

Supplementary materials for “*Metamorphic conditions of the Mesoarchean Amikoq Layered Complex, SW Greenland*”

S1. Detailed methods descriptions

S1.1. Electron microprobe analyses

Polished thin sections were carbon coated and analysed on a JEOL JXA 8200 EMP at the Department of Geosciences and Natural Resource Management, University of Copenhagen. The analyses were carried out with a microprobe setup of 15 kV accelerating voltage, 15 nA electron current and a beam size of 5 μm . Peak and background counting times for Si, Fe, Mg, Al, Ca, Ti, Mn, Ni, Cr and Cl (when analysed) were 20 s and 10 s, respectively, whereas Na and K was 10 s and 5 s, respectively. Internal silicate and oxide standards were run in-between the sessions to monitor analytical accuracy (**Table S1**). An analytical session was dedicated rim-to-rim traverses (20-40 μm steps) across garnets, run with 100 nA electron current to ensure superior data quality with Si, Fe, Mg and Al standardized to an almandine standard (H12-garnet). Each reported mineral analysis comprise an average of 1-3 analyses. Accepted analyses required pyroxene totals within $100 \pm 2\%$, plagioclase totals within $100 \pm 0.5\%$, oxides with <0.1 wt.% SiO_2 , olivine with Si_{apfu} deviating less than 1% from stoichiometric ideal, garnet and sillimanite totals within $100 \pm 1\%$ while amphibole analyses were required not to violate any of the crystallographic considerations outlined in Locock (2014). Owing to frequent vacancies in chlorite and the uncertainty of Fe^{3+} in biotite (e.g., Dymek, 1983; Zane et al., 1998) all analytical data on these minerals was retained, except where other mineral phases obviously resulted in a mixed analysis. Compositional maps were created with setups ranging from 600x600 to 1024x1024 pixels, pixel sizes of 2-5 μm and dwell times between 12-60 ms.

S1.2. Pseudosection modelling

Phase equilibria modelling was undertaken to evaluate the metamorphic conditions experienced by the ALC. Phase relations were modelled in the $\text{Na}_2\text{O}-\text{CaO}-\text{K}_2\text{O}-\text{FeO}-\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}-\text{TiO}_2-\text{Fe}_2\text{O}_3$ (NCKFMASHTO) system using an updated version of THERIAK-DOMINO v. 01.08.09 (de Capitani and Brown, 1987), and an updated version of the Holland and Powell (2011) dataset (ds62, February 2012). Activity models for solid solution minerals used in the calculations are Green et al. (2016) clinopyroxene, amphibole, and melt models; White et al. (2014) orthopyroxene, biotite, chlorite, and garnet models; Holland and Powell (2011) olivine and epidote models; Holland and Powell (2003) plagioclase model; White et al. (2002) magnetite model; and the White et al. (2000) ilmenite model. Files for phase equilibria calculations are the same as those used in Jørgensen et al. (2019). An estimate for bulk rock H_2O was made on the premise that no free H_2O is present in the suprasolidus as it partitions into the melt (e.g., Clemens and Vielzeuf, 1987). Temperature-composition ($T-\text{M}_{\text{H}_2\text{O}}$) diagrams were constructed at 3, 5, 7, and 10 kbar to constrain a reasonable amount of H_2O to ensure no excess fluid in the suprasolidus. Bulk composition Fe is allocated as 80% Fe^{2+} and 20% Fe^{3+} .

S1.3. SHRIMP U-Pb zircon geochronology

A ~4 mm thick slice of the zircon obtained from drill core FF-DDH-06-39 (188.7 m) was cast in epoxy along with grains the U-Pb calibration standard FC1 and polished. Zircons from gabbroic and noritic samples FOS2 and QUA, respectively, were separated using standard density and magnetic susceptibility techniques. Zircon concentrates were handpicked using a binocular microscope and were mounted onto double-sided sticky tape along with grains of the U-Pb calibration standard Temora and then cast into an epoxy resin disc. After the epoxy had cured, the disc was ground to reveal median sections through the grains and then polished with 1 μm diamond paste. Samples were documented by cathodoluminescence (CL) imaging prior to analysis. Analysis of the large zircon from drill core FF-DDH-06-39 was undertaken on the Beijing SHRIMP 2 #1 instrument at the Chinese Academy of Geological Sciences. Analysis of the FOS2 and QUA zircons was undertaken on the SHRIMP-RG at the Australian National University (ANU). In both cases, the analytical protocol followed that of Williams (1998). Data reduction was undertaken by the Mac (pre-OS10) ANU software PRAWN and LEAD. The reduced and calibrated data were assessed and plotted using the Excel™ plug-in ISOPLOT (Ludwig, 2003). All mean ages given are at the 95% confidence level.

S2. Results

Detailed petrography and additional mineral composition descriptions are available as Supplementary Material. Additional petrographic photos and chemical maps are available as Supplementary Figures (**Figs. S4-13**). All mineral data is available as Supplementary Material (**Tables S2-11**). Zircon geochronology results are provided in **Table S12**.

S2.1. Petrography

Amikoq rocks display a wide range of textures from igneous to metamorphic. Because all thin sections display metamorphic features to some extent, the *meta*-prefix is assumed throughout this work. While granoblastic textures can develop in cumulates as the system approaches ideal mineral surface energies in a high temperature environment (Hunter, 1996; Wager et al., 1960), much of the granoblastic textures observed at Amikoq are likely metamorphic given the Meso-Neoproterozoic polymetamorphic history of the Akia terrane (**Section 2**). We utilize the nomenclature of Le Maitre (2002) to describe the current mineralogy of ALC lithologies while that of the SCMR (Fettes and Desmons, 2011) is used for the host rocks. Retrograde mineralogy is largely ignored in the nomenclature adopted here, e.g., extensively retrogressed norites are simply termed *retrogressed norites*.

S2.1.1. Host rocks

S2.1.1.1. *Foliated plagioclase-spinel-bearing ilmenite-biotite-sillimanite-garnet-cordierite gneiss (sil-grt-crd gneiss; 191142): Marginal floor gneiss*

The sil-grt-crd gneiss, in places forms the boundary between the ALC and TTG gneisses and has been suggested to represent a reaction zone (Armitage, 2009) similar to what is observed at the nearby Ulamertoq ultramafic complex (Guotana et al., 2018). A sedimentary origin was proposed by

Garde (1997), but the term metavolcanoclastic is preferred here (**Section 5.1.1**). Alternatively, such rocks may represent metasomatic products with reaction between felsic orthogneiss and norite or peridotite.

The complex mineralogy consists of cordierite (~45%) + porphyroblastic garnet (~20%) + sillimanite (~10%) + biotite (~10%) + ilmenite (~5%) + plagioclase (<5%) + spinel (*s.l.* ~1-2%) + quartz (~1-2%) and accessories includes Fe-sulfides (likely pyrite), zircon and REE-rich apatite.

S2.1.1.2. Metavolcanic-subvolcanic host rocks

The host rocks are usually plagioclase + amphibole ± clinopyroxene ± orthopyroxene ± biotite ± garnet ± quartz metabasites with occasional intermediate to felsic compositions (i.e., up to ~64 wt.% SiO₂; Kristensen, 2006). A marginal quartz-rich host rock of this study (191143) may represent a meta-andesite/dacite or silicified amphibolite. Where unspecified, the host rocks (excluding sil-grt-crd gneiss 191142) will simply be referred to as amphibolites, so as not to break with convention (e.g., Garde, 1997) or metabasites, although two-pyroxene-amphibole mafic granulite commonly would be the more correct term (e.g., 193500).

S2.1.1.3. K-hastingsite-bearing clinopyroxene-magnetite-ilmenite-orthopyroxene-plagioclase-quartz gneiss (K-hastingsite-bearing gneiss; 191143): Marginal “floor” gneiss

Located immediately above the sil-grt-crd gneiss, a quarzo-feldspathic amphibole-orthopyroxene-spinel (*s.l.*)-ilmenite-clinopyroxene gneiss occurs (**Fig. S5**), and is abbreviated K-hastingsite-bearing gneiss henceforth. This quartz (~40%) and plagioclase (~30-40%) dominated rock displays an inequigranular granoblastic texture with some quartz porphyroblasts (up to 5 mm) while quartz, plagioclase, oxide (~10-20%), orthopyroxene (~5-10%), clinopyroxene (~5%) and amphibole (~1-5%) matrix minerals typically are <1.0 mm. Accessory minerals include rare biotite associated with oxide, zircon and hexagonal to rounded Cl-apatite.

S2.1.1.4. Gneissic ilmenite-clinopyroxene-orthopyroxene-amphibole mafic granulite (193500): Intrusion “roof” mafic granulite

The main mineral constituents are plagioclase (~40%), amphibole (~30%), orthopyroxene (~10-15%) and clinopyroxene (~10-15%) and subordinate oxides (<10%), all usually 0.1-2 mm in size (**Fig. S6**). Accessory minerals comprise zircon and rod-shaped euhedral apatite without detectable Cl-spikes commonly as inclusions in plagioclase and amphibole and occasionally in pyroxenes (**Fig. S6f**).

S2.1.2. Amikoq Complex rocks

S2.1.2.1. Olivine-rich and orthopyroxene-rich rocks (191147, 191150 and 181584): Ultramafic cumulates

The ultramafic sample 191147 dominated by olivine is an amphibole-bearing, spinel (*s.l.*) dunite/harzburgite and sample 191150 is a spinel-bearing (*s.l.*) olivine hornblende orthopyroxenite, forming the intermediary link between the peridotitic/dunite bodies and orthopyroxenite cumulate of 181584 (**Fig. S7-8**).

Optically homogeneous, inequigranular and granoblastic olivine (30 to >90%) and orthopyroxene (<10% to 60%) dominate, followed by amphibole (up to 30%). Dark green to opaque spinel (*s.l.*

<10%) and fringed chlorite (<5%) are minor phases. Some Cr-magnetite grains enveloped in chlorite contain relict spinel (s.s.) cores (**Figs. 4d, S7-8**).

The orthopyroxene cumulate comprise hornblende orthopyroxenite (181584) dominated by orthopyroxene (~80-95%) with subordinate green Ca-amphibole (~5-20%) and accessories of chromite to Cr-rich hercynite (<1%) and apatite (**Fig. S9**). Minor metamorphic overprint consist of (Fe,Mg,Mn)-amphibole.

S2.1.2.2 Orthopyroxene-rich rocks (191146 and 191234): Orthopyroxene cumulates

These rocks are hornblende melanorites (191146 and 191234, abbreviated melanorites henceforth; **Figs. 2, S1**). They are the least metamorphosed rocks in this study, dominated by orthopyroxene (~70-90%), subordinate amphibole (~5-20%) and plagioclase (~5-15%; **Figs. 4e-f, S10**). Accessories comprise rare small biotite laths, minor apatite, minor Fe-Ni- and Fe-sulfides. A minor metamorphic overprint consists of chlorite and (Fe,Mg,Mn)-amphibole. Sample 191234 shows euhedral-anhedral coarse orthopyroxene crystals (up to 2 cm), representing characteristic magmatic textures along with interstitial plagioclase showing preserved zonation patterns and localized poikilitic textures. This yields an orthopyroxene (~80%) mesocumulate with intercumulus poikilitic and occasionally zoned plagioclase (~10%) and anhedral-euhedral amphibole (~10%; **Figs. 4e-f, S10**). Orthopyroxene frequently contain small green subhedral-euhedral amphibole inclusions and amphibole + anhedral plagioclase. Cumulate textures are not well preserved in sample 191146 (**Fig. S10**).

S2.1.2.3 Orthopyroxene- and plagioclase-rich rocks (181760, 181785 and 181791): Mafic noritic cumulates

Mafic samples from ALC have modal compositions of hornblende leuco- to melanorites. They are principally composed of granoblastic plagioclase (~30-70%), orthopyroxene (~30-50%) and amphibole (~5-45%) with accessory biotite, rare ilmenite, sulphides (likely pyrite and pentlandite) and apatite. Magnetite is significant only in retrogressed samples along with chlorite (**Figs. 4g-h, S11-12**). The ALC mafic lithologies are generally devoid of clinopyroxene in contrast to the hosting amphibolites.

The fabric is characterized by pinkish orthopyroxene-stringers in parallel layers (**Fig. 5g, S11**) intercalated with plagioclase-rich layers similar to what is observed in some supracrustal amphibolites (compare **Fig. S6** with **S11**). Small euhedral amphibole inclusions are common both in plagioclase and orthopyroxene (**Fig. 4g**). Ilmenite rarely occurs as inclusions in orthopyroxene and amphibole in the non-retrogressed sample. Samples 181785 and 181791 are intensely retrogressed with near-complete replacement of orthopyroxene by colourless to stained (Fe,Mg,Mn)-amphibole (cummingtonite), magnetite, pale-green high-Al Ca-amphibole and locally chlorite (**Figs. 4h, S12**), while green Ca-amphibole appears to have been stable, indicating a hydration event that resulted in a stable mineral assemblage consisting of amphiboles + plagioclase + magnetite + chlorite. One hornblende composition (the green Ca-amphibole) equilibrated with a higher temperature assemblage; a different hornblende composition + (Fe-Mg-Mn) cummingtonite was the result of hydration and replacement of orthopyroxene. Please note that we use the term 'stained amphibole' for the above-mentioned 'patchy' nature of the retrogressive Fe-Mg-Mn amphibole that has this characteristic appearance.

S2.1.2.4 Hornblende melagabbro (181600): Amphibole cumulate?

This sample stem from a minor occurrence at South Margin (**Fig. S1**). Dominant granoblastic green Ca-amphibole (~80-90%) tend to be larger (usually 1-2 mm) than subordinate plagioclase (~10-20%; <1 mm) (**Figs. 4c, S13**). Accessory minerals comprise oxides, decomposed Fe-Ni sulfides and relatively large apatite crystals with detectable Cl-spikes commonly hosted in amphibole but also in plagioclase. Pyroxenes are not observed. Replacement of green Ca-amphibole ± plagioclase to colourless-stained fine-grained fibrous nematoblastic aggregates of (Fe,Mg,Mn)-amphibole + pale-green high-Al Ca-amphibole + chlorite ± quartz (**Figs. 4c, S13**) is significant locally, indicative of retrograde conditions.

S2.2. Mineral Chemistry

S2.2.1. Olivine and pyroxenes

Pyroxene endmember calculations follow Lindsley (1983) in **Figure 6**; whereas, orthopyroxene endmember calculations in **Figure S15** follow Morimoto (1988). No obvious variation was detected in core and rim compositions of olivine and pyroxenes.

Olivine and orthopyroxene in ultramafic dunite cores from West Margin are less magnesian ($Fe_{82.2-82.8}$, $En_{81.8-82.6}$) than the adjacent olivine-orthopyroxenite ($Fe_{84.0-84.9}$, $En_{83.1-84.1}$; **Figs. 5, S14-15**); whereas, olivine NiO wt.% ranges 0.21-0.28 and 0.21-0.30, respectively. Orthopyroxene from orthopyroxenites are more magnesian ($En_{84.1-84.9}$) than orthopyroxene in melanorites. Orthopyroxene of melanorite 191146 is more Fe-rich ($En_{69.9-71.0}$) relative to melanorite 191234 ($En_{81.2-81.9}$). The mafic norites of West Margin have more Fe-rich orthopyroxene ($En_{62.0-67.5}$; **Figs. 6, S15**). All of these ALC orthopyroxenes are generally more magnesian than host metabasites ($\leq En_{65}$; **Fig. S15**).

The marginal K-hastingsite-bearing gneiss, sandwiched between noritic rocks and sil-grt-crd gneiss at the West Margin (**Fig. 2b**), have Fe-rich orthopyroxene ($En_{46.8-49.1}$) and diopside ($Wo_{44.3-46.3}En_{34.4-39.1}Fs_{15.6-19.3}$; **Fig. S15**). The most Fe-rich pyroxenes are found in the roof mafic granulite with Fe-rich orthopyroxene ($En_{41.5-42.8}$) and diopside ($Wo_{45.4-47.5}En_{30.5-32.8}Fs_{20.6-22.9}$). The abundant exsolution lamellae observed in clinopyroxene of the roof mafic granulite are Fe-rich pigeonite ($Wo_{8.1-9.9}En_{37.5-38.6}Fs_{53.7-56.0}$; **Figs. 6, S15**).

S2.2.3. Plagioclase

Plagioclase in melanorite 191146 tend to be homogenous and bytownitic ($An_{83.3-85.8}$; **Fig. 7**). Melanorite 191234 have complex relict zoned intercumulus plagioclase where reverse zonation have slightly more calcic centres ($An_{80.4-84.9}$) relative to the margins ($An_{74.8-77.0}$) and vice versa for normal zonation ($An_{69.3-71.2}$ and $An_{74.5}$, respectively). Rarely, labradoritic compositions in zoned plagioclase ($An_{52.9-57.9}$) in intensely fractured domains of the thin section is encountered, indicating that the most sodic compositions are secondary in origin. The slight variation between centres and margins seen elsewhere are deemed relict.

Norites have homogenous plagioclase on a sample scale and a restricted compositional range ($An_{83.1-88.5}$) though quartz-plagioclase-chlorite veins have andesine compositions ($An_{31.2}$; **Fig. 7**). Plagioclase from the hornblende melagabbro is bytownitic ($An_{81.7-83.0}$) with one centre that was anomalously anorthositic ($An_{91.8}$).

Plagioclase in the marginal K-hastingsite-bearing gneiss is homogeneous and displays intermediate compositions ($An_{45.5-46.8}$), as well as elevated K_2O contents ($Or_{1.2-2.6}$). The sil-grt-crd

gneiss and roof mafic granulite are less potassic ($Or_{0-0.7}$ and $Or_{0.9-1.5}$, respectively) than the K-hastingsite-bearing gneiss in spite of their more sodic plagioclase compositions ($An_{36.7-40.0}$ and $An_{39.6-41.2}$, respectively; **Fig. 7**).

S2.2.4. Amphibole

The Amikoq amphibole is divided into four types: i) green Ca-amphibole, ii) pale-green high-Al Ca-amphibole, iii) colourless-stained amphibole (Fe,Mg,Mn-amphibole), iv) brownish Ca-amphibole (K-hastingsite). Classification is according to Hawthorne et al. (2012) and has been calculated using the spreadsheet of Locock (2014). Distinction between monoclinic and orthorhombic (Fe,Mg,Mn)-amphibole were done according to ^{42}Ca content that tend to be >0.15 apfu in monoclinic variants (Schumacher, 2007). They are presented in **Figure 8**, modified after Wones and Gilbert (1982) to include Fe^{3+} , to better illustrate hastingsite compositions, while **Figure S16** is according to Hawthorne et al. (2012). These amphiboles are collectively referred to as 'hornblende' in the following.

S2.2.4.1. Green Ca-amphibole: Igneous amphibole, recrystallites and metamorphic amphibole

Plausible igneous amphibole is exclusively of the green Ca-amphibole variety. Although this is present throughout the Amikoq ultramafic-mafic complex with hues ranging from light green (olivine-rocks) to dark green (host metabasite). Textural evidence suggestive of a magmatic origin is only preserved in some noritic lithologies. Thus, it is possible that some green Ca-amphibole from the ultramafic bodies or noritic lithologies is of metamorphic origin, or recrystallized locally. Mg-rich green Ca-amphibole (Mg#: 0.90-0.97) is the principal amphibole type in dunite/harzburgite, olivine-orthopyroxenite, orthopyroxenite and melanorite 191234, and classifies as magnesio-hornblende and magnesio-ferri-hornblende (ferri: $^{56}Fe^{3+} > ^{27}Al, ^{52}Cr, ^{55}Mn^{3+}$). Pargasite and magnesio-hastingsite are exclusive to melanorite 191146, coinciding with slightly lower amphibole Mg# (0.81-0.84) (**Figs. 8, S16**). Sodium and K are highly variable resulting in variable $\sum^{Na+K+2Ca}$ that range from 0.14-0.18 apfu in orthopyroxenite, 0.08-0.17 apfu in intimately associated dunite and olivine-orthopyroxenite and 0.46-0.55 apfu in melanorite 191146 (**Figs. 8, S16**). Melanorite 191234 have intermediate values ranging from 0.32-0.45. Cr_2O_3 is elevated in orthopyroxenite green Ca-amphibole (1.17-1.67 wt.%) relative to green Ca-amphibole in related olivine-rich rocks (0.38-0.69 wt.%), but similar to melanorite 191234 (1.04-1.42 wt.%). Green Ca-amphibole in melanorite 191146 have lower Cr_2O_3 of 0.68-1.05 wt.% (**Fig. S16b**). All other green Ca-amphibole analyses yield <0.3 wt.% Cr_2O_3 .

Green Ca-amphibole in norite rocks classifies as magnesio-ferri-hornblende, ferri-tschemmakite, magnesio-hastingsite and ferri-sadanagaite reflecting the higher calculated $^{56}Fe^{3+}$ in these amphibole (**Fig. 8**). Despite similar Mg# between norite samples (0.76-0.90, avg. 0.81), the alkali and Ti content vary markedly from sample to sample (albeit is similar intra-sample). The resulting variation in $\sum^{Na+K+2Ca}$ that range from 0.18-0.51 apfu and $\sum^{Al+Fe^{3+}+2Ti}$ from 1.48-1.90 apfu (**Figs. 8, S16**) reflecting metamorphic retrogression as discussed in **Section 5.1.2**.

The hornblende melagabbro is principally composed of green Ca-amphibole classifying as magnesio-hornblende and magnesio-ferri-hornblende with similar Mg# (0.82-0.92) to green Ca-amphibole in the norites. Potassium is generally low yielding 0.14-0.28 $\sum^{Na+K+2Ca}$ (**Figs. 8, S16**), whereas TiO_2 is moderate (0.58-0.81 wt.%) and more comparable to the melanorites and ultramafics (0.46-0.99 wt.%) than norite rocks of West Margin (0.75-1.81 wt. %).

The last green Ca-amphibole is dark green to weakly brownish and hosted in the roof mafic granulite. It has a Mg# ranging 0.51-0.57, elevated K₂O from 0.70-0.81 wt.% (other green Ca-amphibole: K₂O <0.61 wt.%), TiO₂ from 1.2-2.0 wt.% and low Cr₂O₃ from 0.02-0.11 wt.% (**Figs. 8, S16**), classifying as magnesio-ferri-hornblende and magnesio-hastingsite. This green Ca-amphibole type is metamorphic.

S2.2.4.2. Pale-green high-Al Ca-amphibole: Metamorphic

This amphibole type grows at amphibole-plagioclase or orthopyroxene-plagioclase boundaries, and typically has low SiO₂ (40.7-42.1 wt.%) and TiO₂ (0.17-0.45 wt.%) coupled with high Al₂O₃ content (17.3-20.6 wt.%) and consequently classify as sadanagaite (0.4 wt.% CaO; **Fig. 8**). These amphiboles are collectively referred to as 'secondary hornblende'.

S2.2.4.3. Colorless-stained amphibole ((Fe,Mg,Mn)-amphibole): Metamorphic

The (Fe,Mg,Mn)-amphiboles are typical breakdown products of orthopyroxene and have compositions of cummingtonite. Cr₂O₃ range from <0.08 wt.% in retrogressed norite and hornblende melagabbro to 0.19-0.80 wt.% in ultramafic samples (**Fig. S16b**).

S2.2.4.4. Brownish Ca-amphibole (Cl-bearing K-hastingsite): Metasomatic

Exclusive to marginal rocks and rocks interpreted as extensively 'tectonised' norites (Armitage, 2009) this Fe-rich (Mg#: 0.45-0.54) amphibole-species is SiO₂-undersaturated (39.4-40.1 wt.%) with a high content of K₂O and TiO₂ (1.4-2.4 and 2.0-2.4 wt.%, respectively; **Figs. 8, S16**) resulting in hastingsite, K-hastingsite (potassic: ^AK > ^ANa, ^ACa, ^A□) and potassic-magnesio-hastingsite. It is the only amphibole-species in this study that contains significant Cl (0.98-1.27 wt.%, all other: <0.09 wt.%; **Table S5**).

S2.2.5 Oxides

Classification of spinels follows the procedure of Bosi et al. (2018) with Fe³⁺ calculated according to Droop (1987). Chromite (s.s.) is only observed in a West Margin orthopyroxenite (Mg#: 0.39, Cr#: 0.50) and consists of a few rare grains that compositionally extend into hercynite compositions (Mg#: 0.26-0.38, Cr#: 0.43-0.48). The closely associated dunite and olivine-orthopyroxenite of the West Margin contain Al-spinel (spinel, s.s.) with lower Cr-content (Cr#: 0.11-0.17 and 0.15-0.18, respectively) and higher Mg# (0.52-0.54 and 0.52-0.57, respectively; **Fig. 9a**). Magnetite in the dunite and olivine-orthopyroxenite have elevated Cr₂O₃ (4.4-10.7 and 10.9-12.56 wt.%, respectively). One exception is a single magnetite inclusion in olivine from the olivine-orthopyroxenite with 0.2 wt.% Cr₂O₃. Hercynite in the dunite and olivine-orthopyroxenite range in Mg# from 0.43-0.48 and in Cr# from 0.19-0.23 and is compositionally intermediate between spinel (s.s.) and magnetite. One spinel from an olivine-orthopyroxenite features an ilmenite rim (**Table S7**).

Retrogressed norites host near pure end-member magnetite with negligible amounts of Cr₂O₃ (<0.08 wt.%) along with relatively pure ilmenite (**Fig. 9b**); oxides were not encountered in the non-retrogressed norite. The opaque phase of the hornblende melagabbro at the South Margin are usually enveloped or intensely infiltrated by quartz, explaining the high SiO₂ (1.9-3.6 wt.%) of these analyses (**Table S6**). Along with high FeO^{tot} (74-79 wt.%) and NiO (0.1-5.9 wt.%), this suggest that the oxides (no detectable S) represent decomposed Fe- and FeNi-sulfides.

Spinel in marginal K-hastingsite-bearing gneiss are near pure magnetite with minor TiO₂ (0.09-2.98 wt.%) that co-exist with ilmenite (**Fig. 9b**). Low Cr₂O₃ (<0.03 wt.%) is distinctly different from

the high Cr₂O₃-content in hercynite and magnetite (7.2-11.9 and 5.6-5.7 wt.%, respectively) of the closely associated sil-grt-crd gneiss. One rare magnetite grain from the roof mafic granulite had slightly elevated Cr₂O₃ and TiO₂ (1.16 and 7.2 wt.%, respectively), a Cr-content not shared by co-existing ilmenite (<0.06 wt.%). All ilmenite are relatively pure with only a minor hematite component (0.00-0.06; **Fig. 9b**).

S2.2.6. Biotite, garnet and cordierite

Biotites are Mg-rich with Mg ranging 3.0 to 5.0 apfu and (Fe_{tot} + Mn + Ti – Al^{VI}) ranging 0.05 to 1.59 apfu, consequently classifying as Mg-biotite to phlogopite according to the classification scheme of Tischendorf et al. (1997). Biotite with the highest Mg# is observed in melanorite 191234 (0.88-0.95). The biotite Mg# in norite samples (0.75-0.86) largely overlaps with values from garnet-hosted biotite inclusions in sil-grt-crd gneiss (0.75-0.89). Biotite in the matrix or close to garnet rims of the sil-grt-crd gneiss yield the lowest Mg# (0.64-0.75). TiO₂ is highly variable with 1.0-2.4 wt.% in melanorite 191234, 1.0-3.3 wt.% in norite samples and 1.6-2.4 wt.% in the sil-grt-crd gneiss though one biotite-inclusion in garnet have anomalously low TiO₂ of 0.1 wt.%.

Garnets are Ca-poor, Al-rich almandines with insignificant MnO (<1 wt.%). The cores of large garnet grains show systematic core to rim variation with core compositions of Pyr_{0.34}Alm_{0.62}Grs_{0.04} to rim compositions of Pyr_{0.22}Alm_{0.74}Grs_{0.04}; whereas, smaller garnet grains do not record the more Mg-rich part of the large garnet cores (**Fig. S17**). Cordierite of the sil-grt-crd gneiss may have slightly lower Mg# cores (0.73) relative to rims (0.75-0.76). Cordierite from a fracture in an orthopyroxenite has high Mg# (0.93).

S2.3. Zircon petrography and geochronology

The zircon megacryst (~5 mm), obtained from a felsic patch hosted in an orthopyroxene-rich rock, displays fine-scale oscillatory zoning typical of igneous crystallization (position marked as AM1 in **Fig. 1**). Zoning is most strongly developed around the fringes of the crystal and around interior lobate inclusions occupied by quartz + plagioclase + biotite ± amphibole consistent with crystallized melt inclusions (**Fig. S3**). This suggests zircon grew coevally with a silicate liquid. All zircon analyses yielded ²⁰⁶Pb/²³⁸U and ²⁰⁷Pb/²⁰⁶Pb ages indistinguishable within error from each other (**Fig. S3b**). The weighted mean ²⁰⁷Pb/²⁰⁶Pb age is 3004 ± 9 Ma (MSWD = 0.55), interpreted as the time of growth of the zircon coeval with a silica-saturated silicate melt.

The zircons separated from FOS2 and QUA (see positions in **Fig. 1**), a gabbroic and noritic rock from north of Amikoq, respectively, are of similar morphology and are described together (**Fig. S3d**). The grains are typically 150-200 µm long, and range from equant/oval, to prismatic or euhedral. The grains commonly display oscillatory zoning, but this is commonly disrupted by recrystallization domains, and for many grains it is truncated at the grain boundaries. Some grains display complex sector zoning. Eleven analyses were undertaken on 11 FOS2 zircons and 13 analyses on 13 QUA zircons (**Table S12; Fig. S3d-h**). All FOS2 analyses yield U-Pb ages indistinguishable from concordant, with minimal or undetectable common Pb (based on measured ²⁰⁴Pb). All analyses yield a weighed mean ²⁰⁷Pb/²⁰⁶Pb age of 3008 ± 7 Ma (MSWD = 1.3), i.e., distribution beyond that expected from analytical error alone. A frequency distribution plot (**Fig. S3f**) shows this is due to a bimodal age distribution, rather than high kurtosis in a single age population. Eight of the analyses are in the older age group, and are sites with better-preserved oscillatory zoning. These yield a weighted mean ²⁰⁷Pb/²⁰⁶Pb age of 3013 ± 6 Ma (MSWD = 0.43) and are interpreted as giving an age of magmatic zircon crystallization. The three analyses (#7.1, #9.1 and #10.1) with younger apparent

$^{207}\text{Pb}/^{206}\text{Pb}$ ages are on recrystallization domains on the zircons and yield a weighted mean $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2995 ± 10 Ma (MSWD = 0.19) and are interpreted as giving an age of (high temperature) zircon recrystallization. All QUA analyses yield U-Pb ages indistinguishable from concordant, with minimal or undetectable common Pb (based on measured ^{204}Pb). All analyses yield a weighed mean $^{207}\text{Pb}/^{206}\text{Pb}$ age of 3008 ± 5 Ma (MSWD = 0.31). Although all analyses have $^{207}\text{Pb}/^{206}\text{Pb}$ ages indistinguishable from each other, a frequency distribution plot (**Fig. S3h**) shows the binned ages are not distributed equally around the mean. Two of the analyses (#4.1 and #7.1) with younger apparent $^{207}\text{Pb}/^{206}\text{Pb}$ ages are on recrystallization domains and yield a weighted mean $^{207}\text{Pb}/^{206}\text{Pb}$ age of 2999 ± 14 Ma (MSWD <0.01) and are interpreted as giving the age of (high temperature) zircon recrystallization. The remaining 11 analyses display better-preserved oscillatory zoning and yield a weighted mean $^{207}\text{Pb}/^{206}\text{Pb}$ age of 3009 ± 6 Ma (MSWD = 0.22) and are interpreted as giving an age of magmatic zircon crystallization. Pooling the data from FOS2 and QUA into groups with better-preserved oscillatory zoning and those displaying recrystallization yield weighted mean $^{207}\text{Pb}/^{206}\text{Pb}$ ages of 3011 ± 4 Ma (MSWD = 0.34) for likely magmatic zircon and 2997 ± 8 Ma (MSWD = 0.15) for recrystallized zircon.

S3. Discussion of K-CI fluid metasomatism

3.1. Cl-bearing K-hastingsite in marginal silicified amphibolite gneiss: Evidence for K-CI fluid metasomatism

Hastingsitic Ca-amphibole (silica-poor and Fe^{3+} -rich; Hawthorne et al., 2012) of igneous origin tends to be hosted in evolved, silica-saturated to silica-undersaturated intrusive rocks of nepheline-syenite, granite, quartz-bearing syenite, calc-alkaline syenites, granodiorites and their metamorphic equivalents, and appear to have restricted sodic ranges in $\text{K}/(\text{K}+\text{Na})$ of 0.25 to 0.46 (Holm, 1971; Thomas, 1982). Chlorine-bearing metamorphic K-hastingsite, however, is usually associated with metasomatic skarn-type rocks, such as calc-silicate- and sphalerite skarns (1.97-3.69 wt.% Cl; Dick and Robinson, 1979; Shiraishi et al., 1994; Suwa et al., 1987; Thomas, 1982), hydrothermal fluid alteration of gabbroic to granitic assemblages (0.71-3.07 wt.% Cl; Bonin and Tatu, 2016) and rocks in geothermal oceanic systems (0.93-2.65 wt.% Cl), as well as submarine metabasites (up to 6.5 wt.% Cl; cf. Enami et al., 1992). A common trait is elevated alkali content and especially elevated K with $\text{K}/(\text{K}+\text{Na}) > 0.5$ where amphibole alkali content, i.e. edenite substitution, tend to correlate positively with Cl-content (Enami et al., 1992). Amikoq Cl-bearing K-hastingsites (0.98-1.27 wt.% Cl) appear exclusive to marginal host rocks that, based on the high quartz and plagioclase content (~40% each; **Section 4.1.1.2.**), either may be considered of meta-andesitic origin or as a silicified version of a host 'amphibolite'. Kristensen (2006) analysed amphibole of K-hastingsite composition (Cl was not measured) from a rock layer occurring in the noritic succession at the West Margin mapped as 'tectonised leuconorite' (**Fig. 2b**); however, a sample from this tectonised zone have major- and trace-elements indistinguishable from the amphibolites. This indicates that the 'tectonised leuconorite' is more akin to the marginal silicified amphibolite with Cl-bearing K-hastingsites than the norites and could either have been tectonically imbricated into the noritic succession or represent a trapped sliver of the country rock into which the ALC intruded. Amikoq K-hastingsites range from 0.49-0.63 $\text{K}/(\text{K}+\text{Na})$ (barring one hastingsite outlier with 0.32) combined with high edenite-component (0.67-0.77 apfu; **Figs. 8, 12**). They are unlikely to be igneous in origin as Cl appears not to partition into amphibole in magmatic systems (Thomas, 1982), implying a metasomatic origin. Apatite with significant Cl-spikes (qualitative WDS-screening), have also been observed in the Cl-

bearing K-hastingsite gneiss and a link is probable. The Cl-bearing K-hastingsites bear all the geochemical hallmarks of a high temperature origin (see **Sections 5.2.1, 5.2.3** and **Figs. 12, 13**) corroborated by plagioclase-hornblende temperatures of 795-846°C. Amphibole, biotite and minor apatite encountered in the ALC lithologies do not show elevated Cl-content, nor the REE-rich apatite in the sil-grt-crd gneiss and the abundant apatite of the roof mafic granulite. This suggests either that the K-Cl metasomatic event predated the intrusion or that the fluid was of magmatic origin. Cl-spikes in apatite was encountered in the hornblende melagabbro at the South Margin, although amphibole analysed immediately adjacent to Cl-bearing apatite (physically touching) were devoid of Cl (<0.01 wt.%). The hornblende melagabbro layer of the South Margin is in itself controversial; bulk rock major- and trace-elements of other rocks from the layer are distinctly of the host amphibolitic variety (unpublished data); whereas, the green Ca-amphibole and plagioclase of the investigated thin section have primitive compositions ($Mg\#_{Green\ amp}: 0.82-0.92$, $An_{81.7-83.0}$) unlike typical host amphibolitic compositions ($Mg\#_{Green\ amp}: 0.45-0.58$, $An_{39.6-46.8}$; **Figs. 7, 12**). Obtained hornblende melagabbro plagioclase-hornblende temperatures (875-924°C) are most easily related to the ALC, possibly indicating that this sample represent an amphibole cumulate. Decomposed Fe-Ni sulphides may support this notion (**Section 4.2.5**). If cumulative in origin, the Cl-spike in apatite provide evidence that the ALC was capable of producing Cl-bearing fluids during differentiation since Cl preferentially partitions in apatite over biotite and amphibole (Nijland et al., 1993). Cl-rich apatites have also been observed in the Bushveld and Stillwater Complexes (Boudreau et al., 1986; Hanley et al., 2008; Schiffries, 1982). The occurrence of Cl-fluids in the ALC could have important implications for the petrogenesis of pegmatoidal parts of the complex and the occurrence of PGE reefs (Armitage, 2010) as inferred for Stillwater (Boudreau et al., 1986; Hanley et al., 2008).

At least two alternative K-Cl fluid origins can be envisaged. 1) Prograde dehydration (and melting?) of the adjacent sil-grt-crd gneiss, which provide cordierite-garnet temperatures (~820°C) indistinguishable from the K-hastingsite-bearing gneiss. Melting of the sil-grt-crd gneiss may be indicated by the absence of K-feldspar, possibly consumed during melting if the rock melted at the granite minimum. Although no clear evidence exists for this scenario (e.g. textural or Cl-spike in apatite), it is difficult to exclude a prograde origin for the Cl-K fluid at present. 2) K-Cl fluid derived from host TTG intrusion (Nishio et al., 2019). This option is unattractive given the occurrence of K-hastingsite-bearing lithologies hosted within the ALC; had the TTGs supplied the K-Cl fluid the ALC should have been affected as well (assuming similar permeability), which is not apparent.

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