

Revision 2

The Mantle Source of Thermal Plumes: Trace and Minor Elements in Olivine & Major Oxides of Primitive Liquids (And Why the Olivine Compositions Don't matter)

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ABSTRACT

We estimate the mantle source compositions for mantle plumes, and by implication Earth's lower mantle, by: (a) measuring trace (e.g. Sc, V, Cu) and minor (e.g., Ca, Mn, Ni) element concentrations of high forsterite olivine grains from several plume localities, (b) estimating the parent liquid compositions from which they crystallized, (c) calculating mantle potential temperatures and degrees of partial melting and (d) estimating trace element compositions of depleted and enriched mantle sources. Our sample set includes two continental flood basalt provinces (Emeishan and Deccan), a flood basalt that erupted in a continental rift setting (Baffin Island), our type example of a thermal mantle plume (Hawaii) and lavas from the Siqueiros Transform at the East Pacific Rise, which represent the mid-ocean ridge system. We also present olivine compositions for the peridotite xenoliths from Kilbourne Hole, New Mexico, USA, which are commonly used as primary and secondary analytical standards. We find that trace elements in lava-hosted olivine grains are too far removed from their mantle source to provide anything but greatly hindered views of such. Olivine compositions reflect not only evolving liquid compositions (including partial melting conditions and later fractionation), but also evolving Ol+liq partition coefficients, which mostly increase with decreasing T during crystallization. Mantle compositions, delimited by maximum forsterite contents and estimates of parental magmas (and experimentally determined partition coefficients) indicate that our selected plumes reflect some combination of (1) a depleted mantle source that is quite similar to that obtained by other methods, and (2) a variably enriched plume source that is more enriched than current estimates of pyrolite. The enriched plume mantle sources can be explained remarkably well as a mixture of subducted mid-ocean ridge basalt (MORB; Gale et al. 2013) and depleted MORB mantle (DM; Salters and Stracke 2004), with MORB:DM ratios of 1:5 to 1:4. These ratios are most sensitive to

46 estimates of melt fraction where plume parental magmas are last equilibrated with their
47 mantle source, but are nonetheless consistent across a wide range of chemically very
48 different elements, and estimates of MORB and DM obtained by very different means.
49 Baffin Island is of particular interest. Like prior studies, we verify a high mantle potential
50 temperature (T_p) of 1630°C (compared to $T_p = 1320$ -1420°C for MORB from Cottrell and
51 Kelley 2011 for Ol of Fo89.3-91.4). The Baffin source is also within error the same as
52 DM with respect to trace elements, although still isotopically distinct; Baffin appears to
53 be sourced in something that is akin to DM that lies at the base of the mantle, where
54 plumes acquire their excess heat. Thus while part of our analysis supports the concept of
55 a “slab graveyard” at the bottom of the lower mantle (e.g., Wyession 1996), that cemetery
56 is by no means ubiquitous at the CMB: subducted slabs are either unevenly interred, or
57 efficiently excavated by later upwellings.

58

59 INTRODUCTION

60 An enduring obscurity concerns the composition and mineralogy of Earth’s lower
61 mantle—we may indeed have a better understanding of Pluto’s surface (Moore et al.
62 2016) than Earth’s lower reaches. As to the upper mantle, mantle xenoliths are a key
63 source of information, since they are just one step removed (they are often residues of
64 prior melt extraction) from the mantle source that gives rise to basalts (e.g., McDonough
65 and Sun 1995; Salters and Stracke 2004; Workman and Hart 2005). Basalts, if
66 sufficiently primitive, are also just one step removed from a pre-melt extraction mantle,
67 and their compositions have been used to infer, for example, that subducted crust may be
68 admixed into the sources of MORB (Hirschmann and Stolper, 1996; Waters et al. 2011)
69 and ocean island basalts (OIB) (e.g., Hawkesworth et al. 1979; Hofmann and White
70 1982; Prytulak and Elliott 2007; Jackson et al. 2008). OIB are in turn thought to derive as
71 deep-seated thermal plumes from the core-mantle boundary (Morgan 1972), and so their
72 sources may provide our most direct view of the lower mantle.

73 Olivine (Ol) phenocrysts have been offered as an additional, perhaps less biased, view
74 of MORB and OIB mantle sources (e.g., Sobolev et al. 2007). One obvious advantage is
75 that primitive Ol grains are highly resistant to the mixing processes that homogenize

76 magmatic liquids, and so might preserve what is otherwise lost to magma mixing.
77 Clearly, diffusion may later homogenize phenocrysts (Thomson and Maclennan 2013),
78 but the diversity of disequilibrium Ol grains demonstrates that the process is quite
79 incomplete. As a test, we present new analyses of trace (Ti, V, Cr, Cu, etc.) and minor
80 elements (Ni, Ca, Mn, etc.; for simplicity, herein referred to as “trace” elements) in
81 olivine grains from several plume localities, and a MORB from the East Pacific Rise. Our
82 data show that olivine compositions provide a rather opaque view of mantle composition,
83 especially compared to their host lavas. For some elements (Ni and Cr) olivine
84 compositions reveal quite little about mantle source regions (instead recording how their
85 parent magmas were generated, i.e., at high P or T or high melt fraction), while for others
86 (Sc and Ti, and possibly Ca) signatures of the mantle source(s) may be preserved, but
87 through a connection that is not perfectly clear.

88 We first review some crucial, perhaps under-appreciated, controls on oceanic volcanic
89 compositions, and how olivine compositions respond to T and liquid composition. Our
90 analysis, which includes some new estimates of mantle-equilibrated parental magmas and
91 the mantle potential temperatures at which they were formed, indicate that the lower
92 mantle contains a depleted component similar to depleted MORB mantle, which is herein
93 noted as DMM [“DM” in Stracke and Salters (2004) and “DMM” in Workman and Hart
94 (2005) with further variations in Shimizu et al. 2016]. This finding is tentative to be sure,
95 but not new, as the FOZO component of Hart et al. (1992) has near DMM-like $^{87}\text{Sr}/^{86}\text{Sr}$
96 (0.70248) and $^{143}\text{Nd}/^{144}\text{Nd}$ (0.5131) and so is clearly quite depleted. What remains clear
97 is that progress will require us to resurrect a pan-periodic chart view of mantle-derived
98 liquids (e.g., Stracke 2012; White 2015), and direct comparisons (e.g., MORB-

99 normalized, rather than the less-useful but habitual chondrite normalized trace element
100 diagrams) of mantle sources, the latter of which are sorely lacking. Ultimately, we plan
101 to show the efficacy of using combined rock composition, experimental and phase
102 chemical data to better constrain mantle plume sources.

103

104 **BACKGROUND**

105 **Approach & Motivation**

106 Our new trace element analyses of high forsterite (Fo; Mg_2SiO_4) olivine phenocrysts
107 (ca. Fo>85) derive from (1) our archetype of a mantle plume, Hawaii, (2) two continental
108 flood basalt provinces, Deccan and Emeishan, (3) a flood basalt province at a rifted
109 margin, Baffin Island, and (4) a “D-MORB” or “depleted MORB” (derived from an
110 especially chemically depleted DMM, see Waters et al. 2011) from the Siqueiros
111 Transform (Perfit et al. 1996; Putirka et al. 2011), which is part of the East Pacific Rise.
112 We also present new data for olivine grains from Kilbourne Hole mantle xenoliths (often
113 known as “KLB”), collected by the lead author, which we use as both an analytical and
114 petrologic reference.

115 Our choice of samples is predicated on the idea that oceanic islands and flood basalts
116 are created by thermal mantle plumes, that these plumes have excess heat (Herzberg
117 1995; Herzberg and Gazel 2009; Li et al. 2012; Putirka 2016), and that they gain much of
118 this heat from the underlying core (e.g., Hess, 1962; Morgan 1971; Lay et al. 2008), or
119 from U-, K- and Th- enriched regions of the lowermost mantle (Garnero et al. 2016).
120 Flood basalts and OIB thus may entrain deep-seated mantle material (e.g., Kellogg et al.

121 1999; Campbell and O'Neill 2012; Li et al. 2014). In contrast, mid-ocean ridge basalts
122 (MORB) derive from passive upwelling of shallow upper mantle (McKenzie 1967).

123 A key motivating work is that of Sobolev et al. (2007), who observe that olivine grains
124 from many plumes exhibit peculiarly high Ni contents; they suggest that high Ni-in-Ol
125 requires parental liquids that formed by partial melting of a pyroxenite (rather than
126 peridotite) source. Their pyroxenite source is putatively created by mixing ambient
127 mantle peridotite with partial melts of deeply subducted crust; this reaction may convert a
128 former peridotite source to a near olivine-free, pyroxene-rich lithology. Some dispute this
129 interpretation (e.g., Matzen et al. 2009, 2013; Putirka et al 2011), arguing that high Ni-in-
130 Ol can be obtained by crystallization of mafic magmas over temperatures ranging from
131 1500°C for moderate Ni contents to 1375°C for the highest Ni-in-Ol (Putirka et al. 2011,
132 their Fig. 5). Others suggest that high Ni-in-Ol reflects mass exchange between a mantle
133 source and Earth's Ni-rich core (Herzberg et al. 2013, but see also Herzberg et al. 2016
134 for a moderating view). Modeling efforts suggest that pyroxenite- and peridotite-derived
135 partial melts may have similar major oxide compositions (Jennings et al. 2016; Lambart
136 et al. 2016), and our tests indicate the same, although some pyroxenite-derived melts
137 (e.g., Kogiso et al. 2001) range to very low Al₂O₃ (<12 wt. %, with SiO₂/Al₂O₃ ratios >4)
138 and very high CaO (>13.8 wt. %), that are less than typical for peridotite partial melts
139 (mostly >12% Al₂O₃, <14% CaO, SiO₂/Al₂O₃ < 5). And surely, Ol compositions derived
140 from such liquids should inherit these characteristics. In addition, trace elements that
141 feature significant contrasts in partitioning behavior between Ol and either of pyroxene or
142 garnet should provide reliable tests. For example, a pyroxenite source should yield Sc-
143 depleted liquids compared to a peridotite source, and Ol grains should inherit such a

144 mineralogy-dependent influence. In a similar manner, if high Ni contents derive from a
145 core-contaminated mantle, then other (easily analyzed) siderophile elements, such as Co,
146 Cr, Zn and P should also be enriched.

147

148 **Essential Contrasts Between OIB and MORB: The Influences of Pressures and** 149 **Temperatures on Partial Melting**

150 A long-observed truth (e.g., Langmuir and Hanson 1980) is that primitive OIB have
151 higher FeO compared to MORB (Fig. 1A). This fact has necessary consequences.

152 1) Higher FeO in OIB almost certainly results from higher P - T conditions of partial
153 melting. It is experimentally well-verified that as P increases, peridotite partial melts shift
154 towards olivine in composition (Stolper 1980; Longhi 2002) and so *liquids generated at*
155 *higher P-T conditions are enriched in elements compatible in olivine*, such as FeO and
156 MgO (see Langmuir and Hanson 1980), as well as Co and Ni, regardless of source
157 composition. If MORB and OIB are generated from peridotites then major oxide
158 compositions imply OIB genesis at $P = 20$ -50 kbar (Fig. 1; Putirka et al. 2011), while
159 MORB are generated at 8-15 kbar (Fig. 1). The necessary effect is that primitive OIBs—
160 and their Ol phenocrysts—have higher Co, Fe and Ni contents relative to MORB as a
161 consequence of partial melting of an olivine-bearing source.

162 2) Because FeO and other compatible elements are affected by the P - T conditions of
163 partial melting, seemingly anomalous ratios of Zn/Fe, Mn/Fe, Co/Fe etc., are as likely to
164 reflect mantle melting conditions (e.g., low Zn/Fe = high melt fractions at high P - T
165 conditions, since these yield high FeO^{liq}) and not necessarily source composition or
166 mineralogy. Our review of peridotite partial melting experiments show that FeO/MnO

167 ratios are far from constant during partial melting, ranging from 45-60 at $P = 40$ to 70
168 kbar, and up to >85 at $P = 20$ to 35 kbar P (Fig. 1C), and new experiments by Matzen et
169 al. (2017) yield the same result. Undoubtedly, FeO/MnO, and other such ratios also vary
170 with source mineralogy (e.g., Humayun et al. 2004; Le Roux et al. 2010; Davis et al.
171 2013)—but only variance outside the ranges illustrated in Fig. 1C can disallow a
172 peridotite source, and such variances are insufficiently explored—particularly for a
173 pyroxenite source.

174 3) It is a grave error to assign an equal T or fractionation extent to olivine grains at an
175 equivalent Fo content. Since OIB have greater FeO than MORB, their Ol grains can only
176 reach an equivalent Fo (as Ol from MORB) when parent magmas have higher MgO, and
177 consequently higher T . The comparison of equivalent Fo contents is tempting, given our
178 familiarity with simple systems (e.g., Bowen and Schairer 1935). But Green et al. (1999)
179 incorrectly hypothesized that MORB and OIB derive from partial melting at equivalent T
180 as both yield Fo91 olivine; their interpretation is flawed because Hawaiian magmas have
181 greater FeO (FeO^{liq}) than MORB (Putirka 2005). For example, if we apply an Fe-Mg
182 exchange coefficient, $K_D(\text{Fe-Mg})^{\text{ol-liq}}$, of 0.30 (where $K_D(\text{Fe-Mg})^{\text{ol-liq}} =$
183 $[\text{X}_{\text{FeO}}^{\text{ol}}/\text{X}_{\text{MgO}}^{\text{ol}}]/[\text{X}_{\text{FeO}}^{\text{liq}}/\text{X}_{\text{MgO}}^{\text{liq}}]$, and X_i^j are mole fractions of i in the phase j), then to
184 crystallize Fo91 olivine from a liquid with 8 wt. % FeO^{liq} (typical of MORB) we must
185 have 13.6% MgO^{liq} . But a liquid with 11 wt. % FeO (typical at Hawaii) must have 18.7%
186 MgO^{liq} to crystallize the very same olivine. Liquids with 13.6 and 18.7% MgO can be
187 produced from the same mantle, but only at very different P - T conditions and melt
188 fractions (F), and the parent liquids (and their equilibrium Ol) will thus have very
189 different trace element contents (Fig. 1).

190 These and other errors befall any analysis of olivine-compatible elements, as the P - T
191 conditions of partial melting are likely to exert a primary control.

192

193 **Methods**

194 We analyzed major oxides as well as trace elements in olivine that range from
195 lithophile (Ca, Ti, Al, Na, Cu, Sc) to moderately siderophile (Mn, V, Cr, Zn, Co, Ni, P)
196 (see Electronic Appendix 1). Olivine compositions were measured by LA-ICP-MS at the
197 State Key Laboratory of Ore Deposit Geochemistry, Institute of Geochemistry, Guiyang,
198 China; see Liu et al. (2008) for details of methods. In Fig. 2, elements are arranged in
199 order of increasing Ol/(silicate)liq compatibility and metal/silicate compatibility, using
200 Laubier et al. (2014), Le Roux et al. (2015), Siebert et al. (2011; their Table 6) and the
201 GERM website (<https://earthref.org/GERM/>).

202 To understand the origin of our Ol compositions we consider how Ol saturated liquids
203 may have evolved, by calculating Liquid and Crystal Lines of Descent (LLDs and CLDs
204 respectively), which trace the evolution of co-existing liquid and crystal compositions
205 (our high Fo Ol grains are from mafic lavas that are Ol saturated only). These curves are
206 derived from olivine/liquid elemental partition coefficients. For the partitioning of Ni
207 between olivine and liquid we use Matzen et al. (2013), supplemented by Jurewicz et al.
208 (1993) and Taura et al. (1998); the latter two studies are mutually consistent with the
209 high-precision work of Matzen et al. (2013) and allow calibration of $D_{Ni}^{ol-liq} = f(T)$ to be
210 extended to both higher and lower temperatures (note that Herzberg et al. (2016) provide
211 a precise model $D_{Ni}^{ol-liq} = f(D_{MgO}^{ol-liq})$, but our key objective is to make the T -sensitivity
212 of D_{Ni} explicit). Using Jurewicz et al. (1993) only, we obtain:

$$213 \quad D_{Ni}^{ol-liq} = e^{\left(\frac{8414}{T} - 4.9\right)} \quad (1a)$$

214 Equation (1a) is calibrated over the range $T = 1140\text{-}1325^\circ\text{C}$ at $P = 0.0001$ GPa, and MgO
215 = 5-10 wt. %. It reproduces the calibration data ($n=17$) with a standard error of estimate
216 of ± 1.3 , with R^2 of 0.82. Using Matzen et al. (2013) (0.0001-3 GPa; 1400-1600°C), we
217 obtain:

$$218 \quad D_{Ni}^{ol-liq} = e^{\left(\frac{3349}{T} - 0.79\right)} \quad (1b)$$

219 Combining the Matzen et al. (2013) and Jurewicz et al. (1993) we have:

$$220 \quad D_{Ni}^{ol-liq} = e^{\left(\frac{4639}{T} - 1.71\right)} \quad (1c)$$

221 which reproduces the calibration data ($n=29$) to ± 1.2 with $R^2 = 0.85$. And combining
222 Matzen et al. (2013), Jurewicz et al. (1993) and Taura et al. (1998) data (3.0-14.4 GPa;
223 1600-2000°C) yields:

$$224 \quad D_{Ni}^{ol-liq} = e^{\left(\frac{5853}{T} - 2.7\right)} \quad (1d)$$

225 and reproduces the calibration data ($n=40$) to ± 1.0 with $R^2 = 0.92$. Equation 1d is valid
226 over the ranges $T = 1140\text{-}2000^\circ\text{C}$, $P = 1$ atm – 14.4 GPa, and $\text{MgO}^{liq} = 5.1\text{-}37.5$ wt. %.

227 We also created models using Wang and Gaetani (2008; 0.0001 GPa; 1200-1325°C),
228 whose eclogite liquid-equilibrated olivines yield very high D_{Ni}^{ol-liq} ($D_{Ni}^{ol-liq} = 22$), as do
229 some other experiments cited by Le Roux et al. (2015). But as we show below, D_{Ni} from
230 Jurewicz et al. (1993) (Eqn. 1a) appear to better explain certain of our observed plume Ni
231 v. Fo trends.

232 Because D_{Ni} is sensitive to T, it is possible to use D_{Ni} to estimate T, which for
233 statistical reasons requires a separate calibration. Using Matzen et al. (2013) and Jurewicz
234 et al. (1993) we obtain:

$$235 \quad T(^{\circ}\text{C}) = (4.21 \times 10^{-4} + 1.87 \times 10^{-4} [\ln(D_{Ni})])^{-1} \quad (2)$$

236 which reproduces T for the calibration data to $\pm 58^\circ\text{C}$ with $R^2 = 0.86$.

237 To facilitate the calculations of LLD and CLD curves, we also employ two additional
238 empirical equations. First, from stoichiometric and mass balance constraints, we have:

$$239 \quad Fo^{ol} = 23.55 + 1.3462MgO^{ol} \quad (3)$$

240 which reproduces Fo of olivine to ± 0.05 , with $R^2 = 1.0$. And to predict MgO in Ol we
241 use:

$$242 \quad MgO^{ol} = a - \frac{b}{MgO^{liq}} \quad (4)$$

243 where $a = 55.45$ and $b = 106.1$, when FeO^{liq} (of the equilibrated liquid) is 11.5 wt. %.

244 Equation (4) is specific to FeO^{liq} content and reproduces MgO^{Ol} to ± 0.1 wt. % with $R^2 =$
245 1.0. For $FeO^{liq} = 8\%$ (e.g., MORB), $a = 55.63$; $b = 75.78$; for $FeO^{liq} = 13\%$ (high FeO
246 OIB), $a = 55.45$; $b = 120.0$).

247 We also construct LLD and CLD curves for several of the other trace and minor
248 elements in Ol (Ca, Ti, Al, Na, Cu, Sc, Mn, V, Cr, Zr, Co and P); these also make use of
249 Eqns. (3) and (4), and olivine/melt partition coefficients from Laubier et al. (2014), Le
250 Roux et al. (2015) and the GERM website (<https://earthref.org/GERM/>). While some of
251 these elements quite likely have partition coefficients that are sensitive to T , only the
252 partition coefficients for Mn, Ti and Cr are known with sufficient experimental precision
253 to calibrate $D_i^{ol-liq} = f(T)$, and so for all other elements we use $D_i = \text{constant}$.

254 Finally, we calculate mantle source compositions, and test whether such mantle
255 sources were contaminated by a metallic core component. To calculate a mantle source,
256 we use the approach of Putirka (2016) by using observed whole rock compositions (see
257 Fig. 3, caption) as input, to predict an equilibrium Ol composition, and then test whether
258 any such liquids can yield our observed maximum Fo contents observed at each volcanic

259 system (we assume $fO_2 = \text{Ni-NiO}$; see Putirka 2016). Output from the Putirka (2016)
260 models yields the degree of partial melting, F , and the P - T conditions at which such melt
261 is generated, and an MgO^{liq} for a mantle-equilibrated parental magma. The estimate of F
262 is one of the least certain and yet most important variables when calculating a mantle
263 source, and so we consider a range of predicted values of F in such calculations (see
264 electronic supplements). We then infer the trace element contents of such a liquid from
265 whole rock variation diagrams (see Fig. 3, caption for data). As an example, the
266 concentration of Ni in a parental liquid, Ni^{liq} , is the median concentration of Ni in whole
267 rocks having the calculated values of MgO^{liq} . Table 1 contains a summary of P - T and
268 melt fraction estimates, and the predicted Fo contents of Ol (using Putirka 2016) for our
269 calculated parental magma compositions (additional outputs are in electronic supplement
270 2). Finally, we use these trace element contents, our predicted T of melting (to estimate
271 D_i when $D_i = f(T)$), our range of predicted values of F , and D_i values from Putirka et al.
272 (2011), updated with partition coefficients from Davis et al. (2013), Laubier et al. (2014),
273 and Le Roux et a. (2015), to calculate a mantle source composition. Putirka et al. (2011)
274 assume batch melting for this calculation, as the error associated with choice of melt
275 model is deemed trivial compared to the errors on estimating a parental magma
276 composition (e.g., Langmuir et al. 1992; Putirka et al. 2011).

277

278

279 **Results**

280 Table 1 summarizes our estimates for the P - T conditions of parental magma extraction
281 from the mantle, the melt fraction at extraction, and mantle potential temperatures (T_p)

282 [assuming fO_2 is buffered at NNO; Putirka 2016)]. Melt fractions are slightly higher than
283 in Putirka et al. (2011), as they are based on new models in Putirka (2016); T_p estimates
284 are slightly lower than in Putirka (2008) because a careful look at just the high precision
285 data of Putirka et al. (2011) indicates slightly lower maximum Fo contents for Ol
286 phenocrysts (at Hawaii, Fo90.5 instead of Fo91.3 in Putirka et al.'s (2011) Table 3 and
287 appendices), and these lower values are consistent with our new results (Fo90.0 for
288 Hawaii; Table 1; electronic appendix 1).

289 Figure 2 provides a graphical summary of our trace element results. Here, our trace
290 element analyses of Ol grains are compared at a constant value of Fo89 (the highest Fo
291 content common to all our analysis suites), with each plume locality normalized to the
292 trace element contents of Fo89 Ol from our MORB sample (Siqueiros transform). To
293 create Fig. 2, we find the trace element content of Fo89 Ol by taking the average of
294 observed olivine compositions that fall (mostly) in the range Fo87-Fo90 (weighted to
295 yield a mean of Fo89), or for Deccan (where all our Ol compositions are <Fo89), by
296 projecting M v. Fo (where M = a metal cation, such as Co, Na, Ti, etc.) trends back to
297 Fo89, as Klein and Langmuir (1987) correct MORB compositions.

298 As we discuss below, these trace elements in olivine are controlled by both parent
299 liquid compositions (in turn affected by P - T conditions of partial melting), and the P - T
300 conditions of Ol crystallization, and can be divided along lines of partitioning behavior:
301 1) Elements highly incompatible in olivine ($D_i^{ol-liq} \ll 1$). For these (e.g., Ca, Ti, V, Al, Na,
302 Sc), concentrations in Ol tend to be uncorrelated with Fo contents, perhaps because of
303 post-crystallization diffusion for at least some fast-diffusing elements (e.g., Thomson and
304 Maclennan 2013), and because of this, they might not always reliably track liquid

305 compositions, or source composition. To give an example of the problem, consider that
306 Prytulak and Elliott (2007) observe that plume lavas have elevated Ti relative to
307 MORB—and our plume-derived Ol (except Baffin) are similarly enriched in Ti (Fig. 3A,
308 B); so far, so good. Published experimental values of $D_{\text{Ti}}^{\text{ol-liq}}$ range widely (0.003 to
309 0.21), and we find that only a very low and constant $D_{\text{Ti}}^{\text{ol-liq}}$ (=0.007 in Fig. 3A, B)
310 simultaneously yields a Liquid line of Descent (LLD) that reproduces whole rock MgO-
311 TiO₂ trends (Fig. 3A) and a Crystal Line of Descent (or CLD, which tracks crystal
312 compositions with fractionation; see Putirka et al. 2011) that approximates the mean Ti in
313 olivine (but is otherwise insensitive to Fo content; Fig. 3B). So here, we capture the
314 broader inter-suite Ti contents (Fig. 3A), but not at all the intra-suite variations of Ti-in-
315 Ol (Fig. 3B), which suggests that Ti-in-Ol is not controlled by fractionation. And for Sc,
316 we find that at Deccan, and especially Emeishan, Sc-in-Ol is quite high compared to
317 Baffin or Hawaii, despite equivalent or lower Sc in corresponding whole rocks (Fig. 3C,
318 D). This may require a greater $D_{\text{Sc}}^{\text{ol-liq}}$ for our continental flood basalt samples, but may
319 reflect non-equilibrium processes (e.g., Shea et al. 2015a, b) or sub-solidus equilibration
320 of Sc-in-Ol with Sc-rich pyroxenitic cumulates. In either case, contrasts in Sc-in-Ol do
321 not directly translate to equivalent contrasts in co-existing liquids, let alone
322 corresponding mantle sources.

323 2) Elements compatible in Ol ($D_i^{\text{ol-liq}} \geq 1$). Concentrations of some of these elements can
324 closely track liquid compositions (e.g., Ni, Cr). Others (e.g., Mn, Zn) can change
325 dramatically in Ol, even at high Fo contents, even as co-existing liquid compositions
326 change little (Fig. 4A, B)—largely because the concentrations of elements like Mn and
327 Zn in Ol are strongly controlled by temperature. For example, $D_{\text{Mn}}^{\text{ol-liq}}$ and $D_{\text{Zn}}^{\text{ol-liq}}$ are

328 close to unity (respectively ranging from 0.8–1.2 and 0.9–1.2; Davis et al. 2013),
329 becoming more compatible at lower T . This change towards increasing compatibility is
330 readily evident as Ol crystals become more Mn- and Zn-rich with decreasing Fo (Fig. 4),
331 despite parent liquids having nearly identical and unchanging Mn and Zn contents at high
332 MgO; Figure 4 shows a calculated Ol-only LLD, demonstrating that during fractional
333 crystallization, the evolving, T -sensitive, $D_{\text{Mn}}^{\text{ol-liq}}$ and $D_{\text{Zn}}^{\text{ol-liq}}$ more greatly affect Ol than
334 coexisting liquids. Manganese is an interesting case as parental liquids at Baffin, Hawaii
335 and Emeishan have indistinguishable MnO contents at high MgO, but Ol grains from all
336 three plume localities yield distinct Mn-Fo arrays (Fig. 4B). Examining Ol compositions
337 alone, one could deduce a contrast in mantle source composition, but the whole rock data
338 belie such an interpretation; contrasts in Mn-in-Ol more likely reflect subtle contrasts in
339 crystallization environments. For instance, both the enriched (Emeishan) and depleted
340 (Hawaiian) Mn-in-Ol trends (Fig. 4B) can be produced from a liquid with 1317 ppm Mn
341 (or MnO = 0.17 wt. %), but applying a lower- T (1050-1280°C) model of $D_{\text{Mn}}^{\text{ol-liq}} = -3.6 +$
342 $4500/T(^{\circ}\text{C})$ for the enriched trend, and a higher- T (1150-1400°C) model of $D_{\text{Mn}}^{\text{ol-liq}} = -6.2$
343 $+ 7800/T(^{\circ}\text{C})$ for the depleted trend (higher T yielding lower $D_{\text{Mn}}^{\text{ol-liq}}$). Other elements
344 change in more complex ways. Chromium, for example (Fig. 4E, F), is similarly
345 compatible as Mn and Zn ($D_{\text{Cr}}^{\text{ol-liq}} = 0.8$; Le Roux et al. 2015), but Cr-in-Ol decreases
346 with decreasing Fo, following parallel changes in liquid MgO-Cr values. Here, Cr
347 contents respond to a rapidly changing liquid composition. These liquids are likely
348 controlled by spinel, and so Cr^{liq} is modeled empirically from whole rock compositions
349 and then Cr-in-Ol is predicted from $D_{\text{Cr}} = f(T)$, and $T = f(\text{MgO}^{\text{liq}})$, using: $\text{Cr}^{\text{liq}}(\text{ppm}) = -$
350 $118.1 + 56[\text{MgO wt. \%}]$, and $\ln[D_{\text{Cr}}^{\text{ol-liq}}] = -2.51 + 3003.3/T(^{\circ}\text{C})$, which describes the

351 mean of plume values with an error of ± 0.31 . Uncertainty on D_{Cr}^{ol-liq} (D_{Cr}^{ol-liq} varies from
352 1.3 to 2.2 at 1500-1525°C; Longhi 2002 and Salters and Longhi 1999) is alone sufficient
353 to encompass all observed Cr-in-Ol values.

354

355 **Discussion**

356 **Ni, Mn and Fe: Measures of Partial Melting Conditions, not Source Composition**

357 The origin of high Ni in olivine from plume lavas has been accredited to a mantle
358 pyroxenite mineralogy (Sobolev et al. 2007), a core-contaminated source at the base of
359 the mantle (Herzberg et al. 2013), or Ni-enriched melts derived by pyroxene fractionation
360 in the crust (Putirka et al. 2011). Our new data support none of these.

361 To the extent that compatible elements in high Fo olivine might act as a proxy for a
362 mantle source, the predicted patterns of enrichment (or depletion, since the core could
363 just as well absorb siderophile elements from the base of the mantle) deriving from core-
364 mantle equilibration are absent. Cobalt and Zn are nearly as siderophile as Ni (Siebert et
365 al. 2011), but neither are enriched relative to MORB. Perhaps Co and Zn are depleted due
366 to the existence of sulfide phases in evolved lavas, but Cu is enriched (Fig. 2) in our
367 mafic samples, and high T near-primary plume magmas are in any case not expected to
368 be sulfide saturated. Meanwhile, mildly siderophile V and Mn, and highly siderophile P
369 (Siebert et al. 2011), are depleted relative to MORB (Fig. 2). We suggest that Ni in
370 plume-derived olivines reflects the high P - T conditions of plume genesis, and Ol
371 crystallization conditions, as can be discerned from Figs. 5A, B. The key observation is
372 that OIB and MORB fall on the same Ni v. MgO trend (Fig. 5A,B), and so at a given
373 MgO content, OIB and MORB have the same Ni contents. The Baffin plume seems to be

374 slightly depleted in Ni compared to Emeishan and Hawaii at >18% MgO, but the contrast
375 is deceptive: at high MgO (>22 wt. %), whole rock Ni-MgO trends appear to be
376 especially influenced by mixing, between high MgO magmas and olivine grains (Fig 5B),
377 and so contrasts in whole rock Ni contents at high MgO are not necessarily reflective of
378 contrasting mantle sources.

379 Can Any Whole Rocks be Proxies for Liquids?

380 The case for magma mixing at Hawaii, our type example of a mafic volcanic system,
381 is long standing (e.g., Wright and Fiske 1971; Maaloe 1979; Rhodes and Vollinger 2004),
382 but it does not obviate the need for considerable crystallization differentiation. And we
383 can be near certain that liquids exist at nearly all points along the whole rock trend at
384 MgO < 22%. The reason is that Ol compositions occur as a continuum, from Fo71.7 to
385 Fo90.5 (Fig. 5C), requiring a corresponding continuum of liquids with 6.3%-20.8%
386 MgO (Fig. 5D); to deny such is to argue that some fraction of observed Ol phenocrysts
387 have no relationship to Hawaiian magmatism. But then where along the continuum of
388 Fig. 5C does one draw a line to say that “these Ol compositions are irrelevant to
389 Hawaiian magma genesis”?

390 This is not to say, however, that mixing is irrelevant. A range of Hawaiian Ol
391 compositions can be generated by liquids that occupy any part of the gray shaded field in
392 Fig 5D. But because most whole rocks are themselves mostly “liquid” (generally having
393 <30% phenocrysts), it would be nearly impossible to create the observed whole rock
394 trend if the liquids were quite different. And indeed, Hawaiian glass compositions
395 overlap the whole rock trend, to at least 15% MgO (Fig. 5D). So why is the whole rock
396 trend so linear? A large part of the reason is that Ol-only LLDs are quite linear (e.g., Figs.

397 3-5). And so mixing need do little work to straighten out what begin as rather straight
398 trends.

399 Rhodes and Vollinger (2004) further, and in our view correctly, recognize that mixing
400 is not restricted to two end-member compositions, such as one Ol composition, and a
401 single, low-MgO magma. Maaloe (1979) also appreciated this complexity, and
402 emphasized that Hawaiian whole rock trends do not intercept Ol compositional trends at
403 a consistent Fo content. For example, the Rhodes and Volinger (2004) whole rock
404 compositions project to Fo82 for MgO versus Ni, Fo83.7 for MgO versus MnO, and
405 Fo87.2 for MgO versus FeO (using Putirka et al. 2011 Ol compositions). This
406 inconsistency is also visible at the thin-section scale: individual samples (Putirka et al.
407 2011) contain Ol phenocrysts with median and mean Fo contents ranging from at least
408 Fo81 to Fo89. The mixing vectors that produce any one particular sample are thus not
409 necessarily the same as the mixing vectors that produce a compositionally adjacent
410 sample.

411 Rhodes and Vollinger's (2004) solution is to posit that evolved magmas (7% MgO)
412 mix with a range of magmas, having up to 40% MgO (Ol slurries), with liquids ranging
413 to 15% MgO. We would only add that liquids probably range to 21% MgO (to explain
414 high Fo olivine grains), and that the evolved end-member need not be fixed (so in our
415 view, anything can mix with anything). Nonetheless, Fo86 Ol grains are the most
416 common in our new dataset (Fig. 5C). These can be obtained from a liquid with 13.3%
417 MgO, which also is near the mean and median of Hawaiian whole rocks (Fig. 5D). Dry
418 Hawaiian liquids of this composition have densities of 2.8-2.9 g/cm³ (a little higher if
419 they carry crystals) and so are just the right density to be neutrally buoyant in Hawaii's

420 middle crust, where thermobarometers place the majority of Hawaiian magma partial
421 crystallization (Putirka 2017, their Fig. 1B). Olivines of Fo86 may be the most common
422 because their equilibrium liquids are the most likely to be trapped in the middle crust. In
423 any case, whole rocks appear to serve as accurate proxies for liquid compositions, up to
424 some upper limit of MgO content, which falls near 21-22% at Hawaii, and which can be
425 determined for any system based on whole rock FeO_t values and maximum Fo contents
426 of phenocrysts (see supplementary files for values inferred for the systems studied here
427 and Putirka 2005 for the approach).

428 Plume-derived olivine grains, then, range to higher Ni compared to MORB because
429 they crystallize from liquids (electronic appendix 2) that are generated at higher F , due to
430 higher temperatures of partial melting. Their mantle sources have similar Ni (electronic
431 appendix 3) but higher F at plumes leads to greater Ni in the liquids (and their co-existing
432 Ol) that are generated (Table 1). To illustrate, consider a liquid that falls on the Ni-MgO
433 trend, having 20% MgO and 1050 ppm Ni (Fig. 6B); this point forms the “starting point”
434 to the solid gray and black curves in Fig. 6B. To construct the CLDs (gray and black
435 curves) in Fig. 6A and the LLDs (gray and black curves) in Fig. 6B, we use that starting
436 composition and evaluate Eqns. (1a; gray curve) and (1b; black curve) by (i) calculating
437 T using the Helz and Thornber MgO-based thermometer (1987; 1416°C at 20% MgO);
438 (ii) we then remove olivine in 2% increments, re-calculating T , and (iii) at each step, re-
439 evaluating Eqns. (1a-b) to obtain Ni^{liq}, and using Eqns. (3) and (4), and mass balance to
440 obtain MgO^{liq}, MgO^{Ol} and Fo contents of Ol. Equation (1b) yields Ol with up to 4100
441 ppm Ni at Fo91, a CLD that reproduces the high-Fo portion of the Hawaiian array (Fig.
442 6A; black curve), and a LLD that nicely matches Hawaiian whole rocks (Fig. 6B; black

443 curve). Using Eqn. (1a), and the very same starting liquid, we obtain a flatter CLD that
444 reproduces the Ni v. Fo trends at Hawaii, Deccan and Baffin (Fig. 6A; gray curves), and
445 an LLD that reproduces some high Ni vs. MgO whole rock compositions at Hawaii (Fig.
446 6B; gray curves). Equation (1b) also yields a CLD and LLD that nicely reproduces
447 MORB-derived high Fo crystals and whole rock compositions (Figs. 6A, B; blue curve),
448 in this case using a starting liquid with 14.4% MgO^{liq} , $\text{Ni}^{\text{liq}} = 500$ ppm, and $T = 1301^\circ\text{C}$.
449 This begs the question of why MORB-derived magmas have lower Ni in the first place,
450 and as we will show, it is because their parental magmas are produced at lower P-T
451 conditions, at lower melt fractions, compared to OIB (Matzen et al. 2009; Putirka et al.
452 2011).

453 These data suggest that not only is the pyroxenite model unnecessary, it is untenable:
454 Ni-enriched sources yield Ni-enriched liquids, which are unobserved (Fig. 6B), and a
455 pyroxenite source predicts Cr-in-Ol that is $\gg 2000$ ppm, when Ni contents approach
456 observed values (Fig. 6D). To take the Ni-enrichment case, we show a CLD derived
457 using a parental liquid with 1500 ppm Ni at 20% MgO (from a pyroxenite, or Ni-enriched
458 peridotite, it does not matter); this explains high Ni-in-Ol at $\text{Fo} \leq \text{Fo}_{88}$ deceptively well
459 (red dashed CLD, Fig. 6A). But the suite of liquids implied by such a CLD have higher-
460 than-observed Ni^{liq} compared to any known whole rock at any MgO (Fig. 6B) when we
461 apply Eqn. 1b and the Helz and Thornber (1987) MgO^{liq} thermometer. Other
462 combinations of thermometers and D_{Ni} models fare no better (Figure 6C). Use of Eqn. 1c
463 does not change the results perceptibly, and if we apply the MgO^{liq} thermometer of
464 Putirka (2008; Eqn. 13), Ni^{liq} contents are further above observed values. If we instead
465 apply Eqn. 1a and the Helz and Thornber (1987) thermometer we can use a parental

466 magma with just 1200 ppm Ni^{liq} (at 20% MgO) to predict Ni-in-Ol at Fo88, but this curve
467 is still displaced to high Ni^{liq} (Fig 6C), and is worse still if the Putirka (2008)
468 thermometer is applied, and does not even explain Ni-in-Ol patterns. The Cpx-dominated
469 LLD of Putirka et al. (2011) fails more dramatically, for similar reasons, as it predicts
470 liquids with remarkably high Ni contents at lower MgO (Figs. 6A, B, green dashed
471 curves).

472 Perhaps an even more compelling reason to reject pyroxenite as a source is that Cr and
473 Ni in olivine are very poorly predicted by such. To illustrate, we compare natural Cr and
474 Ni in Ol crystals, to those that would precipitate from a pyroxenite, here using mineral-
475 liquid partition coefficients of Le Roux et al. (2015) and each of the wide ranging
476 pyroxenite mineralogies and melt fractions of Table S3 in Sobolev et al. (2007) (where
477 partial melting ranges from 8 to 79%), as well as their bulk pyroxenite Cr and Ni contents
478 for pyroxenite (e.g., 1710 and 1000 ppm respectively). The predicted Cr-in-Ol contents
479 that would precipitate from pyroxenite-derived liquids are nearly 6 times greater than
480 observed, and plot well off scale in Fig. 6D. If we instead assume that pyroxenites are
481 subducted MORB, and so use the much lower bulk Cr and Ni contents for such (Cr =
482 296.1 ppm, Ni = 152 ppm; see Putirka et al. 2011), the liquids generated have much
483 lower Cr, as do the equilibrated Ol, but Ni contents are too low (Fig. 6D). In contrast, Ol
484 that precipitates from partial melts of a peridotite work quite well: using a mean bulk
485 peridotite composition having Cr = 2617 ppm and Ni = 2198 ppm (Putirka et al. 2011),
486 and allowing melt fraction to range as $F = 0$ to 0.3, yields Ol crystals that precisely match
487 observed values (Fig. 6D).

488 Another issue involves the very high Ni-in-olivine at Hawaii at \leq Fo88, where Ni
489 contents are even higher than at Deccan (Fig. 6A). This might be a signal of Ni-enriched
490 magmas, but the crystal compositions can also be derived from fractionated, FeO-
491 enriched magmas instead (at $\text{MgO} \leq 8$ wt. % MgO; Fig. 1A), as shown by the orange
492 curves in Figs. 7A, B. These orange curves connect these high Ni-in-Ol crystals (at
493 \leq Fo88) to a corresponding whole rock MgO^{liq} - Ni^{liq} composition that can serve as a viable
494 equilibrated liquid, at least in the case of Fe-Mg exchange equilibrium. The solutions are
495 not unique, and depend upon the Fe/Mg ratio of the liquid. For this model we use
496 observed Fo in Ol and FeO^{liq} to determine an equilibrium MgO^{liq} (see caption to Fig. 7).
497 And then from the whole rock MgO^{liq} - Ni^{liq} trend (Fig. 7B) we obtain a putative Ni^{liq}
498 value, which we assume is the equilibrium value for $\text{Ni}^{\text{Ol}} \leftrightarrow \text{Ni}^{\text{liq}}$ equilibrium. (Fig.
499 7A) (see caption for equations and details). These results allow us to calculate a putative
500 $D_{\text{Ni}}^{\text{ol-liq}}$ for each Ol (Fig. 7A), and to calculate T using Eqn. (2). The temperatures from
501 Eqn. (2) are remarkably close to those obtained from MgO^{liq} using Helz and Thornber
502 (1987) (Fig 7A). An exception involves the lowest Fo Ol in this test (Fo73), where the
503 D_{Ni} value yields a T from Eqn. (2) that is nearly 300°C lower than that obtained from
504 MgO^{liq} . However, the two T estimates are within 20°C if $\text{FeOt}^{\text{liq}} = 18\%$ (Fig. 7B; black
505 connecting line). This is much higher than the FeOt of the host rocks we investigated, but
506 such high values are exhibited by some low-MgO lavas at Hawaii, which may have
507 mixed with our more primitive samples. A test would be to examine high FeOt samples,
508 looking for high Ni Ol grains. In any case, all but the lowest Fo, high Ni-in-Ol crystals
509 have quite little to do with high Ni^{liq} : in diagrams like Figs. 6A or 7A, we can see that it's

510 not so much that Ni is enriched at a given Fo content, but rather that for a given Ni
511 content, Fo is low.

512 **Peridotite as Plausible Source**

513 Finally, Fig. 7B shows that it is possible to obtain the requisite magmas that can be
514 parental to these high Ni-in-Ol grains by partially melting a peridotite, having 1900 ppm
515 Ni (and 65% Ol, 20% Opx and 15% Cpx), as shown by black and gray curves. These
516 curves show the case for $F = 0.15$, assuming a T of 50°C above the Hirschmann et al.
517 (2002) solidus. To link T and F , we use experimental results from Takahashi et al.
518 (1993), Kushiro (1996) and Pickering-Witter and Johnston (2000), from which we obtain
519 ≈ 0.18 to 0.31% melt/°C at $F < 30\%$. At $F > 30\%$, the results diverge greatly, either
520 maintaining a fertile rate of melt production, or reducing to 0.1% melt/°C; we assume a
521 fertile source (0.3% melt/°C), for reasons discussed below. The Hirschmann et al. (2000)
522 solidus yields array of P - T partial melting conditions, which are then used in Eqn. (1d) to
523 evaluate $D_{\text{Ni}} = f(T)$. From partial melting experiments, we also calibrate MgO^{liq} just
524 above the solidus as $\text{MgO}^{\text{liq}} = -55.96 + 0.0532[T(^{\circ}\text{C})] - 2.16[P(\text{GPa})]$, and then also $D_{\text{Ni}} =$
525 $f(\text{MgO}^{\text{liq}}) = \exp(13.83/\text{MgO}^{\text{liq}} - 0.0147)$. These equations simultaneously yield a Hawaiian
526 primitive magma ($\text{MgO}^{\text{liq}} = 18.8$ Wt. %; $\text{Ni}^{\text{liq}} = 815$ ppm; Fig. 7B), at $P = 3.4$ GPa, $T =$
527 1521°C. The value of F provides the most important source of error, but at $F = 0.2$, at
528 66°C above the solidus we still obtain a magma that falls on the observed whole rock
529 trend ($\text{MgO}^{\text{liq}} = 19.3$ Wt. %; $\text{Ni}^{\text{liq}} = 850$ ppm; $P = 3.5$ GPa, $T = 1526^{\circ}\text{C}$). The results are
530 by no means unique, but they illustrate that peridotites are a plausible mineralogy for
531 plume-source lavas from the localities examined here.

532

533 **Melting Conditions Assuming a Peridotite Source**

534 As in Putirka (2011), we compare natural whole rock compositions at Hawaii and the
535 EPR to the partial melts generated in experimental studies. Our model parent magmas are
536 combinations of experimentally-derived liquids (see Fig. 1, caption) that, as precisely as
537 possible, fall on a whole rock compositional trend for all of the major oxides, as in Fig. 1.
538 This approach yields conditions of melt extraction of 10 kbar, 1325°C for MORB and
539 37.5 kbar and 1540°C at Hawaii. The experiments being averaged to achieve a match to
540 observed whole rocks exhibit a 80 to 160°C temperature range for MORB and plumes
541 respectively, and a pressure range of 10-15 kbar for Hawaii; the experimental starting
542 compositions vary as well. We resort to such combinations as no single experiment
543 describes MORB or Plume magmas precisely for every major oxide, while our
544 hypothetical mixtures, provide a very close match. But how then to interpret the *P-T*
545 range? Besides experimental error (e.g., reproducibility, which translated to *P-T*
546 conditions is about $\pm 30^\circ\text{C}$ and $\pm 1\text{-}2$ kbar), this almost certainly represents the challenge
547 of searching *P-T*-composition space to find the “just so” conditions to explain natural
548 magmas. But crucially, melting is not an isothermal, isobaric process. Our *P-T* estimates
549 represent a mean set of conditions over which melt was equilibrated within the mantle,
550 and the starting compositions of the experiments we employ represent an average
551 composition of what we know to be a heterogeneous source. For our Plume 1 model then,
552 the *P-T* ranges exhibited by our averaged experiments (45-30 kbar; 1620-1460°C) might
553 represent very real variations in what is assuredly a polybaric, polythermal partial melting
554 process.

555 While Emieshan and Deccan have major oxides that are quite similar to Hawaii,
556 Baffin Island is clearly a special case, since except for FeO_t and SiO₂, its lavas are
557 otherwise quite similar to our MORB samples—for example Baffin lavas have elevated
558 Al₂O₃ compared to other plumes (Fig. 8C). The Baffin Al₂O₃ contents might seem to
559 contradict our high T_p , since this should also require elevated pressures, and at high- P
560 garnet may be present in the solid residue. But high- P partial melting experiments exhibit
561 more than sufficient scatter to accommodate Baffin lavas. For example, the 30 kbar
562 model partial melts of Longhi (2002) match Al₂O₃ and other oxides at Baffin quite
563 nicely, as do yet higher P experiments by Takahashi et al. (1993; 46 kbar, 1800°C) and
564 Walter (1998; 45 kbar, 1650°C), which both fall on the high-MgO end of the Baffin
565 compositional spectrum. A problem with Al₂O₃, then is that it decreases with increasing
566 MgO. So while experimental data do indeed show decreasing Al₂O₃ in equilibrated melts
567 as P increases, there is enough scatter in these to cover both the Baffin and Hawaiian
568 lavas at high MgO. So the key question is whether we must match Al₂O₃ at the high or
569 low MgO end of an observed whole rock trend? The answer is found with FeO_t. High
570 pressures are required to explain Baffin's high FeO_t compared to MORB (10-11 wt. % at
571 Baffin, compared to 8% for MORB, when MgO>10%). So we find that, like Hawaii, high
572 T_p and melting pressures in the 30-45 kbar range are most consistent with observed Ol
573 and lava compositions.

574 These P - T conditions, though, leave unexplained the fact that Baffin and MORB share
575 similar, elevated Al₂O₃ contents relative to Hawaii and other plumes (Fig. 8), as well as
576 lower TiO₂, and slightly elevated CaO, the latter being perhaps also in common with

577 Deccan. We hypothesize that these contrasts reflect differences in source regions, as
578 indicated by other elements as well (see following discussion of Fig. 9).

579

580 **Tests of a Pyroxenite Source: Ca, Al and Sc**

581 We might expect that pyroxenite-influenced melts and crystals would be enriched in
582 Ca and Al (or perhaps depleted if a high mode of Cpx during melting overrides the higher
583 bulk concentrations): pyroxenite source materials have 7-22 wt. % Al_2O_3 and 2-14% CaO
584 (Kogiso et al. 1998; Kogiso and Hirschmann 2001; Petermann et al. 2004; Lambart et al.
585 2009; Tsuno and Dasgupta 2011), while peridotites have lower Al_2O_3 and CaO (3.6-4.3
586 wt. % and 3.2-3.5 wt. % respectively; Walter 1998). However, peridotite partial melts
587 alone yield a wide range of CaO^{liq} and $\text{Al}_2\text{O}_3^{\text{liq}}$. Ocean Island Basalts have lower CaO^{liq}
588 and $\text{Al}_2\text{O}_3^{\text{liq}}$ than MORB, and the contrasts are expected given current experimental
589 studies, if the contrasting pressures required by Fe and Mn apply (Fig. 1; see also Putirka
590 et al. 2011), i.e., $P = 30\text{-}50$ kbar for Hawaii and 10 kbar for MORB. Our low- P MORB
591 model (Fig. 1) yields 11.6 wt. % CaO^{liq} and 14% $\text{Al}_2\text{O}_3^{\text{liq}}$ at 13.1 wt. % MgO^{liq} (and 8%
592 FeO^{liq}), while our “Plume Model 2” (Fig. 1) has 8% CaO^{liq} and 9% $\text{Al}_2\text{O}_3^{\text{liq}}$ at 20.6%
593 MgO^{liq} (and 11.5% FeO^{liq}). It seems clear that Ca and Al are lower in high Fo Hawaiian
594 Ol compared to MORB (Figs. 8B, D) as each reflects their contrasting parent liquids (Fig.
595 8A).

596 At Baffin, despite having a much greater mantle potential T_p than MORB (Table 2),
597 these lavas exhibit similar Ca and Al contents to MORB for both whole rocks and
598 olivine. Since the whole rock CaO and Al_2O_3 contents of both MORB and Baffin are
599 explained by peridotite partial melting, at low and high P - T conditions respectively, we

600 cannot exclude a pyroxenite source, but have no reason to propose one. At Emeishan,
601 however, olivine compositions range to distinctly high Ca despite their whole rocks
602 having no more CaO than other lavas. Might the elevated Ca-in-Ol at Emeishan indicate
603 the kind of garnet-pyroxenite source that Sobolev et al. (2007) suggested for Hawaii?
604 This may be the case, as Emeishan lavas also have greater Sc-in-Ol and greater Sc in
605 their whole rocks compared to Hawaii (Figs. 3C, D). Perhaps partial melting of a Ca- and
606 Sc-rich source, e.g., pyroxene-rich, might be the cause. But there are problems with this
607 model. Emeishan parent liquids are not the most Sc-enriched (Fig. 3C), ranging to 30
608 ppm Sc at 20% MgO, compared to 35 ppm Sc at Baffin (Fig. 3C), even though Emeishan
609 Ol grains have the highest Sc-in-Ol values (22 ppm on average; Fig. 3D). No
610 experimentally determined values for $D_{\text{Sc}}^{\text{ol-liq}}$ can explain these contrasts. For example,
611 $D_{\text{Sc}}^{\text{ol-liq}}$ ranges to about 0.25 (Davis et al. 2013; Laubier et al. 2014), and for a liquid with
612 30 ppm Sc, a $D_{\text{Sc}}^{\text{ol-liq}}$ of 0.73 is required to obtain Sc-in-Ol of 22 ppm—nearly three times
613 the highest experimental value (and Laubier et al. see no dependency of $D_{\text{Sc}}^{\text{ol-liq}}$ on T or
614 $f\text{O}_2$, so we cannot at the present call on changes in such to increase $D_{\text{Sc}}^{\text{ol-liq}}$). Likewise,
615 applying $D_{\text{Sc}}^{\text{ol-liq}} = 0.25$ requires a liquid with 89 ppm Sc (more than double observed
616 Emeishan whole rock values; Fig. 3C) to precipitate Ol with 22 ppm Sc. So it would be a
617 tenuous conclusion at best to link Emeishan Sc-in-Ol contents to enriched liquids, let
618 alone enriched source materials. Finally, Lambart et al. (2009) report very low Al-in-Ol
619 for their pyroxenite-equilibrated Ol, well below anything we observe at Emeishan,
620 Deccan, Siqueiros or Hawaii (Fig. 8D). Given these problems for a pyroxenite source, we
621 posit that high Ca-in-Ol at Emeishan might instead reflect lower P - T conditions of
622 crystallization (e.g., Stormer 1973; Koehler and Brey 1988), consistent with higher Mn-

623 in-Ol at Emeishan (Fig. 4B). But thermobarometry (Tao et al. 2015) at Emeishan
624 indicates high P - T equilibration conditions for picrites, so Sc and Ca in Ol might instead
625 reflect non-equilibrium processes (e.g. Shea et al. 2015a, b).

626

627 **Mantle Source Compositions from Parental Liquids (and maximum Fo in Ol)**

628 Although Ol trace element compositions do not easily translate to a mantle
629 composition, their Fo contents delimit parent liquid compositions, which provide a more
630 direct inference of mantle source. Applying Putirka et al.'s (2011) melting models with
631 Putirka (2016) for melt fraction estimates, we obtain mantle compositions as in Figures 9-
632 10, where elements are arranged in order of decreasing enrichment at Hawaii relative to
633 our calculated Siqueiros-MORB mantle, moving from left to right. To model the MORB
634 source we rely on the very primitive lavas from the East Pacific Rise (Siqueiros
635 Transform; Perfit et al., 1996; Hays, 2004) as they are closer to putative primary magmas,
636 and so minimize error in reconstructing a MORB mantle source.

637 We find that the mantle source regions of Hawaii, Deccan and Emeishan share similar
638 patterns and magnitudes of enrichments in incompatible elements relative to the MORB
639 source mantle (DMM) (except for lower Cr; Fig. 9). This is not to say that our inferred
640 plume sources are identical; they are not (e.g., Willbold and Stracke 2006), but we affirm
641 prior work that indicates broadly shared enrichments among plumes sources (e.g.,
642 Willbold and Stracke, 2006; Arevalo et al. 2013). Our Hawaiian source is also enriched
643 relative to the pyrolite models of McDonough and Sun (1995) and Lyubetskaya and
644 Korenaga (2003) (Fig. 9B). Baffin, despite having plume-like T_p (Table 1), interestingly
645 has a mantle source with trace element contents that are quite similar to that for MORB,

646 except for being slightly enriched relative to our estimate of Cr (Fig. 9A); however,
647 within uncertainties (see error bars in figures) the Baffin Island plume source overlaps
648 with the sources for the other three plumes examined here. Interestingly, Jackson and
649 Jellinek (2013) find that the Baffin source is slightly less depleted in Large Ion Lithophile
650 Elements (LILE) than DMM than in the model used here. However, they infer lower
651 degrees of partial melting: 14%, instead of 24% in this study (Table 1). Nonetheless, the
652 Baffin Island plume composition considered here is within error of the composition
653 calculated in Jackson and Jellinek (2013) and requires no addition of enriched sources to
654 explain trace element concentrations. We should perhaps emphasize that, due to its very
655 high $^3\text{He}/^4\text{He}$, Baffin is not the same as MORB; but the trace element similarities of the
656 Baffin source and DM provides an apparent striking affinity.

657 In contrast, trace elements are enriched relative to DM (and the Baffin source) at
658 Hawaii, Deccan and Emeishan, and by applying methods of Putirka et al. (2011) we
659 invert our calculated melt compositions to estimate source compositions. We find that
660 Hawaiian, Deccan and Emeishan enrichments can be explained by adding 18-24%
661 average ocean crust (MORB) to DMM of Workman and Hart (2004). Our estimate of the
662 amount of crust added back to the non-Baffin plume-source mantle is greater than in
663 Putirka et al. (2011), as that earlier work assumed smaller degrees of partial melting
664 (10%) for the Hawaiian source. Melt fraction estimates are quite uncertain, and so the
665 actual amounts, if the general approach is correct, probably lie somewhere between those
666 earlier values and our current estimates (see electronic appendix 3). What is especially
667 compelling, though, is that nearly identical proportions of MORB and DMM
668 simultaneously explain elements that are chemically quite different. The errors in such

669 mantle estimates are non-trivial (20-140% for compatible to incompatible elements), but
670 within such error our Hawaiian source estimates match our MORB + DMM pyrolite
671 model for all elements. Moreover, calculated MORB:DMM ratios are similar using
672 differently-derived estimates of both MORB and DMM (Fig. 10A).

673 So why then does Baffin lavas range to higher $^3\text{He}/^4\text{He}$ than, say, Hawaii? It has long
674 been recognized that high $^3\text{He}/^4\text{He}$ at Hawaii is inversely correlated with $^{87}\text{Sr}/^{86}\text{Sr}$ (Hauri
675 and Kurz 2002), and other studies, e.g., Jackson et al. (2008) and Grapic et al. (2015),
676 propose that the Baffin source is the receiver of subducted materials and the source of
677 high $^3\text{He}/^4\text{He}$ in plumes and that subducted oceanic crust provides the enriched TITAN
678 character as proposed by Jackson et al. (2008).

679 Because so many different elements (lithophile, chalcophile and moderately
680 siderophile) at Hawaii, Emeishan and Deccan are explained by a pyrolite-like source, this
681 would seem to argue against a peculiar mineralogy (e.g., pyroxenite) or intra-mantle
682 fractionation (e.g., bridgmanite) to explain plume compositions inferred from the lavas in
683 this study. Zhang et al. (2016) nonetheless intimate that wadsleyite may preferentially
684 partition Ti, V and Cr, while Bindi et al. (2017) report on a Ti-rich form of bridgmanite.
685 To be fair, neither of these studies attempt to explain plume compositions. Perhaps even
686 more interesting is the possibility of an Fe-enriched lower mantle (Kaminsky and Lin,
687 2017). But Fe-enrichments in OIB appear to be not so great as to be produced from a
688 mantle with a major element composition similar to MORB, and we find no pronounced
689 coupled enrichments in Ti and V that accompany Cr. Nor is it clear that the accumulation
690 of Ti-bridgmanite might yield the complete array of enrichments of Fig. 9A. This is not
691 to say that intra-mantle fractionation might not influence plume composition, but it would

692 be an odd coincidence that such a process was global in nature and would precisely
693 mimic the addition of oceanic crust to a DM source.

694

695 **Implications**

696 Our Baffin Island results are perhaps the most interesting. The Baffin source is quite
697 hot, and we infer that it is rooted in as deep a part of the mantle as any other mantle
698 plume. But the Baffin source is also depleted, as reflected in both its major oxides and
699 trace elements, in a way that remarkably mimics the MORB source. Plumes other than
700 Baffin, by contrast, require at least 20-25% subducted MORB-crust to be mixed back into
701 a depleted MORB mantle source, to explain their bulk compositions. We appreciate that
702 the Baffin and MORB sources cannot be equivalent: MORBs have low $^3\text{He}/^4\text{He}$ (~8 Ra)
703 compared to the Baffin mantle (up to 50 Ra). Additionally, if we accept W isotope results
704 (Rizo et al. 2016), that indicate that the Baffin source has been isolated from the rest of
705 the mantle for 4.5 Ga, then the Baffin source was depleted at that early date. The
706 depletion of the MORB source, by contrast, occurred much later, as it is a residue
707 remaining after a presumably much later and prolonged continental crust extraction
708 event. There are several options to explain the MORB-Baffin contrast: The Baffin source
709 could be 1) perfectly unique—an ancient depleted source at the base of the mantle, with
710 an as yet unknown enriched counterpart, that otherwise matches MORB only by
711 coincidence, 2) a depleted matrix that pervades the lowermost mantle that elsewhere
712 receives subducted crust, but beneath Baffin, has been strangely resistant to such
713 accumulation, 3) related to the MORB source, where the materials that would eventually
714 form the crust were extracted into the upper part of the mantle at or before 4.5 Ga only to

715 be gradually tapped over the next 4.5 Ga from relatively shallow mantle only, leaving a
716 shallow depleted MORB source, and a deeper Baffin source (again, resistant to subducted
717 crust accumulation), 4) is not from a source that feeds other mantle plumes—representing
718 something deeper still. None of these are terribly satisfying, or mutually exclusive. But
719 Baffin is isotopically close to PREMA (Zindler and Hart 1986; having just slightly lower
720 $^{206}\text{Pb}/^{204}\text{Pb}$) and FOZO (Hart et al. 1992)—and so if Baffin can be shown to be *uniquely*
721 *old* among plumes (being presumably *uniquely unaffected* by admixtures of young
722 subducted crust), Baffin may then represent a rare sampling of Earth's most ancient
723 mantle (Jackson et al., 2010).

724 Some of these results are not entirely new: Herzberg et al. (2007) infer that Baffin is
725 hot, while Starkey et al. (2012) demonstrate that, despite having plume-like high $^3\text{He}/^4\text{He}$,
726 Baffin is depleted compared to most plumes. Garnero et al. (2016) suggest that
727 geochemical anomalies among ocean island basalts carry no information about source
728 depth, and with one exception, we concur—that exception being high $^3\text{He}/^4\text{He}$, which
729 along with high T_p are the only viable indicators of deep-seated plumes, past or present
730 (Putirka 2008). And we agree with Jackson et al. (2017) that to hide something like the
731 Baffin source for an extended period of time, it must be quite dense, and quite deep.
732 Given that $^{87}\text{Sr}/^{86}\text{Sr}$ is negatively correlated with $^3\text{He}/^4\text{He}$ at Hawaii, and trends toward a
733 Baffin-like composition (see Fig. 1A of Hauri and Kurz 2002), we propose that a Baffin-
734 like source, or perhaps the Baffin source itself, is the wellspring of high $^3\text{He}/^4\text{He}$ in
735 plumes, and whose $^3\text{He}/^4\text{He}$ values are slightly diluted due to mixing of subducted crust.

736 Despite the Baffin enigma, we appear to know the depleted MORB source with
737 confidence. Estimates derived from very different mass balances of primitive melt and

738 peridotite samples (Salters and Stracke 2004; Workman and Hart 2005; Putirka et al.
739 2011) yield quite similar results. This should be expected, since residual peridotites and
740 parental magmas are complementary derivatives of a common source and process. But
741 the convergence also confirms that we have, at least roughly, correctly identified parental
742 magmas and the processes and conditions under which they are created.

743 Finally, we confirm the current view of the deep mantle as a “graveyard” of subducted
744 MORB crust (Wyession 1996; White 2015), but we reject the idea that this graveyard is
745 necessarily resurrected with a pyroxenite mineralogy (e.g., Hofmann and White, 1982);
746 we estimate that plumes entrain up to 25% subducted oceanic crust, rather than the nearly
747 100% indicated in the original model (Baffin providing a 0% end-member case). This
748 material seems to be thoroughly admixed, with no clear evidence of a mineralogically
749 distinct signal. Kogiso et al (2004) suggest otherwise, but as we show in an electronic
750 supplement (ES1), their pyroxenite end-member is arbitrary and trace element ratios do
751 not indicate a mineralogically distinct source for elevated $^{187}\text{Os}/^{188}\text{Os}$.

752 The deep mantle plume source is, however, significantly enriched compared to earlier
753 “pyrolite” models (McDonough and Sun 1995; Lyubetskaya and Korenaga 2010) and
754 may well be what is perceived in seismic studies (e.g., Zhao et al. 2015). Our estimates,
755 perhaps not coincidentally, are also quite close to Ringwood’s (1962) basalt:dunite ratio
756 of 1:4. Such high amounts of added crust may create an untenably “hyperfertile” source,
757 with too much garnet to explain certain trace element ratios (Norman and Garcia 1999;
758 Putirka et al. 2011). But our higher MORB:DMM ratios emanate from higher estimates
759 of melt fraction (using Putirka 2016 instead of Putirka et al. 2007), which may allow
760 sufficiently high dissolution of garnet to allay the key objection: Walter’s (1998)

761 experiments yield just 10% garnet (of all phases, including liquid) at 20% melting at 6
762 GPa, and no garnet at all at the same degrees of melting at 3 GPa. In any case, Baffin
763 Island shows that this slab graveyard is not ubiquitous (i.e., recycled slabs do not
764 permeate all deep mantle reservoirs), either because subducted slabs are unevenly
765 interred, or they can be efficiently excavated in later plume upwelling events, or perhaps
766 because the Baffin source lies below this graveyard, as a dense under-layer (e.g., Jackson
767 et al. 2017) that for some reason is not tapped at yet hotter flood basalt provinces.

768

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776

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1091

1092 **Figures**

1093 **Figure 1.** Weight % MgO for MORB (East Pacific Rise, Siqueiros) and Hawaiian
1094 (HSDP2; Rhodes and Vollinger 2004) whole rocks are compared to (A) FeO, (B) MnO,
1095 (C) FeO/MnO and (D) TiO₂. Whole rock compositions are also compared to liquids from
1096 peridotite partial melting experiments, sorted by pressure and using depleted and pyrolite
1097 mantle model peridotites from Takahashi and Kushiro (1983; KLB-1); Takahashi et al.
1098 (1993, KLB-1); Baker et al. (1995, MM3); Herzberg and Zhang (1996, KLB-1); Walter
1099 (1998, KR4003); Longhi (2002, Model Depleted and Primitive Upper Mantle, DPUM
1100 and PUM). Highlighted with a beige fill are 20-30 kbar partial melts of a high FeO
1101 peridotite from Kushiro (1996, PHN1611), noted as “fertile” but lower in Al₂O₃ and CaO
1102 than the other peridotites noted. Plume model 1 is a 50-50 mixture of Walter (1998; 45
1103 kbar, 1620°C) and Kushiro (1996; 30 bar, 1460°C). Plume model 2 is a 40-60
1104 mixture of peridotite partial melts from Takahashi and Kushiro (1983; 35 kbar,
1105 1600°C), and Walter (1998; 45 kbar, 1620°C). The MORB model is a 56-44 mixture
1106 of Kushiro (1996; 10 kbar, 1360°C), and Baker et al. (1995; 10 kbar, 1280°C).

1107

1108 **Figure 2.** Trace element concentrations in Fo₈₉ olivine, from average or projected
1109 values, normalized to Fo₈₉ olivine compositions from MORB (Siqueiros). From Ca to
1110 Mn, elements are listed in order of increasing Ol/Liq partition coefficients (element [D]):
1111 Ca [0.01], Ti [0.01], Al [0.015], Na [0.015], Cu [0.1], Sc [0.2], Mn [0.9]. From Mn to P,
1112 elements are listed in order of increasing core/mantle partitioning using the mid-point of
1113 minimum and maximum values of Siebert et al. (2011; their Table 6) (element [D]): Mn
1114 [1], V [1.9], Cr [3], Zn [4], Co [24.5], Ni [26], P [35].

1115

1116 **Figure 3.** Comparison of the highly incompatible elements (in Ol), Ti (A) and (B) and Sc
1117 (C) and (D) for host liquids and olivine grains (data from GEOROC; <http://georoc.mpch-mainz.gwdg.de/georoc/>); Hawaii (Rhodes and Vollinger 2004); MORB (PetDB;
1118 <http://www.earthchem.org/petdb>). Black curves are high-T plume LLD and CLDs; gray

1119

1120 curves are low-T mid-ocean ridge LLD and CLDs (see text for discussion and calculation
1121 details). We find that an enriched mantle source and very low partition coefficients for Ti
1122 are required to simultaneously explain Ti in whole rocks and co-existing olivine; see text
1123 for discussion. Symbols as in Fig. 2; $D_{Ti} = \exp(-5.2 + 1979/[T^{\circ}C])$, initial $TiO_2^{liq} = 1.6$ wt.
1124 % or 9592 ppm Ti, at 20% MgO^{liq} ; $T(^{\circ}C) = 1014 + 20.1[MgO^{liq}]$.

1125
1126 **Figure 4.** Comparison of compatible elements (Ol-liq partition coefficients near 1.0), Mn
1127 (A) and (B), Zn (C) and (D) and Cr (E) and (F), for published whole rocks (putative
1128 liquids; see caption to Fig. 3) and our new data for olivine grains. When D_i^{ol-liq} is close to
1129 1, the trace element contents of Ol can be quite sensitive to T. For example, in Mn and
1130 Zn, liquid compositions don't vary at $MgO^{liq} > 10\%$, and yet Zn-in-Ol and Mn-in-Ol
1131 increase greatly, as D_i^{ol-liq} increases (from slightly < 1 to slightly > 1) with decreasing T.
1132 On the other hand, Cr-in-Ol mostly decreases as MgO decreases, as other phases (most
1133 likely spinel) deplete coexisting liquids in Cr. Symbols as in Fig. 2. $D_{Cr} = \exp(-2.51 +$
1134 $3003.7/[T^{\circ}C])$, initial $Cr^{liq} = 680$ ppm at 20% MgO^{liq} ; $D_{Mn} = \exp(-3.6 + 4500/[T^{\circ}C])$, initial
1135 $MnO^{liq} = 0.17$ wt. % at 18% MgO^{liq} ; see caption to Fig. 3 for similar calculations of LLD
1136 and CLD curves (black for plume, gray for MORs). Curves are not shown for Zn, as
1137 current partitioning experiments are too few and fail to reproduce Zn-in-Ol trends.

1138
1139
1140 **Figure 5.** (A) Whole rock Ni (ppm) vs. MgO (wt. %) compositions from Emeishan,
1141 Baffin Island, Deccan and MORB (see caption to Fig. 3). (B) Whole rock Hawaii and
1142 MORB compositions are compared to olivine compositions from Putirka et al. (2011).
1143 (C) Histogram of Hawaiian Ol compositions; (D) Cation fractions of MgO and FeO in
1144 Hawaiian whole rocks (triangles), with isopleths that show the positions of liquids in
1145 equilibrium with a given Fo content, when $K_D(Fe-Mg)^{ol-liq} = 0.34$; isotherms are from
1146 Putirka (2005); red circles indicate Hawaiian liquids needed to produce maximum,
1147 minimum and median Ol compositions in panel C; dashed line in D shows the position of
1148 Hawaiian glass compositions. Other symbols as in Fig. 2. In (A), it is clear that at a given
1149 MgO content, Ni contents for plumes are not enriched relative to MORB. In (B), whole
1150 rock Ni contents at $MgO > 21\%$, are controlled by mixing with Ol, which likely explains
1151 subtle contrasts between Ni contents at Baffin compared to Emeishan, Hawaii or Deccan
1152 in panel (A). Panel (D), with isotherms and Ol isopleths from Putirka (2005), similarly
1153 shows the range of liquids required to produce observed Hawaiian Ol compositions (gray
1154 area, Fo71.7-Fo90.5, as in panel C). Such liquids must have up to 20.8% MgO, with a
1155 continuum down to 6.3% MgO, to create the continuum of Ol compositions in panel C.

1156
1157 **Figure 6.** (A) New data for Olivine Ni v. Forsterite (Fo) contents. (B) and (C) Whole
1158 rock Ni v. MgO. (D) New Cr vs. Ni in Ol compositions. Symbols are as in Fig. 2; whole
1159 rock data are as in Fig. 3 caption; see electronic supplement for new Ol data. Panels (A)
1160 and (B) show models that explain high Ni-in-Ol at moderate to low Fo ($\leq Fo88$). For (A)
1161 and (B) Crystal Lines of Descent (CLDs) and Liquid Lines of Descent (LLDs) are
1162 calculated by selecting as starting compositions, $MgO^{liq}-Ni^{liq}$ values that fall on the
1163 observed whole rock trends of (B), and then subtracting calculated Ol compositions in
1164 2% increments using mass balance. Gray curves use Eqn. (1e), black and red curves Eqn.
1165 1b, and the blue curve Eqn. (1c); in panel (A) we use Eqn. (3) to obtain Fo contents in Ol;

1166 in panel (B) we use Eqn. (4) for $\text{FeO}^{\text{liq}} = 11.5 \text{ wt.}\%$ to obtain MgO^{Ol} , and then calculate
1167 MgO^{liq} from mass balance. To evaluate all of Eqns. (1b,e,c), (3) and (4), T is re-
1168 calculated at each step using Helz and Thornber (1987) [$T(^{\circ}\text{C}) = 20.1[\text{MgO}^{\text{liq}}] + 1014$]
1169 for both MORB and Hawaiian lavas, although a separate calibration for MORB, $T(^{\circ}\text{C}) =$
1170 $17.5[\text{MgO}^{\text{liq}}] + 1054$ (calibrated from Bender et al. 1984; Grove and Juster 1989; Grove
1171 and Bryan 1983) yields similar results at high T . Dashed curves are “failed” models: they
1172 explain some Ni vs. Fo trends in olivine, but not Ni vs. MgO in whole rocks (nominal
1173 liquids) or Liq + Ol mixtures (see Fig. 5B for Liq + Ol trajectories). Green dashed curve
1174 is a modified version of the Cpx-dominated CLD and LLD from Putirka et al. (2011),
1175 while red dashed curve uses Eqn. (1b) and assumes that high Ni in Fo88 olivine derives
1176 from a Ni-enriched liquid. Predicting observed Ni contents for olivines with <Fo88 yield
1177 coexisting liquids displaced even further from observed whole rock trends. In (C), we
1178 show the difficulty of explaining whole rock Ni contents if we assume Ni-enriched parent
1179 liquids: the red curve from (B) is reproduced as Ol LLD1 (with 1500 ppm Ni at 20%
1180 MgO), and is compared to Ol LLD2, which uses the same starting liquid, but uses Eqn.
1181 1c and the thermometer of Putirka (2008), and Ol LLD3, which uses Eqn. 1a with the
1182 Helz and Thornber (1987) thermometer, and a liquid with 1200 ppm Ni at 20% MgO;
1183 each model successfully predicts Ni-in-Ol at Fo88, but fails to reproduce Ni contents of
1184 whole rocks. The models are derived assuming that Ol precipitates from melts derived by
1185 partial melting of pyroxenite (gray curves) or peridotite (green curves). Le Roux et al.
1186 (2011) suggest that $D_{\text{Ni}}^{\text{Cpx-liq}} = 22$ in some circumstances, but we see little evidence for
1187 such high values in experimental data, and we find that $D_{\text{Ni}}^{\text{Cpx-liq}} = 3$ fits observed Ni-in-
1188 Ol compositions much better (see discussion for details).

1189
1190 **Figure 7.** Magnified view of Hawaiian (and Siqueiros) samples as in Figure 5, to
1191 compare olivine Ni v Fo (A), and whole rock Ni vs. MgO (B), for olivines with moderate
1192 Fo contents but high Ni. Olivine grains at $\leq \text{Fo}88$ cannot form from Ni-enriched
1193 magmas—such magmas do not exist (Fig. 6B). But they can form from observed liquids (B) at
1194 lower temperatures (where D_{Ni} is elevated) that are Fe-rich (which decreases Fo at a
1195 given MgO^{liq}). To determine these liquids, we take those Ol grains with the highest NiO
1196 at a given Fo content (empirically, $\text{Ni}_{\text{max}}^{\text{Ol}}(\text{ppm}) = 65.8[\text{Fo}] - 2271$, and $\text{Fo} =$
1197 $100[(X_{\text{MgO}}^{\text{Ol}}/(X_{\text{MgO}}^{\text{Ol}} + X_{\text{FeO}}^{\text{Ol}}))]$) and then use Fo to calculate MgO^{liq} , which (in a variation
1198 of Eqn. 4) can be found empirically, if FeO^{liq} is known: for $\text{FeO}^{\text{liq}} = 13 \text{ wt.}\%$, $\text{MgO}^{\text{liq}} =$
1199 $(0.603 - 0.0061[\text{Fo}])^{-1}$; for $\text{FeO}^{\text{liq}} = 18 \text{ wt.}\%$, $\text{MgO}^{\text{liq}} = (0.383 - 0.0039[\text{Fo}])^{-1}$; Ni-in-liq
1200 can be determined from the whole rock trend: $\text{Ni}^{\text{liq}}(\text{ppm}) = 63.3\text{MgO}^{\text{liq}} - 364.8$. These
1201 calculations provide what we need to determine D_{Ni} —in effect forcing the high Ni Ol
1202 crystals (orange circles in panel A) to form from liquid compositions in panel (B) (orange
1203 circles), connected by straight lines. Having D_{Ni} , and MgO^{liq} , the tables in (A) use such as
1204 input to compare the implied T using Helz and Thornber (1987) with the T implied from
1205 D_{Ni} using Eqn. (2). Temperature estimates are close at high MgO, but diverge as MgO
1206 decreases, where an olivine with Fo73 and 2,530 ppm Ni requires an impossibly high D_{Ni}
1207 ($=59$) and low T^{Ni} ($<900^{\circ}\text{C}$). But rare Hawaiian samples range to 18 wt. % FeO; in such
1208 a liquid this same olivine can form from $\text{MgO}^{\text{liq}} = 10.2 \text{ wt.}\%$, with a resulting $D_{\text{Ni}} = 9.1$
1209 and temperature estimates coincide within error. Black and gray curves in (B) are batch
1210 partial melts, $C_{\text{Ni}}^{\text{liq}} = C_{\text{Ni}}^{\text{source}}/[F + D^{\text{bulk}}(1-F)]$, where $C_{\text{Ni}}^{\text{source}} = 1900$ (for a peridotite
1211 source; Sobolev et al. 2007). Melt compositions are calculated at 25°C above the solidus

1212 (Hirschmann 2000), which approximates a melt fraction (F) of 10% (e.g., Baker et al.
1213 1995). We calculate MgO^{liq} (wt. %) = $-55.96 + 0.0532[T(^{\circ}\text{C})] - 2.16[P(\text{GPa})]$, using this
1214 “solidus + 25°C” curve as input; this expression for MgO^{liq} reproduces calibration data
1215 (Takahashi et al. 1993; Baker et al. 1995; Herzberg and Zhang 1996; Kushiro 1996;
1216 Robinson et al. 1998; Walter 1998; Pickering-Witter and Johnston 2000) to ± 2 wt. %. For
1217 Ni-in-Liq we use $F = 0.1$, and D^{bulk} assumes: 20% Opx, 15% Cpx and 65% Ol, $D_{\text{Ni}}^{\text{cpx-liq}} =$
1218 3.2 and $D_{\text{Ni}}^{\text{opx-liq}} = 3.7$ (from Le Roux et al. 2015). For $D_{\text{Ni}}^{\text{ol-liq}}$, the gray curve uses Eqn.
1219 (1a) and the black curve uses (1b).

1220

1221 **Figure 8.** A comparison of CaO (A and B) and Al or Al_2O_3 (C and D) between whole
1222 rock MgO (see Fig. 3 caption) and Fo from new olivine compositions (see electronic
1223 supplements). In (E) and (F), Ni in Ol is compared to Ca and Al in Ol respectively
1224 from these same data sets. The experimentally-derived MORB and Plume model 2
1225 peridotite partial melts are determined as in the caption to Fig 1. (A) and (C) show
1226 that MORB have higher CaO and Al_2O_3 compared to plume derived lavas, and that in
1227 both cases, such contrasts likely result from contrasts in melting conditions of a
1228 peridotite source. Such partial melts with low Al_2O_3 are mostly garnet saturated,
1229 indicating that deep-seated partial melting, within the garnet stability field, is much
1230 more important at Hawaii, Emeishan and Deccan, compared to Baffin or MORB.
1231 Olivine Ca and Al concentrations reflect these contrasts in liquid composition. Also
1232 plotted in (B) is the field of Ca for olivines precipitated from experimental
1233 pyroxenite partial melts, from Kogiso et al. (1998), Kogiso and Hirschmann (2001)
1234 and Lambart et al. (2009). Only the Ca contents of olivine from Emeishan and
1235 Deccan are consistent with a pyroxenite derivation. But all natural plume-derived
1236 olivine grains have vastly more Al than expected compared to pyroxenite-melt
1237 equilibrated Ol from Lambart et al. (2009) (D).

1238

1239 **Figure 9.** The trace element concentrations in our selected plume sources are
1240 calculated from parental magma compositions (see appendix tables 2 and 3) as in
1241 Putirka et al. (2011), and normalized to (A) our MORB-source mantle (“MORB-
1242 Siqueiros”; calculated from Siqueiros parental magmas) and (B) various estimates of
1243 depleted and pyrolite mantle, where “Pyrolite(M&S)” is the pyrolite model of
1244 McDonough and Sun (20015); “Baffin” is our estimate of the Baffin Island source;
1245 “DMM (W&H)” is “depleted MORB mantle of Workman and Hart (2005); and
1246 “DM(S&S) is the MORB Depleted mantle source of Salters and Stracke (2004). The
1247 pyrolite model of Lyubetskaya and Korenaga (2010) is not plotted, but is effectively
1248 identical to the curve for McDonough and Sun (1995). The “anticipated” errors are
1249 calculated as the maximum – minimum (range) normalized concentration ratios,
1250 calculated using maximum and minimum ranges of F at both MORB and Hawaii, and
1251 maximum and minimum partition coefficients for MORB and Hawaii, as in Putirka et
1252 al. (2011) (i.e., the highest normalized ratio for Th is that obtained by allowing F at
1253 Hawaii to be 0.25, so requiring more Th in the Hawaiian source, and using $F = 0.08$
1254 for MORB, to minimize calculated Th in the MORB source). As noted in the text, our
1255 Cr estimate for DM (1,100 ppm) is much lower than Salters and Stracke (2004;
1256 2,500 ppm). We assume 600 ppm in mantle-equilibrated MORB (Fig. 4E), and a bulk
1257 D_{Cr} during mantle melting of 1.92. We can obtain 2,500 ppm by increasing Cr^{liq} to

1258 1400 ppm, which seems vastly too high compared to primitive MORB, or by
1259 increasing bulk D_{Cr} to 4.5, which can only be reached or exceeded in garnet at high T
1260 and would require very high residual garnet in the source (e.g., Putirka 1998).

1261

1262 **Figure 10.** (A) Our Hawaiian trace element source is compared to our model plume
1263 sources, obtained by mixing MORB crust with depleted MORB-source mantle
1264 (effectively the “pyrolite” of Ringwood (1962)). “HMS” = Hawaiian mantle source.
1265 “SMS” is our depleted Siqueiros mantle source (appendix Table 3); “DM1” is
1266 depleted mantle from Workman and Hart (2005); “DM2” is depleted mantle from
1267 Salters and Stracke (2004); “BMS” is our calculated Baffin mantle source. The
1268 models that make use of DM1 and BMS depleted mantles are mixed with ocean crust
1269 that is equal to the mean MORB estimate of Gale et al. 2013), while the DM and DM2
1270 models are mixed with MORB from Putirka et al. (2011). (B) Our calculated depleted
1271 mantle source for Baffin Island (BMS) is compared directly to SMS, DM1, DM2 as in
1272 panel (A), and the pyrolite model of McDonough and Sun (1995).

1273

Table 1. Estimates of mantle potential temperature and related inp

<i>Plumes and MORB</i>	<i>T_p (°C) (1)</i>	Source
		<i>Olivine Fo (2)</i>
Emeishan (Low Ti)	1700(67)	92.5
Emeishan (High Ti)	1480(84)	89.5
Deccan (H&G 2009)	1620(54)	91.5
Hawaii (SR0061-0)	1630(77)	91.0
Baffin (Hole 2015)	1630(65)	92.0
MORB-Siqueiros	1420(40)	91.4
MORB Mean Mean Cottrell and Kelley (2011)	1320(39)	89.3
<i>Other plume and MORB estimates</i>		
Siberian Traps	1590(37)	92.0
Ferropicrites	1780(155)	92.0
Samoa	1620(66)	91.7
Iceland	1520(46)	91.7
MORB, 9°N; Cottrell & Kelley (2011)	1400(37)	90.5
MORB (Herzberg)	1400(38)	91.3
MORB Cottrell and Kelley	1390(45)	90.7

Notes: (1) T_p = mantle potential temperature; T_p is calculated as in Putirka (2007)
(2) Olivine forsterite (Fo) contents are calculated from liquid compositions using the method of Herzberg (2000). Olivine and liquid compositions are not arbitrary. Liquid compositions are selected to be consistent with the mantle potential temperature estimates.

out parameters

Putirka (2008) Eqn. 42 <i>P</i> (GPa)	$\log[fO_2]$ at NNO	Putirka (2016) Eqn. 8b $K_D(\text{Fe-Mg})^{\text{ol-liq}}$	$T^{\text{ol-liq}}$ Putirka et al. (2007) <i>T</i> (°C)
3.23	-3.18	0.350	1597
2.05	-4.51	0.342	1468
2.63	-3.78	0.347	1539
2.51	-3.75	0.349	1549
2.41	-4.05	0.347	1512
1.59	-5.38	0.342	1376
1.14	-6.17	0.341	1311
2.26	-4.18	0.347	1502
3.96	-2.32	0.347	1696
3.16	-3.27	0.347	1582
1.97	-4.71	0.344	1447
1.39	-5.60	0.342	1363
1.55	-5.49	0.342	1367
1.50	-5.52	0.340	1392

16); see electronic appendix for input liquid compositions, and olivine + liquid temperature, using the calculated oxygen fugacity, and Fe³⁺/Fe²⁺ ratios as determined using equation 16. The calculated *T* values are listed so that the calculated Fo contents for olivine match observed maximum Fo contents.

Melt Fraction
Putirka (2016)
Avg. Eqns. 14a-c
0.233(0.042)
0.066(0.054)
0.183(.004)
0.189(0.009)
0.239(0.041)
0.106(0.028)
0.032(0.029)
0.189(0.017)
0.212(0.095)
0.136(0.024)
0.158(0.027)
0.091(0.005)
0.097(0.026)
0.069(0.022)

tures
 is in Putirka (2016).
nts of high Ca olivine phenocrysts.

Figure 1

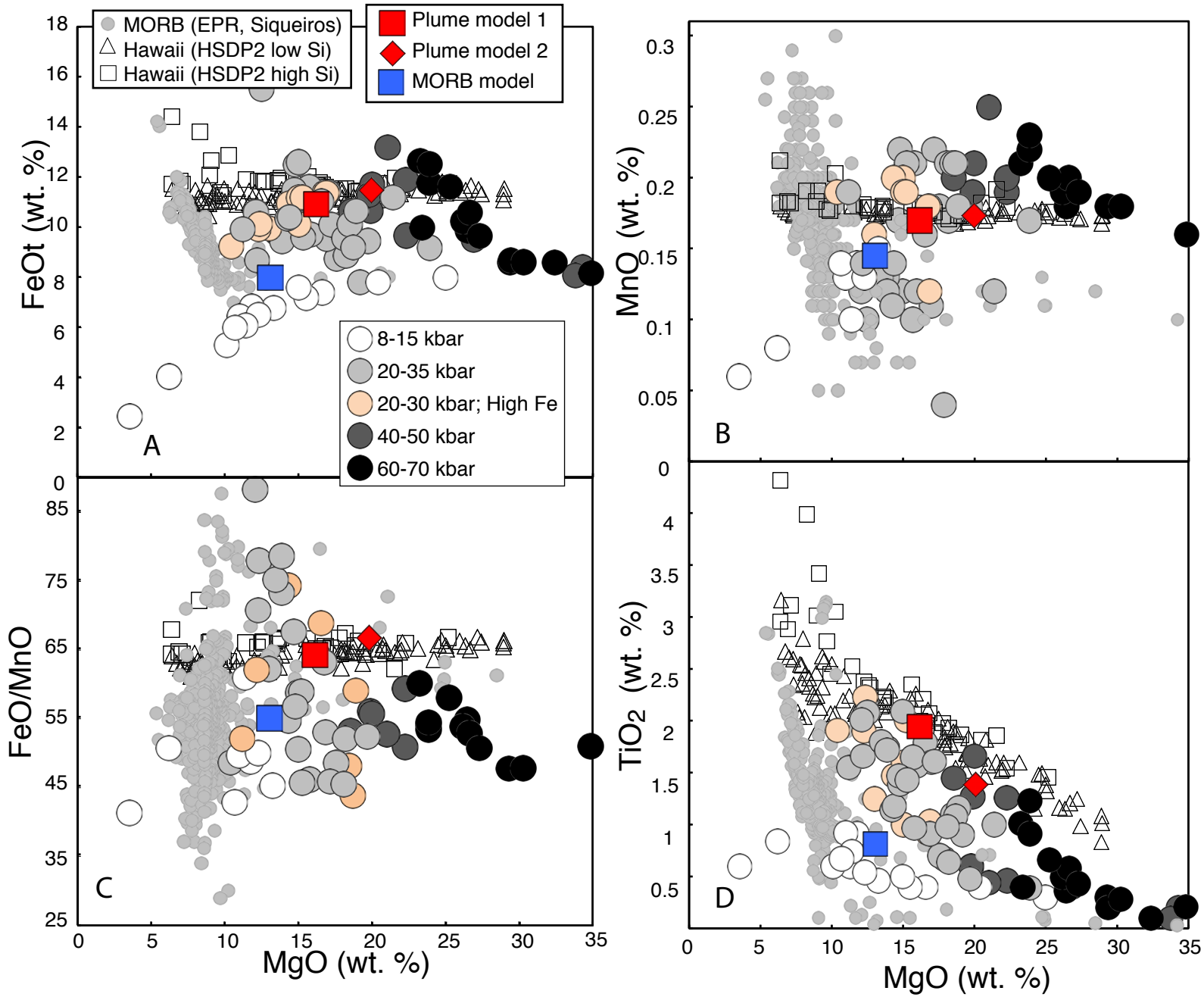


Figure 2

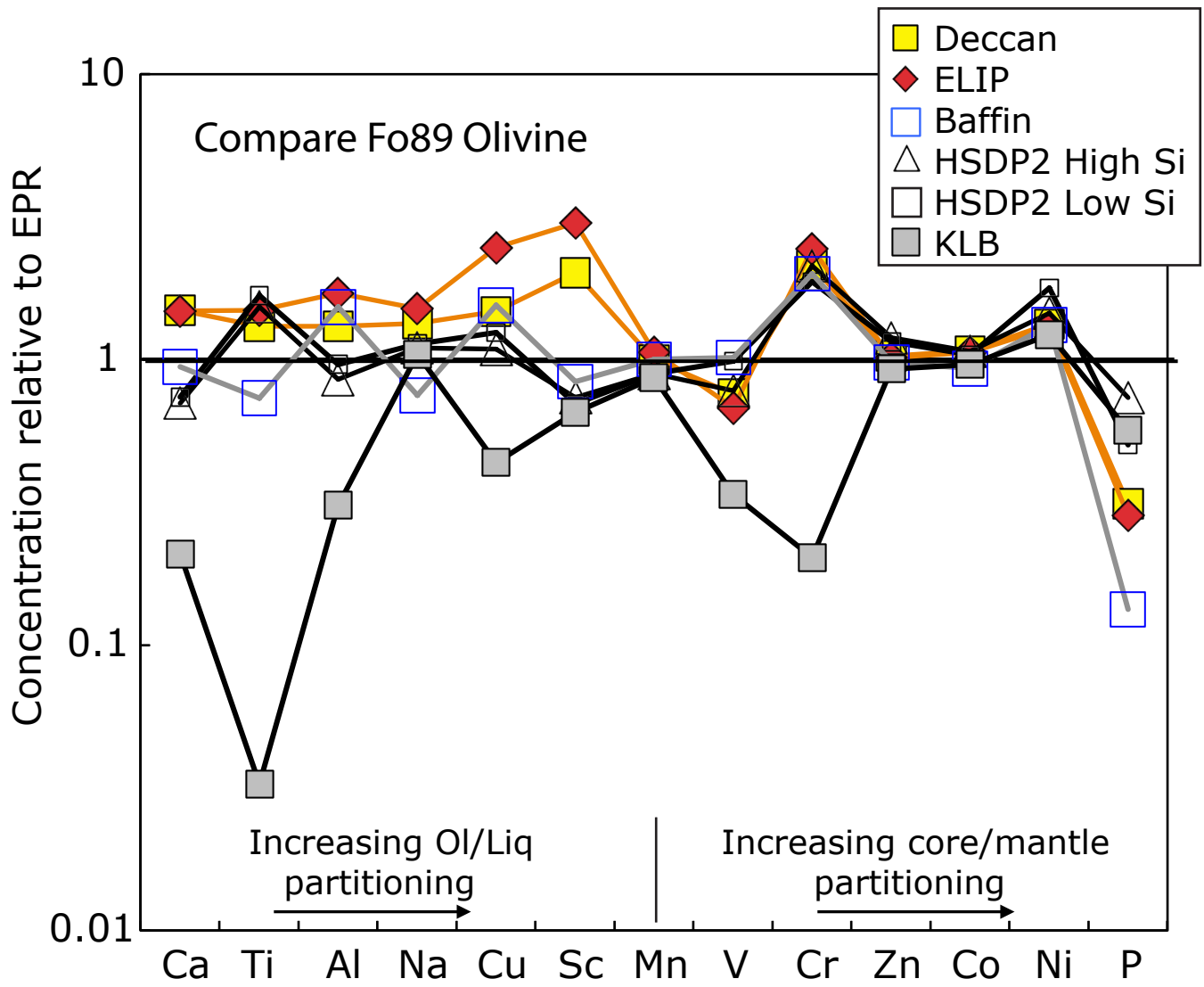


Figure 3

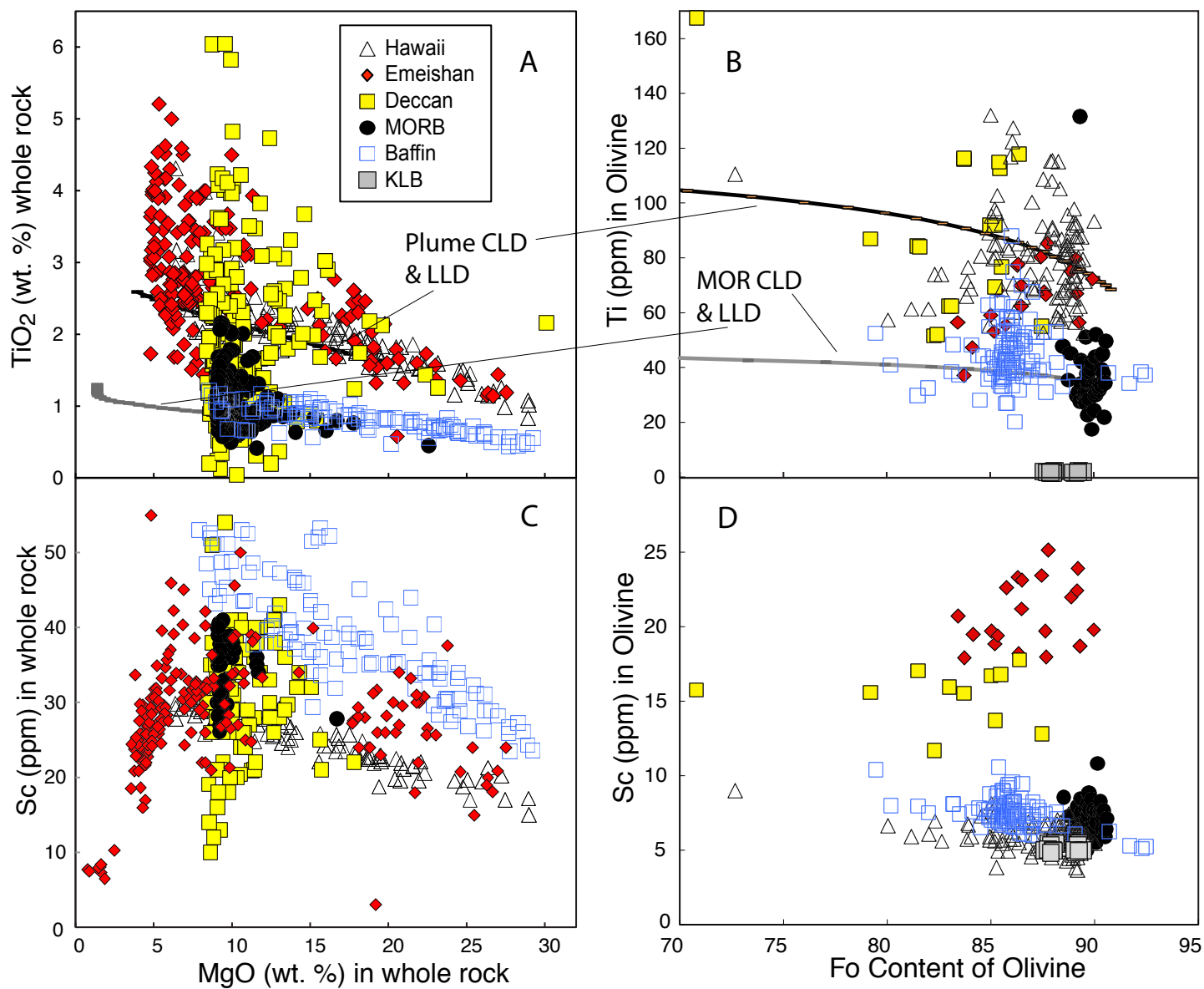


Figure 4

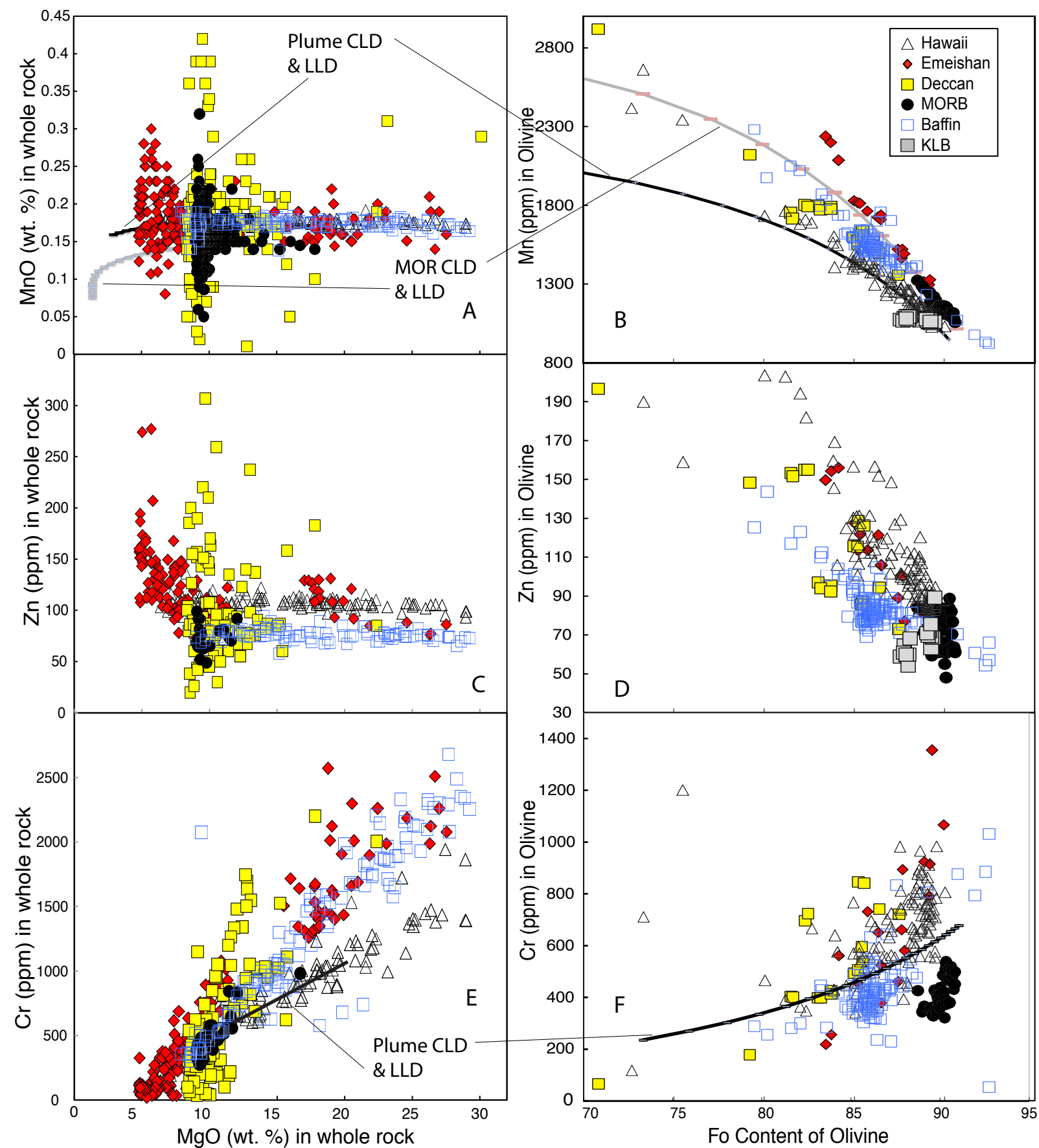


Figure 5

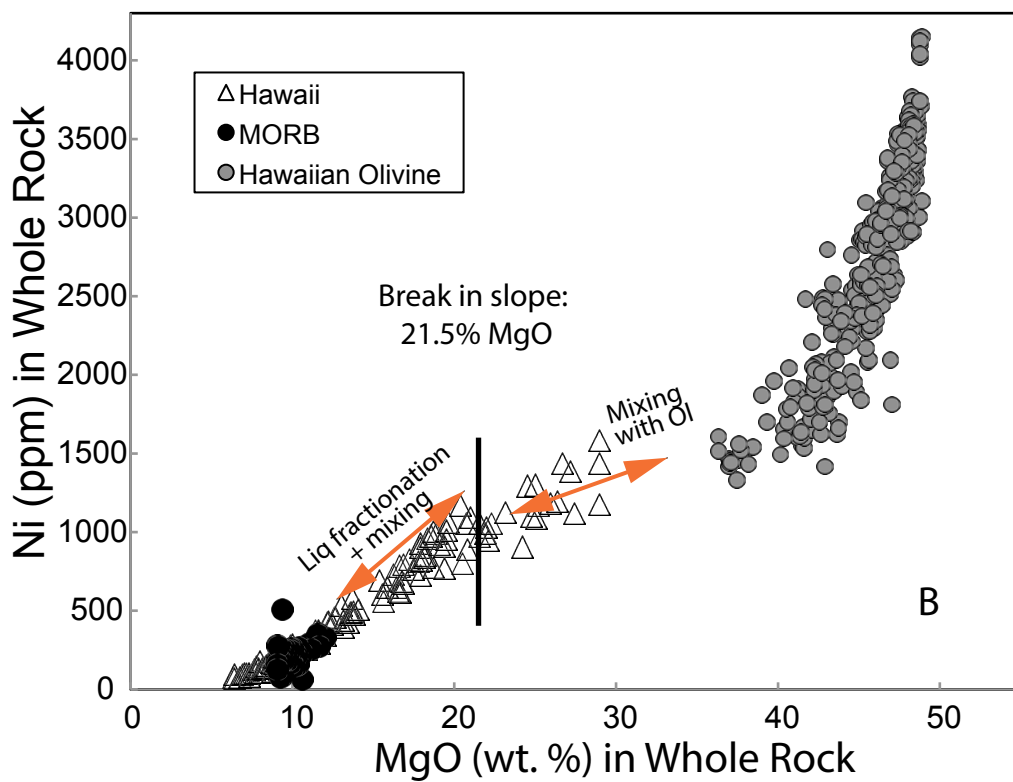
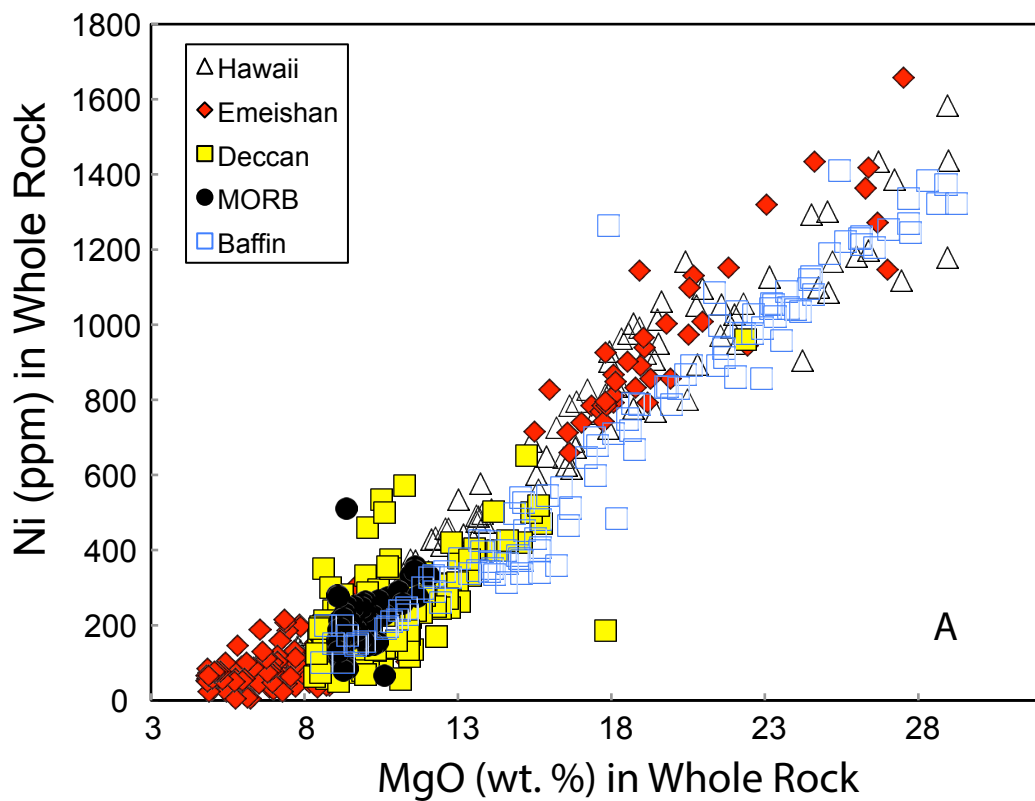


Figure 6E

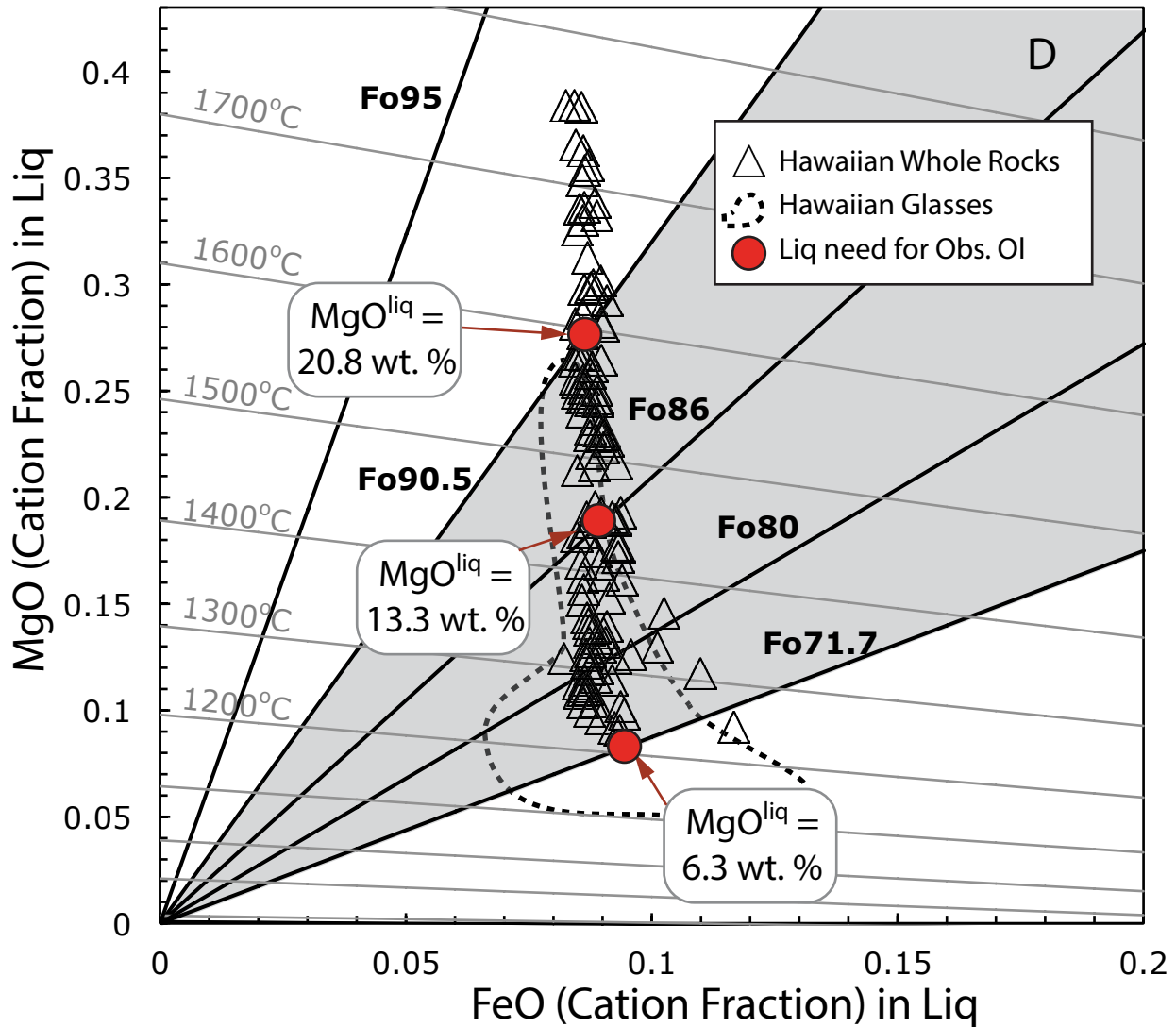
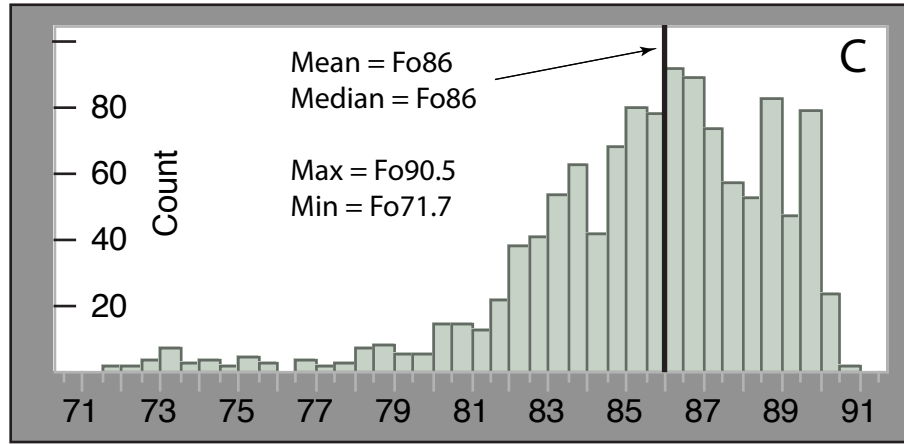


Figure 6

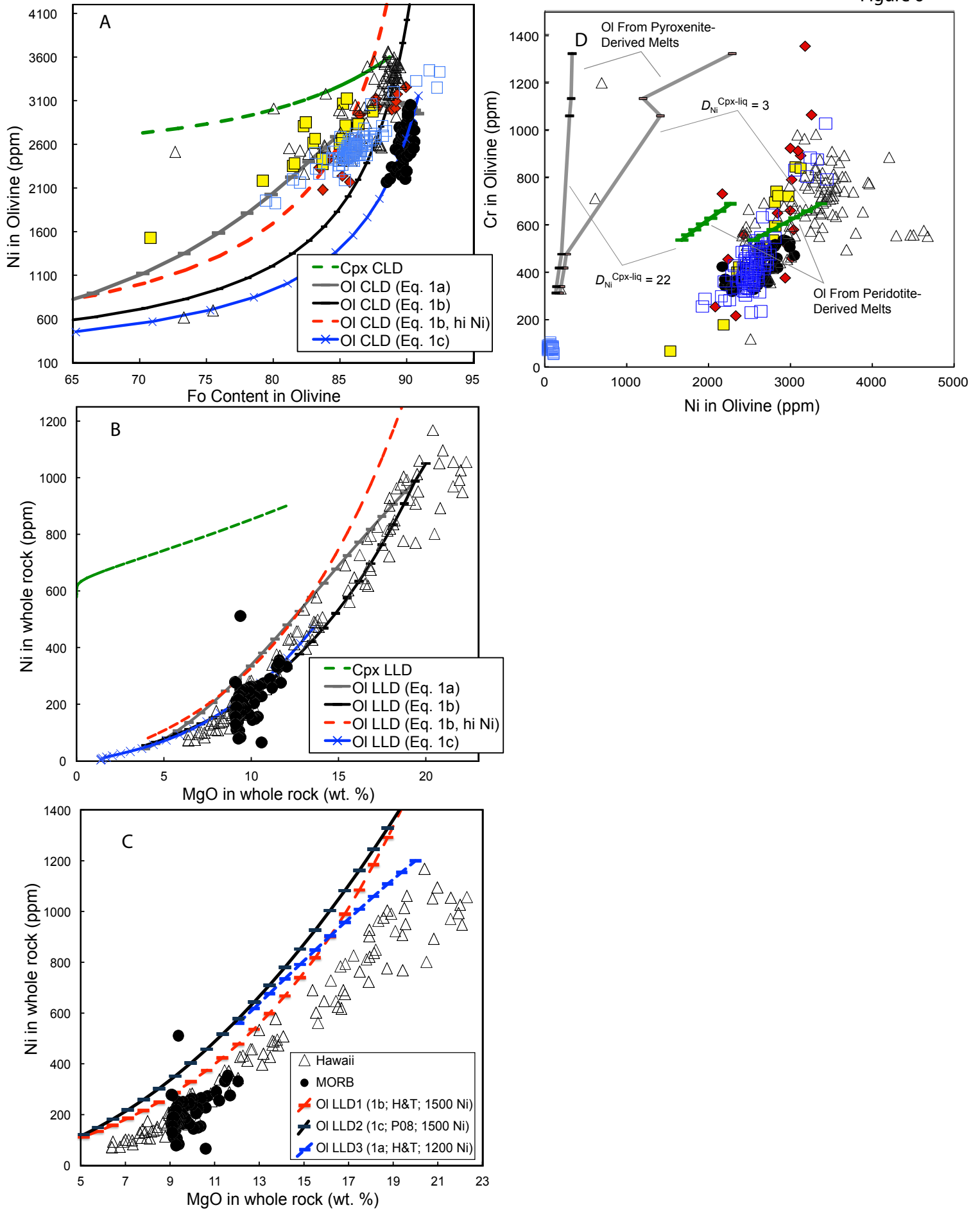


Figure 7

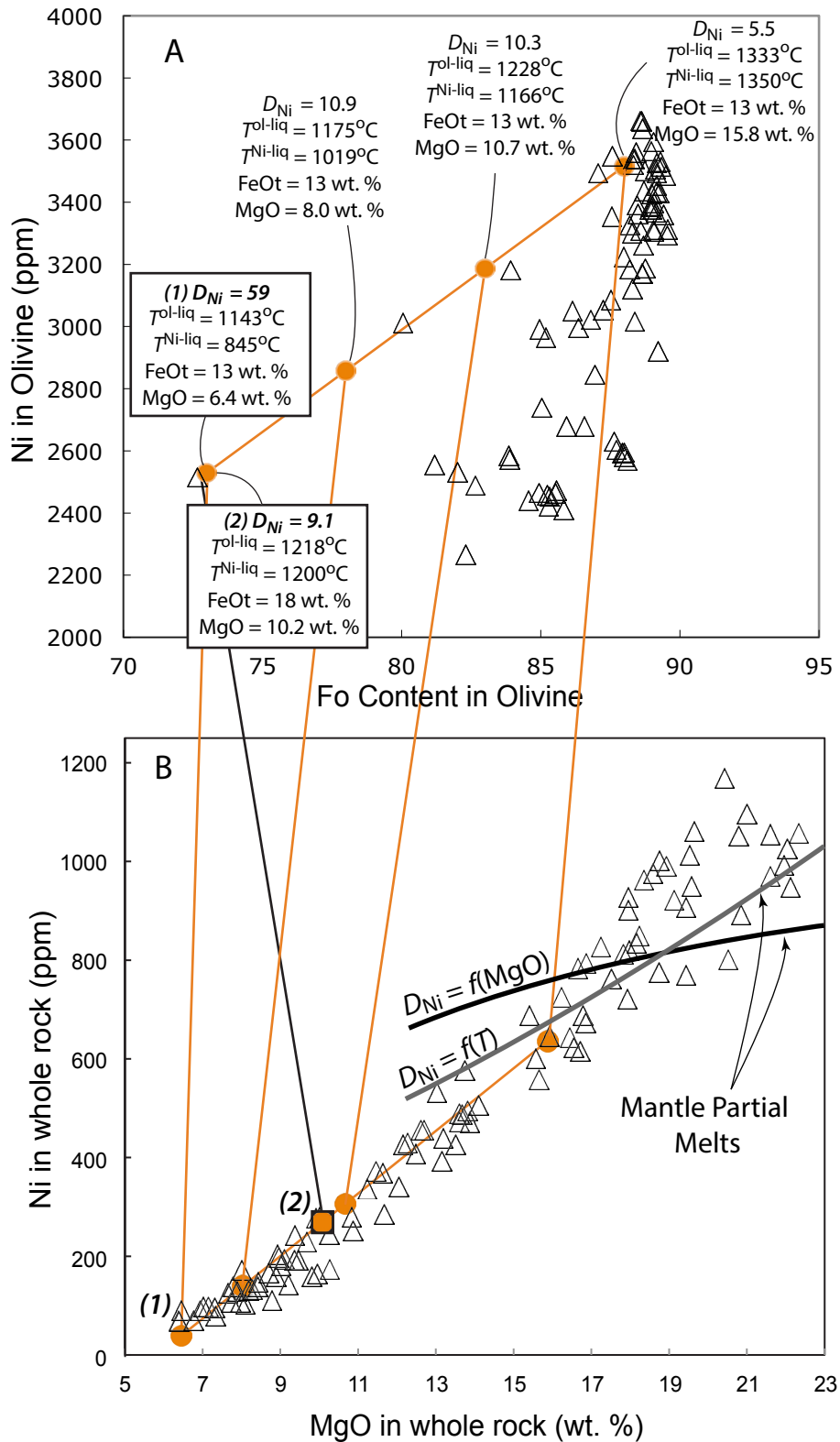


Figure 8

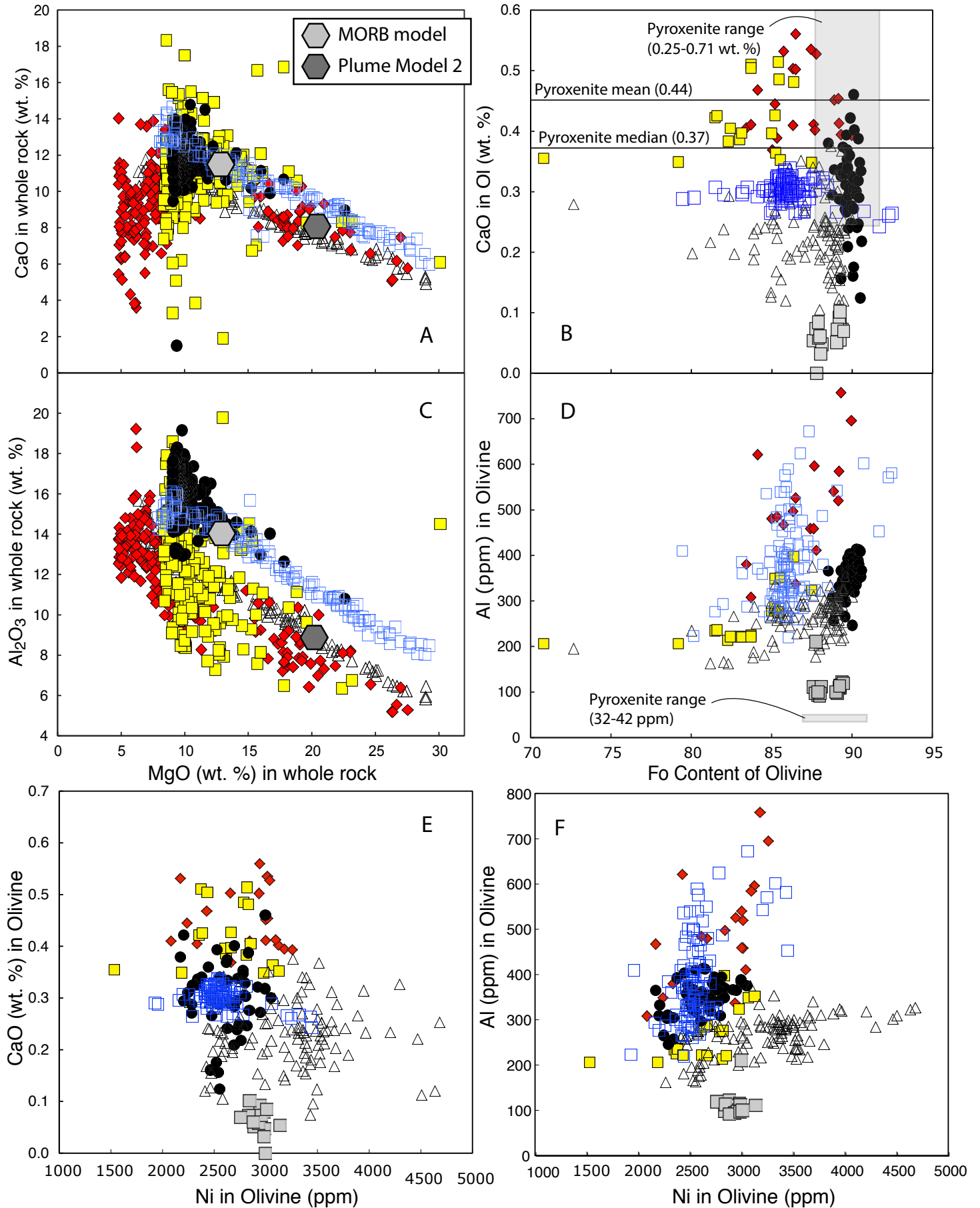


Figure 9

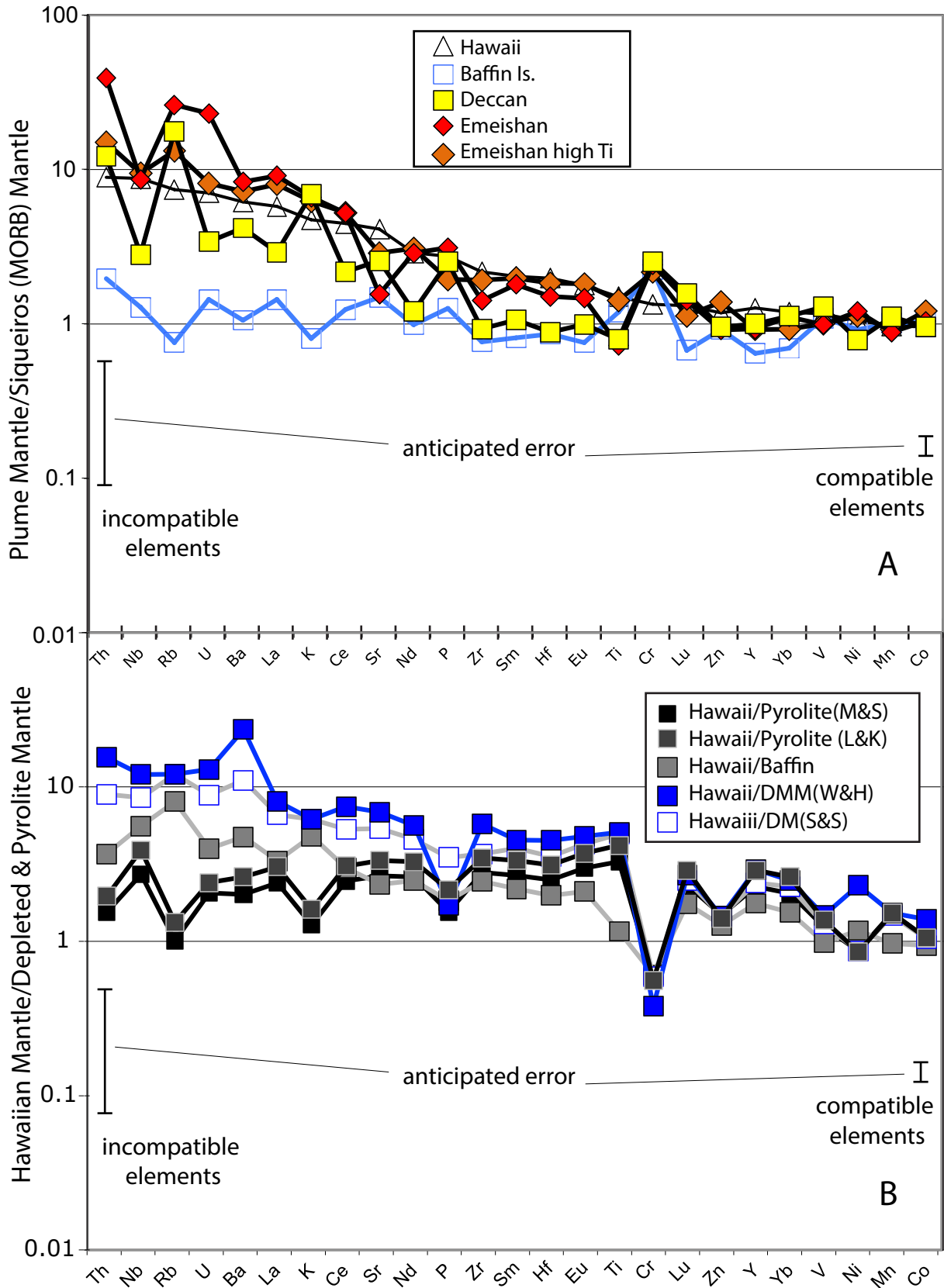
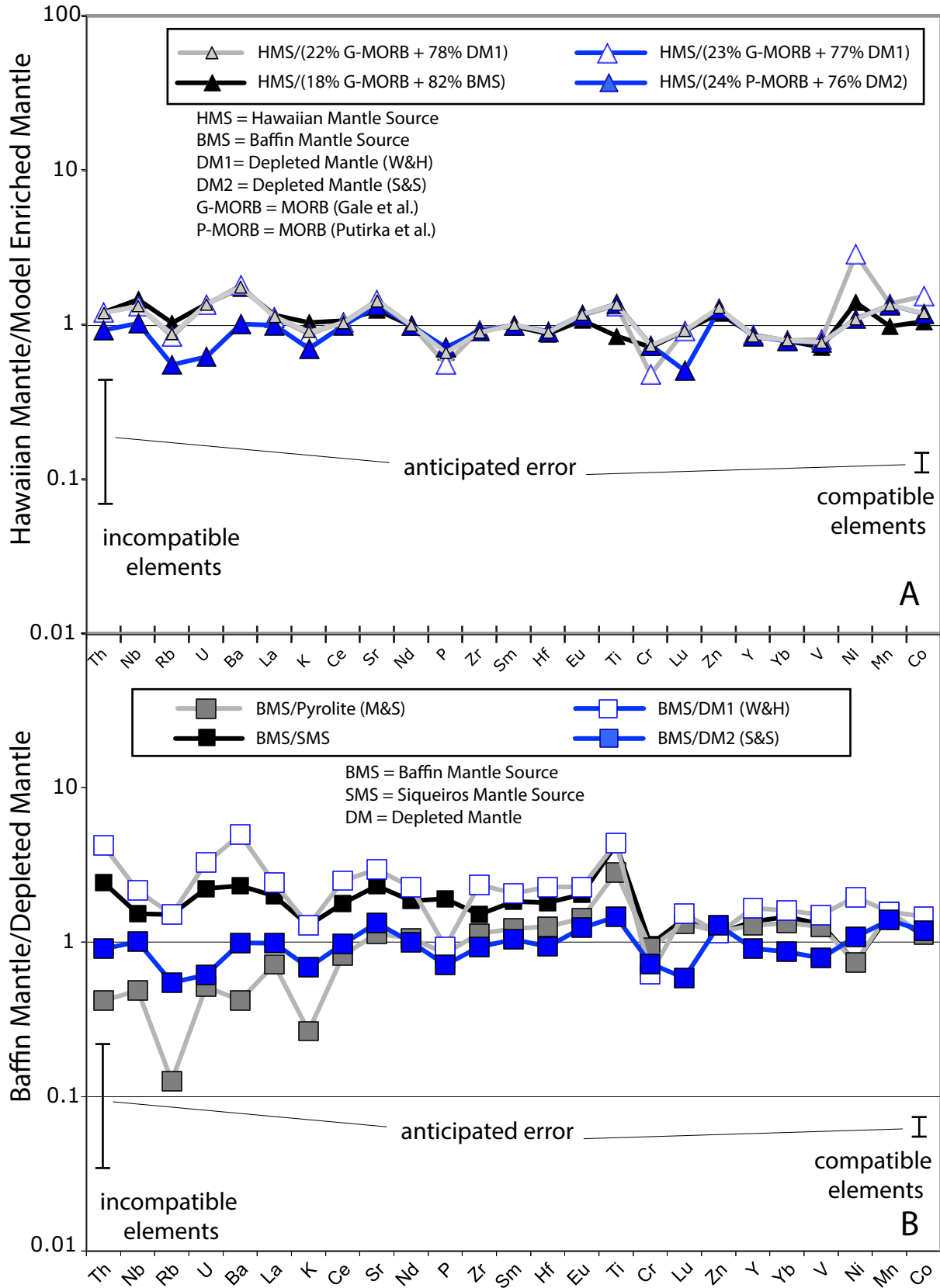


Figure 10



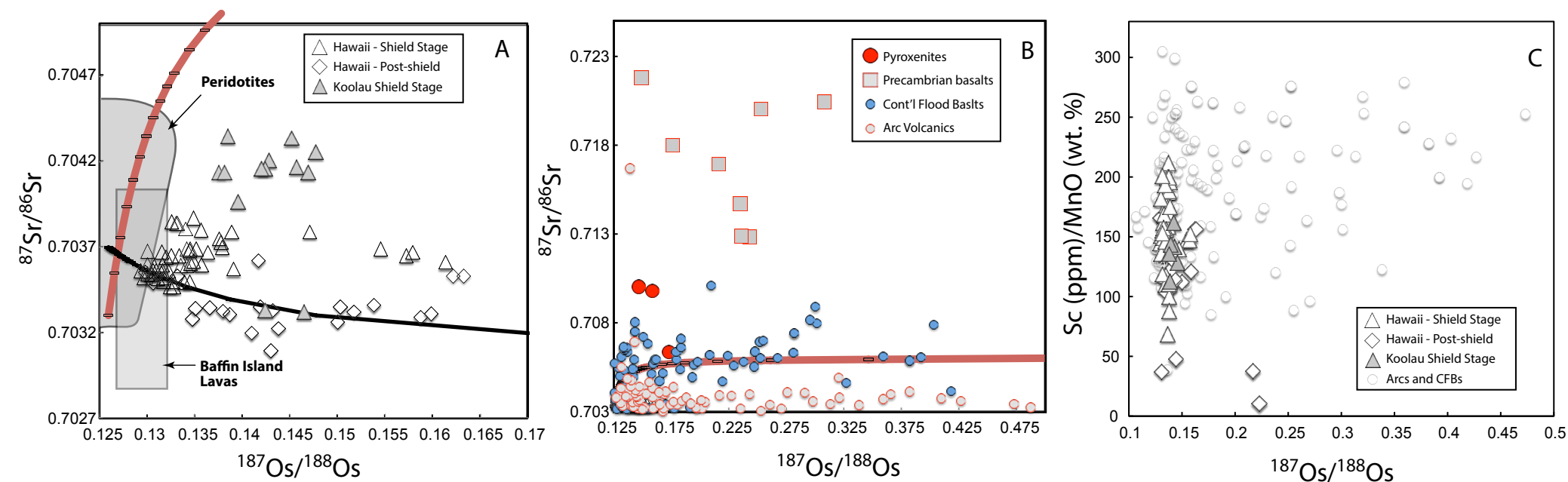


Figure ES1. $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{187}\text{Os}/^{188}\text{Os}$ ratios for (A) Hawaiian shield stage and post-shield lavas, with shield stage Koolau lavas highlighted; also shown are fields for peridotites and lavas from Baffin Island, and (B) Arc lavas, Continental Flood Basalts, Pyroxenites, and Precambrian lavas. Orange curves show mixing between the Kogiso et al. (2004) components of peridotite ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7033$, $[\text{Sr}] = 10$ ppm, $^{187}\text{Os}/^{188}\text{Os} = 0.126$, and $[\text{Os}] = 300$ ppt), and a pyroxenite ($^{87}\text{Sr}/^{86}\text{Sr} = 0.706$, $[\text{Sr}] = 100$ ppm, $^{187}\text{Os}/^{188}\text{Os} = 2$, and $[\text{Os}] = 10$ ppt). The black curves shows mixing between this same peridotite source, and a hypothetical source similar to the pyroxenite, but with $^{87}\text{Sr}/^{86}\text{Sr} = 0.702$ ($[\text{Sr}] = 100$ ppm, $^{187}\text{Os}/^{188}\text{Os} = 2$, and $[\text{Os}] = 10$ ppt). In (C), $^{187}\text{Os}/^{188}\text{Os}$ is compared to Sc/MnO, which should be either fractionated by pyroxene- or garnet-rich source materials. And in (D) we see that Al_2O_3 contents are not any higher, relative to degree of fractionation (MgO) for Hawaiian lavas with elevated $^{187}\text{Os}/^{188}\text{Os}$. These plots collectively show that a pyroxenite source, or any ancient source material, is an unlikely candidate for high $^{187}\text{Os}/^{188}\text{Os}$ ratios, as these would likely have much greater $^{87}\text{Sr}/^{86}\text{Sr}$ than supposed by Kogiso et al. (2004), and because there is no distinctive mineralogic signal with respect to major oxides or trace elements. An intriguing pattern is that CFBs and arcs are both enriched in $^{187}\text{Os}/^{188}\text{Os}$ with CFBs ranging to higher $^{87}\text{Sr}/^{86}\text{Sr}$, and with Hawaiian lavas sharing a muted arc-like $^{187}\text{Os}/^{188}\text{Os}$ enrichment. These patterns point to a shallow source for high $^{187}\text{Os}/^{188}\text{Os}$, one that may be driven by assimilation/mixing processes, with a crustal influence for CFBs (and so their elevated $^{87}\text{Sr}/^{86}\text{Sr}$), but not so (or much less so) for oceanic islands and arcs.