

## Manuscript Details

<b>Manuscript number</b>	EARTH_2017_210_R1
<b>Title</b>	Mineralogical and geochemical evidence for polybaric fractional crystallization of continental flood basalts and implications for identification of their source lithologies
<b>Article type</b>	Review Article

### Abstract

Continental flood basalts undergo crystallization at a variety of pressures in the crust and sometimes even in the mantle. Polybaric fractionation, when magmas may pause and undergo crystallization at different pressures, results in complex fractionation of major elements. Crystallization at high pressures where clinopyroxene is an early crystallizing phase can result in erupted compositions that have major element characteristics which mimic those expected for melts derived from pyroxenite-rich sources. The trace element compositions of early-crystallizing olivine can add further detail to crystallization histories and potentially allow an examination of the crystallization of basalts from melt segregation to the surface. The North Atlantic Igneous Province (NAIP) comprises sub-regions which had diverse crystallization histories. Plateau basalts of the British Palaeogene Igneous Province (BPIP) were generated by partial melting of mantle peridotite starting at ~3.8 GPa with melting ceasing by ~2.7 GPa. Major elements indicate that some basalts crystallized at <1 GPa with plagioclase joining the liquidus before clinopyroxene. However, the majority of BPIP magmas crystallized clinopyroxene before plagioclase feldspar over the pressure range 1.0-2.0 GPa. Trace elements in olivine indicate crystallization of olivine + clinopyroxene over the pressure interval 1.6-2.0 GPa. However, olivine data also show that some near-primary magmas reached near-surface pressures without substantial modification by fractional crystallization. Olivines formed at  $\geq 1.6$  GPa have Ni and Ca contents that are consistent with an origin by partial melting of mantle peridotite with no role for pyroxenite being detected. This contrasts with the low pressure dominated crystallization histories exhibited by lavas from West Greenland and Iceland. Whole-rock data for many BPIP lavas exhibit CaO depletion at a given MgO content compared with those from West Greenland and Iceland, which might be cited as an indicator of pyroxenite in their source, but this is solely a consequence of augite fractionation at depth. An absence of augite phenocrysts in lavas may have resulted either from augite crystallization in the mantle or from a change from augite saturation to under-saturation with decreasing pressure, and consequent augite dissolution. The lack of any significant contribution from recycled oceanic crust to magmatism in the NAIP suggests that the petrological structure of the mantle source from which NAIP lavas were derived does not conform with the generally accepted models for mantle plumes such as those responsible for magmatism in Hawaii and the Siberian Traps.

<b>Keywords</b>	Continental flood basalts; polybaric fractional crystallization; olivine chemistry; pyroxenite; model primary magmas
<b>Corresponding Author</b>	Malcolm Hole
<b>Order of Authors</b>	Malcolm Hole
<b>Suggested reviewers</b>	Jussi Heinonen, Claude Herzberg, Tyrone Rooney, Sally Gibson

## Submission Files Included in this PDF

### File Name [File Type]

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Hole-ESR-Response.doc [Response to Reviewers]

Hole-MJ-ESR-Final-Revisions.doc [Revised Manuscript with Changes Marked]

Hole-Abstract.doc [Abstract]

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Data for: Mineralogical and geochemical evidence for polybaric fractional crystallization of continental flood basalts and implications for identification of their source lithologies

Primary magma models for Skye and Mull picrites

Mineralogical and geochemical evidence for polybaric fractional crystallization of continental flood basalts and implications for identification of their source lithologies  
Malcolm J. Hole.

A revised manuscript has been uploaded. In addition to the reviewer's minor comments (all of which have been addressed in the revised manuscript) the following more substantial revisions have been made;

- 1) A map of the NAIP and BPIP has been included as Fig. 1.
- 2) Sub-figures have been re-lettered so that they are referred to in a more logical order
- 3) Fig. 9 has been modified to show the same samples as Fig. 8. This has been done to improve the discussion on Fe/Mn and clinopyroxene crystallization (reviewer's comment 17).
- 4) **Pressure of crystallization of clinopyroxene (major comment).** This has been clarified in a number of places in the text, including in the abstract; in lines 238 to 251 and in the conclusions section in the revised manuscript. In addition, the fact that the Ol+Pl+Cpx barometer used by Herzberg (2004) is only calibrated to 1.0 GPa has been noted, and Fig. 3b has been modified to show data for Skye Cone Sheets which have been demonstrated to have crystallized at low pressure (1atm) by other workers.
- 5) **Discussion and conclusions.** In the light of the reviewer's comments (19) I have expanded the scope of the discussion and included section 8 entitled "The NAIP – a peridotite-dominated LIP". This section outlines the important differences between the NAIP and continental and oceanic LIPs that are dominated by pyroxenite and addresses the issues relating to the larger geodynamical implications of monolithological peridotite source melting displayed by the NAIP, as suggested by Prof. Herzberg.
- 6) Lambart (2017; *Geochem. Persp. Lett.* 4, 7-12.) was published while this paper was in review, and this has added a timely context for the discussion.
- 7) Fig. 13 has been added and is a summary model showing that the Ni contents of olivine from Iceland does not require, but does not preclude and role for pyroxenite in the petrogenesis of Icelandic lavas. This addresses some issues arising from Lambart (2017)

I hope that this is all satisfactory.

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4 **1 Mineralogical and geochemical evidence for polybaric fractional crystallization of**  
5 **2 continental flood basalts and implications for identification of peridotite and pyroxenite**  
6 **3 source lithologies.**

8  
9 **M.J. Hole<sup>a</sup>**

10 a. Department of Geology & Petroleum Geology, University of Aberdeen, Aberdeen AB243UE, UK.  
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16 and undergo crystallization at different pressures, results in complex fractionation of major  
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## 84 38 **1. Introduction**

87 39 Continental flood basalts (CFB) must traverse continental lithosphere before eruption at  
88  
89 40 the surface. For the North Atlantic Igneous Province (NAIP; Fig. 1), where the mantle was  
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91 41 anomalously hot ( $T_p \sim 1450-1550^\circ\text{C}$ ; Herzberg and Gazel, 2009; Hole and Millett, 2016;  
92  
93 42 Matthews et al. 2016) melting of mantle peridotite was initiated at  $\sim 3.7-4.5$  GPa (Hole and  
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95 43 Millett, 2016; Hole, 2015) and melting ceased when magmas rising by adiabatic ascent along  
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97 44 the olivine liquidus reached the asthenosphere-lithosphere boundary (LAB), which is also the  
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99 45 final pressure of melting ( $P_f$ ). The depth to the LAB varies throughout the province from  
100  
101 46  $\sim 100$  km beneath Disko Island (west of Greenland) to  $\sim 60$  km beneath Baffin Island (Hole  
102  
103 47 and Millett, 2016; Matzen et al., 2017). **During ascent from the initial melting pressure ( $P_i$ )  
104  
105 48 to the LAB, it is generally accepted that magmas crystallize olivine alone (e.g. Herzberg and  
106  
107 49 Asimow, 2008; 2015; Putirka 2008; 2011). However, Herzberg and Asimow (2008) state that  
108  
109 50 augite fractionation can occur in the mantle, an assertion that is corroborated by the  
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111 51 existence of clinopyroxene-bearing xenoliths in Hawaiian lavas that are consistent with  
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113 52 crystallization from a tholeiitic liquid at 110-150 km depth (Keshav et al., 2007). Magmas  
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121 53 may pause at the LAB or within the lithosphere prior to eruption, and in the British  
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123 54 Palaeogene Igneous Province (BPIP), which is a sub-province of the NAIP, there is a large  
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125 55 body of evidence that suggests that magmas paused at the Moho at about 30km depth, or  
126  
127 56 equivalent to  $\sim 0.9 \pm 0.15$  GPa (Thompson, 1974; 1982; Hole et al., 2015a; Millett et al., 2016).  
128  
129 57 Individual batches of magma may have undergone polybaric fractionation. Thompson (1974;  
130  
131 58 1982) on the basis of melting experiments, as well as major, trace element and isotopic  
132  
133 59 compositions of lavas, proposed that some mafic BPIP lavas underwent fractionation at 1.6-  
134  
135 60 1.7 GPa as well as at  $\sim 0.9$  GPa. Conversely, at Disko Island, very primitive and near primary  
136  
137 61 magmas were erupted through  $\sim 100$ km of lithosphere and seem not to have paused to  
138  
139 62 fractionate to any great extent in the crust (Larsen and Pedersen, 2000; 2009; Hole and  
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141 63 Millett, 2016; Matzen et al., 2017).

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145 64 O'Hara (1968) showed that with increasing pressure of crystallization, the liquidus  
146  
147 65 fields of olivine and clinopyroxene contracted and that the field for clinopyroxene  
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149 66 crystallization expanded at the expense of olivine. Experimental phase equilibria (e.g.  
150  
151 67 Villiger et al., 2004; 2007; Whitaker et al., 2007) show that for fractional crystallization of  
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153 68 peridotite-derived magmas at  $\sim 1$  GPa, clinopyroxene (Cpx) crystallizes in compositions with  
154  
155 69 Mg# (molecular %  $\text{MgO}/(\text{MgO} + \text{FeO} + \text{Fe}_2\text{O}_3)$ )  $\leq 65$  whereas the same magma crystallizing at 1  
156  
157 70 atm would have olivine as the sole liquidus phase at Mg# 65. With increasing pressure of  
158  
159 71 fractionation, Cpx becomes a liquidus phase at progressively higher Mg# (Fig. 2) which  
160  
161 72 results in an expansion of the Cpx stability field at the expense of olivine as noted by O'Hara  
162  
163 73 (1968). For a typical BPIP primary magma generated by volatile-free melting of mantle  
164  
165 74 peridotite (see Hole and Millett, 2016 for a compilation of primary magma compositions) at 1  
166  
167 75 atm,  $\sim 25\%$  olivine crystallization would be expected before plagioclase joins the liquidus,  
168  
169 76 whereas at 1.6 GPa, only about 10% crystallization of olivine alone would occur in the same  
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171 77 magma before Cpx joins olivine on the liquidus (Fig. 2). This also means that the minimum  
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180 78 Mg# (or Fo content) of olivine when it is the sole liquidus phase also increases with  
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182 79 increasing pressure (Fig. 2). Derivative liquids formed by Ol+Cpx fractionation at ~1 GPa,  
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184 80 which may ultimately erupt at the surface as lavas, will also have a lower CaO content for a  
185  
186 81 given MgO content than lavas that crystallize olivine alone because Cpx is a CaO-rich phase  
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188 82 and strongly fractionates MgO from CaO (Fig. 3A).

191 83 It is generally accepted that the source lithology for CFB and their oceanic equivalents  
192  
193 84 is not exclusively mantle peridotite, and that magmas derived from partial melting of  
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195 85 pyroxenite are an important component of CFB magmatism (e.g Herzberg, 2011). The  
196  
197 86 pyroxenite source components in LIPs is generally considered to have been derived from  
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199 87 oceanic crust recycled to lower mantle depths as a result of subduction tectonics (Herzberg,  
200  
201 88 2011; Sobolev et al., 2007). In the broadest terms, two distinct lithologies of pyroxenite can  
202  
203 89 be recognized on the basis of SiO<sub>2</sub> content. Primary or very primitive melts derived from  
204  
205 90 high SiO<sub>2</sub> pyroxenite (silica+pyroxene) are recognized because they plot on the SiO<sub>2</sub>-rich  
206  
207 91 side of the orthopyroxene-calcium Tschermaks component (Opx-Cpx-CaTs) join in the  
208  
209 92 molecular projection of the system olivine-Opx-CaTs-silica (Ol-Opx-CaTs-SiO<sub>2</sub>;  
210  
211 93 Herzberg, 2011). Conversely, melts derived from low SiO<sub>2</sub> pyroxenite (olivine+pyroxene)  
212  
213 94 plot to the SiO<sub>2</sub>-poor side of the same plane. Since the Opx-Cpx-CaTs join is a thermal  
214  
215 95 divide at pressures associated with mantle melting i.e. 3-7 GPa, derivative magmas from the  
216  
217 96 melting of high and low SiO<sub>2</sub> pyroxenite have divergent fractionation histories. At near  
218  
219 97 melting pressures, low SiO<sub>2</sub> pyroxenite may undergo mixing with melts of mantle peridotite  
220  
221 98 to form hybrid magmas, whereas melt derived from high SiO<sub>2</sub> pyroxenite cannot (Herzberg,  
222  
223 99 2011)

227 100 Distinguishing between pyroxenite- and peridotite-derived magmas in the lavas erupted  
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229 101 at LIPs using major or trace major element geochemical data is fraught with problems. A  
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231 102 commonly used indicator of the involvement of pyroxenite-derived melt in basalt  
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239 103 petrogenesis is that pyroxenite-derived magmas tend to be too deficient in CaO at a given  
240  
241 104 MgO content to be derived from melting of volatile-free mantle peridotite (Herzberg and  
242  
243 105 Asimow, 2008). However, fractionation of CaO relative to MgO is sensitive to the bulk  
244  
245 106 mineral/melt partition coefficient for CaO ( $DCaO$ ) in a magma, and some pyroxenites appear  
246  
247 107 to have  $DCaO > 1$  and others  $< 1$  regardless of the initial concentration of CaO in the solid  
248  
249 108 pyroxenite (e.g. Herzberg, 2011; Jennings and Holland 2015; [Jennings et al., 2016](#)). Residual  
250  
251 109 clinopyroxene during melting of pyroxenite is likely to result in the generation of CaO  
252  
253 110 deficient derivative lavas and therefore depends on the extent of melting at which  
254  
255 111 clinopyroxene is partially consumed (Lambart et al., 2016). A significant difficulty is using  
256  
257 112 CaO and MgO contents of mafic lavas to identify pyroxenite involvement is that lavas  
258  
259 113 derived from CaO-poor pyroxenite will fall in the same region of an MgO-CaO diagram as  
260  
261 114 peridotite-derived melts that have fractionated Ol+Cpx at crustal pressures (Herzberg and  
262  
263 115 Asimow, 2008; Fig. 3A). Another effect of crystallization of peridotite-derived magmas at  
264  
265 116 the Ol+Cpx+L cotectic is to drive liquids to progressively more SiO<sub>2</sub>-undersaturation with  
266  
267 117 decreasing Mg#. Consequently, crystallization of a peridotite-derived melt at ~1.0 GPa can  
268  
269 118 result in a CaO depleted, Si-undersaturated liquid composition that is indistinguishable from  
270  
271 119 a magma derived from the melting of low SiO<sub>2</sub> pyroxenite such as those found at the Canary  
272  
273 120 Islands and Madeira (Herzberg, 2011; Gurenko et al., 2013). Furthermore, recycled  
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275 121 pyroxenite is also known to be far more heterogeneous than mantle peridotite, both  
276  
277 122 chemically and mineralogically, and produces a diverse range of derivative magmas that may  
278  
279 123 be very similar to peridotite-derived magmas (Keshav et al., 2004; Herzberg, 2011; Lambart  
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281 124 et al., 2012; 2013; 2016;). For most high SiO<sub>2</sub> pyroxenite-derived magmas, covariations  
282  
283 125 between SiO<sub>2</sub> and CaO provide a reasonable discriminant from peridotite-derived magmas,  
284  
285 126 because most high Mg#, high SiO<sub>2</sub> lavas derived from mantle peridotite are silica saturated or  
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287 127 silica oversaturated olivine or quartz tholeiites, and they owe their high SiO<sub>2</sub> contents to  
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298 128 olivine fractionation at near-surface pressures; as a consequence, any deficiency in CaO at a  
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300 129 given MgO content is most likely attributable to high SiO<sub>2</sub> pyroxenite.

302  
303 130 Olivine phenocrysts in mafic lavas provide a mineralogical narrative about magmatic  
304  
305 131 processes in Earth's crust and mantle and olivine records in its chemistry the widespread  
306  
307 132 occurrence of magma chamber fractional crystallization, recharge, and mixing (Herzberg and  
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309 133 O'Hara, 2002; Herzberg, 2011; Herzberg et al., 2016; Sobolev et al., 2007). Olivine contains  
310  
311 134 readily measurable concentrations of trace elements Ni, Ca and Mn and the partitioning of  
312  
313 135 these element between olivine and coexisting melt is well-understood (e.g. Beattie et al.  
314  
315 136 1991; Beattie, 1994; 1995; Herzberg and O'Hara, 2002; Matzen et al., 2013; 2017).  
316  
317 137 Fractional crystallization can never be perfect in natural systems and so olivine in mafic lavas  
318  
319 138 may be able to record episodes of fractionation that are not evident from whole-rock data,  
320  
321 139 particularly in magmatic provinces such as the BPIP where polybaric fractionation is most  
322  
323 140 likely to have occurred. Olivine also records the melting of recycled crust as a distinct  
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325 141 pyroxenite lithology. In the case of Hawaii, many of the shield-forming lavas (e.g. Mauna  
326  
327 142 Kea) have Ni contents that are too high, and Ca contents that are too low, to have been  
328  
329 143 derived by partial melting of mantle peridotite and they are more likely to be derived from a  
330  
331 144 pyroxenite-rich source (Sobolev et al., 2005; Herzberg, 2006; 2011).

334 145 Before an understanding of the possible source lithology for MPLF lavas can be made, it  
335  
336 146 is clearly necessary to firstly constrain the fractionation histories of the magmas to enable an  
337  
338 147 assessment of the role of Cpx fractionation in causing CaO depletion in basic. Only if this is  
339  
340 148 understood can the contribution of low CaO pyroxenite to the source of the basalts be  
341  
342 149 assessed. To this end, a combination of whole-rock and component olivine chemistry has  
343  
344 150 been utilized to examine the fractionation history of some continental flood basalts from the  
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346 151 British Palaeogene Igneous Province (BPIP) which is part of the wider North Atlantic  
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348 152 Igneous Province (NAIP).  
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357 **153 2. Magmatism in the BPIP**  
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360 154 The BPIP is a sub-province of the large NAIP. Magmatism in the BPIP began at ~60 Ma and  
361  
362 155 continued until ~58 Ma. The majority of the lavas of the BPIP are mildly alkaline slightly *Ne-*  
363  
364 156 normative alkali olivine basalts and *Hy*-normative olivine tholeiites. The Mull Plateau Lava  
365  
366 157 Formation (MPLF; Williamson and Bell, 2012) is made of a sequence of ~1 km thickness of  
367  
368 158 plateau-forming lavas dominated by picrites and basalts with local occurrences of hawaiiite,  
369  
370 159 mugearite and trachyte (Kerr 1998; Kerr et al. 1995; 1999). The MPLF is overlain locally by  
371  
372 160 the Central Mull Formation (CMF; Williamson and Bell, 2012) which comprises olivine  
373  
374 161 tholeiite lavas and associated hypabyssal intrusions. The Skye Main Lava Series (SMLS;  
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376 162 Thompson, 1982) is petrologically very similar to the MPLF although it is probably slightly  
377  
378 163 older.  
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380  
381 164 A number of the mafic (>8 wt% MgO) CMF tholeiites which equilibrated at ~1 atm,  
382  
383 165 reached their erupted compositions by crystallization of olivine alone. For 49 of these lavas  
384  
385 166 and intrusions Hole & Millett (2016) generated model primary magma compositions using  
386  
387 167 PRIMELT3 (Herzberg and Asimow, 2008; 2015). Primitive parental melts to CMT erupted  
388  
389 168 compositions required a mantle potential temperature ( $T_p$ ) ~1500°C and MgO ~17.5 wt%.  
390  
391 169 The initial pressure of melting ( $P_i$ ) was ~3.6 GPa with melting stopping by ~2.7 GPa giving a  
392  
393 170 melt fraction for accumulated fractional melting (F-AFM) of ~0.16. Spice et al. (2016)  
394  
395 171 applied the aluminium-spinel-olivine thermometer of Coogan et al. (2014) to Skye and Mull  
396  
397 172 picrites (Mg# 78-85) which yielded crystallization temperatures of 1410°C and 1380°C  
398  
399 173 relative to MORB primitive magmas, which are in excellent agreement with olivine-liquid  
400  
401 174 crystallization temperatures for MPLF lavas given by Hole et al. (2015a). Four of the whole-  
402  
403 175 rock samples of Spice et al. (2016) also provide PRIMELT3 solutions (Appendix 1)  
404  
405 176 indicating  $T_p$  in the range 1524-1568°C and with  $P_f$  of 3.4-3.6 GPa. However, MPLF lavas  
406  
407 177 do not yield PRIMELT3 solutions, although olivine-hosted melt inclusions do provide model  
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416 178 primary magmas which indicate  $T_p \sim 1480^\circ\text{C}$  (Hole and Millett, 2016). The reason why  
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418 179 MPLF lavas do not allow the generation of model primary magmas is because they are too  
419  
420 180 deficient in CaO at a given MgO content to have been derived by olivine fractionation alone  
421  
422 181 from mantle peridotite (Fig. 2). As has already been noted, such CaO deficiency can be  
423  
424 182 caused augite fractionation or can be a characteristic of the source region for pyroxenite-  
425  
426 183 derived magmas.

#### 430 184 *2.1 Estimates of equilibration pressure of BPIP plateau lavas from major element chemistry.*

432 185 Based on experimentally determined phase relationships for SMLS lavas containing  
433  
434 186 11.1 wt% MgO, Thompson (1974; 1982) proposed that most Mg-rich picrites of the SMLS  
435  
436 187 and MPLF reached their erupted compositions at pressures up to about 1.7 GPa, and that the  
437  
438 188 remaining basic magmas of the BPIP have compositions related to cotectic equilibria at  
439  
440 189 approximately  $0.9 \pm 0.15$  GPa, which is near the base of the crust beneath the Province  
441  
442 190 (Thompson 1982). The melting experiments of Thompson (1974) showed that the general  
443  
444 191 sequence of phase relationships with decreasing T and MgO at  $\sim 1.0$  GPa is Ol+liquid (L);  
445  
446 192 Ol+Cpx+L; Ol+Cpx+Pl+L; whereas at 1 atm, plagioclase is a liquidus phase before  
447  
448 193 clinopyroxene. With increasing pressure, the stability field for clinopyroxene expands at the  
449  
450 194 expense of olivine, and at  $\sim 1.7$  GPa olivine is replaced by clinopyroxene as first crystallizing  
451  
452 195 phase in the system for compositions with  $>11$  wt% MgO. However, augite in typical  
453  
454 196 Plateau-type basalts occurs as groundmass poikilitic anhedral and never forms phenocrysts  
455  
456 197 (Thompson, 1982) and the dominant phenocryst phase in mafic plateau lavas is olivine.

459 198 Fig. 3 illustrates the relationships between CaO, MgO and Mg# for MPLF lavas with  $\geq 8$   
460  
461 199 wt% MgO. The liquid lines of descent (LLD) for a model primary magma at varying  
462  
463 200 pressures from 1 atm to 2.0 GPa is also shown. The primary magma composition chosen for  
464  
465 201 the forward model was the PRIMELT3 solution to Skye lava SK982, which is representative  
466  
467 202 of a typical peridotite-derived magma in this region (Hole and Millett, 2016). Additionally,  
468  
469



473  
474  
475 203 SK982 has  $\epsilon\text{Nd}_{58} = +8.9$  and  $^{206}\text{Pb}/^{204}\text{Pb}=17.5$  which means that it is largely unmodified by  
476  
477 204 interaction with the continental crust (Thompson et al., 1986; Hole et al., 2015a). Forward  
478  
479 205 models of fractional crystallization have been generated using Petrolg3 (Danyushevsky and  
480  
481 206 Plechov 2011) using the parameters given in the caption to Fig 2. Forward modelling of all  
482  
483 49 BPIP model primary magmas shows that the relationship between T, MgO, Mg# and the  
484  
485 207 pressure at which clinopyroxene appears on the liquidus is given by the approximations;  
486  
487 208

$$488 \quad 209 \quad \text{Mg\#} = -0.554\text{P}^2 + 9.81\text{P} + 55.9 \quad (1)$$

$$489 \quad 210 \quad \text{T} = 6.277\text{P}^2 + 141.9\text{P} + 1154.4 \quad (2)$$

492 211 For MgO the uncertainty is  $\pm 0.6\%$  and for Mg#  $\pm 2.0$ . Thus, for a BPIP magma crystallizing  
493  
494 212 at 1.0 GPa, clinopyroxene would join olivine on the liquidus at MgO =  $10.6 \pm 0.6$  wt% or Mg#  
495  
496 213 =  $65.2 \pm 2.0$ .

498 214 Inspection of Fig. 3B reveals the situation where CaO decreases with decreasing Mg# for  
499  
500 215 the assemblage Ol+Cpx+L and thereafter, CaO may increase or decrease with Mg#,  
501  
502 216 depending on pressure, for the assemblage Ol+Cpx+Pl+L. However, the overall variation in  
503  
504 217 CaO once clinopyroxene joins the crystallizing assemblage is not great. Thus, for the SK982  
505  
506 218 primary magma crystallizing at 1.0 GPa, CaO decreases from  $\sim 12.6$  to 12.1 wt% for a change  
507  
508 219 in Mg# of 61-65 and then remains nearly constant (CaO=11.7-12.0 wt%) along the  
509  
510 220 Ol+Cpx+Pl+L cotectic. This feature of Mg#-CaO covariations will be returned to later in the  
511  
512 221 discussion regarding olivine chemistry. It is also noticeable from Fig. 3 that data for the  
513  
514 222 majority of MPLF plateau lavas plots to the left and below the 1 atm Ol+L LLD and close to  
515  
516 223 the Ol+Cpx cotectics for various pressures. Data for CMF and Baffin Island lavas, all of  
517  
518 224 which yield PRIMELT3 solutions, plot close to the predicted olivine liquidus. Since  
519  
520 225 PRIMELT3 only provides solutions for the melting of mantle peridotite for erupted  
521  
522 226 compositions that have crystallized only olivine, then data for such lavas must plot on or  
523  
524 227 above the Ol+L LLD, as they do in Fig. 3. It is logical to assume, therefore, that the reason  
525  
526  
527  
528  
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531

532  
533  
534 228 for the MPLF lavas yielding few PRIMELT3 solutions is that they underwent augite  
535  
536 229 fractionation at  $\geq 1.0$  GPa, as proposed by Thompson (1974; 1982).

538  
539 230 Herzberg (2004) provided a method of estimating pressure of equilibration of MORB  
540  
541 231 tholeiitic basalts using a molecular projection in the system Anorthite-Diopside-Enstatite.  
542  
543 232 However, this method has an inherent uncertainty of  $\pm 0.28$  GPa for olivine tholeiites, can  
544  
545 233 only be applied to liquids that fall along the Ol+Pl+Cpx+L cotectic, and is calibrated only up  
546  
547 234 to  $\sim 1.0$  GPa. It has already been shown that for BPIP lavas, the initiation of clinopyroxene  
548  
549 235 crystallization is pressure and composition dependent. Consideration of Fig. 3B shows that  
550  
551 236 the predicted plagioclase-in position has a curvilinear relationship with Mg#. The equation  
552  
553 237 for this curve for the forward models of SK982 is;

$$555 \quad \text{CaO} = 0.0658[\text{Mg\#}]^2 - 6.99[\text{Mg\#}] + 194.8 \quad (3)$$

556 238  
557  
558 239 Data for most Central Mull Formation lavas which are predicted to have been simultaneously  
559  
560 240 saturated in olivine + plagioclase + augite fall above the 0.8 GPa crystallization trajectory in  
561  
562 241 Fig. 3A. Application of the Herzberg (2004) method for estimating  $P_{\text{eq}}$  gives values in the  
563  
564 242 range 0.0-1.0 GPa which are in good agreement with the estimates from Fig. 3B. Most CMF  
565  
566 243 lavas also yield PRIMELT3 solutions for melting of dry peridotite (Hole and Millet, 2016)  
567  
568 244 and therefore have CaO contents at a given MgO content that are consistent with their  
569  
570 245 derivation from mantle peridotite. Bell et al. (1994) presented extensive mineral chemical  
571  
572 246 and geochemical evidence for near 1 atm fractionation of the Skye cone sheets and data for  
573  
574 247 these intrusions and these plot close to the 1 atm Ol+Pl+Cpx+L cotectic in Fig. 3B. There is,  
575  
576 248 therefore, ample evidence of fractionation of MPLF and SMLS lavas in the range 1atm to  $>1$   
577  
578 249 GPa, which is consistent with the results from the melting experiments of Thompson (1974)  
579  
580  
581 250 and other published estimates (e.g. Scarrow and Cox, 1995; Kerr et al., 1999; Hole et al.,  
582  
583 251 2015a). However, many MPLF lavas plot in positions in Fig. 3 that suggest they underwent  
584  
585 252 crystallization of Ol+Cpx at pressures up to 2.0 GPa.

591  
592  
593 253 Interaction between mafic lavas and sialic crust is an integral part of the fractionation  
594  
595 254 history of BPIP magmas. Kerr et al. (1995) provided isotopic evidence for crustal  
596  
597 255 assimilation during turbulent magma ascent for MPLF lavas and there is strong evidence to  
598  
599 256 support the interaction between at least two discrete compositions of sialic crust and mantle  
600  
601 257 derived magma in the BPIP (Thompson et al., 1982; 1986; Hole et al., 2015a). The isotopic  
602  
603 258 compositions of BPIP lavas exhibit extreme variability, with  $\epsilon\text{Nd}_{60}$  varying from  $-30$  to  $+10$   
604  
605 259 and  $^{206}\text{Pb}/^{204}\text{Pb}$  14.3-18.5 which reflects the antiquity of the contaminants involved in  
606  
607 260 petrogenesis. For MPLF lavas, variations in  $\epsilon\text{Nd}_{60}$  are a more modest  $-8$  to  $+8$ . Because the  
608  
609 261 Archean sialic component had  $\epsilon\text{Nd}_{60} \sim -50$ , and was light rare earth element (LREE)  
610  
611 262 enriched, whereas many BPIP magmas are LREE depleted, the total amount of crustal  
612  
613 263 contamination required to reach  $\epsilon\text{Nd}_{60} -8$  was small and possibly only a few % (Hole et al.,  
614  
615 264 2015a). However, Thompson et al. (1986) noted that interaction with Archean granulite  
616  
617 265 facies crust was a feature of plateau lavas that fractionated at  $\sim 1.0$  GPa, whereas upper-  
618  
619 266 crustal contamination occurred during crystallization along the 1 atm Ol+Pl+Cpx cotectic.  
620  
621 267 Peate et al. (2012), using data from olivine-hosted melt inclusions, showed that the crustal  
622  
623 268 assimilation in some magmas must have taken place prior to growth of the olivines, but in  
624  
625 269 others the inclusions have less contaminated compositions than the whole-rocks and crustal  
626  
627 270 assimilation must have taken place both during and after growth of the olivines. Necessarily,  
628  
629 271 the evidence of crustal interaction is entirely from trace element and isotopic compositions.  
630  
631 272 During this study, no relationship was found between isotopic compositions and major  
632  
633 273 element chemistry of lavas, except for concentrations of  $\text{K}_2\text{O}$  and  $\text{TiO}_2$  which tend to be  
634  
635 274 elevated and reduced respectively in lavas that suffered crustal contamination relative to  
636  
637 275 uncontaminated lavas.

642 276 Major element data therefore suggest that the majority MPLF lavas crystallized  
643  
644 277 Ol+Cpx at  $\geq 1$  GPa. Consequently, the position occupied by these lavas in Fig. 3B, which is

650  
651  
652 278 below the peridotite-pyroxenite divide of Herzberg and Asimow (2008) could either be  
653  
654 279 because of their fractionation history or because they were derived from a CaO depleted  
655  
656 280 pyroxenite source. Consequently, major elements alone are not sufficient to identify the  
657  
658 281 course lithology of many MPLF lavas.  
659  
660  
661

### 662 282 **3. Models for trace element behaviour during olivine crystallization**

#### 663 664 283 *3.1 Calculation methods – Ni in olivine*

665  
666 284 The methods for calculating model olivine Ni and Ca compositions of primary magmas used  
667  
668 285 here follow those of Beattie et al. (1991), Beattie (1994; 1995), Herzberg and O’Hara (2002),  
669  
670 286 Toplis (2005), Herzberg (2011) and Matzen et al. (2013; 2017) and Gavrilenko et al. (2016).  
671  
672 287 Equations and assumptions pertinent to the current study are given below.  
673

674  
675 288 Fractional crystallization of primary magmas produces olivines that are expected to  
676  
677 289 crystallize along a liquid line of descent (LLD); such olivine compositions have been  
678  
679 290 appropriately termed “crystal line of descent,” or CLD (Putirka et al. 2011). Forward models  
680  
681 291 to define a CLD for olivine have been generated using Petrolog3 (Danyuchesvky and  
682  
683 292 Plechov, 2011) for model BPIP primary magmas (Hole and Millett, 2016) to give an LLD.  
684  
685 293 The predicted CaO, NiO and MnO concentrations in olivines formed in equilibrium with  
686  
687 294 liquids along the LLD have then been calculated using the parameters embedded in  
688  
689 295 PRIMELT3, or modifications of these, which are detailed below.  
690

691  
692 296 The forsterite (Fo) content of olivine in equilibrium with a melt is calculated using the  
693  
694 297 partitioning of Fe and Mg between olivine and melt;

$$695  
696 298 \quad K_D^{Ol/L}_{(FeO/MgO)} = D^{Ol/L}_{FeO} / D^{Ol/L}_{MgO} \quad (4)$$

697  
698  
699 299  $K_D^{Ol/L}_{(FeO/MgO)}$  varies of a function of pressure, temperature and composition ( $P$ ,  $T$ ,  $X$ ; e.g.  
700  
701 300 Toplis 2005). Herzberg and O’Hara (2002) provide a method for calculating  $K_D^{Ol/L}_{(FeO/MgO)}$   
702  
703 301 for the crystallization of a primary magma along the olivine liquidus from the pressure of  
704  
705  
706  
707  
708

709  
710  
711 302 melting to the surface, and this calculation method is available to the user in Petrolog3.  
712  
713 303 However, for magmas that pause in the crust, then it is  $K_D^{O/L}_{(FeO/MgO)}$  at the depth at which  
714  
715 304 olivine crystallization occurs that is relevant and not necessarily the ascent path to the surface  
716  
717  
718 305 along the olivine liquidus. The thermodynamical parameterization of Toplis (2005) allows  
719  
720 306 for the calculation of  $K_D^{O/L}_{(FeO/MgO)}$  at a given pressure for a known magma composition. The  
721  
722 307 Toplis (2005) model has been investigated using Petrolog3 for BPIP primary magma  
723  
724 308 compositions and the results are illustrated in Fig. 4 for three BPIP magmas with different  
725  
726 309 MgO contents which cover the range of  $T_p$  estimates for the region (Hole and Millet, 2016).  
727  
728 310 The final pressure of melting ( $P_f$ ) was used as a proxy for pressure of melt segregation which  
729  
730 311 for BPIP primary magmas is typically  $\sim 2.7$  GPa (Hole and Millett, 2016). This gives values  
731  
732 312 of  $K_D^{O/L}_{(FeO/MgO)} \sim 0.338$  for derivative magmas at 2.7 GPa formed from primary magmas  
733  
734 313 containing 17.5 wt% MgO or equivalent to  $T_p=1500^\circ\text{C}$  (Fig. 4A). For near-surface  
735  
736 314 crystallization,  $K_D^{O/L}_{(FeO/MgO)}$  for the same magma would be 0.30-0.31. At a fixed pressure  
737  
738 315  $K_D^{O/L}_{(FeO/MgO)}$  exhibits a negative covariation with  $\text{Mg}/\text{Fe}_{\text{Liq}}$  (cationic Mg/Fe of the liquid) for  
739  
740 316 liquids with Mg/Fe in the range 1.5-3.3 (Fig. 4B), which is equivalent to the composition of  
741  
742 317 near-primary magmas to moderately evolved basalts with  $\sim 8.0$  wt% MgO. Conversely, the  
743  
744 318 Herzberg and O'Hara (2002) calculation method defines a relationship between  $\text{Mg}/\text{Fe}_{\text{liq}}$  and  
745  
746 319  $k_D^{O/L}_{(FeO/MgO)}$  that is necessarily positive and curvilinear to account for  $P$ ,  $T$ ,  $X$  variability  
747  
748 320 during ascent along the olivine liquidus, but without magma pausing in the crust. The  
749  
750 321 practical use of this is that the point of intersection between the Herzberg and O'Hara (2002)  
751  
752 322 crystallization trajectory and a Toplis (2005) crystallization contour allows both  
753  
754 323  $K_D^{O/L}_{(FeO/MgO)}$  and  $\text{Mg}/\text{Fe}_{\text{liq}}$  at the contoured pressure to be estimated, which are unique to a  
755  
756 324 given primary magma. For example, considering a magma that segregated at 2.7 GPa and  
757  
758 325 then paused in the crust and crystallized olivine at 1.0 GPa, it can be assumed that the magma  
759  
760 326 follows the Herzberg and O'Hara (2002) curve in Fig. 4B from the point of magma  
761  
762  
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766  
767

768  
769  
770 327 segregation to 1.0 GPa, crystallizing olivine along the CLD. The melt then pauses at 1.0  
771  
772 328 GPa and crystallizes more olivine. The point at which the Herzberg and O'Hara (2002) LLD  
773  
774 329 intersects the 0.5 GPa Toplis (2005) contour, which in this example is at  $Mg/Fe_{liq}=1.7$  and  
775  
776 330  $K_D^{Ol/L}_{(FeO/MgO)}=0.317$  (Fig. 4B), is taken as the compositional starting point for 1 GPa  
777  
778 331 olivine crystallization. From that point onwards,  $K_D^{Ol/L}_{(FeO/MgO)}$  is estimated from  $Mg/Fe_{Liq}$   
779  
780 332 using the Toplis (2005) parameterization at a fixed pressure of 1.0 GPa. Whereas the  
781  
782 333 intersection point in Fig. 4b is unique to a specific primary magma, it is mainly influenced by  
783  
784 334  $Mg/Fe$  (and therefore also  $T_p$  and  $P_i$ ) of the primary magma and  $P_f$ . Consequently, for a  
785  
786 335 magmatic province with relatively consistent values  $T_p$  and  $P_f$  such as the BPIP (Hole and  
787  
788 336 Millett 2016) a single crystallization model is probably sufficient to describe the LLD of most  
789  
790 337 basalts and the CLD of their component olivines, and that is the approach which has been  
791  
792 338 taken here.  
793  
794

795  
796 339 An additional complication comes from the expansion of the clinopyroxene stability  
797  
798 340 field at the expense of olivine with increasing pressure (Fig. 2; Whitaker et al., 2007; Villger  
799  
800 341 et al., 2007). Magmas that pause in the crust are likely to crystallize on the Ol+Cpx+Liq  
801  
802 342 cotectic rather than along the olivine liquidus. Investigations using Petrolog3 show that  
803  
804 343  $K_D^{Ol/L}_{(FeO/MgO)}$  remains nearly constant for the Ol+Cpx+L crystallization interval, but the role  
805  
806 344 of olivine in the crystallizing assemblage diminishes substantially with continuing  
807  
808 345 crystallization (Fig. 2). Consequently, for crystallization at elevated crustal pressures, olivine  
809  
810 346 should be less abundant than for crystallization of the same magma near the surface. This  
811  
812 347 probably naturally limits the number of olivine analyses that can be generated for olivines  
813  
814 348 that crystallized on the Ol+Cpx+L cotectic. For the purposes of this study it has been  
815  
816 349 assumed that for crystallization of Ol+Cpx+L,  $K_D^{Ol/L}_{(FeO/MgO)}$  is constant at the pressure of  
817  
818 350 interest.  
819  
820  
821  
822  
823  
824  
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826

827  
828  
829 351 The method used for estimating the Ni content of a primary magma was that of Herzberg  
830  
831 352 (2011);

$$833 \quad 834 \quad 353 \quad \text{Ni ppm} = 21.6\text{MgO} - 0.32\text{MgO}^2 + 0.051\text{MgO}^3 \quad (5)$$

835  
836  
837 354  $D^{\text{O}/\text{L}}_{\text{NiO}}$  was determined using the method of Beattie et al. (1991) which was re-examined by  
838  
839 355 Herzberg et al. (2016) and found to remain applicable in the light of new experimental  
840  
841 356 results;

$$842 \quad 843 \quad 357 \quad D^{\text{O}/\text{L}}_{\text{NiO}} = 3.346 D^{\text{O}/\text{L}}_{\text{MgO}} - 3.665 \quad (6)$$

844  
845  
846 358 Because  $D^{\text{O}/\text{L}}_{\text{MgO}}$  is determined from  $K_{\text{D}}^{\text{O}/\text{L}}_{(\text{FeO}/\text{MgO})}$  then magmas which pause at depth in the  
847  
848 359 crust are pressure-compensated for  $D^{\text{O}/\text{L}}_{\text{NiO}}$  by the  $K_{\text{D}}^{\text{O}/\text{L}}_{(\text{FeO}/\text{MgO})}$  calculation method outlined  
849  
850 360 above and in Fig. 4.

851  
852 361 Matzen et al. (2013; 2017) showed that the temperature dependence of  $D^{\text{O}/\text{L}}_{\text{NiO}}$  can  
853  
854 362 ultimately result in olivine crystallizing from near-primary magmas at near-surface pressures,  
855  
856 363 having higher Ni contents than olivine crystallizing from the same magma at the pressure of  
857  
858 364 melt segregation. The relationship between the NiO content of an olivine crystallizing from  
859  
860 365 the same magma at 1 bar and at the pressure of melt segregation is given by;

$$861 \quad 862 \quad 366 \quad \text{NiO}^{\text{O}/\text{L}}_{\text{T}_{\text{P}0}}/\text{NiO}^{\text{O}/\text{L}}_{\text{T}_{\text{P}f}} = \exp(4505 - (1/\text{T}_{\text{P}0} - 1/\text{T}_{\text{P}f})) \quad (7)$$

863  
864  
865  
866 367 Where  $\text{Ni}^{\text{O}/\text{L}}_{\text{T}_{\text{P}0}}$  and  $\text{NiO}^{\text{O}/\text{L}}_{\text{T}_{\text{P}f}}$  are the NiO contents, in weight units, of olivine crystallizing  
867  
868 368 from magma at the surface and close to the pressure of melt segregation respectively, and  $\text{T}_{\text{P}0}$   
869  
870 369 and  $\text{T}_{\text{P}f}$  are the temperatures of the olivine saturated magma at 0 GPa and at the pressure of  
871  
872 370 melt segregation respectively. Olivine-saturated  $\text{T}_{\text{P}f}$  was taken from  $\text{T}_{\text{O}1}$  of the PRIMELT3  
873  
874 371 solution for the magma of interest (Herzberg and Asimow, 2015; Hole and Millett, 2016).  
875  
876 372 Since subsequent crystallization was assumed to take place near to the surface  
877  
878 373  $K_{\text{D}}^{\text{O}/\text{L}}_{(\text{FeO}/\text{MgO})}=0.30$  was used in all calculations. Application of equation (7) to primary  
879  
880 374 magma compositions therefore yields the maximum possible Ni content of an olivine  
881  
882  
883  
884  
885

886  
887  
888 375 crystallizing from a primary magma at near-surface pressure. Such olivines are referred to  
889  
890 376 hereafter as ‘Matzen olivines’.

892  
893 377 *3.2 Calculation methods – Ca in olivine*

894  
895  
896 378 Three different methods are available for calculating the model olivine Ca compositions of  
897  
898 379 primitive magmas. Two of these (Beattie et al., 1991; Herzberg and O’Hara, 2002) are based  
899  
900 380 on variation in  $D^{O/L}_{CaO}$  as a function of  $D^{O/L}_{MgO}$ . The Beattie et al. (1991) method, which is  
901  
902 381 appropriate to near-surface pressures, and is given by;

903  
904  
905 382 
$$D^{O/L}_{CaO} = 0.0056 D^{O/L}_{MgO} + 0.0135 \quad (8)$$

906  
907 383 And for pressures close to those of melt segregation in the range 3-7 GPa, the equation of  
908  
909 384 Herzberg and O’Hara (2002) can be applied;

910  
911  
912 385 
$$D^{O/L}_{CaO} = -0.019 D^{O/L}_{MgO} + 0.007 D^{O/L}_{MgO} + 0.0063 / D^{O/L}_{MgO} \quad (9)$$

913  
914 386 Gavrilenko et al. (2016) provided a further method of calculating  $D^{O/L}_{CaO}$  using only the  
915  
916 387 MgO content of an anhydrous magma based on new melting experiments. This method is  
917  
918 388 applicable over a wide range of pressures (1atm to 14 GPa) and temperatures (1150-2050°C).  
919  
920 389 For magmas containing <11.1 wt% MgO;

921  
922  
923 390 
$$D^{O/L}_{CaO} = -0.0043 MgO + 0.072 \quad (10)$$

924  
925 391 And for magmas containing >11.1 wt% MgO;

926  
927  
928 392 
$$D^{O/L}_{CaO} = 0.00042 MgO + 0.0196 \quad (11)$$

929  
930 393 Fig. 5A shows Petrolog3 forward crystallization models of two BPIP model primary magmas  
931  
932 394 (SK982 and MU1.1) at a nominal pressure of 1 atm for the three olivine-liquid CaO  
933  
934 395 partitioning models given in equations (8)-(11). In addition, the CaO contents of olivine in  
935  
936 396 equilibrium with 49 model primary BPIP magmas calculated for equations (8)-(11) are also  
937  
938 397 shown in Fig. 5A. The uncertainty in  $D^{O/L}_{CaO}$  ( $\pm 0.004$ ) for the Gavrilenko et al. (2016) model  
939  
940 398 for compositions with >11.0 wt% MgO propagates to  $\pm 300$  ppm Ca in Fig. 5. The Beattie et  
941  
942  
943  
944



945  
946  
947 399 al. (1991) and Gavrilenko et al. (2016) models predict very similar primary equilibrium  
948  
949 400 olivine compositions for BPIP magmas, but exhibit significant differences in slope of the  
950  
951 401 CLD, with the Gavrilenko et al. (2016) model initially predicting lower  $D^{Ol/L}_{CaO}$  than the  
952  
953 402 Beattie et al. (1991) model for the same range of Fo contents. However, at around 85-87%  
954  
955 403 Fo, equivalent to a whole rock MgO content of 11 wt% (see equations (10) and (11) above)  
956  
957 404 the Gavrilenko et al. (2016) model exhibits a sharp increase in olivine Ca content with  
958  
959 405 decreasing Fo compared to the Beattie et al. (1991) model. The Herzberg and O'Hara (2002)  
960  
961 406 model predicts lower Ca contents of primary olivine than either of the other two models.  
962

963  
964 407 Fig 4b illustrates the effects of crystallization of magmas at varying pressures on Ca and  
965  
966 408 Fo contents of olivine. A single model primary BPIP magma (SK982) has been selected and  
967  
968 409 Petrolog3 forward models at pressures in the range 1atm to 2.2 GPa generated. CLDs for  
969  
970 410 olivine have then been calculated from equations (9)-(11). The Beattie et al. (1991) model  
971  
972 411 has not been investigated because it is applicable only to crystallization at 1 atm. The key  
973  
974 412 features of Fig. 5 are that both the Herzberg and O'Hara (2002) and Gavrilenko et al. (2016)  
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976 413 models develop an inflection on the CLD at the point at which clinopyroxene joins the  
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978 414 crystallizing assemblage, which is also reflected in the major element data for BPIP lavas  
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980 415 (Fig. 3). According to the Gavrilenko et al. (2016) model, crystallization of the assemblage  
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982 416 Ol+Cpx+L causes the Ca contents of equilibrium olivine to decrease relative to the primary  
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984 417 olivine compositions, whereas the Herzberg and O'Hara (2002) model suggests a buffering of  
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986 418 Ca content of equilibrium olivine for the same assemblage. Additionally, for the Gavrilenko  
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988 419 et al. (2016) model, the CLD for Fo and Ca for the assemblage Ol+L has a relatively shallow  
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990 420 slope for Fo contents in the range 85.5-91.5, which is equivalent to about 11 wt% MgO in the  
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992 421 whole rock (see equation (11)). For the purposes of consistency, the most recent  
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994 422 parameterizations for  $D^{Ol/L}_{CaO}$  of Gavrilenko et al. (2016) are primarily used below, but the  
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996 423 Herzberg and O'Hara model (2002) is also considered if necessary.  
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1006 424 Also shown in Fig. 5B are data for natural olivines in near-primary Siqueiros Fracture  
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1008 425 Zone MORB. These have  $\text{Ca}=2060\pm 63$  ppm at  $\text{Fo}=89.9\pm 0.5$  (64 samples from Gavrilenko et  
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1010 426 al., 2016). Olivine in equilibrium with PRIMELT3 model primary magma compositions  
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1012 427 (Herzberg and Asimow 2015) have predicted  $\text{Ca}=2204\pm 58$  ppm and  $\text{Fo}= 89.9\pm 0.3$  using the  
1013  
1014 428 method of Gavrilenko et al. (2016), which is lower than the observed abundances. However,  
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1016 429 for MORB, predicted and observed data fall within the limits of the regressions used in the  
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1018 430 Gavrilenko et al. (2016) model.  
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#### 1022 431 **4. Olivine in MPLF lavas**

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1024 432 Data for olivine in MPLF lavas are taken from Sobolev et al. (2007) who analyzed olivine  
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1026 433 grains from 14 different samples giving a total data set of 842 individual determinations.  
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1028 434 Spice et al. (2016) provide a further 64 analyses from Skye, Mull and Rum picrites all of  
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1030 435 which have whole-rock MgO contents in the range 24.8-31.9 wt% MgO. These picrites will  
1031  
1032 436 be treated as a separate group of rocks from the MPLF because their stratigraphical affinity is  
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1034 437 unclear they are unlike any samples found within the main MPLF lava pile.  
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1037 438 Covariations between Fo content and Ni content for selected analyses are shown in Fig. 6.  
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1039 439 The data have been selected to be representative of all MPLF olivine, and particularly the  
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1041 440 range of Fo contents which is from ~75-90, although the bulk of the data have  $\text{Fo} > 80$ .  
1042  
1043 441 Olivine with compositions in the range Fo 80-90 would be in equilibrium with whole-rock  
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1045 442 compositions with Mg# 54-73 and MgO 7.7-15.8 wt% at 1 atm based on forward models of  
1046  
1047 443 the primary magma to SK982. This range in whole-rock compositions encompasses the  
1048  
1049 444 majority of that for MPLF lavas (Fig. 3). The CLDs in Fig. 6 were calculated both for the  
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1051 445 Beattie et al. (1991) and the Herzberg and O'Hara (2002) models. For olivine in MPLF  
1052  
1053 446 lavas, Ni correlates positively with Fo and the data falls into two distinct groups which  
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1055 447 relate to the position of data relative to the 1 atm Beattie CLD; data for samples BR2, BR6  
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1057 448 and BHL15 (Fig. 6A) plots closer to the 1 atm CLD than the remaining samples, the latter all  
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449 having a higher Ni for a given Fo content than, for example, sample BR2. Olivine from  
450 picrites from Mull, Skye and Rum (Spice et al. 2016) plot close to the position expected of  
451 olivine in equilibrium with primary magmas. Using these data and equation (7) above, gives  
452 Matzen olivines with Fo 91.2–91.8 and Ni = 3560-3570 ppm which are within the range of  
453 the analyzed values for olivine in the Skye and Mull picrites. Indeed, Matzen olivines  
454 calculated to be in equilibrium with all 49 BPIP PRIMELT3 primary magmas (Fig. 6B) show  
455 considerable overlap with the olivine data from Spice et al. (2016). The picrites of Spice et  
456 al. (2016) which contain high Fo and high Ni olivines must therefore have been formed by  
457 rapid emplacement of a near-primary magma at the surface followed by olivine  
458 crystallization at close to 1 atm. This allows the maximum Ni content of BPIP olivines to be  
459 constrained to ~3600 ppm at Fo ~91.5, values which are within the normal range expected for  
460 dry melting of mantle peridotite at  $T_p \sim 1500^\circ\text{C}$  (Matzen et al., 2017) and within the range of  
461 those calculated for lavas from west Greenland (Herzberg et al., 2016; Matzen et al., 2017).

462 Since it is likely that MPLF lavas equilibrated at pressures  $\geq 1.6$  GPa, the CLD for SK982  
463 at 1.6 GPa has been calculated and is shown in in Fig. 6. It shows a remarkable  
464 correspondence with the data from MPLF samples BR2, BR6 and BHL15, including an  
465 apparent inflection in the data trend which corresponds to clinopyroxene joining the liquidus.  
466 The inflection occurs because  $D^{\text{Cpx/L}}_{\text{NiO}} \sim 0.25 \times D^{\text{Ol/L}}_{\text{NiO}}$ , (Herzberg et al., 2016) and  
467 therefore crystallization along the Ol+Cpx+L cotectic results in a smaller bulk mineral-melt  
468 distribution coefficient for Ni than for olivine crystallization alone. This, in turn, results in  
469 liquid compositions that have fractionated Ol+Cpx having higher Ni contents than for the  
470 same liquid fractionating only olivine, at the same Mg#. It is also noticeable that the data for  
471 near CLD MPLF lavas terminates at the Ol+Cpx+Pl+L cotectic. Mixing between primary  
472 and derivative magmas along the LLD and their subsequent crystallization of olivine would  
473 produce olivine compositions that fall on the mixing lines shown in Fig. 6. The remaining

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474 data for MPLF olivines fall along these mixing lines. This is a similar situation to that  
475 described by Herzberg et al. (2016) for mixing relationships in olivines and primary and  
476 derivative magmas from Theistareykir, Iceland. The mixing lines shown in Fig. 5 relate to  
477 specific MgO contents of the derivative magma along the LLD. So, for sample BR5, mixing  
478 between a primary magma and a derivative magma with ~6 wt% MgO could produce the  
479 observed array of olivine compositions, whereas sample BHL34 would require a derivative  
480 magma with higher MgO content to produce its component olivine compositions.

481 Fig. 7 shows covariations between Fo and Ca content of the same olivines as shown in  
482 Fig. 6. Because Ca is incompatible with respect to olivine ( $D^{Ol/L}_{CaO}$  typically 0.025-0.031)  
483 then a negative covariation between Fo and Ca content might be expected, but this is not  
484 observed. A feature of Fo-Ca covariations in MPLF olivines is the relatively consistent Ca  
485 content of 1800-2400 ppm over a range of Fo = 80-89. The predicted range of olivine  
486 compositions calculated for the fractional crystallization of 49 BPIP PRIMELT3 solutions  
487 from primary MgO contents to 8 wt% MgO, using the Beattie et al. (1991) 1atm  
488 crystallization and Gavrilenko et al. (2016) olivine liquidus models are also shown in Fig. 7.  
489 The field for olivine calculated using the Beattie *et al.* (1991) model has a considerably  
490 steeper slope than that for the Gavrilenko et al. (2016) model. It is noticeable that olivine in  
491 the Skye picrites mostly fall within the range of the Beattie et al. (1991) model and close to  
492 primary olivine compositions predicted by the Gavrilenko et al. (2016) model, which is also  
493 consistent with their position in Ni-Fo space (Fig. 6). Olivine in Mull picrites appear to have  
494 consistently lower Ca for a given Fo content than the Skye olivines.

495 Very few of the data for olivine grains from MPLF lavas fall within the region reserved  
496 for 1 atm olivine crystallization from primary and derivative magmas using the Gavrilenko et  
497 al. (2016) model. CLDs at varying pressures, calculated as described earlier, are also shown  
498 in Fig. 7, and mirror the data trends shown in Fig. 3 such that once Cpx joins olivine on the

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1183 499 liquidus, the CaO content of olivine decreases slightly at a given pressure. Therefore, as in  
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1185 500 Fig. 4 for whole-rock data, some of the MPLF olivines exhibit patterns of distribution of Fo  
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1187 501 and CaO that are consistent with crystallization along the Ol+Cpx+L cotectic. However, the  
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1189 502 position of isobars in Fig. 7 cannot be predicted with certainty because the CaO abundances  
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1191 503 of BPIP primary magmas vary considerably, such that at ~1.6 GPa the variation within  
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1193 504 primary magmas is equivalent to a variation of ~500 ppm Ca in the olivine data set. In  
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1195 505 addition, slow diffusion of Ca in olivine can result in consistent Ca but variable Fo contents  
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1197 506 in individual grains, which is not evident for faster-diffusing Ni data (Coogan et al., 2005;  
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1199 507 Chakraborty, 2010). Consequently, the apparent sub-horizontal arrays delineated by olivines  
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1201 508 from individual MPLF samples could in part be a result of slow Ca diffusion, and it is the Ca  
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1203 509 content of the most Fo-rich samples that best reflects crystallization conditions. However,  
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1205 510 the Ca and Fo content of the MPLF lavas shown in Fig. 7 seems to require crystallization at  
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1207 511  $\geq 1.4$  GPa  
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1211 512 Covariations between NiO and CaO are shown in Fig. 8 for all the Ca partitioning  
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1213 513 models in equations (8)-(11) based on the primary magma to Skye lava SK982, and for the  
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1215 514 Herzberg and O'Hara (2002) and Gavrilenko et al. (2016) methods, at varying pressures. Ca-  
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1217 515 Ni covariations for the Gavrilenko et al. (2016) model exhibit a series of negative 'spikes'  
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1219 516 which relate to the composition at which clinopyroxene joins olivine in the crystallizing  
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1221 517 assemblage, which is a predictable consequence of augite fractionation. Because  
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1223 518 clinopyroxene joins the crystallizing assemblage at progressively higher MgO contents with  
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1225 519 increasing pressure (Fig. 3), the negative 'spikes' occur at higher Ni contents with increasing  
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1227 520 pressure. However, an added complication is that for pressures >1.2 GPa, clinopyroxene joins  
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1229 521 the crystallizing assemblage at >11.1wt% MgO so equation (11) applies to the assemblages  
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1231 522 Ol+L and Ol+Cpx+L, whereas for pressures <1.2 GPa clinopyroxene joins the crystallizing  
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1233 523 assemblage at <11.1wt% MgO and so equation (11) only applies to the olivine liquidus.  
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1242 524 Once crystallization takes place along the Ol+Cpx+Pl+L cotectic Ni-Ca variations define  
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1244 525 shallow concave upwards trends, Ca contents decreasing with increasing pressure of  
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1246 526 fractionation. The Herzberg and O'Hara (2002) model is less complex and predicts lower Ca  
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1249 527 for a given Ni content than the Gavrilenko et al. (2016) model.  
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1251 528 Data for olivine in Skye picrites falls close to the extension of the trajectory for low  
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1253 529 pressure (~1 atm) crystallization of Ol+L and close to the position of Matzen olivines  
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1255 530 calculated to be in equilibrium with BPIP primary magmas at 1 atm. Data for olivine in Mull  
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1257 531 picrites generally have lower Ca for a given Fo than those from Skye picrites, and lower than  
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1259 532 the model Matzen olivines. This may suggest that the Mull picrites may have experienced  
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1261 533 clinopyroxene fractionation in the mantle (Herzberg and Asimow, 2008). The data for MPLF  
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1263 534 olivines shown in Fig 7c plots significantly below the olivine liquidus of Gavrilenko et al.  
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1265 535 (2016) and close to, or below, the olivine liquidus for the Herzberg and O'Hara (2002)  
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1267 536 model. Consequently, data for the samples shown in Figs 6 and 7 are consistent with  
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1269 537 crystallization of Ol+Cpx+L at elevated pressures up to ~2.2 GPa. Given that the final  
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1271 538 pressure of melting for many BPIP lavas is ~2.7 GPa (Hole and Millett, 2016) then in some  
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1273 539 cases, crystallization of olivine must have commenced at or close to the lithosphere-  
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1275 540 asthenosphere boundary, as suggested by Matzen et al. (2017). In Fig. 8A data are shown for  
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1277 541 MPLF olivines that falls between the Gavrilenko et al. (2016) and Herzberg and O'Hara  
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1279 542 (2002) olivine liquidii, and therefore close to the expected range of Ca and Ni content for  
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1281 543 crystallization of the assemblage Ol+L. BPIP Matzen olivines fall on the extension of the  
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1283 544 Ol+L trajectory and have higher Fo contents ~92 (Fig. 7) than the observed values for MPLF  
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1285 545 lavas (maximum ~Fo88). Crystallization of ~15-20% olivine from a Skye picrite  
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1287 546 composition could produce the observed Ni, Ca and Fo contents of the MPLF olivines shown  
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1289 547 in Figs 7A and 8A. On the basis of Ni and Ca distributions in olivines discussed above all  
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1291 548 MPLF lavas could have been generated by crystallization from a peridotite-derived magma at  
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1301 549 variable, but elevated pressures of up to ~2.2 GPa. Consequently, there is no evidence to  
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1303 550 suggest the involvement of pyroxenite in the source of the MPLF lavas.

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1305 551 Herzberg (2011) showed that  $Fe/Mn > 70$  at any Fo content is characteristic of olivine  
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1307 552 that crystallizes from pyroxenite compared to  $Fe/Mn$  60-70 for olivine in peridotite-derived  
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1309 553 magmas. The mean  $Fe/Mn$  for MPLF lavas is  $69.2 \pm 2.6$  (Fig. 9) but two samples (BCH14 and  
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1311 554 AM7a) fall exclusively above 70 with a maximum  $Fe/Mn$  of 75 (Fig. 9A). However,  
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1313 555 Herzberg et al. (2013) demonstrated that clinopyroxene fractionates Fe from Mn causing  
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1315 556 elevated  $Fe/Mn$  in lavas that undergo clinopyroxene fractionation. Consequently, the elevated  
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1317 557  $Fe/Mn$  in the MPLF lavas that crystallized at the highest pressures (Fig. 9B) may solely be  
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1319 558 due to clinopyroxene fractionation and not because they were derived from pyroxenite.  
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1321 559 Furthermore, the lavas shown in Fig. 9B contain olivine which carry a lower pressure  
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1323 560 crystallization signature than those in Fig. 9A, and also have  $Fe/Mn < 70$ . Low and high  $SiO_2$   
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1325 561 pyroxenite-derived lavas from Hawaii (HSDP-2) contain olivines with  $Fe/Mn = 73.8 \pm 1.7$  and  
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1327 562 up to a maximum of 81 all of which are higher than the MPLF samples again suggesting that  
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1329 563 pyroxenite was not an important component of the course of MPLF lavas.

## 1330 1331 564 **5. Olivine in magmas from Iceland, Baffin Island and west Greenland**

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1333 565 The distribution of Ni in olivines in lavas from Thesitareykir (Iceland), Baffin Island and  
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1335 566 Disko Island (west of Greenland) were discussed in detail by Herzberg et al. (2016). A  
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1337 567 feature of olivines within lavas from Theistareykir are the existence of two populations, one  
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1339 568 primitive population crystallizing close to the 1 atm CLD and the other falling on mixing  
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1341 569 lines with derivative magmas along the CLD (Herzberg et al., 2016). However, the two  
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1343 570 populations of olivines are mutually exclusive with respect to their host lavas. When the same  
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1345 571 data are considered in terms of Ni-Ca covariations (Fig. 10A), olivines that exhibit evidence  
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1347 572 of an origin by magma mixing in terms of Fo-Ni correlations, have lower CaO contents over  
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1349 573 the same range of Ni contents than the more primitive olivines that fall close to the 1 atm  
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1360 574 CLD. Simple magma mixing cannot account for these variations because mixing lines for  
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1362 575 reasonable MgO contents of derivative magmas ( $\geq 7$  wt% MgO) do not intersect both groups  
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1364 576 of olivines. Indeed, mixing between a primary magma formed at depth with magmas formed  
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1366 577 at progressively shallower levels would necessarily fall along the predicted pathway for the  
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1368 578 Herzberg and O'Hara (2002) Ni-Ca partitioning model (Fig. 9). Similarly, mixing between  
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1370 579 primitive or near primary magmas (those which would crystallize Matzen olivines) and  
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1372 580 derivative magmas close to the surface would follow the predicted pathway for the Beattie et  
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1374 581 al. (1991) Ni-Ca partitioning model.

1377 582 Inspection of Fig. 7 shows that for BPIP lavas crystallizing only olivine, Ca contents of  
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1379 583 olivines should be lower for crystallization occurring at higher pressures compared to those at  
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1381 584 lower pressures. MacLennan et al. (2003) showed that some lavas at Theistareykir  
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1383 585 crystallized at  $>0.8$  GPa and showed evidence of crystallization along the Ol+Cpx+L  
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1385 586 cotectic. So, rather than being a result of mixing it is more likely the low CaO olivines found  
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1387 587 in some Theistareykir lavas are those which crystallized at  $\sim 1.0$  GPa. The variation in  
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1389 588  $D^{Ol/L}_{NiO}$  because of fractionation at higher T (Matzen et al. 2017) would not be great enough  
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1391 589 to cause any discernible variation in Ni content between high and lower pressure olivines.  
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1393 590 Nevertheless,  $K_D^{Ol/L}_{(FeO/MgO)}$  would be higher ( $\sim 0.310$ ) for the higher pressure olivines than  
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1395 591 those which crystallized at 1 atm ( $\sim 0.298$ ) thus resulting in differences in distribution of the  
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1397 592 data in terms of Ni-Fo covariations. Also, because crystallization along the Ol+Cpx+L  
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1399 593 cotectic results in a smaller bulk mineral-melt distribution coefficient for Ni than for olivine  
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1401 594 crystallization alone, liquid compositions that have fractionated Ol and Cpx have higher Ni  
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1403 595 contents than for the same liquid fractionating only olivine, resulting in higher Ni contents in  
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1405 596 olivine for a given Fo content, as is observed at Theistareykir.

1409 597 Covariations between Ca and Ni in data for olivines from Baffin Island (Fig. 10B)  
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1411 598 follow the predicted CLDs defined by the Beattie et al. (1991) and Gavrilenko et al. (2016)  
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1419 599 models and the data therefore follow trends that are consistent with crystallization at or near  
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1421 600 the surface. The most Ni-rich and Ca-poor olivines overlap with the composition of Matzen  
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1423 601 olivines calculated to be in equilibrium with PRIMELT3 primary magmas. Consequently,  
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1425 602 some of the olivines in Baffin Island lavas can be considered to be truly primary and  
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1427 603 crystallized after rapid emplacement of magma close to the surface. Importantly, there is no  
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1429 604 evidence for crystallization along the Ol+Cpx+L cotectic, the projection of which falls well  
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1431 605 below the observed data in Fig. 10B. Olivines in lavas from Disko Island (Fig. 10C) have  
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1433 606 systematically lower Ca contents at a given Fo content compared to the values predicted by  
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1435 607 the Gavrilenko et al. (2016) model for crystallization at 1 atm, and lower Ca contents than  
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1437 608 olivines from Baffin Island with similar Fo contents. Hole and Millett (2016) showed that at  
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1439 609 Baffin Island  $P_f \sim 2.1$  GPa whereas at Disko island  $P_f \sim 2.8$  GPa which most likely reflects a  
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1441 610 deeper asthenosphere-lithosphere boundary at Disko Island Baffin Island (Matzen et al.,  
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1443 611 2017). One possibility is that Disko Island magmas underwent minor clinopyroxene  
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1445 612 fractionation in the mantle before being emplaced, whereas the Baffin Island magmas only  
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1447 613 crystallized olivine and underwent rapid emplacement to shallow levels. Alternatively, the  
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1449 614 low Ca and high Ni contents of some Disko Island olivines could be interpreted as a result of  
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1451 615 their derivation from a low CaO pyroxenite source. However, the involvement of pyroxenite  
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1453 616 cannot be substantiated by the Fe/Mn for Disko Island olivines (Fig. 9). Consequently, the  
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1455 617 observed differences between Baffin and Disko island olivines might relate to the differences  
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1457 618 in magmatic plumbing at the two locations rather than to source heterogeneities.  
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## 1463 619 **6. Clinopyroxene stability during polybaric fractionation**

1466 620 MPLF and SMLS lavas do not contain augite phenocrysts, only olivine. Augite tends  
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1468 621 to form as groundmass poikilitic anhedral in sparsely olivine-phyric lavas regardless of Mg#  
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1470 622 (Thompson, 1982; Bailey et al., 1923). Nevertheless, there appears to be ample evidence to  
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623 suggest that augite was an important crystallizing phase during the evolution of the BPIP  
624 plateau lavas, which sets them apart from other NAIP lavas. On a regional scale, there were  
625 large volumes of pyroxene gabbro intruded throughout the lifetime of the BPIP. The Ben  
626 Buie gabbro complex on Mull forms part of the intrusive complex beneath the Mull volcano  
627 and is considered to be a product of the crystallization of a tholeiitic magma (Thompson  
628 1982).

629         The estimates of ~1.0 GPa crystallization pressure of some of the more evolved lavas  
630 along the Ol+Cpx+Pl+L cotectic, coupled with the high pressure Ol+L or Ol+Cpx+L  
631 crystallization trends evident in the olivine data, strongly suggests that polybaric fractional  
632 crystallization was important in the BPIP, as was originally postulated by Thompson *et al.*  
633 (1980) and developed further by other workers (e.g. Kerr *et al.*, 1999; Fowler *et al.*, 2003;  
634 Font *et al.*, 2008; Hole *et al.*, 2015a). Herzberg and Asimow (2008) noted that deep  
635 crystallization of high CaO augite can have the same effect as shallow augite fractionation on  
636 the CaO contents of high MgO primary magmas, and Keshav *et al.* (2007) argued that  
637 clinopyroxene can accumulate in the mantle and evidence of its existence is found in mantle  
638 xenoliths. Consequently, the Ni-Ca relationships shown in Fig. 8 for MPLF lavas could  
639 result from initial crystallization of augite at mantle depths, close to the top of the melting  
640 column (~2.7 GPa) in the region. The distribution of the most Fo-rich olivines (Fo~92) from  
641 Mull in Fig. 8B, and the fact that they have lower Ca content but the same Ni contents as  
642 Matzen olivine compositions suggests that mantle augite accumulation is a possibility.  
643 Furthermore, olivines in Skye picrites exhibit Ni-Ca covariations that are suggestive of  
644 limited or no mantle augite accumulation (Fig. 8B). As discussed above, the sub-horizontal  
645 arrays delineated by the MPLF lavas in Fig. 8B might be a result of slower diffusion of Ca  
646 relative to Ni in olivine (Coggan *et al.*, 2005; Chakraborty, 2010) and the Ca contents of these  
647 olivines reflect the accumulation of augite in the mantle.

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1537 648 An alternative explanation for the lack of early-formed augite phenocrysts in MPLF  
1538  
1539 649 lavas relates to differences in clinopyroxene and olivine stability in primitive melts.  
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1541 650 Sugawara (2000) and Chen and Zhang (2008; 2009) provide empirical relationships between  
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1543 651 pressure, temperature and the MgO and CaO content of olivine and pyroxene saturated  
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1545 652 liquids. The Sugarawa (2000) parameterizations show that for a given melt MgO content and  
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1547 653 pressure, augite saturation occurs at ~50°C lower temperature than olivine saturation. The  
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1549 654 Chen and Zhang (2008; 2009) studies relate to the dissolution of mantle clinopyroxene  
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1551 655 (diopside  $\text{Wo}_{49.5}\text{En}_{48.5}\text{Fs}_{2.0}$ ) in basaltic melt. It is appreciated that extrapolating such a study  
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1553 656 to pyroxenes with compositions of around  $\text{Wo}_{45}\text{En}_{41}\text{Fs}_{14}$  such as those found in mafic BPIP  
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1555 657 magmas (Hole et al. 2015a) may not be strictly valid, but these are the most applicable  
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1557 658 parameterizations in the literature. Chen and Zhang (2009) showed that saturation of Mg-  
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1559 659 and Ca-rich clinopyroxene may be determined by the three major components MgO, CaO and  
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1561 660  $\text{SiO}_2$ . Chen and Zhang (2009) developed a relationship between  $C_0^{\text{MgO}} \times C_0^{\text{CaO}}$ , pressure and  
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1563 661 temperature to define clinopyroxene stability in a basaltic melt, where  $C_0^{\text{MgO}}$   $C_0^{\text{CaO}}$  are the  
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1565 662 concentrations of MgO and CaO in a liquid at the point of saturation. Fig. 11 is a plot of CaO  
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1567 663 *versus* MgO for the MPLF whole-rock samples from which olivine data were derived plotted  
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1569 664 alongside clinopyroxene and olivine saturation curves calculated from Chen and Zhang  
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1571 665 (2008; 2009). Here it is assumed that polybaric fractional crystallization took place in two  
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1573 666 distinct phases; firstly at 1.5 GPa with Cpx joining the liquidus at ~1390°C (the Cpx-in  
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1575 667 temperature for SK982 at 1.5 GPa) and secondly at 1.0 GPa with an accompanying fall in  
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1577 668 temperature to 1365°C (~55°C/GPa<sup>-1</sup>; Sugawara, 2000; Matzen et al., 2017). Olivine  
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1579 669 saturation is independent of CaO content of the liquid, and over the temperature interval in  
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1581 670 question, olivine saturation is little effected. However, augite saturation occurs at  
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1583 671 considerably higher CaO content for a given MgO at 1.0 GPa compared to its saturation at  
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1585 672 1.5 GPa. The whole-rock analyses of MPLF lavas fall close to the join the between olivine  
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1596 673 and clinopyroxene saturation curves at 1.5 GPa. However, at 1.0 GPa the same liquid  
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1598 674 composition is saturated with respect to olivine, but not with respect to clinopyroxene.  
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1600 675 Consequently, is it possible during polybaric fractional crystallization under the range of  
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1602 676 pressure conditions applicable to the MPLF, to render clinopyroxene unstable. Consequently,  
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1604 677 the lack of observed phenocrysts of augite in MPLF lavas might result from their dissolution  
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1606 678 during magma storage and fractionation at  $\leq 1.0$  GPa.  
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## 1610 679 **7. Predicting pyroxenite in the source of continental flood basalts**

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1612 680 Olivines from low SiO<sub>2</sub>-pyroxenite-derived magmas from Mauna Loa (HSDP-2) have Ni  
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1614 681 contents of ~4000 ppm at Fo ~90 whereas high SiO<sub>2</sub> pyroxenite derived magmas from  
1615  
1616 682 Koolau (Makapuu stage) have even more extreme Ni contents of up to 4660 ppm at Fo ~89  
1617  
1618 683 (Herzberg, 2006; 2011; Sobolev et al., 2005). Both of these Ni contents are in excess of the  
1619  
1620 684 highest Ni Matzen olivines from Disko Island which have ~3600 ppm Ni at Fo ~91.5. Since  
1621  
1622 685 the Matzen olivines from West Greenland represent the highest possible Ni contents  
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1624 686 attainable for olivines in the NAIP, the values of up to 3000 ppm Ni at Fo ~90 for MPLF  
1625  
1626 687 lavas does not require a high Ni pyroxenite source. Additionally, CaO contents of low and  
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1628 688 high SiO<sub>2</sub> pyroxenite-derived magmas are <2000 and <1700 ppm respectively which are  
1629  
1630 689 lower than the most CaO-poor MPLF lavas (Figs 5 & 6).  
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1633 690 Olivines from mafic magmas in some Karoo LIP lavas (e.g. N356 of Sobolev et al.,  
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1635 691 2007) which have whole-rock with ~6wt% CaO at ~15wt% MgO (Ellam and Cox 1989)  
1636  
1637 692 exhibit extreme depletions in Ca (100-1400 ppm), enrichment in Ni (up to 4500 ppm at Fo  
1638  
1639 693 88) and have Fe/Mn 75-82. However, sample N356 comes from the Lebombo monocline and  
1640  
1641 694 is one of a suite of high TiO<sub>2</sub> lavas with  $\epsilon\text{Nd}_{190} = -4$  to  $-10$  and  $\epsilon\text{Hf}_{190} = -3$  to  $-11$  and is likely  
1642  
1643 695 to be a sample of Proterozoic lithosphere (Ellam and Cox, 1989; 1992; Ellam et al., 1992;  
1644  
1645 696 Ellam, 2006) and not necessarily recycled oceanic crust in the sense of Sobolev (2005) and  
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1647 697 Herzberg (2006). Extreme depletions in CaO (~8 wt% at MgO ~9.5 wt%) in Si-oversaturated  
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698 (SiO<sub>2</sub> ~54 wt%) whole rock samples from part of the Karoo LIP in the Falkland Islands were  
699 also reported by Hole et al. (2015b) and these too have εNd<sub>190</sub> as low as -11, although they  
700 are olivine-free. It is beyond the scope of the current study to discuss the role of pyroxenite  
701 in continental LIPs, but a picture is emerging that the continental lithosphere might also be a  
702 source of pyroxenite which contributes to CFB magmatism. Data for olivines in lavas from  
703 the Siberian Traps (Sobolev et al. 2007; 2009) have Ni contents of ~3200ppm at Fo<sub>83.0</sub>  
704 which would project to higher Ni contents than those of Matzen olivines (~3200ppm at  
705 Fo<sub>91.8</sub>) calculated from PRIMELT3 solutions at the same Fo content, regardless of pressure of  
706 crystallization. These same olivine phenocrysts contain <2000 ppm Ca. Consequently, the  
707 olivines must have crystallized from partial melt of pyroxenite, and not from mantle  
708 peridotite (Sobolev et al. 2009).

709         Given the intricacies of interpreting the whole-rock and olivine record of fractionation  
710 of magmas in the crust, it is not surprising that the positive identification of pyroxenite  
711 involvement in magmagenesis at LIPs using major elements is highly problematical. It is  
712 quite clear that any attempts to utilize CaO content of lavas to identify pyroxenite sources  
713 will be ambiguous in their outcomes. Perhaps a step closer can be made by considering the  
714 fractionation of basaltic magmas at varying pressures without utilizing CaO abundances. Fig.  
715 12 shows SiO<sub>2</sub>/MgO versus Mg# for basalts from Hawaii (HSDP-2), the MPLF and  
716 Theistareykir. LLDs at 1 atm have been calculated from PRIMELT3 primary magmas using  
717 Petrolog3, including for HSDP samples, a few of which are derived from mantle peridotite  
718 (Herzberg and Gazel 2009). Because Cpx is saturated with respect to silica, when it joins the  
719 liquidus it fractionates SiO<sub>2</sub> and MgO relative to one another at a different rate from olivine  
720 crystallization resulting in infections on the LLD as seen in Fig. 12. Data for HSDP lavas  
721 mostly fall on the 1 atm LLD implying olivine crystallization alone is responsible for their  
722 evolution. The majority of HSDP samples have lower CaO for a given MgO than would be

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1714 723 expected for olivine crystallization alone and consequently the source from which they were  
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1716 724 derived must be similarly CaO deficient indicating a role for pyroxenite which is also  
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1718 725 consistent with olivine trace element data (e.g. Sobolev et al., 2005; Herzberg, 2006). By  
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1720 726 contrast, MPLF lavas show significant deviations from the 1 atm LLD as a result of  
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1722 727 crystallization of Ol+Cpx. Consequently, the CaO deficiency of these lavas need not be  
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1724 728 attributed to the involvement of pyroxenite, which the olivine trace element data also  
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1726 729 indicates. For Theistareykir, the majority of lavas follow the 1 atm LLD with some deviation  
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1728 730 at Mg# <65 towards lower SiO<sub>2</sub>/MgO than the 1 atm LLD, which is most likely a  
1729  
1730 731 consequence of the crystallization of Ol+Cpx. Crystallization of Ol+Cpx at ~0.8 GPa is  
1731  
1732 732 consistent with the melt inclusion studies of MacLennan et al., (2003) as well as the olivine  
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1734 733 trace element data discussed above. So, whereas the representation of data in Fig. 12 cannot  
1735  
1736 734 be used to identify pyroxenite in the source of LIPs, it can be used to identify lavas that have  
1737  
1738 735 crystallized Cpx+Ol at depth, without the use of CaO data, which therefore removes some  
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1740 736 ambiguity from the interpretation of MgO and CaO data for continental flood basalts.  
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## 1745 737 **8. The NAIP – a peridotite-dominated LIP**

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1747 738 It is generally accepted that many of the shield-building lavas of Hawaii are likely to have  
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1749 739 been derived from melting of a pyroxenite-rich source lithology (e.g. Herzberg 2006; 2011;  
1750  
1751 740 Sobolev et al., 2005; 2007). This pyroxenite is thought to be derived from the recycling of  
1752  
1753 741 oceanic crust at depth and was subsequently entrained in a mantle plume. The  
1754  
1755 742 transformations that oceanic crust may undergo during recycling are complex and involve  
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1757 743 rock-melt, melt-melt and rock-rock interactions (e.g. Herzberg, 2011; Lambart et al., 2016;  
1758  
1759 744 Lambart, 2017). Sobolev et al. (2005; 2007) argued that the rising mantle plume beneath  
1760  
1761 745 Hawaii contains eclogite bodies that start melting at about 190–180km depth. Melting of  
1762  
1763 746 eclogite produces high SiO<sub>2</sub> initial melts which infiltrate into, and react with, the adjacent  
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1765 747 peridotite eliminating olivine and producing a solid pyroxenite. Both the reaction pyroxenite  
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748 and unreacted peridotite melt at depths between 140 and 100 km, producing hybrid magmas  
749 by mixing in conduits and crustal magma chambers. Nevertheless, data from Mauna Kea  
750 reveal that low- and high-SiO<sub>2</sub> pyroxenite-sourced magmas coexist as discrete batches along  
751 with peridotite-derived magmas (Rhodes et al., 2012; Sobolev et al., 2005; 2007) leading  
752 Herzberg (2011) to promote the hypothesis that melts of pure pyroxenite may segregate from  
753 their source without mixing and be erupted at the surface as lavas.

754 In the case of LIPs formed above thick continental lithosphere (e.g. Siberian Traps and  
755 parts of the NAIP) the composition of magmas should be almost exclusively controlled by the  
756 melting of reaction pyroxenite because thick lithosphere restricts the amount of melt that can  
757 be derived from mantle peridotite (Sobolev et al., 2005; 2007; 2009; Hole and Millett, 2016).  
758 The model proposed by Sobolev et al. (2005; 2009) for the petrogenesis of lavas of the  
759 Siberian Traps relies on the fact that SiO<sub>2</sub> rich olivine-free pyroxenite (that which plots to the  
760 right of the Cpx-CaTs-Opx plane) has a lower solidus temperature at a given pressure than  
761 peridotite (Sobolev et al., 2005; Lambart et al., 2012; 2016; Lambart 2017) and consequently  
762 for a given T<sub>p</sub>, pyroxenite will melt at a higher pressure than peridotite. On this basis,  
763 Sobolev et al. (2009) concluded that early tholeiites from the Gudchikhinsky Formation of  
764 the Siberian Traps were 100% pyroxenite melts with the contribution from pyroxenite  
765 decreasing to 40-50% (Tuklonsky and Nadezhdinsky formations) as peridotite was entrained  
766 in the melting zone. Sobolev et al. (2009) further argued that differences in the compositions  
767 of lavas from the Hawaii and Siberian LIPs was simply a function of lithospheric thickness  
768 and not the amount of recycled material present in the mantle, implying that all mantle  
769 plumes could have, and should have, a similar petrological structure.

770 Matzen et al. (2017) provided estimates of 100 and 32 km for the depths to the LAB  
771 beneath Disko Island and Iceland rift zones respectively. Hole and Millett (2016) showed  
772 that P<sub>f</sub> was ~2.7 GPa for BPIP magmas and was not significantly different from that for

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773 Disko Island, West Greenland ( $P_f \sim 2.8$  GPa) suggesting a similar depth to the LAB in both  
774 areas. A pyroxenite melting signature would therefore be expected to be most evident in lavas  
775 of the MPLF and Disko Island (Fig. 13). However, it is apparent that none of the NAIP lavas  
776 under consideration here provide any evidence of melting of 100% pyroxenite, or indeed any  
777 significant proportion of pyroxenite, once the effect of high pressure fractionation on major  
778 element compositions is accounted for. In the case of Iceland, existing studies provide  
779 estimates for the overall contribution from pyroxenite which vary from <5% (Brown and  
780 Leshner, 2014) through 10% (Sobolev et al., 2007; Lambart, 2017) to 30% (Matthews et al.,  
781 2016). Lambart (2017) showed that the observed crust thickness of the Icelandic rift zones is  
782 consistent with about 10% of recycled crust in the form of a *low* SiO<sub>2</sub> pyroxenite lithology  
783 and that diversity of trace element and isotopic compositions observed in Icelandic rift zone  
784 basalts does not require a contribution from melts derived directly from a recycled basalt  
785 component, but results from the melting of hybrid olivine-bearing lithologies formed by solid  
786 state reactions between recycled crust and peridotite (Lambart, 2017).

787 Low SiO<sub>2</sub> olivine pyroxenites (those which plot to the left of the Cpx-CaTs-Opx plane)  
788 melt under P-T conditions that are not much different from that of peridotite but are  
789 characterized by higher melt productivity than peridotite at the same temperature and  
790 pressure (e.g. Lambart et al., 2016; Lambart, 2017). A contribution from low SiO<sub>2</sub> pyroxenite  
791 is therefore most likely to take the form of a hybrid lithology formed by solid state reactions  
792 (Lambart, 2017) and the resultant melt may not have an identifiable ‘pyroxenite signature’.  
793 Melting experiments at 2.0 and 2.5 GPa on low SiO<sub>2</sub> pyroxenite SL77-582 (Keshav et al.,  
794 2004) show that large extents of melting (>70%) of pyroxenite can produce magmas with  
795 whole-rock compositions that are similar to peridotite-derived magmas, and in particular,  
796 such melts occupy the same position as some peridotite-derived melts in terms of CaO and  
797 MgO contents (Fig. 2). Lambart (2017) further showed that most of the variations in Ni



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798 content of olivine in Icelandic lavas reported by Sobolev et al. (2007) and Shorttle and  
799 Maclennan (2011) can be explained by varying the sampling pressure of the aggregated melts  
800 in the melting column, with the compositions richest in Ni sampled at the highest pressure  
801 where the contribution of pyroxenite is the largest. However, modelling carried out during  
802 the current study does not require a pyroxenite source to explain the Ni-Ca-Fo relationships  
803 of Icelandic basalts, but merely requires fractionation of olivines at varying pressures.  
804 Nevertheless, a contribution from a pyroxenite lithology is not necessarily precluded on the  
805 basis of Ni contents of olivines in Icelandic basalts.

806 Fig. 13 shows the predicted CLDs for Matzen olivines with 0.365 and 0.600 wt% NiO  
807 which are in equilibrium with experimental melt #370 (Keshav et al., 2004) which itself  
808 represents 70% melt of low SiO<sub>2</sub> pyroxenite SL77-582 at 2.0 GPa and 1385°C. Herzberg et  
809 al. (2016) showed that olivine with 0.365 wt% NiO was characteristic of high-precision  
810 analysis of olivines having an Mg number of 89.5 that represent near solidus melts of mantle  
811 peridotite. Pyroxenite melts, and their near solidus olivines, have considerably higher NiO  
812 than melts from peridotite (e.g. Sobolev et al., 2007) and a value of 0.600 wt% NiO in near  
813 solidus pyroxenite olivine has been chosen here for illustrative purposes. The CLD for 0.365  
814 wt% NiO intersects the data for olivine from Theistareykir which crystallized at low  
815 pressures (~0 GPa) whereas the CLD for 0.600 wt% NiO intersects the data field for  
816 pyroxenite-derived magmas from Mauna Kea recovered during the Hawaiian Scientific  
817 Drilling Programme (HSDP-2; Sobolev et al., 2007; Herzberg, 2006). In addition, the 0.600  
818 wt% NiO CLD falls immediately below the field for olivines in lavas from Koolau (Hawaii)  
819 which were derived by melting of a high SiO<sub>2</sub> pyroxenite. Reverse modelling from the  
820 Matzen olivines shown in Fig. 13 to whole-rock Ni contents gives primary magma  
821 compositions which contain 463ppm and 792ppm Ni for 0.365 and 0.600 wt% NiO in olivine  
822 respectively; the former is within the estimates for near-primary high SiO<sub>2</sub> pyroxenite-

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823 derived magmas given in Sobolev et al. (2005) and the latter is within the estimates for  
824 primary peridotite-derived magmas. Hybridization of melts derived from peridotite and  
825 pyroxenite must therefore fall between the 0.600 and 0.365 wt% NiO CLDs and this is indeed  
826 the region where data for all olivines from the NAIP are found. However, the Theistareykir  
827 olivines with the higher Ni for a given Fo content are also those which fractionated at ~1.0  
828 GPa (Fig. 10) and while their relatively elevated Ni content does not *require* an input from a  
829 pyroxenite-derived melt it does not preclude it. Consequently, there is no necessity to involve  
830 a significant proportion of pyroxenite in the petrogenesis of any NAIP lavas on the basis of  
831 the trace element content of olivine.

832 It is interesting to speculate on the fact that more than 320 lavas from the NAIP yield  
833 PRIMELT3 solutions for melting of dry peridotite (Hole and Millett 2016), which does not  
834 include peridotite-derived magmas that crystallized at elevated pressures, compared to ~18  
835 lavas from the Siberian Traps and ~14 lavas from Maua Loa and Mauna Kea, even though  
836 the data bases for each area are of a similar magnitude. This is probably also an indication of  
837 the dominance of peridotite-derived melt in the NAIP. Why then, if the NAIP was the site of  
838 a mantle plume that has been active from ~61 Ma until the present, do lavas show an  
839 overwhelming signature of peridotite melting? Variations in the depth to the LAB cannot  
840 provide a viable explanation which leads to the conclusion that pyroxenite was never a  
841 significant component in the source of the NAIP magmas, and where present, it had  
842 thoroughly hybridized with peridotite. If pyroxenite was not present in any significant  
843 volume, then this suggests that the NAIP did not have the same petrological structure as that  
844 envisaged for the commonly accepted mantle plume models such as those for Hawaii or the  
845 Siberian Traps. In this connection, it may be beneficial to consider alternative models for  
846 magmatism at LIPs which do not require large scale recycling of oceanic lithosphere (e.g.  
847 Foulger 2012; Anderson and Natland 2014).

2007  
2008  
2009 848 **9. Conclusions**  
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2012 849 Some plateau forming lavas of the BPIP have major element compositions that indicate they  
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2014 850 crystallized at <1 GPa with plagioclase joining the liquidus before clinopyroxene, the  
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2016 851 majority crystallized clinopyroxene before plagioclase feldspar over the pressure range 1.0-  
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2018 852 2.0 GPa. However, the majority crystallized olivine and clinopyroxene before plagioclase  
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2020 853 feldspar and therefore must have crystallized at >~1 GPa. Olivine within the plateau lavas  
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2022 854 have Ni and Ca contents that suggests that magmas paused at >1.6 GPa and crystallized  
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2024 855 Ol±Cpx at that depth. A few examples of near-primary magmas that underwent  
2025  
2026 856 crystallization of olivine near the surface are found on Skye and their component olivines can  
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2028 857 be successfully modelled using the method of Matzen et al. (2017) giving a maximum Ni  
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2030 858 content for BPIP olivine as 3600 ppm. MPLF lavas which fractionated Ol+Cpx in the crust  
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2032 859 exhibit a CaO deficiency for a given MgO compared to lavas from west of Greenland and  
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2034 860 Iceland which crystallized olivine alone near the surface. Low CaO high MgO in lavas is  
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2036 861 often considered to be a characteristic sign of the involvement of pyroxenite in the source of  
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2038 862 basalts. However, olivines within MPLF lavas have Fe/Mn <70 and Ni and Ca contents that  
2039  
2040 863 are entirely consistent with and origin solely from mantle peridotite. Therefore, apparent CaO  
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2042 864 deficiency in these lavas results from augite crystallization within the crust. The absence of  
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2044 865 augite phenocrysts in lavas may be a consequence of change from augite saturation to under-  
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2046 866 saturation with decreasing pressure, and consequent augite dissolution. The commonly  
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2048 867 accepted petrological structure of mantle plumes involving entrainment of recycled oceanic  
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2050 868 crust and significant contributions to magmatism from pyroxenite-rich sources cannot be  
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2052 869 applied to the flood basalts of the NAIP which is dominated by peridotite-derived melts.  
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2058 871 **Acknowledgements.**  
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2541 1072 **Figure Captions.**  
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2543 1073 Fig. 1. (A) Reconstruction of the North Atlantic region at about 65Ma showing the locations  
2544 referred to in the text. Pecked lines labelled A6 etc., are seafloor magnetic anomalies. Black dots  
2545 1074 numbered 55-70 are plume-head positions at the time indicated and are taken from Lawver and  
2546 1075 Müller (1994). JMFZ, Jan Mayen Fracture Zone. After Hole and Millett (2016). (B) detail of the  
2547 British Pelaeocene Igneous Province showing the position of the main magmatic centres  
2548 1076 mentioned in the text and the main structural features of the region. After Hole et al. (2015a).  
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2553 1079 Fig. 2. Schematic representation of the pressure, temperature and composition ( $P, T, X$ )  
2554 relationships for the crystallization of a typical BPIP primary magma. (a) Phase relationships  
2555 1080 calculated using Petrolog3 for the primary magma to Skye lava SK982 (Thompson, 1982; Hole &  
2556 1081 Millett, 2016). The white dots are the approximate Fo content (Mg#) of olivine that crystallizes  
2557 1082 at the Ol+Cpx+L cotectic, and the grey dots the same for the Ol+Cpx+Pl+L cotectic. % figures on  
2558 the arrows are the approximate % of olivine that can crystallize on the Ol+L liquidus at a given  
2559 1083 pressure before reaching the Ol+Cpx+L cotectic. The olivine saturation curve was calculated  
2560 1084 using the method of Sugarawa (2000) with the final pressure of melting ( $P_f$ ) from PRIMELT3  
2561 solutions (Hole & Millett, 2016) acting as a proxy for the pressure of melt segregation.  
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2567 1088 Fig. 3 (A) CaO (wt%) *versus* Mg# and b) CaO (wt%) *versus* MgO (wt%) for and Mull Plateau  
2568 Lava Formation (MPLF) lavas (grey dots), Central Mull Formation (CMF; triangles) and Baffin  
2569 1089 Island lavas (white dots). The Baffin Island and CMF data shown yield PRIMELT3 solutions for  
2570 1090 melting of dry peridotite. Liquid lines of descent (LLD) were generated using Petrolog3  
2571 (Danyushevsky & Plechov, 2011) using the QFM buffer of Kress & Carmichael (1988) and the  
2572 1091 plagioclase and clinopyroxene melt liquidus associations of Danyuchevsky (2001). Black  
2573 1092 diamonds; Cpx-in at the pressure indicated; grey diamonds, Plag-in. The LLD shown is for the  
2574 PRIMELT3 solution for Syke lava SK982 (Thompson, 1982) which has the following  
2575 1093 characteristics;  $Fe^{2+}/Fe_T=0.905$  ( $Fe_2O_3/TiO_2=1.0$ );  $T_p=1500^\circ C$ ;  $MgO=17.4$  wt%;  $P_f=3.5$  GPa;  
2576 1094  $P_f=2.9$  GPa;  $F-AFM=0.17$  (Hole & Millett, 2016). In a) note that for pressures  $<0.6$  GPa the  
2577 crystallization order is  $Ol+L \rightarrow Ol+Pl+L \rightarrow Ol+Pl+Cpx+L$  whereas for pressures  $>0.6$  GPa the  
2578 1095 order is  $Ol+L \rightarrow Ol+Cpx+L \rightarrow Ol+Cpx+Pl+L$ . In (B) the dividing lines between pyroxenite- and  
2579 1096 peridotite-derived magmas and the range of possible peridotite-derived primary magmas (fine  
2580 pecked lines) are taken from Herzberg & Asimow (2008). The black dots in each diagram are  
2581 1097 the samples for which high-precision olivine trace element data is available (Sobolev *et al.*  
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1104 Fig. 4. (A)  $K_D^{Ol/L}_{(FeO/MgO)}$  versus pressure for three BPIP model primary magmas calculated using  
1105 Petrolog3 for the thermodynamical model of Toplis (2005). MgO contents of the model primary  
1106 magmas were derived from PRIMELT3 the final pressure of melting ( $P_f$ ) being used as a proxy  
1107 for the pressure of melt segregation PRIMELT3 solutions for S006 (16.0 wt% MgO) and SK982  
1108 (17.5 wt% MgO) are given in Hole & Millett (2016) and for MU1.1 (20.0 wt% MgO) in the  
1109 electronic appendix. (B) variation in  $K_D^{Ol/L}_{(FeO/MgO)}$  and Mg/Fe<sub>Liq</sub> (cationic Mg/Fe of the liquid)  
1110 during crystallization of the model primary magma to SK982 for the Toplis (2005) model at  
1111 fixed pressures from 0.0 to 2.0 GPa (solid lines). Also shown is the parameterization of Herzberg  
1112 & O'Hara (2002) SK982 which assumes magma ascent and crystallization along the olivine  
1113 liquidus from the pressure of melt segregation to the surface. The grey diamonds represent the  
1114 intersections of the Toplis (2005) contours for a fixed pressure with the Herzberg & O'Hara  
1115 (2002) ascent path – see text for details. The pecked line is the approximate position of Cpx-in,  
1116 with the vertical shading on the Ol+Cpx+L side of the cotectic.

1117 Fig. 5 (A) Calculated Fo and Ca content for crystal lines of descent (CLD) for equilibrium olivines  
1118 crystallizing from two BPIP model primary magmas (SK982 and MU1.1) at 1atm pressure for  
1119 the models given in equations (8)-(11) in the text. Crystallization models were calculated using  
1120 Petrolog3 with the following parameters; QFM Buffer of Kress and Carmichael (1988);  
1121 plagioclase and clinopyroxene equilibria of Danyushevsky (2001). Crosses are at 1%  
1122 crystallization intervals. The composition of 49 olivines in equilibrium with model BPIP primary  
1123 magmas (Hole and Millett 2016) are also shown for each calculation method. (B) CLDs for the  
1124 model primary magma to SK982 at variable pressures for; grey lines, Herzberg and O'Hara  
1125 (2002); black lines, Gevrienko et al. (2016) calculation methods (equations (9)-(11) in the text).  
1126 Pressure in GPa is indicated for each CLD. Grey squares, points at which clinopyroxene joins the  
1127 crystallizing assemblage for each pressure. SQFZ, Siqueiros Fracture Zone MORB measured  
1128 olivine compositions (filled triangles) from Gavrienko et al. (2016) and compositions calculated  
1129 (open triangles) using the Gavrienko et al. (2016) model. Vectors relating to various  
1130 petrological parameters are from Gavrienko et al. (2016). The line with dot terminations is the  
1131 approximate uncertainty in Ca content propagated from equation (10) in the text.

1132 Fig. 6. (A) & (B) Ni content (ppm) versus Fo content for olivines in MPLF lavas. The CLDs shown  
1133 were calculated for the primary magma to SK982 and are for; (i) the Beattie *et al.* (1991) model  
1134 at 1 atm (short dashes line); (ii) the Herzberg & O'Hara (2002) model for crystallization and  
1135 ascent along the olivine liquidus (long dashes) and (iii) the Herzberg & O'Hara (2002) model  
1136 until 1.6 GPa and then at a fixed pressure of 1.6 GPa (solid back line). For model (iii)  
1137  $K_D^{Ol/L}_{(FeO/MgO)}$  values were calculated from Mg/Fe<sub>Liq</sub> as shown in Fig. 2(A), propagating to give  
1138  $D^{Ol/L}_{NiO}$  from equation (3) in the text. The large white diamond represents the Fo content for

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1139 Cpx-in, and at this point  $D^{Cpx/L}_{NiO}$  was assumed to be  $0.25 D^{Ol/L}_{NiO}$ . Grey curves are mixing lines  
1140 between the primary magma composition and derivative magmas along the CLD (see also  
1141 Herzberg *et al.*, 2016) with the approximate MgO content of the derivative magma indicated. It  
1142 was assumed that mixing was complete and that neither liquid contained phenocrysts at the  
1143 time of mixing. 'Matzen olivines' are model olivine compositions for BPIP primary magmas  
1144 segregating at  $P_f$ , undergoing rapid emplacement at the surface without crystallizing any  
1145 olivine, and then crystallizing olivine at 1 atm (equation (7) in the text. The cross-hatched field  
1146 in (a) is for olivines from Siqueiros Fracture Zone MORB (Sobolev *et al.* 2007; Gavrilenko *et al.*  
1147 2016) and is shown for comparison. In (b) Skye and Mull olivines are near-primary olivines in  
1148 picrites (Spice *et al.*, 2016). These include sample MU1.1 for which a Toplis model is shown in  
1149 Fig. 2.

1150 Fig. 7. (A) and (B) Ca content (ppm) *versus* Fo content for olivines for the same MPLF lavas as in  
1151 Fig. 6. The cross-hatched area delimits the range of Ca and Fo contents of olivines for 49  
1152 forward crystallization models of BPIP primary magmas at 1 atm using the Beattie *et al.* (1991)  
1153 parameterization for  $D^{Ol/L}_{CaO}$  given in equation (4) in the text. The stippled field uses the same  
1154 primary magma data but for the Gavrilenko *et al.* (2016) method of calculating  $D^{Ol/L}_{CaO}$  given in  
1155 equations (10) and (11) in the text. Grey squares are the composition at which clinopyroxene  
1156 joins the crystallizing assemblage. The field labelled SQFZ encompasses measured olivine  
1157 compositions for Siqueiros Fracture Zone MORB from Gavrilenko *et al.* (2016)

1158 Fig. 8. (A) calculated CLDs for Ca and Ni content of olivines at variable pressures using the Ni  
1159 and Ca model of Herzberg and O'Hara (2002) and the Ca model of Gavrilenko *et al.* (2016). The  
1160 Ni model has been modified for appropriate final pressure of equilibration as discussed in the  
1161 text and shown in Fig. 3. The black lines with arrows are for the Herzberg and O'Hara (2002) Ca  
1162 model at the pressures indicated in GPa. Fine lines with various ornaments are for the Ni  
1163 model of Herzberg and O'Hara (2002) with Ca calculated according to the Gavrilenko *et al.*  
1164 (2016) model, and the thick grey line is for 1 atm pressure, which is the same as the olivine. Grey  
1165 squares are the compositions at which clinopyroxene joins the crystallizing assemblage for a  
1166 given pressure and the white squares the same for plagioclase feldspar. The curved inflections  
1167 giving negative 'spikes' for pressures  $>1.2$  GPa result from clinopyroxene joining the liquidus  
1168 before 11 wt% MgO, the MgO content at which the calculation  $D^{CaO}_{Ol/L}$  changes from equations  
1169 (10) to (11) in the text. Matzen olivines are for olivine in equilibrium with 49 BPIP model  
1170 primary magmas. The double-headed arrow illustrates the effect of crystallization of  $\sim 15\%$   
1171 olivine at 1 atm. (B) and (C) Ni and Ca contents of MPLF olivines. The cross hatched area  
1172 represents the range of compositions for forward crystallization of BPIP model primary  
1173 magmas at 1 atm and therefore represents the olivine liquidus. The thick grey line is the CLD for

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2717 1174 sample SK982 at 1 atm, and the lines labelled 'Beattie' and 'H&O' are 1atm CLDs for the Beattie  
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2719 1175 et al. (1991) and Herzberg and O'Hara (2002) calculation methods.  
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2721 1176 Fig. 9. Fe/Mn (ppm) *versus* Fo content for olivine in BPIP, Baffin Island and West Greenland  
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2723 1177 (Disko Island) lavas. The fields for primary olivines derived from melts of mantle peridotite are  
2724 1178 labelled with their source mineralogy i.e. harzburgite or lherzolite. The two pecked horizontal  
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2726 1179 lines (Fe/Mn 60 and 70) are the upper and lower boundaries respectively for olivines that  
2727 1180 crystallize from peridotite-derived magmas. After Herzberg (2011).  
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2729 1181  
2730 1182 Fig. 10. covariations between Ni and Ca content of lavas from (A) Theistareykir, Iceland, (B)  
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2732 1183 Baffin Island and (C) Disko Island, west Greenland. Stippled areas are the range of predicted Ni  
2733 1184 and Ca contents for the crystallization of model primary magmas (Hole and Millett 2016) at 1  
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2735 1185 atm from each location. The grey line on each diagram is the calculated Ol+L CLD at 1 atm for a  
2736 1186 model primary magma from each location. Model primary magmas are as follows; Disko Island,  
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2738 1187 #410188 (Larsen and Pedersen 2000); Baffin Is, BI/CS/14 (Starkey et al. 2009); Theistareykir,  
2739 1188 #9302 (MacLennan et al. 2003). In (c) grey symbols are olivines considered by Herzberg et al.  
2740  
2741 1189 (2016) to be near primary, and open circles are olivines considered by Herzberg et al. (2016) to  
2742 1190 be the result of crystallization from mixed magmas. The Ni content of Matzen olivines were  
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2744 1191 calculated from PRIMELT3 solutions using equation (7) and the Ca content using equation (10).  
2745 1192 The cross-hatched area is the field for olivine data from MPLF sample BHL15 for comparison  
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2747 1193 with Fig. 7. Data are from Sobolev *et al.* (2007) and Herzberg *et al.* (2016).  
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2749 1195 Fig. 11 Olivine (vertical lines) and clinopyroxene (curves) saturation calculated using the  
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2751 1196 method of Chen & Zhang (2008; 2009). To the left of the curves, liquids are undersaturated with  
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2753 1197 respect to olivine or clinopyroxene. At 1.5 GPa, the temperature estimate of 1390°C is the  
2754 1198 temperature estimate immediately prior to the Cpx-in temperature from a Petrolog3 forward  
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2756 1199 crystallization model for the primary magma to lava SK982. The temperature at 1.0 GPa  
2757 1200 assumes a 55°C/GPa<sup>-1</sup> gradient on the olivine liquidus (Sugawara 2000; Matzen et al. 2017).  
2758 1201 Black dots are the whole-rock compositions of the MPLF lavas for which olivine data are  
2759  
2760 1202 available. The shaded area is the region of instability of Cpx in the melt caused by  
2761  
2762 1203 decompression.  
2763 1204  
2764 1205 Fig. 12. SiO<sub>2</sub>/MgO versus Mg# for (A) Hawaii Scientific Drilling Project lavas, (B) MPLF lavas and  
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2766 1206 (C) lavas from Theistareykir. For each sample set forward crystallization models have been  
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2768 1207 generated at varying pressures from 0.0 to 1.8 GPa using Petrolog3, and starting with  
2769 1208 PRIMELT3 primary magma compositions. The shaded area on each diagram is the field for  
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2772 Hole, MJ. Polybaric fractionation CFB  
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1209 Ol+Cpx+L. The insets are CaO *versus* MgO for the same lavas, the black line indicating the  
1210 dividing line between peridotite- (above the line) and pyroxenite-derived magmas taken from  
1211 Herzberg & Asimow (2008). The CaO-MgO relationships for MPLF lavas are shown in Fig. 2.  
1212 Note the extended scale in (C).

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1214 Fig. 13. Ni (ppm) *versus* %Fo in olivine for basalts from Theistareykir, and pyroxenite-derived  
1215 lavas recovered during the Hawaii Scientific Drilling Project (HSDP-2) and from Koolau (shield  
1216 stage). Only olivine with Fo>86 are given in Sobolev et al. (2007) for HSDP-2. The open  
1217 diamond is the composition of Matzen olivine, assuming an NiO content 0.365 wt%, in  
1218 equilibrium with experimentally generated melt #370 of Keshav et al. (2004) which represents  
1219 70% melt of pyroxenite SL77-582. The whole-rock content of this magma would be ~460 ppm  
1220 Ni which is within the range for natural olivine in peridotite-derived magmas. Black diamond is  
1221 the composition of Matzen olivine, assuming and NiO content of 0.600 wt%, in equilibrium with  
1222 melt #370. In this case the whole-rock composition would be 792 ppm Ni which is within the  
1223 range of parental melts to Hawaiian pyroxenite-derived magmas (Sobolev et al., 2005). The  
1224 pecked lines represent the CLD at 1 atm from the Matzen olivine indicated.



**Abstract** Continental flood basalts undergo crystallization at a variety of pressures in the crust and sometimes even in the mantle. Polybaric fractionation, when magmas may pause and undergo crystallization at different pressures, results in complex fractionation of major elements. Crystallization at high pressures where clinopyroxene is an early crystallizing phase can result in erupted compositions that have major element characteristics which mimic those expected for melts derived from pyroxenite-rich sources. The trace element compositions of early-crystallizing olivine can add further detail to crystallization histories and potentially allow an examination of the crystallization of basalts from melt segregation to the surface. The North Atlantic Igneous Province (NAIP) comprises sub-regions which had diverse crystallization histories. Plateau basalts of the British Palaeogene Igneous Province (BPIP) were generated by partial melting of mantle peridotite starting at  $\sim 3.8$  GPa with melting ceasing by  $\sim 2.7$  GPa. Major elements indicate that some basalts crystallized at  $< 1$  GPa with plagioclase joining the liquidus before clinopyroxene. However, the majority of BPIP magmas crystallized clinopyroxene before plagioclase feldspar over the pressure range 1.0-2.0 GPa. Trace elements in olivine indicate crystallization of olivine + clinopyroxene over the pressure interval 1.6-2.0 GPa. However, olivine data also show that some near-primary magmas reached near-surface pressures without substantial modification by fractional crystallization. Olivines formed at  $\geq 1.6$  GPa have Ni and Ca contents that are consistent with an origin by partial melting of mantle peridotite with no role for pyroxenite being detected. This contrasts with the low pressure dominated crystallization histories exhibited by lavas from West Greenland and Iceland. Whole-rock data for many BPIP lavas exhibit CaO depletion at a given MgO content compared with those from West Greenland and Iceland, which might be cited as an indicator of pyroxenite in their source, but this is solely a consequence of augite fractionation at depth. An absence of augite phenocrysts in lavas may have resulted either from augite crystallization in the mantle or from a change

from augite saturation to under-saturation with decreasing pressure, and consequent augite dissolution. The lack of any significant contribution from recycled oceanic crust to magmatism in the NAIP suggests that the petrological structure of the mantle source from which NAIP lavas were derived does not conform with the generally accepted models for mantle plumes such as those responsible for magmatism in Hawaii and the Siberian Traps.

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4 1 **Mineralogical and geochemical evidence for polybaric fractional crystallization of**  
5 2 **continental flood basalts and implications for identification of peridotite and pyroxenite**  
6 3 **source lithologies.**  
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9 4 **M.J. Hole<sup>a</sup>**  
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14 8 **Abstract** Continental flood basalts undergo crystallization at a variety of pressures in the  
15 9 crust and sometimes even in the mantle. Polybaric fractionation, when magmas may pause  
16 10 and undergo crystallization at different pressures, results in complex fractionation of major  
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24 18 (BPIP) were generated by partial melting of mantle peridotite starting at ~3.8 GPa with  
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33 27 being detected. This contrasts with the low pressure dominated crystallization histories  
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62 28 exhibited by lavas from West Greenland and Iceland. Whole-rock data for many BPIP lavas  
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64 29 exhibit CaO depletion at a given MgO content compared with those from West Greenland  
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66 30 and Iceland, which might be cited as an indicator of pyroxenite in their source, but this is  
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68 31 solely a consequence of augite fractionation at depth. An absence of augite phenocrysts in  
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70 32 lavas may have resulted either from augite crystallization in the mantle or from a change  
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72 33 from augite saturation to under-saturation with decreasing pressure, and consequent augite  
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76 35 magmatism in the NAIP suggests that the petrological structure of the mantle source from  
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78 36 which NAIP lavas were derived does not conform with the generally accepted models for  
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80 37 mantle plumes such as those responsible for magmatism in Hawaii and the Siberian Traps.  
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## 84 38 **1. Introduction**

87 39 Continental flood basalts (CFB) must traverse continental lithosphere before eruption at  
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89 40 the surface. For the North Atlantic Igneous Province (NAIP; Fig. 1), where the mantle was  
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91 41 anomalously hot ( $T_p \sim 1450-1550^\circ\text{C}$ ; Herzberg and Gazel, 2009; Hole and Millett, 2016;  
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93 42 Matthews et al. 2016) melting of mantle peridotite was initiated at  $\sim 3.7-4.5$  GPa (Hole and  
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95 43 Millett, 2016; Hole, 2015) and melting ceased when magmas rising by adiabatic ascent along  
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97 44 the olivine liquidus reached the asthenosphere-lithosphere boundary (LAB), which is also the  
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99 45 final pressure of melting ( $P_f$ ). The depth to the LAB varies throughout the province from  
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101 46  $\sim 100$  km beneath Disko Island (west of Greenland) to  $\sim 60$  km beneath Baffin Island (Hole  
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103 47 and Millett, 2016; Matzen et al., 2017). During ascent from the initial melting pressure ( $P_i$ )  
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105 48 to the LAB, it is generally accepted that magmas crystallize olivine alone (e.g. Herzberg and  
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107 49 Asimow, 2008; 2015; Putirka 2008; 2011). However, Herzberg and Asimow (2008) state that  
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109 50 augite fractionation can occur in the mantle, an assertion that is corroborated by the  
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111 51 existence of clinopyroxene-bearing xenoliths in Hawaiian lavas that are consistent with  
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113 52 crystallization from a tholeiitic liquid at 110-150 km depth (Keshav et al., 2007). Magmas  
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121 53 may pause at the LAB or within the lithosphere prior to eruption, and in the British  
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123 54 Palaeogene Igneous Province (BPIP), which is a sub-province of the NAIP, there is a large  
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125 55 body of evidence that suggests that magmas paused at the Moho at about 30km depth, or  
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127 56 equivalent to  $\sim 0.9 \pm 0.15$  GPa (Thompson, 1974; 1982; Hole et al., 2015a; Millett et al., 2016).  
128  
129 57 Individual batches of magma may have undergone polybaric fractionation. Thompson (1974;  
130  
131 58 1982) on the basis of melting experiments, as well as major, trace element and isotopic  
132  
133 59 compositions of lavas, proposed that some mafic BPIP lavas underwent fractionation at 1.6-  
134  
135 60 1.7 GPa as well as at  $\sim 0.9$  GPa. Conversely, at Disko Island, very primitive and near primary  
136  
137 61 magmas were erupted through  $\sim 100$ km of lithosphere and seem not to have paused to  
138  
139 62 fractionate to any great extent in the crust (Larsen and Pedersen, 2000; 2009; Hole and  
140  
141 63 Millett, 2016; Matzen et al., 2017).

144  
145 64 O'Hara (1968) showed that with increasing pressure of crystallization, the liquidus  
146  
147 65 fields of olivine and clinopyroxene contracted and that the field for clinopyroxene  
148  
149 66 crystallization expanded at the expense of olivine. Experimental phase equilibria (e.g.  
150  
151 67 Villiger et al., 2004; 2007; Whitaker et al., 2007) show that for fractional crystallization of  
152  
153 68 peridotite-derived magmas at  $\sim 1$  GPa, clinopyroxene (Cpx) crystallizes in compositions with  
154  
155 69 Mg# (molecular %  $\text{MgO}/(\text{MgO} + \text{FeO} + \text{Fe}_2\text{O}_3)$ )  $\leq 65$  whereas the same magma crystallizing at 1  
156  
157 70 atm would have olivine as the sole liquidus phase at Mg# 65. With increasing pressure of  
158  
159 71 fractionation, Cpx becomes a liquidus phase at progressively higher Mg# (Fig. 2) which  
160  
161 72 results in an expansion of the Cpx stability field at the expense of olivine as noted by O'Hara  
162  
163 73 (1968). For a typical BPIP primary magma generated by volatile-free melting of mantle  
164  
165 74 peridotite (see Hole and Millett, 2016 for a compilation of primary magma compositions) at 1  
166  
167 75 atm,  $\sim 25\%$  olivine crystallization would be expected before plagioclase joins the liquidus,  
168  
169 76 whereas at 1.6 GPa, only about 10% crystallization of olivine alone would occur in the same  
170  
171 77 magma before Cpx joins olivine on the liquidus (Fig. 2). This also means that the minimum  
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179  
180 78 Mg# (or Fo content) of olivine when it is the sole liquidus phase also increases with  
181  
182 79 increasing pressure (Fig. 2). Derivative liquids formed by Ol+Cpx fractionation at ~1 GPa,  
183  
184 80 which may ultimately erupt at the surface as lavas, will also have a lower CaO content for a  
185  
186 81 given MgO content than lavas that crystallize olivine alone because Cpx is a CaO-rich phase  
187  
188 82 and strongly fractionates MgO from CaO (Fig. 3A).

191 83 It is generally accepted that the source lithology for CFB and their oceanic equivalents  
192  
193 84 is not exclusively mantle peridotite, and that magmas derived from partial melting of  
194  
195 85 pyroxenite are an important component of CFB magmatism (e.g Herzberg, 2011). The  
196  
197 86 pyroxenite source components in LIPs is generally considered to have been derived from  
198  
199 87 oceanic crust recycled to lower mantle depths as a result of subduction tectonics (Herzberg,  
200  
201 88 2011; Sobolev et al., 2007). In the broadest terms, two distinct lithologies of pyroxenite can  
202  
203 89 be recognized on the basis of SiO<sub>2</sub> content. Primary or very primitive melts derived from  
204  
205 90 high SiO<sub>2</sub> pyroxenite (silica+pyroxene) are recognized because they plot on the SiO<sub>2</sub>-rich  
206  
207 91 side of the orthopyroxene-calcium Tschermaks component (Opx-Cpx-CaTs) join in the  
208  
209 92 molecular projection of the system olivine-Opx-CaTs-silica (Ol-Opx-CaTs-SiO<sub>2</sub>;  
210  
211 93 Herzberg, 2011). Conversely, melts derived from low SiO<sub>2</sub> pyroxenite (olivine+pyroxene)  
212  
213 94 plot to the SiO<sub>2</sub>-poor side of the same plane. Since the Opx-Cpx-CaTs join is a thermal  
214  
215 95 divide at pressures associated with mantle melting i.e. 3-7 GPa, derivative magmas from the  
216  
217 96 melting of high and low SiO<sub>2</sub> pyroxenite have divergent fractionation histories. At near  
218  
219 97 melting pressures, low SiO<sub>2</sub> pyroxenite may undergo mixing with melts of mantle peridotite  
220  
221 98 to form hybrid magmas, whereas melt derived from high SiO<sub>2</sub> pyroxenite cannot (Herzberg,  
222  
223 99 2011)

227 100 Distinguishing between pyroxenite- and peridotite-derived magmas in the lavas erupted  
228  
229 101 at LIPs using major or trace major element geochemical data is fraught with problems. A  
230  
231 102 commonly used indicator of the involvement of pyroxenite-derived melt in basalt  
232  
233  
234

237  
238  
239 103 petrogenesis is that pyroxenite-derived magmas tend to be too deficient in CaO at a given  
240  
241 104 MgO content to be derived from melting of volatile-free mantle peridotite (Herzberg and  
242  
243 105 Asimow, 2008). However, fractionation of CaO relative to MgO is sensitive to the bulk  
244  
245 106 mineral/melt partition coefficient for CaO ( $DCaO$ ) in a magma, and some pyroxenites appear  
246  
247 107 to have  $DCaO > 1$  and others  $< 1$  regardless of the initial concentration of CaO in the solid  
248  
249 108 pyroxenite (e.g. Herzberg, 2011; Jennings and Holland 2015; Jennings et al., 2016). Residual  
250  
251 109 clinopyroxene during melting of pyroxenite is likely to result in the generation of CaO  
252  
253 110 deficient derivative lavas and therefore depends on the extent of melting at which  
254  
255 111 clinopyroxene is partially consumed (Lambart et al., 2016). A significant difficulty is using  
256  
257 112 CaO and MgO contents of mafic lavas to identify pyroxenite involvement is that lavas  
258  
259 113 derived from CaO-poor pyroxenite will fall in the same region of an MgO-CaO diagram as  
260  
261 114 peridotite-derived melts that have fractionated Ol+Cpx at crustal pressures (Herzberg and  
262  
263 115 Asimow, 2008; Fig. 3A). Another effect of crystallization of peridotite-derived magmas at  
264  
265 116 the Ol+Cpx+L cotectic is to drive liquids to progressively more SiO<sub>2</sub>-undersaturation with  
266  
267 117 decreasing Mg#. Consequently, crystallization of a peridotite-derived melt at ~1.0 GPa can  
268  
269 118 result in a CaO depleted, Si-undersaturated liquid composition that is indistinguishable from  
270  
271 119 a magma derived from the melting of low SiO<sub>2</sub> pyroxenite such as those found at the Canary  
272  
273 120 Islands and Madeira (Herzberg, 2011; Gurenko et al., 2013). Furthermore, recycled  
274  
275 121 pyroxenite is also known to be far more heterogeneous than mantle peridotite, both  
276  
277 122 chemically and mineralogically, and produces a diverse range of derivative magmas that may  
278  
279 123 be very similar to peridotite-derived magmas (Keshav et al., 2004; Herzberg, 2011; Lambart  
280  
281 124 et al., 2012; 2013; 2016;). For most high SiO<sub>2</sub> pyroxenite-derived magmas, covariations  
282  
283 125 between SiO<sub>2</sub> and CaO provide a reasonable discriminant from peridotite-derived magmas,  
284  
285 126 because most high Mg#, high SiO<sub>2</sub> lavas derived from mantle peridotite are silica saturated or  
286  
287 127 silica oversaturated olivine or quartz tholeiites, and they owe their high SiO<sub>2</sub> contents to  
288  
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296  
297  
298 128 olivine fractionation at near-surface pressures; as a consequence, any deficiency in CaO at a  
299  
300 129 given MgO content is most likely attributable to high SiO<sub>2</sub> pyroxenite.

302  
303 130 Olivine phenocrysts in mafic lavas provide a mineralogical narrative about magmatic  
304  
305 131 processes in Earth's crust and mantle and olivine records in its chemistry the widespread  
306  
307 132 occurrence of magma chamber fractional crystallization, recharge, and mixing (Herzberg and  
308  
309 133 O'Hara, 2002; Herzberg, 2011; Herzberg et al., 2016; Sobolev et al., 2007). Olivine contains  
310  
311 134 readily measurable concentrations of trace elements Ni, Ca and Mn and the partitioning of  
312  
313 135 these element between olivine and coexisting melt is well-understood (e.g. Beattie et al.  
314  
315 136 1991; Beattie, 1994; 1995; Herzberg and O'Hara, 2002; Matzen et al., 2013; 2017).  
316  
317 137 Fractional crystallization can never be perfect in natural systems and so olivine in mafic lavas  
318  
319 138 may be able to record episodes of fractionation that are not evident from whole-rock data,  
320  
321 139 particularly in magmatic provinces such as the BPIP where polybaric fractionation is most  
322  
323 140 likely to have occurred. Olivine also records the melting of recycled crust as a distinct  
324  
325 141 pyroxenite lithology. In the case of Hawaii, many of the shield-forming lavas (e.g. Mauna  
326  
327 142 Kea) have Ni contents that are too high, and Ca contents that are too low, to have been  
328  
329 143 derived by partial melting of mantle peridotite and they are more likely to be derived from a  
330  
331 144 pyroxenite-rich source (Sobolev et al., 2005; Herzberg, 2006; 2011).

334 145 Before an understanding of the possible source lithology for MPLF lavas can be made, it  
335  
336 146 is clearly necessary to firstly constrain the fractionation histories of the magmas to enable an  
337  
338 147 assessment of the role of Cpx fractionation in causing CaO depletion in basic. Only if this is  
339  
340 148 understood can the contribution of low CaO pyroxenite to the source of the basalts be  
341  
342 149 assessed. To this end, a combination of whole-rock and component olivine chemistry has  
343  
344 150 been utilized to examine the fractionation history of some continental flood basalts from the  
345  
346 151 British Palaeogene Igneous Province (BPIP) which is part of the wider North Atlantic  
347  
348 152 Igneous Province (NAIP).



355  
356  
357 **153 2. Magmatism in the BPIP**  
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359  
360 154 The BPIP is a sub-province of the large NAIP. Magmatism in the BPIP began at ~60 Ma and  
361  
362 155 continued until ~58 Ma. The majority of the lavas of the BPIP are mildly alkaline slightly *Ne-*  
363  
364 156 normative alkali olivine basalts and *Hy-*normative olivine tholeiites. The Mull Plateau Lava  
365  
366 157 Formation (MPLF; Williamson and Bell, 2012) is made of a sequence of ~1 km thickness of  
367  
368 158 plateau-forming lavas dominated by picrites and basalts with local occurrences of hawaiiite,  
369  
370 159 mugearite and trachyte (Kerr 1998; Kerr et al. 1995; 1999). The MPLF is overlain locally by  
371  
372 160 the Central Mull Formation (CMF; Williamson and Bell, 2012) which comprises olivine  
373  
374 161 tholeiite lavas and associated hypabyssal intrusions. The Skye Main Lava Series (SMLS;  
375  
376 162 Thompson, 1982) is petrologically very similar to the MPLF although it is probably slightly  
377  
378 163 older.  
379

380  
381 164 A number of the mafic (>8 wt% MgO) CMF tholeiites which equilibrated at ~1 atm,  
382  
383 165 reached their erupted compositions by crystallization of olivine alone. For 49 of these lavas  
384  
385 166 and intrusions Hole & Millett (2016) generated model primary magma compositions using  
386  
387 167 PRIMELT3 (Herzberg and Asimow, 2008; 2015). Primitive parental melts to CMT erupted  
388  
389 168 compositions required a mantle potential temperature ( $T_p$ ) ~1500°C and MgO ~17.5 wt%.  
390  
391 169 The initial pressure of melting ( $P_i$ ) was ~3.6 GPa with melting stopping by ~2.7 GPa giving a  
392  
393 170 melt fraction for accumulated fractional melting (F-AFM) of ~0.16. Spice et al. (2016)  
394  
395 171 applied the aluminium-spinel-olivine thermometer of Coogan et al. (2014) to Skye and Mull  
396  
397 172 picrites (Mg# 78-85) which yielded crystallization temperatures of 1410°C and 1380°C  
398  
399 173 relative to MORB primitive magmas, which are in excellent agreement with olivine-liquid  
400  
401 174 crystallization temperatures for MPLF lavas given by Hole et al. (2015a). Four of the whole-  
402  
403 175 rock samples of Spice et al. (2016) also provide PRIMELT3 solutions (Appendix 1)  
404  
405 176 indicating  $T_p$  in the range 1524-1568°C and with  $P_f$  of 3.4-3.6 GPa. However, MPLF lavas  
406  
407 177 do not yield PRIMELT3 solutions, although olivine-hosted melt inclusions do provide model  
408  
409  
410  
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414  
415  
416 178 primary magmas which indicate  $T_p \sim 1480^\circ\text{C}$  (Hole and Millett, 2016). The reason why  
417  
418 179 MPLF lavas do not allow the generation of model primary magmas is because they are too  
419  
420 180 deficient in CaO at a given MgO content to have been derived by olivine fractionation alone  
421  
422 181 from mantle peridotite (Fig. 2). As has already been noted, such CaO deficiency can be  
423  
424 182 caused augite fractionation or can be a characteristic of the source region for pyroxenite-  
425  
426 183 derived magmas.

### 430 184 *2.1 Estimates of equilibration pressure of BPIP plateau lavas from major element chemistry.*

432 185 Based on experimentally determined phase relationships for SMLS lavas containing  
433  
434 186 11.1 wt% MgO, Thompson (1974; 1982) proposed that most Mg-rich picrites of the SMLS  
435  
436 187 and MPLF reached their erupted compositions at pressures up to about 1.7 GPa, and that the  
437  
438 188 remaining basic magmas of the BPIP have compositions related to cotectic equilibria at  
439  
440 189 approximately  $0.9 \pm 0.15$  GPa, which is near the base of the crust beneath the Province  
441  
442 190 (Thompson 1982). The melting experiments of Thompson (1974) showed that the general  
443  
444 191 sequence of phase relationships with decreasing T and MgO at  $\sim 1.0$  GPa is Ol+liquid (L);  
445  
446 192 Ol+Cpx+L; Ol+Cpx+Pl+L; whereas at 1 atm, plagioclase is a liquidus phase before  
447  
448 193 clinopyroxene. With increasing pressure, the stability field for clinopyroxene expands at the  
449  
450 194 expense of olivine, and at  $\sim 1.7$  GPa olivine is replaced by clinopyroxene as first crystallizing  
451  
452 195 phase in the system for compositions with  $>11$  wt% MgO. However, augite in typical  
453  
454 196 Plateau-type basalts occurs as groundmass poikilitic anhedral and never forms phenocrysts  
455  
456 197 (Thompson, 1982) and the dominant phenocryst phase in mafic plateau lavas is olivine.

459 198 Fig. 3 illustrates the relationships between CaO, MgO and Mg# for MPLF lavas with  $\geq 8$   
460  
461 199 wt% MgO. The liquid lines of descent (LLD) for a model primary magma at varying  
462  
463 200 pressures from 1 atm to 2.0 GPa is also shown. The primary magma composition chosen for  
464  
465 201 the forward model was the PRIMELT3 solution to Skye lava SK982, which is representative  
466  
467 202 of a typical peridotite-derived magma in this region (Hole and Millett, 2016). Additionally,  
468  
469

473  
474  
475 203 SK982 has  $\epsilon\text{Nd}_{58} = +8.9$  and  $^{206}\text{Pb}/^{204}\text{Pb}=17.5$  which means that it is largely unmodified by  
476  
477 204 interaction with the continental crust (Thompson et al., 1986; Hole et al., 2015a). Forward  
478  
479 205 models of fractional crystallization have been generated using Petrolg3 (Danyushevsky and  
480  
481 206 Plechov 2011) using the parameters given in the caption to Fig 2. Forward modelling of all  
482  
483 49 BPIP model primary magmas shows that the relationship between T, MgO, Mg# and the  
484  
485 207 pressure at which clinopyroxene appears on the liquidus is given by the approximations;  
486  
487 208

$$488 \quad 209 \quad \text{Mg\#} = -0.554\text{P}^2 + 9.81\text{P} + 55.9 \quad (1)$$

$$489 \quad 210 \quad \text{T} = 6.277\text{P}^2 + 141.9\text{P} + 1154.4 \quad (2)$$

492 211 For MgO the uncertainty is  $\pm 0.6\%$  and for Mg#  $\pm 2.0$ . Thus, for a BPIP magma crystallizing  
493  
494 212 at 1.0 GPa, clinopyroxene would join olivine on the liquidus at MgO =  $10.6 \pm 0.6$  wt% or Mg#  
495  
496 213 =  $65.2 \pm 2.0$ .

498 214 Inspection of Fig. 3B reveals the situation where CaO decreases with decreasing Mg# for  
499  
500 215 the assemblage Ol+Cpx+L and thereafter, CaO may increase or decrease with Mg#,  
501  
502 216 depending on pressure, for the assemblage Ol+Cpx+Pl+L. However, the overall variation in  
503  
504 217 CaO once clinopyroxene joins the crystallizing assemblage is not great. Thus, for the SK982  
505  
506 218 primary magma crystallizing at 1.0 GPa, CaO decreases from  $\sim 12.6$  to 12.1 wt% for a change  
507  
508 219 in Mg# of 61-65 and then remains nearly constant (CaO=11.7-12.0 wt%) along the  
509  
510 220 Ol+Cpx+Pl+L cotectic. This feature of Mg#-CaO covariations will be returned to later in the  
511  
512 221 discussion regarding olivine chemistry. It is also noticeable from Fig. 3 that data for the  
513  
514 222 majority of MPLF plateau lavas plots to the left and below the 1 atm Ol+L LLD and close to  
515  
516 223 the Ol+Cpx cotectics for various pressures. Data for CMF and Baffin Island lavas, all of  
517  
518 224 which yield PRIMELT3 solutions, plot close to the predicted olivine liquidus. Since  
519  
520 225 PRIMELT3 only provides solutions for the melting of mantle peridotite for erupted  
521  
522 226 compositions that have crystallized only olivine, then data for such lavas must plot on or  
523  
524 227 above the Ol+L LLD, as they do in Fig. 3. It is logical to assume, therefore, that the reason  
525  
526  
527  
528  
529

532  
533  
534 228 for the MPLF lavas yielding few PRIMELT3 solutions is that they underwent augite  
535  
536 229 fractionation at  $\geq 1.0$  GPa, as proposed by Thompson (1974; 1982).

538  
539 230 Herzberg (2004) provided a method of estimating pressure of equilibration of MORB  
540  
541 231 tholeiitic basalts using a molecular projection in the system Anorthite-Diopside-Enstatite.  
542  
543 232 However, this method has an inherent uncertainty of  $\pm 0.28$  GPa for olivine tholeiites, can  
544  
545 233 only be applied to liquids that fall along the Ol+Pl+Cpx+L cotectic, and is calibrated only up  
546  
547 234 to  $\sim 1.0$  GPa. It has already been shown that for BPIP lavas, the initiation of clinopyroxene  
548  
549 235 crystallization is pressure and composition dependent. Consideration of Fig. 3B shows that  
550  
551 236 the predicted plagioclase-in position has a curvilinear relationship with Mg#. The equation  
552  
553 237 for this curve for the forward models of SK982 is;

$$555 \quad \text{CaO} = 0.0658[\text{Mg\#}]^2 - 6.99[\text{Mg\#}] + 194.8 \quad (3)$$

556 238  
557  
558 239 Data for most Central Mull Formation lavas which are predicted to have been simultaneously  
559  
560 240 saturated in olivine + plagioclase + augite fall above the 0.8 GPa crystallization trajectory in  
561  
562 241 Fig. 3A. Application of the Herzberg (2004) method for estimating  $P_{\text{eq}}$  gives values in the  
563  
564 242 range 0.0-1.0 GPa which are in good agreement with the estimates from Fig. 3B. Most CMF  
565  
566 243 lavas also yield PRIMELT3 solutions for melting of dry peridotite (Hole and Millet, 2016)  
567  
568 244 and therefore have CaO contents at a given MgO content that are consistent with their  
569  
570 245 derivation from mantle peridotite. Bell et al. (1994) presented extensive mineral chemical  
571  
572 246 and geochemical evidence for near 1 atm fractionation of the Skye cone sheets and data for  
573  
574 247 these intrusions and these plot close to the 1 atm Ol+Pl+Cpx+L cotectic in Fig. 3B. There is,  
575  
576 248 therefore, ample evidence of fractionation of MPLF and SMLS lavas in the range 1atm to  $>1$   
577  
578 249 GPa, which is consistent with the results from the melting experiments of Thompson (1974)  
579  
580 250 and other published estimates (e.g. Scarrow and Cox, 1995; Kerr et al., 1999; Hole et al.,  
581  
582 251 2015a). However, many MPLF lavas plot in positions in Fig. 3 that suggest they underwent  
583  
584 252 crystallization of Ol+Cpx at pressures up to 2.0 GPa.

591  
592  
593  
594 253 Interaction between mafic lavas and sialic crust is an integral part of the fractionation  
595  
596 254 history of BPIP magmas. Kerr et al. (1995) provided isotopic evidence for crustal  
597  
598 255 assimilation during turbulent magma ascent for MPLF lavas and there is strong evidence to  
599  
600 256 support the interaction between at least two discrete compositions of sialic crust and mantle  
601  
602 257 derived magma in the BPIP (Thompson et al., 1982; 1986; Hole et al., 2015a). The isotopic  
603  
604 258 compositions of BPIP lavas exhibit extreme variability, with  $\epsilon\text{Nd}_{60}$  varying from  $-30$  to  $+10$   
605  
606 259 and  $^{206}\text{Pb}/^{204}\text{Pb}$  14.3-18.5 which reflects the antiquity of the contaminants involved in  
607  
608 260 petrogenesis. For MPLF lavas, variations in  $\epsilon\text{Nd}_{60}$  are a more modest  $-8$  to  $+8$ . Because the  
609  
610 261 Archean sialic component had  $\epsilon\text{Nd}_{60} \sim -50$ , and was light rare earth element (LREE)  
611  
612 262 enriched, whereas many BPIP magmas are LREE depleted, the total amount of crustal  
613  
614 263 contamination required to reach  $\epsilon\text{Nd}_{60} -8$  was small and possibly only a few % (Hole et al.,  
615  
616 264 2015a). However, Thompson et al. (1986) noted that interaction with Archean granulite  
617  
618 265 facies crust was a feature of plateau lavas that fractionated at  $\sim 1.0$  GPa, whereas upper-  
619  
620 266 crustal contamination occurred during crystallization along the 1 atm Ol+Pl+Cpx cotectic.  
621  
622 267 Peate et al. (2012), using data from olivine-hosted melt inclusions, showed that the crustal  
623  
624 268 assimilation in some magmas must have taken place prior to growth of the olivines, but in  
625  
626 269 others the inclusions have less contaminated compositions than the whole-rocks and crustal  
627  
628 270 assimilation must have taken place both during and after growth of the olivines. Necessarily,  
629  
630 271 the evidence of crustal interaction is entirely from trace element and isotopic compositions.  
631  
632 272 During this study, no relationship was found between isotopic compositions and major  
633  
634 273 element chemistry of lavas, except for concentrations of  $\text{K}_2\text{O}$  and  $\text{TiO}_2$  which tend to be  
635  
636 274 elevated and reduced respectively in lavas that suffered crustal contamination relative to  
637  
638 275 uncontaminated lavas.  
639  
640  
641

642  
276 Major element data therefore suggest that the majority MPLF lavas crystallized  
643  
644  
277 Ol+Cpx at  $\geq 1$  GPa. Consequently, the position occupied by these lavas in Fig. 3B, which is

650  
651  
652 278 below the peridotite-pyroxenite divide of Herzberg and Asimow (2008) could either be  
653  
654 279 because of their fractionation history or because they were derived from a CaO depleted  
655  
656  
657 280 pyroxenite source. Consequently, major elements alone are not sufficient to identify the  
658  
659 281 course lithology of many MPLF lavas.  
660

### 662 282 **3. Models for trace element behaviour during olivine crystallization**

#### 663 664 283 *3.1 Calculation methods – Ni in olivine*

665  
666 284 The methods for calculating model olivine Ni and Ca compositions of primary magmas used  
667  
668 285 here follow those of Beattie et al. (1991), Beattie (1994; 1995), Herzberg and O’Hara (2002),  
669  
670 286 Toplis (2005), Herzberg (2011) and Matzen et al. (2013; 2017) and Gavrilenko et al. (2016).  
671  
672 287 Equations and assumptions pertinent to the current study are given below.  
673

674  
675 288 Fractional crystallization of primary magmas produces olivines that are expected to  
676  
677 289 crystallize along a liquid line of descent (LLD); such olivine compositions have been  
678  
679 290 appropriately termed “crystal line of descent,” or CLD (Putirka et al. 2011). Forward models  
680  
681 291 to define a CLD for olivine have been generated using Petrolog3 (Danyuchesvky and  
682  
683 292 Plechov, 2011) for model BPIP primary magmas (Hole and Millett, 2016) to give an LLD.  
684  
685 293 The predicted CaO, NiO and MnO concentrations in olivines formed in equilibrium with  
686  
687 294 liquids along the LLD have then been calculated using the parameters embedded in  
688  
689 295 PRIMELT3, or modifications of these, which are detailed below.  
690

691  
692 296 The forsterite (Fo) content of olivine in equilibrium with a melt is calculated using the  
693  
694 297 partitioning of Fe and Mg between olivine and melt;

$$696 \quad 298 \quad K_D^{Ol/L}_{(FeO/MgO)} = D^{Ol/L}_{FeO} / D^{Ol/L}_{MgO} \quad (4)$$

698  
699 299  $K_D^{Ol/L}_{(FeO/MgO)}$  varies of a function of pressure, temperature and composition ( $P$ ,  $T$ ,  $X$ ; e.g.  
700  
701 300 Toplis 2005). Herzberg and O’Hara (2002) provide a method for calculating  $K_D^{Ol/L}_{(FeO/MgO)}$   
702  
703 301 for the crystallization of a primary magma along the olivine liquidus from the pressure of  
704  
705

709  
710  
711 302 melting to the surface, and this calculation method is available to the user in Petrolog3.  
712  
713 303 However, for magmas that pause in the crust, then it is  $K_D^{O/L}_{(FeO/MgO)}$  at the depth at which  
714  
715 304 olivine crystallization occurs that is relevant and not necessarily the ascent path to the surface  
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717  
718 305 along the olivine liquidus. The thermodynamical parameterization of Toplis (2005) allows  
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720 306 for the calculation of  $K_D^{O/L}_{(FeO/MgO)}$  at a given pressure for a known magma composition. The  
721  
722 307 Toplis (2005) model has been investigated using Petrolog3 for BPIP primary magma  
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724 308 compositions and the results are illustrated in Fig. 4 for three BPIP magmas with different  
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726 309 MgO contents which cover the range of  $T_p$  estimates for the region (Hole and Millet, 2016).  
727  
728 310 The final pressure of melting ( $P_f$ ) was used as a proxy for pressure of melt segregation which  
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730 311 for BPIP primary magmas is typically  $\sim 2.7$  GPa (Hole and Millett, 2016). This gives values  
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732 312 of  $K_D^{O/L}_{(FeO/MgO)} \sim 0.338$  for derivative magmas at 2.7 GPa formed from primary magmas  
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734 313 containing 17.5 wt% MgO or equivalent to  $T_p=1500^\circ\text{C}$  (Fig. 4A). For near-surface  
735  
736 314 crystallization,  $K_D^{O/L}_{(FeO/MgO)}$  for the same magma would be 0.30-0.31. At a fixed pressure  
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738 315  $K_D^{O/L}_{(FeO/MgO)}$  exhibits a negative covariation with  $\text{Mg}/\text{Fe}_{\text{Liq}}$  (cationic Mg/Fe of the liquid) for  
739  
740 316 liquids with Mg/Fe in the range 1.5-3.3 (Fig. 4B), which is equivalent to the composition of  
741  
742 317 near-primary magmas to moderately evolved basalts with  $\sim 8.0$  wt% MgO. Conversely, the  
743  
744 318 Herzberg and O'Hara (2002) calculation method defines a relationship between  $\text{Mg}/\text{Fe}_{\text{liq}}$  and  
745  
746 319  $k_D^{O/L}_{(FeO/MgO)}$  that is necessarily positive and curvilinear to account for  $P$ ,  $T$ ,  $X$  variability  
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748 320 during ascent along the olivine liquidus, but without magma pausing in the crust. The  
749  
750 321 practical use of this is that the point of intersection between the Herzberg and O'Hara (2002)  
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752 322 crystallization trajectory and a Toplis (2005) crystallization contour allows both  
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754 323  $K_D^{O/L}_{(FeO/MgO)}$  and  $\text{Mg}/\text{Fe}_{\text{liq}}$  at the contoured pressure to be estimated, which are unique to a  
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756 324 given primary magma. For example, considering a magma that segregated at 2.7 GPa and  
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758 325 then paused in the crust and crystallized olivine at 1.0 GPa, it can be assumed that the magma  
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760 326 follows the Herzberg and O'Hara (2002) curve in Fig. 4B from the point of magma  
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770 327 segregation to 1.0 GPa, crystallizing olivine along the CLD. The melt then pauses at 1.0  
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772 328 GPa and crystallizes more olivine. The point at which the Herzberg and O'Hara (2002) LLD  
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774 329 intersects the 0.5 GPa Toplis (2005) contour, which in this example is at  $Mg/Fe_{liq}=1.7$  and  
775  
776 330  $K_D^{Ol/L}_{(FeO/MgO)}=0.317$  (Fig. 4B), is taken as the compositional starting point for 1 GPa  
777  
778 331 olivine crystallization. From that point onwards,  $K_D^{Ol/L}_{(FeO/MgO)}$  is estimated from  $Mg/Fe_{Liq}$   
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780 332 using the Toplis (2005) parameterization at a fixed pressure of 1.0 GPa. Whereas the  
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782 333 intersection point in Fig. 4b is unique to a specific primary magma, it is mainly influenced by  
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784 334  $Mg/Fe$  (and therefore also  $T_p$  and  $P_i$ ) of the primary magma and  $P_f$ . Consequently, for a  
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786 335 magmatic province with relatively consistent values  $T_p$  and  $P_f$  such as the BPIP (Hole and  
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788 336 Millett 2016) a single crystallization model is probably sufficient to describe the LLD of most  
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790 337 basalts and the CLD of their component olivines, and that is the approach which has been  
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792 338 taken here.  
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796 339 An additional complication comes from the expansion of the clinopyroxene stability  
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798 340 field at the expense of olivine with increasing pressure (Fig. 2; Whitaker et al., 2007; Villger  
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800 341 et al., 2007). Magmas that pause in the crust are likely to crystallize on the Ol+Cpx+Liq  
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802 342 cotectic rather than along the olivine liquidus. Investigations using Petrolog3 show that  
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804 343  $K_D^{Ol/L}_{(FeO/MgO)}$  remains nearly constant for the Ol+Cpx+L crystallization interval, but the role  
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806 344 of olivine in the crystallizing assemblage diminishes substantially with continuing  
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808 345 crystallization (Fig. 2). Consequently, for crystallization at elevated crustal pressures, olivine  
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810 346 should be less abundant than for crystallization of the same magma near the surface. This  
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812 347 probably naturally limits the number of olivine analyses that can be generated for olivines  
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814 348 that crystallized on the Ol+Cpx+L cotectic. For the purposes of this study it has been  
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816 349 assumed that for crystallization of Ol+Cpx+L,  $K_D^{Ol/L}_{(FeO/MgO)}$  is constant at the pressure of  
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818 350 interest.  
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829 351 The method used for estimating the Ni content of a primary magma was that of Herzberg  
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831 352 (2011);

$$833 \quad 834 \quad 353 \quad \text{Ni ppm} = 21.6\text{MgO} - 0.32\text{MgO}^2 + 0.051\text{MgO}^3 \quad (5)$$

835  
836  
837 354  $D^{\text{O}/\text{L}}_{\text{NiO}}$  was determined using the method of Beattie et al. (1991) which was re-examined by  
838  
839 355 Herzberg et al. (2016) and found to remain applicable in the light of new experimental  
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841 356 results;

$$842 \quad 843 \quad 357 \quad D^{\text{O}/\text{L}}_{\text{NiO}} = 3.346 D^{\text{O}/\text{L}}_{\text{MgO}} - 3.665 \quad (6)$$

844  
845  
846 358 Because  $D^{\text{O}/\text{L}}_{\text{MgO}}$  is determined from  $K_D^{\text{O}/\text{L}}_{(\text{FeO}/\text{MgO})}$  then magmas which pause at depth in the  
847  
848 359 crust are pressure-compensated for  $D^{\text{O}/\text{L}}_{\text{NiO}}$  by the  $K_D^{\text{O}/\text{L}}_{(\text{FeO}/\text{MgO})}$  calculation method outlined  
849  
850 360 above and in Fig. 4.

851  
852 361 Matzen et al. (2013; 2017) showed that the temperature dependence of  $D^{\text{O}/\text{L}}_{\text{NiO}}$  can  
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854 362 ultimately result in olivine crystallizing from near-primary magmas at near-surface pressures,  
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856 363 having higher Ni contents than olivine crystallizing from the same magma at the pressure of  
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858 364 melt segregation. The relationship between the NiO content of an olivine crystallizing from  
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860 365 the same magma at 1 bar and at the pressure of melt segregation is given by;

$$861 \quad 862 \quad 366 \quad \text{NiO}^{\text{O}/\text{L}}_{\text{T}_{\text{P}0}}/\text{NiO}^{\text{O}/\text{L}}_{\text{T}_{\text{P}f}} = \exp(4505 - (1/\text{T}_{\text{P}0} - 1/\text{T}_{\text{P}f})) \quad (7)$$

863  
864  
865  
866 367 Where  $\text{Ni}^{\text{O}/\text{L}}_{\text{T}_{\text{P}0}}$  and  $\text{NiO}^{\text{O}/\text{L}}_{\text{T}_{\text{P}f}}$  are the NiO contents, in weight units, of olivine crystallizing  
867  
868 368 from magma at the surface and close to the pressure of melt segregation respectively, and  $\text{T}_{\text{P}0}$   
869  
870 369 and  $\text{T}_{\text{P}f}$  are the temperatures of the olivine saturated magma at 0 GPa and at the pressure of  
871  
872 370 melt segregation respectively. Olivine-saturated  $\text{T}_{\text{P}f}$  was taken from  $\text{T}_{\text{O}1}$  of the PRIMELT3  
873  
874 371 solution for the magma of interest (Herzberg and Asimow, 2015; Hole and Millett, 2016).  
875  
876 372 Since subsequent crystallization was assumed to take place near to the surface  
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878 373  $K_D^{\text{O}/\text{L}}_{(\text{FeO}/\text{MgO})}=0.30$  was used in all calculations. Application of equation (7) to primary  
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880 374 magma compositions therefore yields the maximum possible Ni content of an olivine  
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888 375 crystallizing from a primary magma at near-surface pressure. Such olivines are referred to  
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890 376 hereafter as ‘Matzen olivines’.

892  
893 377 *3.2 Calculation methods – Ca in olivine*

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896 378 Three different methods are available for calculating the model olivine Ca compositions of  
897  
898 379 primitive magmas. Two of these (Beattie et al., 1991; Herzberg and O’Hara, 2002) are based  
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900 380 on variation in  $D^{O/L}_{CaO}$  as a function of  $D^{O/L}_{MgO}$ . The Beattie et al. (1991) method, which is  
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902 381 appropriate to near-surface pressures, and is given by;

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905 382 
$$D^{O/L}_{CaO} = 0.0056 D^{O/L}_{MgO} + 0.0135 \quad (8)$$

906  
907 383 And for pressures close to those of melt segregation in the range 3-7 GPa, the equation of  
908  
909 384 Herzberg and O’Hara (2002) can be applied;

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912 385 
$$D^{O/L}_{CaO} = -0.019 D^{O/L}_{MgO} + 0.007 D^{O/L}_{MgO} + 0.0063 / D^{O/L}_{MgO} \quad (9)$$

913  
914 386 Gavrilenko et al. (2016) provided a further method of calculating  $D^{O/L}_{CaO}$  using only the  
915  
916 387 MgO content of an anhydrous magma based on new melting experiments. This method is  
917  
918 388 applicable over a wide range of pressures (1atm to 14 GPa) and temperatures (1150-2050°C).  
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920 389 For magmas containing <11.1 wt% MgO;

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$$D^{O/L}_{CaO} = -0.0043 MgO + 0.072 \quad (10)$$

924  
925 391 And for magmas containing >11.1 wt% MgO;

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928 392 
$$D^{O/L}_{CaO} = 0.00042 MgO + 0.0196 \quad (11)$$

929  
930 393 Fig. 5A shows Petrolog3 forward crystallization models of two BPIP model primary magmas  
931  
932 394 (SK982 and MU1.1) at a nominal pressure of 1 atm for the three olivine-liquid CaO  
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934 395 partitioning models given in equations (8)-(11). In addition, the CaO contents of olivine in  
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936 396 equilibrium with 49 model primary BPIP magmas calculated for equations (8)-(11) are also  
937  
938 397 shown in Fig. 5A. The uncertainty in  $D^{O/L}_{CaO}$  ( $\pm 0.004$ ) for the Gavrilenko et al. (2016) model  
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940 398 for compositions with >11.0 wt% MgO propagates to  $\pm 300$  ppm Ca in Fig. 5. The Beattie et  
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947 399 al. (1991) and Gavrilenko et al. (2016) models predict very similar primary equilibrium  
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949 400 olivine compositions for BPIP magmas, but exhibit significant differences in slope of the  
950  
951 401 CLD, with the Gavrilenko et al. (2016) model initially predicting lower  $D^{Ol/L}_{CaO}$  than the  
952  
953 402 Beattie et al. (1991) model for the same range of Fo contents. However, at around 85-87%  
954  
955 403 Fo, equivalent to a whole rock MgO content of 11 wt% (see equations (10) and (11) above)  
956  
957 404 the Gavrilenko et al. (2016) model exhibits a sharp increase in olivine Ca content with  
958  
959 405 decreasing Fo compared to the Beattie et al. (1991) model. The Herzberg and O'Hara (2002)  
960  
961 406 model predicts lower Ca contents of primary olivine than either of the other two models.  
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963  
964 407 Fig 4b illustrates the effects of crystallization of magmas at varying pressures on Ca and  
965  
966 408 Fo contents of olivine. A single model primary BPIP magma (SK982) has been selected and  
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968 409 Petrolog3 forward models at pressures in the range 1atm to 2.2 GPa generated. CLDs for  
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970 410 olivine have then been calculated from equations (9)-(11). The Beattie et al. (1991) model  
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972 411 has not been investigated because it is applicable only to crystallization at 1 atm. The key  
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974 412 features of Fig. 5 are that both the Herzberg and O'Hara (2002) and Gavrilenko et al. (2016)  
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976 413 models develop an inflection on the CLD at the point at which clinopyroxene joins the  
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978 414 crystallizing assemblage, which is also reflected in the major element data for BPIP lavas  
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980 415 (Fig. 3). According to the Gavrilenko et al. (2016) model, crystallization of the assemblage  
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982 416 Ol+Cpx+L causes the Ca contents of equilibrium olivine to decrease relative to the primary  
983  
984 417 olivine compositions, whereas the Herzberg and O'Hara (2002) model suggests a buffering of  
985  
986 418 Ca content of equilibrium olivine for the same assemblage. Additionally, for the Gavrilenko  
987  
988 419 et al. (2016) model, the CLD for Fo and Ca for the assemblage Ol+L has a relatively shallow  
989  
990 420 slope for Fo contents in the range 85.5-91.5, which is equivalent to about 11 wt% MgO in the  
991  
992 421 whole rock (see equation (11)). For the purposes of consistency, the most recent  
993  
994 422 parameterizations for  $D^{Ol/L}_{CaO}$  of Gavrilenko et al. (2016) are primarily used below, but the  
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996 423 Herzberg and O'Hara model (2002) is also considered if necessary.  
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1006 424 Also shown in Fig. 5B are data for natural olivines in near-primary Siqueiros Fracture  
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1008 425 Zone MORB. These have  $\text{Ca}=2060\pm 63$  ppm at  $\text{Fo}=89.9\pm 0.5$  (64 samples from Gavrilenko et  
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1010 426 al., 2016). Olivine in equilibrium with PRIMELT3 model primary magma compositions  
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1012 427 (Herzberg and Asimow 2015) have predicted  $\text{Ca}=2204\pm 58$  ppm and  $\text{Fo}= 89.9\pm 0.3$  using the  
1013  
1014 428 method of Gavrilenko et al. (2016), which is lower than the observed abundances. However,  
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1016 429 for MORB, predicted and observed data fall within the limits of the regressions used in the  
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1018 430 Gavrilenko et al. (2016) model.

#### 1022 431 **4. Olivine in MPLF lavas**

1023  
1024 432 Data for olivine in MPLF lavas are taken from Sobolev et al. (2007) who analyzed olivine  
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1026 433 grains from 14 different samples giving a total data set of 842 individual determinations.  
1027  
1028 434 Spice et al. (2016) provide a further 64 analyses from Skye, Mull and Rum picrites all of  
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1030 435 which have whole-rock MgO contents in the range 24.8-31.9 wt% MgO. These picrites will  
1031  
1032 436 be treated as a separate group of rocks from the MPLF because their stratigraphical affinity is  
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1034 437 unclear they are unlike any samples found within the main MPLF lava pile.

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1036 438 Covariations between Fo content and Ni content for selected analyses are shown in Fig. 6.  
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1038 439 The data have been selected to be representative of all MPLF olivine, and particularly the  
1039  
1040 440 range of Fo contents which is from ~75-90, although the bulk of the data have  $\text{Fo} > 80$ .  
1041  
1042 441 Olivine with compositions in the range  $\text{Fo} 80-90$  would be in equilibrium with whole-rock  
1043  
1044 442 compositions with  $\text{Mg}\# 54-73$  and  $\text{MgO} 7.7-15.8$  wt% at 1 atm based on forward models of  
1045  
1046 443 the primary magma to SK982. This range in whole-rock compositions encompasses the  
1047  
1048 444 majority of that for MPLF lavas (Fig. 3). The CLDs in Fig. 6 were calculated both for the  
1049  
1050 445 Beattie et al. (1991) and the Herzberg and O'Hara (2002) models. For olivine in MPLF  
1051  
1052 446 lavas, Ni correlates positively with Fo and the data falls into two distinct groups which  
1053  
1054 447 relate to the position of data relative to the 1 atm Beattie CLD; data for samples BR2, BR6  
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1056 448 and BHL15 (Fig. 6A) plots closer to the 1 atm CLD than the remaining samples, the latter all

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449 having a higher Ni for a given Fo content than, for example, sample BR2. Olivine from  
450 picrites from Mull, Skye and Rum (Spice et al. 2016) plot close to the position expected of  
451 olivine in equilibrium with primary magmas. Using these data and equation (7) above, gives  
452 Matzen olivines with Fo 91.2–91.8 and Ni = 3560-3570 ppm which are within the range of  
453 the analyzed values for olivine in the Skye and Mull picrites. Indeed, Matzen olivines  
454 calculated to be in equilibrium with all 49 BPIP PRIMELT3 primary magmas (Fig. 6B) show  
455 considerable overlap with the olivine data from Spice et al. (2016). The picrites of Spice et  
456 al. (2016) which contain high Fo and high Ni olivines must therefore have been formed by  
457 rapid emplacement of a near-primary magma at the surface followed by olivine  
458 crystallization at close to 1 atm. This allows the maximum Ni content of BPIP olivines to be  
459 constrained to ~3600 ppm at Fo ~91.5, values which are within the normal range expected for  
460 dry melting of mantle peridotite at  $T_p \sim 1500^\circ\text{C}$  (Matzen et al., 2017) and within the range of  
461 those calculated for lavas from west Greenland (Herzberg et al., 2016; Matzen et al., 2017).

462 Since it is likely that MPLF lavas equilibrated at pressures  $\geq 1.6$  GPa, the CLD for SK982  
463 at 1.6 GPa has been calculated and is shown in in Fig. 6. It shows a remarkable  
464 correspondence with the data from MPLF samples BR2, BR6 and BHL15, including an  
465 apparent inflection in the data trend which corresponds to clinopyroxene joining the liquidus.  
466 The inflection occurs because  $D^{\text{Cpx/L}}_{\text{NiO}} \sim 0.25 \times D^{\text{Ol/L}}_{\text{NiO}}$ , (Herzberg et al., 2016) and  
467 therefore crystallization along the Ol+Cpx+L cotectic results in a smaller bulk mineral-melt  
468 distribution coefficient for Ni than for olivine crystallization alone. This, in turn, results in  
469 liquid compositions that have fractionated Ol+Cpx having higher Ni contents than for the  
470 same liquid fractionating only olivine, at the same Mg#. It is also noticeable that the data for  
471 near CLD MPLF lavas terminates at the Ol+Cpx+Pl+L cotectic. Mixing between primary  
472 and derivative magmas along the LLD and their subsequent crystallization of olivine would  
473 produce olivine compositions that fall on the mixing lines shown in Fig. 6. The remaining

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474 data for MPLF olivines fall along these mixing lines. This is a similar situation to that  
475 described by Herzberg et al. (2016) for mixing relationships in olivines and primary and  
476 derivative magmas from Theistareykir, Iceland. The mixing lines shown in Fig. 5 relate to  
477 specific MgO contents of the derivative magma along the LLD. So, for sample BR5, mixing  
478 between a primary magma and a derivative magma with ~6 wt% MgO could produce the  
479 observed array of olivine compositions, whereas sample BHL34 would require a derivative  
480 magma with higher MgO content to produce its component olivine compositions.

481 Fig. 7 shows covariations between Fo and Ca content of the same olivines as shown in  
482 Fig. 6. Because Ca is incompatible with respect to olivine ( $D^{Ol/L}_{CaO}$  typically 0.025-0.031)  
483 then a negative covariation between Fo and Ca content might be expected, but this is not  
484 observed. A feature of Fo-Ca covariations in MPLF olivines is the relatively consistent Ca  
485 content of 1800-2400 ppm over a range of Fo = 80-89. The predicted range of olivine  
486 compositions calculated for the fractional crystallization of 49 BPIP PRIMELT3 solutions  
487 from primary MgO contents to 8 wt% MgO, using the Beattie et al. (1991) 1atm  
488 crystallization and Gavrilenko et al. (2016) olivine liquidus models are also shown in Fig. 7.  
489 The field for olivine calculated using the Beattie *et al.* (1991) model has a considerably  
490 steeper slope than that for the Gavrilenko et al. (2016) model. It is noticeable that olivine in  
491 the Skye picrites mostly fall within the range of the Beattie et al. (1991) model and close to  
492 primary olivine compositions predicted by the Gavrilenko et al. (2016) model, which is also  
493 consistent with their position in Ni-Fo space (Fig. 6). Olivine in Mull picrites appear to have  
494 consistently lower Ca for a given Fo content than the Skye olivines.

495 Very few of the data for olivine grains from MPLF lavas fall within the region reserved  
496 for 1 atm olivine crystallization from primary and derivative magmas using the Gavrilenko et  
497 al. (2016) model. CLDs at varying pressures, calculated as described earlier, are also shown  
498 in Fig. 7, and mirror the data trends shown in Fig. 3 such that once Cpx joins olivine on the

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1183 499 liquidus, the CaO content of olivine decreases slightly at a given pressure. Therefore, as in  
1184  
1185 500 Fig. 4 for whole-rock data, some of the MPLF olivines exhibit patterns of distribution of Fo  
1186  
1187 501 and CaO that are consistent with crystallization along the Ol+Cpx+L cotectic. However, the  
1188  
1189 502 position of isobars in Fig. 7 cannot be predicted with certainty because the CaO abundances  
1190  
1191 503 of BPIP primary magmas vary considerably, such that at ~1.6 GPa the variation within  
1192  
1193 504 primary magmas is equivalent to a variation of ~500 ppm Ca in the olivine data set. In  
1194  
1195 505 addition, slow diffusion of Ca in olivine can result in consistent Ca but variable Fo contents  
1196  
1197 506 in individual grains, which is not evident for faster-diffusing Ni data (Coogan et al., 2005;  
1198  
1199 507 Chakraborty, 2010). Consequently, the apparent sub-horizontal arrays delineated by olivines  
1200  
1201 508 from individual MPLF samples could in part be a result of slow Ca diffusion, and it is the Ca  
1202  
1203 509 content of the most Fo-rich samples that best reflects crystallization conditions. However,  
1204  
1205 510 the Ca and Fo content of the MPLF lavas shown in Fig. 7 seems to require crystallization at  
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1207 511  $\geq 1.4$  GPa  
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1211 512 Covariations between NiO and CaO are shown in Fig. 8 for all the Ca partitioning  
1212  
1213 513 models in equations (8)-(11) based on the primary magma to Skye lava SK982, and for the  
1214  
1215 514 Herzberg and O'Hara (2002) and Gavrilenko et al. (2016) methods, at varying pressures. Ca-  
1216  
1217 515 Ni covariations for the Gavrilenko et al. (2016) model exhibit a series of negative 'spikes'  
1218  
1219 516 which relate to the composition at which clinopyroxene joins olivine in the crystallizing  
1220  
1221 517 assemblage, which is a predictable consequence of augite fractionation. Because  
1222  
1223 518 clinopyroxene joins the crystallizing assemblage at progressively higher MgO contents with  
1224  
1225 519 increasing pressure (Fig. 3), the negative 'spikes' occur at higher Ni contents with increasing  
1226  
1227 520 pressure. However, an added complication is that for pressures >1.2 GPa, clinopyroxene joins  
1228  
1229 521 the crystallizing assemblage at >11.1wt% MgO so equation (11) applies to the assemblages  
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1231 522 Ol+L and Ol+Cpx+L, whereas for pressures <1.2 GPa clinopyroxene joins the crystallizing  
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1233 523 assemblage at <11.1wt% MgO and so equation (11) only applies to the olivine liquidus.  
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1242 524 Once crystallization takes place along the Ol+Cpx+Pl+L cotectic Ni-Ca variations define  
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1244 525 shallow concave upwards trends, Ca contents decreasing with increasing pressure of  
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1246 526 fractionation. The Herzberg and O'Hara (2002) model is less complex and predicts lower Ca  
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1249 527 for a given Ni content than the Gavrilenko et al. (2016) model.  
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1251 528 Data for olivine in Skye picrites falls close to the extension of the trajectory for low  
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1253 529 pressure (~1 atm) crystallization of Ol+L and close to the position of Matzen olivines  
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1255 530 calculated to be in equilibrium with BPIP primary magmas at 1 atm. Data for olivine in Mull  
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1257 531 picrites generally have lower Ca for a given Fo than those from Skye picrites, and lower than  
1258  
1259 532 the model Matzen olivines. This may suggest that the Mull picrites may have experienced  
1260  
1261 533 clinopyroxene fractionation in the mantle (Herzberg and Asimow, 2008). The data for MPLF  
1262  
1263 534 olivines shown in Fig 7c plots significantly below the olivine liquidus of Gavrilenko et al.  
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1265 535 (2016) and close to, or below, the olivine liquidus for the Herzberg and O'Hara (2002)  
1266  
1267 536 model. Consequently, data for the samples shown in Figs 6 and 7 are consistent with  
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1269 537 crystallization of Ol+Cpx+L at elevated pressures up to ~2.2 GPa. Given that the final  
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1271 538 pressure of melting for many BPIP lavas is ~2.7 GPa (Hole and Millett, 2016) then in some  
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1273 539 cases, crystallization of olivine must have commenced at or close to the lithosphere-  
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1275 540 asthenosphere boundary, as suggested by Matzen et al. (2017). In Fig. 8A data are shown for  
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1277 541 MPLF olivines that falls between the Gavrilenko et al. (2016) and Herzberg and O'Hara  
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1279 542 (2002) olivine liquidii, and therefore close to the expected range of Ca and Ni content for  
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1281 543 crystallization of the assemblage Ol+L. BPIP Matzen olivines fall on the extension of the  
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1283 544 Ol+L trajectory and have higher Fo contents ~92 (Fig. 7) than the observed values for MPLF  
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1285 545 lavas (maximum ~Fo88). Crystallization of ~15-20% olivine from a Skye picrite  
1286  
1287 546 composition could produce the observed Ni, Ca and Fo contents of the MPLF olivines shown  
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1289 547 in Figs 7A and 8A. On the basis of Ni and Ca distributions in olivines discussed above all  
1290  
1291 548 MPLF lavas could have been generated by crystallization from a peridotite-derived magma at  
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1301 549 variable, but elevated pressures of up to ~2.2 GPa. Consequently, there is no evidence to  
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1303 550 suggest the involvement of pyroxenite in the source of the MPLF lavas.  
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1305 551 Herzberg (2011) showed that  $Fe/Mn > 70$  at any Fo content is characteristic of olivine  
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1308 552 that crystallizes from pyroxenite compared to  $Fe/Mn$  60-70 for olivine in peridotite-derived  
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1310 553 magmas. The mean  $Fe/Mn$  for MPLF lavas is  $69.2 \pm 2.6$  (Fig. 9) but two samples (BCH14 and  
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1312 554 AM7a) fall exclusively above 70 with a maximum  $Fe/Mn$  of 75 (Fig. 9A). However,  
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1314 555 Herzberg et al. (2013) demonstrated that clinopyroxene fractionates Fe from Mn causing  
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1316 556 elevated  $Fe/Mn$  in lavas that undergo clinopyroxene fractionation. Consequently, the elevated  
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1318 557  $Fe/Mn$  in the MPLF lavas that crystallized at the highest pressures (Fig. 9B) may solely be  
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1320 558 due to clinopyroxene fractionation and not because they were derived from pyroxenite.  
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1322 559 Furthermore, the lavas shown in Fig. 9B contain olivine which carry a lower pressure  
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1324 560 crystallization signature than those in Fig. 9A, and also have  $Fe/Mn < 70$ . Low and high  $SiO_2$   
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1326 561 pyroxenite-derived lavas from Hawaii (HSDP-2) contain olivines with  $Fe/Mn = 73.8 \pm 1.7$  and  
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1328 562 up to a maximum of 81 all of which are higher than the MPLF samples again suggesting that  
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1330 563 pyroxenite was not an important component of the course of MPLF lavas.  
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## 1333 564 **5. Olivine in magmas from Iceland, Baffin Island and west Greenland**

1334  
1335 565 The distribution of Ni in olivines in lavas from Thesitareykir (Iceland), Baffin Island and  
1336  
1337 566 Disko Island (west of Greenland) were discussed in detail by Herzberg et al. (2016). A  
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1339 567 feature of olivines within lavas from Theistareykir are the existence of two populations, one  
1340  
1341 568 primitive population crystallizing close to the 1 atm CLD and the other falling on mixing  
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1343 569 lines with derivative magmas along the CLD (Herzberg et al., 2016). However, the two  
1344  
1345 570 populations of olivines are mutually exclusive with respect to their host lavas. When the same  
1346  
1347 571 data are considered in terms of Ni-Ca covariations (Fig. 10A), olivines that exhibit evidence  
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1349 572 of an origin by magma mixing in terms of Fo-Ni correlations, have lower CaO contents over  
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1351 573 the same range of Ni contents than the more primitive olivines that fall close to the 1 atm  
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1360 574 CLD. Simple magma mixing cannot account for these variations because mixing lines for  
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1362 575 reasonable MgO contents of derivative magmas ( $\geq 7$  wt% MgO) do not intersect both groups  
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1364 576 of olivines. Indeed, mixing between a primary magma formed at depth with magmas formed  
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1366 577 at progressively shallower levels would necessarily fall along the predicted pathway for the  
1367  
1368 578 Herzberg and O'Hara (2002) Ni-Ca partitioning model (Fig. 9). Similarly, mixing between  
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1370 579 primitive or near primary magmas (those which would crystallize Matzen olivines) and  
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1372 580 derivative magmas close to the surface would follow the predicted pathway for the Beattie et  
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1374 581 al. (1991) Ni-Ca partitioning model.

1377 582 Inspection of Fig. 7 shows that for BPIP lavas crystallizing only olivine, Ca contents of  
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1379 583 olivines should be lower for crystallization occurring at higher pressures compared to those at  
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1381 584 lower pressures. MacLennan et al. (2003) showed that some lavas at Theistareykir  
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1383 585 crystallized at  $>0.8$  GPa and showed evidence of crystallization along the Ol+Cpx+L  
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1385 586 cotectic. So, rather than being a result of mixing it is more likely the low CaO olivines found  
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1387 587 in some Theistareykir lavas are those which crystallized at  $\sim 1.0$  GPa. The variation in  
1388  
1389 588  $D^{Ol/L}_{NiO}$  because of fractionation at higher T (Matzen et al. 2017) would not be great enough  
1390  
1391 589 to cause any discernible variation in Ni content between high and lower pressure olivines.  
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1393 590 Nevertheless,  $K_D^{Ol/L}_{(FeO/MgO)}$  would be higher ( $\sim 0.310$ ) for the higher pressure olivines than  
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1395 591 those which crystallized at 1 atm ( $\sim 0.298$ ) thus resulting in differences in distribution of the  
1396  
1397 592 data in terms of Ni-Fo covariations. Also, because crystallization along the Ol+Cpx+L  
1398  
1399 593 cotectic results in a smaller bulk mineral-melt distribution coefficient for Ni than for olivine  
1400  
1401 594 crystallization alone, liquid compositions that have fractionated Ol and Cpx have higher Ni  
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1403 595 contents than for the same liquid fractionating only olivine, resulting in higher Ni contents in  
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1405 596 olivine for a given Fo content, as is observed at Theistareykir.

1409 597 Covariations between Ca and Ni in data for olivines from Baffin Island (Fig. 10B)  
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1411 598 follow the predicted CLDs defined by the Beattie et al. (1991) and Gavrilenko et al. (2016)  
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1419 599 models and the data therefore follow trends that are consistent with crystallization at or near  
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1421 600 the surface. The most Ni-rich and Ca-poor olivines overlap with the composition of Matzen  
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1423 601 olivines calculated to be in equilibrium with PRIMELT3 primary magmas. Consequently,  
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1425 602 some of the olivines in Baffin Island lavas can be considered to be truly primary and  
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1427 603 crystallized after rapid emplacement of magma close to the surface. Importantly, there is no  
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1429 604 evidence for crystallization along the Ol+Cpx+L cotectic, the projection of which falls well  
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1431 605 below the observed data in Fig. 10B. Olivines in lavas from Disko Island (Fig. 10C) have  
1432  
1433 606 systematically lower Ca contents at a given Fo content compared to the values predicted by  
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1435 607 the Gavrilenko et al. (2016) model for crystallization at 1 atm, and lower Ca contents than  
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1437 608 olivines from Baffin Island with similar Fo contents. Hole and Millett (2016) showed that at  
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1439 609 Baffin Island  $P_f \sim 2.1$  GPa whereas at Disko island  $P_f \sim 2.8$  GPa which most likely reflects a  
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1441 610 deeper asthenosphere-lithosphere boundary at Disko Island Baffin Island (Matzen et al.,  
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1443 611 2017). One possibility is that Disko Island magmas underwent minor clinopyroxene  
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1445 612 fractionation in the mantle before being emplaced, whereas the Baffin Island magmas only  
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1447 613 crystallized olivine and underwent rapid emplacement to shallow levels. Alternatively, the  
1448  
1449 614 low Ca and high Ni contents of some Disko Island olivines could be interpreted as a result of  
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1451 615 their derivation from a low CaO pyroxenite source. However, the involvement of pyroxenite  
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1453 616 cannot be substantiated by the Fe/Mn for Disko Island olivines (Fig. 9). Consequently, the  
1454  
1455 617 observed differences between Baffin and Disko island olivines might relate to the differences  
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1457 618 in magmatic plumbing at the two locations rather than to source heterogeneities.  
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## 1463 619 **6. Clinopyroxene stability during polybaric fractionation**

1466 620 MPLF and SMLS lavas do not contain augite phenocrysts, only olivine. Augite tends  
1467  
1468 621 to form as groundmass poikilitic anhedral in sparsely olivine-phyric lavas regardless of Mg#  
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1470 622 (Thompson, 1982; Bailey et al., 1923). Nevertheless, there appears to be ample evidence to  
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623 suggest that augite was an important crystallizing phase during the evolution of the BPIP  
624 plateau lavas, which sets them apart from other NAIP lavas. On a regional scale, there were  
625 large volumes of pyroxene gabbro intruded throughout the lifetime of the BPIP. The Ben  
626 Buie gabbro complex on Mull forms part of the intrusive complex beneath the Mull volcano  
627 and is considered to be a product of the crystallization of a tholeiitic magma (Thompson  
628 1982).

629 The estimates of ~1.0 GPa crystallization pressure of some of the more evolved lavas  
630 along the Ol+Cpx+Pl+L cotectic, coupled with the high pressure Ol+L or Ol+Cpx+L  
631 crystallization trends evident in the olivine data, strongly suggests that polybaric fractional  
632 crystallization was important in the BPIP, as was originally postulated by Thompson *et al.*  
633 (1980) and developed further by other workers (e.g. Kerr *et al.*, 1999; Fowler *et al.*, 2003;  
634 Font *et al.*, 2008; Hole *et al.*, 2015a). Herzberg and Asimow (2008) noted that deep  
635 crystallization of high CaO augite can have the same effect as shallow augite fractionation on  
636 the CaO contents of high MgO primary magmas, and Keshav *et al.* (2007) argued that  
637 clinopyroxene can accumulate in the mantle and evidence of its existence is found in mantle  
638 xenoliths. Consequently, the Ni-Ca relationships shown in Fig. 8 for MPLF lavas could  
639 result from initial crystallization of augite at mantle depths, close to the top of the melting  
640 column (~2.7 GPa) in the region. The distribution of the most Fo-rich olivines (Fo~92) from  
641 Mull in Fig. 8B, and the fact that they have lower Ca content but the same Ni contents as  
642 Matzen olivine compositions suggests that mantle augite accumulation is a possibility.  
643 Furthermore, olivines in Skye picrites exhibit Ni-Ca covariations that are suggestive of  
644 limited or no mantle augite accumulation (Fig. 8B). As discussed above, the sub-horizontal  
645 arrays delineated by the MPLF lavas in Fig. 8B might be a result of slower diffusion of Ca  
646 relative to Ni in olivine (Coggan *et al.*, 2005; Chakraborty, 2010) and the Ca contents of these  
647 olivines reflect the accumulation of augite in the mantle.

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1536  
1537 648 An alternative explanation for the lack of early-formed augite phenocrysts in MPLF  
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1539 649 lavas relates to differences in clinopyroxene and olivine stability in primitive melts.  
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1541 650 Sugawara (2000) and Chen and Zhang (2008; 2009) provide empirical relationships between  
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1543 651 pressure, temperature and the MgO and CaO content of olivine and pyroxene saturated  
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1545 652 liquids. The Sugarawa (2000) parameterizations show that for a given melt MgO content and  
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1547 653 pressure, augite saturation occurs at ~50°C lower temperature than olivine saturation. The  
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1549 654 Chen and Zhang (2008; 2009) studies relate to the dissolution of mantle clinopyroxene  
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1551 655 (diopside  $\text{Wo}_{49.5}\text{En}_{48.5}\text{Fs}_{2.0}$ ) in basaltic melt. It is appreciated that extrapolating such a study  
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1553 656 to pyroxenes with compositions of around  $\text{Wo}_{45}\text{En}_{41}\text{Fs}_{14}$  such as those found in mafic BPIP  
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1555 657 magmas (Hole et al. 2015a) may not be strictly valid, but these are the most applicable  
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1557 658 parameterizations in the literature. Chen and Zhang (2009) showed that saturation of Mg-  
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1559 659 and Ca-rich clinopyroxene may be determined by the three major components MgO, CaO and  
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1561 660  $\text{SiO}_2$ . Chen and Zhang (2009) developed a relationship between  $C_0^{\text{MgO}} \times C_0^{\text{CaO}}$ , pressure and  
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1563 661 temperature to define clinopyroxene stability in a basaltic melt, where  $C_0^{\text{MgO}}$   $C_0^{\text{CaO}}$  are the  
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1565 662 concentrations of MgO and CaO in a liquid at the point of saturation. Fig. 11 is a plot of CaO  
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1567 663 *versus* MgO for the MPLF whole-rock samples from which olivine data were derived plotted  
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1569 664 alongside clinopyroxene and olivine saturation curves calculated from Chen and Zhang  
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1571 665 (2008; 2009). Here it is assumed that polybaric fractional crystallization took place in two  
1572  
1573 666 distinct phases; firstly at 1.5 GPa with Cpx joining the liquidus at ~1390°C (the Cpx-in  
1574  
1575 667 temperature for SK982 at 1.5 GPa) and secondly at 1.0 GPa with an accompanying fall in  
1576  
1577 668 temperature to 1365°C (~55°C/GPa<sup>-1</sup>; Sugawara, 2000; Matzen et al., 2017). Olivine  
1578  
1579 669 saturation is independent of CaO content of the liquid, and over the temperature interval in  
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1581 670 question, olivine saturation is little effected. However, augite saturation occurs at  
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1583 671 considerably higher CaO content for a given MgO at 1.0 GPa compared to its saturation at  
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1585 672 1.5 GPa. The whole-rock analyses of MPLF lavas fall close to the join the between olivine  
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1596 673 and clinopyroxene saturation curves at 1.5 GPa. However, at 1.0 GPa the same liquid  
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1598 674 composition is saturated with respect to olivine, but not with respect to clinopyroxene.  
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1600 675 Consequently, is it possible during polybaric fractional crystallization under the range of  
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1602 676 pressure conditions applicable to the MPLF, to render clinopyroxene unstable. Consequently,  
1603  
1604 677 the lack of observed phenocrysts of augite in MPLF lavas might result from their dissolution  
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1606 678 during magma storage and fractionation at  $\leq 1.0$  GPa.  
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## 1610 679 **7. Predicting pyroxenite in the source of continental flood basalts**

1611  
1612 680 Olivines from low SiO<sub>2</sub>-pyroxenite-derived magmas from Mauna Loa (HSDP-2) have Ni  
1613  
1614 681 contents of ~4000 ppm at Fo ~90 whereas high SiO<sub>2</sub> pyroxenite derived magmas from  
1615  
1616 682 Koolau (Makapuu stage) have even more extreme Ni contents of up to 4660 ppm at Fo ~89  
1617  
1618 683 (Herzberg, 2006; 2011; Sobolev et al., 2005). Both of these Ni contents are in excess of the  
1619  
1620 684 highest Ni Matzen olivines from Disko Island which have ~3600 ppm Ni at Fo ~91.5. Since  
1621  
1622 685 the Matzen olivines from West Greenland represent the highest possible Ni contents  
1623  
1624 686 attainable for olivines in the NAIP, the values of up to 3000 ppm Ni at Fo ~90 for MPLF  
1625  
1626 687 lavas does not require a high Ni pyroxenite source. Additionally, CaO contents of low and  
1627  
1628 688 high SiO<sub>2</sub> pyroxenite-derived magmas are <2000 and <1700 ppm respectively which are  
1629  
1630 689 lower than the most CaO-poor MPLF lavas (Figs 5 & 6).  
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1632

1633 690 Olivines from mafic magmas in some Karoo LIP lavas (e.g. N356 of Sobolev et al.,  
1634  
1635 691 2007) which have whole-rock with ~6wt% CaO at ~15wt% MgO (Ellam and Cox 1989)  
1636  
1637 692 exhibit extreme depletions in Ca (100-1400 ppm), enrichment in Ni (up to 4500 ppm at Fo  
1638  
1639 693 88) and have Fe/Mn 75-82. However, sample N356 comes from the Lebombo monocline and  
1640  
1641 694 is one of a suite of high TiO<sub>2</sub> lavas with  $\epsilon\text{Nd}_{190} = -4$  to  $-10$  and  $\epsilon\text{Hf}_{190} = -3$  to  $-11$  and is likely  
1642  
1643 695 to be a sample of Proterozoic lithosphere (Ellam and Cox, 1989; 1992; Ellam et al., 1992;  
1644  
1645 696 Ellam, 2006) and not necessarily recycled oceanic crust in the sense of Sobolev (2005) and  
1646  
1647 697 Herzberg (2006). Extreme depletions in CaO (~8 wt% at MgO ~9.5 wt%) in Si-oversaturated  
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698 (SiO<sub>2</sub> ~54 wt%) whole rock samples from part of the Karoo LIP in the Falkland Islands were  
699 also reported by Hole et al. (2015b) and these too have εNd<sub>190</sub> as low as -11, although they  
700 are olivine-free. It is beyond the scope of the current study to discuss the role of pyroxenite  
701 in continental LIPs, but a picture is emerging that the continental lithosphere might also be a  
702 source of pyroxenite which contributes to CFB magmatism. Data for olivines in lavas from  
703 the Siberian Traps (Sobolev et al. 2007; 2009) have Ni contents of ~3200ppm at Fo<sub>83.0</sub>  
704 which would project to higher Ni contents than those of Matzen olivines (~3200ppm at  
705 Fo<sub>91.8</sub>) calculated from PRIMELT3 solutions at the same Fo content, regardless of pressure of  
706 crystallization. These same olivine phenocrysts contain <2000 ppm Ca. Consequently, the  
707 olivines must have crystallized from partial melt of pyroxenite, and not from mantle  
708 peridotite (Sobolev et al. 2009).

709         Given the intricacies of interpreting the whole-rock and olivine record of fractionation  
710 of magmas in the crust, it is not surprising that the positive identification of pyroxenite  
711 involvement in magmagenesis at LIPs using major elements is highly problematical. It is  
712 quite clear that any attempts to utilize CaO content of lavas to identify pyroxenite sources  
713 will be ambiguous in their outcomes. Perhaps a step closer can be made by considering the  
714 fractionation of basaltic magmas at varying pressures without utilizing CaO abundances. Fig.  
715 12 shows SiO<sub>2</sub>/MgO versus Mg# for basalts from Hawaii (HSDP-2), the MPLF and  
716 Theistareykir. LLDs at 1 atm have been calculated from PRIMELT3 primary magmas using  
717 Petrolog3, including for HSDP samples, a few of which are derived from mantle peridotite  
718 (Herzberg and Gazel 2009). Because Cpx is saturated with respect to silica, when it joins the  
719 liquidus it fractionates SiO<sub>2</sub> and MgO relative to one another at a different rate from olivine  
720 crystallization resulting in infections on the LLD as seen in Fig. 12. Data for HSDP lavas  
721 mostly fall on the 1 atm LLD implying olivine crystallization alone is responsible for their  
722 evolution. The majority of HSDP samples have lower CaO for a given MgO than would be

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1713  
1714 723 expected for olivine crystallization alone and consequently the source from which they were  
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1716 724 derived must be similarly CaO deficient indicating a role for pyroxenite which is also  
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1718 725 consistent with olivine trace element data (e.g. Sobolev et al., 2005; Herzberg, 2006). By  
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1720 726 contrast, MPLF lavas show significant deviations from the 1 atm LLD as a result of  
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1722 727 crystallization of Ol+Cpx. Consequently, the CaO deficiency of these lavas need not be  
1723  
1724 728 attributed to the involvement of pyroxenite, which the olivine trace element data also  
1725  
1726 729 indicates. For Theistareykir, the majority of lavas follow the 1 atm LLD with some deviation  
1727  
1728 730 at Mg# <65 towards lower SiO<sub>2</sub>/MgO than the 1 atm LLD, which is most likely a  
1729  
1730 731 consequence of the crystallization of Ol+Cpx. Crystallization of Ol+Cpx at ~0.8 GPa is  
1731  
1732 732 consistent with the melt inclusion studies of MacLennan et al., (2003) as well as the olivine  
1733  
1734 733 trace element data discussed above. So, whereas the representation of data in Fig. 12 cannot  
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1736 734 be used to identify pyroxenite in the source of LIPs, it can be used to identify lavas that have  
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1738 735 crystallized Cpx+Ol at depth, without the use of CaO data, which therefore removes some  
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1740 736 ambiguity from the interpretation of MgO and CaO data for continental flood basalts.  
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## 1745 737 **8. The NAIP – a peridotite-dominated LIP**

1746  
1747 738 It is generally accepted that many of the shield-building lavas of Hawaii are likely to have  
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1749 739 been derived from melting of a pyroxenite-rich source lithology (e.g. Herzberg 2006; 2011;  
1750  
1751 740 Sobolev et al., 2005; 2007). This pyroxenite is thought to be derived from the recycling of  
1752  
1753 741 oceanic crust at depth and was subsequently entrained in a mantle plume. The  
1754  
1755 742 transformations that oceanic crust may undergo during recycling are complex and involve  
1756  
1757 743 rock-melt, melt-melt and rock-rock interactions (e.g. Herzberg, 2011; Lambart et al., 2016;  
1758  
1759 744 Lambart, 2017). Sobolev et al. (2005; 2007) argued that the rising mantle plume beneath  
1760  
1761 745 Hawaii contains eclogite bodies that start melting at about 190–180km depth. Melting of  
1762  
1763 746 eclogite produces high SiO<sub>2</sub> initial melts which infiltrate into, and react with, the adjacent  
1764  
1765 747 peridotite eliminating olivine and producing a solid pyroxenite. Both the reaction pyroxenite  
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748 and unreacted peridotite melt at depths between 140 and 100 km, producing hybrid magmas  
749 by mixing in conduits and crustal magma chambers. Nevertheless, data from Mauna Kea  
750 reveal that low- and high-SiO<sub>2</sub> pyroxenite-sourced magmas coexist as discrete batches along  
751 with peridotite-derived magmas (Rhodes et al., 2012; Sobolev et al., 2005; 2007) leading  
752 Herzberg (2011) to promote the hypothesis that melts of pure pyroxenite may segregate from  
753 their source without mixing and be erupted at the surface as lavas.

754 In the case of LIPs formed above thick continental lithosphere (e.g. Siberian Traps and  
755 parts of the NAIP) the composition of magmas should be almost exclusively controlled by the  
756 melting of reaction pyroxenite because thick lithosphere restricts the amount of melt that can  
757 be derived from mantle peridotite (Sobolev et al., 2005; 2007; 2009; Hole and Millett, 2016).  
758 The model proposed by Sobolev et al. (2005; 2009) for the petrogenesis of lavas of the  
759 Siberian Traps relies on the fact that SiO<sub>2</sub> rich olivine-free pyroxenite (that which plots to the  
760 right of the Cpx-CaTs-Opx plane) has a lower solidus temperature at a given pressure than  
761 peridotite (Sobolev et al., 2005; Lambart et al., 2012; 2016; Lambart 2017) and consequently  
762 for a given T<sub>p</sub>, pyroxenite will melt at a higher pressure than peridotite. On this basis,  
763 Sobolev et al. (2009) concluded that early tholeiites from the Gudchikhinsky Formation of  
764 the Siberian Traps were 100% pyroxenite melts with the contribution from pyroxenite  
765 decreasing to 40-50% (Tuklonsky and Nadezhdinsky formations) as peridotite was entrained  
766 in the melting zone. Sobolev et al. (2009) further argued that differences in the compositions  
767 of lavas from the Hawaii and Siberian LIPs was simply a function of lithospheric thickness  
768 and not the amount of recycled material present in the mantle, implying that all mantle  
769 plumes could have, and should have, a similar petrological structure.

770 Matzen et al. (2017) provided estimates of 100 and 32 km for the depths to the LAB  
771 beneath Disko Island and Iceland rift zones respectively. Hole and Millett (2016) showed  
772 that P<sub>f</sub> was ~2.7 GPa for BPIP magmas and was not significantly different from that for

1830  
1831  
1832 773 Disko Island, West Greenland ( $P_f \sim 2.8$  GPa) suggesting a similar depth to the LAB in both  
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1834 774 areas. A pyroxenite melting signature would therefore be expected to be most evident in lavas  
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1836 775 of the MPLF and Disko Island (Fig. 13). However, it is apparent that none of the NAIP lavas  
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1838 776 under consideration here provide any evidence of melting of 100% pyroxenite, or indeed any  
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1840 777 significant proportion of pyroxenite, once the effect of high pressure fractionation on major  
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1842 778 element compositions is accounted for. In the case of Iceland, existing studies provide  
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1844 779 estimates for the overall contribution from pyroxenite which vary from <5% (Brown and  
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1846 780 Leshner, 2014) through 10% (Sobolev et al., 2007; Lambart, 2017) to 30% (Matthews et al.,  
1847  
1848 781 2016). Lambart (2017) showed that the observed crust thickness of the Icelandic rift zones is  
1849  
1850 782 consistent with about 10% of recycled crust in the form of a *low* SiO<sub>2</sub> pyroxenite lithology  
1851  
1852 783 and that diversity of trace element and isotopic compositions observed in Icelandic rift zone  
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1854 784 basalts does not require a contribution from melts derived directly from a recycled basalt  
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1856 785 component, but results from the melting of hybrid olivine-bearing lithologies formed by solid  
1857  
1858 786 state reactions between recycled crust and peridotite (Lambart, 2017).

1861  
1862 787 Low SiO<sub>2</sub> olivine pyroxenites (those which plot to the left of the Cpx-CaTs-Opx plane)  
1863  
1864 788 melt under P-T conditions that are not much different from that of peridotite but are  
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1866 789 characterized by higher melt productivity than peridotite at the same temperature and  
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1868 790 pressure (e.g. Lambart et al., 2016; Lambart, 2017). A contribution from low SiO<sub>2</sub> pyroxenite  
1869  
1870 791 is therefore most likely to take the form of a hybrid lithology formed by solid state reactions  
1871  
1872 792 (Lambart, 2017) and the resultant melt may not have an identifiable ‘pyroxenite signature’.  
1873  
1874 793 Melting experiments at 2.0 and 2.5 GPa on low SiO<sub>2</sub> pyroxenite SL77-582 (Keshav et al.,  
1875  
1876 794 2004) show that large extents of melting (>70%) of pyroxenite can produce magmas with  
1877  
1878 795 whole-rock compositions that are similar to peridotite-derived magmas, and in particular,  
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1880 796 such melts occupy the same position as some peridotite-derived melts in terms of CaO and  
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1882 797 MgO contents (Fig. 2). Lambart (2017) further showed that most of the variations in Ni  
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1891 798 content of olivine in Icelandic lavas reported by Sobolev et al. (2007) and Shorttle and  
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1893 799 Maclennan (2011) can be explained by varying the sampling pressure of the aggregated melts  
1894  
1895 800 in the melting column, with the compositions richest in Ni sampled at the highest pressure  
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1897 801 where the contribution of pyroxenite is the largest. However, modelling carried out during  
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1899 802 the current study does not require a pyroxenite source to explain the Ni-Ca-Fo relationships  
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1901 803 of Icelandic basalts, but merely requires fractionation of olivines at varying pressures.  
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1903 804 Nevertheless, a contribution from a pyroxenite lithology is not necessarily precluded on the  
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1905 805 basis of Ni contents of olivines in Icelandic basalts.

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1907  
1908 806 Fig. 13 shows the predicted CLDs for Matzen olivines with 0.365 and 0.600 wt% NiO  
1909  
1910 807 which are in equilibrium with experimental melt #370 (Keshav et al., 2004) which itself  
1911  
1912 808 represents 70% melt of low SiO<sub>2</sub> pyroxenite SL77-582 at 2.0 GPa and 1385°C. Herzberg et  
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1914 809 al. (2016) showed that olivine with 0.365 wt% NiO was characteristic of high-precision  
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1916 810 analysis of olivines having an Mg number of 89.5 that represent near solidus melts of mantle  
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1918 811 peridotite. Pyroxenite melts, and their near solidus olivines, have considerably higher NiO  
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1920 812 than melts from peridotite (e.g. Sobolev et al., 2007) and a value of 0.600 wt% NiO in near  
1921  
1922 813 solidus pyroxenite olivine has been chosen here for illustrative purposes. The CLD for 0.365  
1923  
1924 814 wt% NiO intersects the data for olivine from Theistareykir which crystallized at low  
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1926 815 pressures (~0 GPa) whereas the CLD for 0.600 wt% NiO intersects the data field for  
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1928 816 pyroxenite-derived magmas from Mauna Kea recovered during the Hawaiian Scientific  
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1930 817 Drilling Programme (HSDP-2; Sobolev et al., 2007; Herzberg, 2006). In addition, the 0.600  
1931  
1932 818 wt% NiO CLD falls immediately below the field for olivines in lavas from Koolau (Hawaii)  
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1934 819 which were derived by melting of a high SiO<sub>2</sub> pyroxenite. Reverse modelling from the  
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1936 820 Matzen olivines shown in Fig. 13 to whole-rock Ni contents gives primary magma  
1937  
1938 821 compositions which contain 463ppm and 792ppm Ni for 0.365 and 0.600 wt% NiO in olivine  
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1940 822 respectively; the former is within the estimates for near-primary high SiO<sub>2</sub> pyroxenite-

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823 derived magmas given in Sobolev et al. (2005) and the latter is within the estimates for  
824 primary peridotite-derived magmas. Hybridization of melts derived from peridotite and  
825 pyroxenite must therefore fall between the 0.600 and 0.365 wt% NiO CLDs and this is indeed  
826 the region where data for all olivines from the NAIP are found. However, the Theistareykir  
827 olivines with the higher Ni for a given Fo content are also those which fractionated at ~1.0  
828 GPa (Fig. 10) and while their relatively elevated Ni content does not *require* an input from a  
829 pyroxenite-derived melt it does not preclude it. Consequently, there is no necessity to involve  
830 a significant proportion of pyroxenite in the petrogenesis of any NAIP lavas on the basis of  
831 the trace element content of olivine.

832 It is interesting to speculate on the fact that more than 320 lavas from the NAIP yield  
833 PRIMELT3 solutions for melting of dry peridotite (Hole and Millett 2016), which does not  
834 include peridotite-derived magmas that crystallized at elevated pressures, compared to ~18  
835 lavas from the Siberian Traps and ~14 lavas from Maua Loa and Mauna Kea, even though  
836 the data bases for each area are of a similar magnitude. This is probably also an indication of  
837 the dominance of peridotite-derived melt in the NAIP. Why then, if the NAIP was the site of  
838 a mantle plume that has been active from ~61 Ma until the present, do lavas show an  
839 overwhelming signature of peridotite melting? Variations in the depth to the LAB cannot  
840 provide a viable explanation which leads to the conclusion that pyroxenite was never a  
841 significant component in the source of the NAIP magmas, and where present, it had  
842 thoroughly hybridized with peridotite. If pyroxenite was not present in any significant  
843 volume, then this suggests that the NAIP did not have the same petrological structure as that  
844 envisaged for the commonly accepted mantle plume models such as those for Hawaii or the  
845 Siberian Traps. In this connection, it may be beneficial to consider alternative models for  
846 magmatism at LIPs which do not require large scale recycling of oceanic lithosphere (e.g.  
847 Foulger 2012; Anderson and Natland 2014).

2007  
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2009 848 **9. Conclusions**  
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2012 849 Some plateau forming lavas of the BPIP have major element compositions that indicate they  
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2014 850 crystallized at <1 GPa with plagioclase joining the liquidus before clinopyroxene, the  
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2016 851 majority crystallized clinopyroxene before plagioclase feldspar over the pressure range 1.0-  
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2018 852 2.0 GPa. However, the majority crystallized olivine and clinopyroxene before plagioclase  
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2020 853 feldspar and therefore must have crystallized at >~1 GPa. Olivine within the plateau lavas  
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2022 854 have Ni and Ca contents that suggests that magmas paused at >1.6 GPa and crystallized  
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2024 855 Ol±Cpx at that depth. A few examples of near-primary magmas that underwent  
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2026 856 crystallization of olivine near the surface are found on Skye and their component olivines can  
2027  
2028 857 be successfully modelled using the method of Matzen et al. (2017) giving a maximum Ni  
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2030 858 content for BPIP olivine as 3600 ppm. MPLF lavas which fractionated Ol+Cpx in the crust  
2031  
2032 859 exhibit a CaO deficiency for a given MgO compared to lavas from west of Greenland and  
2033  
2034 860 Iceland which crystallized olivine alone near the surface. Low CaO high MgO in lavas is  
2035  
2036 861 often considered to be a characteristic sign of the involvement of pyroxenite in the source of  
2037  
2038 862 basalts. However, olivines within MPLF lavas have Fe/Mn <70 and Ni and Ca contents that  
2039  
2040 863 are entirely consistent with and origin solely from mantle peridotite. Therefore, apparent CaO  
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2042 864 deficiency in these lavas results from augite crystallization within the crust. The absence of  
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2044 865 augite phenocrysts in lavas may be a consequence of change from augite saturation to under-  
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2046 866 saturation with decreasing pressure, and consequent augite dissolution. The commonly  
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2048 867 accepted petrological structure of mantle plumes involving entrainment of recycled oceanic  
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2050 868 crust and significant contributions to magmatism from pyroxenite-rich sources cannot be  
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2052 869 applied to the flood basalts of the NAIP which is dominated by peridotite-derived melts.  
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2058 871 **Acknowledgements.**  
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2543 1073 Fig. 1. (A) Reconstruction of the North Atlantic region at about 65Ma showing the locations  
2544 referred to in the text. Pecked lines labelled A6 etc., are seafloor magnetic anomalies. Black dots  
2545 1074 numbered 55-70 are plume-head positions at the time indicated and are taken from Lawver and  
2546 1075 Müller (1994). JMFZ, Jan Mayen Fracture Zone. After Hole and Millett (2016). (B) detail of the  
2547 British Pelaeocene Igneous Province showing the position of the main magmatic centres  
2548 1076 mentioned in the text and the main structural features of the region. After Hole et al. (2015a).  
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2553 1079 Fig. 2. Schematic representation of the pressure, temperature and composition ( $P, T, X$ )  
2554 relationships for the crystallization of a typical BPIP primary magma. (a) Phase relationships  
2555 1080 calculated using Petrolog3 for the primary magma to Skye lava SK982 (Thompson, 1982; Hole &  
2556 1081 Millett, 2016). The white dots are the approximate Fo content (Mg#) of olivine that crystallizes  
2557 1082 at the Ol+Cpx+L cotectic, and the grey dots the same for the Ol+Cpx+Pl+L cotectic. % figures on  
2558 the arrows are the approximate % of olivine that can crystallize on the Ol+L liquidus at a given  
2559 1083 pressure before reaching the Ol+Cpx+L cotectic. The olivine saturation curve was calculated  
2560 1084 using the method of Sugarawa (2000) with the final pressure of melting ( $P_f$ ) from PRIMELT3  
2561 solutions (Hole & Millett, 2016) acting as a proxy for the pressure of melt segregation.  
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2567 1088 Fig. 3 (A) CaO (wt%) *versus* Mg# and b) CaO (wt%) *versus* MgO (wt%) for and Mull Plateau  
2568 Lava Formation (MPLF) lavas (grey dots), Central Mull Formation (CMF; triangles) and Baffin  
2569 1089 Island lavas (white dots). The Baffin Island and CMF data shown yield PRIMELT3 solutions for  
2570 1090 melting of dry peridotite. Liquid lines of descent (LLD) were generated using Petrolog3  
2571 (Danyushevsky & Plechov, 2011) using the QFM buffer of Kress & Carmichael (1988) and the  
2572 1091 plagioclase and clinopyroxene melt liquidus associations of Danyuchevsky (2001). Black  
2573 1092 diamonds; Cpx-in at the pressure indicated; grey diamonds, Plag-in. The LLD shown is for the  
2574 PRIMELT3 solution for Syke lava SK982 (Thompson, 1982) which has the following  
2575 1093 characteristics;  $Fe^{2+}/Fe_T=0.905$  ( $Fe_2O_3/TiO_2=1.0$ );  $T_p=1500^\circ C$ ;  $MgO=17.4$  wt%;  $P_f=3.5$  GPa;  
2576 1094  $P_f=2.9$  GPa;  $F-AFM=0.17$  (Hole & Millett, 2016). In a) note that for pressures  $<0.6$  GPa the  
2577 1095 crystallization order is  $Ol+L \rightarrow Ol+Pl+L \rightarrow Ol+Pl+Cpx+L$  whereas for pressures  $>0.6$  GPa the  
2578 1096 order is  $Ol+L \rightarrow Ol+Cpx+L \rightarrow Ol+Cpx+Pl+L$ . In (B) the dividing lines between pyroxenite- and  
2579 1097 peridotite-derived magmas and the range of possible peridotite-derived primary magmas (fine  
2580 1098 pecked lines) are taken from Herzberg & Asimow (2008). The black dots in each diagram are  
2581 1099 the samples for which high-precision olivine trace element data is available (Sobolev *et al.*  
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1104 Fig. 4. (A)  $K_D^{Ol/L}_{(FeO/MgO)}$  versus pressure for three BPIP model primary magmas calculated using  
1105 Petrolog3 for the thermodynamical model of Toplis (2005). MgO contents of the model primary  
1106 magmas were derived from PRIMELT3 the final pressure of melting ( $P_f$ ) being used as a proxy  
1107 for the pressure of melt segregation PRIMELT3 solutions for S006 (16.0 wt% MgO) and SK982  
1108 (17.5 wt% MgO) are given in Hole & Millett (2016) and for MU1.1 (20.0 wt% MgO) in the  
1109 electronic appendix. (B) variation in  $K_D^{Ol/L}_{(FeO/MgO)}$  and Mg/Fe<sub>Liq</sub> (cationic Mg/Fe of the liquid)  
1110 during crystallization of the model primary magma to SK982 for the Toplis (2005) model at  
1111 fixed pressures from 0.0 to 2.0 GPa (solid lines). Also shown is the parameterization of Herzberg  
1112 & O'Hara (2002) SK982 which assumes magma ascent and crystallization along the olivine  
1113 liquidus from the pressure of melt segregation to the surface. The grey diamonds represent the  
1114 intersections of the Toplis (2005) contours for a fixed pressure with the Herzberg & O'Hara  
1115 (2002) ascent path – see text for details. The pecked line is the approximate position of Cpx-in,  
1116 with the vertical shading on the Ol+Cpx+L side of the cotectic.

1117 Fig. 5 (A) Calculated Fo and Ca content for crystal lines of descent (CLD) for equilibrium olivines  
1118 crystallizing from two BPIP model primary magmas (SK982 and MU1.1) at 1atm pressure for  
1119 the models given in equations (8)-(11) in the text. Crystallization models were calculated using  
1120 Petrolog3 with the following parameters; QFM Buffer of Kress and Carmichael (1988);  
1121 plagioclase and clinopyroxene equilibria of Danyushevsky (2001). Crosses are at 1%  
1122 crystallization intervals. The composition of 49 olivines in equilibrium with model BPIP primary  
1123 magmas (Hole and Millett 2016) are also shown for each calculation method. (B) CLDs for the  
1124 model primary magma to SK982 at variable pressures for; grey lines, Herzberg and O'Hara  
1125 (2002); black lines, Gevrienko et al. (2016) calculation methods (equations (9)-(11) in the text).  
1126 Pressure in GPa is indicated for each CLD. Grey squares, points at which clinopyroxene joins the  
1127 crystallizing assemblage for each pressure. SQFZ, Siqueiros Fracture Zone MORB measured  
1128 olivine compositions (filled triangles) from Gavrienko et al. (2016) and compositions calculated  
1129 (open triangles) using the Gavrienko et al. (2016) model. Vectors relating to various  
1130 petrological parameters are from Gavrienko et al. (2016). The line with dot terminations is the  
1131 approximate uncertainty in Ca content propagated from equation (10) in the text.

1132 Fig. 6. (A) & (B) Ni content (ppm) versus Fo content for olivines in MPLF lavas. The CLDs shown  
1133 were calculated for the primary magma to SK982 and are for; (i) the Beattie *et al.* (1991) model  
1134 at 1 atm (short dashes line); (ii) the Herzberg & O'Hara (2002) model for crystallization and  
1135 ascent along the olivine liquidus (long dashes) and (iii) the Herzberg & O'Hara (2002) model  
1136 until 1.6 GPa and then at a fixed pressure of 1.6 GPa (solid back line). For model (iii)  
1137  $K_D^{Ol/L}_{(FeO/MgO)}$  values were calculated from Mg/Fe<sub>Liq</sub> as shown in Fig. 2(A), propagating to give  
1138  $D^{Ol/L}_{NiO}$  from equation (3) in the text. The large white diamond represents the Fo content for

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1139 Cpx-in, and at this point  $D^{Cpx/L}_{NiO}$  was assumed to be 0.25  $D^{Ol/L}_{NiO}$ . Grey curves are mixing lines  
1140 between the primary magma composition and derivative magmas along the CLD (see also  
1141 Herzberg *et al.*, 2016) with the approximate MgO content of the derivative magma indicated. It  
1142 was assumed that mixing was complete and that neither liquid contained phenocrysts at the  
1143 time of mixing. 'Matzen olivines' are model olivine compositions for BPIP primary magmas  
1144 segregating at  $P_f$ , undergoing rapid emplacement at the surface without crystallizing any  
1145 olivine, and then crystallizing olivine at 1 atm (equation (7) in the text. The cross-hatched field  
1146 in (a) is for olivines from Siqueiros Fracture Zone MORB (Sobolev *et al.* 2007; Gavrilenko *et al.*  
1147 2016) and is shown for comparison. In (b) Skye and Mull olivines are near-primary olivines in  
1148 picrites (Spice *et al.*, 2016). These include sample MU1.1 for which a Toplis model is shown in  
1149 Fig. 2.

1150 Fig. 7. (A) and (B) Ca content (ppm) *versus* Fo content for olivines for the same MPLF lavas as in  
1151 Fig. 6. The cross-hatched area delimits the range of Ca and Fo contents of olivines for 49  
1152 forward crystallization models of BPIP primary magmas at 1 atm using the Beattie *et al.* (1991)  
1153 parameterization for  $D^{Ol/L}_{CaO}$  given in equation (4) in the text. The stippled field uses the same  
1154 primary magma data but for the Gavrilenko *et al.* (2016) method of calculating  $D^{Ol/L}_{CaO}$  given in  
1155 equations (10) and (11) in the text. Grey squares are the composition at which clinopyroxene  
1156 joins the crystallizing assemblage. The field labelled SQFZ encompasses measured olivine  
1157 compositions for Siqueiros Fracture Zone MORB from Gavrilenko *et al.* (2016)

1158 Fig. 8. (A) calculated CLDs for Ca and Ni content of olivines at variable pressures using the Ni  
1159 and Ca model of Herzberg and O'Hara (2002) and the Ca model of Gavrilenko *et al.* (2016). The  
1160 Ni model has been modified for appropriate final pressure of equilibration as discussed in the  
1161 text and shown in Fig. 3. The black lines with arrows are for the Herzberg and O'Hara (2002) Ca  
1162 model at the pressures indicated in GPa. Fine lines with various ornaments are for the Ni  
1163 model of Herzberg and O'Hara (2002) with Ca calculated according to the Gavrilenko *et al.*  
1164 (2016) model, and the thick grey line is for 1 atm pressure, which is the same as the olivine. Grey  
1165 squares are the compositions at which clinopyroxene joins the crystallizing assemblage for a  
1166 given pressure and the white squares the same for plagioclase feldspar. The curved inflections  
1167 giving negative 'spikes' for pressures >1.2 GPa result from clinopyroxene joining the liquidus  
1168 before 11 wt% MgO, the MgO content at which the calculation  $D^{CaO}_{Ol/L}$  changes from equations  
1169 (10) to (11) in the text. Matzen olivines are for olivine in equilibrium with 49 BPIP model  
1170 primary magmas. The double-headed arrow illustrates the effect of crystallization of ~15%  
1171 olivine at 1 atm. (B) and (C) Ni and Ca contents of MPLF olivines. The cross hatched area  
1172 represents the range of compositions for forward crystallization of BPIP model primary  
1173 magmas at 1 atm and therefore represents the olivine liquidus. The thick grey line is the CLD for

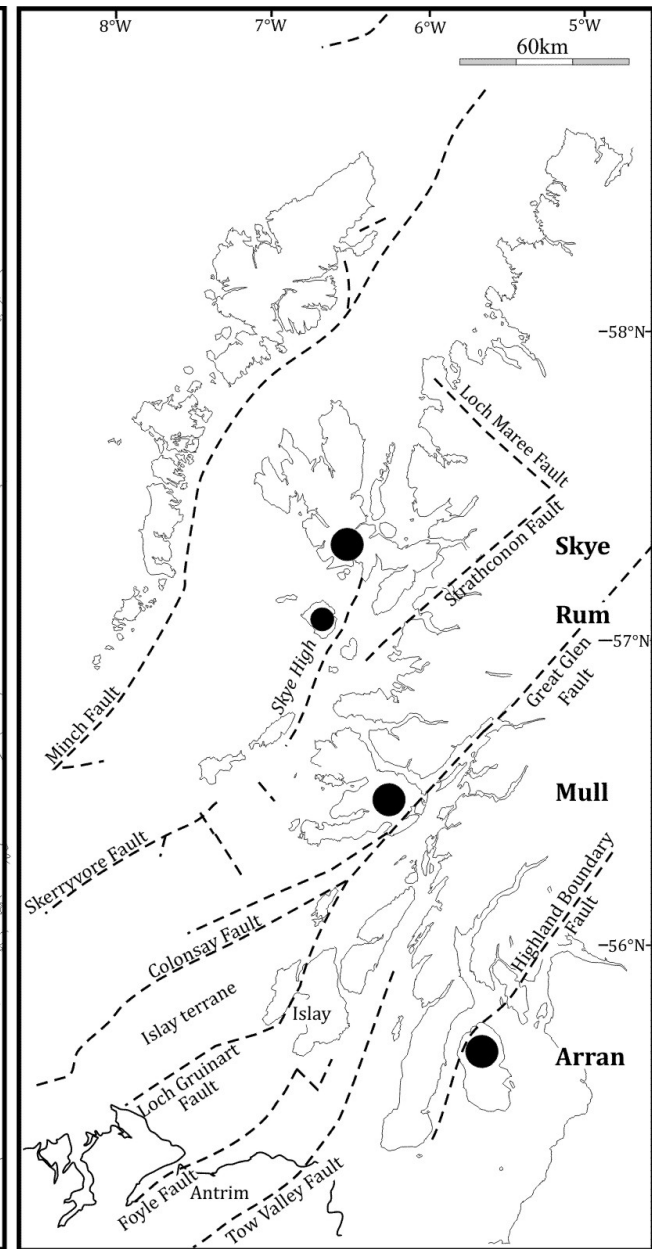
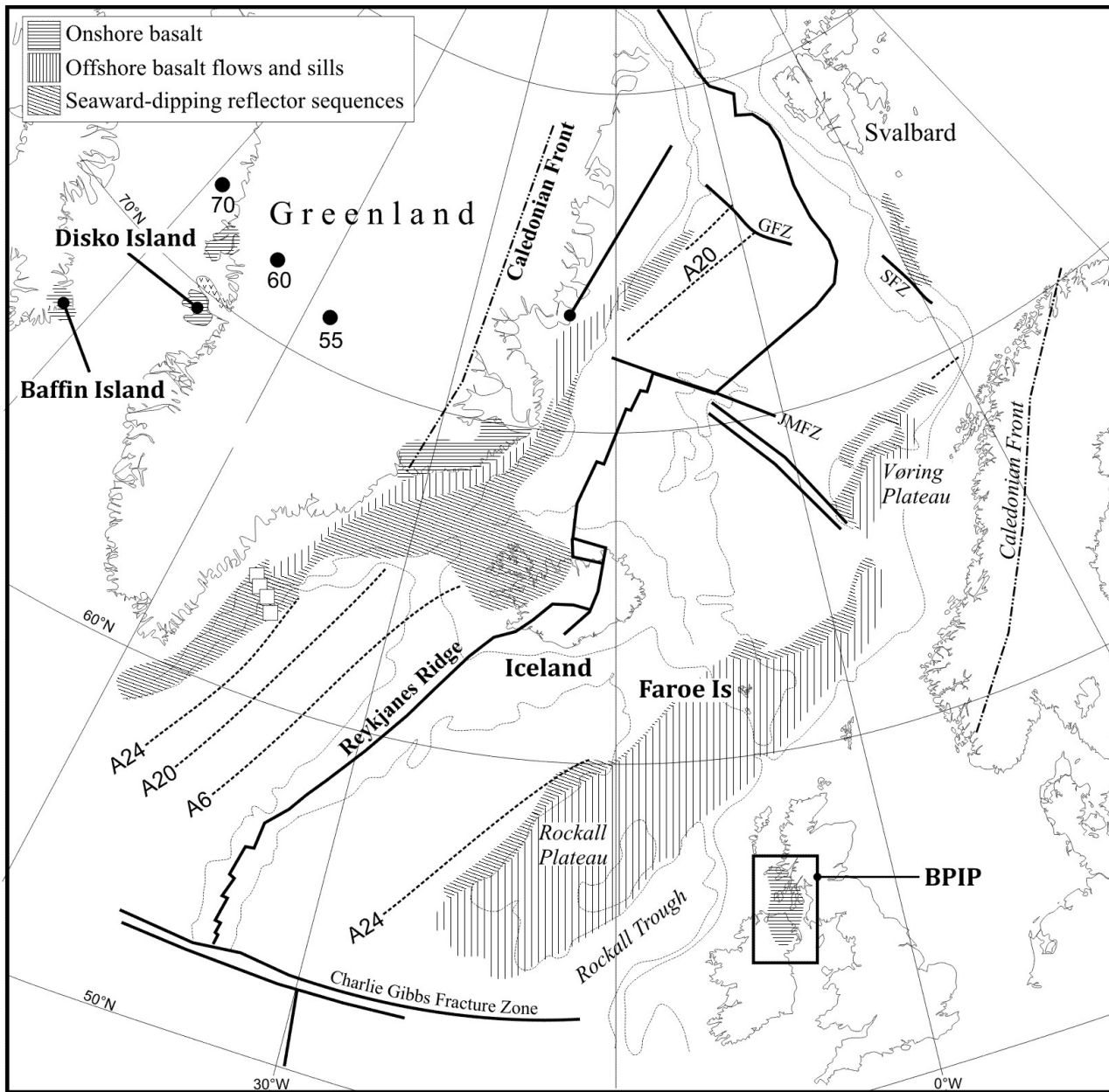


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2717 1174 sample SK982 at 1 atm, and the lines labelled 'Beattie' and 'H&O' are 1atm CLDs for the Beattie  
2718  
2719 1175 et al. (1991) and Herzberg and O'Hara (2002) calculation methods.  
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2721 1176 Fig. 9. Fe/Mn (ppm) *versus* Fo content for olivine in BPIP, Baffin Island and West Greenland  
2722  
2723 1177 (Disko Island) lavas. The fields for primary olivines derived from melts of mantle peridotite are  
2724 1178 labelled with their source mineralogy i.e. harzburgite or lherzolite. The two pecked horizontal  
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2726 1179 lines (Fe/Mn 60 and 70) are the upper and lower boundaries respectively for olivines that  
2727 1180 crystallize from peridotite-derived magmas. After Herzberg (2011).  
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2729 1181  
2730 1182 Fig. 10. covariations between Ni and Ca content of lavas from (A) Theistareykir, Iceland, (B)  
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2732 1183 Baffin Island and (C) Disko Island, west Greenland. Stippled areas are the range of predicted Ni  
2733 1184 and Ca contents for the crystallization of model primary magmas (Hole and Millett 2016) at 1  
2734  
2735 1185 atm from each location. The grey line on each diagram is the calculated Ol+L CLD at 1 atm for a  
2736 1186 model primary magma from each location. Model primary magmas are as follows; Disko Island,  
2737  
2738 1187 #410188 (Larsen and Pedersen 2000); Baffin Is, BI/CS/14 (Starkey et al. 2009); Theistareykir,  
2739 1188 #9302 (MacLennan et al. 2003). In (c) grey symbols are olivines considered by Herzberg et al.  
2740  
2741 1189 (2016) to be near primary, and open circles are olivines considered by Herzberg et al. (2016) to  
2742 1190 be the result of crystallization from mixed magmas. The Ni content of Matzen olivines were  
2743  
2744 1191 calculated from PRIMELT3 solutions using equation (7) and the Ca content using equation (10).  
2745 1192 The cross-hatched area is the field for olivine data from MPLF sample BHL15 for comparison  
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2747 1193 with Fig. 7. Data are from Sobolev *et al.* (2007) and Herzberg *et al.* (2016).  
2748 1194  
2749 1195 Fig. 11 Olivine (vertical lines) and clinopyroxene (curves) saturation calculated using the  
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2751 1196 method of Chen & Zhang (2008; 2009). To the left of the curves, liquids are undersaturated with  
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2753 1197 respect to olivine or clinopyroxene. At 1.5 GPa, the temperature estimate of 1390°C is the  
2754 1198 temperature estimate immediately prior to the Cpx-in temperature from a Petrolog3 forward  
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2756 1199 crystallization model for the primary magma to lava SK982. The temperature at 1.0 GPa  
2757 1200 assumes a 55°C/GPa<sup>-1</sup> gradient on the olivine liquidus (Sugawara 2000; Matzen et al. 2017).  
2758 1201 Black dots are the whole-rock compositions of the MPLF lavas for which olivine data are  
2759  
2760 1202 available. The shaded area is the region of instability of Cpx in the melt caused by  
2761  
2762 1203 decompression.  
2763 1204  
2764 1205 Fig. 12. SiO<sub>2</sub>/MgO versus Mg# for (A) Hawaii Scientific Drilling Project lavas, (B) MPLF lavas and  
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2766 1206 (C) lavas from Theistareykir. For each sample set forward crystallization models have been  
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2768 1207 generated at varying pressures from 0.0 to 1.8 GPa using Petrolog3, and starting with  
2769 1208 PRIMELT3 primary magma compositions. The shaded area on each diagram is the field for  
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2772 Hole, MJ. Polybaric fractionation CFB  
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1209 Ol+Cpx+L. The insets are CaO *versus* MgO for the same lavas, the black line indicating the  
1210 dividing line between peridotite- (above the line) and pyroxenite-derived magmas taken from  
1211 Herzberg & Asimow (2008). The CaO-MgO relationships for MPLF lavas are shown in Fig. 2.  
1212 Note the extended scale in (C).

1213  
1214 Fig. 13. Ni (ppm) *versus* %Fo in olivine for basalts from Theistareykir, and pyroxenite-derived  
1215 lavas recovered during the Hawaii Scientific Drilling Project (HSDP-2) and from Koolau (shield  
1216 stage). Only olivine with Fo>86 are given in Sobolev et al. (2007) for HSDP-2. The open  
1217 diamond is the composition of Matzen olivine, assuming an NiO content 0.365 wt%, in  
1218 equilibrium with experimentally generated melt #370 of Keshav et al. (2004) which represents  
1219 70% melt of pyroxenite SL77-582. The whole-rock content of this magma would be ~460 ppm  
1220 Ni which is within the range for natural olivine in peridotite-derived magmas. Black diamond is  
1221 the composition of Matzen olivine, assuming and NiO content of 0.600 wt%, in equilibrium with  
1222 melt #370. In this case the whole-rock composition would be 792 ppm Ni which is within the  
1223 range of parental melts to Hawaiian pyroxenite-derived magmas (Sobolev et al., 2005). The  
1224 pecked lines represent the CLD at 1 atm from the Matzen olivine indicated.



Temperature °C

