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Abstract: Metamorphosed Meso- to Neoarchaealuminous rocks in the central part of the island of Storø (Nuuk region, southern West Greenland), show field- and geochemical evidence of premetamorphic chemical alteration. This alteration changed basaltic precursors into aluminous lithologies, and following amphibolite grade metamorphism and penetrative ductile deformation, these garnet-biotite schists now resemble adjacent metapelitic schists of sedimentary origin. Mass balance calculations (isocon method), suggests that most major elements (Si, Fe, Mg, Ca, Na and P) were leached during alteration. The calculated overall net mass changes are between -18% and -45%, consistent with breakdown of olivine, pyroxenes, plagioclase and apatite in the basaltic precursor rocks. Major and trace elements such as, K, Cs, Rb, Ba, Pb, Zn, La, Ce were added during this alteration process, whereas high field strength elements (Ti, Al, Zr, Hf and Nb) remained essentially immobile and were thus residually enriched. Interestingly, Th which is generally assumed to be immobile in fluids, was also added during this process. These chemical changes reflect interaction between a basaltic protolith and hydrous fluids that established a new equilibrium and thus a different mineral assemblage. It is proposed that the premetamorphic alteration at Storø was due to low-temperature interaction between seawater and oceanic crust, and thus essentially represents in situ submarine seafloor weathering. This interpretation is consistent with the mass balances reported from well-documented examples in younger settings.

New U-Pb zircon geochronology from the arc-related mafic sequences at Storø shows that they comprise at least two distinct age groups: an older anorthosite complex dated at 3051.3 ± 2.6 Ma and a younger supracrustal sequence with age brackets between 2840-2710 Ma. The allochthonous nature of these two mafic igneous to sedimentary stacks is consistent with accretionary processes in island arc complexes and a compressional Archaean tectonic setting.

*Highlights (for review)

- Garnet-rich gneisses on Storø have similar immobile element ratios as amphibolites.
- These garnet-rich gneisses were derived from basalts by premetamorphic alteration.
- Isocon mass-balance calculations show similarities to modern seafloor weathering.

Mesoarchaealuminous rocks at Storø, southern West Greenland: new age data and evidence of premetamorphic seafloor weathering of basalts

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different mineral assemblage. It is proposed that the premetamorphic alteration at Storø was due to low-temperature interaction between seawater and oceanic crust, and thus essentially represents *in situ* submarine seafloor weathering. This interpretation is consistent with the mass balances reported from well-documented examples in younger settings.

New U-Pb zircon geochronology from the arc-related mafic sequences at Storø shows that they comprise at least two distinct age groups: an older anorthosite complex dated at 3051.3 ± 2.6 Ma and a younger supracrustal sequence with age brackets between 2840–2710 Ma. The allochthonous nature of these two mafic igneous to sedimentary stacks is consistent with accretionary processes in island arc complexes and a compressional Archaean tectonic setting.

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1. Introduction

Metavolcanic belts in Archaean cratons constitute a major source of Au and other metals worldwide (Goldfarb et al., 2001). The origin of the supracrustal host rocks and their common Au deposits have been much debated, especially in polymetamorphosed and deformed rocks where the protoliths are not easily recognisable (e.g. Groves et al., 2003). For instance, the metavolcanic rocks are commonly associated with garnet-rich aluminous schists that superficially appear to be of sedimentary origin, but which lack lithological layering akin to transposed bedding.

Both epithermal, orogenic and volcanogenic-massive-sulfide mineralisation processes are well understood and closely associated with specific types of hydrothermal alteration and controlled by host rock compositions and fabrics including porosity and permeability, as well as heat and fluid

50 sources, ambient temperature and hydrostatic pressure gradients (Hedenquist and Lowenstein, 1994;
51 Eilu and Groves, 2001; Herrington et al., 2005). Various low-temperature subaqueous and subaerial
52 weathering processes can also lead to significant chemical alteration (e.g. Alt and Teagle, 2003;
53 Polat et al., 2012). Due to this difficulty in identifying different alteration processes through
54 metamorphic overprint, several orogenic deposits have for example now been reinterpreted as
55 metamorphosed epithermal (and in a few cases volcanogenic-massive-sulfide) deposits (Hallberg,
56 1993; Penczak and Mason, 1999; Bonnet et al., 2005; McFarlane et al., 2007; Owens and Pasek,
57 2007; Garde et al., 2012). Such distinctions are important not least for future Au exploration.

58 Identifying different types of alteration in highly deformed and metamorphosed rocks can be
59 challenging, because the bulk composition of leached rocks may be similar to that of rocks derived
60 by sedimentary processes. Archaean aluminous schists and gneisses within supracrustal belts in the
61 North Atlantic craton of southern West Greenland have therefore been interpreted as
62 metasedimentary rocks (e.g. Rivalenti and Rossi, 1972), as various kinds of chemically altered
63 (meta)volcanic rocks (e.g. Beech and Chadwick, 1980; Dymek and Smith, 1990; Garde et al., 2007,
64 2012), or as a combination of both (Smith et al., 1992; Bolhar et al., 2005).

65 In this paper we describe and discuss the nature of chemical alteration of Archaean arc-related
66 mafic metavolcanic rocks on the island of Storø in Godthåbsfjord, in the central North Atlantic
67 craton of southern West Greenland. It is shown that a conspicuous component of aluminous rocks,
68 which superficially resembles adjacent metasedimentary rocks, likely formed during the interaction
69 between mafic rocks and cold seawater, implying that the combined sequence of mafic
70 metavolcanic and aluminous lithologies are likely to represent a section through Archaean oceanic
71 crust.

72 We also present new geochronological data, which corroborate that the central part of Storø
73 comprises at least three different tectonic slices of Eoarchaeoan to Neoarchaeoan crust including at

74 least two different age groups of mafic rock sequences that were juxtaposed during Neoarchaean
75 tectonic amalgamation of older magmatic arcs (e.g. Nutman and Friend, 2007).

76

77 **2. Geological setting**

78

79 The island of Storø is located in the central part of the largely Mesoarchaean North Atlantic
80 craton that forms most of southern Greenland and continues as the Nain Province in eastern
81 Labrador (**Fig. 1**; Schiøtte et al., 1989; Schiøtte and Bridgwater, 1990; St-Onge et al., 2009). In
82 southern West Greenland the craton has recently been divided into six tilted crustal blocks that
83 expose the root zones of several convergent andesitic island arc – continental arc systems and
84 oceanic supra-subduction zones (Garde, 2007; Polat et al., 2007; Windley and Garde, 2009; Szilas
85 et al., 2012, 2013a, 2013b). Storø itself is located at the south-eastern margin of the >3000 Ma
86 Fiskefjord block of Windley and Garde (2009), previously referred to as the Akia terrane (e.g.
87 Friend et al., 1996). The complex, collisional Godthåbsfjord–Ameralik belt to its south-east, which
88 also comprises the eastern part of Storø, consists of several smaller terranes of different origin and
89 age that were tectonically assembled and folded at around 2720 Ma (Friend et al., 1996). Following
90 the assembly of the Fiskefjord block and the terranes to the south-east, Storø and adjacent parts of
91 the Godthåbsfjord–Ameralik belt were intruded by c. 2650–2550 Ma granitoid crustal melts
92 including the 2550 Ma Qôrqt granite complex (Brown et al. 1981; Friend et al. 1996; Nutman and
93 Friend, 2007; Nutman et al., 2007, 2010). This resulted in an extensive network of granitic
94 pegmatites throughout the supracrustal rocks in central Storø.

95 The central part of Storø consists of a stack of supracrustal rocks of different age and
96 composition that have been referred to as the 'Storø supracrustal belt' or 'Storø greenstone belt' in
97 the literature, most recently by Scherstén et al. (2012). Existing age data quoted below are from

98 Scherstén et al. (2012) and stem from GEUS reports cited therein. The supracrustal rocks constitute
99 a major, in part refolded anticlinal structure with a NNW–SSE-trending hinge zone on the mutually
100 facing slopes of the Qingaaq and Aappalaartoq mountains (**Fig. 2**) and are cut by an inclined,
101 Neoarchaeal shear zone along the north-western side of the island. The northern part of the stacked
102 supracrustal and associated metagabbroic and anorthositic rocks on Aappalaartoq have been
103 intruded by Mesoarchaeal tonalitic orthogneiss. The south-eastern side of the stack is in presumed
104 tectonic contact with Eoarchaeal orthogneiss (previously termed Amîtsoq gneisses, now recognised
105 as part of the Itsaq gneiss complex; Nutman et al., 1996). The supracrustal stack includes (1)
106 metavolcanic tholeiitic amphibolite with island-arc geochemical signature yielding a metamorphic
107 U-Pb zircon age of c. 2630 Ma but with a currently unknown protolith age, (2) undated garnet-rich,
108 biotite- and sillimanite-bearing rocks that have previously been interpreted as either sedimentary or
109 altered metavolcanic rocks, and (3) metasedimentary mica schist and biotite gneiss with detrital
110 zircon grains yielding ages of ~2840 Ma, as well as both older and younger metamorphic grains
111 (van Gool et al., 2007). This supracrustal stack is in tectonic contact with (4) remnants of a
112 metagabbro–anorthosite complex intruded by c. 3050 Ma tonalitic orthogneiss.

113 The supracrustal rocks on Storø host a large, subeconomic Au occurrence that has been
114 investigated in detail since the 1990s by surface mapping, geochemical and isotopic studies, drilling
115 and 3D modelling (Grahl-Madsen, 1994; Trepka-Bloch, 1996; Skyseth, 1996; Appel et al., 2003;
116 Hollis, 2005; Scherstén et al., 2012). The occurrence of Au in the supracrustal rocks at central Storø
117 has been investigated in detail in recent years by the exploration company NunaMinerals A/S. Gold
118 mainly occurs in two zones on the north-east-facing slope of the mountain of Qingaaq (**Fig. 2**;
119 Østergaard and van Gool, 2007; Scherstén et al., 2012). The first, 'BD zone' is strata bound within
120 the above mentioned garnet-rich rocks. The second, 'Main zone' is located in the hinge zone of a
121 nearby Neoarchaeal anticlinal fold, where the Au is mainly confined to late quartz veins

122 (Østergaard and van Gool, 2007). The latter authors proposed a structural model that links
123 formation of the anticline with the above mentioned Neoarchaeon shear zone flanking the
124 supracrustal stack. Their model is supported by Re-Os age data from arsenopyrite, which yielded
125 different ages for the two zones, suggesting that Au was introduced into the BD zone at c. 2710 Ma
126 and partly remobilised into the 'Main zone' at c. 2630 Ma (Scherstén et al., 2012). Nutman et al.
127 (2007) also obtained the latter age from U-Pb ion probe geochronology of zircon associated with
128 sulphides in the 'Main zone' and argued that this represents the main Au mineralisation event.

129 The new Re-Os age data overrule a previously proposed age of ~2863 Ma for the Au
130 mineralisation (Juul-Pedersen et al. 2007), which was based on Pb-Pb isotope data. The latter age
131 likely represents the deposition of the younger volcanic-sedimentary supracrustal sequence on
132 Storø, in agreement with the maximum deposition age of c. 2840 Ma for the metasedimentary rocks
133 reported by van Gool et al. (2007).

134 Garnet-biotite thermobarometry of the supracrustal rocks on Storø has yielded (minimum) peak
135 metamorphic temperatures of 521-624°C and pressures of 4.5–6.1 kbar (Hollis, 2005; Persson,
136 2007). This metamorphic event took place at c. 2630 Ma, based on U-Pb data from zircon rims
137 (Hollis, 2005; Nutman et al., 2007).

138

139 **3. Field relationships and petrography**

140

141 Detailed observations from a traverse across the Au-hosting 'BD zone' and its wall rocks in the
142 central part of the supracrustal stack on the Qingaaq mountain in central Storø between localities 79
143 and 83 are presented here (**Fig. 2**). These localities are located in the west-facing slope of 'Lille
144 Qingaaq' below the main mountain top, which can be traversed by foot. The geochemical
145 compositions and element variations in ten samples along the traverse are presented and discussed

146 in subsequent sections. The samples are grouped into the following units from top to bottom: (1)
147 fragmental to massive amphibolite, (2) garnet-rich gneiss, (3) biotite gneiss and (4) layered
148 amphibolite.

149 The traverse begins in fragmental to massive mafic amphibolite in the hanging wall, moves into
150 garnet-rich gneiss of the 'BD zone', crosses the biotite-rich gneiss from which we present new
151 detrital zircon U-Pb age data, and ends in layered amphibolite of variable, mafic to intermediate
152 composition. The latter rocks contain dacitic layers for which we also present new U-Pb age data.
153 The lithological units as employed here are essentially the same as those used in central Storø by
154 van Gool et al. (2007).

155 The hanging wall amphibolite (unit 1) of the traverse consists of a dark, fine- to medium grained,
156 homogeneous to locally layered, and in places fragmental rock mainly composed of hornblende and
157 plagioclase and up to 10% titanite (**Fig. 3a-b**). The metamorphic textures appear well-equilibrated.
158 We interpret these rocks mainly as mafic lava flows, although evidence for a pyroclastic origin has
159 also been documented for rocks from one locality (van Gool et al., 2007). The analysed samples
160 (477610–12) are fresh, homogeneous, fine- to medium-grained hornblende-plagioclase-titanite
161 bearing rocks. Small proportions of garnet and biotite are commonly also observed, especially near
162 the contact with unit 2. Amphibolite sample 477613 at the contact with the garnet-rich gneiss (unit
163 2) contains a few per cent almandine garnet.

164 The garnet-rich gneiss of unit 2 is 1–4 m thick and displays a characteristic, highly variable but
165 diffuse layering. Its contact with the hanging wall amphibolite appears knife-sharp in the field (**Fig.**
166 **3b**). The rock has a granular texture. Garnet is mostly evenly distributed within each layer and
167 contains inclusions of staurolite. Foliation is weak or absent, but the crude layering is folded into
168 irregular, open to tight, decimetre-scale folds.

169 The lithological layering and predominant aluminous mineralogy of unit 2 have previously led to
170 the interpretation that these rocks represented metamorphosed pelitic schists of clastic sedimentary
171 origin (Pedersen, 1996; Persson, 2007). However, these rocks do not display any distinct schistosity
172 and do not contain any anatectic granitic veins, unlike genuine metaclaystone or metagreywacke
173 where anatectic veins would be common and widespread at this metamorphic grade. The rocks of
174 unit 2 possess a non-sedimentary aluminous composition that prevents upper amphibolite facies
175 partial melting.

176 Darker parts of the garnet-rich gneiss reveal rounded to elongate, and in some places folded pods
177 and layers of mafic amphibolite about 10–20 cm thick, which contain small proportions of evenly
178 distributed garnet, and gradational boundaries into the predominant garnet-rich, amphibole-absent
179 lithologies (**Fig. 3c-d**). These observations suggest that (1) the garnet-rich gneiss has been derived
180 by chemical alteration of a mafic volcanic precursor, and (2) the alteration took place prior to
181 metamorphism and folding. It has previously been proposed, based on their geochemistry, that such
182 rocks in central Storø represent alteration products (Eilu et al., 2006; Knudsen et al., 2007; Szilas,
183 2008).

184 The biotite gneiss of unit 3 is well-foliated, compositionally layered and fine to medium grained,
185 and comprises biotite, quartz, plagioclase, sillimanite and minor garnet. The biotite gneiss (**Fig. 3e-**
186 **f**) displays gradational compositional layering with sharp internal boundaries suggestive of
187 flattened, transposed and recrystallised graded bedding. Pods and stringers of quartz are common
188 and interpreted as relict premetamorphic quartz veins. Migmatitic melt seams are abundant, as
189 would be expected during high grade metamorphism of claystone. A sedimentary origin is
190 corroborated by a detrital zircon population in sample 477609, with new age data presented below
191 (**section 5.2**). The contact to the hanging wall garnet-rich gneiss is sharp, but it is difficult to
192 determine if it is primary or tectonic. The biotite gneiss may be equivalent to the biotite (-garnet)

gneiss sample 487416 collected approximately 500 m to the north-east (still on the western slope of 'Lille Qingaaq'), which was described by van Gool et al. (2007).

Layered amphibolite (unit 4) forms the footwall sequence below the biotite gneiss (unit 3), with a possible or likely tectonic contact. This unit mainly consists of strongly deformed, grey, biotite-bearing amphibolite which occasionally displays a uniform, millimetre- to centimetre-scale layering (**Fig. 3g**). The rocks of unit 4 have a mafic to dacitic composition (van Gool et al. 2007) and are interpreted by us as deformed and metamorphosed pyroclastic deposits. A deformed, medium-grained, felsic layer (25 cm thick) within this unit was sampled for age determination (**Fig. 3h**; sample 477618), see **section 5.2**. Due to intense deformation and apparent concordance with the adjacent rocks it is difficult to determine if this sheet is a metamorphosed felsic tuff or an intrusive granitoid rock. We interpret the entire unit 4 as pyroclastic in origin, however this has no bearing on the interpretation of the age data from the felsic layer presented in section 5.2.

Granitic sheets and pegmatites related to the 2550 Ma Qôrqut Granite Complex form an extensive network throughout all supracrustal rocks in central Storø (Fig. 2).

4. Analytical methods

4.1. Whole rock major and trace element analysis

Major element geochemical data were acquired by X-ray fluorescence (XRF) at the Geological Survey of Denmark and Greenland (GEUS). The data are presented in **Table 1** in the online supplementary material. See Kystol and Larsen (1999) for a detailed description of the analytical procedures.

Trace element data were obtained by inductively coupled plasma mass-spectrometry (ICP-MS) methods likewise at GEUS, using the fused borate discs from the XRF analysis. The use of the borate disc for trace element analysis ensures complete dissolution of all mineral phases but at the expense of analytical precision. However, the verification (supplementary data, Table 1, including 32 runs of the Disko-1 standard) shows that the Storø data set is sufficiently accurate and precise for their present purpose.

The trace element procedure is as follows: 0.5–1.0 g of the borate disc used for XRF-analysis is dissolved by nitric and hydrofluoric acid. The open Savillex vessel is placed on a hotplate at 180° C until the acids have evaporated. Nitric acid is then added and evaporated to dryness in two cycles. Nitric acid, internal standard solution (Ge, Rh, Re) and water are added. The vessel is closed and placed on a hotplate at 130° C for at least 12 hours. The sample is then diluted to 50 ml and just before measurement the sample is diluted further 11 times. For calibration of the instrument, certified solutions containing REE's and some additional elements are used. For verification an in-house standard (Disko-1), three international reference samples (BE-N, GA and GH), two blanks and one borate blank are prepared at the same time as the samples. The trace elements are measured using a PerkinElmer Elan 6100DRC ICP-MS instrument. The set-up, data acquisition, calibration and calculations are done using the Elan software version 2.3.2.

4.2. Zircon U-Pb isotope analysis

Separated zircons were hand-picked, mounted in transparent epoxy together with a reference zircon, and polished sufficiently to reveal cores. Scanning electron microscope (SEM) backscatter and cathode luminescence images of the internal structure of the grains were made using a Philips instrument at GEUS. The U-Th-Pb analyses were performed using a single-collector, magnetic

sectorfield LA-ICP-MS instrument at GEUS (sample 477609) and with the Cameca ims1270 instrument at the Nordsim laboratory, Swedish Museum of Natural History (sample 477618). The analytical procedures for the LA-ICP-MS and ion probe work closely followed those described by Frei and Gerdes (2009) and Whitehouse et al. (1999) and Whitehouse and Kamber (2005), respectively, with the 1065 Ma Geostandards zircon 91500 used as a reference for calculating U/Pb ratios. Where the amount of ^{204}Pb indicates the likely presence of common Pb, this was corrected using the present day terrestrial Pb composition of Stacey and Kramers (1975). Age regression was done using Isoplot of Ludwig (2003), with results presented at the 95% confidence level.

The U-Pb geochronological data are shown in **Fig. 7b-7c** and listed in **Table 2** of the supplementary data.

250

251 **5. Analytical results**

252

253 **5.1. Whole-rock geochemical data**

254

The same lithological groups as outlined in section 3, based on the field appearance and mineralogy, were employed in this section. The MFW weathering index of Ohta and Arai (2007) was used to identify which of the samples show signs of weathering and/or alteration relative to the igneous evolution trend. This is shown as the magnitude of the W index in **Figure 4**). The whole-rock geochemical composition is recalculated on a volatile-free basis and we use recalculated compositions throughout this paper. All diagrams are plotted using the software GCDkit of Janoušek et al. (2006).

The amphibolite samples (n=4) are basaltic with 48.6–49.8 wt.% SiO_2 , 5.8–6.8 wt.% MgO and 1.7–1.9 wt.% TiO_2 , and plot as Fe-tholeiites in various discrimination diagrams (e.g. Irvine and

264 Baragar, 1971; Jensen, 1976; Miyashiro, 1974). As seen in **Fig. 5**, their trace element patterns are
265 relatively flat with no distinct anomalies in the elements that are generally fluid-immobile, except
266 for a slight enrichment in Th and a small positive Zr anomaly relative to primitive mantle. Their
267 chondrite-normalised (Boynton, 1984) $\text{La}_{\text{CN}}/\text{Sm}_{\text{CN}}$ ratio is 0.78-0.89. The amphibolites have a W-
268 index of 4.2-4.5 with one outlier (477613) at a W index of 9.1 and a Zr/TiO_2 ratio varying between
269 63 and 74.

270 The garnet-rich gneisses (n=4) contain 49.6–53.2 wt.% SiO_2 , 2.3–3.0 wt.% MgO and 1.80–2.75
271 wt.% TiO_2 , and have fractionated REE with $\text{La}_{\text{CN}}/\text{Sm}_{\text{CN}}$ of 1.48–2.11. They are distinctly
272 peraluminous with 19.1–23.1 wt.% Al_2O_3 . Their W index is 19.2–27.0. They have Zr/TiO_2 ratios of
273 66–78, with one outlier at 113 (sample 477607).

274 The analysed sample of biotite gneiss contains 70.8 wt.% SiO_2 , 2.69 wt.% MgO and 0.71 wt.%
275 TiO_2 and is peraluminous with 14.2 wt.% Al_2O_3 and a W index of 36. The sample has an enriched
276 trace element pattern with $\text{La}_{\text{CN}}/\text{Sm}_{\text{CN}}$ of 3.3, a distinctly negative Nb anomaly, a small negative Ti
277 anomaly (**Fig. 5**) and a Zr/TiO_2 ratio of 154.

278 The analysed sample of layered amphibolite has 68.1 wt.% SiO_2 , 1.05 wt.% MgO and 0.35 wt.%
279 TiO_2 and is thus of dacitic composition, whereas the overall unit appears to be andesitic due to
280 predominance of more mafic layers (**section 3**). Normalised to primitive mantle it has a distinctly
281 enriched trace element pattern with a negative Nb anomaly, a large positive Zr anomaly (**Fig. 5**) and
282 Zr/TiO_2 of 741. It has a $\text{La}_{\text{CN}}/\text{Sm}_{\text{CN}}$ ratio of 6.1, which is distinctly different from the mafic
283 amphibolites. Concentrations of compatible elements are low, with 6.9 ppm Ni and of 3.6 ppm Cr.

284 The geochemical profile from massive amphibolite through garnet-rich gneiss and into biotite
285 gneiss shows that although the garnet-rich gneiss is distinctly peraluminous, its immobile element
286 ratios are comparable to those of the amphibolite, as seen on plots of TiO_2 , Nb, Lu and Hf versus Zr

(**Fig. 8**). However, one sample of garnet-rich gneiss (477607) has immobile element ratios that are intermediate between biotite gneiss and amphibolite.

5.2. U-Pb zircon geochronology

Sample 477618 from the layered amphibolite in the tectono-stratigraphically lowest part of the traverse contains stubby prismatic zircon grains with generally well-preserved oscillatory and sector zonation (**Fig. 7a**). Metamorphic overgrowths occur but are only a few microns thick, too narrow to be analysed. Eighteen out of 28 analysed grains yield a mean $^{207}\text{Pb}/^{206}\text{Pb}$ age of 3051.3 ± 2.6 Ma (2σ), MSWD = 3.7 (**Fig. 7b**; **Table 2** of electronic appendix), which is interpreted as a magmatic age. The dated population also comprises a couple of older (inherited?) grains, as well as a tail of younger grains yielding dates down to c. 2800 Ma (**Fig. 7b**). We interpret these as due to partial resetting during subsequent metamorphism (see discussion). The new age of 3051.3 ± 2.6 Ma demonstrates that the tectono-stratigraphically lowest part of the supracrustal rocks on Qingaaq (and presumably including the adjacent metagabbro and anorthosite) is contiguous with the lowest part of the mafic sequence on Aappalaartoq (**Fig. 2**; Hollis et al. 2005).

The detrital $^{207}\text{Pb}/^{206}\text{Pb}$ zircon age data from biotite gneiss sample 477609 are shown in **Fig. 7c** and **Table 2** of the online supplementary material. The age distribution is comparable to a previous data set from another metasedimentary rock that probably represents a tectono-stratigraphic equivalent (sample 484601; Scherstén et al., 2012) and corroborates that the central and possibly also upper part of the supracrustal sequence was deposited at or after 2840 Ma. Assuming that the biotite gneiss is *in situ* with respect to the chemically altered garnet-rich gneiss, this age is also the maximum age of the alteration and initial Au mineralisation.

311 6. Discussion

312

313 6.1. Whole-rock geochemical signatures

314

315 Three of the four amphibolite samples appear to represent isochemically metamorphosed basaltic
316 rocks. Ordóñez-Calderón et al. (2011) showed that the amphibolites on Storø are generally tholeiitic
317 basalts with island arc chemical affinity. Our amphibolite samples are similar to the ‘upper
318 amphibolites’ presented in their study and confirm the island arc tholeiitic signature of these
319 metavolcanic rocks. Sample 467613 has a W index of 9.1, which is slightly elevated relative to
320 values of 4.2–4.5 for the essentially unaltered amphibolites. This sample also has slight enrichment
321 in light rare earth elements (LREE) and K₂O. This is consistent with the observation that this
322 particular sample is located at the contact to the garnet-rich gneiss unit and contains some garnet
323 and biotite (**section 3**). Thus, the chemical boundary between the two units is not as sharp as it
324 might appear in outcrop (Fig. 3b). This sample has likely been affected by element mobility during
325 the alteration that affected the garnet-rich gneiss precursor (**section 6.2**).

326 The dacitic component of the layered amphibolite at locality 83 at the base of the traverse has a
327 trace element pattern that is distinctly different from the mafic amphibolites of unit 1. The observed
328 REE fractionation with a La_{CN}/Sm_{CN} ratio of 6.1 is too great to have been derived from melts
329 represented by amphibolite unit 1 by fractional crystallisation. This is consistent with the older U-
330 Pb zircon age of ~3050 Ma obtained from the layered amphibolite unit (**section 5.2**). The two
331 amphibolite units therefore belong to juxtaposed igneous mafic and volcanosedimentary sequences
332 of different origin.

333 The garnet-rich gneisses plot with a pelitic composition in sedimentary discrimination diagrams
334 of Herron (1988; not shown), which is based on their major element compositions. Although the

335 garnet-rich gneiss and biotite gneiss units resemble each other mineralogically, they possess
336 significant geochemical differences. In particular, the garnet-rich gneisses are enriched in high field
337 strength elements (HFSE) and heavy rare earth elements (HREE). The fact that the HFSE and
338 HREE ratios of the garnet-rich gneisses overlap with those of the tholeiitic amphibolites (**Fig. 8**)
339 strongly supports the field evidence that the garnet-rich gneiss stems from a basaltic precursor
340 (**section 3**). This is also seen from the total REE patterns, where the HREE are elevated in the
341 residual alteration product relative to the basaltic precursor, whereas the LREE were apparently
342 mobilised and become slightly fractionated and enriched. We also note that the biotite gneisses have
343 similar Zr/Hf ratios as the amphibolites, which suggest a significant contribution from a mafic
344 protolith to these metasedimentary rocks.

345 The new observations and geochemical data presented here corroborate previous suggestions that
346 the garnet-rich gneiss represents an alteration product of a mafic volcanic precursor (Eilu et al.,
347 2006; Knudsen et al., 2007; Szilas, 2008), although the specific type of alteration was not defined in
348 these earlier studies.

349 It can be ruled out that the chemical alteration represents palaeosol formation on basalt, because
350 of the remnants of unaltered amphibolite within the garnet-rich gneiss (**Fig. 3c-d**). In addition, the
351 adjacent biotite gneiss is interpreted as sedimentary and likely to represent a subaqueous deposit.
352 Additionally, the presence of amphibolite remnants also rule out the possibility that the garnet-rich
353 gneiss represents erosion products derived from a mafic source. On the other hand, the garnet-rich
354 gneiss could conceivably have been derived by subaqueous alteration of a basaltic precursor, either
355 by low-temperature seafloor weathering prior to or during deposition of the metasedimentary biotite
356 gneiss unit, or by subsequent infiltration of hydrothermal fluids along this lithological contact.

357 In the following section we focus on this alteration from amphibolite to garnet-rich gneiss and
358 provide mass balance calculations for this process.

359

360 6.2. Isocon mass balance calculations

361

362 The hypothesis that the garnet-rich gneiss is an alteration product from the massive
363 amphibolite precursor can be tested with mass balance calculations, considering that the field
364 relationships indicate that the alteration occurred prior to metamorphism and assuming that only
365 minor and insignificant element changes occurred during the metamorphism. For this test we use
366 the isocon method of Grant (1986, 2005). This method relies on the fact that certain elements
367 remain immobile during alteration and define a so-called isocon line, which has a slope that is
368 proportional to the net mass change. The change can be quantified by comparing an altered sample
369 with its precursor (Gresens, 1967). The average composition of the three least altered amphibolites
370 (477610, 477611 and 477612) was chosen as a hypothetical precursor in order to minimise the
371 effects of possible crystal fractionation. It was assumed that TiO₂, Zr, Hf, Nb and Lu remained
372 essentially immobile during the alteration, which is supported by a high regression coefficient of the
373 isocon lines. The results of the isocon calculations are shown in **Fig. 9** and **Table 3** in the online
374 supplementary material. Even small discrepancies between the true precursor and the chosen
375 median value might affect the mass calculation. Such discrepancies could arise from fractional
376 crystallisation through the mafic sequence or addition of a possible detrital component, given the
377 presence of genuine sedimentary rocks within the supracrustal pile. Therefore the actual numerical
378 results are considered less significant than the qualitative information describing which elements
379 were mobilised on a broader scale.

380 As already noted, the amphibolite located immediately next to the alteration zone (sample
381 477613) contains some garnet and biotite. This sample has slightly elevated LREE and K₂O and
382 could thus represent a transitional stage to an alteration product such as the garnet-rich gneiss, in

383 agreement with the calculated mass changes. There is a systematic progression in the intensity of
384 alteration from the slightly altered amphibolite (477613), which has a net mass loss of -18 %, to the
385 garnet-rich gneisses (477614, 477608 and 477615) with net mass losses of -25%, -27% and -45 %,
386 respectively.

387 As mentioned above one garnet-rich gneiss (477607) has immobile element ratios that are
388 intermediate between the amphibolites and the biotite gneiss. This is interpreted as reflecting a
389 sedimentary component in this particular sample. The composition of sample 477607 can be
390 modelled by mixing a basaltic protolith with a sedimentary source, using relative proportions of
391 about 35/65 as estimated by the Zr/TiO₂ ratios (**Fig. 8**). Although it is theoretically possible to use
392 the isocon method to quantify mass changes in such binary rock mixtures, this has not been
393 attempted due to the uncertainty about the actual precursor of sample 477607.

394 The modelled alteration process results in broadly consistent mass changes for all of the relevant
395 samples, which are intensified as the alteration progresses (**Fig. 9**). There is a general depletion of
396 most major oxides, including SiO₂, Fe₂O₃^T, MgO, CaO, Na₂O and P₂O₅. This is compatible with the
397 breakdown of olivine, pyroxenes, plagioclase and apatite in a basaltic precursor. Sr and Ni, which
398 are compatible in plagioclase and olivine, respectively, are also lost. There appears to be a
399 consistent enrichment in K₂O, Cs, Rb, Ba, Pb, Zn, LREE and Th.

400

401 6.3. Geological setting and fluid conditions during alteration

402

403 A loss of most major elements would be expected if a corrosive fluid reacted with the basaltic
404 precursor rock, and this is also seen in the isocon modelling (**section 6.2**). The observed addition of
405 large ion lithophile elements (LILE) and LREE could represent a second stage of fluid activity,
406 because these elements can be dissolved and mobilised in some aqueous fluids, regardless of the

407 concentration in the rock with which they are in equilibrium (Mottl and Holland, 1978; Ludden and
408 Thompson, 1979; Staudigel and Hart, 1983; Seyfried et al., 1998). Although, it is not possible to
409 resolve multiple events of alteration with the isocon method, because only the resulting changes can
410 be calculated between two samples, the effects of metamorphism are unlikely to affect the results,
411 because both precursor and altered rocks likely experienced the same metamorphic fluxes.

412 The mass change calculations suggests that Th was mobile. In this context it should be noted that
413 correct analysis of Th can be difficult, but the fused glass disc method of trace element analysis
414 employed in this study ensures full dissolution of all minerals. Besides, the measured concentrations
415 are well above the detection limit and the Th enrichment is consistently high for all garnet-rich
416 gneiss samples. It is therefore assumed that the calculated Th enrichment is a genuine feature of the
417 alteration process and that it can be used to constrain the conditions of the fluid-rock reaction.
418 Mobility of Th is generally assumed to require the presence of high-temperature supercritical fluids
419 (Kessel et al., 2005). Interestingly, Alt and Teagle (2003) reported addition of Th along with LILE
420 enrichment in the uppermost alteration zone of oceanic seafloor at a fast-spreading ridge in the
421 western Pacific and estimated a temperature below 100°C for the reaction. Below this zone the
422 oceanic crust was also characterised by significant loss of most major elements and the presence of
423 silica-iron deposits. Metasedimentary biotite gneisses on Storø adjacent to the garnet-rich gneiss
424 alteration zone at Storø have Th contents between 4–7.8 ppm (van Gool et al., 2007). These rocks
425 could therefore represent the source of the elevated Th in the garnet-rich gneisses and could also
426 have provided the LILE and LREE enrichment.

427 The solubility of silica is generally proportional to the temperature of the fluid system (von
428 Damm et al., 1991; Rimstidt, 1997). It is therefore likely that the addition of LILE and LREE took
429 place at low-temperature conditions, since SiO₂ was not added together with these components.

430 The loss of Fe suggests that the fluid conditions may have been reducing during the alteration,
431 enabling transport of iron as Fe^{2+} . This is consistent with the widely accepted assumption that the
432 Archaean atmosphere and oceans were reduced (Holland, 1984). Furthermore, the presence of an
433 anastomosing network of Fe-rich garnetite within the metasedimentary sequence adjacent to the
434 garnet-rich gneiss (see 'quartz-garnet-magnetite unit', van Gool et al., 2007), could possibly
435 represent redox-induced re-precipitation of Fe that was mobilised during the alteration of the
436 basaltic precursor. Szilas (2008) showed from mass balance considerations that the amount of Fe
437 that was lost in the garnet-rich gneiss alteration zone could indeed account for the observed volume
438 of magnetite precipitated within the metasedimentary sequence as a garnet-magnetite rock. The
439 above mentioned garnet-magnetite rocks possess a seawater-like REE pattern (Szilas, 2008),
440 compatible with the idea that LREE, LILE and Th were added during interaction between seawater
441 and the protolith for the garnet-rich gneiss, after the basaltic precursor had been leached of its major
442 elements during an earlier stage of alteration.

443 Hydrothermal seafloor alteration (spilitisation) of Archaean basalts has been described by
444 Kitajima et al. (2001) and by Polat et al., (2003, 2007). Generally, the upper alteration zones are
445 strongly carbonated in this type of seafloor alteration, whereas the lower zone is characterised by
446 epidotisation. This bears some resemblance to recent seafloor hydrothermal systems, which are
447 commonly characterised by variable enrichment and depletion of e.g. MgO (chlorite) or Na_2O
448 (albite), depending on what part of the fluid plumbing system is sampled, and on the age of the
449 oceanic crust (Hart, 1970; Hart et al., 1974; Humphris and Thompson, 1978; Seyfried and Bischoff,
450 1979; Harper, 1999; Paul et al., 2006). This also resembles observations from volcanogenic massive
451 sulfide (VMS) systems, where fluid-rock reaction is controlled by faults and circulation of heated
452 seawater (Franklyn et al., 1981; Lydon, 1984). However, the isocon modelling of the rocks from

453 Storø reported here does not show evidence of either of these two types of alteration, but rather
454 dissolution of the major rock forming minerals.

455 Hofmann (2011) summarised evidence for low-temperature (<150°C) seafloor alteration in the
456 Archaean Barberton greenstone belt in the Kaapvaal craton and demonstrated loss of most major
457 elements and enrichment of LILE and La. However, in this case the altered rocks were also affected
458 by significant silicification. However, no significant addition of SiO₂ is observed for the garnet-rich
459 gneisses at Storø, except in the form of local quartz veins, which may be contemporary with
460 subsequent deformation and metamorphism. A possible explanation of the lack of SiO₂ enrichment
461 is that any dissolved SiO₂ was re-precipitated in the metasedimentary rocks immediately above the
462 alteration zone, whereas Th and LILE were mobilised from the latter rocks to the leached basalts.
463 Alternatively, as discussed above, the fluid temperature might have been low at Storø, SiO₂ being
464 mostly immobile in such conditions.

465 The main remaining question is whether the very significant early leaching of the basaltic
466 precursor (with up to 45% net mass loss) was caused by a low- or high-temperature fluid. Given the
467 occurrence of Au in these supracrustal rocks, it could be suggested that hot epithermal fluids in
468 volcanic systems might have produced the observed net mass changes by advanced argillic alteration
469 (Hedenquist and Lowenstein, 1994). However, such alteration is commonly also associated with
470 silicification in a vuggy silica cap, which is not observed. Epithermal leaching would also likely
471 have been discordant and would have affected the sedimentary rocks as well. More importantly, the
472 exact age of the volcanic rocks at Storø (bracketed between 2840-2710 Ma) in relation to the
473 earliest Re-Os arsenopyrite age of 2710 Ma (Scherstén et al. 2012) remains unknown, and it is
474 likewise unknown if the initial Au mineralisation might have been epithermal. Primary, discordant
475 hydrothermal plumbing systems might be expected to become transposed into parallelism with the
476 host rocks during ductile deformation and metamorphism, but such systems have nevertheless been

477 recognised in rocks affected by regional deformation and high-grade metamorphism just as intense
478 as in central Storø. A reworked epithermal, Au mineralised alteration system of this type has
479 recently been described from the Qussuk area in northern Godthåbsfjord, only 25–50 km from
480 central Storø (Garde et al., 2012).

481 Deep sea drilling of oceanic crust has documented low-temperature hydrous alteration of oceanic
482 basalts in a process termed seafloor weathering (Robinson et al., 1977; Humphris et al., 1980; Alt,
483 2003). A stepwise increase in the intensity of alteration is observed, with secondary, low-
484 temperature phyllosilicates in the upper crust and hydrous greenschist facies minerals deeper down
485 in the volcanic pile. The alteration process presumably takes place along a chemical gradient that
486 develops during reaction with seawater over millions of years (Wilson et al., 2006). The actual
487 seafloor weathering at any given time is most intense near the free contact to the water column. This
488 is in strong contrast to the fracture- and fluid-flow-controlled types of alteration described above,
489 such as epithermal alteration or spilitisation, which require a convection cell to develop. The
490 qualitative chemical changes deducted from garnet-rich gneiss at central Storø resemble those seen
491 in modern dredged samples from the ocean floor, which consistently show enrichment in LILE and
492 Th, but variable changes in LREE (e.g. Alt and Teagle, 2003). In particular, mass balance
493 calculations of dredged, low-temperature seafloor weathered basalt described by Bienvenu et al.
494 (1990) show a net mass loss of -36% and removal of most major elements in combination with
495 enrichments in LILE and Th (**Fig. 10**).

496 There is also a striking resemblance between the mass changes calculated for the Storø alteration
497 zone (**section 6.2**) and alteration in both recent ocean floor and Permian garnet-chloritoid rocks in
498 Taiwan, which were likewise interpreted to be metamorphosed, seafloor weathered basalts (Yui et
499 al., 1994). **Fig. 10** shows the remarkable similarities between the isocon mass balances for these
500 two alteration zones and those calculated for Storø (**Fig. 9**). The Permian example displays a net

501 mass loss of -38%, which is in the same range as that calculated for the garnet-rich gneisses on
502 Storø. In both systems most elements were lost, and the enrichments consist of LILE and Th,
503 whereas LREE were either lost or enriched. These complex features may serve to evaluate the
504 specific type of alteration that affected the garnet-rich gneisses at Storø.

505 We would like to emphasize that the isocon method that we have employed to study these altered
506 rocks can only be applied to the net mass changes when comparing two specific samples. Although
507 it is a possibility that the main mass loss was associated with an earlier stage than the addition of
508 alkali elements, this addition of alkalis is also observed in the two analogue studies to which we
509 compare our data. It therefore seems likely that this type of alteration represents a single commonly
510 occurring process. We simply note that it remains a possibility that the resulting alteration was
511 represented by two events, because we cannot distinguish between the two situations with the
512 applied method. However, we would argue that the entire alteration was caused during low-
513 temperature seawater-rock reaction and we do not invoke a hot hydrothermal system at any stage of
514 the alteration of the mafic rocks to produce these highly aluminous garnet-rich gneisses.

515 We propose that the main primary alteration product on the island of Storø was montmorillonite,
516 as suggested from the alteration vectors seen in **Fig. 11 and 12**. Montmorillonite is also the
517 predominant alteration mineral in recently drilled samples of modern oceanic crust (e.g. Talbi and
518 Honnorez, 2003; Banerjee et al., 2004). The stability of sericite could account for the enrichment in
519 K₂O, Cs, Rb and Ba, whereas early allanite is a likely accessory phase that could explain the
520 stability of Th and LREE in the altered assemblage. These minerals are typical alteration products
521 and their former presence compatible with the mass changes observed by us from the isocon
522 modelling.

523 Based on the above characteristics for the alteration system on Storø, we suggest that long-term
524 (several million years) interaction between cold seawater and Meso- to Neoarchaeon oceanic crust

525 of island arc affinity resulted in the formation of a clay-rich alteration assemblage during seafloor
526 weathering of the type discussed above. Clastic sediment may have been deposited during the
527 alteration, as indicated by the suggested mixing of altered basalt with sediment displaying distinctly
528 different immobile element ratios (sample 477607). Given the abundance of quartzite in the
529 supracrustal sequence proximity to a continental source seems likely.

530 After accretion of this lithological sequence and adjacent island arc complexes, these rocks
531 were brought to amphibolite facies metamorphic conditions, where the current biotite-garnet-
532 sillimanite assemblage formed. Although such rocks are commonly interpreted in the field as
533 metasedimentary, the present study shows that this may not always be the case. Careful field and
534 geochemical studies may reveal similar seafloor weathered rocks and aluminous rocks affected by
535 other types of premetamorphic alteration in Achaean supracrustal belts world-wide.

536

537 6.4. Implications of the U-Pb zircon geochronology

538

539 The new zircon age data reported here provide evidence that the mafic rocks at central Storø are
540 in fact part of (at least) two distinct sequences. The younger sequence is 2840–2710 Ma in age, as
541 indicated by the youngest detrital U-Pb zircon ages in the metasedimentary biotite gneiss () and the
542 Re-Os age of highly radiogenic arsenopyrite associated with the Au occurrence (van Gool et al.,
543 2007; Scherstén et al., 2012). We propose the formal name the ‘Storø Supracrustal Belt’ for this
544 younger sequence (<2840 Ma). The older sequence is c. 3050 Ma in age and thus likely genetically
545 related to anorthosite and layered gabbros at Aappalaartoq, from which a similar age is inferred,
546 based on an intrusive tonalitic rock yielding an age of 3053 ± 3.4 Ma (sample 481271, see Hollis et
547 al. 2005). We propose that this older sequence should be termed the ‘Storø Anorthosite Complex’,

548 so that the allochthonous nature and the different age groups of the mafic and sedimentary rocks on
549 Storø are distinguished in future studies.

550 Together with structural observations reported by van Gool et al. (2007) there is therefore evidence
551 for tectonic juxtaposition of at least two mafic sequences with different ages and tectono-
552 metamorphic histories, as also suggested by Scherstén et al. (2012). A similar tectonic accretion of
553 arc-related supracrustal sequences has also been proposed for the Eoarchaeon Isua supracrustal belt
554 north of Godthåbsfjord, supporting an uniformitarian model of continental crust formation for the
555 Godthåbsfjord region (e.g. Nutman and Friend, 2009).

556

557 **7. Summary of key observations and conclusions**

558

- 559 • Field observations show that garnet-rich gneisses on central Storø contain relict pods and
560 elongate lenses of mafic amphibolite similar to that found in the immediate hanging wall.
- 561 • Immobile elements (TiO_2 , Zr, Hf, Nb, Lu) have similar ratios in the amphibolites and the
562 garnet-rich gneisses, but were residually enriched in the latter rocks due to a net mass loss
563 during alteration.
- 564 • Isocon mass balance calculations between mafic amphibolite and garnet-rich gneisses
565 suggest that SiO_2 , $\text{Fe}_2\text{O}_3^{\text{T}}$, MgO, CaO, Na_2O , P_2O_5 , Sr and Ni were lost during the alteration,
566 consistent with breakdown of rock-forming olivine, pyroxenes, plagioclase and apatite in a
567 basaltic precursor. There appears to be a rather uniform enrichment in K_2O , Cs, Rb, Ba, Pb,
568 Zn, LREE and Th. The net mass loss ranges from -18% to -45%.
- 569 • Similarities between the calculated mass changes for modern seafloor weathered oceanic
570 basalts and the Archaean rocks studied in this profile at Storø suggest that they represent a

section of fossil ocean floor that was subjected to weathering by long term reaction with cold seawater.

- Seafloor weathering is most intense near the contact with the water column and gradually diminished in intensity with depth. This suggests that the hanging wall amphibolite (unit 1) at central Storø is located stratigraphically below the metasedimentary rocks (unit 3), implying that the sequence might be inverted.
- New U-Pb zircon geochronology confirms that the mafic rocks at Storø comprise at least two rock sequences with distinctly different ages: 1) 'The Storø Supracrustal Belt' (<2840 Ma) and 2) 'The Storø Anorthosite Complex' (c. 3050 Ma).

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816

817 **Figure text**

818 Fig. 1. Geological map of the Godthåbsfjord region in southern West Greenland with the location of
 819 the study area on the island of Storø. Based in part on Garde (1987, 1989).

820

821 Fig. 2. Geological map of central Storø, modified from Fig. 1 in van Gool et al. (2007). The
822 positions of localities 79, 80 and 83 mark the geographical top, middle and bottom of the profile
823 from which field observations and geochemical data are presented.

824

825 Fig. 3. Field photographs of metasedimentary, metavolcanic and garnet-rich, chemically altered
826 aluminous rock units at 'Lille Qingaaq' (lower peak of Qingaaq mountain), central Storø. **a.**
827 Fragmental amphibolite of likely volcanoclastic origin. Loc. 79. **b.** Contact between mafic
828 metavolcanic amphibolite and altered, garnet-rich aluminous rock. Loc. 79. The garnet-rich gneiss
829 visible in the right part of Fig. 3b contains up to 40% almandine garnet, 20–30% plagioclase, biotite
830 and commonly sillimanite, and is generally quartz-poor. **c-d.** Pods of relict amphibolite in altered,
831 garnet-rich rocks. Note gradational boundaries, variable garnet content in relict pods, absence of
832 partial melting, and fold in Fig. 3d. Loc. 80. **e-f.** Metagreywacke with quartz veins and partial melt
833 veins. Loc. 80. **g.** Finely layered amphibolite of andesitic composition, interpreted as having a
834 pyroclastic protolith. Loc. 83. **h.** Felsic layer in finely layered amphibolite. Loc. 83. The size of the
835 coin is 24.5 mm in diameter.

836

837 Fig. 4. MFW-weathering index of Ohta and Arai (2007), showing that the amphibolites and the
838 layered amphibolite plot along the igneous trend, whereas the biotite gneiss and the garnet-rich
839 gneisses plot in the field of weathered and/or altered rocks.

840

841 Fig. 5. Primitive mantle-normalised (Sun and McDonough, 1989) trace element diagrams for the
842 various lithological units presented in this study. a) Amphibolite, b) garnet-rich gneiss, c) biotite

gneiss and d) dacitic component within layered amphibolite. The shaded area is the total range of all samples.

Fig. 6. Profile from the least altered amphibolite, across the garnet-rich gneiss and into the biotite gneiss, showing variations in Zr/TiO_2 and A/CNK ($Al/(Ca+Na+K)$).

Fig. 7. **a.** CL (cathodoluminescence) and BSE (backscattered electron) images showing typical morphology of igneous zircon in sample 477618 (felsic layer in layered amphibolite) and detrital and metamorphic zircon in sample 477609 (metasedimentary biotite gneiss). Note igneous-type oscillatory zoning and sector zoning in 477618, and zoned crystal fragments with homogeneous metamorphic overgrowths as well as entire homogeneous grains in 477609. **b-c.** U-Pb concordia diagrams for samples 477618 and 477609 showing age distributions. Scale bar is 100 microns in all images.

Fig. 8. Plots of various high field strength elements showing the similar trend for the amphibolites and the garnet-rich gneisses. Biotite gneiss shown for comparison, shows a distinctly different trend. TiO_2 is in wt.% and Zr, Hf, Nb and Lu are in ppm.

Fig. 9. Isocon mass balance calculations for the alteration of the garnet-rich gneisses from a median basaltic precursor. It is assumed that TiO_2 , Zr, Hf, Nb and Lu remained immobile during the alteration. The calculations were performed in the EASYGRESGRANT Microsoft Excel spread sheet program of López-Moro (2012). The results are plotted here as loss/gain relative to the precursor ($\Delta C_i/C_i^0$). The absolute loss/gain (ΔC_i) in wt.% or ppm are reported in the supplementary Table 3.

867
868 Fig. 10. Isocon mass balance calculations for a sample pair from Yui et al. (1994) and Bienvenu et
869 al. (1990). The results are plotted here as loss/gain relative to the precursor ($\Delta C_i/C_i^0$).

870
871 Fig. 11. Amphibolites (squares), Garnet-rich gneisses (triangles) and montmorillonite and sericite
872 end-members (crosses) showing possible trends from of the garnet-rich gneisses from the
873 amphibolites towards these common alteration products. End-members are taken from Herrmann
874 and Berry (2002) and normalised to 100% on a volatile-free basis.

875
876 Fig. 12. Ternary diagram of the amphibolite, garnet-rich gneisses and possible alteration end-
877 members. The distinct trends towards the clay end-members suggests equilibrium change towards
878 these components during the alteration process.

879
880 **Supplementary Data Tables**

881 Table 1. Major and trace element geochemical data for the samples.

882
883 Table 2. U-Pb isotope data for zircon.

884
885 Table 3. Isocon mass balance calculations.

Figure 1

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Table 2 Background dataset for online publication only

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