Jaspers of the Løkken ophiolite

Jaspers are typically bedded cherts (silica-rich, microcrystalline rocks) rich in haematite \pm magnetite \pm Fe-silicate minerals. Stratiform occurrences of jasper with pillow basalts and volcanogenic massive sulphide deposits imply an origin from lithified hydrothermal precipitates ^{21,27,29}, formed in submarine settings analogous to those of modern midocean ridges or back-arc sea-floor spreading centres, intra-plate seamounts, or flanks and calderas of arc volcanoes ^{22,23,26}. Si-Fe colloidal-gel protoliths of jaspers can be attributed to several processes, including *in situ* precipitation from low-temperature (20-100°C ⁶⁰) fluid flow through oceanic crust ⁶¹, material derived from low-temperature (2-50°C) vent chimneys ⁶¹, and proximal or distal sedimentation from hydrothermal plumes ⁶².

The Early Ordovician (Tremadoc) Løkken ophiolite was deformed and emplaced during the Caledonian orogeny, reaching metamorphic grades of lower greenschist facies; the original setting was dominated by back-arc rifting in a marginal basin ⁶³. The ophiolite contains laterally extensive jaspilitic beds that cluster at stratigraphic levels equivalent to those of the Løkken and Høydal volcanogenic massive sulphide (VMS) deposits and are in places intimately associated with the Fe-Cu-Zn sulphide ore. The jasper, which formed originally as sea-floor gels, displays myriad exceptionally well-preserved primary textures, including haematitized bacteria remains, haematitic tubes, and a variety of textures attributed to gel maturation and more advanced diagenetic changes ⁶³. The gel was deposited by particle fallout from hydrothermal plumes, in which silica flocculation and rapid settling of colloidal particles was promoted by the bridging effect of Fe oxyhydroxide ²⁷. Fe was sourced from white smoker-type vents with high Fe/S ratios ⁶⁴, whereas silica is thought to be primarily of ambient seawater origin based on the interpretation of silica saturation or supersaturation in pre-Late Cretaceous oceans ²⁷. Trace element variations in the gel precursor were controlled by coprecipitation and/or adsorption by Fe oxyhydroxide particles within the plume(s), REE patterns (positive Eu and negative Ce anomalies) reflect mixing of hydrothermal solutions with seawater at dilution ratios of $\sim 10^2$ to 10^4 .

The Nuvvuagittuq Supracrustal belt (NSB)

The NSB is located in the Northeastern Superior Province, in northern Québec, Canada. It is surrounded and intruded by multiple generations of Eoarchaean to Palaeoarchaean tonalite-trondhjemite-granodiorite (TTG) gneisses dated at 3760 Myr, 3660 Myr, 3500 Myr, and 3350 Myr ^{65,66}, and forms a core enclave within the Neoarchaean TTG gneisses of the Inukjuak domain. The NSB is dominated by mafic amphibolites composed of cummingtonite-plagioclase-biotite±garnet¹⁷.

The amphibolite unit has been interpreted to represent mafic volcanic deposits, with compositions ranging from basaltic to basaltic andesite ¹¹. In some locations, pillow lava structures are preserved within the amphibolite ¹¹. These amphibolites are divided into three geochemical groups displaying distinct Al/Ti ratios and following a chemostratigraphy within the NSB ¹¹.The base of the sequence is characterised by low Al/Ti

metabasaltic rocks with tholeiitic affinities. They exhibit relatively flat trace element profiles and their chemistry is consistent with derivation from an undepleted mantle source and fractionation under relatively dry conditions¹¹. The top of the stratigraphy comprises amphibolites having geochemical compositions consistent with derivation from a reenriched depleted mantle and fractionation under elevated water pressure ¹¹. They comprise high Al/Ti basaltic amphibolite relatively depleted in incompatible trace elements and displaying characteristic concave-up REE profiles typical of modern boninites. They are overlaid by intermediate Al/Ti amphibolite of basaltic to andesitic compositions with calcalkaline affinities displaying LREE-enrichments and flat HREE profiles. Chemical sedimentary rocks (BIF and chert-like silica rocks) mark the transition in the stratigraphy between the low Al/Ti tholeiitic amphibolite and the higher Al/Ti boninitic and calcalkaline amphibolite. This transition and chemo-stratigraphy led some to suggest that the NSB metavolcanic rocks were formed in subduction initiation settings similar to those of modern convergent tectonic settings ⁶⁷. Locally, the NSB amphibolite is composed of a cordierite-orthoamphibole mineral assemblage and is characterised by high Mg and K concentrations with depleted Ca, Na, and Si. This mineralogical assemblage and geochemical signature are consistent with seawater hydrothermal alteration of the oceanic crust ⁶⁸. Together with the presence of pillow lavas and BIF, this pattern suggests that the NSB formed through submarine volcanism with important hydrothermal activity.

On the basis of mineralogy, two broad types of BIF lithologies are recognized from the NSB including quartz + ferrous silicates (banded silicate formation (BSF) and quartz + magnetite + ferrous silicates (BIF) ¹⁶. Other minor lithologies that have been identified in the belt are jasper-quartz-carbonate, quartz-biotite schists (with possible meta-conglomerates), and chromite-bearing silica rock ^{69, 12, 18, 19}.

Outcrops of BIF vary from 5 to 30m in thickness, with the jasper forming thinner beds of 1 m thickness and less, commonly interbedded with layers of amphibolite. In places, small veins, typically a few cm wide of jasper infiltrate and cut the amphibolite ¹⁹, suggesting Fe-Si gels infilled networks within the (interpreted) pyroclastics during deposition. The depositional depth of the NSB jaspers is uncertain from the associated lithologies or textures in rocks of the belt; however, the occurrence of fine-scale, iron-oxide lamination (Fig. E3A) is similar to that in other younger jaspers and IFs suggestive of a calm depositional environment below wave base.

The NSB was mostly metamorphosed to upper amphibolite facies with temperatures reaching 650 °C and 4-5 Kbar ^{17, 20}. Sm-Nd isotopic compositions of garnets in the NSB amphibolites suggest that the peak metamorphic event occurred in the Neoarchaean ¹⁴ contemporaneous with intrusion of pegmatites dated at 2688 ± 2 Myr ⁶⁶. This is also consistent with the regional metamorphism occurring at 2705–2680 Myr ⁷⁰. The amphibolites in the southwest and southeast corners of the NSB are characterised by lower metamorphic grade assemblages of chlorite+epidote+actinolite. To the southeast, chlorite preserves the shape of what appears to have been garnet crystals, suggesting that the lower greenschist assemblage is retrograde ¹⁷. No evidence of retrogressed garnet is observed in the lower grade facies to the southwest, which may never have reached upper amphibolite facies. Very local layers in Fe-cherts preserve primary chert, calcite rhombohedra, and well-formed minnesotaite needles in jasper, suggesting some rare layers escaped severe recrystallisation during peak metamorphism. Furthermore, the crystallisation temperatures estimated for graphitic carbon in rocks from the southwest corner of the NSB do not exceed 550°C (Table S9-10), suggesting that the metamorphic facies did not exceed much past greenschist facies.

The geochronology of the NSB is highly debated and two ages have been proposed for the contained metavolcanic rocks: an Eoarchaean age of \sim 3750 Myr ⁶⁹ and a Hadaean age of \sim 4280 Myr ¹⁵. Here we present an overview of this geochronological debate.

One of the challenges in precisely dating the formation of the NSB is the quasiabsence of zircon-bearing felsic rocks and their equivocal relationship to the mafic metavolcanic rocks. Rare bands of felsic rocks with trondhjemitic composition have yielded zircons dated by U-Pb between 3750 and 3774 Myr^{69, 66, 65, 13, 71}. These trondjhemitic rocks consist of thin discontinuous bands 25-40 cm in width found only at the southern edge of the NSB 17. Other interpretations placed these rocks as felsic intrusions due to cross-cutting relationships observed in the field ⁶⁹, thus giving a minimum age for the NSB. Other field evidence such as the presence of felsic intrusions within gabbro sills were used to reach to the same conclusion about their intrusive nature ^{14,65}. Other works, however, interpreted these rocks as felsic volcanites ⁷¹, hence providing the actual age for the NSB. Significantly, this is inconsistent with the cross-cutting relationships observed by ⁶⁹. Fuchsitic quartz-rich rocks in the NSB have been interpreted as detrital quartzites by some works ¹² which yielded 13 zircons with ²⁰⁷Pb/²⁰⁶Pb ages ranging from 3718 Myr to 3780 Myr. These therefore put a maximum age for the NSB at 3780 Myr. However, this interpretation is inconsistent with the principle of inclusion, because the maximum age for the detrital quartzite should be constrained by the youngest 3718 Myr detrital zircon. This, however, is in conflict with the minimum age of 3770 Myr for the NSB supported by the age of the cross-cutting felsic rocks ⁶⁹. Other studies ascertained the same U-Pb age of 3794 ± 16 Myr for the fuchsitic rocks, which they conversely interpreted as metasomatically altered felsic orthogneiss of intrusive origin ¹³, hence providing a minimum age for the NSB. A Hadaean age for the NSB is supported by the ¹⁴²Nd isotopic composition of the mafic amphibolites. ¹⁴²Nd is the daughter product of ¹⁴⁶Sm, and because the parent isotope has a short half-life of 103 Myr, any deviation in ¹⁴²Nd compared to modern terrestrial standards implies Sm-Nd fractionation before ca.

4000 Myr, prior to extinction of ¹⁴⁶Sm. A significant number of NSB amphibolites exhibit anomalous ¹⁴²Nd compositions compared to modern terrestrial standards. The correlation

between the ¹⁴²Nd/¹⁴⁴Nd and Sm/Nd ratios for all three groups of NSB amphibolite is interpreted by some ^{14,15} to represent an isochron giving an igneous age of ca. 4290 Myr for the NSB metavolcanic rocks. This Hadaean age has been challenged by some who interpreted the ¹⁴²Nd/¹⁴⁴Nd vs. Sm/Nd correlation to reflect mixing with a Hadaeanenriched reservoir ⁷². However, this correlation is preserved between the mafic amphibolite and their ultramafic co-genetic cumulates, which can be used to argue against mixing as a cause of the ¹⁴²Nd/¹⁴⁴Nd vs. Sm/Nd correlation ¹⁴. The composition of these ultramafic rocks is controlled by olivine fractionation and they are interpreted to be co-genetic cumulates to the mafic amphibolites ¹¹. The ¹⁴²Nd/¹⁴⁴Nd vs. Sm/Nd correlation between the hypothesised mafic liquids and ultramafic cumulates can only be produced if the igneous fractionation occurs while ¹⁴⁶Sm was still decaying, i.e. prior to 4000 Myr. A Hadaean age for the NSB is also supported by the ¹⁴⁷Sm-¹⁴⁴Nd isotopic composition of intruding gabbro sills that yield an isochron age of ca. 4100 ± 100 Myr, which would establish a minimum age for the NSB metavolcanic rocks ¹⁴.

Hence, there is still no consensus on the exact age of the NSB, but nonetheless, it is at least 3770 Ma and possibly as old as 4290 Myr, which makes the NSB BIF among the oldest, if not the oldest, chemical sedimentary rocks known on Earth.

Jaspers and BIFs metamorphosed at the upper greenschist/lower amphibolite facies

Eoarchaean supracrustal belts worldwide are generally metamorphosed to at least upper amphibolite facies. The metamorphic mineral assemblages of BIFs are well-documented ⁷³. Petrographic and quantitative analysis of elements in Fe-silicate minerals within BIFs can be used to constrain metamorphic facies. In the BIFs of the NSB, the highest-grade samples exhibit extensive recrystallization and dissemination of coarse magnetite layers, and contain fayalite and pyroxene that indicate amphibolite- to granulite-facies conditions. With decreasing grades, the BIFs lack fayalite and instead contain minor pyroxene, amphiboles (predominantly grunerite-cummingtonite), and retrograde phyllosilicates (clinochlore). The low-grade BIFs contain patches of fine and coarse grained quartz. The very lowest grade BIFs have acicular stilpnomelane, minnesotaite, and rhombohedral ferroan calcite (Fig. E3D). The stilpnomelane in lower grade BIF is likely prograde, because this mineral does not form massive pseudomorphs after higher grade minerals, but rather fine euhedral laths. An upper stability field of 430-470°C and 5-6 Kbar is proposed for stilpnomelane ⁷⁴. Furthermore, the presence of chert suggests that the P-T conditions of these prograde stilpnomelane--bearing samples did not exceed much past upper greenschist facies.

Null hypothesis and abiotic formation of haematite tubes and filaments

The null hypothesis in the context of early life studies requires one to assess the likelihood of plausible abiotic explanations for observations purporting to support a biological phenomenon. If all plausible abiotic explanations are considered and assessed to be significantly unlikely, then one can reject the null hypothesis leaving a biological interpretation for the observed features. Below we consider the possible abiogenic mechanisms that could have created haematite tubes and filaments akin to those documented in the NSB.

First, the NSB tubes rarely branch, are commonly straight, and exhibit parallel orientation that may reflect preferred mineral growth. If iron was remobilised along crystal boundaries of acicular minerals as they grew during diagenesis and metamorphism, tube-like structures of haematite could have formed. However, if the precursor was not opal then the mineral would need to have been replaced completely, because only quartz is found inside the tubes. Secondly, acicular mineral growth in rocks is not uniform in shape, in contrast to the consistent morphology observed for the tubes.

Likewise, metamorphic processes may elongate quartz-hosted haematite to produce aligned, tubular structures like the tubes in the NSB, through fluctuating volume change in the rock during silica remobilisation or metamorphic stretching. Such processes would lead to the structures and minerals in the local environment being preferentially and consistently aligned, yet the orientation of ribbons and tubes of nanoscopic haematite (Fig. 1C) in the NSB jasper is highly variable over scales from hundreds of microns to millimetres. However, the presence of 500-µm gaps in iron-oxide layers (Fig. E3B) showing strong alignment of tubes and ribbons of haematite raises the possibility that during metamorphism elevated strain pressure pried apart these iron-oxide layers, which subsequently were infilled with silica and haematite that nucleated on the iron-oxide layers and were elongated during continued layer separation. In addition, layers and pre-existing amorphous structures of haematite may have, under pressure, undergone a process similar to neurulation, whereby layers of haematite are pressed together to form tubes. The two mechanisms above could, in theory, form consistently sized tubes in the range of $16-30 \,\mu m$ by reaching a critical size limit, but it is highly unlikely that such mechanisms can facilitate the growth of multiple tubes from a single haematite knob at varying angles (Fig. 1A, E4), both horizontally and vertically during prograde metamorphism, together with the formation of internal filaments and similarly coiled, branched, and twisted filaments (Fig. E4) with delicate haematite envelopes (Fig. 1C). In addition, their close spatial association with carbonate and graphite (Fig. E5D) is most compelling and consistent with a biogenic origin for these tubes and filaments.

Because formation of the NSB tubes via the above metamorphic processes fails to fully explain the NSB tubular structures as metamorphic products, a primary origin of the tubular structures is considered more probable. Nonetheless, there still exist exotic processes that may produce pseudo-microfossils under certain conditions⁷⁵. However, these known exotic processes form filaments of silica and barium carbonate and form under very alkaline conditions, which are unlikely to be relevant for the sea-floor environment of deposition of the NSB jaspers (which lack barite) and formation of the contained tubes. Similar iron-oxide tubes like those present in the NSB have been synthesised using iron-ammonium-sulphate solutions and electric currents, or via mixing

of hydrogen with ammonia bubble streams ⁷⁶. Notably, these synthesised tubes are demonstrably different from those in the NSB and younger jaspers. Firstly, the experimental tubes have solid walls of iron oxide and consist of a complex mixture of Fe minerals that vary depending on the nature of the bubble solution, whereas the NSB tube walls are composed only of dispersed, individual, nanoscale grains of haematite, consistent with the recrystallization of primary Fe-oxyhydroxide precipitates from seawater. Secondly, the experimental tubes are variable in size, which contrasts with the generally consistent size of the NSB tubes; yet, a number of tubes in the NSB jasper are strongly deformed and display irregular forms and sizes (Fig. 2D). Finally, the synthesised tubes lack internal chains of platy haematite or any other phase, unlike the NSB tubes.

Whereas tubular structures in modern and lithified hydrothermal precipitates are interpreted almost exclusively as microfossils, some branching filaments have been attributed to inorganic, self-organized mineral growth of Fe-oxyhydroxide, controlled by redox fronts in a silica gel⁷⁷. However, such structures are highly branching and are not reported to occur as tubes, but instead as solid Fe-oxyhydroxide filaments, and hence are debated in terms of biological origin²⁸. Similarly, Fe- oxyhydroxide filaments could form abiogenically in a Fe-Si gel and then be coated subsequently by colloidal silica and Feoxyhydroxide to form tubes with internal filaments, like those observed in the NSB. However, this process fails to explain the tubes that lack internal filaments, because silica has never been proven to replace haematite, and fails to explain their associations with carbonate and CM; thus, the tubes are unlikely to have formed via this process. Rather, in our interpretation, silica precipitated on and replaced bacterial filaments, which were then coated with nanoscale Fe-oxyhydroxide to form the tubes, while minor amounts of microbial organic matter from the filaments was oxidised to carbonate that are now preserved as carbonate-graphite associations with the filaments (Fig. E5). In summary, the NSB haematite tubes and filaments cannot be explained fully by any known abiotic mechanism, and in light of their association with multiple, independent lines of evidence supporting biological activity, the simple uniting explanation is that these tubes and filaments represent fossilised microbial remains.

Table S1 Laser Ablation parameters

Wavelength Energy fluency	193 nm Pulse duration 2.5 J/cm2	20 ns
Ablation spot size	25 μm	
Ablation time / spot	10 Hz 25 / 30 s (U-Pb / trace element	nts)

LA-ICP-MS instrument parameters

RF power 1340 W Ar Carrier Gas ow 1.04 l/min He gas ow 950 ml/min Sweep Time 400 / 900 ms (U-Pb / trace elements)

Table S2 Acquired masses for U-Pb dating. 235U was ommitted, natural 238U/235U ratio of 137.88 is assumed.

Mass	Element	Dwell time
29	Si	5ms
43	Ca	5ms
44	Ca	5ms
206	Pb	100ms
207	Pb	200ms
208	Pb	100ms
221	bkg	5ms
232	Th	100ms
238	U	100ms

Table S3 Acquired masses for trace elements.

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Mass	Element	Dwell time
23	Na	5ms
27	Al	5ms
29	Si	5ms
43	Ca	5ms
44	Ca	5ms
55	Mn	10ms
88	Sr	50ms
89	Y	50ms
137	Ва	50ms
139	La	50ms
140	Ce	50ms
141	Pr	50ms
146	Nd	50ms
147	Sm	50ms
153	Eu	50ms
157	Gd	50ms
159	Tb	50ms
163	Dy	50ms
165	Но	50ms
166	Er	50ms
169	Tm	50ms
172	Yb	50ms
175	Lu	50ms
221	background	5ms

Formation	Samples	Age	General rock	Optical	Raman	SEM/	FIB	LA- ICP-	Samples	locations
			type	meroscopy	meroscopy	LFIMA		MS	Northings	Eastings
Løkken ophiolite, Høydal jaspers, Norway	HO6A, HO7B, TG-82-S6, TG-82- H19 and H16, Jah-1 and 2	0.485 Ga	Jasper	х	x	х	K Mine tail		ailings	
Animike, Mesabi range, Biwabik Iron Formation, USA	MARY-ELLEN1	1.8 Ga	Stromatolitic X X jasper X X		Minet	ailings				
Brockman formation, Dales gorge group, Australia	DGH1_273'1"	2.5 Ga	BIF/ Fe- cherts	x	х	х			DGH1 drill core	
	PC0822, PC0823, PC0824								58 16 50.9	77 44 14.7
	PC0825								58 17 08.7	77 44 12.2
NSB	PC028/A	>3.77 Ga	BIF/ jasper	х	х	х	х	х	58 17 09.1	77 44 10.8
	PC0844								58 16 53.8	77 44 14.1

Table. S4 List of samples and their locations and methods used in this study

Table S5. Iron-oxide tubes and filaments

Locality	Age	Host rock	Diameter Length	Mineralogy	Additonal	Reference
Lau hydrothermal basin, Pacific ocean	Present day	Fe-Si hydrothermalvent sediments	0.5-1.5μm 10-100μm	Iron-oxide-silica	Straight, helicaland curved varieties. Core of filaments iron-oxide coated by amorphoussilica	78
Chocolate pot hot springs, Yellowstone, USA	Present day	Iron-rich hydrothermal springs	1.5μm >100μm	Ferrihydrite	Very low Total organic carbon present in buried microbial mats	79
Southwest Indian Ridge, Indian Ocean	Present day	Fe-Si oxyhydroxide	2-10μm 10-100μm	Ferrihydrite	Filaments can be curved and intertwining or form bundles of rod-like filaments with the same orientations, diameter and length	80
Franklin Seamount, Woodlark Basin, Papua New Guinea	Present day	Fe-Si-Mn oxyhydroxides	0.5-5µm	Iron-oxide-silica	Filaments have hollow structure with walls of iron oxyhydroxide. Filaments branch and some occur as orientated parallel to one another	24
Various ophiolites and modern day vent sites	Late Jurassic to present day	Inter-pillow cherts, jasper, Fe-Si seafloor gels and inactive chimney fragments	1-10 µm	Iron-oxide-silica-pyrite- barite		22
Numerous deposits	490 Ma to Present day	Japsers	1.2-30 μm 20-3000 μm	haematite-iron-oxide	See tables 1, 2 and 3 in Little et al., 2014. Tubes with internal platy haematite chains (Løkken deposit)	28
Qigebulake Formation, Northwestern Tarim Basin, China	551–541 Ma	Hydrothermal quartz veins	1-5 μm 20-200μm	Goethite-haematite	No organic carbon detected by Raman	80
Jerome mining district, central Arizona, USA	1738 Ma	Jasper-IF-VMS	1-3 μm 30-50 μm	haematite		81
Frere formation, Australia	1880 Ma	Jasper/IF	Tubes 4-9 μm Filament 0.5-2 μm 100s μm	Nanoscale haematite	Filaments orientated parallel to oncolites	50
Gunflint iron-formation, North America	1880 Ma	Jasper/IF	0.5-8 μm Can exceed 300 μm	Nanoscale haematite	Same filament morphologies in other areas of the formation are preserved by organic matter	82,83
Kittilä jaspers, Finland	ca. 2000 Ma	Jasper	?	Nanoscale 'dusty' haematite		84
Hamersley BIF, Dales gorge member, Australia	2500 Ma	BIF (haematite + quartz + magnetite + stilpnomelane + ankerite + minnesotaite + riebeckite)	5 μm 100 μm	haematite-dolomite	Filaments preserved as carbonate moulds attached to iron spheres	85
Nuvvuagittuq supracrustal belt, Canada (filaments)	3750 Ma	Jasper (haematite + quartz + magnetite + ripidolite + chalcopyrite ± calcite ± apatite)	1-7 μm 35-500 μm	Platy-haematite, acicular, grey haematite, moulds in massive nanoscale haematite	Filaments commonly encapsulated by nanoscale haematite. Some have a forked-end. Others exhibit branching	This study
Nuvvuagittuq supracrustal belt, Canada (tubes)	3750 Ma	Jasper (haematite + quartz + magnetite + ripidolite + calcite + chalcopyrite + apatite)	20-30 μm 80-300 μm	Nanoscale haematite	Some contain a central chain of platy haematite	This study

Table S6. Carbonate rosettes in IFs

Locality	Age	Hostrock	Diameters	Carbonate	Additional	Reference
Høydal, Norway	480 Ma	Jaspers associated with VMS deposits	100-250 μm	Ankerite	Form circular and rhombohedra rosettes, overgrow haematite filaments; can contain inclusions of CM and associated with apatite	26; This study
Gunflint iron-formation, North America	1880 Ma	Jasper/IF	20-40 μm	Siderite	Associated with CM and sometimes contain cores of apatite	85, 86, 41, 87
Hamersley BIF, Dales gorge member, Australia	2500 Ma	BIF (haematite + quartz + magnetite + stilpnomelane +	100-300 μm	Ankerite-siderit	Generally rhombohedralin shape, with poikibitic e textures developing, which form a sphere-shaped centre. Associated with CM layers	This study
		ankerite + minnesotaite + riebeckite)	60-80 μm	Dolomite- ankerite	Sometimes have a rosette structure in the middle	88
Nuvvuagittuq supracrustal belt, Canada	>3770 Ma	BIF (Calcite + quartz + magnetite + stilpnomelane + haematite + apatite + chalcopyrite)	20-180µm) Fe-calcite	Poikiolitic, rosettes generally rounded, associated with apatite, can contain inclusions of CM and haematite	This study 18

Table S7. Iron-oxide rosettes

Locality	Age	Host rock	Diameters	Mineralogy	Additional	Reference
Lau hydorthermal basin, Pacific ocean	Present day	Fe-Si hydorthermal vent sediments	0.1-2 µm	Si-Ferrihydrite	Associated branching filaments	90, 23
Edmond vent field, Indian ocean	Present day	Inner surface of sulphide chimney	1 µm	Non-crystalline silica and acicular iron oxides	Mineralized cells, with cytoplasm preserved	91
Milos, Greece	2Ma	Hydrothernal vent iron- formation	30 µm	haematite	Associated filaments, crumpled cores	33
Lokken-Hoydal, Norway	480Ma	Jaspers associated with VMS deposits	10-100 µm	haematite-quartz	Forms mm thick layers, large diversity, haematite cores, multi- layered varieties	27
Mount Windsor, Australia	ca. 485Ma	Jaspers associated with VMS deposits	100 µm	haematite-quartz	Multi-layered varieties	29
Frere formation, Australia	1850 Ma	Jasper/ IF	20-50 μm	haematite-quartz	haematite cores and associated haematite filaments	This study
Gunflint iron-formation, North America	1880 Ma	Jasper/ IF	3-30 µm	haematite-quartz		82, 86
Sokoman formation, Canada	1900 Ma	Jasper	3-25 μm	haematite-quartz		86, 92, 93
Great Slave group	Paleoproterozo ic	IF	10-15 μm	haematite-quartz		This study
Kittilä jaspers, Finland	ca. 2000 Ma	Jasper	?	haematite		84
Boolgeeda formation, Australia	2400 Ma	BIF (haematite + quartz + magnetite + riebeckite + stilpnomelane + goethite + apatite)	10-15 µm	haematite-quartz	Forms mm thick layers	This study
		BIF (haematite + quartz +		haematite-quartz		94
Hamersley BIF, Dales gorge member, Australia	2500 Ma	magnetite + stilpnomelane + ankerite + minnesotaite + riebeckite)	5-20µm	Stilpnomelane and opaque pigments		89
Hamersley BIF, Marra Mamba member, Australia	2500 Ma	BIF (haematite + quartz + magnetite + stilpnomelane + ankerite + minnesotaite + riebeckite)	120-200 nm	haematite-quartz		95
Carajás formation, Brazil	2740 Ma	BIF (haematite + quartz + magnetite)	20-30 µm	haematite-quartz-kerogen		96
Nuvvuagittuq supracrustal belt, Canada	>3770 Ma	BIF (haematite + quartz + magnetite + ripidolite + calcite + chalcopyrite + apatite)	15-30 μm 60-100 μm	haematite-quartz	Small and large varieties. Some have haematite cores and concentric layers	This study

Table S8 Wavelength-dispersive spectroscopy (WDS) analysis of various minerals in the NSB BIF and jaspers.

										FeO				
Mineral (Sample*)	F	Cl	Na ₂ O	K ₂ O	MgO	SiOz	Al ₂ O ₃	CaO	P2O5	(Total Fe)	MnO	ZnO	TiO ₂	Total
Fluro-apatite (E-F)	1.6	0.1	0.0	0.0	0.0	0.2	0.0	56.2	25.3	0.1	0.0	0.0	0.0	82.9
Fluro-apatite (E-F)	1.4	0.3	0.0	0.0	0.0	0.5	0.0	53.2	26.0	0.2	0.0	0.0	0.0	81.1
Stilpnomelane (E-F)	0.0	0.0	0.0	1.1	4.6	47.9	4.7	0.0	0.0	33.8	0.3	0.0	0.0	92.6
Stilpnomelane (E-F)	0.0	0.0	0.0	1.2	4.5	48.3	4.8	0.0	0.0	34.3	0.3	0.0	0.0	93.4
Magnetite (E-F)	0.0	0.0	0.0	0.0	0.0	0.6	0.1	0.2	0.0	94.3	0.0	0.1	0.0	95.3
Fe-calcite (E-F)	0.0	0.0	0.1	0.0	0.3	0.3	0.3	69.9	0.0	2.8	1.0	0.1	0.0	74.7
Ankerite (D)	0.0	0.0	0.0	0.0	0.4	0.0	0.0	69.6	0.0	4.8	1.6	0.0	0.2	76.4
Mg-ankerite (D)	0.0	0.0	0.0	0.0	16.2	0.2	0.0	30.5	0.0	16.9	1.1	-	-	64.8

*Sample makes reference to thin sections in Figure E3. Low totals attributed to no analysis for REE, hydroxyl and carbon.

Table S9 Raman parameters of graphitic carbon in carbonate rosettes (Fig. E6C).

											T estimate
	G-band	G-band	D-band	D-band	2D-band	2D-band	D-band	G+D2 band	D/G	2D/ G	Beyssac ⁵⁵
Grain No.	position	FWHM	position	FWHM	position	FWHM	area	area	intensity	intensity	± 50°C
G1	1576	9.78	1339	6.25	N.D	N.D	222.69	749.98	1.32	N.D	539
G2	1567	9.80	1341	9.97	2702	5.41	238.21	803.49	0.32	0.23	539
G3	1565	10.57	1350	6.13	2704	4.71	358.58	640.57	0.41	0.48	481
G4	1579	7.31	1345	11.25	2704	5.54	299.53	369.09	0.56	0.11	441
G5	1557	10.61	1337	9.58	2674	7.94	87.27	355.61	0.13	0.2	553
G6	1569	12.04	1334	15.74	2684	9.22	66.72	158.05	0.61	0.36	509
SG1	1584	5.63	1343	5.77	2708	4.46	140.11	672.22	0.08	0.05	564
SG2	1569	8.98	1331	6.60	2680	5.58	160.64	586.33	0.13	0.12	545
SG3	1577	6.91	1343	12.06	2702	5.66	162.30	500.62	0.18	0.14	532
SG4	1577	6.80	1334	7.68	2696	4.67	167.80	515.40	0.56	0.13	532
SG5	1583	5.65	1347	13.18	2702	5.26	284.12	831.28	0.24	0.05	528
SG6	1569	10.56	1344	6.87	2705	4.95	127.96	367.99	0.44	0.44	526
SG7	1570	10.22	1346	9.73	2725	6.82	122.76	278.79	0.34	0.22	505
SG8	1577	6.29	1345	15.36	2700	5.09	365.76	711.13	0.30	0.16	490
SG9	1565	7.64	1342	7.43	2668	5.21	1203.96	2265.20	0.23	0.08	487
SG10	1580	7.19	1349	7.26	2704	4.58	227.73	371.57	0.28	0.21	472
SG11	1570	8.28	1339	6.63	2705	5.06	604.43	937.62	0.29	0.31	467
SG12	1579	7.32	1345	11.30	2704	5.63	322.68	346.98	0.56	0.12	442
SG13	1583	6.17	1344	12.66	2717	6.58	105.02	119.06	0.37	0.20	432
SG14	1581	10.16	1344	14.20	2676	6.20	657.67	537.29	1.10	0.27	396

* Grains starting with G- are from the Raman scan in Fig 3D and grains starting with SG- are from the Raman scan in Fig E6.

Table S10 Raman parameters of graphitic carbon from NSB jasper sample (Fig. E8E).

G-band position	G-band FWHM	D-band position	D-band FWHM	2D-band position	2D-band FWHM	D-band area	G+D2 band area	D/G intensity	2D/ G intensity	T estimate Beyssac⁵⁵ ±50°C
1581	6.18	1359	12.70	2715	9.01	63.77	260.57	0.10	0.24	554
1583	7.29	1348	14.30	2709	10.33	87.76	141.04	0.46	0.51	470
1581	7.03	1346	11.50	2714	12.43	79.70	442.69	0.15	0.25	573

Table S11 Raman parameters of poorly crystalline graphite from NSB. NR - Not resolvable (Fig. E8F).

											T estimate
Spectrum No.	G-band position	G-band FWHM	D-band position	D-band FWHM	2D-band position	2D-band FWHM	D-band area	G+D2 band area	D/G intensity	2D/ G intensity	Beyssac ⁵⁵ ± 50°C
1	1585	9.38	1351	8.98	2692	17.55	2614.56	1701.18	1.66	0.45	371
2	1591	7.30	1352	6.15	2701	4.66	1238.30	788.34	2.14	0.14	369
3	1591	7.47	1355	10.10	NR	NR	169.90	87.62	1.91	NR	347

Supplementary data references

- 60 Mills, R. A., Clayton, T. & Alt, J. C. Low-temperature fluid flow through sulfidic sediments from TAG: modification of fluid chemistry and alteration of mineral deposits. *Geophysical research letters* **23**, 3495–3498 (1996).
- 61 Alt, J. C. Hydrothermal oxide and nontronite deposits on seamounts in the eastern Pacific. *Marine Geology* **81**, 227-239 (1988).
- 62 Mills, R. A., Elderfield, H. & Thomson, J. A dual origin for the hydrothermal component in a metalliferous sediment core from the Mid-Atlantic Ridge. *Journal of Geophysical research* **98**, 9671-9681 (1993).
- 63 Grenne, T. & Slack, J. F. Paleozoic and Mesozoic silica-rich seawater: Evidence from hematitic chert (jasper) deposits. *Geology* **31**, 319-322 (2003).
- 64 Grenne, T. & Slack, J. F. Geochemistry of Jasper Beds from the Ordovician Løkken Ophiolite, Norway: Origin of Proximal and Distal Siliceous Exhalites. *Economic Geology* **100**, 1511-1527 (2005).
- 65 O'Neil, J., Boyet, M., Carlson, R. W. & Paquette, J.-L. Half a billion years of reworking of Hadean mafic crust to produce the Nuvvuagittuq Eoarchean felsic crust. *Earth and Planetary Science Letters* **379**, 13-25, doi:10.1016/j.epsl.2013.07.030 (2013).
- 66 David, J., Godin, L., Stevenson, R., O'Neil, J. & Francis, D. U-Pb ages (3.8-2.7 Ga) and Nd isotope data from the newly identified Eoarchean Nuvvuagittuq supracrustal belt, Superior Craton, Canada. *Geological Society of America Bulletin* **121**, 150-163, doi:Doi 10.1130/B26369.1 (2009).
- 67 Turner, S., Rushmer, T., Reagan, M. & Moyen, J. F. Heading down early on? Start of subduction on Earth. *Geology* **42**, 139-142, doi:10.1130/g34886.1 (2014).
- 68 Franklin, J. M., Gibson, H. L., Jonasson, I. R. & Galley, A. G. in *Economic Geology: One Hundredth Anniversary Volume* (eds J.W Hedenquist, J.F.H Thompson, R.J Goldfarb, & J.P Richards) 523-560 (The Economic Geology Publishing Company, 2005).
- 69 Cates, N. L. & Mojzsis, S. J. Pre-3750 Ma supracrustal rocks from the Nuvvuagittuq supracrustal belt, northern Québec. *Earth and Planetary Science Letters* **255**, 9-21, doi:10.1016/j.epsl.2006.11.034 (2007).
- 70 Boily, M., Leclair, A., Maurice, C., Bédard, J. H. & David, J. Paleo- to Mesoarchean basement recycling and terrane definition in the Northeastern Superior Province, Québec, Canada. *Precambrian Research* 168, 23-44, doi:10.1016/j.precamres.2008.07.009 (2009).
- 71 Augland, L. E. & David, J. Protocrustal evolution of the Nuvvuagittuq Supracrustal Belt as determined by high precision zircon Lu–Hf and U–Pb isotope data. *Earth and Planetary Science Letters* **428**, 162-171, doi:10.1016/j.epsl.2015.07.039 (2015).
- 72 Roth, A. S. G. *et al.* Inherited 142Nd anomalies in Eoarchean protoliths. *Earth and Planetary Science Letters* **361**, 50-57, doi:10.1016/j.epsl.2012.11.023 (2013).
- 73 Klein, C. Some Precambrian banded iron-formations (BIFs) from around the world: Their age, geologic setting, mineralogy, metamorphism, geochemistry, and origins. *American Mineralogist* **90**, 1473-1499, doi:10.2138/am.2005.1871 (2005).
- 74 Miyano, T. & Klein, C. Phase equilibria in the system K20 FeO MgO AlzO3 SiO2 H20 CO2 and the stability limit of stilpnomelane in metamorphosed Precambrian iron-formations. *Contributions to Mineralogoy and Petrology* **102**, 478-491 (1989).
- 75 Garcia-Ruiz, J. M. *et al.* Self-assembled silica-carbonate structures and detection of ancient Microfossils. *Science* **302**, 1194-1197 (2003).
- 76 Stone, D. A. & Goldstein, R. E. Tubular precipitation and redox gradients on a bubbling template. *Proceedings of the national academy of science* **101**, 11537-11541, doi:10.1073/pnas.0404544101 (2004).
- 77 Hopkinson, L., Roberts, S., Herrington, R. & Wilkinson, J. Self-organization of submarine hydrothermal siliceous deposits: Evidence from the TAG hydrothermal mound, 26N Mid-Atlantic ridge. *Geology* 26 (1998).
- 78 Li, J. *et al.* Microbial diversity and biomineralization in low-temperature hydrothermal iron-silica-rich precipitates of the Lau Basin hydrothermal field. *FEMS Microbiology Ecology* **81**, 205-216, doi:10.1111/j.1574-6941.2012.01367.x (2012).
- 79 Parenteau, M. N. & Cady, S. L. Microbial Biosignatures in Iron-Mineralized Phototrophic Mats at Chocolate Pots Hot Springs, Yellowstone National Park, United States. *Palaios* **25**, 97-111, doi:10.2110/palo.2008.p08-133r (2010).

- Zhou, X. *et al.* Biogenic Iron-Rich Filaments in the Quartz Veins in the Uppermost Ediacaran
 Qigebulake Formation, Aksu Area, Northwestern Tarim Basin, China: Implications for Iron Oxidizers in
 Subseafloor Hydrothermal Systems. *Astrobiology* 15, 523-537, doi:10.1089/ast.2014.1234 (2015).
- 81 Slack, J. F., Grenne, T., Bekker, A., Rouxel, O. J. & Lindberg, P. A. Suboxic deep seawater in the late Paleoproterozoic: Evidence from hematitic chert and iron formation related to seafloor-hydrothermal sulfide deposits, central Arizona, USA. *Earth and Planetary Science Letters* **255**, 243-256, doi:10.1016/j.epsl.2006.12.018 (2007).
- 82 Barghoorn, E. S. & Tyler, S. A. Microorganisms from Gunflint Chert These Structurally Preserved Precambrian Fossils from Ontario Are Most Ancient Organisms Known. *Science* **147**, 563-&, doi:DOI 10.1126/science.147.3658.563 (1965).
- 83 Shapiro, R. S. & Konhauser, K. O. Hematite-coated microfossils: primary ecological fingerprint or taphonomic oddity of the Paleoproterozoic? *Geobiology* **13**, 209-224, doi:10.1111/gbi.12127 (2015).
- 84 Kinnunen, K. A. Primary sedimentary features in Kittila jasper, Finnish Lapland. *The geological society of Finland bulletin* **54**, 69-76 (1982).
- 85 Karkhanis, S. N. Fossil iron bacteria may be preserved in Precambrian feroan carbonate. *Nature* **261**, 406-407 (1976).
- Laberge, G. L. Possible Biological Origin of Precambrian Iron-Formations. *Economic Geology* 68, 1098-1109 (1973).
- 87 Winter, B. L. & Knauth, L. P. Stable isotope geochemistry of cherts and carbonates from the 2.0 Ga Gunflint Iron Formation: implications for the depositional setting, and the effects of diagenesis and metamorphism. *Precambrian Research* **59**, 283-313 (1992).
- 88 Carrigan, W. J. & Cameron, E. M. Petrological and stable isotope studies of carbonate and sulfide minerals from the Gunflint Formation, Ontario: evidence for the origin of early Proterozoic ironformation. *Precambrian Research* **52**, 347-380 (1991).
- Rasmussen, B., Meier, D. B., Krapez, B. & Muhling, J. R. Iron silicate microgranules as precursor sediments to 2.5-billion-year-old banded iron formations. *Geology* 41, 435-438, doi:10.1130/g33828.1 (2013).
- 90 Sun, Z. *et al.* Mineralogical characterization and formation of Fe-Si oxyhydroxide deposits from modern seafloor hydrothermal vents. *American Mineralogist* **98**, 85-97, doi:10.2138/am.2013.4147 (2012).
- 91 Peng, X. *et al.* Intracellular and extracellular mineralization of a microbial community in the Edmond deep-sea vent field environment. *Sedimentary Geology* **229**, 193-206, doi:10.1016/j.sedgeo.2010.06.003 (2010).
- 92 Knoll, A. H. & Barghoor, E. S. Ambient Pyrite in Precambrian Chert: New Evidence and a Theory. *Proceedings of the national academy of science* **71**, 2329-2331 (1974).
- 93 Klein, C. & Fink, P. R. Petrology of the Sokoma Iron Formation in the Howells R iver area, at the western edge of the Labrador trough. *Economic Geology* **71**, 453-487 (1976).
- 94 Ayres, D. E. Genesis of Iron-bearing Minerals in Banded Iron Formation Mesobands in The Dales Gorge Member, Hamersley Group, Western Australia. *Economic Geology* **67**, 1214-1233 (1972).
- Ahn, J. H. & Buseck, P. R. Hematite Nanospheres of Possible Colloidal Origin from a Precambrian
 Banded Iron Formation. *Science* 250, 111-113, doi:DOI 10.1126/science.250.4977.111 (1990).
- 96 Ribeiro da Luz, B. & Crowley, J. K. Morphological and chemical evidence of stromatolitic deposits in the 2.75Ga Carajás banded iron formation, Brazil. *Earth and Planetary Science Letters* **355-356**, 60-72, doi:10.1016/j.epsl.2012.08.028 (2012).