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1 Revision 2

2 A New Clinopyroxene-liquid Barometer, and Implications for Magma
3 Storage Pressures under Icelandic Rift Zones

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25 **Abstract**

26 Pressure is one of the key variables that controls magmatic phase equilibria. However, estimating magma
27 storage pressures from erupted products can be challenging. Various barometers have been developed
28 over the past two decades that exploit the pressure-sensitive incorporation of jadeite (Jd) into
29 clinopyroxene. These Jd-in-clinopyroxene barometers have been applied to rift zone magmas from
30 Iceland, where published estimates of magma storage depths span the full thickness of the crust, and
31 extend into the mantle. Tests performed on commonly used clinopyroxene-liquid barometers with data
32 from experiments on H₂O-poor tholeiites in the 1 atm to 10 kbar range reveal substantial pressure-
33 dependent inaccuracies, with some models overestimating pressures of experimental products equilibrated
34 at 1 atm by up to 3 kbar. The pressures of closed-capsule experiments in the 1–5 kbar range are also
35 overestimated, and such errors cannot be attributed to Na loss, as is the case in open furnace experiments.
36 The following barometer was calibrated from experimental data in the 1 atm to 20 kbar range to improve
37 the accuracy of Jd-in-clinopyroxene barometry at pressures relevant to magma storage in the crust:

$$P(\text{kbar}) = -26.27 + 39.16 \frac{T(\text{K})}{10^4} \ln \left[\frac{X_{\text{Jd}}^{\text{cpx}}}{X_{\text{NaO}_{0.5}}^{\text{liq}} X_{\text{AlO}_{1.5}}^{\text{liq}} (X_{\text{SiO}_2}^{\text{liq}})^2} \right] - 4.22 \ln(X_{\text{DiHd}}^{\text{cpx}}) + 78.43 X_{\text{AlO}_{1.5}}^{\text{liq}} + 393.81 (X_{\text{NaO}_{0.5}}^{\text{liq}} X_{\text{KO}_{0.5}}^{\text{liq}})^2$$

38 This new barometer accurately reproduces its calibration data with a standard error of estimate (SEE) of
39 ± 1.4 kbar, and is suitable for use on hydrous and anhydrous samples that are ultramafic to intermediate in
40 composition, though should be used with caution below 1100 °C and at oxygen fugacities greater than one
41 log unit above the QFM buffer. Tests performed using with data from experiments on H₂O-poor tholeiites
42 revealed that 1 atm runs were overestimated by less than the model precision (1.2 kbar); the new
43 calibration is significantly more accurate than previous formulations. Many current estimates of magma
44 storage pressures may therefore need to be reassessed. To this end, the new barometer was applied to
45 numerous published clinopyroxene analyses from Icelandic rift zone tholeiites that were filtered to
46 exclude compositions affected by poor analytical precision or collected from disequilibrium sector zones.
47 Pressures and temperatures were then calculated using the new barometer in concert with equation 33
48 from Putirka (2008). Putative equilibrium liquids were selected from a large database of Icelandic glass

49 and whole-rock compositions using an iterative scheme because most clinopyroxene analyses were too
50 primitive to be in equilibrium with their host glasses. High-Mg# clinopyroxenes from the highly primitive
51 Borgarfraun eruption in north Iceland record a mean pressure in the lower crust (4.8 kbar). All other
52 eruptions considered record mean pressures in the mid-crust, with primitive clinopyroxene populations
53 recording slightly higher pressures (3.5–3.7 kbar) than evolved populations (2.5–2.8 kbar). Thus, while
54 some magma processing takes place in the shallow crust immediately beneath Iceland’s central volcanoes,
55 magma evolution under the island’s neovolcanic rift zones is dominated by mid-crustal processes.

56

57 **Key words:** thermobarometry; clinopyroxene-liquid equilibria; Iceland; magma plumbing

58

59

INTRODUCTION

Clinopyroxene-liquid Barometry—Background and Applications

61 Alongside composition, crystallinity, temperature and oxygen fugacity, pressure is one of the primary
62 intensive variables that controls magmatic phase equilibria (Yoder and Tilley 1962; Blundy and Cashman
63 2008). This raises the possibility of estimating magma storage pressures using observed phase equilibria
64 relations and information from other intensive variables. Indeed, determining magma storage pressures,
65 and hence depths, is essential for various reasons. For example, understanding the distribution of magma
66 storage depths within the lithosphere provides information about crustal formation mechanisms in both
67 oceanic and continental settings (Henstock et al. 1993; Kelemen et al. 1997; Annen et al. 2006).
68 Estimating pre-eruptive magma storage depths is also essential for integrating petrological records of
69 magmatism with expressions of ongoing unrest such as seismicity, ground deformation and gas emission
70 in volcanically active regions (Edmonds 2008; Sigmundsson et al. 2010; Tarasewicz et al. 2014). With the
71 aim of improving the accuracy with which magma storage pressures can be estimated, we assessed the
72 performance of various published clinopyroxene-liquid barometers. To anticipate our results, we find that
73 some barometers overestimate reported experimental pressures at <7 kbar. We thus develop a new jadeite-
74 in-clinopyroxene barometer optimized for use at crustal pressures on hydrous and anhydrous

75 compositions. We then recalculate storage pressures for a series of Icelandic rift tholeiites from which
76 diverse clinopyroxene-liquid pressures have been reported in recent years.

77
78 The number of barometric tools available for estimating magma storage pressures has grown considerably
79 over the past 25 years. Current barometric methods range from exploiting volatile solubility laws (e.g.,
80 Newman and Lowenstern 2002; Moore 2008) or equations of state (Hansteen and Klügel 2008) to
81 determine melt and fluid inclusion entrapment pressures, through to calibrating pressure-sensitive phase
82 equilibria relations using experimental data (e.g., Putirka 2008). However, these various barometric
83 methods are subject to numerous assumptions and unavoidably return pressure estimates with
84 considerable uncertainties. For example, while the solubilities of H₂O and CO₂ in silicate melts are
85 generally well understood (Dixon and Stolper 1995; Lesne et al. 2011; Shishkina et al. 2014), many recent
86 studies have demonstrated that interpreting melt inclusion entrapment pressures can be complicated by
87 post-entrapment processes such as diffusive reequilibration, shrinkage bubble formation and the
88 precipitation of solid carbon phases (Bucholz et al. 2013; Hartley et al. 2014; Moore et al. 2015; Wallace
89 et al. 2015). Although condensed phase barometry (i.e., barometry based on solid-solid or solid-liquid
90 phase equilibria) can avoid the challenges presented by melt and fluid inclusions, it is fundamentally
91 limited in other ways. For example, determining magma storage pressures is complicated not only by the
92 mostly small volume contrasts associated with most mineral-mineral or mineral-liquid equilibria, but also
93 by the small number of phases present in many magmas (e.g., melt + olivine ± plagioclase ±
94 clinopyroxene in the case of many mafic magmas; Grove et al. 1992), which limits the number of
95 reactions available for barometric use.

96
97 Fortunately for igneous petrologists, the incorporation of jadeite (NaAlSi₂O₆; Jd) into clinopyroxene is
98 strongly pressure dependent, with high Jd contents stabilized at high pressures as a result of the large
99 partial molar volume change associated with the formation of the Jd component (Putirka et al. 1996;
100 Holland and Powell 1998; Putirka 2016). About twenty years ago, Putirka et al. (1996) presented a series

101 of equations describing the pressure-dependent Jd-liquid (liq) reaction, as well as the strongly
102 temperature-dependent Jd into diopside-hedenbergite ($\text{Ca}(\text{Mg,Fe})\text{Si}_2\text{O}_6$; DiHd) and calcium Tschermak's
103 component (CaAlAlSiO_6 ; CaTs) into DiHd exchange reactions. These thermobarometric equations were
104 subsequently reformulated in 2003 to extend their applicability to felsic and hydrous systems (Putirka et
105 al. 2003), before being reviewed further five years later (Putirka 2008).

106

107 Perhaps unsurprisingly, these clinopyroxene-liquid thermobarometers have now seen extensive use for
108 some years. A far from exhaustive list of barometric studies that have used clinopyroxene-liquid equilibria
109 would include the description of deep, multi-level magmatic differentiation under the Canary Islands
110 (Hansteen et al. 1998; Klügel et al. 2005; Stroncik et al. 2009), as well as the identification and
111 subsequent re-evaluation of deep fractionation recorded by high-Al clinopyroxene crystals in alkaline
112 magmas from Haleakala, Hawaii (Chatterjee et al. 2005; Hammer et al. 2016). Clinopyroxene-liquid
113 equilibria have also been used to define trans-crustal magma plumbing systems under Mt. Etna amongst
114 other locations (Giacomoni et al. 2016). To improve the suitability of clinopyroxene-liquid
115 thermobarometers for studying evolved alkaline systems, a series of recalibrated thermobarometric
116 expressions have been developed and applied to Mt. Vesuvius and Campi Flegrei in Italy (Masotta et al.
117 2013), as well as to Nemrut in Turkey (Macdonald et al. 2015).

118

119 Clinopyroxene-liquid thermobarometers have also been applied to the products of several Icelandic
120 eruptions. For example, clinopyroxene-liquid storage pressure from the primitive Borgarhraun lava in the
121 on-axis Northern Volcanic Zone (NVZ) of Northern Iceland indicate that clinopyroxene crystallization
122 occurred close to the Moho (>20 km; MacLennan et al. 2003a; Winpenny and MacLennan 2011). These
123 high storage pressures are also consistent with phase equilibria experiments and estimates from olivine-
124 plagioclase-augite-melt (OPAM) boundary barometry (Yang et al. 1996; MacLennan et al. 2012). In the
125 southern, off-axis extension of Iceland's Eastern Volcanic Zone (sEVZ), a bimodal distribution of storage
126 depths has been inferred for the 2010 Eyjafjallajökull-Fimmvörðuháls eruption, with the basalts erupted

127 from the flank thought to have been sourced from substantially greater depths (16–18 km) than the
128 benmoreites erupted from the summit (2–5 km; Keiding and Sigmarsson 2012). Similar vertically
129 extensive magma plumbing systems have also been inferred from the products of the neighboring Katla
130 volcanic system (Budd et al. 2016).

131

132 The importance of mid-crustal storage has also been demonstrated in the products of the 1783–1784 Laki
133 and 10 ka Grímsvötn tephra series eruptions from the on-axis portion of the Eastern Volcanic Zone (EVZ;
134 Neave et al. 2013, 2015). However, these studies of the EVZ proper used substantially different
135 approaches from those used in studies of the sEVZ. Firstly, instead of using the carrier melt as the
136 equilibrium liquid for thermobarometric calculations, putative equilibrium liquids were selected from a
137 large database of Icelandic glass and whole-rock analyses on the basis of being in Fe-Mg, Ti and CaTs
138 component equilibrium with measured clinopyroxenes (e.g., Wippeny and MacLennan 2011). In other
139 words, no *a priori* assumptions about which liquids would be in equilibrium with clinopyroxene crystals
140 were made. Avoiding such assumptions was deemed to be essential because of the extensive
141 disequilibrium within and between the different components of Icelandic magmas (Halldórsson et al.
142 2008; MacLennan 2008; Neave et al. 2014). For example, even though Keiding and Sigmarsson (2012)
143 and Budd et al. (2016) performed thermobarometric calculations on clinopyroxene compositions in Fe-
144 Mg equilibrium with co-erupted glass or whole-rock compositions, only the outermost rims of analyzed
145 crystals may have been co-genetic with the melts that carried them to the surface; magma mixing could
146 have decoupled earlier growth phases of such crystals from their equilibrium melts. Besides, even if a
147 system establishes Fe-Mg exchange equilibrium, the equilibration of pressure-dependent NaAl-CaMg
148 exchange is by no means guaranteed; different components may equilibrate at different rates. Filtering for
149 Fe-Mg equilibrium alone may thus be an insufficient test for equilibrium in systems where mixing is
150 prevalent (Zellmer et al. 2014). Moreover, Fe-Mg exchange is insensitive to the disequilibrium growth of
151 clinopyroxene, and testing for equilibrium across multiple clinopyroxene components is probably more
152 robust (Mollo et al. 2013a). Secondly, the accuracy of the barometric equations used in the EVZ studies

153 was tested by applying them to experiments on mafic compositions in the 1–7 kbar range. The pressures
154 from experimental clinopyroxene-liquid pairs were generally found to exceed the pressures at which
155 experiments were carried out, so empirical corrections were applied to the results of calculations
156 performed in clinopyroxene from the Laki and 10 ka Grímsvötn tephra series eruptions (Neave et al.
157 2013, 2015).

158
159 The magnitudes of empirical corrections proposed by Neave et al. (2013, 2015) (–1.5 and –2.7 kbar
160 respectively at a true pressure of 3 kbar) are comparable with the 2–3 kbar overestimation of most 1 atm
161 experimental data noted by Putirka (2008) when testing various Jd-in-clinopyroxene barometers (Putirka
162 et al. 1996, 2003). This overestimation was initially attributed to Na loss during open furnace experiments
163 artificially shifting the equilibrium constant of Jd formation in favor of higher pressure estimates (Tormey
164 et al. 1987; Putirka 2008). However, the persistence of this overestimation when barometers are applied to
165 the products of higher pressure experiments carried out in closed capsules suggests that Na volatilization
166 alone cannot account for the poor accuracy of Jd-in-clinopyroxene barometers at low pressures.
167 Unfortunately, the tests carried out by Neave et al. (2013, 2015) are insufficient to evaluate the
168 performance of currently available barometers robustly: they were performed using an incomplete
169 experimental database that was not filtered for the attainment of clinopyroxene-liquid equilibrium. To
170 provide more reliable evaluation of published barometers, we therefore carried out a series of tests on a
171 much larger experimental database.

172

173 **Evaluating the Performance of Published Clinopyroxene-liquid Barometers**

174 When the first Jd-in-clinopyroxene barometer was proposed by Putirka et al. (1996), few high-
175 temperature phase equilibria data were available for mafic systems above 1 atm and below 7 kbar—
176 arguably, the pressure interval where most magma reservoirs are located (e.g., Singh et al. 2006; Cashman
177 and Sparks 2013; Tarasewicz et al. 2014; Gudmundsson et al. 2016)—because of the experimental
178 challenges of working under these conditions. Many 1 atm experiments carried out before the mid-1990s

179 also experienced Na loss, limiting their use in calibrating Jd exchange equilibria (Tormey et al. 1987).
180 However, a range of technological developments over the past few decades have greatly expanded the
181 number of low- to moderate-pressure (≤ 7 kbar) experiments available for calibrating and testing
182 thermobarometers. Notable advancements include: introducing procedures for minimizing Na loss during
183 open furnace experiments (Tormey et al. 1987; Yang et al. 1996); developing rapid quench devices for
184 internally heated pressure vessels (IHPVs; Berndt et al. 2002; Holloway et al. 1992; Moore and
185 Carmichael 1998); and modifying piston cylinder assemblies to achieve reproducible pressures when
186 operating below 7 kbar (Moore et al. 2008).

187
188 Phase equilibria experiments from the following sources were thus compiled to evaluate published
189 clinopyroxene-liquid barometers: Berndt et al. (2005), Botcharnikov et al. (2008), Grove et al. (1992),
190 Husen et al. (2016), Thy et al. (2006), Toplis and Carroll (1995), Villiger et al. (2004, 2007), Whitaker et
191 al. (2007, 2008) and Yang et al. (1996). We only include low-H₂O (H₂O ≤ 1 wt.%) experiments that were
192 carried out at ≤ 10 kbar with an fO_2 between the C-CO₂-CO buffer (i.e., in equilibrium with graphite
193 capsules) and one log unit above the QFM buffer (QFM+1). These conditions encompass the anticipated
194 magma storage conditions for tholeiitic basalts in oceanic settings (Michael 1995; Cottrell and Kelley
195 2011). They also exclude the oxidizing conditions ($fO_2 > \text{QFM}+1$) under which the presence of Fe³⁺ in
196 clinopyroxene can stabilize an aegirine component (NaFe³⁺Si₂O₆; Aeg) that complicates the solution of
197 Na into pyroxene, with an unknown impact on the true Jd fraction, compared to that calculated from
198 major element oxide data (Blundy et al. 1995). Furthermore, only clinopyroxene-liquid pairs with
199 $K_{d \text{ Fe-Mg}}^{\text{cpx-liq}}$ values within 20% of equilibrium values calculated using equation 35 from Putirka (2008) were
200 considered. The compiled experimental data are provided in the electronic appendix.

201
202 The results of our tests on previously published clinopyroxene-liquid barometers are summarized in
203 Figure 1. All barometric calculations were performed using experimental temperatures and quoted H₂O

204 contents where available. Where no H₂O contents were provided, experiments were assumed to be
205 anhydrous. Calculations using data from Villiger et al. (2004, 2007), as well as data from composition 70-
206 002 of Grove et al. (1992), returned extremely variable pressure estimates and were therefore excluded
207 from regressions performed to evaluate the performance of the barometers. We also excluded the results
208 of calculations from a few 1 atm experiments that returned spurious pressures of less than -8 kbar.

209
210 Figure 1a shows the performance of model P1 from Putirka et al. (1996), the first formulation of the Jd-
211 in-clinopyroxene barometer (standard error of estimate = 1.4 kbar). Although the test dataset can be fitted
212 reasonably well using a simple linear model ($r^2 = 0.81$, SEE = 1.1 kbar), the regression does not pass
213 through the origin. Specifically, while the estimates for high pressure (≥ 7 kbar) experiments lie within one
214 SEE of experimental values (i.e., within 1.4 kbar), estimates for low-pressure experiments are
215 significantly overestimated (intercept = 3.0 kbar), resulting in a mean residual between calculated and
216 experimental pressures of 2.3 kbar. However, given that this model was only calibrated using ≥ 8 kbar
217 experiments, the poor accuracy at lower pressures is perhaps not surprising.

218
219 Figure 1b shows the performance of equation 30 from Putirka (2008), a recalibrated Jd-in-clinopyroxene
220 barometer (calibration SEE = 1.6 kbar) designed for use with evolved and hydrous compositions as well
221 as with primitive and nominally anhydrous compositions (cf., Putirka et al. 2003). Despite being
222 calibrated with a larger and more compositionally diverse dataset, this barometer performed similarly to
223 model P1 from Putirka et al. (1996) in our tests: $r^2 = 0.82$, SEE = 0.9 kbar, intercept = 3.0 kbar and mean
224 residual = 2.1 kbar. Nevertheless, the low SEE is consistent with the analysis of Putirka (2008) that this is
225 the most precise formulation of the Jd-in-clinopyroxene barometer to date for mafic systems.

226
227 Figure 1c shows the performance of equation 32a from Putirka (2008), a barometer that, following the
228 approach of Nimis (1995), is based on temperature and clinopyroxene composition alone (calibration SEE
229 = 3.1 kbar). Although the test data are fitted well by a simple linear model ($r^2 = 0.89$; SEE = 0.9), pressure

230 estimates are consistently ~1–2 kbar too high, with the magnitude of the overestimation being greatest at
231 1 atm (intercept = 2.3 kbar, mean residual 2.0 kbar). We therefore suggest that the systematic
232 overestimation of pressure noted by Putirka (2008) when applying this model to hydrous compositions
233 may extend to H₂O-poor compositions at low to moderate pressures (<10 kbar).

234

235 Figure 1d shows the performance of equation 32c from Putirka (2008), a barometer based on Al
236 partitioning between clinopyroxene and liquid (SEE = 1.5 kbar). This barometer performed worst out of
237 the four barometric expressions tested: $r^2 = 0.70$, SEE = 1.5 kbar, intercept = 2.6 kbar and mean residual
238 1.8 kbar. Despite exploiting Al partitioning instead of Jd formation, this barometer shows a similar
239 pressure-dependent accuracy to the two Jd-in-clinopyroxene barometers shown in Figures 1a and 1b: the
240 barometer is accurate at high pressures (≥ 7 kbar), but increasingly inaccurate at lower pressures.

241

242 A crucial observation from Figure 1 is that pressure overestimation is not restricted to experiments carried
243 out at 1 atm in which Na loss by volatilization may have occurred. Applying Jd-in-clinopyroxene
244 barometers to data from 1–4 kbar experiments carried out in zirconium-hafnium-molybdenum (ZHM)
245 cold seal, IHPV and piston cylinder equipment also results in values 1.5–3.0 kbar above experimental
246 pressures. Pressure overestimations for these closed-capsule experiments cannot be attributed to Na loss
247 from their coexisting liquids, and the cause of the Jd-in-clinopyroxene barometers' spuriously high
248 pressure estimates at low pressures must lie elsewhere. Encouragingly, though, these existing barometers
249 qualitatively, at least, recover the sense of pressure change in the test dataset: taking the mean calculated
250 pressure of the various isobaric datasets, it is possible to place these isobaric sets in the correct order. This
251 indicates that there is merit to the clinopyroxene-liquid approach to barometry, which might be resolved
252 by a suitable recalibration.

253

254

METHOD

255 The performance of a barometer depends strongly on how calibration data are selected. To calibrate a new

256 clinopyroxene-liquid barometer that is accurate at crustal pressures, it is thus essential to include low- to
 257 moderate-pressure experiments (≤ 7 kbar) in the calibration dataset. Furthermore, given that even small
 258 amounts of H₂O can have a strong effect on mineral-liquid equilibria (e.g., Almeev et al. 2007, 2012;
 259 Médard et al. 2008), it is also important to include hydrous experiments. A calibration database was thus
 260 compiled from the following sources: Blatter and Carmichael (2001), Kinzler and Grove (1992), Moore
 261 and Carmichael (1998), Putirka et al. (1996), Sisson and Grove (1993) and Yang et al. (1996). The
 262 calibration database contains a total of 113 experiments saturated in clinopyroxene \pm olivine \pm plagioclase
 263 \pm orthopyroxene \pm spinel \pm hornblende \pm magnetite \pm ilmenite. The inclusion of 1 atm experiments from
 264 Yang et al. (1996) represents a key development from the calibration datasets used in previous barometer
 265 calibrations. Importantly, this set of 1 atm experiments experienced negligible Na loss. Further details are
 266 summarized in Table 1, and the complete dataset is provided in the electronic appendix.

267
 268 Liquid and clinopyroxene components were calculated following the methods outlined in Tables 1 and 3
 269 from Putirka (2008) respectively. All the clinopyroxene compositions in the calibration dataset return six-
 270 oxygen cation sums close to 4 (3.97–4.05), confirming that they are stoichiometric. Linear regression was
 271 performed following the procedures outlined in Putirka et al. (1996) and Putirka (2008) to determine the
 272 regression coefficients. Following the qualitative success of prior models (Figure 1), the regression
 273 equation is based on a thermodynamic description of Jd formation, and includes empirical terms to
 274 improve the precision of pressure estimates. The new Jd-in-clinopyroxene barometer has the following
 275 form:

$$276 \quad P(\text{kbar}) = -26.27 + 39.16 \frac{T(\text{K})}{10^4} \ln \left[\frac{X_{\text{Jd}}^{\text{cpX}}}{X_{\text{NaO}_{0.5}}^{\text{liq}} X_{\text{AlO}_{1.5}}^{\text{liq}} (X_{\text{SiO}_2}^{\text{liq}})^2} \right] - 4.22 \ln(X_{\text{DiHd}}^{\text{cpX}}) + 78.43 X_{\text{AlO}_{1.5}}^{\text{liq}} + 393.81 (X_{\text{NaO}_{0.5}}^{\text{liq}} X_{\text{KO}_{0.5}}^{\text{liq}})^2 \quad (1)$$

277 This new barometer is calibrated for ultramafic to intermediate compositions in the 0.001–20 kbar and
 278 950–1400 °C ranges. Although this study is focused primarily on H₂O-poor tholeiites, the inclusion of
 279 H₂O-rich experiments in the calibration dataset means that the barometer is also suitable for use with
 280 hydrous compositions. However, because of Aeg component formation at high $f\text{O}_2$ (Blundy et al. 1995),

281 we advise caution when using this barometer under oxidizing conditions ($fO_2 \geq QFM+1$); we do not
282 calculate an Fe^{3+} or Aeg component, but pressures may be overestimated if Na is accidentally assigned to
283 Jd when it should be assigned to Aeg. A spreadsheet for calculating pressures is provided in the electronic
284 appendix.

285

286

RESULTS

287 Calibration data are fitted well by a simple model (Figure 2a; $r^2 = 0.95$; SEE = 1.3 kbar). Importantly, the
288 accuracy of the new barometer does not vary systematically as a function of pressure (intercept = 0.3
289 kbar); the inaccuracies visible in Figure 1 appear to have been avoided. Given that Jd-in-clinopyroxene
290 barometers depend on temperature as well as composition, they are usually solved iteratively using a
291 complementary thermometric expression, which may itself be pressure dependent. Equation 33 from
292 Putirka (2008) is the most precise clinopyroxene-liquid thermometer currently available (calibration SEE
293 = 45 °C). This thermometer was calibrated using a large global dataset that has minimal overlap with the
294 dataset used to calibrate equation 1. Figure 2b shows pressures estimated for the calibration dataset by
295 solving our new Jd-in-clinopyroxene barometer iteratively with equation 33 from Putirka (2008).
296 Encouragingly, iteratively calculated pressures are very similar to those calculated using experimentally
297 reported temperatures; neither the accuracy nor the precision of the pressure estimates suffer from being
298 calculated iteratively ($r^2 = 0.95$; SEE = 1.4 kbar; intercept = 0.6 kbar). Corresponding iteratively
299 calculated temperatures are shown in Figure 2c. Although temperatures appear to be calculated more
300 precisely than the thermometer's quoted uncertainty (SEE = 28 °C versus 45 °C from Putirka, 2008), they
301 are systematically overestimated below 1100 °C. While equation 33 from Putirka (2008) performs well
302 when paired with our new barometer at the temperatures of interest for this study (1100–1300 °C), we
303 advise caution when using this barometer-thermometer combination at low temperatures (≤ 1100 °C).

304

305 A direct comparison between pressures calculated with the original Jd-in-clinopyroxene calibration from
306 Putirka et al. (1996) and our new calibration is shown in Figure 2d. While the original model reproduces

307 the low pressures of hydrous experiments in the 0.5–2.5 kbar range (Sisson and Grove 1993; Blatter and
308 Carmichael 2001), the 1 atm experiments of Yang et al. (1996) are overestimated by ~4 kbar and do not
309 lie on a regression through the dataset. Comparing Figure 2a with Figure 2d suggests that our new
310 barometer does not suffer from the same inaccuracies at low pressures present in other clinopyroxene-
311 liquid barometers.

312

313

DISCUSSION

314 Evaluating the Jd-in-clinopyroxene Barometer

315 Although regression statistics from calibration datasets provide important information about the precision
316 of thermobarometric models, it is important to verify model performance with independent datasets that
317 do not contain calibration data. Figure 3 shows the results of tests carried out using a global dataset of
318 clinopyroxene-liquid pairs from experiments conducted above 1 atm and below 20 kbar to avoid the
319 effects of Na loss in legacy datasets while remaining within the barometer's calibration range. This global
320 dataset does not include the data from experiments on H₂O-poor tholeiites used to produce Figure 1, and
321 is provided in the electronic appendix. Clinopyroxene-liquid pairs were also filtered on the basis of being
322 within 20% of $K_{d\text{Fe-Mg}}^{\text{cpx-liq}}$ equilibrium according to equation 35 from Putirka (2008). A total of 624
323 clinopyroxene-liquid pairs fitted these criteria (Bender et al. 1978; Johnston 1986; Kelemen et al. 1990;
324 Meen 1990; Bartels et al. 1991; Vander Auwera and Longhi 1994; Patiño-Douce and Beard 1996; Falloon
325 et al. 1999, 2001, Grove et al. 1997, 2003; Kinzler 1997; Falloon et al. 1997; Gaetani and Grove 1998;
326 Johnson 1998; Kogiso et al. 1998; Robinson et al. 1998; Takahashi et al. 1998; Vander Auwera et al.
327 1998; Draper and Green 1999; Pickering-Witter and Johnston 2000; Kogiso and Hirschmann 2001;
328 Müntener et al. 2001; Berndt et al. 2001; Schwab and Johnston 2001; Dann et al. 2001; Pichavant et al.
329 2002; Bulatov et al. 2002; Wasylenki et al. 2003; Elkins-Tanton and Grove 2003; Barclay 2004; Laporte
330 et al. 2004; Médard et al. 2004; Parman and Grove 2004; Kägi et al. 2005; Scoates et al. 2006; Di Carlo et
331 al. 2006; Ganino et al. 2013).

332

333 Figure 3a shows pressures calculated for the global dataset using imposed experimental temperatures.
334 Although simple linear fits to the global dataset are worse than similar fits to the calibration dataset ($r^2 =$
335 0.51 ; $SEE = 3.8$ kbar; intercept = 1.5 kbar), the mean residual between experimental and calculated
336 pressures is slightly smaller (-1.3 kbar). Barometer performance was marginally improved when
337 pressures were calculated iteratively (Figure 3b; $r^2 = 0.58$, $SEE = 3.6$ kbar; intercept = 0.8 kbar; mean
338 residual = -1.3 kbar). The performance of our new barometer is nonetheless comparable with the
339 performance of previous barometers subjected to testing against global datasets (Putirka 2008). However,
340 the usefulness of performing global tests is somewhat unclear given that many of the experiments in the
341 global dataset were performed on compositions either beyond the range of barometer calibration or of
342 little relevance to the investigation of magmatic processes in the crust. For example, low-pressure ($2-4$
343 kbar), high- fO_2 (NNO to NNO+2.3) experiments on phonolitic compositions return pressures ~ 10 kbar
344 higher than experimental pressures (Figure 3; Berndt et al. 2001). This discrepancy is perhaps
345 unsurprising given that there are no highly alkaline or highly oxidized compositions in the calibration
346 dataset, and may serve as an example of how pressures may be overestimated when Na partitions as Aeg
347 rather than Jd. We therefore carried out a more specific evaluation of barometer performance in basaltic
348 systems using the tholeiitic test dataset used to produce Figure 1. The experiments of Yang et al. (1996)
349 were, however, excluded because they were used in the barometer calibration. Calculations performed on
350 data from Villiger et al. (2004, 2007) and composition 70-002 from Grove et al. (1992) were also
351 excluded because of the large pressure ranges estimated for these experiments.

352

353 Figure 4a shows pressures calculated for test data with imposed experimental temperatures and Figure 4b
354 shows pressures for the same data calculated iteratively using equation 33 from Putirka (2008). Figure 4c
355 shows the iteratively calculated temperatures corresponding to Figure 4b. Pressure estimates for the test
356 dataset were determined with similar precisions using both imposed and iteratively calculated
357 temperatures ($SEE = 1.1$ and 1.2 kbar respectively). Encouragingly, the mean residuals between

358 experimental and calculated pressures are also small (0.6 and 0.8 kbar respectively). The barometer's
359 ability to reproduce the calibration dataset (SEE = 1.4 kbar; Figure 2b) is a conservative and thus more
360 appropriate estimate of barometer precision for tholeiitic systems.

361
362 Regressions through pressures from the test dataset do not pass through the origin as closely as
363 regressions through pressures from the calibration dataset do: intercepts = 1.1 and 1.2 kbar when using
364 imposed and iteratively calculated temperatures respectively. The significance of this inaccuracy is
365 unclear. While the specter of Na loss could still haunt the 1 atm data used here (Toplis and Carroll 1995;
366 Thy et al. 2006), it is important to note that the magnitude of pressure overestimation at 1 atm is
367 indistinguishable from the barometer's inherent uncertainty. This contrasts with the inaccuracies
368 identified in Figure 1, where the pressures of 1 atm experiments are overestimated by more than 2 SEE
369 (~3 kbar). Indeed, our new barometer returns systematically lower pressures for the test dataset than
370 model P1 from Putirka et al. (1996) (Figure 4d). However, improving the precision and accuracy of
371 thermobarometric expressions of the type we present here is challenging for two reasons. Firstly, both the
372 calibration and application of Jd-in-clinopyroxene barometers are strongly affected by the ability to
373 measure Na₂O in glasses and clinopyroxenes by electron probe precisely: Na₂O can be mobile under
374 electron beams and is often present at only 0.1–0.5 wt.% levels in clinopyroxenes. Secondly, barometry
375 using condensed phases (i.e., solids and liquids) is limited by the small absolute magnitudes of partial
376 molar volume changes associated with reactions that can be exploited as barometers, i.e., a precision of
377 1.4 kbar may be close to the limit of what can be achieved using current approaches (Putirka 2016).

378

379 **Magma Storage Pressures under Icelandic Rift Zones**

380 **Selecting Appropriate Clinopyroxene Analyses.** Magma storage pressures have been estimated using
381 the products of a number of tholeiitic eruptions from Iceland's neovolcanic rift zones using Jd-in-
382 clinopyroxene barometry (Maclennan et al. 2001; Winpenny and Maclennan 2011; Hartley and
383 Thordarson 2013; Neave et al. 2013, 2015; Geiger et al. 2016). However, most studies followed different

384 methodologies, making inter-eruption comparisons unreliable. For example, different studies used
385 different barometers and different methods of selecting equilibrium clinopyroxene-liquid pairs, and while
386 some studies applied empirical corrections to account for the systematic errors highlighted in Figure 1,
387 others did not. In order to investigate magma storage pressures of Icelandic tholeiites systematically, we
388 compiled clinopyroxene data from the highly primitive Borgarhraun lava ($Mg\#_{liq} \sim 69$, assuming $Fe^{3+}/\Sigma Fe$
389 $= 0.85$, Shorttle et al. 2015), the moderately primitive Thjórsá and Skuggafjöll flows ($Mg\#_{liq} \sim 50-54$) and
390 the evolved Laki flow and 10 ka Grímsvötn tephra series ($Mg\#_{liq} \sim 45$). We also included data from the
391 moderately primitive Holuhraun 1 and 2 eruptions ($Mg\#_{liq} \sim 50$) that took place at the same location as the
392 2014–2015 eruption in 797 and 1862–1864 (Hartley and Thordarson 2013). Data sources are listed in
393 Table 2.

394
395 Compiled Icelandic clinopyroxene compositions are summarized in Figure 5 using components calculated
396 according to the methods outlined in Table 3 from Putirka (2008). Individual eruptions form distinct
397 arrays in both DiHd-EnFs space and DiHd-CaTs space (Figures 5a and 5b). While some eruptions form
398 single arrays in $Ca/(Ca+Mg+Fe)$ -Al space, others form multiple arrays (Figure 5c). Eruptions also form
399 single arrays in $Ca/(Ca+Mg+Fe)$ -Jd space (Figure 5d). The large compositional ranges in Figure 5
400 represent not only the pressure and temperature conditions at which the crystals formed, but also
401 analytical errors, the occurrence of sector zoning and the diversity of primary melt compositions supplied
402 to Icelandic magmatic systems. To calculate storage pressures robustly, the effects of these three factors
403 on barometric calculations must be considered.

404
405 Analytical errors can strongly affect the results of thermobarometric calculations. We therefore applied
406 two filters to screen our dataset for poor analyses before performing any calculations. Firstly, we excluded
407 all clinopyroxene compositions that returned non-stoichiometric values when cation sums were calculated
408 on a six-oxygen basis (6O). Specifically, analyses with 6O sums of <3.99 or >4.02 were discarded.
409 Secondly, we excluded all analyses with $Jd < 0.01$, which represent Na_2O contents at or below the

410 detection limit of EPMA. The few analyses with Jd contents below 0.01 lie away from the bulk of the
411 data from their respective eruptions, confirming the poor quality of these analyses.

412

413 Sector zoning resulting from the variable partitioning of certain elements onto different crystal faces
414 during growth is extremely common in igneous clinopyroxenes (Strong 1969; Nakamura 1973; Downes
415 1974). The rate of crystal growth, itself closely related to the degree of undercooling (ΔT), exerts a first
416 order control on the style of sector zoning (Kouchi et al. 1983). For example, at high degrees of
417 undercooling ($\Delta T \sim 45$ °C), pressure-sensitive Na and Al partition onto $\{-111\}$ faces of clinopyroxene
418 crystal that record compositions strongly out of equilibrium (Welsch et al. 2016). In contrast, at low
419 degrees of undercooling ($\Delta T \sim 20$ °C), the sense of partitioning can be reversed, with Al partitioning
420 variably onto $\{100\}$, $\{010\}$ and $\{110\}$ faces depending on the exact conditions experienced.
421 Clinopyroxenes from tholeiitic systems often display strong sector zoning in their Ca/(Ca+Mg+Fe)
422 contents, which results from the disequilibrium partitioning of Mg and Fe onto $\{100\}$ faces causing
423 concurrent dilutions of Ca and Al (Nakamura 1973). Before performing thermobarometric calculations it
424 is thus essential to evaluate which sector zones were closest to being in equilibrium at the time of
425 crystallization (Hammer et al. 2016). For example, low-Al zones in high-Al clinopyroxenes from the
426 Haleakala ankaramite, Hawai'i, which crystallized at high degrees of undercooling, record the
427 compositions closest to being in equilibrium at the time of formation (Hammer et al. 2016; Welsch et al.
428 2016).

429

430 In the Icelandic clinopyroxene populations where it is present, sector zoning is clearly resolved in
431 Ca/(Ca+Mg+Fe)-Al space (Figure 5c): one set of sectors defines a high-Al field with high-
432 Ca/(Ca+Mg+Fe) while the other defines a low-Al field with variable-Ca/(Ca+Mg+Fe). This type of sector
433 zoning is common in clinopyroxenes formed at low degrees of undercooling in tholeiitic magmas
434 (Nakamura 1973). Following Nakamura (1973), we conclude that the low-Al sectors experienced Ca and
435 Al dilution during growth as a result of the disequilibrium incorporation of Mg and Fe, and that the high-

436 Al sectors preserve the compositions that were closest to equilibrium during their near-isothermal
437 crystallization (Neave et al. 2013). We therefore applied a third filter to our clinopyroxene dataset before
438 carrying out thermobarometry: all compositions with Al (6O) < 0.11 were excluded (Figure 5c).
439 Furthermore, almost all un-zoned clinopyroxenes from a series of recent equilibrium crystallization
440 experiments carried out on tholeiitic starting compositions at a range of pressures (1–7 kbar) have Al
441 (6O) > 0.11 (Husen et al. 2016), confirming that high-Al zones preserve the best record of clinopyroxene-
442 liquid equilibrium at the time of crystallization in the samples considered here. It is nonetheless important
443 to note that diffusive pile-up during crystallization at high degrees of undercooling can result in Al, Na
444 and Ti enrichments that compromise the identification of near-equilibrium zones (Mollo et al. 2013b).

445

446 **Performing Thermobarometric Calculations.** The selection of appropriate equilibrium liquids is a pre-
447 eminent consideration when calculating storage pressures by Jd-in-clinopyroxene barometry. In some
448 cases, textural relationships between crystals and their carrier liquids (i.e., matrix glasses) indicate that
449 these two phases were in equilibrium at the time of quenching and can be used for thermobarometric
450 calculations. However, many clinopyroxenes are concentrically zoned, reflecting growth in different
451 magmatic environments because of magma mixing and fractionation crystallization prior to eruption.
452 Indeed, many instances of crystal-liquid disequilibrium have been described in Icelandic tholeiites (e.g.,
453 Hansen and Grönvold 2000; Halldórsson et al. 2008; Thomson and MacLennan 2013; Neave et al. 2014),
454 demonstrating that equilibrium clinopyroxene-liquid pairs must be selected with care.

455

456 Kernel density estimates (KDEs) of $Mg\#_{\text{cpx}}$ values calculated with a bandwidth corresponding to the 1σ
457 precision of $Mg\#_{\text{cpx}}$ determination (± 0.5 mol.%) are shown in Figure 6a (Rudge 2008; Thomson and
458 MacLennan 2013). The compositions of clinopyroxene crystals predicted to be in equilibrium with the
459 average carrier liquid compositions from each eruption are shown as vertical bars in Figure 6a. These
460 compositions were calculated using $K_{\text{d Fe-Mg}}^{\text{cpx-liq}}$ values from equation 35 from Putirka (2008), liquid

461 $\text{Fe}^{3+}/\Sigma\text{Fe}$ values of 0.85, temperatures from equation 15 from Putirka (2008) and matrix glass
462 compositions collated from the following sources: Holuhraun 1 and 2, Hartley and Thordarson (2013);
463 Laki, Hartley et al. (2014); 10 ka Grímsvötn tephra series, Neave et al. (2015), Skuggafjöll, Neave et al.
464 (2014); and Borgarhraun Sigurdsson et al. (2000). No matrix glasses are known from the crystalline
465 Thjórsá lava, so the average composition of groundmass separates, which may have experienced olivine
466 and clinopyroxene accumulation, was used instead (Halldórsson et al. 2008). Hence, the equilibrium
467 $\text{Mg}\#_{\text{cpx}}$ value calculated for the Thjórsá carrier liquid was probably overestimated. Figure 6a reveals that
468 the majority of clinopyroxene compositions in our compilation crystallized from liquids significantly
469 more primitive than those which carried them to the surface, mirroring similar observations from olivine
470 and plagioclase populations (Hansen and Grönvold 2000; Thomson and MacLennan 2013). Therefore,
471 matrix glass compositions are not plausible equilibrium liquids for most of the analyses in our
472 compilation of Icelandic clinopyroxenes.

473

474 In order to increase the number of available clinopyroxene-liquid pairs and hence estimate the storage
475 pressures of primitive clinopyroxenes, we used an iterative equilibrium liquid-matching algorithm similar
476 to those used by Wimpenny and MacLennan (2011) and Neave et al. (2013, 2015). Putative equilibrium
477 liquids were selected from a large database of Icelandic whole-rock and matrix glass compositions
478 collated by Shorttle and MacLennan (2011), to which further additions have been made from more recent
479 studies (Jude-Eton et al. 2012; Koornneef et al. 2012; Hartley and Thordarson 2013; Hartley et al. 2014;
480 Neave et al. 2014, 2015; Streeter and Dugmore 2014). To avoid selecting geologically implausible alkali
481 basalt compositions, all data from flank zone eruptions were excluded. Furthermore, highly primitive
482 compositions ($\text{MgO} > 12$ wt.%) liable to be affected by olivine accumulation were also excluded from the
483 database.

484

485 In the first iteration of the liquid-matching algorithm, equilibrium melts were selected on the basis of
486 being within 20% of Fe-Mg equilibrium with each clinopyroxene analysis according to the temperature-

487 independent model of Wood and Blundy (1997). Initial guesses for equilibrium pressures and
488 temperatures were then calculated for every successfully matched clinopyroxene-liquid pair by iteratively
489 solving our new Jd-in-clinopyroxene barometer with equation 33 from Putirka (2008). Pressures and
490 temperatures were subsequently refined by performing several further iterations of this procedure with
491 more stringent criteria for selecting equilibrium melts (cf., Mollo et al. 2013a). Specifically,
492 clinopyroxene-liquid pairs were selected on the basis of being within 10% of Fe-Mg, DiHd component,
493 CaTs component and Ti equilibrium according to equation 35 from Putirka (2008), the DiHd and CaTs
494 equilibrium models from Putirka (1999) and the Ti partitioning model from Hill et al. (2011) respectively.
495 The inclusion of criteria for Ti and CaTs component equilibrium is important to account for the effects of
496 mantle-derived variability in liquid compositions (Shorttle and MacLennan 2011). Median pressures and
497 temperatures for each clinopyroxene analysis were calculated at the end of each iteration for use as
498 starting guesses in the next iteration.

499

500 **Interpreting the Results of Thermobarometric Calculations.** Out of the initial compilation of 1037
501 clinopyroxene analyses, a total of 603 were successfully matched to between 1 and 1167 glass and whole-
502 rock compositions (median = 184 matches). $Mg\#_{\text{cpx}}$ values to which equilibrium liquids were successfully
503 matched are summarized in Figure 6b. Although sufficient matches were made to estimate magma storage
504 pressures for each eruption, no matches were found for some ranges of clinopyroxene composition. For
505 example, no matches were found for the most primitive clinopyroxene analyses from the Borgarhraun
506 lava. In the cases of the Laki and Thjórsá lavas, and the 10 ka Grímsvötn tephra series, matched $Mg\#_{\text{cpx}}$
507 values have bimodal distributions. Given that this bimodality represents the crystallization of different
508 crystal populations under different conditions (Halldórsson et al. 2008; Neave et al. 2013, 2015), storage
509 pressure estimates for the evolved and primitive populations were considered independently. The
510 compositions of successfully matched clinopyroxene analyses are summarized in Figure 7 and form much
511 tighter clusters than the input data shown in Figure 5.

512

513 Iteratively calculated pressures and temperatures are summarized in Figure 8 and provided in the
514 electronic appendix. The full dataset of clinopyroxene-liquid pairs is available from the corresponding
515 author on request. As expected, there is an appreciable positive correlation between $Mg\#_{cpx}$ and
516 equilibrium temperature ($r^2 = 0.64$; Figure 8a). There is no strong relationship between $Mg\#_{cpx}$ and
517 equilibrium pressure, with the exception of high- $Mg\#$ clinopyroxenes from the Borgarhraun lava
518 returning higher pressures ($r^2 = 0.29$ with Borgarhraun, but 0.13 without; Figure 8b). Conversely, while
519 there is probably no relationship between clinopyroxene Al (6O) and temperature ($r^2 = 0.14$; Figure 8c),
520 there may be a very weak positive correlation between Al (6O) and pressure ($r^2 = 0.2$; Figure 8d).

521
522 In some cases, large ranges of pressures calculated for individual clinopyroxene populations have been
523 interpreted as evidence for vertically extensive magma storage regions (e.g., Budd et al. 2016; Giacomoni
524 et al. 2016). However, the certainty with which pressures can be calculated using clinopyroxene-liquid
525 barometers is limited (Figures 1–4). For example, the new Jd-in-clinopyroxene calibration presented here
526 has an SEE of 1.4 kbar, indicating that only 67% of the calibration data lie within 1.4 kbar of a regression
527 through the calibration dataset. In order to test whether the large pressure ranges observed in Figure 8
528 represent polybaric storage or merely the inherent uncertainty of the barometric calculations, data were
529 converted to KDEs that were in turn fitted with Gaussian functions (Figure 9; Rudge 2008). KDEs were
530 calculated with an imposed bandwidth of 1.4 kbar, which is equivalent to the estimated SEE of the
531 barometer calibration. Gaussian functions fitted to pressure KDEs calculated for each clinopyroxene
532 population identified in Figure 6b have standard deviations of 1.5–1.8 kbar that are only slightly larger
533 than the SEE of the barometer itself (Table 3). The SEEs of pressures calculated for each clinopyroxene
534 population, which may be better estimates of precision than standard deviations in such datasets (e.g.,
535 Putirka 2016), are also substantially smaller than the SEE of the barometer (0.1–0.4 kbar versus 1.4 kbar;
536 Table 3). Thus, the variability in the pressures calculated for individual clinopyroxene populations
537 feasibly reflects uncertainties in either the barometric model or the equilibrium liquid-matching procedure
538 rather than crystallization over a large pressure range. In other words, while different clinopyroxene

539 populations may have crystallized at different pressures, individual populations are likely to have
540 crystallized within a narrow pressure window of 0.1–0.4 kbar.

541
542 The means of Gaussian fits to the results of thermobarometric calculations represent our best magma
543 storage pressure estimates (Table 3). The highest storage pressures were calculated for the highly
544 primitive ($Mg\#_{\text{cpx}} > 85$) Borgarhraun lava ($4.8 \pm 1.8(1\sigma)$ kbar). However, these values are significantly
545 lower than previous estimates ($8.1 \pm 1.1(1\sigma)$ kbar; Maclennan et al. 2012; Winpenny and Maclennan 2011).
546 We identify two possible reasons for this discrepancy: firstly, we obtained few matches to the most
547 primitive clinopyroxenes ($Mg\#_{\text{cpx}} > 90$) that are most likely to have crystallized at high pressures; and
548 secondly, although the Putirka et al. (1996) barometer used by Winpenny and Maclennan (2011)
549 performed best at 8–10 kbar when tested with experimental data (Figure 1a), it appears to converge
550 towards high pressures when performing iterative calculations on natural data (Supplementary Figure 1).
551 Assuming an average Icelandic crustal density of $2.86 \text{ Mg}\cdot\text{m}^{-3}$ (Carlson and Herrick 1990), a storage
552 pressure of $4.8 \pm 1.8(1\sigma)$ kbar corresponds to lower crustal depths of $13.7 \pm 5.1(1\sigma)$ km (Moho depth ~ 20
553 km; Darbyshire et al. 2000).

554
555 Moderately primitive clinopyroxenes ($Mg\#_{\text{cpx}} \sim 80\text{--}86$) in the Laki, Thjórsá and Skuggafjöll lavas, and the
556 10 ka Grímsvötn tephra series from the EVZ returned very similar mean pressures of 3.5–3.7 kbar with
557 1σ uncertainties of 1.5–1.8 kbar. These values are broadly consistent with previous estimates of 2–5.4
558 kbar and 2.5–5.5 kbar for the Laki lava and 10 ka Grímsvötn tephra series calculated by applying
559 empirical corrections to published barometers (Neave et al. 2013, 2015). Pressures of 3.5–3.7 kbar
560 correspond to mid-crustal depths of $10.0\text{--}10.6 \pm 4.3\text{--}5.1(1\sigma)$ km (Moho depth $\sim 25\text{--}35$ km; Darbyshire et
561 al. 2000). More evolved clinopyroxenes ($Mg\#_{\text{cpx}} < 80$) from the same samples of the Laki and Thjórsá
562 lavas and the 10 ka Grímsvötn tephra series returned slightly lower pressures of $2.5\text{--}2.8 \pm 1.5\text{--}1.7(1\sigma)$
563 kbar, which corresponds to depths of $7.2\text{--}8.0 \pm 4.3\text{--}4.9(1\sigma)$ km. The ~ 1 kbar difference in mean storage
564 pressures between primitive and evolved clinopyroxene populations suggests that crystallization may

565 have occurred at different depths. Indeed, pressures of final equilibration based on OPAM barometry
566 suggest that the evolved assemblage crystallized at low pressures ($1\text{--}2\pm 1$ kbar), though a low-pressure
567 overprint could have been imposed during final ascent (Neave et al. 2013, 2015).

568
569 Moderately primitive clinopyroxenes ($\text{Mg}\#_{\text{cpx}} \sim 80$) from the Holuhraun 1 and 2 lavas returned the lowest
570 storage pressures of $2.4\pm 1.7(1\sigma)$ kbar, which are substantially lower than previously reported values (2.3--
571 7.6 kbar; Hartley and Thordarson 2013). The pressures we calculated from the Holuhraun 1 and 2 lavas
572 are also substantially lower than recent estimates from the petrologically analogous 2014–2015
573 Holuhraun lava (4.7 kbar; Geiger et al. 2016). These discrepancies probably reflect the inaccuracies in
574 previously published barometers (e.g., Supplementary Figure 1). Interestingly, a pressure of $2.4\pm 1.7(1\sigma)$
575 kbar translates to a depth of $6.9\pm 4.9(1\sigma)$ km, which corresponds closely to the depth of dyke
576 emplacement during the 2014–2015 eruption ($5\text{--}7$ km; Ágústsdóttir et al. 2015) as well as to geodetically
577 inferred magma storage depths under the Bárðarbunga caldera ($8\text{--}12$ km; Gudmundsson et al. 2016).

578

579

IMPLICATIONS

580

The Accuracy and Precision of Jd-in-clinopyroxene Barometry

581 Tests performed on a range of clinopyroxene-liquid barometers reveal that currently published models
582 routinely overestimate the pressure of experimental clinopyroxene-liquid pairs at crustal storage
583 conditions, i.e., ≤ 7 kbar. The accuracy of current Jd-in-clinopyroxene barometers is pressure dependent,
584 with overestimates being greatest at 1 atm. Although these tests were designed to investigate the accuracy
585 and precision of barometers at the conditions relevant to magma evolution under Iceland's rift zones (i.e.,
586 the storage of low- H_2O tholeiitic basalts), they demonstrate the importance of evaluating barometer
587 performance at the conditions of interest before interpreting any results (e.g., Masotta et al. 2013).
588 Therefore, numerous storage pressures calculated using clinopyroxene-liquid barometers may have been
589 overestimated by up to 3 kbar, and pressure estimates from MORB and OIB settings are likely to have
590 been affected with the greatest certainty. It may thus be necessary to reassess a number of published

591 magma storage pressure estimates, particularly in situations where they are used to inform monitoring
592 strategies in volcanically active regions.

593
594 Our new Jd-in-clinopyroxene barometer offers a significant improvement in accuracy over previous
595 versions of the barometer: calibration data are reproduced with a high degree of accuracy and testing
596 against a global dataset reveals a maximum overestimation of 1.5 kbar at 1 atm; testing against
597 experiments on H₂O-poor tholeiites reveals a lower maximum overestimation of 1.2 kbar at 1 atm.
598 Although our new calibration is a substantial improvement on its forebears, it should nevertheless be
599 tested further as experimental databases continue to expand. The new barometer's precision (SEE = 1.4
600 kbar) is probably close to the limit of what can be achieved with the Jd-liquid reaction. It is thus essential
601 to consider the inherent imprecision of Jd-in-clinopyroxene barometers when interpreting calculation
602 results. For example, pressure distributions calculated from multiple analyses that approximate Gaussian
603 bell curves with standard deviations of ~1.4 kbar may only reflect model uncertainties rather than
604 polybaric distributions of magma storage. New approaches, such as integrating numerous mineral-mineral
605 and mineral-liquid thermobarometric equations, as is commonly undertaken in thermobarometric studies
606 of metamorphic rocks (Powell and Holland 2008), will probably be necessary to improve the barometric
607 precision in magmatic systems.

608

609 **Magma Storage Pressures under Iceland's Neovolcanic Rift Zones**

610 Our calculations provide an internally consistent insight into the storage pressures of a range of Icelandic
611 rift zone tholeiites. In line with previous observations, high-Mg# clinopyroxenes from the Borgarhraun
612 lava record the highest pressures in our dataset ($4.8 \pm 1.8(1\sigma)$ kbar; $13.7 \pm 5.1(1\sigma)$ km), suggesting that near-
613 primary melts undergo substantial processing in the lower crust prior to eruption or intrusion to shallower
614 levels (Winpenny and MacLennan 2011). Indeed, patterns of microseismicity under the NVZ provide
615 strong evidence for the formation of intrusions in the lower crust (Hooper et al. 2011; Greenfield and
616 White 2015), where much of the initial compositional variability of primary mantle melts is probably

617 destroyed by mixing (Maclennan 2008). However, the discrepancy between the pressures calculated here
618 ($4.8 \pm 1.8(1\sigma)$ kbar) and those reported previously ($8.1 \pm 1.1(1\sigma)$ kbar) should be noted (Winpenny and
619 Maclennan 2011). Further experiments on primitive basalts should help to elucidate the causes of this
620 inconsistency, though an inability to identify equilibrium melts for highly primitive clinopyroxenes may
621 have also biased our calculations towards lower pressures. A storage pressure of $4.8 \pm 1.8(1\sigma)$ kbar thus
622 represents a robust minimum value for the Borgarhraun magma.

623
624 Most of the eruption products we considered show evidence for crystallization in the Icelandic mid-crust.
625 Those containing compositionally bimodal clinopyroxene populations suggest that crystallization may
626 have occurred at two pressure intervals, $3.5\text{--}3.7 \pm 1.5\text{--}1.8(1\sigma)$ kbar and $2.5\text{--}2.8 \pm 1.5\text{--}1.7(1\sigma)$ kbar.
627 However, given the large inherent uncertainties in Jd-in-clinopyroxene pressure estimates, it is currently
628 unclear whether these different pressures genuinely reflect polybaric crystallization. Nevertheless, the
629 conclusion that Icelandic rift zone tholeiites are primarily stored and processed in the mid-crust ($2.5\text{--}3.7$
630 kbar; $7.2\text{--}10.6$ km) is robust. While a depth range of $7.2\text{--}10.6$ km overlaps with some geodetic and
631 geophysical estimates of magma storage depths under Icelandic rift zones (Reverso et al. 2014;
632 Guðmundsson et al. 2016), many estimates from central volcanoes are significantly shallower ($2\text{--}4$ km;
633 Alfaro et al. 2007; de Zeeuw-van Dalssen et al. 2012; Hreinsdóttir et al. 2014). However, with the
634 possible exception of the 10 ka Grímsvötn tephra series, none of the volcanic products we considered
635 were erupted from central volcanoes, indicating that they probably bypassed shallow storage zones *en*
636 *route* to the surface, as occurs in Hawai'i (Poland 2015).

637
638 Although our calculations compress the pressure ranges reported by previous studies using the same
639 clinopyroxene analyses (e.g., $2.8\text{--}3.8$ kbar for Laki in contrast with $2.0\text{--}5.4$ kbar; Neave et al. 2013),
640 integrating results across multiple eruptions and clinopyroxene populations indicates that magma storage
641 occurs across a range of depths under Icelandic rift zones (at least $2.5\text{--}4.8$ kbar). Thus, accretion of the
642 Icelandic crust ostensibly proceeds in a similar manner to that outlined by stacked sill-type models from

643 mid-ocean ridges (Boudier et al. 1996; Kelemen et al. 1997; MacLennan et al. 2001).

644

645

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1027

1028 **Figure 1** Tests performed on published clinopyroxene-liquid barometers with data from experiments
1029 carried out on H₂O-poor tholeiites (dark red points). Test data were collated from the following sources:
1030 Berndt et al. (2005), Botcharnikov et al. (2008), Grove et al. (1992), Husen et al. (2016), Thy et al.
1031 (2006), Toplis and Carroll (1995), Villiger et al. (2004, 2007), Whitaker et al. (2007, 2008) and Yang et al.
1032 (1996). One-to-one lines between experimental and calculated pressures are shown in pale grey.
1033 Regression lines through the test dataset are shown in black. Data from Villiger et al. (2004, 2007) and
1034 composition 70-002 from Grove et al. (1992) that were excluded from the regressions used to summarize
1035 barometer performance are shown in grey. The following barometers were tested: (a) model P1 from
1036 Putirka et al. (1996), a Jd-in-clinopyroxene barometer; (b) equation 30 from Putirka (2008), a Jd-in-
1037 clinopyroxene barometer; (c) equation 32a from Putirka (2008), a clinopyroxene composition barometer;
1038 and (d) equation 32c from Putirka (2008), an Al partitioning barometer.

1039
1040 **Figure 2** Calibration of a new Jd-in-clinopyroxene barometer. The barometer's abilities to reproduce
1041 experimental pressures with (a) imposed and (b) iteratively calculated temperatures are shown, alongside
1042 (c) the ability of equation 33 from Putirka (2008) to reproduce experimental temperatures during iterative
1043 calculations. The new barometer is directly compared with model P1 from Putirka et al. (1996) in (d).
1044 Note that the new barometer faithfully reproduces the pressure of a wider range of 1 atm experiments than
1045 model P1 from Putirka et al. (1996).

1046
1047 **Figure 3** Tests performed on the new Jd-in-clinopyroxene barometer using a global dataset of
1048 clinopyroxene-saturated experiments performed at ≤ 20 kbar (Bender et al. 1978; Johnston 1986; Kelemen
1049 et al. 1990; Meen 1990; Bartels et al. 1991; Vander Auwera and Longhi 1994; Patiño-Douce and Beard
1050 1996; Falloon et al. 1999, 2001; Grove et al. 1997, 2003; Kinzler 1997; Falloon et al. 1997; Gaetani and
1051 Grove 1998; Johnson 1998; Kogiso et al. 1998; Robinson et al. 1998; Takahashi et al. 1998; Vander
1052 Auwera et al. 1998; Draper and Green 1999; Pickering-Witter and Johnston 2000; Kogiso and
1053 Hirschmann 2001; Müntener et al. 2001; Berndt et al. 2001; Schwab and Johnston 2001; Dann et al.

1054 2001; Pichavant et al. 2002; Bulatov et al. 2002; Wasylenki et al. 2003; Elkins-Tanton and Grove 2003;
1055 Barclay 2004; Laporte et al. 2004; Médard et al. 2004; Parman and Grove 2004; Kägi et al. 2005; Scoates
1056 et al. 2006; Di Carlo et al. 2006; Ganino et al. 2013). The barometer's abilities to reproduce experimental
1057 pressures with (a) imposed temperatures and (b) iteratively calculated temperatures are shown. High- f_{O_2}
1058 experiments on phonolitic compositions that return significantly overestimated pressures are marked
1059 (Berndt et al. 2001).

1060

1061 **Figure 4** Tests performed on the new Jd-in-clinopyroxene barometer using the same test dataset of
1062 experiments performed on H₂O-poor tholeiites plotted in Figure 1. The barometer's abilities to reproduce
1063 experimental pressures with (a) imposed temperatures and (b) iteratively calculated temperatures are
1064 shown, alongside (c) its ability to reproduce experimental temperatures with equation 33 from Putirka
1065 (2008) during iterative calculations. The performance of the new barometer is directly compared with that
1066 of model P1 from Putirka et al. (1996) in (d).

1067

1068 **Figure 5** A summary of the clinopyroxene compositions used to investigate magma storage pressures
1069 under Icelandic rift zones. Clinopyroxene compositions and components were calculated on a six oxygen
1070 (6O) basis following the methods outlined in Table 3 from Putirka (2008). Horizontal black bars on (c)
1071 and (d) show the lower limits of Al (6O) and Jd considered to represent equilibrium sector zone
1072 compositions and reliable analyses respectively. See the main text for further discussion. Unzoned
1073 experimental clinopyroxene compositions from Husen et al. (2016) are also shown to aid in the
1074 identification of equilibrium sector zone compositions. Low- and high-Al sector zone arrays are also
1075 marked on (c).

1076

1077 **Figure 6** Kernel density estimates (KDEs) of clinopyroxene Mg# distributions for (a) the full
1078 clinopyroxene compilation and (b) clinopyroxene compositions to which equilibrium liquid compositions
1079 were successfully matched. KDEs were calculated with a bandwidth comparable to the 1σ precision of

1080 Mg# determinations (± 0.5 mol.%). Vertical red lines show the Mg# of clinopyroxenes calculated to be in
1081 equilibrium with erupted liquids (see the main text for details). Only whole-rock analyses of groundmass
1082 separates, which are an inexact reflection of liquid compositions, were available for the Thjórsá lava
1083 (Halldórsson et al. 2008), so a tentative equilibrium clinopyroxene composition is marked with a vertical
1084 dashed line. Distinct clinopyroxene populations from the Laki and Thjórsá lavas, and the 10 ka Grímsvötn
1085 tephra series are marked with different colors in (b). KDEs of the full clinopyroxene compilation are also
1086 shown in pale grey behind the KDEs of matched clinopyroxenes. Matches were found for most
1087 clinopyroxene compositions, with the notable exception of the most primitive clinopyroxenes from
1088 Borgarhraun ($Mg\#_{\text{cpx}} > 90$), which grew from melts that were much more primitive than any known to
1089 have erupted in Iceland (Winpenny and MacLennan 2011)

1090

1091 **Figure 7** A summary of the clinopyroxene compositions to which equilibrium liquid compositions were
1092 successfully matched. Symbols as in Figure 5.

1093

1094 **Figure 8** A summary of calculated temperatures and pressures as functions of clinopyroxene Mg# ((a) and
1095 (b) respectively) and Al (6O) ((c) and (d) respectively). Regressions through the results are shown in
1096 black. The red regression in (b) excludes results from the Borgarhraun lava. Symbols as in Figure 5.

1097

1098 **Figure 9** KDEs of pressures and temperatures calculated for the clinopyroxene populations identified in
1099 Figure 6b. (a) Pressure KDEs calculated with a bandwidth of 1.4 kbar, which is comparable to the SEE of
1100 the new Jd-in-clinopyroxene barometer (Figure 2b). (b) Temperature KDEs calculated with a bandwidth
1101 of 17 °C, which is comparable with the SEE of equation 33 from Putirka (2008) when applied to
1102 experiments on low-H₂O tholeiites in conjunction with the new barometer (Figure 4c).

1103

1104 **Supplementary Figure 1** A comparison of pressures returned from iterative thermobarometric
1105 calculations performed on Icelandic clinopyroxene compositions using our new barometer and equation

1106 33 from Putirka (2008), and expressions from Putirka et al. (1996). A one-to-one line and a regression
1107 through the calculation results are shown in grey and black respectively.

1108

TABLES1109 **Table 1** A summary of the experimental data used to calibrate the new Jd-in-clinopyroxene barometer

Source	n	Pressure (kbar)	Temperature (°C)	H ₂ O (wt.%)
Blatter and Carmichael (2001)	13	0.548–2.28	930–1000	3–6
Kinzler and Grove (1992)	30	9–16	1250–1350	anhydrous
Moore and Carmichael (1998)	10	0.441–2.496	950–1075	3–5
Putirka et al. (1996)	23	8–20	1100–1390	anhydrous
Sisson and Grove (1993)	17	1–2	965–1082	3.8–6.2
Yang et al. (1996)	20	0.001	1110–1190	anhydrous

1110

1111 **Table 2** Icelandic clinopyroxene data sources

Eruption	Sources
Holuhraun 1 and 2	Hartley and Thordarson (2013)
Laki	Neave et al. (2013)
10 ka Grímsvötn tephra series	Neave et al. (2015)
Skuggafjöll	Neave et al. (2014)
Thjórsá	Passmore (2009)
Borgarhraun	Maclennan et al. (2001), Maclennan et al. (2003a) and Slater et al. (2001)

1112

1113 **Table 3** Barometric results and properties of Gaussian fits to pressures calculated for individual

1114 clinopyroxene populations

Clinopyroxene population	Mean	1 σ	1SEE	Mean _{Gaussian}	1 σ _{Gaussian}
Holuhraun 1 and 2	2.4	0.9	0.3	2.4	1.7
Laki, Mg# _{cpx} > 80	3.8	0.6	0.1	3.7	1.5
Laki, Mg# _{cpx} < 80	2.7	1.0	0.2	2.8	1.6
Grímsvötn tephra series, Mg# _{cpx} > 80	3.5	1.0	0.4	3.5	1.6
Grímsvötn tephra series, Mg# _{cpx} < 80	2.9	0.9	0.2	2.8	1.6
Skuggafjöll	3.6	1.3	0.1	3.6	1.8
Thjórsá, Mg# _{cpx} > 80	3.7	1.3	0.4	3.6	1.7
Thjórsá, Mg# _{cpx} < 80	2.6	1.2	0.3	2.5	1.8
Borgarhraun	4.8	1.4	0.1	4.8	1.8

1115

Figure 1

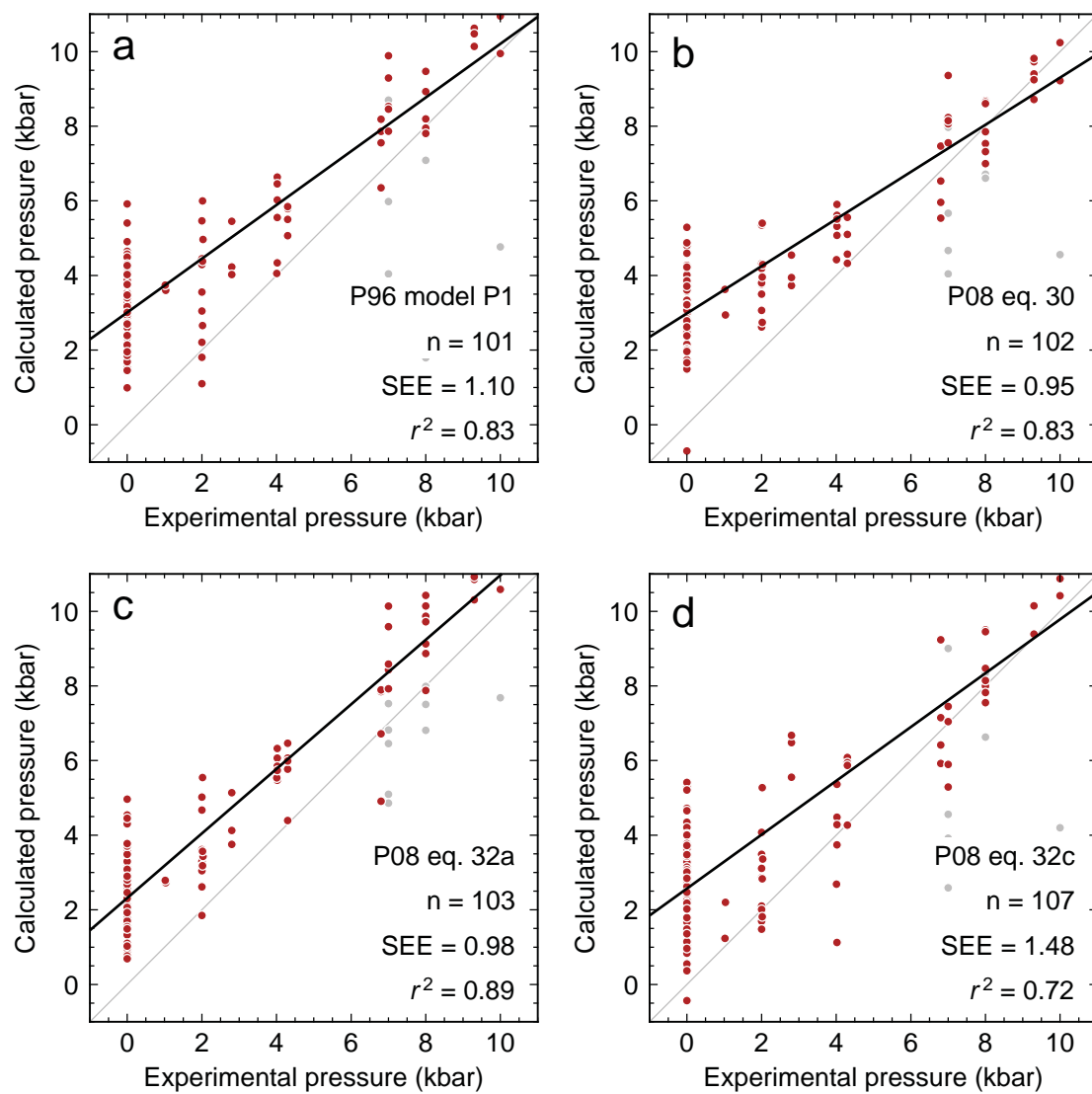


Figure 2

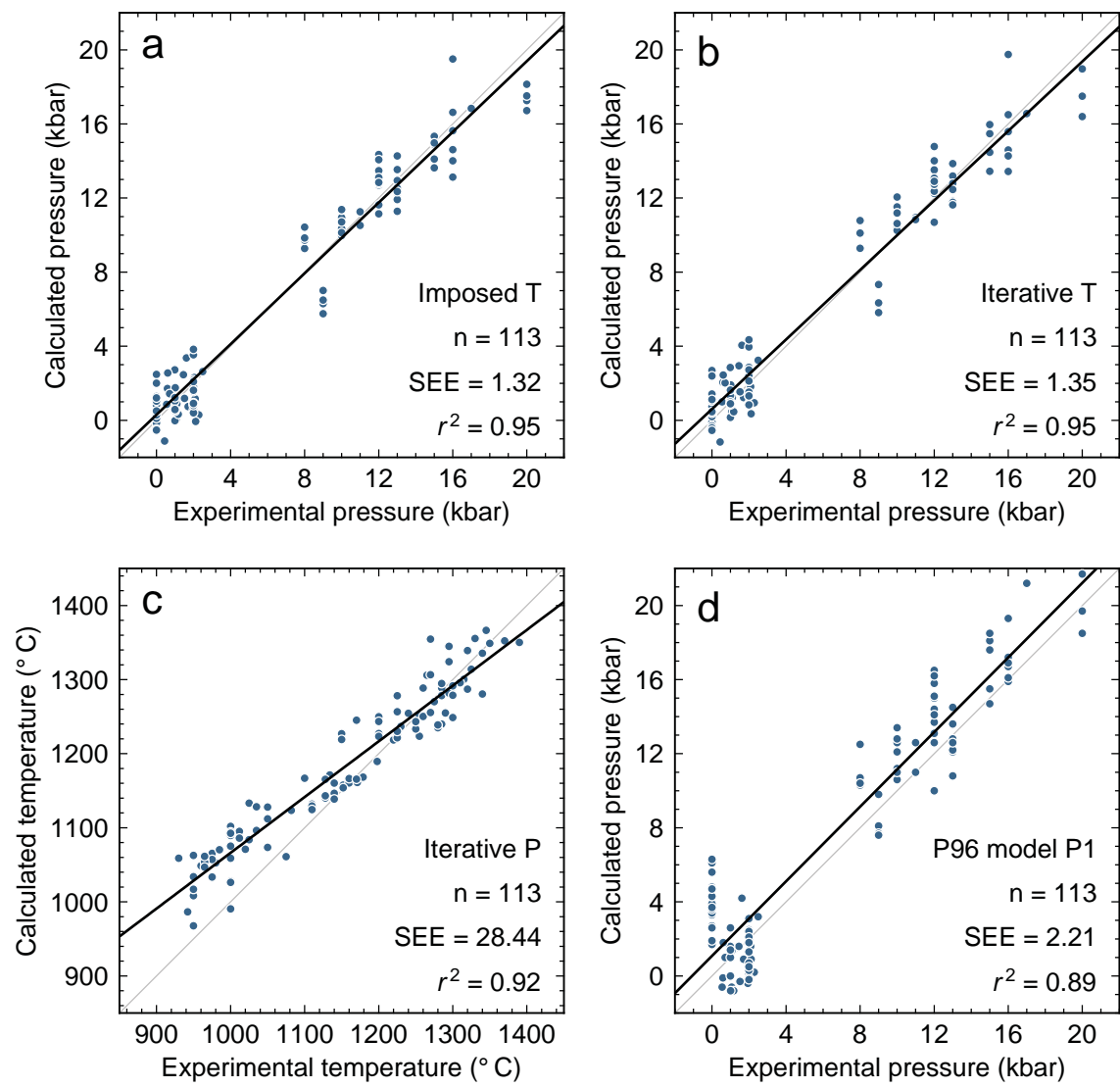


Figure 3

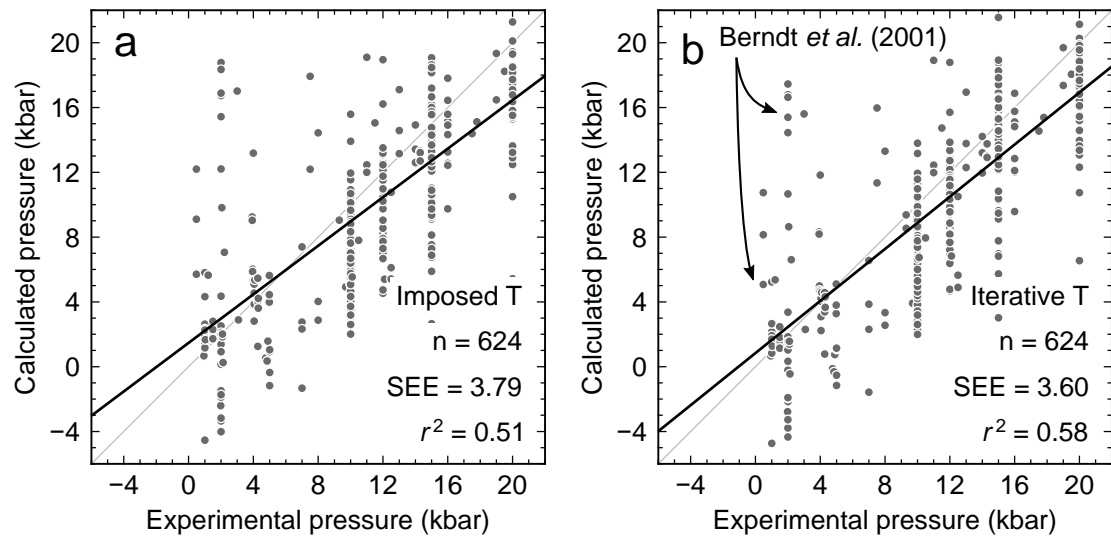


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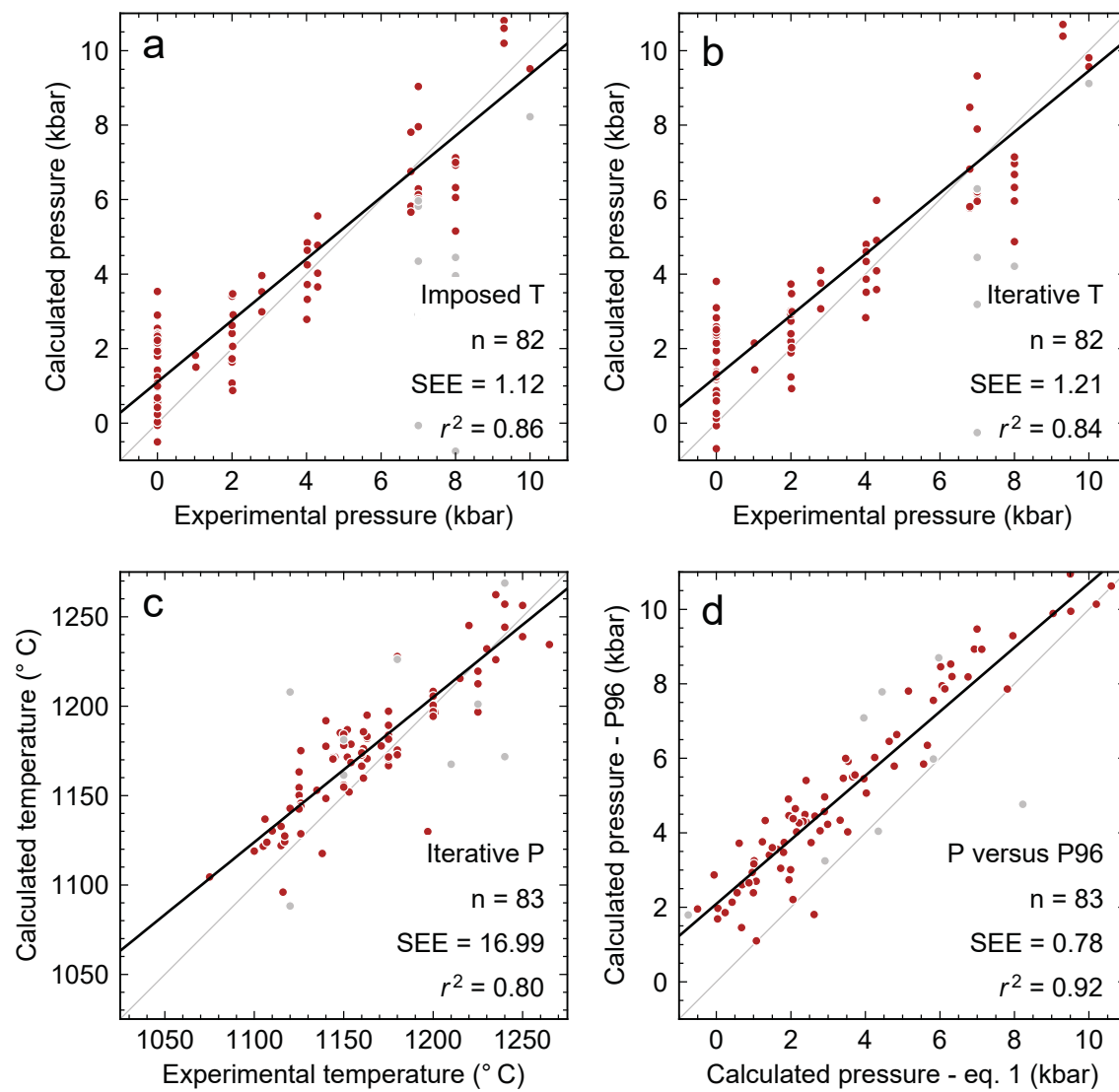


Figure 5

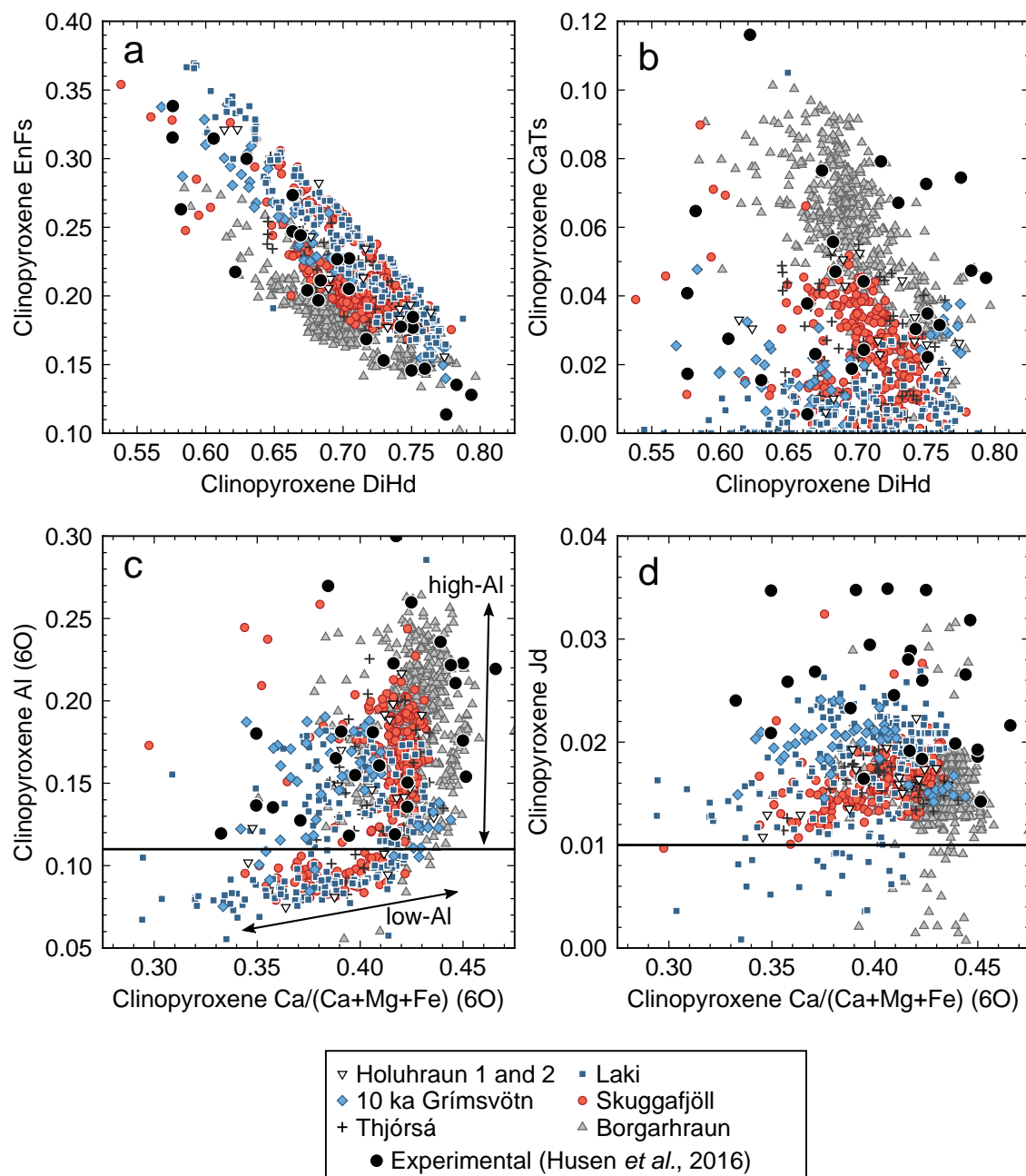


Figure 6

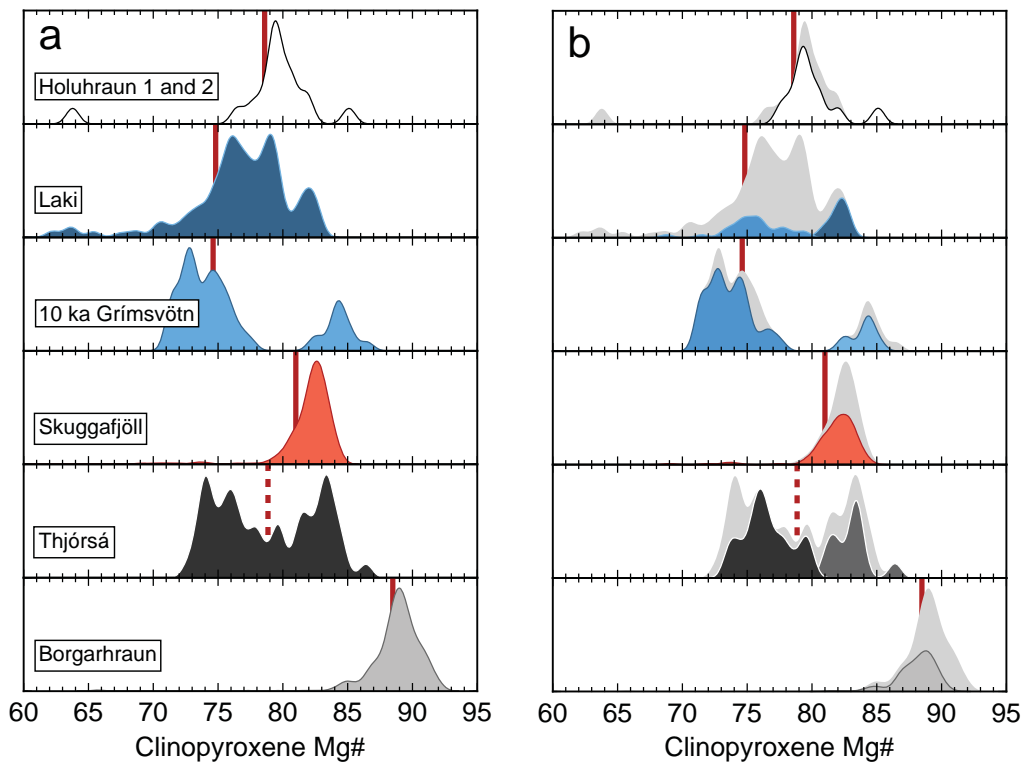


Figure 7

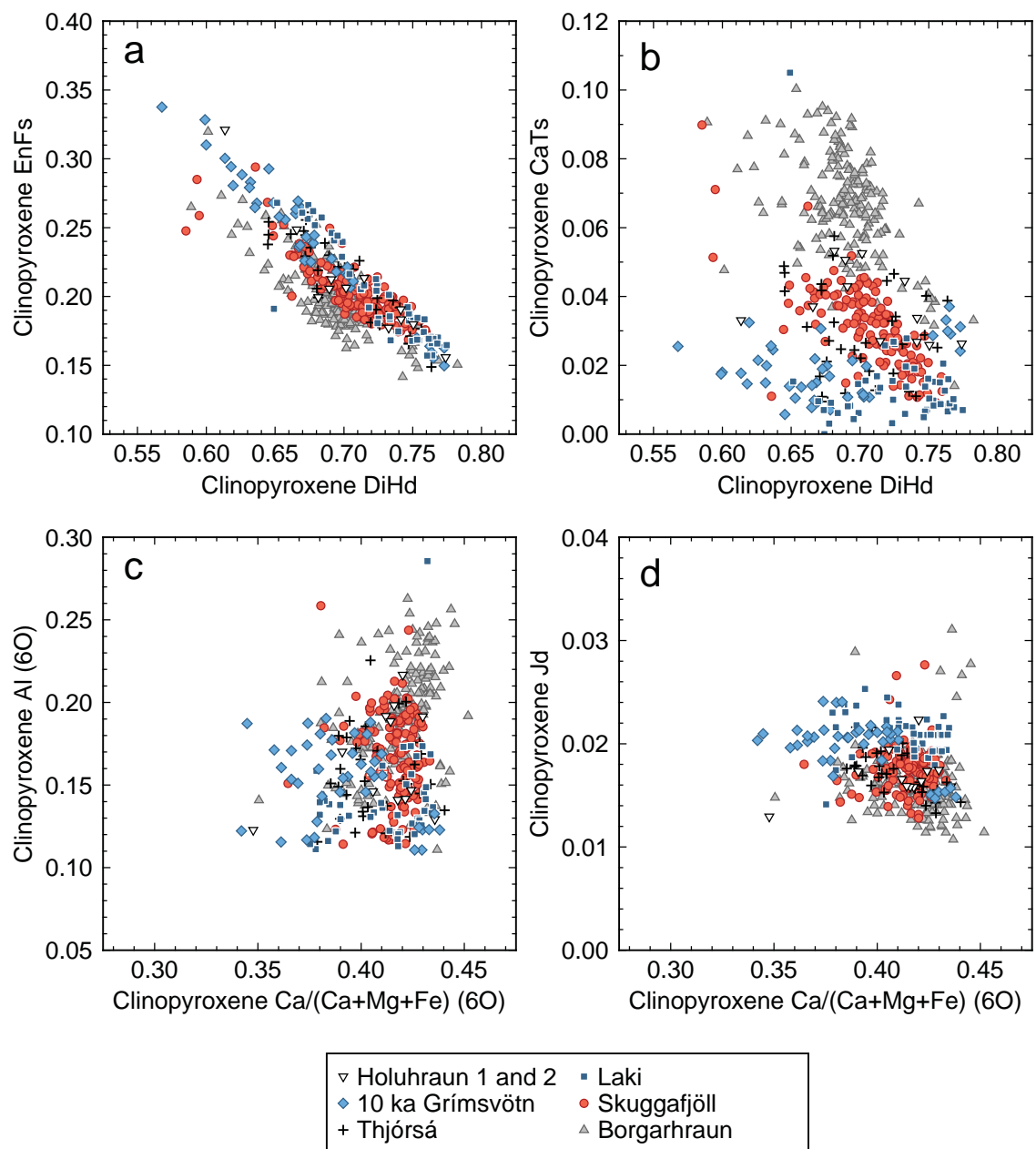


Figure 8

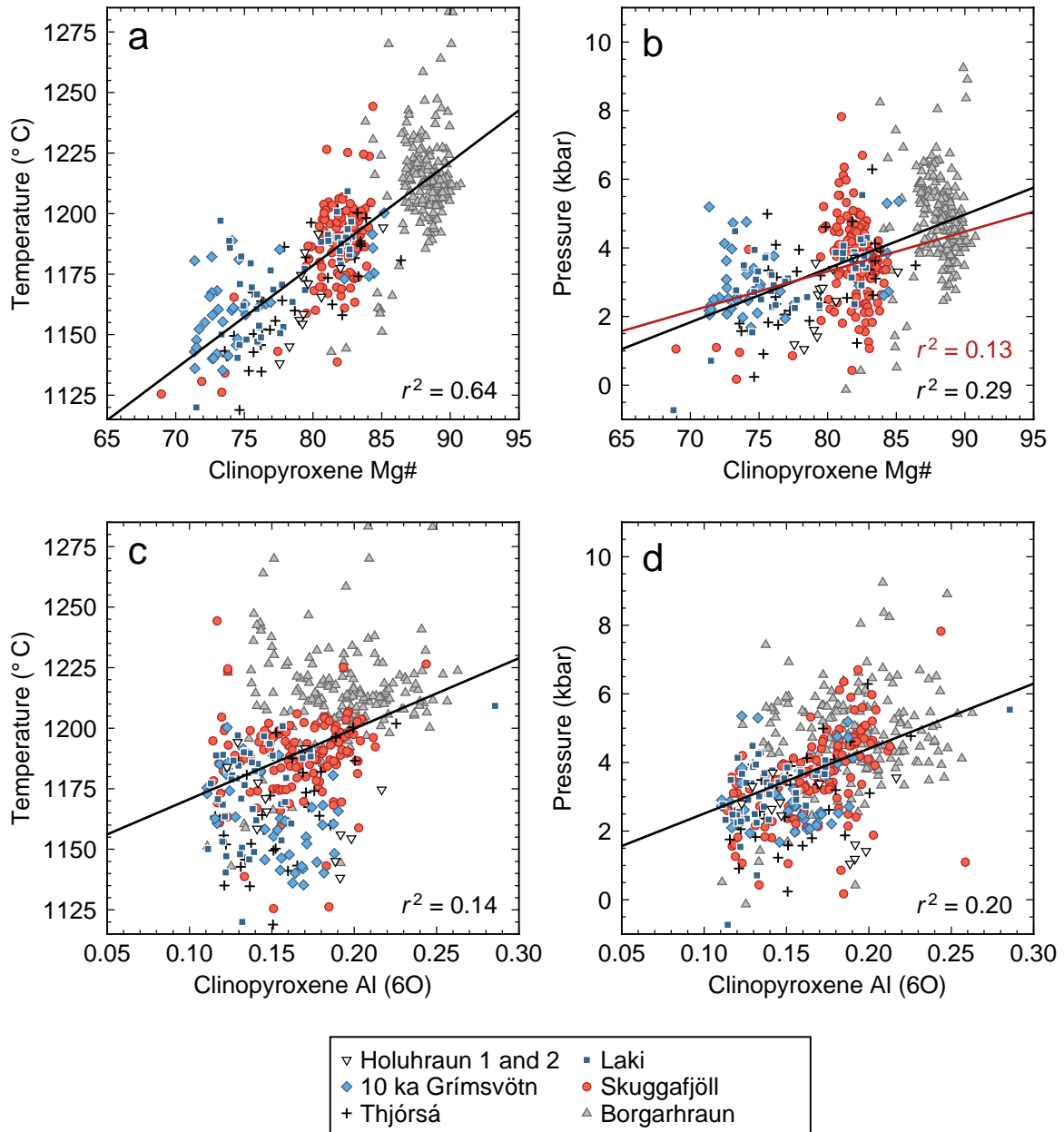


Figure 9

