrounding TTG gneisses, proposing a so-called ‘Iqaluk-Uivak’ succession of prolonged granitoid formation between 3.9 and 3.6 Ga. This extrapolation assumes correlation between the ‘Iqaluk Gneiss’ and nearby outcrops, which cannot be demonstrated in the field because the outcrop is completely isolated, occurring in a low-lying area of poor exposure (Fig. 2b, c). Nor is evidence presented that the metamorphism and deformation recorded in this, or nearby outcrops, is not related to the widespread ca. 2.7 Ga granulite-grade event that has been demonstrated by numerous studies to have produced the dominant gneissosity in most of the Saglek Block (e.g. Kusiak et al., 2018, and references therein).

The ‘Iqaluk’ site also provides the sole field relationship on which the  $>3.95$  Ga life claim relies: namely, the presence of mafic gneiss of a supposed earlier (‘first’) generation, at the southeast end of the exposure (Fig. 2). This mafic portion is interpreted as ‘supracrustal’ by Shimojo et al. (2016), and as belonging to the Nulliak supracrustals by Tashiro et al. (2017). Both interpretations are entirely unsupported by evidence. There are no associated metasediments present; indeed, the nearest graphite-bearing lithologies are hundreds of metres away (Fig. 1D), and are separated from the Iqaluk site by large areas of vegetation (Fig. 2c; also see maps in Tashiro et al., 2017). There is no evidence that the undated mafic gneiss predates the ‘Iqaluk’ gneiss – it shares the folded gneissosity of the latter, but is separated from it by granite that intrudes across gneissosity (Fig. 2d, e) and was injected into fold axial planes (‘sixth generation’ of Shimojo et al., 2016; Fig 2a). In fact, there is no reason to assume that the mafic gneiss is anything but a more mafic component of the TTG-like gneiss that dominates the outcrop.

In summary, without justification provided by either field association or geochronology, Tashiro et al. (2017) uncritically assume that the mafic orthogneiss present in the ‘Iqaluk’ outcrop is related to mafic components of the Nulliak supracrustal assemblage. They further assume that any mafic or metasedimentary gneiss that contains remnants of mafic metadykes must belong to the Nulliak assemblage. Neither of these assumptions holds up to scrutiny.

#### *Age constraints from geochronological data*

As explained above, the specific age constraint of  $>3.95$  Ga for the isotopically light-carbon bearing metasediments described by Tashiro et al. (2017) relies entirely on the dating of one outcrop of ‘Iqaluk Gneiss’ by Shimojo et al. (2016). Tashiro et al. (2017) claim that this is “...the oldest supracrustal rock in the world, intruded by the more than 3.95 Ga Uivak-Iqaluk Gneiss.” This statement is misleading, since Shimojo et al. (2016) nowhere claim that they have evidence for the Iqaluk Gneiss being older than 3.95 Ga; instead, they claim an age of “~3.9 Ga”, and speculate that “...the precursor tonalitic magma... was possibly formed at  $\geq$ ca. 3.9 Ga.” The ‘ $>3.95$  Ga’ claim is, in fact, repeated from Komiya et al. (2015), who made

the claim based on unpublished data. The interpretation of zircon data presented by Shimojo et al. (2016) will be discussed further below.

Recent dating of gneisses on Nulliak Island and the adjacent coast (Komiya et al., 2017; Morino et al., 2017; Vezinet et al., 2018) highlights the problem with assuming age equivalence between similar lithologies, both in the case of TTG gneisses (especially the fine-grained ‘grey’ variety that is widely assumed to belong to Uivak I), and between separate tectonic enclaves of supracrustal rocks with similar assemblages. Ultramafic rocks in multiple ‘supracrustal’ enclaves, mapped as Nulliak by Ryan and Martineau (2012), have yielded bulk-rock Sm-Nd regression ages of both ca. 3.3 and 3.8 Ga (albeit from sample suites that exhibit scatter beyond that attributed to analytical uncertainties, as shown by MSWDs of 4.5 and 15, respectively; Morino et al., 2017). On Nulliak Island, grey gneisses that host supracrustal enclaves, previously assumed to belong to the Eoarchean Uivak I gneiss (Ryan and Martineau, 2012), include both ca. 3.3 Ga and 3.7 Ga protoliths (Komiya et al., 2017). Post-Uivak I ages for the protoliths of ‘grey gneiss’ have also been obtained from Little Island (Krogh and Kamo, 2006), Big Island (Komiya et al., 2017), Tigigakyuk Inlet (Sałacińska et al., 2018) and south of St. John’s Harbour (Sałacińska et al., submitted). Considering such variation of protolith ages for TTG gneisses, it cannot be assumed from composition or texture that ages can be extrapolated from dated to undated outcrops. Nor can it be assumed that grey gneisses observed to intrude supracrustal rocks belong to the Eoarchean Uivak I or supposed older (‘Iqaluk’) TTG gneisses. Zircon dating is required where intrusive relationships are clearly observed, and this has not been undertaken in the studies that Tashiro et al. (2017) rely upon.

The St. John’s Harbour East locality (Fig. 1C) identified by Tashiro et al. (2017) exposes graphite-bearing pelitic (Komiya et al., 2015), conglomeritic (Tashiro et al., 2017) and siliceous (Schjøtte et al., 1992) metasediments. Detrital zircon from the locality where Tashiro et al. (2017) obtained graphite-bearing samples LAD849A-C and LAD852 (Fig. 1C) previously yielded ages between 3.35 Ga and ca. 2.83 Ga (sample DB82.4X, Schjøtte et al., 1992; see Fig. 3a for replotting of isotopic data), giving a Mesoarchean maximum age of deposition that is also consistent with a multigrain zircon Lu-Hf chondritic model age of 3.05 Ga (Stevenson and Patchett, 1990). Metasediments from nearby Torr Bay yielded similar results. Neither Tashiro et al. (2017), nor the field and geochronological studies by the same group that they rely upon (Komiya et al., 2015; 2017; Shimojo et al., 2016), address the significance of these earlier data to their claims. These zircon data can only be reconciled with Eoarchean deposition of the metasediments if they either: (1) result from severe Pb-loss from much older zircon during the ~2.7 Ga granulite facies metamorphism or, (2) represent post-depositional zircon growth. As shown in Fig. 2a for metasedimentary sample DB-82.4X from St. John’s Harbour East (Schjøtte et al., 1992), zircon analyses plot on or close to the concordia curve around 3 Ga. If such zircon had originally crystallised at ~3.95 Ga, and experienced Pb-loss during high-grade (granulite facies) metamorphism at ~2.7 Ga, the analyses should lie along a discordia line that deviates markedly from concordia at the  $^{207}\text{Pb}/^{206}\text{Pb}$  ages represented by these zircon analyses, which is clearly not the case. Furthermore, a plot of the  $\alpha$ -dose (from U and Th decay) accumulated between hypothetical

zircon crystallization at  $\sim 3.95$  Ga and metamorphism at  $\sim 2.7$  Ga (Fig. 3b) shows that only a few grains in DB-82.4X would be fully metamict (“stage III” of Murakami et al., 1991, using the revised  $\alpha$ -doses of Pidgeon, 2014) at  $\sim 2.7$  Ga, the majority having sufficiently low U and Th contents that they would remain mostly crystalline (stage I) and hence, are unlikely to have undergone profound Pb loss. The Lu-Hf data obtained on a population of zircon from sample DB-82.4X (Stevenson and Patchett, 1990) are also difficult to reconcile with a  $>3.95$  Ga age. When plotted on a Hf isotope evolution diagram (Fig. 3c) at the average  $^{207}\text{Pb}/^{206}\text{Pb}$  age of  $\sim 3.0$  Ga obtained from the ion microprobe U-Pb data, back extrapolation along a zircon Pb-loss trajectory ( $\text{Lu}/\text{Hf} \approx 0$ ) from this point to hypothetical zircon crystallization at  $\sim 3.95$  Ga implies an improbable highly supra-chondritic ( $\epsilon_{\text{Hf}3950\text{Ma}} \approx +20$ ) source. Alternatively, if the zircon represents metamorphic growth at  $\sim 3$  Ga, the Hf isotope composition should lie on a typical crustal evolution trend at this time (reference trends are shown in Fig. 3c for mafic and felsic rocks originating from a chondritic source at 3.95 Ga), which clearly is not the case. Thus, both alternatives that would permit a ca. 3.95 Ga age are contradicted by the published U-Pb and Lu-Hf isotope data.

So far, the only detrital zircon study of metasediments previously mapped as part of the Nulliak assemblage yielded a population of ca. 3845 Ma ages (sample 83-187, Nutman and Collerson, 1991; see Fig 1B for location). As with sample DB-82.4X, accumulated radiation damage in this sample at 2.7 Ga (Fig. 3b) indicates that only a few grains would be susceptible to Pb-loss at this time and the reported ages are therefore likely to provide a maximum age of deposition, as was suggested by Nutman and Collerson (1991). However, Komiya et al. (2015) suggest that they have all experienced Pb loss, and consequently argue that the data instead provide a *minimum* depositional age, with the oldest concordant datum having the least Pb loss. This is a similar argument to that in Shimojo et al. (2016), where the oldest single analysis from the ‘Iqaluk Gneiss’ is used to suggest that the protolith is older than 3.95 Ga. Such arguments are illogical; while the possibility of Pb loss is always present in metamorphosed rocks, it cannot be just assumed, nor should such assumptions form the basis of interpretation without independent evidence, for example from non-equivalence of multiple *in situ* analyses in the same zircon growth phase (e.g. Whitehouse et al., 2017). Detrital ages are, by definition, older than the deposition of the host sediment, and data affected by Pb loss do not provide real ages; therefore, no datum could ever be used to provide a minimum age of deposition in a sediment, regardless of Pb loss.

As discussed in the previous section on field relationships, Tashiro et al.’s (2017) age of  $>3.95$  Ga is based on the dating of zircon at a single outcrop (Fig. 2) of ‘Iqaluk Gneiss’ (Shimojo et al., 2016). Two of the seven magmatic generations proposed by Shimojo et al., (2016) were dated by laser ablation–inductively coupled plasma mass spectrometry (LA-ICPMS) on zircon grains characterized by cathodoluminescence. Both samples yielded similarly-scattered ages that reflect a complex magmatic and metamorphic history with significant Pb loss. The data are rather imprecise, with individual  $2\sigma$  uncertainties on  $^{207}\text{Pb}/^{206}\text{Pb}$  ages averaging  $\pm 67$  Ma (range  $\pm 18$  to  $\pm 136$  Ma). The  $^{238}\text{U}/^{206}\text{Pb}$  ratios also range widely in precision, from 0.9 to 8.5 % ( $2\sigma$ ), with random dispersion of the data away from concordia in both directions, suggesting that there was large secular variation in isotope count rates during many analyses,

or that external precision has been grossly underestimated. Even if one accepts the assertion that the single oldest analysis of  $3953 \pm 54$  Ma dates the formation of the magmatic protolith and that all other data are younger due to the effects of later metamorphism, the uncertainty of this single datum undermines the claim for an age *in excess* of 3.95 Ga. Furthermore, this datum is not a unique statistical outlier; pooling of a large subset of the oldest analyses (within 5% discordance limits) yields a statistically significant ( $p > 0.05$ ) population with a mean  $^{207}\text{Pb}/^{206}\text{Pb}$  age of  $3865 \pm 4$  Ma ( $n = 142$  of 304 analyses). Such an analysis includes data that Shimojo et al. (2016) interpret as both igneous and metamorphic, based on CL observations (oscillatory versus unzoned) and Th/U values (“metamorphic” with Th/U  $< 0.48$ ). However, criteria to distinguish igneous and metamorphic zircon are inappropriately applied. Not all igneous zircon will have strong oscillatory zoning, and the higher U content of what appears to be ‘unzoned’ zircon dampens CL response and obscures growth zoning (Hoskin and Schaltegger, 2003). In addition, although Th/U values are commonly used to distinguish metamorphic from igneous zircon, with a threshold value far lower ( $\sim 0.1$ ) than that employed by Shimojo et al. (see review by Rubatto, 2017 and references therein), the use of Th/U is unreliable, particularly in granulite facies rocks (Möller et al., 2003). Inspection of data from both samples LAA994 and LAA995 shows almost complete overlap of ages between zircon ascribed to metamorphic and igneous growth; as such, the two types are not statistically distinguishable. Furthermore, Shimojo et al. (2016) use ca. 3870 Ga data from sample LAA994 of granitic gneiss, whose protolith intruded across gneissosity, to infer that all metamorphism and deformation occurred at this time, providing a minimum age for all generations of magmatism in the outcrop. However, they do not provide an age estimate for the gneissic protolith, instead relying on the interpretation of unzoned, low-CL zircon as metamorphic. This interpretation ignores the possibility of zircon inheritance in granitic melts produced during high-grade metamorphic events that elsewhere in the Saglek Block, along with granitic magmatism during and after these events, have been dated at ca. 2.7 Ga and 2.5 Ga (e.g. Baadsgaard and Collerson, 1979; Krogh and Kamo, 2006; Kusiak et al., 2018; Schiøtte et al., 1989; Sałacińska et al., 2018; Vezinet et al., 2018).

Faced with such uncertainties in this critical age constraint, both statistically and in terms of field relationships, it cannot be convincingly demonstrated that graphite occurs in metasediments that predate the ‘Iqaluk’ gneiss. The use of ‘ $>3.95$  Ga’ by Tashiro et al. (2017) is not even supported by the Shimojo et al. (2016) paper itself, where it is merely suggested as a possible interpretation, without substantial evidence. The assumption that multiple exposures of TTG gneisses in the Saglek Bay area (renamed as Iqaluk-Uivak Gneiss in Tashiro et al., 2017) are of similar age is completely unsupported by the evidence. Finally, the interpretation that the graphite-bearing metasediments belong to supracrustal assemblages that predate the TTG gneisses is not only unsupported by zircon geochronology, in one locality it is clearly contradicted by previous work (Schiøtte et al., 1992; Stevenson and Patchett, 1990). In light of these many weaknesses in the chain of evidence, the claim by Tashiro et al. (2017) to have found the oldest traces of life remains unsubstantiated.

## Conclusions

It was not the purpose of this study to present new evidence to add to the long-running debate about the presence and age of life in the earliest part of the Earth's history. Instead, we have presented an assessment of such claims, with information that is available to any reader of the literature, in which we stress the vital importance of unambiguous and reliable interpretations of field relationships, tied to equally reliable geochronology. Without rigorously applying these criteria, claims of evidence for early and/or earliest life will remain questionable. This is especially the case in studies like that of Tashiro et al. (2017), where such essential evidence is not provided, instead exclusively relying on the veracity of work already published. Such subordination of supporting evidence puts the onus on the reader to delve into the literature to extract the facts, or otherwise take the authors presentation of such at face value. Instead, it should be the scientific responsibility of the claimant to present the evidence in a way that the reader can objectively assess.

To end on a more positive note, when examined against the key geological and geochronological criteria, there is good evidence for isotopically light, potentially biogenic carbon in the Eoarchean, particularly from the relatively well-preserved and laterally extensive Isua Greenstone Belt (Rosing et al., 1999). Likewise, if precipitation of banded iron formation truly requires a biologically mediated process, the IGB again provides an abundance of unambiguous, geochronologically well-constrained lithology (Whitehouse and Fedo, 2007). The (trace) fossil evidence from the Eoarchean remains more circumspect and, with the exception of a claim from the Nuvvuagittuq Supracrustal Belt (Dodd et al., 2017), has been robustly challenged. This is not to say that such evidence cannot exist; simply that all such claims need a foundation of good geology and geochronology to support them. Without these, the presence of life in the Earth's oldest rocks will remain speculative.

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## Figure and table captions

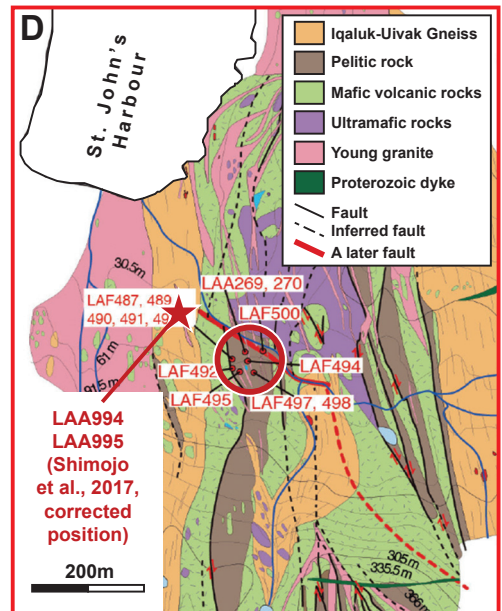
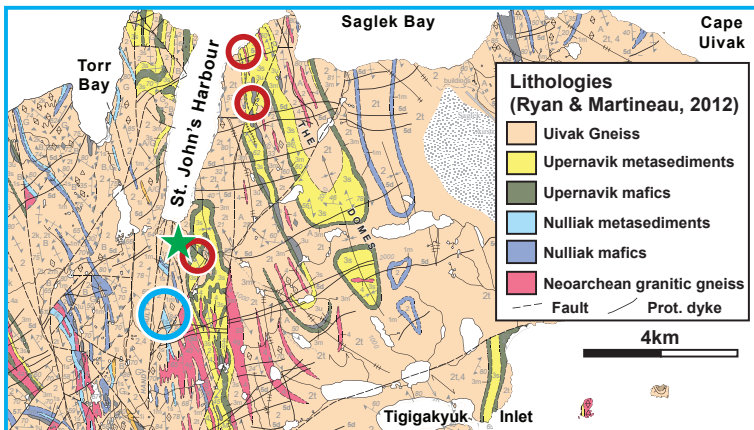
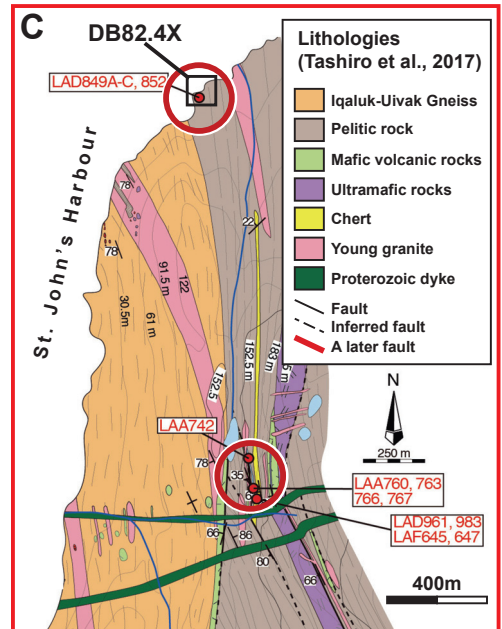
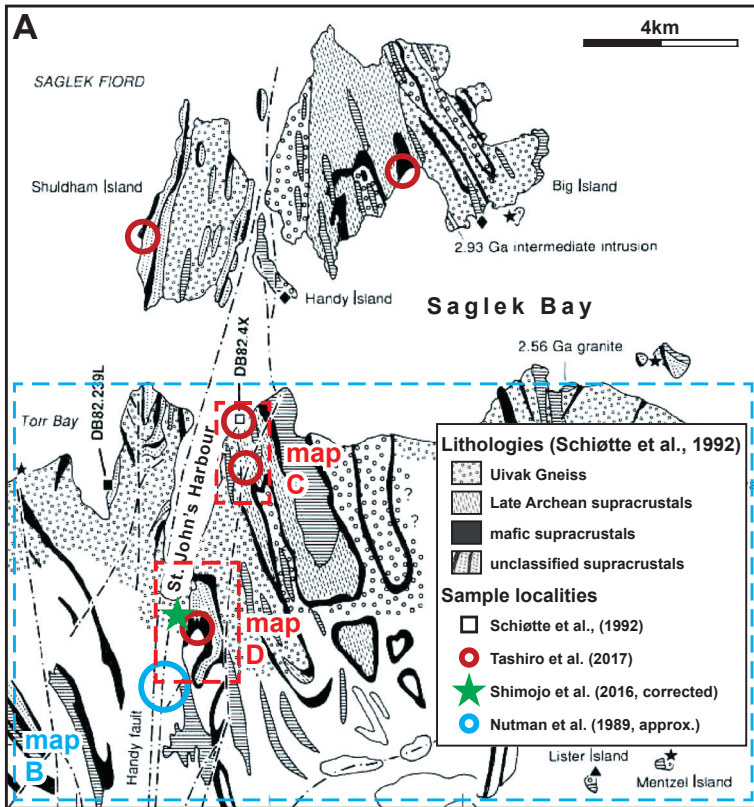
Fig. 1. Maps of the Saglek Bay/St. John's Harbour area from various publications, showing differing interpretations of supracrustal lithologies. (A) from Schiøtte et al. (1992); (B) part of the Ryan & Martineau (2012) map at same scale as map A, showing division of supracrustals into Upernavik and Nulliak assemblages. Sample positions shown from various publications; location of outcrop in Shimojo et al. (2016) is incorrect; correct position taken from Komiya et al. (2017) and field observations by authors in 2017. (C) map from Tashiro et al. (2017) of St. John's Harbour East area, showing localities of graphite-bearing samples; (D) map from Tashiro et al. (2017) of the St. John's Harbour South area. All 'pelitic rocks' are regarded by Tashiro et al. as belonging to the >3.95Ga Nulliak supracrustal assemblage, contrary to earlier publications.

Fig. 2. The outcrop described in Shimojo et al. (2016). (A) interpretative sketch map after Shimojo et al. (2016), showing seven magmatic generations, including the Iqaluk Gneiss (generation 3) and mafic gneisses (hornblendite and amphibolite, generations 1 and 2, respectively) assigned to the Nulliak supracrustal assemblage by Tashiro et al. (2017). (B) photograph of outcrop from Shimojo et al. (2016) with mafic gneisses indicated by green arrow. Area and view direction of photograph D shown in red. (C) view of outcrop looking east towards the closest outcrops of supracrustals, which are over 150 m away across a significant area of no-exposure. (D) view of SE corner of outcrop, showing banded gneiss ('Iqaluk') separated from mafic gneiss by deformed metagranite. Area and view direction of photo E shown in blue. (E) view of mafic gneiss intruded by metagranite to the right. Photograph B after Shimojo et al. (2016); C-E taken during visit by the authors in 2017.

Fig. 3. Isotope systematics of U-Pb and Lu-Hf in zircon from samples DB-82.4X (siliceous metasediment, Upernavik supracrustal assemblage, east entrance St. John's Harbour; Schiøtte et al., 1992) and 83-187 (quartzite, Nulliak supracrustal assemblage, 2 km south of St. John's Harbour; Nutman and Collerson, 1991). The former is from the same locality where Tashiro et al. (2017) identify graphite in associated metapelite; see Fig. 1 for both sample localities. (A) Concordia plot of U-Pb ion microprobe data for zircon from sample DB-82.4X. Data plot around 3 Ga on the concordia; if such zircon had originally crystallised at 3.95 Ga (black star), and experienced Pb-loss during high-grade metamorphism at 2.7 Ga, the data should lie along a discordia line (dashed), which they do not. (B) Plot of hypothetical accumulated  $\alpha$ -dose (from U and Th decay) in 3.95 Ga zircon at the time of metamorphism (2.7 Ga), for U-Th compositions of zircon from samples DB-82.4X and 83-187. Shaded areas show the stages of radiation damage due  $\alpha$ -recoil (Murakami et al., 1991). Most zircon grains plot in the stage 1 field and would have been crystalline and hence are unlikely to have undergone profound Pb-loss. (C) Plot of Hf isotope evolution of a bulk zircon separate from sample DB-82.4X (Stevenson & Patchett, 1990) at 3.0Ga, the average  $^{207}\text{Pb}/^{206}\text{Pb}$  age of the ion microprobe data shown in (A). Extrapolating back along a Pb-loss trajectory ( $\text{Lu}/\text{Hf} \approx 0$ ) from this point to 3.95 Ga (black star) would require an improbably high supra-chondritic source. Alternatively, if the ca. 3Ga age is metamorphic, the Hf isotope composition should lie on a typical crustal evolution trend from a chondritic source at 3.95 Ga (for mafic or felsic rocks, as shown), which clearly is not the case.



Table 1. Summary of evidence for early life, revised and updated after Whitehouse & Fedo (2007).



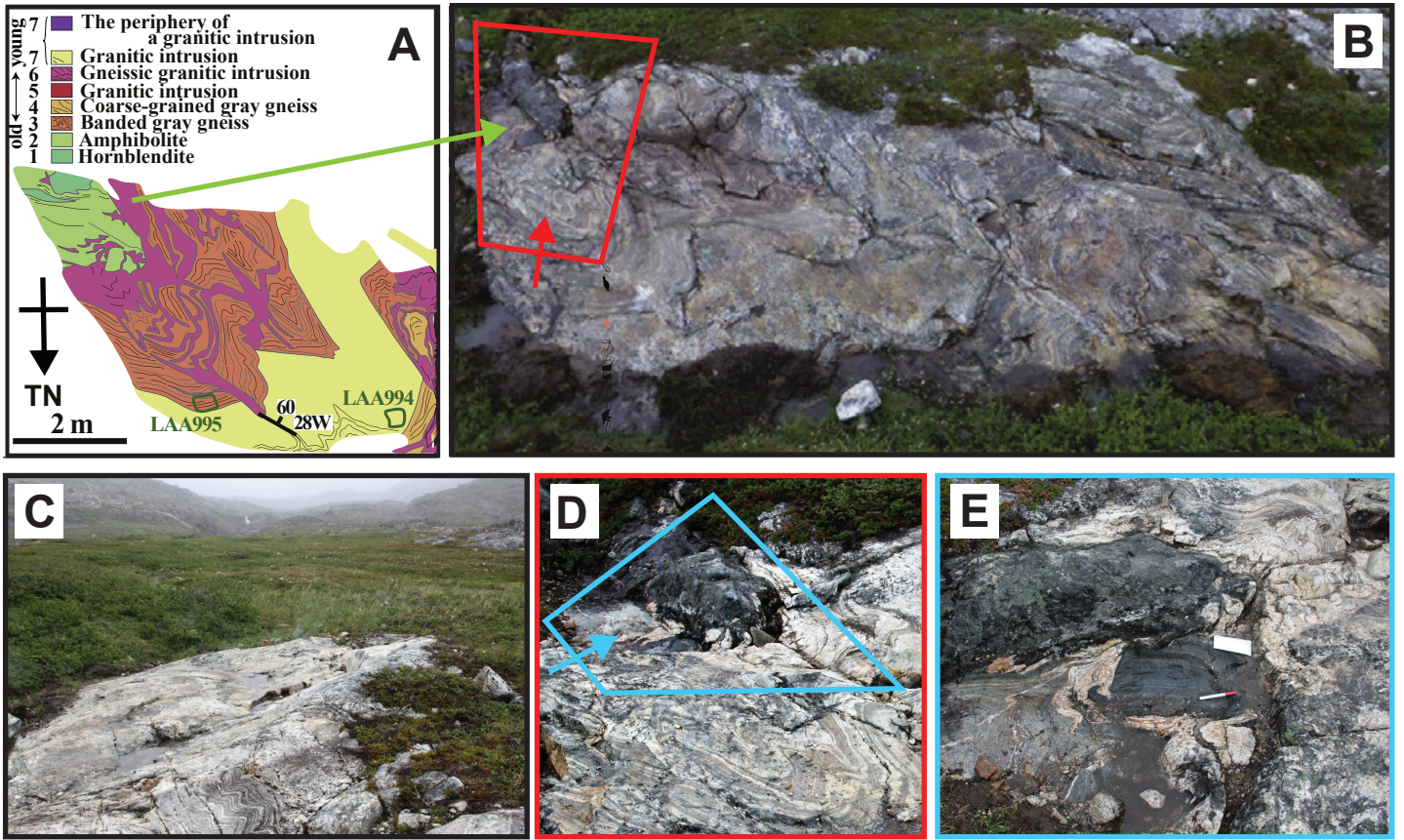
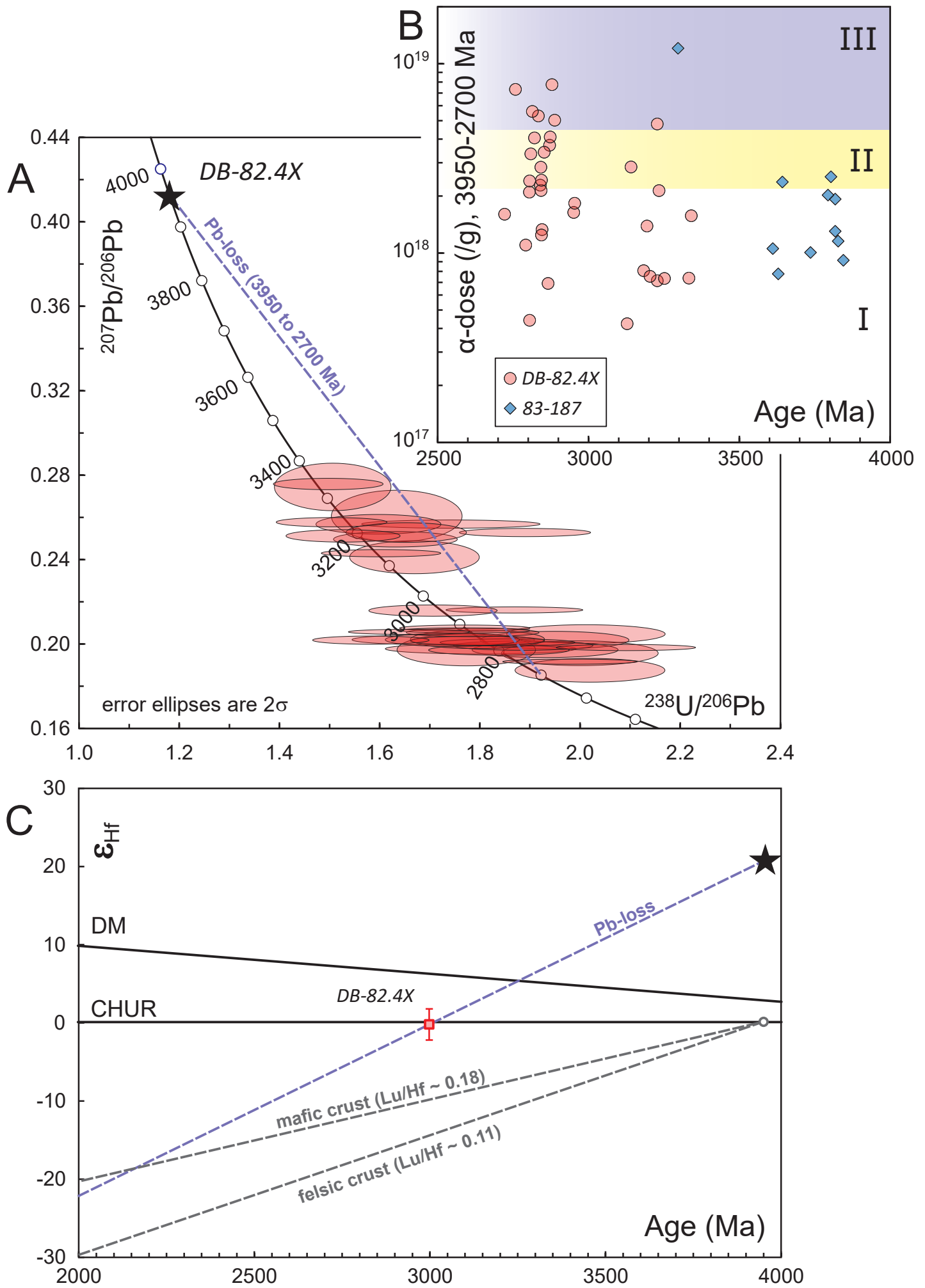


Fig. 2



**Fig.3**

**Table 1. Summary of evidence for terrestrial life before 3.4Ga**

Biomarker	Claim age	Primary reference	Assessment criteria				Reliability
			Field relationships	Geochronological constraints	Suitability of host	Evidence of biogenicity	
<b>PILBARA, WESTERN AUSTRALIA</b>							
Microfossils + microscale (SIMS) C isotopes	3.46 Ga	Schopf et al., 2017	Apex chert in stratigraphic sequence with dated volcanics	Indirect: Between volcanics with zircon ages of ca. 3.47, 3.46 Ga (1)	Microfossils hosted by epiclastics and syndepositional hydrothermal veins in chert (2)	Kerogenous filaments w. variable $\delta^{13}\text{C}$ down to -25‰; filaments could be abiogenic phyllosilicate with later hydrocarbon films (3)	low
Microbial mat fossils	3.48 Ga	Noffke et al., 2013	sediments in well-preserved succession	Direct: ca. 3.48Ga from detrital zircon and overlying volcanic (4)	Intertidal sediments	Organic films draped over ripple beds, akin to modern biomats	viable
<b>BARBERTON, SOUTH AFRICA</b>							
Trace fossils & C isotopes	3.48 Ga	Furnes et al., 2004	Basalt/komatiite in stratigraphic sequence with chemical sediments (chert); Prenite-pumpellyite-facies metamorphism	Direct: Ar-Ar age for metamorphism (ca. 3.48 Ga, 5); detrital zircon dating (ca. 3.48Ga, 6)	Altered basaltic glass in pillow lavas; oncoids in black chert breccia	$\delta^{13}\text{C}$ down to -25‰ in oncoids;	viable
<b>ISUA GREENSTONE BELT, WEST GREENLAND</b>							
Relict microfossils	3.8 Ga	Pflug & Jaeschke-Boyer, 1979	>3.7 Ga metachert in Banded Iron Formation (BIF)	Direct:: ca. 3.7 Ga, Pb-Pb isochron from metaBIF (7) and detrital (volcaniclastic) zircon (8)	Extreme deformation precludes preservation of spherical body fossils (9)	Apparently fluid inclusions (10) or cavities (11). Organic matter present probably recent endolithic (cf. 12)	discredited
Bulk rock carbon isotopes	3.7 Ga	Schidlowski et al., 1979	metacarbonate (replacing sediments)	metacarbonate age unknown (<3.7 Ga)	IGB metacarbonates not original sediments (13, 14)	Range of bulk $\delta^{13}\text{C}$ not replicated (15)	discredited
Microscale (SIMS) carbon isotopes	>3.7 Ga	Mojzsis et al., 1996	metacarbonate (replacing sediments)	metacarbonate age unknown (<3.7 Ga)	IGB metacarbonates not original sediments (13, 14)	Graphite in apatite association not unique to supracrustals (16)	discredited
Bulk rock carbon isotopes	3.78 Ga	Rosing, 1999	Clastic metasediments in sequence with dated volcanics	Direct: ca. 3.78 Ga Sm-Nd age (with mafics); Indirect: zircon dating of felsic volcanics, more reliable (ca. 3.70 Ga; 17)	Clastic metasediments	Bulk $\delta^{13}\text{C}$ of about -19 ‰ consistent with a biogenic origin	viable
Microscale (SIMS) carbon isotopes	3.8 Ga	Ueno et al., 2002	Cherts >3.7 Ga; metacarbonate age unknown (<3.7 Ga)	as above	IGB metacarbonates not original sediments (13, 14); cherts more suitable.	Less extreme fractionation (most $\delta^{13}\text{C}$ >-15 ‰), could be abiogenic (18)	low

**Table 1 (cont.)**

Biomarker	Claim age	Primary reference	Assessment criteria				Reliability
			Field relationships	Geochronological constraints	Suitability of host	Evidence of biogenicity	
ISUA GREENSTONE BELT, WEST GREENLAND (cont.)							
Step-release carbon and nitrogen isotopes	3.8 Ga	Nishizawa et al., 2005	BIF	as above	Quartz magnetite BIF; suitable host	Bulk $\delta^{13}\text{C} \sim -30\%$ from high T release inferred to be in diagenetic Corg in mt.; $\delta^{15}\text{N}$ values inconclusive (cf. 19)	viable
Stromatolites	>3.7Ga	Nutman et al. 2016	metacarbonate (dolomite) in metasediment-metavolcanic sequence	as above	Suitable	Morphological ('cones'); 3D structural analysis indicates ridges produced by deformation (20)	discredited
AKILIA, GOTHÅBSFJORD, WEST GREENLAND							
Microscale carbon isotopes	>3.86 Ga	Mojzsis et al., 1996	metaBIF?	Indirect: ca. 3.65 Ga (21, 22); debatably cut by quartz diorite at ca. 3.86 Ga; age from possibly xenocrystic zircon (21, 23 cf. 24, 25, 26)	Controversial; chemical sediment (21, 27, 28) or metamorphosed igneous rocks (22, 29, 30, 31)?	Graphite inclusions in apatite; graphite not found in some subsequent studies (32, 33 cf. 34); apatite is young (35, 36).	discredited
NUVVUAGITTUQ, QUEBEC							
Microfossils and carbon isotopes	$\geq 3.77$ Ga	Dodd et al., 2017	Altered and metamorphosed ocean floor rocks intruded by trondhjemite	Indirect: zircon dating of intruding trondhjemite: ca. 3.77-3.75 Ga (37)	Fe-rich metasediments (hydrothermal seafloor vent deposits) in ocean floor mafic gneisses and chemical metasediments; amphibolite-facies metamorphism	$\delta^{13}\text{C}$ down to -15‰ in graphite associated with apatite	viable
SAGLEK BAY, LABRADOR							
carbon isotopes	>3.95Ga	Tashiro et al, 2017	Clastic metasediments assigned to pre-3.95 Ga supracrustals, based on unproven intrusion of ca. 3.9 Ga tonalite into correlated mafic rocks	Indirect: zircon dating of ca. 3.9 Ga tonalitic gneiss (38); Direct: pre-3.95 Ga age contradicted by ca. 3.2 Ga detrital zircon in same metasediments (39)	Graphite in clastic metasediments	$\delta^{13}\text{C}$ down to -25‰ in graphite from metapelite; graphite absent in enclaves with ironstone or BIF characteristic of older supracrustals	discredited (this study)

References:

(1) Thorpe et al., 1992; (2) van Kranendonk, 2006; (3) Brasier et al., 2015; (4) van Kranendonk et al., 2008; (5) Lopez-Martinez et al., 1992; (6) de Ronde et al., 1994; (7) Frei et al., 1999; (8) Nutman et al., 2009; (9) Appel et al., 2003; (10) Bridgwater et al., 1981; (11) Roedder, 1981; (12) Westall and Folk, 2003; (13) Rose et al., 1996; (14) Rosing et al., 1996; (15) van Zuilen et al., 2002; (16) Lepland et al., 2002; (17) Nutman and Friend, 2009; (18) Whitehouse and Fedo, 2007; (19) van Zuilien et al., 2005; (20) Allwood et al., 2018; (21) Manning et al., 2006; (22) Fedo et al., 2006; (23) Nutman et al., 1997b; (24) Myers and Crowley, 2000; (25) Whitehouse and Fedo, 2003; (26) Whitehouse and Kamber, 2005; (27) Dauphas et al., 2004; (28) Mojzsis et al., 2003; (29) Bolhar et al., 2004; (30) Fedo and Whitehouse, 2002; (31) Whitehouse et al., 2005; (32) Lepland et al., 2005; (33) Nutman and Friend, 2006; (34) McKeegan et al., 2007; (35) Whitehouse et al., 2009; (36) Chew et al., 2014; (37) Cates et al., 2013; (38); Shimojo et al., 2016; (39) Schiøtte et al., 1992.