

## **Supplementary Materials – AFC modelling for:**

### **”The Mesoarchean Amikoq Layered Complex of SW Greenland: Part 2. Geochemical evidence for high-Mg noritic plutonism through crustal assimilation”**

#### **The Amikoq parental melt: Constraints from trace element modelling**

In the following, we provide a detailed description of the trace element systematics of the ALC, as well as of the assimilation fractional crystallisation (AFC) modelling that we carried out to assess the petrogenesis of the ALC, as well as its relation to the hosting amphibolites and potential contaminants.

#### **Cumulate modelling**

It is important to test whether the ALC trace element characteristics could be caused by cumulate processes, e.g. the U-shaped pattern could in principle be owed to a relatively pure cumulate where the HREE-enrichment is caused by the preference for ferromagnesian minerals to incorporate the HREE (e.g. Kennedy et al., 1993; Stracke et al., 2003) while the moderate LREE enrichment could be caused by small amounts of interstitial LREE-enriched trapped melt. We begin this investigation in simple end-member cumulate models where three assumed melts are, boninitic, komatiitic and contaminated komatiite (SHMB) (AN-1, high-Ti ultramafic sample 193203 and Mt. Hunt sample 331/626 of Sun et al. (1989), respectively), which are mixed with cumulus minerals assumed to be in equilibrium with the respective melt. Though no single model reproduces the sample variation perfectly (see primitive mantle normalised models, Fig. S9) reasonable approximations are obtained (**Fig. 11**).

The trace element patterns of the dunites are approached with ~80% accumulation of olivine and spinel (95:5) and ~20% interstitial melt assumed as the high-Ti ultramafic 193203. It is important to note that the relatively high concentration of HREE preclude an origin from Barberton AUK type komatiites, because these have very low trace element concentrations. To match e.g. Yb concentration the melt would have to dominate the trace element budget (35% trapped melt), inheriting the flat HREE pattern resulting in absence of the characteristic subchondritic Dy/Yb<sub>N</sub>. The flat LREE pattern requires a melt with a flat LREE pattern, since any positively-trending LREE (and resulting high concentration) would effectively overprint the very low LREE concentration of the dunites (although whether primary or secondary is unclear). The only plausible alternative model is using a melt similar to some of the mafic host rocks (e.g. 193150), which displays flat trace element patterns but at higher concentrations (~4-5x primitive mantle – essentially MORB- or IAT-like)

than the Barberton AUK-type komatiites (~ primitive mantle). With such a melt, the dunite trace element patterns may be reproduced as ~97% cumulates with ~3% trapped melt (**Fig. 11**).

The peridotites with U-shaped trace element patterns (191126, 12963, 191198, 193261, 183146) can be approximated in an olivine-orthopyroxene-spinel cumulate model (75:20:5) assuming both a SHMB and AN-1 type melt (**Fig. 11**). The boninitic AN-1 melt model (~24-60% trapped melt) decently reproduce the general trace element pattern with a slight underestimation of the lightest REE, whereas the SHMB model (~4% trapped melt) reproduce the LREE well but underestimates the HREE, and vice versa for higher degrees of trapped melt. Neither of the models reproduce the largely absent Nb-anomaly, because the cumulate models inherit the negative Nb-anomaly of the assumed melts. Samples 191126 and 183146 have higher trace element concentrations with parallel LREE but, curiously, 191126 possess the lower LREE but higher HREE. No melts with LREE depletion or flat trace element patterns, e.g. komatiites, can reproduce the peridotites. Orthopyroxenites with U-shaped trace element patterns (177425, -35, 191232) cannot be reproduced in a komatiitic parental melt model as they are too enriched, exemplified by  $\geq$ chondritic Lu in the orthopyroxenites relative to approximately chondritic Lu in AUK Barberton komatiite (Sossi et al., 2016). Close approximations are obtained in an orthopyroxene-amphibole cumulate model (95:5) with an AN-1 parental melt (~42-60% trapped melt). The distinct negative Sr-anomalies may require plagioclase extraction from AN-1, which will result in higher overall trace element abundances, resulting in lower estimated trapped melt (~24-40%, **Figs. 11**). In spite of the good fit, this model cannot reconcile the low La/Nb of these samples (Fig. S10). SHMB-models either severely under- or overestimate the HREE or LREE with 2% and 10% trapped melt, respectively.

As previously pointed out, the high-Ti ultramafic rocks share many characteristics with komatiites. However, an alternative solution is that they represent cumulates associated with a mafic melt similar to the mafic host rocks that host ALC. Support for this scenario comes from slight negative Sr-Eu anomalies, suggestive of minor plagioclase fractionation, and weakly subchondritic Gd/Yb<sub>N</sub> (0.80-0.86). Plagioclase is not expected to fractionate from a primitive komatiite and the rocks may more easily be reconciled with a mafic melt. Furthermore, both forsterite- and Ni-content of olivine of WM peridotite (Fo<sub>82-85</sub>, 1620-2330 ppm; Aarestrup et al., 2020) is significantly lower than what is expected from average komatiite olivine (Fo<sub>89</sub>, 3400 ppm Ni; Arndt, 1994); using  $D_{Ni}^{ol/melt} \approx 8$  (Canil & Fedortchouk, 2001) would yield a melt Ni-content of ~200-290 ppm consistent with some of the mafic host rocks. However, recent modeling of olivine crystallising from high-MgO melts (20-30 wt.% of primary melt; Herzberg, 2016) suggest that the WM Fo-Ni content in olivine could be consistent with crystallisation from a komatiite liquid, as could most olivine from ultramafic complexes of the Fiskefjord region (Fig. S11; Szilas et al., 2015a, 2018; Guotana et al., 2018; Nishio et al., 2019).

The trace element patterns of the ultramafic rocks may be approximated using an orthopyroxene-amphibole (95:5) cumulate model assuming a melt similar to mafic host rock 193279, which has suffered 20% plagioclase fractionation. The trapped melt component constitute ~25-45%. The model reproduces the trace element characteristics closely and the slight La/Sm misfit between model and rocks could easily be reconciled with melt with flatter LREE such as 193510 (25% trapped melt, not shown). Peridotite 191224 can be generated in an olivine-orthopyroxene-spinel (75:20:5) cumulate model with 35% trapped melt. The Ni-TiO<sub>2</sub> can in principle also be explained by this model, using olivine (2140 ppm Ni) and orthopyroxene (292 ppm Ni) analyses from WM ultramafic rocks (Aarestrup et al., 2020) in a 60:40 ratio, mixed with mafic host rock 193279 (205 ppm Ni, 0.42 wt.% TiO<sub>2</sub>). However, this would require 40-75% (mafic) melt component and should significantly affect the major element budget, which seems unrealistic. Similar results apply to the Sc-TiO<sub>2</sub>, assuming 0 ppm Sc in olivine and 20 ppm Sc in orthopyroxene. While it is not possible to exclude an origin for these samples as basaltic cumulates, cumulates from SHMB or komatiites can easily be dismissed since the former is much too enriched in LREE and the latter have trace element concentrations approximately equal to the ALC high-Ti ultramafic rocks.

To model the melanorites, plagioclase is extracted from AN-1 (**Fig. 6**) using Rayleigh fractionation (Rollinson, 1993). However, this does not imply that plagioclase was the only liquidus phase, it is merely performed to simulate plagioclase extraction. The various samples probably reflect various stages of plagioclase removal and fractionation steps of 40-60-80% has been modelled. This large range in fractionation result in an equally large estimate of trapped melt ranging from 5% to 100%, where the cumulate assemblage is modelled as orthopyroxene with subordinate plagioclase and amphibole (80:10:10). The resulting model reproduce the melanorite trace element pattern well, although minor deviations do occur such as absence of Zr- and Ti-anomalies. The Ti-anomaly could in principle be remedied by minor Fe-Ti oxide fractionation, whereas the Zr-anomaly may reflect that the Zr-enrichment through plagioclase extraction ( $D_{Zr/Sm}^{Plg/melt} \approx 0.01-0.3$ ; e.g. Aigner-Torres et al., 2007; Dunn & Sen, 1994; Sun et al., 2017) was more effective than what the partition coefficients used in this modelling allow (note the tendency of negative Zr-anomalies in norites). Relatively extreme partition coefficients for plagioclase is indeed expected in silicic andesitic systems, as may have been approached for some melanorites, relative to basaltic systems (Dunn & Sen, 1994). While a SHMB model utilizing the same cumulate model superficially reproduces some of the melanoritic features, the model predicts too extreme La/Sm<sub>N</sub> (>2) and a continuous increase from Lu to La resulting in Dy/Yb<sub>N</sub> of >1 unlike the subchondritic values in the melanorites (**Figs. 9, 11**).

Some norites have relatively high trace element concentrations relative to AN-1, implying an overall more evolved melt composition and is approximated here as AN-1 after 60% fractional crystallisation of plagioclase. Such a scenario may physically relate to local expulsion of crystallising AN-1 melt and seems to

have been a more random or episodic process judging from the large range in La/Nb, La/Sm<sub>N</sub> and SiO<sub>2</sub>-TiO<sub>2</sub> of norites relative to the very systematic trending and limited range in melanorites (**Figs. 8-9, S8**). To simulate plagioclase aggregation, a plagioclase composition is assumed as the bulk composition of solids after 5% equilibrium plagioclase crystallisation from an unfractionated AN-1 melt; a solid composition equal to e.g. 40% equilibrium crystallisation of plagioclase will tend to result in models with slightly higher maximum La/Nb (9.8 vs. 7.7) at 50% trapped melt. The 5% equilibrium plagioclase reproduce the Eu-Sr anomalies best.

These models reproduce the norites well with trapped melt ranging ~5-50%, though La/Sm<sub>N</sub> is slightly underestimated in the most trace elementally enriched norites owing to the slight La-Ce depletion in the assumed plagioclase-fractionated AN-1 trapped melt. This could be remedied by e.g. using a less fractionated AN-1 melt or if AN-1 had assimilated some high(er) La/Sm<sub>N</sub> crust. The observed orthopyroxene may mark minor accumulation, which should not affect the results significantly, or it is related to the putative trapped melt. SHMB in a plagioclase cumulate model seems to reproduce the general trace element pattern of the norites, however this is only superficial as once again the model cannot reproduce the subchondritic Dy/Yb<sub>N</sub> and also is unrealistically LREE enriched with La/Sm<sub>N</sub> >3.1 (**Figs. 9, 11**).

### Modelling the effect of apatite fractionation

Apparent leucosomes (Aarestrup et al., 2020) and a crude negative SiO<sub>2</sub>-TiO<sub>2</sub> correlation in Amikoq mafic supracrustal rocks ( $R^2 = 0.21$ ,  $n = 13$ ; excluding outlier 183010) could support this notion, though if the second-most extreme sample is excluded, no correlation exists ( $R^2 = 0.02$ ). The flat and relatively coherent trace element patterns seem to preclude melt extraction given absence of LREE loss over HREE resulting in e.g. low La/Yb. This could be remedied by residual apatite, a commonly observed phase (Aarestrup et al., 2020), which has  $D_{\text{MREE-LREE}}^{\text{Apatite/melt}} \sim 2-28$  (Prowatke & Klemme, 2006) potentially acting as a MREE-LREE sponge. Conversely, HFSE are incompatible (e.g.  $D_{\text{Zr}}^{\text{Apatite/melt}} \sim 0.05$ ; Prowatke & Klemme, 2006) which should yield high Sm/Zr and La/Nb in an apatite-bearing residue. Residual amphibole should exert a similar control on Sm/Zr (e.g. Tiepolo et al., 2007). This scenario was tested in a simple non-modal batch melt model (Shaw, 1970) using the mafic granulite 193500 of Aarestrup et al. (2020); 40% plagioclase, 31% amphibole, 12% ortho- and clinopyroxene are implied. Apatite is varied from 0 to 5%. Amphibole is assumed present, although the amphibole found in Amikoq host mafic rocks probably is of retrograde origin (Aarestrup et al., 2020). The modelled residual rock after partial melt extraction is plotted in **Figure 7**.

### AFC modelling

The endmember models presented above may be too extreme to resolve the process of cumulates related to a continuously contaminated komatiite. To test this we model contamination of a Barberton-type

komatiite with flat REE-spectrum (Sossi et al., 2016) by an average Akia tonalite (Gardiner et al., 2019; **Table S2**, Fig. S9) using an assimilation-fractional-crystallisation model (DePaolo, 1981). This model is essentially identical to Precambrian SHMB petrogenesis (e.g. Sun et al., 1989) and implies that the LREE-enrichment as well as the negative Nb-anomaly is derived from contamination of the komatiite. Age considerations are important when choosing contaminant and it is questionable whether the c. 3 Ga tonalities (Gardiner et al., 2019) were available at the time of ALC intrusion, given the minimum age of 3 Ga. Alternatively, the c. 3050-3020 Ma magmatic phase producing TTGs plus minor diorite (Garde et al., 2014) or the c. 3.2 Ga central Akia nuclei of dioritic composition (e.g. Garde, 1997) may serve as more appropriate contamination candidates.

Differences between these two candidates are significant, evidenced from the heavy depletion of HREE in the average tonalite (subchondritic Yb) relative to the significantly higher HREE of the diorite (Yb ~9x chondrite; Fig. S9). Net result is that contamination by tonalite have near to no impact on the HREE-budget, e.g. after 50% combined AFC ( $r = 0.3$ ) fractionating olivine and orthopyroxene, Yb will have increased from 0.44 ppm to 0.92 ppm, and from 0.44 to 1.30 ppm using a tonalite and diorite contaminant, respectively. Conversely, the tonalite have a higher impact on the LREE budget relative to the diorite (melt La/Sm<sub>N</sub> of ~4.5 vs. ~3.2, respectively, after 50% combined AFC). Nevertheless, the overall results and derivative conclusions will not differ markedly as the modelled trends are similar and merely differ in amplitude.

Here we use the average tonalites as proxies for a crustal contaminant and note the similarity to B-1 of Bushveld and SHMB (Fig. S9). The very systematic behaviour of the noritic lithologies in terms of La/Sm<sub>N</sub>, Dy/Yb<sub>N</sub>, Sc/Y and La/Nb suggest that these ratios are undisturbed by metamorphism and reflect magmatic processes. Furthermore, these ratios span the entire trace element spectrum from the very incompatible elements (La/Nb) to least incompatible (Sc/Y, Dy/Yb), while Sc is also an important element in distinguishing komatiite from contaminated komatiite and in boninite petrogenesis (e.g. Crawford et al., 1989; Sun et al., 1989). Consequently, these ratios plotted against each other as well as against Sm, Yb, Sc and Nb are used to test the AFC-scenario. The modelled melt is then mixed with equilibrium cumulate olivine-orthopyroxene-spinel (75:20:5) in a peridotitic model (**Fig. 12**) and plagioclase-orthopyroxene (60:40) in a noritic model (**Fig. 13**). AFC melt evolution (red lines) are shown for  $r$ -values of 0 to 0.3 (unless otherwise indicated). Green, grey and dark red curves correspond to olivine, plagioclase and orthopyroxene compositions in equilibrium with the red melt evolution curves, whereas the purple curves represent the bulk crystals in equilibrium with the red melt curves. The blue evolution curves are cumulate models showing 1 or 10% trapped melt (light blue) and 15 or 40% trapped melt (dark blue), depending on cumulate model, demonstrating the effect of trapped melt shift (**Figs. 12-13**).

In general, all of the trace element characteristics of most peridotites can be reasonably well reproduced. Dy/Yb<sub>N</sub> in the modelled melt remain essentially uniform at chondritic values and contamination of komatiite

have next to no effect on the HREE (**Fig. 12**), as expected (Puchtel et al., 2013). Consequently, the subchondritic Dy/Yb<sub>N</sub> requires significant cumulus minerals (85-99%), whereas the highest peridotite La/Sm<sub>N</sub> (~2.8) requires up to ~30% or ~50% combined AFC ( $r = 0.3$  or  $0.2$ ); it is questionable whether such high degrees of AFC can produce the very primitive mineral and rock compositions observable at Amikoq (**Figs. 4, S2**). Some discrepancies arise. While the variable Sc/Y and low Sc is reproduced (except for some higher Sc samples), the low Sc/Y combined with low Dy/Yb<sub>N</sub> of the peridotites is difficult to reproduce suggesting that the initial melt had a lower Sc/Y of ~1.6, which is more akin to average MORB (~1.1) and the mafic host rocks (1.4-2.7) relative to komatiites (2.0-8.7; **Figs. 12, S10**). Furthermore, a plot of La/Nb vs. La/Sm<sub>N</sub> reveal that even in the case of 1% trapped melt, the resulting peridotite rock will inherit these ratios from the parent melt owing to the very low concentration of these elements in olivine + orthopyroxene (**Figs. 12, S10**). This implies that the three peridotites with highest La/Sm<sub>N</sub> but La/Nb of ~1 must have inherited these characteristics from their parent melt, as must the low-La/Nb dunites (~0.5), provided they are undisturbed. Because any plausible LREE-enriched contaminant available at the time have markedly elevated La/Nb ratios (**Figs. 12, S9**), it can be inferred that peridotites with La/Nb ~1 were negligibly contaminated, if at all. This supports the presence of uncontaminated high-Mg compositions as parental to at least some peridotites.

Importantly, these AFC models demonstrate that the subchondritic Dy/Yb<sub>N</sub> (0.74), high Sc/Y, Sc (8 and 40, respectively) and elevated La/Nb (2.7) of the AN-1 melt cannot be reproduced through contamination of a parental komatiite (red lines in **Fig. 12**), which is also obvious in Th/Yb vs. Nb/Yb space in Fig. S14. The misfit in terms of HREE and Sc-Y is due to both komatiite and investigated contaminants having Dy/Yb<sub>N</sub> ≥ 1 and Sc/Y < 4, and thus cannot evolve through AFC to the lower Dy/Yb<sub>N</sub> and higher Sc/Y of AN-1. Lastly, in the AFC modelling undertaken, all have model melts trending towards SHMB compositions, lending support to previous inferences that SHMB can be generated through contamination of a primary komatiite (e.g. Sun et al., 1989; Sparks, 1986).

Additional insight is gained from modelling the very systematic trends of the noritic ALC lithologies (**Figs. 13**). Even though the general shape of the melanorite trace element pattern can be reasonably approached by the simple cumulate model described above using AN-1, it is actually difficult to reproduce the trend toward higher Dy/Yb<sub>N</sub>-lower La/Sm<sub>N</sub> through simple fractionation and accumulation because of the modelled equilibrium plagioclase (~1.3 and ~4.4, respectively) and orthopyroxene (~0.3 and ~0.3, respectively). Significant plagioclase extraction (+ subordinate orthopyroxene) from AN-1 will result in a decrease in both ratios while accumulation of orthopyroxene would have the same effect. This could in part be caused by the used partition coefficients given that the depressed La-Ce-Pr of the melanorites are not perfectly reproduced from 40-80% plagioclase extraction from AN-1. Persistently low Dy/Yb<sub>N</sub> in the norites (**Fig. 13**) may suggest that the high-La/Sm<sub>N</sub> solid component actually had slightly lower Dy/Yb<sub>N</sub> than the modelled plagioclase;

however most plagioclase  $D$ 's predict  $D_{\text{Dy/Yb}}^{\text{Pl/melt}} > 1$  (e.g. Aigner-Torres et al., 2007; Sun et al., 2017) except in some calc-alkaline dacitic melts ( $\sim 0.8$ ; Fujikami et al., 1984) and rare basalt ( $\sim 0.6$ ; Schnetzler & Philpotts, 1970). Even if the extreme dacite  $D$ 's are used (which seem unrealistic) the model AN-1 melt  $\text{Dy/Yb}_N$  will only evolve from 0.74 to 0.75 after 50% pure plagioclase fractionation, underlining the inability of simple plagioclase fractionation in explaining the melanorite trend with  $\text{Dy/Yb}_N$  up to 0.96 (**Fig. 13**). Either this reflects the inability of such simple modelling to explain the natural process that separated the norites from their complimentary melanorites, or a different process must be sought.

Since the melanorites most probably are dominated by a melt component, judging from e.g. REE vs.  $\text{TiO}_2$  (**Figs. 8, S8**), then the melanorite trending could be indicative of contamination of AN-1 or mixing with a melt. The co-existence of two discrete melts in the same system is common (e.g. Falloon et al., 1989; Irvine et al., 1983; Szilas et al., 2015b). In this case, the persistent trending of the melanorites from AN-1 towards the average MORB composition suggests input from such a melt/contaminant. However, the slightly elevated  $\text{La/Nb}$  of the melanorites indicate high  $\text{La/Nb}$  of the melt/contaminant, unlike MORB ( $\sim 1$ ). Thus, the best fit is obtained with an IAT type of composition which generally mimics MORB but possess high  $\text{La/Nb}$  ( $> 4$ ; Todd et al., 2012). This model generally reproduces the melanorite trend well (**Fig. 13**), assuming a crystallising assemblage consisting of plagioclase and orthopyroxene (60:40) and that the trace element budget of the melanorites mostly reflect a melt. The  $\text{Sc/Y}$  trend is not well reproduced and could indicate a rather large  $r$ -value (up to 0.7), perhaps even bordering bulk mixing (DePaolo, 1981), suggesting that the IAT was a melt or hot wall rock.

This model reproduces the norites as dominantly plagioclase-orthopyroxene cumulates with the amount of trapped interstitial melt ranging from 0 to 40%, even though a slightly lower  $\text{Dy/Yb}_N$  of the plagioclase seem to be required (**Fig. 13**). In this model, the lower  $\text{La/Sm}_N$  and higher  $\text{Sc/Y}$  of the South Margin norites require a larger contribution from cumulus orthopyroxene relative to the West Margin norites that seems to be dominated by plagioclase. A lower orthopyroxene-content in West Margin norites may reconcile the lower  $\text{Cr-Ni-MgO}$  of these samples (**Fig. 4**). Input of an IAT-like melt/contaminant may help explain the abundance of plagioclase given the classification requirement for true primitive boninites to be plagioclase-free (although groundmass plagioclase may occur; Crawford et al., 1989) and plagioclase only becomes a liquidus phase in more evolved boninites (Taylor et al., 1994; Van der Laan et al., 1992). Contribution from IAT could have aided in pushing the melt composition towards the plagioclase stability field, resulting in the abundant observed plagioclase, which should not have been on the liquidus given an elevated melt  $\text{H}_2\text{O}$ -content and the general instability of plagioclase in primitive boninitic melts (e.g. Van der Laan et al., 1989). It is worth noting that the  $\text{Si-Al-Ca-Mg}$  content of AN-1 plot approximately between the lherzolite and harzburgite derived experimental melts of Falloon & Danyushevsky (2000).

High  $\text{La}/\text{Sm}_\text{N}$  combined with high  $\text{Dy}/\text{Yb}_\text{N}$  preclude both diorite and tonalite as appropriate contaminants, and it is worth noting that in  $\text{La}/\text{Sm}_\text{N}$ - $\text{La}/\text{Nb}$  space the melanorites trend in the direct opposite direction of these compositions as well as SHMB (**Fig. 13**). Some issues does remain such as the HREE- $\text{TiO}_2$  trending of the melanorites (**Figs. 8, S8**) that is difficult to reproduce. In an AFC model, an assimilant of unusually low  $\text{TiO}_2$  (<0.1 wt.%) is required or an unrealistic large amount of amphibole fractionation (>20% in fractionating assemblage and >50% combined AFC) which fail in reproducing other trace element characteristics (e.g.  $\text{La}/\text{Sm}_\text{N}$ - $\text{Dy}/\text{Yb}_\text{N}$ ). Either this require fractionation of Fe-Ti oxide (not modelled here) or a very unusual contaminant of IAT-like character and exceptionally low  $\text{TiO}_2$  – the former is preferred here. It is concluded that the very high  $\text{La}/\text{Sm}_\text{N}$  and  $\text{La}/\text{Nb}$  of the norites are not melt signatures and have been artificially elevated owing to plagioclase accumulation. Consequently, we urge caution in interpreting extreme  $\text{La}/\text{Sm}$  and  $\text{La}/\text{Nb}$  in plagioclase-rich rocks as direct evidence for subduction environments.

The modelling underlines the difference between some of the ultramafic samples (i.e. the high-Ti ultramafic rocks with low  $\text{La}/\text{Nb}$ ,  $\text{La}/\text{Sm}_\text{N}$ ) and the noritic samples. This raises the possibility that some of the ultramafic rocks rather are cumulates associated with the mafic host rocks or komatiites, rather than the noritic rocks. Thus, the rocks may not necessarily be coeval. A temporal distinction could also explain why the reef-like PGE occurrences in the ALC are associated with noritic lithologies rather than the ultramafic peridotitic rocks (Armitage, 2009). Aarestrup et al. (2020) reported mineral data on three samples from the same ultramafic body at WM which range from a more central dunitic/harzburgitic to orthopyroxenite at the margin of the body, with increases in forsterite ( $\text{Fo}_{82.5} \rightarrow \text{Fo}_{84.5}$ ) and enstatite ( $\text{En}_{82.5} \rightarrow \text{En}_{84} \rightarrow \text{En}_{85}$ ) content towards the margin. This may indicate that the peridotite was replaced through interaction with a high Si-Mg melt, such as AN-1, in principle similar to what occurred in the Chilas Complex in Pakistan, i.e. melt + olivine  $\rightarrow$  pyroxenes (Jagoutz et al., 2006) although the parental melt and resulting melt reaction was markedly different. This option opens up for an alternative origin for the trace elemental similarities between the noritic rocks and some ultramafic rocks. The positive Zr-anomalies of some ultramafic samples may have been inherited from such a melt-rock reaction (**Fig. 5**).

### **The position of the ALC in the Archean geotectonic jigsaw puzzle**

Mantle potential temperatures were higher in the early Earth, culminating in the Archean, yielding a higher melt output from ambient mantle resulting in MgO-rich primary basalts (18-24 wt.%) and a considerably thicker oceanic crust (~25-35 km) relative to today, with a complimentary harzburgitic restitic mantle (Herzberg et al., 2010). This may have resulted in long-lived thick basaltic crust prior to the stabilisation of true modern style subduction zones (Moyen & Laurent, 2018) considered present since c. 2.5 Ga (Bédard, 2018; Sizova et al., 2010). Recent zircon Hf-isotope data from the felsic constituents of Akia reveal a



contribution from Eoarchean mafic components supporting a long-lived, mafic crust in the Archean geological history of Akia (Gardiner et al., 2019) with a complimentary dunitic and harzburgitic SCLM (Bernstein et al., 2007; 2013).

Conversely, several studies have highlighted that many >2.7 Ga NAC rock associations depict structural and geochemical signatures typically associated with subduction zones (McGregor et al., 1991; Garde, 1997, 2007; Hoffmann et al., 2012; Klausen et al., 2017; Polat & Longstaffe, 2014; Polat et al., 2015; Steenfelt et al., 2005; Szilas et al., 2012b, 2017; Szilas, 2018; Van Hinsberg et al., 2018). How subduction zones behaved in the Archean is intensely debated (e.g. Bédard, 2018) and geochemical data suggest that modern style subduction zones were rare (Moyen & Laurent, 2018). Instead, it has been suggested that the Archean constitute a pre-subduction transition period where subduction was shallow and with a low dip leading to underthrusting rather than subduction (de Wit, 2004; Sizova et al., 2010). Eventually, the underthrust crust may have become gravitationally unstable and peeled off (Moyen & Laurent, 2018) or the weak oceanic crust may have dripped off into the mantle (Bédard, 2018; Johnson et al., 2013).

Given that modern style island arcs have been proposed to occur in Akia (e.g. Garde, 2007), an alternative scenario involve that ALC may be related to a pulse of melt emplaced into an ageing arc in conjunction with an arc-splitting event (Aarestrup et al., 2020) similar to the intrusive Himalayan Chilas Complex (85 Ma) of Pakistan (Burg et al., 1998; Jagoutz et al., 2006, 2007). If the Chilas Complex rock association (95 vol.% gabbro-norite plus some olivine-gabbro-norite and quartz-gabbro-norite, subordinate dunite with minor pyroxenite and olivine-websterite; Jagoutz et al., 2011) as well as the general geochemistry (parental melt; basaltic andesite, Mg#: 53, La/Sm<sub>N</sub>: 1.86, Dy/Yb<sub>N</sub>: 1.14; Jagoutz et al., 2006) is representative for such an arc splitting event, then we can easily dismiss this possibility, considering the significant lithological and geochemical differences between the ALC and the Chilas Complex. In fact, it seems that the mantle source for ALC was significantly more refractory than that of the Chilas Complex.

The occurrence of highly refractory cumulative ultramafic complexes in Akia (Szilas et al., 2015a) may suggest a role for a very high-Mg melt, possibly komatiitic (Szilas et al., 2018). In this regard, the boninitic AN-1 could be the result of SCLM melting in conjunction with plume impingement (Pearce & Reagan, 2019). However, komatiites *sensu stricto* are so far unknown from NAC (Szilas et al., 2012a, 2018). The high-Ti ultramafic samples discussed as possible komatiites are too ambiguous in their geochemistry to favour a plume model for the AN-1 at present. Furthermore, the occurrence of late igneous hornblende seem to preclude an unusually hot melt and, by inference, source.

Boninite petrogenesis requires 1) previous melt extraction leaving a LILE-LREE-depleted, low Dy/Yb, refractory lithology such as harzburgite, 2) re-introduction of LREE-LILE-Zr into the refractory boninite source lithology (Arndt, 2003; Cameron et al., 1983; Crawford et al., 1989; Hickey & Frei, 1982; König et al., 2010;

Pearce & Reagan, 2019). In the petrogenesis of modern boninites, the LILE-LREE are usually envisaged to originate from fluids (or melt-like slab components; König et al., 2010) released into the refractory mantle wedge by the subducting plate in a subduction zone (e.g. Reagan et al., 2017) although other possibilities have been suggested too (cf. Pearce et al., 1992; Pearce & Reagan, 2019). The positive Zr-anomaly have led workers to suggest that the enriching slab component originated from MORB in amphibolite facies as amphibole has low  $D_{\text{Zr/Sm}}^{\text{amp/melt}}$  (Crawford et al., 1989; König et al., 2010; Li et al., 2013; Pearce et al., 1992; Pearce & Reagan, 2019; Taylor et al., 1994).

The mafic host rocks of the ALC are ambiguous in their geochemistry and cannot be used in conjunction with the boninitic AN-1 to infer a complete supra-subduction scenario (Whattam & Stern, 2011). Furthermore, it is possible that the mafic host rocks and the ALC are temporally distinct. The AN-1 melt shows strong geochemical similarities to boninites from Chichi-jima (Cameron et al., 1983; Taylor et al., 1994). The Chichi-jima boninites are related to oceanic subduction initiation and succeed the first arc tholeiites of MORB-affinity that ultimately evolved to a normal arc basaltic system (Reagan et al., 2010, 2017; Pearce & Reagan, 2019). Hence, the ALC might reflect a subduction initiation system, in accord with the requirement for a high-La/Nb IAT-like contaminant. However, absent LILE-Zr anomalies in AN-1 must record a fundamental difference between Chichi-jima and the ALC. Provided that the low LILE-content is unrelated to metamorphic loss, it may suggest that the LREE re-enrichment of the boninitic AN-1 source was not related to fluids in a typical subduction zone setting. In a flat subduction setting where the oceanic crust is heated initially, perhaps to high-pressure granulite facies conditions, the crust may have lost LILE prior to delamination or dripping into the mantle. Once delamination or dripping does commence, the last fluids or a LILE-depleted melt phase may be released from the dense eclogitic crust to the overlying refractory mantle (Bédard, 2018). This could also reconcile the input of an IAT-like contaminant/mixer, as this model also predicts basaltic magmatism with subduction zone signatures (Bédard, 2018).

Absence of positive Zr-anomalies in AN-1 might indicate that the metasomatising component was derived from an amphibole-free source, consistent with an enriching component originating in an eclogitic or anhydrous high-pressure granulite. Furthermore, if the LREE-enriching component originated from a garnet-rich lithology, the HREE-Sc-Y content is expected to be extremely low (Kessel et al., 2005) and would be dominated by the refractory harzburgitic lithology. Akiyas highly refractory SCLM (Bernstein et al., 2007; 2013; Wittig et al., 2008) may have supplied the refractory source necessary in boninitic petrogenesis. It has been argued that the SCLM of the NAC was formed through subduction stacking processes (Tappe et al., 2011; Wittig et al., 2008). Detailed investigations of the >3 Ga Tartoq greenstone belt in the southern NAC support this scenario, where shallow trending in P-T-t space toward granulite facies is in marked contrast to modern high-P eclogite-facies trending (Van Hinsberg et al., 2018; Palin et al., 2020). Thus, rather than

representing modern style subduction initiation, the ALC may reflect an Archean equivalent involving subduction abortion and stacking. Pearce et al. (1992) argued that the boninite-source metasomatising agent was derived from a partial melt originating from the mafic subducting slab; if plausible, it may be relevant to note the close comparison of the pattern of the most incompatible elements of AN-1 and average Akia tonalite (Fig. S9) considering that these tonalities are thought to be derived from partial melting of mafic crust (Steenfelt et al., 2005; Yakymchuk et al., 2020; Gardiner et al., 2019).

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