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Benjamin Wasilewski, Jonathan O'Neil, Hanika Rizo, Jean-Louis Paquette, Abdelmouhcine Gannoun. Over one billion years of Archean crust evolution revealed by zircon U-Pb and Hf isotopes from the Saglek-Hebron complex. Precambrian Research, 2021, 359, pp.106092. 10.1016/j.precamres.2021.106092 hal-03211908

# HAL Id: hal-03211908 https://uca.hal.science/hal-03211908

Submitted on 4 May 2021

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# Over one billion years of Archean crust evolution revealed by zircon U-Pb and Hf isotopes from the Saglek-Hebron Complex

# 4 **1. Introduction**

5 Although the extent of the early felsic crust is debated, Archean cratons are mostly composed of silica-6 rich rocks from the tonalite-trondhjemite-granodiorite (TTG) suites, which abundance appears to 7 significantly decrease after 2500 Ma (Laurent et al., 2014; Moyen and Laurent, 2018; Moyen and 8 Martin, 2012). The geochemical composition of TTG differs from that of the modern continental crust, 9 suggesting they may have formed through distinct processes (e.g. Moyen and Martin, 2012). The study 10 of ancient terrain containing TTG, therefore, is crucial to our understanding of how continents formed 11 and stabilized. The Saglek-Hebron Complex (SHC), located in Northern Labrador, Canada, is a 12 polymetamorphic terrain dominated by TTG as old as 3900 Ma, and encompassing more than a billion years of the geological record (e.g. Komiya et al., 2017). With such protracted geological history, the 13 SHC is one of the best candidates to decipher ancient crustal processes. Previous studies highlighted 14 the existence of multiple generations of granitoids within the SHC that exhibit a large variation of 15 16 chemical compositions and tonalite ages (Komiya et al., 2017; Krogh and Kamo, 2006; Sałacińska et al., 2018; Schiøtte et al., 1989a; Shimojo et al., 2016; Vezinet et al., 2018). Most of these studies, 17 18 however, largely focused on the oldest Paleo to Eoarchean history of the SHC. This contribution 19 investigates the evolution of the SHC over the whole Archean Eon and combines detailed whole-rock geochemistry of the wide compositional array of SHC granitoids, along with in-situ U-Pb and Lu-Hf 20 21 isotopic compositions of their zircon. The zircon U-Pb geochronology is used here to refine the complex felsic magmatic and metamorphic history of the SHC over more than one billion years, while 22

the in-situ zircon Hf isotopic compositions, combined with the whole-rock geochemical composition of the host rocks, are used to unravel the evolution of the crustal sources involved. Together, these tools contribute to better understand the timing, the formation, and the long-term evolution of the SHC and the overall Archean sialic crust.

#### 27 **2.** Geological context

28 The Saglek-Hebron Complex, located on the east coast of Northern Labrador in Canada, is dominated 29 by orthogneisses from the TTG series along with granitic rocks and includes meter to kilometer scale 30 supracrustal enclaves (Fig. 1). Supracrustal rocks mainly include mantle-derived rocks (mafic 31 metavolcanic and ultramafic rocks), and chemical or clastic metasediments (Baadsgaard et al., 1979; 32 Bridgwater et al., 1975; Komiya et al., 2015; Nutman and Collerson, 1991). The metavolcanic rocks 33 are divided into two distinct units, the Upernavik assemblage interpreted to be Mesoarchean and the Nulliak assemblage interpreted to be Eoarchean (Bridgwater and Schiøtte, 1991; Morino et al., 2018, 34 35 2017; Nutman et al., 1989; Schiøtte et al., 1992). Both units, however, are compositionally similar and 36 interpreted as a series of tholeiitic basaltic flows displaying some extent of differentiation (Wasilewski 37 et al., 2019). The SHC orthogneisses are divided into four main units including the Iqaluk grey gneiss 38 (sometimes referred to as the Nanok gneiss), the Uivak gneiss, the Lister gneiss, and late granitic 39 intrusions. The oldest TTG unit in the SHC is the Iqaluk grey gneiss dated at ~3900 Ma (Collerson, 1983a; Komiya et al., 2017; Regelous and Collerson, 1996; Shimojo et al., 2016). The Uivak gneiss 40 41 are the predominant lithology in the SHC, and were originally subdivided into two units based on their 42 age and mineralogy, with the older tonalitic Uivak I dated between 3863 Ma and 3732 Ma (Sałacińska et al., 2018; Schiøtte et al., 1989b; Vezinet et al., 2018) and the younger granodioritic Uivak II dated at 43 ~3600 Ma (Hurst et al., 1975; Komiya et al., 2017; Sałacińska et al., 2018). However, a recent study 44

argued that the Uivak gneiss rather includes five units produced throughout the Eoarchean, from 3890
to 3610 Ma, and thus form almost continuous protracted magmatism occurring over more than 250
million years (Komiya et al., 2017). The younger granitoids include Paleoarchean TTG called the
Lister gneiss consisting of granodioritic intrusions emplaced between 3240 Ma and 3350 Ma (Komiya
et al., 2017; Schiøtte et al., 1989b) and Neoarchean granitic rocks intruding the SHC with a magmatic
peak occurring at ~2766 Ma (Collerson, 1983a, 1983b; Schiøtte et al., 1989b).

51 The SHC records at least two major protracted thermal episodes leading to some extent of crustal 52 reworking around ~3620 Ma and high-grade metamorphism up to granulite facies around ~2760-2600 Ma (Bridgwater and Collerson, 1976; Hurst et al., 1975; Kusiak et al., 2018; Nutman and 53 54 Collerson, 1991; Sałacińska et al., 2018; Schiøtte et al., 1992, 1986; Van Kranendonk, 1990). Based on 55 detrital zircon, it has also been suggested that the Neoarchean thermal event could have been caused by 56 a massive collision and terrane accretion after the emplacement of the ~3300 Ma Upernavik 57 metasedimentary assemblage (Schiøtte et al., 1992). A recent geochronology investigation on 58 monazites and apatites suggests two thermal closure ages at 2500 and 2200 Ma, interpreted either as 59 cooling ages, or as two separate successive thermal events reaching, respectively, upper amphibolite 60 and greenschist facies metamorphic conditions (Kusiak et al., 2018).

#### 61 **3. Methods**

Forty-seven orthogneiss samples have been collected for this study. All samples have been analysed for whole-rock geochemistry. A subset of eighteen samples have been selected for coupled to U-Pb and Hf in-situ analysis in zircon. These samples have been selected to cover the full compositional range of SHC granitoids and to include all felsic lithologies previously described in the literature. This 66 includes type localities for the Iqaluk gneiss described by Shimojo et al. (2016) in the Kangidluarsuk 67 Inlet (St John's Harbour), the Uivak II gneiss described by Bridgwater et al. (1975) on the opposite coast of Nulliak Island and "White Point" and the Lister gneiss found on Lister Island. The outcrop 68 69 studied by Shimojo et al. (2016) on which they obtained an age of  $3920 \pm 49$  Ma and interpreted as the 70 oldest rock of the complex was resampled because of its significance and given that its exact age was questioned due the relatively large imprecision (Whitehouse et al., 2019). Specific locations 71 72 previously studied by Bridgwater et al., (1975) were also targeted given the paucity of geochronological data on the Uivak II and Lister gneiss from these localities. The detailed 73 74 geochronology on multiple generations of orthogneisses provided the framework to the Hf isotope 75 work and helped targeting the best-preserved zircon grains to analyse for Hf isotopic compositions which are used to better understand the crustal evolution of the SCH over the whole Archean Eon. 76 77 Figure 1 shows the main sample locations and GPS coordinates for all samples are given in Table 1. Selected outcrop photographs are available in the supplementary material Figure S1). The full methods 78 79 regarding whole-rock major and trace element geochemistry can be found in the supplementary 80 material (Supplementary Table S1).

81 Eighteen samples from our set of 47 granitoids were analyzed for both in-situ U-Pb geochronology and 82 Hf isotopes in zircon. Rock samples were crushed using a steel jaw crusher and disk mill and then 83 sieved to collect grain sizes between 250-106 µm. Heavy minerals, such as zircon and metallic oxides, 84 were separated using a water shaking table and methylene iodide heavy liquids. The heavy mineral 85 fractions were then passed through a Frantz magnetic separator to remove the magnetic minerals. Between 100 and 120 zircon grains were then handpicked, mounted on an epoxy resin and polished. 86 87 Cathodoluminescence (CL) images of the polished zircon grains were taken using the JEOL 6610LV 88 scanning electron microscope (SEM) at the University of Ottawa, to identify the different zircon zones

89 and guide the laser ablation work. Both U-Th-Pb and Hf isotope analyses were undertaken at the 90 Laboratoire Magmas et Volcans (LMV) (Clermont-Ferrand, France). U-Th-Pb isotope analyses were performed on a Thermo Scientific Element XR-ICP-MS, and Hf isotope analyses were conducted on a 91 92 Thermo Scientific Neptune Plus multicollector ICP-MS, both instruments were coupled with a 93 Resonetics M50E 193 nm excimer laser ablation (LA) system. The analytical method for isotope 94 dating by LA-ICP-MS described in Hurai et al.(2010) and Paquette et al.(2014) were followed and 95 more details on analytic conditions are available in the supplementary material Table S1. Primary (GJ-96 1) and secondary (91500) zircon reference materials have been measured to ensure the quality of the 97 data over the different analytic sessions and data are found in the supplementary material Table S1 and Table S2. Common Pb was not corrected owing to the large isobaric interference of <sup>204</sup>Hg. The ablated 98 99 zircon zones were carefully chosen in order to avoid any mixed zones and Hf isotopic measurements 100 were performed on the same spots than those chosen for the U-Pb analyses according to analytical 101 techniques described in Moyen et al. (2017) and Paquette et al. (2017). The full Lu-Hf isotope data are available in the supplementary material Table S3. Initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios were calculated using the 102  $\lambda^{176}$ Lu decay constant of 1.867x10<sup>-11</sup>.yr<sup>-1</sup> of Söderlund et al. (2004) and the CHUR parameters of 103  ${}^{176}\text{Hf}/{}^{177}\text{Hf} = 0.282785$  and  ${}^{176}\text{Lu}/{}^{177}\text{Hf} = 0.0336$  (Bouvier et al., 2008) were used for calculation of 104 εHf values. 105

- 106 **4 Results**
- 107

#### 4.1 Petrography & Geochemistry

Although felsic rocks from the SHC have been metamorphosed and are commonly gneisses, we use the term "granitoid" *sensu lato* in this study to refer to the felsic plutonic rocks. Similarly, the terms granite, tonalite, trondhjemite, granodiorite are use to refer to a mineralogical and geochemical

111 composition regardless of metamorphic fabrics. Based on observed mineralogy and CIPW normative 112 compositions, the SHC granitoids can be divided into four distinct groups, including trondhjemite, 113 tonalite, granodiorite (grouped as the TTG, *s.l.*) and granite (Fig. 2). From the TTG studied here, only 114 a few samples of granodiorite have been analyzed, but they are compositionally similar to other SHC 115 granodiorite previously described by Schiøtte et al. (1993) (Fig. 2). The TTG are commonly medium 116 grained and exhibit variable degrees of deformation (Fig. S1 and Fig. S2) with common evidence of 117 migmatization (Fig. S1). Pegmatitic and porphyritic textures can also occasionally be observed (Fig. 118 S1 d. e. f.). Trondhjemite samples (Fig. S1 b. g. i. and Fig S2 e. f. g. h. i.) consist of typical grey gneiss 119 composed of quartz + oligoclase + biotite + titanite  $\pm$  zircon  $\pm$  apatite. Compared to the trondhjemite, 120 rocks that exhibit tonalitic to granodioritic compositions can be described as melanocratic grey gneiss containing higher amounts ferromagnesian minerals such as clinopyroxene and biotite (Fig. S1 a. c. e. 121 122 f. j. k. and S2 k. l. m. n.). The granodiorite locally exhibits augen textures at the outcrop scale (e.g. 123 Iluilik & White Point; Fig S1.e), which is not observed in trondhjemite. The granite is composed of quartz + orthoclase + oligoclase  $\pm$  biotite  $\pm$  zircon  $\pm$  apatite  $\pm$  garnet and typically defines meter to tens 124 125 of meter scale leucocratic units (Fig. S1 d. and S2 a. b. c. d. j.). Granitic rocks commonly occur as 126 migmatite veins but also as larger plutonic bodies. Granite usually shows little fabric and minor 127 deformation compared to TTG.

The TTG display a wide range of silica content (56.6 wt. % to 76.8 wt. %SiO<sub>2</sub>), with most samples having below 72 wt. %SiO<sub>2</sub>. The tonalite exhibits the lowest SiO<sub>2</sub> contents among the TTG and display high Mg concentrations relative to the other SHC granitoids (Fig. 3a), and therefore are here referred to as Mg-rich tonalite. Except for one sample, the granite shows high and uniform silica contents ranging from 73.0 to 77.5wt. %SiO<sub>2</sub> (Fig. 3a). Both the Mg-rich tonalite and the granodiorite display relatively high FeO<sub>t</sub> and TiO<sub>2</sub> contents compared to the granite and the trondhjemite (Fig. 3b). Compared to all TTG, the granite exhibits higher concentrations in  $K_2O$  (from 3 to 6 wt.%; Fig. 3c) and Rb, and lower CaO concentrations (<2 wt. %; Fig. 3e). The tonalite and trondhjemite exhibit higher Na<sub>2</sub>O, CaO, and Al<sub>2</sub>O<sub>3</sub>but lower  $K_2O$  compared to the granite and granodiorite (Fig. 3c-d-e). The Mg-rich tonalite, however, exhibits lower A/CNK ratios relative to the trondhjemite, with the granite showing the highest A/CNK ratios (Fig. 3f).

139 All granitoids exhibit pronounced negative Nb-Ta anomalies (Fig. 4a) along with strong depletion in 140 heavy rare earth elements (HREE) compared to light rare earth elements (LREE, Fig. 4b). Mg-rich 141 tonalite and granodiorite are generally characterized by lower (La/Yb)<sub>N</sub> ratios than most trondhjemite and granite samples (Fig. 5). The trondhjemite samples exhibit higher (La/Yb)<sub>N</sub> ratios (up to 150) and 142 143 most samples display positive Eu anomalies (Fig. 4b; Fig. 5). The trondhjemite samples collected on 144 White Point, however, display higher HREE contents and thus lower (La/Yb)<sub>N</sub> ratios compared to 145 other trondhjemite samples. The trondhjemite sample SG-019 collected on Ukkalek Island exhibits 146 extremely low trace element and REE concentrations, a prominent positive Eu anomaly and mostly 147 plots outside of the compositional field comprising the other trondhjemite samples (Fig. 4). The Lister 148 gneiss (SG-265) is compositionally similar to the Mg-rich tonalite but exhibits the lowest REE 149 concentrations (Fig. 4b). Granitic samples exhibit high Rb and Pb with lower Sr concentrations, 150 relative to all other TTG. In general, granite samples are more enriched in LREE compared to the 151 trondhjemitic rocks with large variability in (La/Yb)<sub>N</sub> ratios ranging from 0-300 (Fig. 5). The granite 152 samples commonly display small positive or negative Eu anomalies while the leucogranitic samples 153 SG-007 and SG-017 exhibit very low REE concentrations with pronounced positive Eu anomalies 154  $(Eu_N/Eu^* \sim 60)$  (Fig. 4b).

155

#### **4.2U-Pb Geochronology**

Zircon grains from 18 granitoids have been analyzed by LA-ICP-MS for U-Pb geochronology. 156 157 Samples were selected to comprise representative samples of each rock types with an effort to include 158 the different generations of granitoids known in the SHC. This selection was based on whole-rock 159 geochemistry, overall quality of sample (no veins, limited weathering, or alteration) and field 160 relationship within the SHC. For each sample, a set of 100 to 120 zircon grains have been mounted for 161 analysis. Data for all individual zircon analyses are available in supplementary material Table S3 and Figure S3. Analyse s that were considered as of lesser quality (e.g. presence of <sup>204</sup>Pb) were discarded. 162 163 Therefore, some sample yielded a smaller number of analyzed zircon grains, especially those including a larger amount of metamict zircon grains (e.g. SG-017, SG-019 and SG-080). Figure 6 shows 164 165 cathodoluminescence (CL) zircon images for 6 samples (all CL images are included in Figure S3) and 166 the Concordia plots for each sample are displayed in Figure 7. Zircon grains exhibiting inconsistent texture in CL imaging with numerous inclusions and a high uranium content have been characterized 167 168 as metamict (e.g. B11, B12 from the SG-143 granite; Fig. 6-f.). Recrystallized zircon grains are mostly 169 rounded and exhibits black color in CL imaging. Recrystallized rims are commonly following the pre-170 existent zonation or irregularly crosscut the zoned core in not fully recrystallized zircon grains. They 171 often exhibit a high uranium content and a younger age than the core. Table 2 presents a summary of 172 the crystallization ages, inherited ages and metamorphic ages obtained for each sample analyzed. 173 Reported ages for each sample in Table 2 were determined from either the Concordia age, for samples with populations of clustered concordant zircon analyses, the weighted mean <sup>207</sup>Pb/<sup>206</sup>Pb age, for 174 populations of less concordant zircon analyses, or the upper intercept age, for populations with mostly 175 176 discordant zircon analyses. For samples with populations showing concordant results over a wide 177 range of ages, indicative of ancient Pb-loss, the oldest concordant zircon analyses were considered as178 the most representative of the crystallization age.

179

#### a. Trondhjemite

Four trondhjemite samples: SG-019, SG-024, SG-025, and SG-122, have been analyzed for U-Pb on
zircon.

182 Zircon grains from sample SG-019 are mostly metamict and rounded. A total of 33 analyses were performed (Fig. 7a). The main population of zircon yields a weighted mean <sup>207</sup>Pb/<sup>206</sup>Pb age of 183 184  $2732 \pm 25$  Ma (MSWD=2.2; *n* =12). These zircon grains consist of homogeneous black core to oscillatory zoned grains, which exhibit an average U concentration of ~470 ppm (All <700 ppm). 185 "Black cores" or "black grains" refer to the black CL imaging rendered in response to a relatively high 186 187 U content. A resolvable second population mostly consists of metamict zircon grains (U concentrations from 600 to 2860 ppm; average ~1600 ppm) and yields a weighted mean <sup>207</sup>Pb/<sup>206</sup>Pb age of 188 189  $2574 \pm 14$  Ma (MSWD= 0.46; n = 14). Two older zircon cores yield Concordia ages at  $3477 \pm 18$  Ma 190 and  $3553 \pm 15$  Ma. Given the large number of metamict zircon grains in this sample, and the relatively 191 small number of analyses compared to other trondhjemite samples, the crystallization age of SG-019 is 192 ambiguous. If the 2732 Ma population is taken as representing the crystallization age, the 2 193 Paleoarchean zircon grains would likely be inherited. Contrastingly, the Neoarchean ages could 194 represent reset ages or secondary recrystallization, in which case the older zircon grains could be more 195 representative of the crystallization age. Nevertheless, the geochronological data from this sample 196 must be taken with caution.

A total of 128 U-Pb analyses have been conducted on sample SG-024 (Fig. 7b). Zircon grains are dominated by two main populations that exhibit U concentrations <1000 ppm, associated with a wide</p>

199 range of Th/U ratios (0.01-2.83). A number of zircon grains from sample SG-024 are subeuhedral to 200 euhedral and exhibit well defined oscillatory zoning (e.g. zircon #A17: Fig. 6a). The few concordant 201 analyses plot between 3800 and 3870 Ma. However, most grains from this population are discordant 202 and spread along a poorly defined discordia line (Fig. 7b). The two oldest concordant cores yield a 203 Concordia age of  $3869 \pm 10$  Ma (MSWD<sub>(Concordance+Equivalence)</sub> =1.5; n = 2) considered as the 204 crystallization age for this sample. A population of Neoarchean zircon is dominated by subeuhedral to 205 rounded zircon grains that show clear sector zoning and exhibits a higher average Th/U ratio of 1.64 (e.g. zircon #A05:Fig. 6a), which altogether yield a weighted mean  ${}^{207}$ Pb/ ${}^{206}$ Pb age of 2721 ± 8 Ma 206 (MSWD= 0.89; n = 42). A few Neoarchean oscillatory zoned rims and cores with lower Th/U ratios 207 (<0.2) are also present. However, these Neoarchean zircon grains exhibit much more scattered ages 208 and yield a weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2769 ± 19 Ma (MSWD= 4; *n* =28). 209

Zircon grains from sample SG-025 are dominated by oscillatory and sector zoned textures to 210 homogeneous black in CL images. A total of 122 U-Pb analyses have been done on sample SG-025. 211 212 All analyses of oscillatory zoned zircon spread along the Concordia curve from 3838 to 3000 Ma (Fig. 213 7c). The older zircon grains from this population are characterized by higher Th/U ratios, up to 0.81, 214 relative to the average Th/U ratio of oscillatory zoned zircon of 0.16, whereas the zircon grains displaying younger measured <sup>207</sup>Pb/<sup>206</sup>Pb ages show more evidence of ancient Pb-loss. Therefore, the 215 216 crystallization age for this sample is best represented by the oldest concordant zircon grain with a 217 Concordia age of  $3838 \pm 10$  Ma. Sample SG-025 also includes a population of sector zoned and black 218 zircon with high Th/U (>0.2) and yielding Neoarchean ages. These grains cluster around a weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2747 ± 5 Ma (MSWD =0.54; *n* =91). 219

A total of 105 U-Pb analyses were conducted on sample SG-122 (Fig. 7d). Zircon grains from this trondhjemite sample are dominated by rounded grains with oscillatory zoned textures and

222 homogeneous black grains in CL images. The population of oscillatory zoned zircon scatters along a 223 discordia line with an upper intercept age at  $3752 \pm 33$  Ma (MSWD=13; n = 62). Moreover, a number 224 of concordant zircon analyses from this population spread along the Concordia curve between ~3790 225 and ~3700 Ma, suggestive of ancient Pb-loss. The oldest concordant zircon from this scattered 226 population gives a Concordia age of  $3781 \pm 12$  Ma, representing the crystallization age for this sample. 227 This euledral zircon grain shows a clear oscillatory zoning and a relatively high Th/U ratio ( $\sim 0.2$ ). A subordinate population of U-rich rims yields a younger weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2804 ± 9 Ma 228 229 (MSWD= 0.81; *n* =28).

230

## **b.** Mg-rich tonalite

Four Mg-rich tonalite samples have been analyzed for U-Pb on zircon, including sample SG-026, SG027, SG-210c and SG-265.

Sample SG-026 is dominated by oscillatory zoned to sector zoned subeuhedral zircon grains (e.g. zircon #B16 & #E12;Fig. 6b) that exhibit variable U concentrations (20 to 1000 ppm) and relatively high Th/U ratios (>0.6) compared to zircon grains from other samples. A total of 115 U-Pb analyses were performed on sample SG-026, which includes a single zircon population defining a discordia line with an upper intercept at  $3820 \pm 20$  Ma, interpreted as the crystallization age, and a lower intercept at  $2514 \pm 71$  Ma (MSWD=1.12; n = 103; Fig. 7e). Most zircon grains exhibit recrystallized black and white rims, that systematically yield younger and more discordant ages compared to the zoned cores.

A total of 138 analyses were done for the sample SG-027, which includes three main zircon populations (Fig. 7f). Zircon grains from the oldest population exhibit oscillatory zoning and igneous Th/U ratios from 0.2 to 0.5. Several zircon gains from this population are concordant, spreading on the Concordia curve between ~3600 and ~3500 Ma, which may reflect ancient Pb-loss. Therefore, the average age of the two oldest concordant zircon analyses of  $3632 \pm 9$  Ma (MSWD<sub>(C+E)</sub> = 1.3; n = 2) is taken as the crystallization age. The other zircon populations are Neoarchean, which include two groups characterized by metamict textures and black CL images, commonly occurring as rims around older cores. One Neoarchean population exhibits lower Th/U ratio and yields a weighted mean  $^{207}$ Pb/<sup>206</sup>Pb age of  $2785 \pm 7$  Ma (MSWD= 1.3: n = 46), while a secondary smaller population with higher Th/U ratios (>0.4) yields a weighted mean  $^{207}$ Pb/<sup>206</sup>Pb age of  $2595 \pm 20$  Ma (MSWD= 0.93; n = 6).

251 Sample SG-210c was collected from the same outcrop and lithology as the sample previously dated at 252  $3920 \pm 49$  Ma (Shimojo et al., 2016), the oldest reported U-Pb age in the SHC. A total of 120 U-Pb analyses were obtained from this sample. A number of zircon analyses scatter along the Concordia 253 254 curve from ~3900 to ~3500 Ma defining two populations (Fig. 7g). The oldest concordant zircon analyses display a relatively homogenous population exhibiting oscillatory zoning (e.g. zircon #A01; 255 Fig. 6c), with high Th/U ratios (> 0.3) and a Concordia age of  $3869\pm6$  Ma (MSWD<sub>(C+E)</sub> = 1.11; n = 34) 256 257 interpreted as the crystallization age. The younger zircon analyses show a fair amount of scattering and 258 a progressive decrease of Th/U ratios (to <0.1) as ages get younger. This population consists of U-rich recrystallized zircon, black on CL images (e.g. zircon #A02; Fig. 6c) and displays a weighted mean 259  $^{207}$ Pb/ $^{206}$ Pb age of 3558±11 Ma (MSWD=0.49; *n* =19). A single zircon yields a Concordia age of 260 261 2720±16 Ma (F02, black rounded grain).

A total of 86 U-Pb analyses were performed on sample SG-265 that was collected on Lister Island. All zircon grains from this sample show clear and well-defined oscillatory zoning as well as relatively high Th/U ratios, between 0.3 and 0.5. Zircon analyses from a single population define a discordia line with an upper intercept age of  $3229 \pm 8$  Ma (MSWD= 1.3; n = 70; Fig. 7h). The most concordant zircon analyses yield a Concordia age of  $3224 \pm 7$  Ma (MSWD<sub>(C+E)</sub> = 0.13; n = 13) and considered as the crystallization age.

268

#### c. Granodiorite

One sample of granodiorite SG-203 has been analyzed for U-Pb with a total of 121 in-situ analyses (Fig. 7i). The main population of zircon exhibits oscillatory zoning and high Th/U ratios ranging between 0.3 and 0.5. They define an upper intercept age of  $3330 \pm 15$  Ma (MSWD=1.2; n = 103) interpreted to be the crystallization age. Two additional minor populations of zircon with lower Th/U ratios (<0.1) yield younger weighted mean  ${}^{207}$ Pb/ ${}^{206}$ Pb ages of  $2975 \pm 17$  Ma (MSWD= 1.02; n = 9) and  $274 = 2703 \pm 30$  Ma (MSWD=0.69; n = 3).

275

## d. Granite

Nine granite samples have been analyzed for U-Pb on zircon, including sample SG-007, SG-017, SG-037, SG-080, SG-084, SG-087, SG-127, SG-143, and SG-208. All granitic samples were collected
from large granite exposures, except for leucogranite sample SG-017 collected from meter scale veins
between tonalite and ultramafic rocks.

280 A total of 98 U-Pb analyses have been conducted on sample SG-007 (Fig. 7j). Zircon grains are 281 dominantly elongated subeuhedral grains exhibiting oscillatory zoning or metamict textures. Most 282 zircon grains with oscillatory zoned cores show thin recrystallized rims. This sample displays zircon 283 grains with variable ages and multiple heterogeneous populations. Concordant zircon analyses display a wide range of ages from 3883 to 3300 Ma which correlates with a decrease of Th/U ratios (from 0.7 284 to <0.1). The three oldest concordant grains consist of oscillatory zoned low U cores with ages 285 spreading on the Concordia from  $3883 \pm 10$  Ma (Concordia age from zircon #C5;Fig. 6d) to 286  $3853 \pm 12$  Ma (Concordia age from zircon #E10-c;Fig. 6d), with a  $^{207}$ Pb/ $^{206}$ Pb weighted mean age = 287

288  $3869 \pm 26$  Ma MSWD=0.41; n = 3). The younger concordant ages mostly correspond to rims around 289 oscillatory zoned older cores. On a Kernel density estimation (KDE) diagram (Fig. 7j), two highdensity peaks of apparent ages yield weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  ages at  $3521 \pm 19$  Ma (MSWD = 5.4; 290 291 n = 35) and  $3815 \pm 22$  Ma (MSWD= 3.1; n = 15) indicative of ancient Pb-loss around 3800 and 292 3500 Ma. A younger concordant population consisting of recrystallized rims to completely metamict 293 grains (e.g. zircon #C6;Fig. 6d) with very high U concentrations (up to 2599 ppm) and a low Th/U ratio (<0.1), yields a weighted mean  ${}^{207}$ Pb/ ${}^{206}$ Pb age of 2769 ± 22 Ma. A third population of U-rich 294 metamict zircon rims (up to 5790 ppm) yields a weighted mean  ${}^{207}$ Pb/ ${}^{206}$ Pb age of 2649 ± 17 Ma 295 296 (MSWD =1.7; *n* =9).

A total of 22 U-Pb analyses were conducted on sample SG-017 (Fig. 7k). It is dominated by round to elongated metamict zircon grains. Two zircon populations with low Th/U ratios (<0.1) yield weighted mean  ${}^{207}$ Pb/ ${}^{206}$ Pb ages of 2802 ± 13 Ma (MSWD= 1.2; *n* =13) and 2712 ± 13 Ma (MSWD= 1.2; *n* =4). A single zircon grain, perhaps inherited, yielded a Concordia age of 3576 ± 14 Ma.

301 A total of 99 U-Pb analyses were obtained on sample SG-037 (Fig. 71), collected within a granitic unit 302 in contact with mafic metavolcanic rocks. Most zircon grains from this granite sample are black on CL 303 images and display metamict cores or rare zoned cores with high Th/U ratios (>0.2). A consistent zircon population (Fig. 7l) yields a weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2744 ± 8 Ma (MSWD=0.6; *n* 304 =44). Younger grains spread along the Concordia curve from 2744 to 2200 Ma and mostly consist of 305 306 metamict grains with high U concentrations (700-3800 ppm) and relatively low Th/U ratios (<0.3). 307 Two older discordant inherited zircon grains are also present, with the oldest one yielding a <sup>207</sup>Pb/<sup>206</sup>Pb 308 age of  $3303 \pm 48$  Ma.

A total of 38 U-Pb analyses were conducted on sample SG-080 (Fig. 7m). The dominant zircon population is characterized by high Th/U ratios and oscillatory zoned grains yielding a weighted mean  $^{207}Pb/^{206}Pb$  age of  $3612 \pm 9$  Ma (MSWD=0.71; n = 26), interpreted as the crystallization age. Two inherited zircon cores appear to exhibit older ages with one concordant analysis (e.g. zircon #C15; Fig. 6e) which yielded a Concordia age of  $3805 \pm 15$  Ma. Two low Th/U (<0.3) homogeneous black zircon rim and core, yield Concordia ages of  $2752 \pm 9$  Ma (MSWD<sub>(C+E)</sub>=1.4: n = 2).

A total of 18 analyses were conducted on the sample SG-084 (Fig. 7n). This granitic sample is intruded by pegmatites and mostly includes metamict zircon grains commonly containing many inclusions, explaining the limited number of data obtained from this sample. Only 10 oscillatory zoned cores yield a poorly defined population with a weighted mean  ${}^{207}$ Pb/ ${}^{206}$ Pb age of 3710 ± 24 Ma (MSWD= 1.9; *n* =10)

A total of 54 analyses were obtained on sample SG-087 (Fig. 7o). Zircon grains from this granite sample are dominated by sector and oscillatory zoned grains that exhibit variable Th/U ratios. Sector zoned grains are grouped in three major clusters. Two younger populations yielded weighted mean  $^{207}Pb/^{206}Pb$  ages of  $2755 \pm 13$  Ma (MSWD=1.2; n = 15) and  $2584 \pm 14$  Ma (MSWD=1.5; n = 14) whereas the oldest population defines a weak discordia line, but with the two oldest zircon analyses yielding a Concordia age of  $3758 \pm 10$  Ma (MSWD<sub>(C+E)</sub>=1.3; n = 2).

A total of 45 U-Pb analyses were conducted on sample SG-127 (Fig. 7p). Zircon grains with high Th/U ratios (>0.2) showing oscillatory zoned cores with few metamict U-rich grains, yield a weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2789 ± 12 Ma (MSWD=0.83; *n* =18) interpreted as the crystallization age. A younger population with lower Th/U ratios yields a weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2662 ± 18 Ma (MSWD=0.68; *n* =8). One inherited zircon grain with clear oscillatory zoning yields a discordant 331 (~4%)  $^{207}$ Pb/ $^{206}$ Pb age of 3661 ± 48 Ma. All other analyzed zircon grains consist in low Th/U ratio 332 metamict grains with  $^{207}$ Pb/ $^{206}$ Pb ages ranging from 2600 to 2200 Ma.

333 A total of 53 U-Pb analyses were done on sample SG-143 (Fig. 7q). This granite is dominated by 334 metamict zircon grains with subordinate euhedral oscillatory zoned textures (e.g. zircon #B11 335 and #B12; Fig. 6f). These zircon grains exhibit a wide range of Th/U ratios (from 0.02 to 0.7) with the main population yielding a weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2780 ± 8 Ma (MSWD=0.78; *n* =42) that 336 corresponds to the crystallization age. A smaller population of U-rich metamict grains yields a slightly 337 younger weighted mean  ${}^{207}\text{Pb}/{}^{206}\text{Pb}$  age of 2706 ± 19 Ma (MSWD=1.7; *n* =7). Three inherited cores 338 yield older <sup>207</sup>Pb/<sup>206</sup>Pb ages, with a single concordant grain exhibiting a Concordia age of 339 340 3655 ± 12 Ma.

A total of 77 U-Pb analyses were obtained on sample SG-208 (Fig.7r). This granitic sample mostly includes oscillatory zoned zircon grains with low U concentrations (31-400 ppm) and igneous Th/U ratios ranging between 0.2 and 0.6. Concordant zircon analyses yield two different populations. The first population gives a Concordia age of  $3330 \pm 7$  Ma (MSWD<sub>(C+E)</sub> = 1.4; *n* =18) and is considered as the crystallization age. The second slightly younger population yields a weighted mean <sup>207</sup>Pb/<sup>206</sup>Pb age of  $3241 \pm 11$  Ma (MSWD=0.68; *n* =23), possibly reflecting age resetting. Four zircon grains also exhibit a younger weighted mean <sup>207</sup>Pb/<sup>206</sup>Pb age around  $2946 \pm 8$  Ma (MSWD= 0.33; *n* =4).

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#### 4.3 Zircon Lu-Hf isotopic compositions

For each rock sample, a subset of 10 to 30 representative zircon grains has been analyzed for in-situ Hf isotopes (supplementary material Table S4) and selected based on concordance ( $\pm 2$  %) and  $^{207}Pb/^{206}Pb$  age (Table 2). Measurements were acquired on the same spot used for the U-Pb analyses (supplementary material Figure S3). Figure 8 shows the initial  $\epsilon$ Hf values for each zircon grains,

calculated by using their corresponding <sup>207</sup>Pb/<sup>206</sup>Pb age. However, given that the studied samples are 353 354 granitoids with a single crystallization age, the initial Hf isotopic composition for all of the zircon 355 grains from a single sample should be at the same crystallization age. Therefore, initial Hf isotope 356 compositions of each zircon analyses from the same sample were also calculated at the age of 357 crystallization reported in Table 2 and the average initial  $\varepsilon$ Hf values ( $\pm$  2SE) for each sample are also 358 shown on Figure 8. This is performed in order to avoid calculating an initial Hf composition at an over or underestimated <sup>207</sup>Pb/<sup>206</sup>Pb age for zircon grains that would have suffered ancient Pb-loss but 359 360 remained on the Concordia curve (Fisher et al., 2014a, 2014b; Kemp et al., 2010) as illustrated in 361 Figure 9. Figure 8 shows that Eoarchean samples (3883 to 3781 Ma) display initial Hf isotope compositions that are suprachondritic, with average initial  $\varepsilon$ Hf values ranging between +3.7 to +1.7. 362 The Mg-rich tonalite sample SG-026 contains zircon grains exhibiting the most radiogenic Hf isotopic 363 364 composition with an average initial EHf value of  $+3.7 \pm 0.2$  at 3820 Ma, falling on the depleted mantle 365 evolution line. Most samples with positive EHf initial values are tonalitic or trondhjemitic in 366 composition, except for granitic sample SG-007 showing a complex zircon population (Fig. 7j) and with initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios between 0.28027 and 0.28042 for all zircon grains analyzed and an 367 average  $\varepsilon$ Hf initial value of +2.0 at 3883 Ma. However, given the complex and heterogeneous age 368 369 populations of this sample, this initial eff must be taken with cautious as its Eoarchean age is based on 370 the three oldest zircon grains, that may be inherited or recrystallized. If the crystallization age of most zircon grains from this sample is rather Neoarchean, recalculation of initial *EHf* values at 3883 Ma for 371 all zircon analyses would not be representative of its source. Five granitoids with Paleoarchean ages 372 from 3632 to 3330 Ma display subchondritic initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios corresponding to average ɛHf 373 initial values from -1.0 to -6.3. The low *EHf* initial value of -4.6 at 3576 Ma for the oldest zircon grain 374 from the sample SG-017, however, is from an inherited grain and the main Neoarchean zircon 375

376 population for this sample shows an average  $\varepsilon$ Hf of -12.7  $\pm$  0.3 at 2802 Ma. Sample SG-203 is the only 377 granodiorite sample analyzed but displays the same 3330 Ma age and Hf isotopic composition as the 378 spatially associated granitic sample SG-208, with initial  $\epsilon$ Hf values of around -6. The later ~3200 Ma 379 Paleoarchean Mg-rich tonalite from Lister Island, sample SG-265, is characterized by a radiogenic initial Hf isotopic composition and does not appear to follow the general EHf trend vs. time that most 380 other granitoids display on Figure 8. This sample has a slightly suprachondritic initial EHf value of 381 382  $+1.0 \pm 0.3$  at 3224 Ma. The late Mesoarchean granitic sample SG-087 is characterized by an initial EHf of  $-2.6 \pm 0.2$  at 2996 Ma. However, this sample has multiple and complex zircon generations (Fig. 7o) 383 384 and geochronology results should be taken with caution as it also includes few inherited older zircon 385 cores with initial  $\varepsilon$ Hf= +1.5 ± 0.4 at 3745 Ma.

Three Neoarchean granite samples dated between 2789 and 2744 Ma have been analyzed for zircon Hf 386 isotopic composition. They all display low initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios corresponding to average eHf 387 initial values of -11.2 to -14.3. These Neoarchean granite samples include a few inherited older grains 388 389 which  $\varepsilon$ Hf values fall within the main time *vs.* initial  $\varepsilon$ Hf array with two ~3660 Ma cores with initial 390  $\varepsilon$ Hf = -0.2 to -1.7 and one 3303 Ma core with initial  $\varepsilon$ Hf = -7.8. The Neoarchean zircon population of 391 the trondhjemitic sample SG-019 yielded an average *eHf* initial value of -11.3 if the 2732 Ma age if 392 considered as the crystallization age. This sample also includes two zircon cores with older ages of 393 3477 and 3553 Ma that respectively yielded initial EHf values of -3.3 and +7.0. However, given the ambiguity of the crystallization age of this sample, these initial EHf values need to be considered with 394 395 prudence. A few Eoarchean to Paleoarchean granitoids display Neoarchean zircon rims (sample SG-024, SG-025, and SG-027) with variable initial eHf values ranging from ~ -15 to ~ -22. These zircon 396 397 grains, however, show variable and often low Th/U ratios, which could be indicative of metamorphic recrystallization. Therefore, the initial  $\epsilon$ Hf values calculated at these ages may not be reflective of their crustal source.

#### 400 **5 Discussion**

#### 401 5.1 Composition and petrogenesis of the SHC felsic crust

402 The geochemical composition of Archean felsic rocks has been widely used to constrain the nature of 403 their sources and the various processes leading to the formation of Archean crust (e.g. Hoffmann et al., 2019; Laurent et al., 2014; Moyen et al., 2001; Moyen and Martin, 2012; Whalen et al., 2002). Rocks 404 405 from the TTG series make up a major part of the Archean cratonic nucleus forming our stable 406 continents. Various petrogenetic models have been proposed to explain the origin of TTG, and several 407 lines of evidence point to an incompatible element enriched mafic source as their precursor (e.g. 408 Hoffmann et al., 2019, 2011; Jayananda et al., 2015; Moyen, 2011; Moyen and Martin, 2012; Smithies 409 et al., 2009). Melting of this mafic source at medium to high-pressures, with the involvement of 410 residual garnet, is the mechanism dominantly proposed to account for the TTG's typical HREE 411 depletion, correlated with high Sr/Y ratios (Moyen, 2011; Nagel et al., 2012). In comparison, granite 412 requires a K-rich source and are generally thought to derive from the melting of felsic lithologies (e.g. 413 Laurent et al., 2014; Moyen, 2011; Moyen and Laurent, 2018).

Given the high-grade metamorphic conditions of the SHC, our interpretation is focused toward the least mobile elements, although the SHC granitoids appear to have relatively well-preserved wholerock geochemical compositions, despite metamorphism reaching up to granulite facies. Granitoids from the SHC display variable geochemical compositions, but the samples analysed here are consistent with geochemical compositions of SHC felsic rocks from the literature (Bridgwater and Collerson, 419 1976; Schiøtte et al., 1993, 1989a) and can be divided into four main rock types including 420 trondhjemite, Mg-rich tonalite, granodiorite and granite. Table 3 and Figure 10 present the main 421 compositional characteristics of each granitoid groups. While there is no systematic relationship 422 between the age of the SCH granitoids and their whole-rock chemical compositions, there appears to 423 be a general temporal evolution of the SHC felsic rocks composition. The TTG are more commonly 424 found in the Eoarchean and the Paleoarchean, whereas most granite samples are often Neoarchean 425 (Table 2). This broad compositional secular evolution is observed in most Archean cratons (Laurent et al., 2014). 426

427 In general, the SHC granitoids exhibit a wide range of HREE depletion with most rocks exhibiting high (La/Yb)<sub>N</sub> ratios typical of Archean TTG (La/Yb<sub>N</sub>>15; Fig. 4 and 5). The HREE depletion in TTG 428 429 is generally attributed to the presence of residual garnet in the source, which seems to be mainly controlled by the pressure of melting (e.g. Moyen, 2011; Nagel et al., 2012). Most SHC trondhjemite 430 samples exhibit a more pronounced HREE depletion compared to the Mg-rich tonalite and 431 432 granodiorite (Fig. 4 and 5), which would suggest that they were produced from melting at higher 433 pressures. This is also supported by the higher Sr/Y ratios of the trondhjemite compared to the Mg-rich 434 tonalite consistent with formation at lower pressures (Fig. S5).

Several trondhjemite samples exhibits pronounced positive Eu anomalies that appear to correlate with La/Yb and Zr/Nd ratios (Fig. 11). Different petrogenetic processes have been proposed to explain positive Eu anomalies in Archean granitoids such as accumulation of feldspars, fractionation of small amounts of allanite or residual rocks from partial melting (Condie et al., 1985; Martin, 1987; Rudnick, 1992). Given the complex reworking history displayed by some SHC granitoids, supported by the high discordance of some Eoarchean zircon (Fig. 7b-c-d & supplementary material Table S3) and the relative abundance of secondary sector zoned zircon in the trondhjemite, it is possible that the 442 trondhjemite which exhibit positive Eu anomalies represent residual rocks after some extent of partial 443 melting. All trondhjemitic samples analyzed here for U-Pb include at least a small proportion of Neoarchean sector zoned igneous zircon, despite their Eoarchean crystallization ages. The main zircon 444 445 population of sample SG-019, which displays the largest Eu anomaly, is Neoarchean with only 2 older zircon cores preserved. Field observations support the relatively high migmatitisation of the Saglek-446 Hebron Complex crust, which exhibits important, if not systematic, leucocratic veins of granitic melt 447 448 (see supplementary material Fig. S1). Contrastingly, the Mg-rich tonalite and granodiorite do not exhibit such positive Eu anomalies and show higher REE contents, suggesting that their whole-rock 449 450 geochemical composition may not have been affected by remelting event(s) to the same extent as the trondhjemitic rocks. The SHC granodiorite have a REE composition similar to modern granite as 451 opposed to the high La/Yb ratios typically displayed by TTG (Fig. 5). 452

Interestingly, the SHC granitic rocks show a wide range of HREE compositions (Fig. 4) and La/Yb 453 ratios similar to, or much higher than the TTG, as opposed to typical modern granitic rocks (Fig. 5). 454 455 This would be expected if they were mostly produced form re-melting of a precursor with a wide 456 compositional range and high La/Yb ratios, such as the older SHC trondhjemite and Mg-rich tonalite. 457 Most granitic samples exhibit negative to slightly positive Eu anomalies (Fig. 4b and 11) consistent 458 with this petrogenetic interpretation. The leucogranite samples SG-007 and SG-017 are the two 459 exceptions that exhibit pronounced positive Eu anomalies with very low REE concentrations (Fig. 4b 460 and 11) and most likely represent restites from the melting of older crust. This is also consistent with 461 their zircon populations. SG-017 includes mainly low Th/U Neoarchean zircon with a single inherited 462 Paleoarchean zircon (Fig. 7k), whereas sample SG-007 displays a complex zircon population with 463 several Eoarchean zircon that show evidence of important Pb-loss between 3800 and 3500 Ma and 464

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abundant Neoarchean re-crystallized zircon (Fig. 7j). This is indicative of a complex protracted crustal reworking history consistent with the restitic compositions of the leucogranite samples.

466 Figure 12 can be used to determine the potential source for felsic melts and therefore, the precursors of Archean granitoids. The SHC TTG appear to result from the melting of a mafic source characterized 467 468 by variable K contents. The SHC granitic rocks, on the other hand, are more consistent with a felsic 469 crustal source. The older Eoarchean and Paleoarchean tonalite and trondhjemite are the likely crustal 470 precursor sources to the Neoarchean granitic rocks, which would explain the wide range of REE 471 compositions of the granite with La/Yb ratios similar to both the Mg-rich tonalite and the trondhjemite. Several granite samples exhibit negative Eu anomalies which could be complementary with the 472 473 positive anomalies found in the trondhjemitic (perhaps restitic) rocks. The ternary diagram shown in 474 figure 12b was proposed by Laurent et al. (2014) to highlight the relative end-member petrogenetic 475 processes involved in the formation of Archean granitoids. A high Na<sub>2</sub>O/K<sub>2</sub>O ratio indicates the melting of mafic rocks producing K-poor granitoids, a high A/CNK ratio is consistent with an Al-rich 476 477 source such as metasediments, and the FMSB [FSBM = (FeO<sub>t</sub> + MgO)  $\times$  (Sr + Ba) wt. %] value is 478 indicative of the interaction with a metasomatized mantle. Applied to the SHC granitoids, this diagram 479 further supports the hypothesis that the likely source of the tonalite and trondhjemite is a mafic crust, 480 whereas the granite is more consistent with derivation from the melting of Al-rich felsic crust. Some 481 Mg-rich tonalite and granodiorite samples plot towards the FMSB end member, suggesting that they 482 may have interacted with, or included, a mantle component. Similar geochemical characteristics are 483 observed in Archean sanukitoid (Laurent et al., 2014) originally defined as diorite to granodiorite with high Mg# (>0.6), Ni (>100 ppm) and Cr contents (200-500 ppm), with variable TiO<sub>2</sub> contents and 484 relatively high K, Sr, Zr and Nb concentrations (Shirey and Hanson, 1984). Despite the geochemical 485 signature that could be supportive of interaction with the mantle and the relatively high abundance of 486

487 Cr-Ni displayed by the SHC Mg-rich tonalite, it still does not reach the typical enrichment in Ni-Cr-Sr 488 found in typical sanukitoid (Heilimo et al., 2010; Martin et al., 2009; Martin and Moyen, 2005). The 489 SHC tonalite, nevertheless, exhibits high Mg, Fe, Cr, Ti and V concentrations relative to the 490 trondhjemite, but perhaps not quite comparable to typical Neoarchean sanukitoid.

# 491 **5.2Geochronology of the SHC granitoids**

Early work on the SHC granitoids associated the main episodes of felsic magmatism with defined rock 492 493 compositions. For example, the Uivak I gneiss was described as ~3700-3800 Ma tonalite, whereas the 494 Uivak II gneiss was defined as ~3600 Ma granodiorite (Baadsgaard et al., 1979; Bridgwater and 495 Schiøtte, 1991; Nutman and Collerson, 1991). It has become clear with the more recent work, 496 however, that specific geochemical compositions are not associated with specific ages. This is also 497 evident from the new dataset we present in this study. While Mesoarchean and Neoarchean felsic rocks 498 are mostly composed of granite (with perhaps subordinate trondhjemite), the Paleo- to Eoarchean 499 granitoids in the SHC include trondhjemitic, tonalitic, granodioritic and granitic rocks, (Table 2). 500 Therefore, we propose here that the terminology for the different SHC units (e.g. "Iqaluk" or "Uivak 501 I", etc...) refers to distinct temporal magmatic events, regardless of geochemical compositions, we will 502 use names such as: "Uivak I Mg-rich tonalite", "Uivak I trondhjemite" or "Iqaluk Mg-rich tonalite", in 503 order to refer to both the age and respective geochemical composition.

To get a statistical overview of the age distribution of the felsic magmatism in the SHC, we have used Kernel density estimation diagrams (KDE; Fig. 13a-b) to highlight the probability of a zircon to be either metamorphic or magmatic, based on the textural and chemical characteristics of each zircon, at a given time. Probabilities from KDE, provide an overview of maximum magmatic production, thermal recrystallization and relative timing between different maximums, as well as show whether magmatic production is punctual (sharp Gaussian peak) or diffused in time (asymmetrical peak or large wavelength Gaussian distribution). It should also be noted that sample bias or missing data can be acaveat for interpretation of these KDE.

512 **5.2.1 Magmatic history** 

513 The SHC is one of the rare geological terrains on Earth preserving Eoarchean rocks. Precise and 514 accurate age determination of Earth's oldest rocks has important implications, as it brings timing 515 constraints on various geological processes to understand the early Earth (e.g. Whitehouse et al., 2019). Shimojo et al. (2016) proposed age of  $3920 \pm 49$  Ma for a banded grey gneiss sample 516 517 (LAA995) they interpreted as from the Iqaluk gneiss. This would represent the second oldest 518 occurrence of felsic crust on Earth, after the Acasta gneiss (Bowring and Williams, 1999; Reimink et 519 al., 2016). However, over 300 spots from 231 zircon were analyzed by LA-ICP-MS for this sample and only the 6 oldest zircon were used to define this age, of which 5 yield over-concordant ages. 520 521 Whitehouse et al. (2019) recently questioned the exactitude of this age and pointed out that a larger 522 subset of analyses from this granitoids yields a statistically significant population with a mean  $^{207}$ Pb/ $^{206}$ Pb age of 3865 ± 4 Ma. Sample SG-210c (this study) is the same banded grey gneiss, collected 523 524 on the same outcrop as sample LAA995 (supplementary material Fig. S4). The Concordia age we 525 obtained for the 34 oldest concordant zircon from sample SG-210c is  $3869 \pm 6$  Ma (Fig. 7g). When we 526 apply the same data filtering method used for SG-210c to select a subset of analyses from sample LAA995 (±2% concordance; Th/U >0.3; <sup>207</sup>Pb/<sup>206</sup>Pb age older than 3800 Ma), it defines a consistent 527 concordant population with a well-defined Gaussian distribution around a Concordia age of 528 529  $3869\pm5$  Ma (MSWD=1.1 *n* =181). Although it still represents one of the oldest rocks on Earth, the 530 oldest Iqaluk granitoids unit is thus more consistent with a <3900 Ma age, which was displayed by 531 other granitoids samples from the SHC, such as  $3860 \pm 10$  Ma (Vezinet et al., 2018);  $3851 \pm 46$  Ma, 532 3829 ± 27 Ma, 3869 ± 31 Ma, 3895 ± 33 Ma and 3897 ± 33 Ma (Komiya et al., 2017); 3849 ± 260 Ma
533 (Collerson, 1979); 3863 ± 12 Ma (Schiøtte et al., 1989a).

534 Figure 13a displays the probability density for concordant zircon ( $\pm 2\%$ ) that exhibit Th/U ratios >0.3 535 and igneous textures (sector/oscillatory zoned), interpreted to represent the probability of 536 crystallization ages. The limit of 0.3 for Th/U ratios was set based on the correlation between the rock 537 crystallization age and the zircon's Th/U ratio. Within our dataset, the zircon grains that are closer in 538 age to the interpreted crystallization age of their host rocks, exhibit higher Th/U ratios that are generally above 0.3. The Concordia age deconvolution analysis shows that three distinct probability 539 540 peaks of zircon production are recorded during the Eoarchean and early Paleoarchean, at 3857 Ma, 541 3744 Ma and 3575 Ma (relative misfit of 0.131). In contrast to the proposed protracted and continuous 542 magmatic activity proposed by Komiya et al. (2017), we suggest that the magmatic activity is most 543 likely represented by three discrete magmatic pulses during the Eoarchean, which would correspond to 544 the units previously defined as the Iqaluk gneiss, the Uivak I gneiss and the Uivak II gneiss.

545 Between 3400 and 3200 Ma, two distinct generations of granitoids intruded the SHC, supported by the 546 Concordia age of  $3224 \pm 7$  Ma for the Lister gneiss sample SG-265 and the ages of  $3330 \pm 15$  Ma and 547  $3330 \pm 7$  Ma, obtained respectively from samples SG-203 and SG-208. The 3224 and 3330 Ma 548 samples have distinct initial EHf values of +1 and -6, and we thus interpret these felsic magmas to be 549 derived from distinct sources produced from two separate magmatic events. We therefore define the 550 Lister gneiss unit as being emplaced at ~3220 Ma and propose a distinct ~3330 Ma magmatic event, 551 referred to as the "Iluilik", the local Inuit name for the area described as the "opposite coast of Nulliak 552 Island" by Komiya et al.(2017, 2015), where the 3330 Ma samples were collected. Although 553 recognizably distinct, the Iluilik magmatic event appears to be a minor component of the SHC mostly 554 located around the Nulliak Island and its Iluilik opposite coast.

A prominent Neoarchean event is dominated by granitic intrusions and defines a single magmatic peak with an age of 2750 Ma (Fig. 13a). This suggests that the Neoarchean thermal event generally associated with peak metamorphism in the SHC (e.g. Komiya et al., 2017; Kusiak et al., 2018; Nutman and Collerson, 1991; Schiøtte et al., 1989b; Van Kranendonk, 1990), was also accompanied by an important magmatic activity.

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## 5.2.2. Metamorphic history

The SHC has been subjected to a complex thermal history with prevalent metamorphism between 561 562 2700-2800 Ma, but later high-temperature events have also been recognized (Komiya et al., 2017; 563 Kusiak et al., 2018; Sałacińska et al., 2019, 2018; Schiøtte et al., 1992; Van Kranendonk, 1990). 564 Figure 13b shows the KDE diagram of the probability density ages for recrystallized zircon grains and 565 those exhibiting evidence of age resetting. Only zircon grains yielding concordant ( $\pm 2\%$ ) ages, and 566 thus fully reset ages, are included on Figure 13b. Zircon grains which exhibit low Th/U ratios (>0.3)567 and homogeneously black texture or heterogeneous patterns (metamict) have also been used as criteria 568 for metamorphic zircon. Eoarchean zircon grains consistent with a metamorphic recrystallization 569 exhibit only one major probability peak that appears to be contemporaneous, if not slightly later, to the ~3600 Ma Uivak II magmatic event. This would be consistent with a thermal event described in 570 571 previous work (Bridgwater et al., 1975; Collerson, 1983b; Sałacińska et al., 2018; Van Kranendonk, 572 1990). These concurrent igneous and metamorphic events are supported by the presence of 3600 Ma 573 metamorphic recrystallization rims surrounding pristine igneous oscillatory zoned 3860 Ma old zircon 574 cores (e.g. SG-210c; Fig. 6c & Fig. 7g; supplementary material Table S3), as well as the presence of 575 inherited 3805 Ma igneous cores surrounded by a Uivak II oscillatory zoned crystallization (e.g. SG-576 080; Fig 6e & Fig 7m; supplementary material Table S3). An older subordinate Eoarchean population 577 of metamorphic recrystallized zircon grains is seen at ~3750 Ma, but we suggest this could result from 578 ancient Pb-loss in zircon from the Iqaluk gneiss rather than reflecting a distinct metamorphic event. 579 Two minor metamorphic peaks can be observed at 3250 Ma and ~3000 Ma (Fig. 13b), but these late 580 Paleo- to Mesoarchean metamorphic events remain equivocal due to the lack of resolution of the KDE 581 diagram and the relative rarity of these populations.

582 The highest probability peak of recrystallization occurs during the Neoarchean (Fig. 13b), which is 583 consistent with the extensive metamorphic event previously suggested at 2700 Ma (e.g. Kusiak et al., 584 2018; Schiøtte et al., 1989b). However, our maximum of recrystallized zircon ages appears to occur 585 closer to 2800 Ma, perhaps shortly before the 2750 Ma Neoarchean peak defined by igneous zircon 586 grains (Fig. 13a). This may suggest a slight delay (~50 Ma) between the maximum intensity of the 587 thermal event and the maximum probability of granitic magmatism, which may correspond to crustal 588 anatexis. The overall distribution of the Neoarchean recrystallized zircon shows an asymmetric peak 589 shape that reaches its maximum at 2800 Ma, before gradually decreasing to lower probabilities until 590 2200 Ma, after which no zircon is produced in the SHC. This asymmetric peak could be an artifact 591 from our selection criteria used to build the KDE diagram, which could comprise zircon grains that 592 experienced incomplete Pb-loss. However, we cannot disregard the possibility that it suggests 593 protracted high-grade metamorphism before gradually decreasing until 2400-2200 Ma. The apparent 594 asynchrony between the igneous and metamorphic zircon ages may therefore be due to some 595 metamorphic zircon grains that have not been fully reset. The zircon grains with ages <2700 show high 596 U concentrations (Fig. 13b top right inset) and could be more susceptible to reopening of the U-Pb 597 system due to the higher rate of U decay causing the breakdown the zircon crystal lattice (Lee et al., 598 1997). Therefore, a protracted thermal event that gradually decreased in intensity would keep the U-599 rich zircon in isotopically open conditions longer than those with lower U contents. The latter would 600 then record older ages compared to the more U-rich zircon grains. This protracted thermal event is also 601 in agreement with what was suggested by Kusiak et al. (2018) based on younger ages obtained on 602 monazite and apatite in the SHC of 2600-2500 and 2200 Ma respectively.

#### **5.3. Crustal sources and reworking history**

604 The U-Pb-Hf isotopic composition of the Eoarchean and Hadean Jack Hills detrital zircons provided 605 invaluable information about the early crust (e.g. Amelin et al., 1999; Blichert-Toft and Albarède, 606 2008; Harrison, 2005; Kemp et al., 2010), but the fact that their host rocks have been eroded away 607 limits the constraints we can put on the earliest crustal history. Possible ancient Pb-loss in detrital zirconscan also result in younger apparent <sup>207</sup>Pb/<sup>206</sup>Pb ages and consequently bias the calculated initial 608 609 Hf isotopic composition (e.g. Amelin et al., 2000; Vervoort and Kemp, 2016). The SHC zircon grains 610 studied here are all from rock samples with a single interpreted crystallization age. As shown on 611 Figure 9, to avoid the possible calculation of the Hf isotopic compositions at an incorrect apparent Pb-612 Pb age, we are considering the average initial EHf value for all magmatic zircon grains from each 613 granitoids, calculated at the crystallization age of their respective host rock (Table 2, Fig. 14). The 614 composition we attribute to the precursor crustal source of the rocks hosting the zircon grains has an 615 important implication on how the same dataset can be interpreted regarding crustal history. For 616 example, a 3.5 Ga zircon with an initial EHf value of -5 could be produced from the remelting of a 3.8 Ga felsic crustal precursor or a 4.3 Ga mafic crustal precursor, assuming <sup>176</sup>Lu/<sup>177</sup>Hf ratios of 0.01 and 617 0.025 respectively, and a chondritic initial <sup>176</sup>Hf/<sup>177</sup>Hf ratio for both sources. To better constrain the 618 619 Lu/Hf ratio for the crustal precursor(s) of the SHC granitoids, we use here the whole-rock geochemical 620 composition of each of the host rock samples as a proxy for the composition of their crustal sources 621 and evolution. As discussed previously, we interpret SHC TTG to be derived from the melting of a mafic precursor, while the granitic rocks are consistent with the melting of a felsic crustal source (Fig.12a).

624 The Eoarchean Iqaluk and Uivak I gneiss exhibit slight positive initial  $\epsilon$ Hf values from +1.7 to +3.7, 625 between 3700 and 3900 Ma (Fig. 14), suggesting that the precursor source of these granitoids had a 626 suprachondritic Hf isotopic composition in the Eoarchean. This contrasts with zircon from most other 627 Eoarchean TTG, generally displaying chondritic or subchondritic initial EHf values (e.g. Guitreau et al. 2012; Iizuka et al., 2009; Næraa et al., 2012; O'Neil et al., 2013; Reimink et al., 2016), leading some 628 629 authors to suggest that widespread chemical depletion of the mantle, did not take place prior to 630 ~3800 Ma (e.g. Vervoort and Kemp, 2016). Vezinet et al. (2018) recently concluded that a 3860 Ma TTG sample from the SHC included zircon with chondritic initial EHf values within uncertainty. 631 632 However, when the  $\varepsilon$ Hf values for each zircon analysed are all calculated at the crystallization age of the host rock of 3860 Ma, such as the approach from study, it yields an average initial EHf value of 633  $+1.6 \pm 0.2$  (2 S.E.). This is consistent with the slight suprachondritic Hf isotopic compositions we have 634 635 obtained for all SHC Eoarchean granitoids (Fig. 14). We, therefore, argue that the oldest granitoids 636 from the SHC were sourced from reservoir characterized by a suprachondritic Lu/Hf ratio. The 3820 Ma Iqaluk sample SG-026 yields the highest initial *EHf* value of +3.2. To evolve to such positive 637 εHf values by 3800 Ma, a reservoir formed at 4568 Ma with chondritic <sup>176</sup>Hf/<sup>177</sup>Hf would need a 638 <sup>176</sup>Lu/<sup>177</sup>Hf ratio of 0.0435. The other SHC Eoarchean granitoids have suprachondritic initial EHf 639 values corresponding to time-integrated <sup>176</sup>Lu/<sup>177</sup>Hf ratios for their source ranging from 0.0393 to 640 641 0.0412. This would suggest that the crustal precursor source of the SHC Eoarchean granitoids was 642 derived from a mantle with a comparable to slightly higher degree of depletion than the present-day depleted mantle with  ${}^{176}Lu/{}^{177}Hf = 0.03933$  (Blichert-Toft and Puchtel, 2010). 643

644 The Uivak gneiss has previously been interpreted to result from the melting of the Nulliak mafic 645 supracrustal rocks (Komiya et al., 2017, 2015; Morino et al., 2018; Nutman and Collerson, 1991; 646 Shimojo et al., 2016). A Lu-Hf isochron, mainly including ultramafic rocks interpreted to be from the 647 Nulliak assemblage, yielded an initial  $\epsilon$ Hf value of +5.1 at 3794 Ma(Morino et al., 2018) with  $\varepsilon$ Hf<sub>(3770Ma)</sub> for individual samples as high +12.8. No zircon analyses from SHC granitoids have 648 649 however yielded such high initial Elf values (this study; Vezinet et al., 2018). The highly positive Elf 650 values of the Nulliak rocks may in part be due to some extent of disturbance of the Lu-Hf isotopic 651 system, as suggested by the high MSWD value of 142 of the Lu-Hf isochron (Morino et al., 2018). It 652 nevertheless suggests that the SHC includes Eoarchean mafic/ultramafic crust with suprachondritic Hf 653 isotopic compositions, which would be a possible crustal source for the Iqaluk and Uivak I TTG. The Iqaluk granitoids, however, appear to predate the Nulliak mafic rocks, which would argue against 654 655 their derivation from melting of the Nulliak metabasalts. All long-lived isotopic systems used to 656 constrain the age of the Nulliak mafic rocks, such as Sm-Nd, Lu-Hf or Re-Os (Collerson et al., 1991; Ishikawa et al., 2017; Morino et al., 2018, 2017), however, show evidence of some degree of 657 658 disturbance with large errors on isochron ages (hundreds of million years) and we therefore cannot rule 659 out the Nulliak mafic rocks as the possible source of the Iqaluk gneiss.

Although trondhjemite and Mg-rich tonalite are more dominant within the Eoarchean Iqaluk and Uivak I gneiss, tonalitic samples also occur within the ~3600 Ma Uivak II and ~3200 Ma Lister gneiss. The Uivak II tonalitic sample has a slight subchondritic initial  $\epsilon$ Hf value of -1.1 at 3632 Ma. While reworking of Eoarchean felsic crust evolving with a low <sup>176</sup>Lu/<sup>177</sup>Hf ratio of ~0.011 could explain the zircon Hf isotopic composition of this Uivak II tonalite sample (Fig. 14), its whole-rock geochemical composition rather suggests derivation from a mafic crustal source (Fig. 12a). Previous work showed that the mafic amphibolites from the SHC display a wide range of <sup>176</sup>Lu/<sup>177</sup>Hf [e,g, 0.016 to 0.032 (Morino et al., 2018)] but assuming a <sup>176</sup>Lu/<sup>177</sup>Hf ratio between 0.020 and 0.026 for this mafic
precursor, more typical of a basaltic crust (Amelin et al., 2000; Blichert-Toft and Albarède, 2008;
Kemp et al., 2010), it would suggest a Hadean age between ~4000 and ~4200 Ma for the crustal source
of the Uivak II tonalite, if this mafic crust was derived from a long-term depleted mantle-like reservoir
(Fig. 14).

672 The initial EHf value of +1.0 for the 3224 Ma Lister tonalitic gneiss contrasts with the lower initial EHf value of ~-6 for the 3330 Ma Iluilik granitoids, suggesting that although these granitoids units were 673 produced only ~100 Ma apart, they were derived from sources with distinct early histories. If we 674 consider a mafic composition for the crustal precursor of the tonalitic Lister gneiss (Fig. 12a), it would 675 676 suggest a ~3500 Ma to ~3600 Ma mafic crustal source (Fig. 14). Morino et al. (2018, 2017) proposed the occurrence of Paleoarchean mafic rocks in the SHC, but their chondritic <sup>176</sup>Hf/<sup>177</sup>Hf composition 677 678 at ~3400 Ma suggest it would not be a suitable precursor source for the Lister gneiss. Other potential 679 mafic crustal sources for the Lister tonalite would include the ~3800 Ma Nulliak mafic supracrustal 680 rocks or perhaps the Paleoarchean Saglek dikes (Baadsgaard et al., 1979). Alternatively, the Lister 681 gneiss could be derived from the melting of older mafic crust, similar to the source of the Iqaluk-Uivak 682 I or Uivak II, but with contribution of juvenile material or interaction with the depleted mantle. The 683 Lister gneiss, however, does not exhibit the geochemical features that are typically interpreted as 684 reflecting an interaction between TTG melts and the mantle, such as high Cr and Ni (e.g. Moyen and 685 Martin, 2012) nor evidence for a high component of metasomatized mantle (Fig 12b). Regardless of 686 the exact source of the Lister gneiss, its zircon Hf isotopic composition denotes the contribution of a 687 more juvenile component at the end of the Paleoarchean.

Both the granite and granodiorite ~3300 Ma Iluilik samples yielded comparable initial εHf values of ~6. While the granitic sample could be produced by reworking of the older SHC Eoarchean felsic crust

(Fig. 14), the whole-rock geochemical composition of the granodiorite would be more consistent with a high-K mafic crustal source (Fig 12a). A mafic crustal reservoir would need to have been formed before ~4.1 Ga to evolve to an initial ɛHf value of -6 at 3300 Ma (Fig. 14). No evidence of Hadean mafic crust has been observed in the SHC, but the existence of Hadean hydrothermally altered enriched mafic crust has been proposed as the source of the Jack Hills detrital zircons (Kemp et al., 2010), the Acasta gneiss (e.g. Reimink et al. 2016, 2019) and the Nuvvuagittuq TTG (O'Neil et al., 2013; O'Neil and Carlson, 2017).

697 Another scenario than melting Hadean mafic crust that could possibly explain the low EHf values of 698 the zircon grains from the ~3600 Ma Uivak II and ~3300 Ma Iluilik TTG, would be derivation from a 699 mixed crustal source. One could imagine that by the Paleoarchean, the SHC crust was a mixture of 700 mafic Nulliak metavolcanic-type rocks and felsic Iqaluk-Uivak I tonalite-trondhjemite. The 701 contribution of an older felsic crustal component in the source of the Paleo- to Mesoarchean TTG 702 could result in lower initial EHf values. Assuming that both mafic and felsic end-member sources were 703 formed at 3880 Ma, with respective Hf concentrations of 1.1 and 3.5 ppm [average Hf concentrations for the Nulliak mafic rocks (Wasilewski et al., 2019) and SHC tonalite-trondhjemite (Table 1)] and 704 evolved with respective <sup>176</sup>Lu/<sup>177</sup>Hf ratios of 0.025 and 0.01, it would, require that the felsic 705 706 component represent nearly 70% of the mixed crustal source to explain the zircon Hf isotopic composition of the 3630 Ma Uivak II tonalite. Even if a lower <sup>176</sup>Lu/<sup>177</sup>Hf ratio of 0.001 707 708 [corresponding to the lowest Lu/Hf ratio measured in the SHC trondhjemite (Table 1)] is considered 709 for the felsic source, more than 50% felsic component is needed to account for the EHf value of the Uivak II tonalite. This is however inconsistent with its whole-rock geochemical composition which 710 711 infers a dominantly mafic source (Fig. 12). Mixing older SHC felsic rocks in the source of the 3300 Ma Iluilik granodiorite to account for its low zircon initial EHf value is also unlikely as the Iluilik 712

713 rocks have the highest Hf concentrations (>6.8 ppm) of all SHC felsic rocks and therefore, felsic crust 714 assimilation or mixing would only have a limited effect on its Hf isotopic composition. Except for 715 Neoarchean sample SG-087, all other SHC granite samples define an initial EHf vs. time trend consistent with the reworking of an Eoarchean felsic reservoir evolving with a  $^{176}Lu/^{177}Hf$  of ~0.011 716 717 that originates from the Iqaluk/Uivak I gneiss (Fig. 14). Most inherited zircon grains are found in 718 granitic samples and follow the same felsic reservoir trend (Fig. 14), which further supports the 719 hypothesis that the granite is produced from the melting of the Eoarchean felsic crust. The combined 720 whole-rock geochemical and Hf isotopic compositions of the granitic rocks of Iqaluk, Uivak I, Uivak 721 II, Iluilik and Neoarchean ages, therefore suggest that they were produced from the remelting of the SHC Eoarchean tonalite/trondhjemite over ~1 billion years. The ~3000 Ma granitic sample SG-087, 722 however, would be consistent with the reworking of the ~3200 Ma Lister gneiss rather than the 723 Eoarchean TTG. Except perhaps for the trondhjemite sample SG-019 with an equivocal crystallization 724 725 age, all Neoarchean granitoids analyzed here are granitic in composition with low initial zircon EHf 726 values. This suggests that the  $\sim$ 2750 Ma felsic magmatism is dominated by remelting older felsic crust, 727 with little to no input from a more juvenile source. The major Neoarchean high-grade metamorphic 728 event that affected the SHC, therefore, appears to also have been accompanied by important crustal 729 reworking and anatexis.

Given that the SHC is part of the North Atlantic Craton, it is often compared to the Itsaq Gneiss Complex of Southwest Greenland, also comprising Eoarchean TTG and supracrustal rocks (Bridgwater et al., 1990; Collerson, 1983a; McGregor, 1973; Morino et al., 2017; Næraa et al., 2012; Wasilewski et al., 2019). Figure 14 shows the evolution of Hf isotopic compositions for detrital and igneous zircon from southwest Greenland (Næraa et al., 2012; 2014) compared to the SHC igneous zircon. The Eoarchean zircon grains from Greenland overall display lower initial eHf values, with

736 mostly chondritic to slightly subchondritic compositions, compared to the SHC where the 3700-737 3900 Ma zircon grains generally display positive initial *EHf* values (Fig. 14). The zircon Hf isotopic compositions from both SW Greenland and the SHC are mostly consistent with reworking of 738 739 Eoarchean crust until the late-Paleoarchean when it abruptly shifts from low initial eHf values around 740 3300 Ma to suprachondritic compositions at ~3200 Ma (Fig. 14; Hoffmann et al., 2011; Næraa et al., 2012). This was interpreted by Næraa et al. (2012) to represent a change of geodynamic setting in SW 741 742 Greenland that involved juvenile crust generation by plate tectonic processes. An important input of 743 juvenile crust was recorded throughout the Mesoarchean in SW Greenland with mostly positive initial zircon EHf values between 3200 and 2800 Ma (Næraa et al., 2012), but the lack of Mesoarchean 744 zircon-bearing rocks in the SHC does not allow to assess if the same processes were involved during 745 that time. Similarly to the SHC Neoarchean granitoids, the Neoarchean Qôrqut Granite Complex of 746 747 SW Greenland displays low zircon EHf values between -12 and -18 at ~2550 Ma. However, these are 748 believed to be derived from an Eoarchean mafic source (Naeraa et al., 2014) rather than from 749 reworking of older felsic crust, such as what we propose for the SHC Neoarchean granitoids. 750 Therefore, the dominant crustal source of the SHC and SW Greenland felsic rocks seems to diverge in 751 the Neoarchean.

752

#### **5.4.** Tectonic context of the SHC

One of the most highly debated subjects about the early Earth concerns the tectonic setting operating and responsible for the formation of the Archean cratons (e.g. Bédard, 2016; de Wit, 1986; Grove et al., 2003; Johnson et al., 2013; Moyen and Laurent, 2018; Nutman and Bennett, 2019; Smithies and Champion, 2000; Wiemer et al., 2018). The geochemical and isotopic compositions of ancient rocks have been widely used to constrain the tectonic environments in which they formed, but there is still

758 no consensus on Earth's early geodynamics. Although the compositions of the SHC metavolcanic 759 rocks do not exhibit geochemical signatures typically found in suprasubduction environments 760 (Wasilewski et al., 2019), Komiya et al. (2015) suggested evidence of Eoarchean subduction settings 761 in the SHC. Based on field observations, they describe the stratigraphy of the Nulliak supracrustal 762 assemblage as an analog to the duplex structures observed in the Japanese trench, interpreted as large 763 accretion prisms. In this model, the Uivak I granitoids would be produced from the melting of basaltic 764 crust within an accretionary complex at shallow depths. However, Uivak I and Igaluk TTG exhibit 765 strong HREE depletion (Fig. 4 and Fig. 5) consistent with the melting of a garnet-bearing precursor at 766 relatively high-pressures, which is at odds with shallow-level melting of a tholeiitic or possibly 767 komatiitic crust. Although U-Pb and Hf isotopes cannot directly be linked to tectonic settings, these 768 data can help to constrain the type of crustal precursor and its reworking history, to better establish the 769 architecture of the reworked crustal sources and evaluate which tectonic context would be more likely. 770 Figure 15 shows the rock or inherited zircon crystallization age vs. mantle extraction Hf model age for 771 the source of the SHG TTG. The mantle extraction model ages are obtained using the zircon average initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios for each of the samples, back-calculated to the depleted mantle evolution line, 772 and assuming a <sup>176</sup>Lu/<sup>177</sup>Hf between 0.020 and 0.026 for a mafic crustal reservoir. Consequently, only 773 774 the samples consistent with derivation from a mafic crustal precursor (i.e. the TTG) and inherited 775 zircon grains from granitic samples (which likely crystallized within their TTG precursor) are included 776 on Figure 15. Although it is more difficult to assess if the inherited zircon grains were affected by 777 ancient Pb-loss, which could lead to erroneous calculation of their initial Hf isotopic composition, data 778 from inherited grains are consistent with the other samples and follow the same trend on Figure 15. 779 The zircon grains from Eoarchean granitoids are the only samples with relatively juvenile 780 compositions, consistent with the remelting of almost contemporaneous mantle-derived mafic crust,

781 potentially the Nulliak basaltic crust. The younger Uivak II and Iluilik granitoids appear to be derived 782 from the melting of pre-Nulliak Hadean mafic crust. Except for the Lister gneiss (sample SG-265), the 783 SHC granitoids derived from the melting of mafic crust show a negative correlation between their 784 crystallization age and their Hf model age, suggesting that the latest granitoids were produced from the 785 melting of the oldest crustal source. In a modern-style subduction setting, it would be unlikely that: -1) 786 the subducting mafic crustal reservoir displayed such a wide age range (from ~3900 to ~4400 Ma) and 787 -2) the oldest portions of the subducting mafic crust melted the latest. While one could suggest that a 788 thick mafic crust, similar to an oceanic plateau, could include mafic crust with the range of ages 789 suggested by the Hf model ages from Figure 15, it would be expected that the base of this thickened 790 mafic crust, *i.e.* the oldest portions, melts before the younger overlying portions. This is inconsistent 791 with the negative trend observed in Figure 15. Some authors have proposed a "subcretion" model as an 792 alternative to modern-style subduction tectonics (Barr et al., 1999; Ducea et al., 2009; Grove et al., 793 2003; Hacker et al., 2015; Taramon et al., 2015). Sequential stacking or tectonic imbrication have been 794 suggested as a mechanism for early Archean crustal growth (e.g. de Wit, 1986; Smithies and 795 Champion, 2000) and the formation of the Eoarchean Itsaq Gneiss Complex of Southwest Greenland 796 (Nutman et al., 2007; Nutman and Bennett, 2019). This tectonic regime has the potential to imbricate 797 portions of mafic crust of variable ages in a configuration where older crust could overlie younger 798 crust and perhaps melt later, in a thickened oceanic crust. Reworking of such an imbricated mafic crustal source could explain the crystallization age vs. Hf model age trend seen for the SHC TTG. 799

### 800 **6** Conclusion

The SHC granitoids record more than 1 billion years of complex crustal history, including several episodes of felsic magmatism. The different generations of gneiss in the SHC were previously 803 associated with certain geochemical characteristics, but they appear to be more geochemically and 804 petrologically diverse. We, therefore, suggest that the different gneissic units should refer to distinct temporal magmatic events regardless of composition. Six distinct felsic magmatic events can be 805 806 identified in the SHC: the oldest 3850 Ma Iqaluk gneiss, the 3750 Ma Uivak I gneiss, the 3600 Ma 807 Uivak II gneiss, the 3330 Ma Iluilik gneiss, the 3230 Ma Lister gneiss, and the 2700-2800 Ma 808 Neoarchean granitoids. The chondritic to subchondritic Hf compositions that Eoarchean zircons from 809 most other early terrains exhibit has led to the suggestion that the depletion of the mantle through 810 extraction of crustal material prior to 3.8 Ga was negligible (e.g. Vervoort and Kemp, 2016). The 811 slightly suprachondritic Hf isotopic compositions of the Eoarchean zircon from the SHC granitoids, however, denote the existence of a long-term depleted source, more consistent with the evidence for an 812 early depleted mantle as suggested from Nd isotopes (e.g. Bennett et al., 2007, 1993; Boyet and 813 814 Carlson, 2006; Caro et al., 2003; O'Neil et al., 2016; Rizo et al., 2011). Most of the juvenile 815 continental crust in the SHC appears to have been formed during the Eoarchean. Considering the 816 geochemical composition of the SHC granitoids to constrain the nature of their crustal precursor, the 817 Hf isotopic compositions of the different generations of TTG suggest the remelting of Eoarchean to Hadean mafic crust. The juvenile Iqaluk and Uivak I TTG are consistent with remelting of ~3800-818 819 3900 Ma mafic crust, while the younger Uivak II and Iluilik TTG would suggest melting of 820 increasingly older (up to ~4300 Ma) mafic sources As observed in SW Greenland, the SHC granitoids 821 show a marked transition between ~3300 and 3200 Ma from relatively unradiogenic to more juvenile 822 compositions. The Neoarchean felsic magmatism in the SHC, however, appears to be dominated by 823 the reworking of the Eoarchean TTG, without contribution of juvenile material.

#### 824 Acknowledgements:

We acknowledge the Nunatsiavut Government and Parks Canada permission to work in the SHC area. 825 826 We are thankful to the staff from the Torngat Mountains Base Camp and Research Station, and 827 particularly J. Goudie and W. Broomfield, for their help during fieldwork, and to Tuumasi Itua 828 Annanack, John, Eli, Joe, Ryan as well as all other bear guards to have kept us safe. We thank B. Ryan 829 from the Depart. of Natural Res. of Newfoundland for sharing samples. This manuscript was improved by comments from A. Bauer and an anonymous reviewer as well as from the editor N. Wodicka. This 830 research was supported by a Natural Sciences and Engineering Research Council of Canada Discovery 831 832 grants to J.O. (RGPIN 435589-2013) and H.R. (RGPIN-2015-03982 and RGPIN-477144-2015), the 833 Ontario Early Researcher Award to J.O. and Carnegie Canada grant to H.R.

### 834 **Captions**

# **Figures:**

- Figure 1: Simplified geological map of the Saglek-Hebron Complex (SHC) modified from Ryan and
  Martineau (2012) and Komiya et al. (2015). Sample locations (yellow circles) show the main localities
  where multiple samples were collected for this study. Coordinates are in UTM NAD 27 zone 20.
- 839
- Figure 2: Ab-An-Or ternary diagram (Barker, 1979) for the SHC granitoids. Shaded fields with smaller
  light colored samples represent SHC granitoids compositions from the literature (Bridgwater and
  Collerson, 1976; Collerson and Bridgwater, 1979; Schiøtte et al., 1993, 1989b).
- 843

Figure 3: Selected major element compositions of the SHC granitoids. a), c) and d) selected elements respectively *vs.* MgO b)  $TiO_2$  *vs.* FeO<sub>t</sub> e) and f) have MgO + FeO<sub>t</sub> on their abscissa showing covariations with CaO (wt. %) and A/CNK (relative aluminum concentration in molar unit calculated as followed A/CNK = Al/(2Ca + Na + K). Symbols are as in Figure 2.

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Figure 4: a) Primitive mantle normalized trace element diagrams , b) Chondrite normalized rare earth element diagrams for the SHC granitoids (McDonough and Sun, 1995). Shaded fields show the data for SHC granitoids from the literature (Bridgwater and Collerson, 1976; Collerson and Bridgwater, 1979; Schiøtte et al., 1993, 1989b). Symbols and colors for fields are as in Figure 2. Normalization values for primitive mantle are from Lyubetskaya and Korenaga (2007) and for chondrite are from McDonough and Sun (1995). Some specific samples discussed in the text are emphasised by thicker lines and are labled.

Figure 5:  $(La/Yb)_N vs$ . Yb<sub>N</sub> diagram reflecting LREE/HREE ratios of all SHC TTG and granite compared to typical TTG, sanukitoid and modern granite from the literature (Moyen and Martin (2012) and references therein). Normalization values are after Masuda et al. (1973). Symbols are as in Figure 2.

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Figure 6: Cathodoluminescence images of zircon grains with U-Pb (red dashed circles) and Lu-Hf (blue dashed circles) LA-ICP-MS spots. a) Trondhjemite sample SG-024: grain #A05 shows typical sector zoning of zircon that crystallized during the 2700-2800 thermal events. Zircon #A17 exhibiting igneous oscillatory zoning and a younger core age relative to the rim. b) Mg-rich tonalite sample SG-

865 026 from Ukkalek Island: grains #B16 and #E12 show respectively oscillatory and sector zoned cores 866 with discordant black colored rims. c) Mg-rich tonalite sample SG-210c from the Kangidluarsuk fjord: 867 grain #A01 exhibits multiple ages decreasing from the core to the rim, suggesting the spread of zircon 868 ages observed on the Concordia diagram can be explained by ancient Pb-loss. Zircon #A02 is a 869 discordant metamict U-rich grain. d) Granite sample SG-007 from Ukkalek Island: grain #C05 yields 870 an Eoarchean magmatic age and grain #C06 from the same sample is a black U-rich metamict zircon. 871 e) Granite sample SG-080 from the Kangidluarsuk fjord: grain #C15 is a good example of an 872 Eoarchean inherited zircon within an early-Paleoarchean oscillatory zoned crystal. f) Granite sample 873 SG-143: U-rich zircon #B11 shows oscillatory zoning and zircon B12 is a metamict crystal typically 874 found in Neoarchean rocks.

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876 Figure 7: Concordia diagrams of trondhjemite (a-b-c-d), Mg-rich tonalite (e-f-g-h), granodiorite (i) and 877 granite (j-k-l-m-n-o-p-q-r) samples from the SHC. All diagrams show interpreted crystallization ages, 878 inherited grains, and metamorphic secondary populations. Colors for the ellipses reflect the different 879 age populations which are classified from the criteria described in the results (section 2.4); green: 880 inherited zircon; red: primary igneous zircon; orange: secondary igneous zircon; yellow: metamorphic 881 re-crystallized zircon; pink: zircon used for Concordia ages; light blue: Concordia ages. Dashed empty 882 ellipses show altered zircon yielding analyses that cannot be grouped in a consistent population. These 883 analyses were rejected for any age calculation. Ellipses represent  $2\sigma$  uncertainties.

884

Figure 8: Initial  $\epsilon$ Hf values *vs*.<sup>207</sup>Pb/<sup>206</sup>Pb ages for the analyzed zircon grains. Grey diamonds represent individual analyses, and the colored symbols show the average initial  $\epsilon$ Hf values calculated at the interpreted crystallization age of the host rocks. Symbols are as in Figure 2. Note that average  $\epsilon$ Hf values only include relevant analysis for the defined population, such that metamorphic and inherited zircon grains are not included in the average epsilon calculation. A ratio of <sup>176</sup>Lu/<sup>177</sup>Hf = 0.03915 was used for the long-term evolution of the depleted mantle, starting with a chondritic <sup>176</sup>Hf/<sup>177</sup>Hf composition at 4568 Ma. 2 SD errors on the average  $\epsilon$ Hf values and age for the colored symbols are smaller than the symbols.

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894 Figure 9: Cartoon showing how average initial EHf values are calculated in Figure 8, from the interpreted age of crystallization of the rocks, to avoid apparent  $\epsilon Hf vs$ ,  ${}^{207}Pb/{}^{206}Pb$  trends caused by 895 896 ancient Pb-loss. Panel a) shows a fictitious example of a sample with a crystallization age of 3880 Ma, which was affected by ancient Pb-loss and for which most zircon analyses are still relatively 897 898 concordant. Panel b) illustrates that for the same fictitious sample, if initial EHf values are calculated 899 for individual zircon grains at their respective "concordant" ages (vellow circles), it produces a steep eHfvs.<sup>207</sup>Pb/<sup>206</sup>Pb age array consistent with the low Lu/Hf of zircon, rather than representative of the 900 901 evolution of a crustal source. The blue cross symbol shows the average  $\varepsilon$ Hf value for all zircon 902 analyses calculated at the 3880 Ma crystallization age.

903

Figure 10: Proposed major element composition discrimination diagram for all granitoids from theSHC. Symbols and colored fields are as in Figure 2.

906

907 Figure 11: Zr/Nd<sub>N</sub> *vs.* Eu<sub>N</sub>/Eu\* diagram showing the correlation between Eu anomalies and
908 incompatible trace element ratio (Zr/Nd). Eu is normalized to chondrite, Zr and Nd are normalized to
909 the primitive mantle. Normalization values are from McDonough and Sun (1995). Symbols are as in
910 Figure 2.

911

Figure 12: Discrimination ternary diagrams for granitoids proposed by Laurent et al. (2014). a) Major element ternary diagram showing the possible crustal source(s) for the granitoids. The arrow labeled "restite" shows the suggested restitic nature of some samples from the SHC that exhibit pronounced positive Eu anomalies (e.g. SG-007; SG-017; SG-019). b) Ternary diagram showing the petrogenetic processes involved in the formation of Archean granitoids. Contribution of mantle component in the melt is highlighted by the FMSB value =[(FeO<sub>t</sub> + MgO)wt.% x (Sr + Ba)wt.%]. Symbols and colored fields are as in Figure 2.

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920 Figure 13: Kernel density estimate (KDE) and frequency diagrams for zircon grains analyzed for this 921 study. Igneous zircon grains (panel a) and metamorphic zircon grains (panel b) have been 922 discriminated based on CL imagining and Th/U ratios. The top right inset on figure 13b shows a U (ppm) vs.<sup>207</sup>Pb/<sup>206</sup>Pb age diagram for all the Neoarchean zircon grains that illustrates the increase of 923 924 the U concentrations in younger zircon grains. KDE analysis was Performed under "IsoplotR" 925 (Vermeesch, 2018) software using a combination of the Botev et al. (2010) bandwidth selector and the 926 Abramson (1982) adaptive kernel bandwidth modifier. More detail available at: https://cran.r-927 project.org/web/packages/IsoplotR/IsoplotR.pdf.

Figure 14: Average zircon initial  $\varepsilon$ Hf values vs.<sup>207</sup>Pb/<sup>206</sup>Pb age diagram for the SHC rocks. The green 929 930 field shows data for the SW Greenland TTG and detrital zircons from Næraa et al. (2012). Data from 931 Vezinet et al. (2018) shows the average  $\varepsilon$ Hf value for all analyzed zircon grains recalculated at the crystallization age of the host TTG. Data from Morino et al. (2018) shows the initial EHf values from 932 933 their whole-rock Lu-Hf isochrons. Evolution arrays for mafic sources are shown in purple. Evolution array for an Eoarchean felsic source is shown in pink. Evolution array for a Paleoarchean felsic source 934 935 starting from the Lister gneiss sample is shown with a dash line. The KDE from Figure 13 for igneous zircon analyses is shown in blue at the bottom of the diagram. A ratio of  ${}^{176}Lu/{}^{177}Hf = 0.03915$  was 936 used for the long-term evolution of the depleted mantle, starting with a chondritic <sup>176</sup>Hf/<sup>177</sup>Hf 937 938 composition at 4568 Ma. This reference line for the depleted mantle corresponds to a present-day <sup>176</sup>Hf/<sup>177</sup>Hf of 0.2833 consistent with modern high degree melt MORB average value (Salters and 939 940 Stracke, 2004) Symbols for the granitoids are as figure 2, except for trondhjemite sample SG-019 941 shown by a yellow diamond given it equivocal Neoarchean age. 2 SD errors on the average EHf values 942 and age are smaller than the symbols.

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Figure 15: Mantle extraction Hf model ages *vs.* crystallization ages of the SHC granitoids and inherited zircon grains from the SHC granite sample. The vertical bars for each sample represent the variation of model ages using  ${}^{176}Lu/{}^{177}$ Hf ratios of 0.020 and 0.026, with the symbols plotted as the average.

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## 948 **Tables:**

Table 1: Whole-rock major (wt. %) and trace (ppm) element analysis for the SHC granitoids. GPS
coordinates are in UTM NAD 27 zone 20. Major element compositions are recalculated as anhydrous
compositions.

952

Table 2: Summary of geochronological data and initial  $\varepsilon$ Hf-zircon for the SHC granitoids. Age type = method used to calculate the age. n = number of zircon analyses used in age calculation. Age= interpreted crystallization, inherited or metamorphic age.  $\varepsilon$ Hf<sub>(i)</sub> is the average initial values of  $\varepsilon$ Hf of all zircon analyses calculated at the crystallization age of their host rock. Full dataset can be found in the supplementary material Tables S3 and S4.

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Table 3: Key characteristics discriminating the four compositional types of granitoids from the SHC,
based on this study and compositions from the literature (Bridgwater and Collerson, 1976; Collerson,
1979; Schiøtte et al., 1993, 1989a).

962

# 963 Supplementary material:

#### 964 FIGURES:

Figure S1 : Field photographs of different rock types and points of interest (rock hammer head is 20 cm and the sledgehammer is 1 meter long). a) Sampling location for sample SG-210c of Iqaluk tonalitic gneiss dated at  $3869 \pm 6$  Ma (the frame size is about a meter). Same outcrop as the sample dated at  $3920 \pm 49$  by Shimojo et al. (2016). b) Banded grey gneiss of trondhjemite from the Nulliak Island (SG-227). c) Mg-tonalite (SG-265; SG-

969 266) dated at 3230 Ma from Lister Island crosscut by granitic migmatite. d) Crosscutting relationship observed 970 at the White Point location showing Mg-tonalite (melaosome e.g. SG-272) of supposedly Lister in age (3200 971 Ma) crosscut by migmatitic melts (leucosome e.g. SG-271), both crosscut by a mafic dike. e) Typical Iluilik 972 augen granodiorite (SG-203; SG-204) described in the literature as the Uivak II (Bridgwater and Schiøtte, 1991; 973 Hurst et al., 1975). f) Iluilik banded granodiorite that exhibits sheared plagioclase and migmatitic veins. g) 974 Trondhjemite observed on Big Island infiltrated by migmatites (SG-260). h) Similar grey gneiss to that in shown 975 in "g", with higher content of Fe and magnetic minerals (SG-258). i) Migmatitized trondhjemite SG-024 found 976 on Ukkalek Island, only melanosome was analysed. j) Migmatitized Mg-rich tonalite (SG-026) found on 977 Ukkalek Island, only melanosome was analysed. k) Iluilik banded granodiorite that exhibits migmatitic veins. 978 Meter scale veins have been analyzed consisting in the sample SG-208 and SG-209.

979 Figure S2: 15 representative photomicrographs of SHC granitoids. a) cross-polarized photomicrograph of the 980 SG-134 granite and plane-polarized pair in "b". c) cross-polarized photomicrograph of a garnet grain in the SG-981 127 granite and plane-polarized pair in "d". e) cross-polarized photomicrograph of a SG-122 trondhjemite and 982 plane-polarized pair in "f". g- h) Good example of sagenitic texture in SG-024 biotite formed by secondary 983 exsolution of titanium oxides in the crystal lattices seen in plane-polarized light. i) Indicator of alteration and 984 high deformation in the feldspar found in the SG-024 trondhjemitic sample. j) Highly deformed quartz in granite 985 SG-080 from the Kangidluasuk inlet. k) Cross-polarized photomicrograph of a SG-026 showing the presence of 986 clinopyroxene and plane-polarized pair in "l". m-n) Cross-polarized photomicrograph of a SG-027 which 987 similarly showing the presence of clinopyroxene. o) Cross-polarized SG-027 shows sub-grain formation in 988 plagioclase. Otz = quartz, Bio = biotite, Apt = apatite, Grt = garnet, Pl = plagioclase, Ep = epidote, Or =989 Orthose, Zr = zircon, Cpx = clinopyroxene, Amph = amphibole.

Figure S.3: Cathodoluminescence images of all analyzed zircon with U-Pb and Hf analyses laserablation spots.

Figure S.4: Schematic of the outcrop from Shimojo et al. (2016) where the Iqaluk gneiss has been described
and dated at 3920 Ma. The precise location of sample SG-210c dated at 3869 Ma (this study) and the Sample
LAA995 (Shimojo et al., 2016) of Iqaluk gneiss are shown respectively in red and green. The top right inset is a
photograph of the drawn outcrop.

Figure S.5: Sr/Y *vs.* La/Yb diagram for the SHC granitoids with the fields for TTG produced by
variable melting pressures (Moyen and Martin, 2012).

998 TABLES:

Table S.1: Detailed summary of the analytic procedure and conditions for the in-situ U-Pb geochronological analyses and the in-situ Lu-Hf isotopic analyses. Primary and secondary standard reproducibility of U-Pb analysis. The primary standard is the GJ-1 and the secondary is the 91500

1002 Table S.2: Full set of U-Pb analysis on zircon primary standard GJ-1 and secondary standard 91500.  $\rho$  is the 1003 error correlation coefficient.

Table S.3: Full set of U-Pb analysis on zircon from the 18 samples analyzed here. Abbreviations: Osci.=Oscillatory; Inh= Inherited; Ext.= External; Int.= Internal; Meta.= Metamict; Z.= Zoned; Conc = concordance. In the internal structure column when the rim or core is not mentionned, the mentioned characteristic describes the whole grain.  $\rho$  is the error correlation coefficient.

1008 Table S.4: Full data set for the in-situ Lu-Hf isotopic analyses of zircon.

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