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4 **Title**

5 The distribution and abundance of halogens in eclogites: an in situ SIMS perspective of the
6 Raspas Complex (Ecuador)

7 **Revision 2**

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Abstract

22 We present in situ secondary ion mass spectrometry (SIMS) and electron microprobe
23 analyses of co-existing garnet, omphacite, phengite, amphibole, and apatite, combined with

24 pyrohydrolysis bulk-rock analyses to constrain the distribution, abundance, and behavior of
25 halogens (F and Cl) in six MORB-like eclogites from the Raspas Complex (Southern Ecuador).
26 In all cases concerning lattice-hosted halogens, F compatibility decreases from apatite (1.47–3.25
27 wt %), to amphibole (563–4727 $\mu\text{g/g}$), phengite (610–1822 $\mu\text{g/g}$), omphacite (6.5–54.1 $\mu\text{g/g}$)
28 and garnet (1.7–8.9 $\mu\text{g/g}$). The relative compatibility of Cl in the assemblage is greatest for
29 apatite (192–515 $\mu\text{g/g}$), followed by amphibole (0.64–82.7 $\mu\text{g/g}$), phengite (1.2–2.1 $\mu\text{g/g}$),
30 omphacite (<0.05–1.0 $\mu\text{g/g}$) and garnet (<0.05 $\mu\text{g/g}$). Congruence between SIMS-reconstructed
31 F bulk abundances and yield-corrected bulk pyrohydrolysis analyses indicates that F is primarily
32 hosted within the crystal lattice of eclogitic minerals. However, SIMS-reconstructed Cl
33 abundances are a factor of five lower, on average, than pyrohydrolysis-derived bulk
34 concentrations. This discrepancy results from the contribution of fluid inclusions, which may
35 host at least 80% of the bulk rock Cl. The combination of SIMS and pyrohydrolysis is highly
36 complementary. Whereas SIMS is well suited to determine bulk F abundances, pyrohydrolysis
37 better quantifies bulk Cl concentrations, which include the contribution of fluid inclusion-hosted
38 Cl. Raspas eclogites contain 145–258 $\mu\text{g/g}$ F and at least 7–11 $\mu\text{g/g}$ Cl. We estimate that ~95% of
39 F is retained in the slab through eclogitization and returned to the upper mantle during
40 subduction, whereas at least 95% of subducted Cl is removed from the rock by the time the slab
41 equilibrates at eclogite facies conditions. Our calculations provide further evidence for the
42 fractionation of F from Cl during high-pressure metamorphism in subduction zones. Although
43 the HIMU mantle source (dehydrated oceanic crust) is often associated with enrichments in Cl/K
44 and F/Nd, Raspas eclogites show relatively low halogen ratios identical within uncertainty to
45 DMM. Thus, the observed halogen enrichments in HIMU ocean island basalts require either

46 further fractionation during mantle processing, or recycling of a halogen-enriched carrier
47 lithology such as serpentinite into the mantle.

48

49 Keywords: eclogite, halogens, subduction, SIMS, nominally anhydrous minerals, HIMU

50 **1. Introduction**

51

52 Our understanding of the abundance and distribution of halogens in subducted slabs is
53 limited. Hydrothermally altered oceanic crust (AOC) is thought to be a major halogen carrier
54 during subduction, where F and Cl substitute into the hydroxyl sites of hydrous minerals such as
55 amphibole and mica (Ito et al. 1983; Philippot et al. 1998; Van den Bleeken and Koga 2015;
56 Barnes et al. 2018). Bulk estimates of pre-subduction AOC vary from 50–253 $\mu\text{g/g}$ Cl and 216–
57 400 $\mu\text{g/g}$ F (Ito et al. 1983; Straub and Layne 2003; Barnes and Cisneros 2012; Van den Bleeken
58 and Koga 2015; Chavrit et al. 2016). More recently, primary halogen measurements of altered
59 oceanic crust from the East Pacific Rise (Penrose-type oceanic crust) and Atlantis Bank (SWIR,
60 ultra-slow spreading) have shown extreme variability with both stratigraphic depth and lithology
61 (Kendrick 2019a; 2019b). Bulk halogen measurements of blueschists and eclogites thought to
62 represent high-pressure metamorphic AOC are also sparse. In a study of mélangé rocks from
63 Syros, Greece, Marschall et al. (2009) measured the bulk Cl content of eclogites (28–60 $\mu\text{g/g}$)
64 and called into question previous estimates of eclogitized AOC Cl abundances, e.g. 100–200
65 $\mu\text{g/g}$ Cl of Philippot et al. (1998), speculating that they may be overestimated. Pagé et al. (2016)
66 analyzed a suite of blueschists from northwest Turkey; their results (8–22 $\mu\text{g/g}$ Cl) also indicate
67 that bulk eclogitized AOC could host less Cl than previously thought. Debret et al. (2016)
68 reconstructed bulk halogen concentrations (57–79 $\mu\text{g/g}$ Cl and 10–62 $\mu\text{g/g}$ F) from in situ

69 secondary ion mass spectrometry (SIMS) analyses for both blueschists and eclogites from the
70 Western Alps, cautioning that typical bulk halogen measurements are not representative of
71 prograde conditions due to potential retrogression during exhumation.

72 In situ halogen data in eclogitized AOC are also limited to a handful of studies, yet provide
73 important first order observations. Debret et al. (2016) suggested that approximately 50% of F
74 and 90% of Cl in the initial bulk-rock are lost to fluids during the first 80 km of subduction.
75 Hughes et al. (2018) analyzed in situ halogen abundances of eclogites from the Western and
76 Central Alps, and provided eclogitized AOC estimates of as low as 3 $\mu\text{g/g}$ Cl. Finally, Pagé et al.
77 (2016) reported low Cl and high F abundances in blueschist-hosted minerals, and concluded that
78 Cl is primarily expelled at shallow depths, whereas F is retained to at least 80 km depth.

79 Estimates of halogen fluxes in subduction zones are typically derived from the difference
80 between the trench inputs and extrusive products of arc magmatism (e.g. (Mather et al. 2006;
81 Sadofsky et al. 2008; Pyle and Mather 2009; Freundt et al. 2014). Straub and Layne (2003)
82 inferred a global arc recycling efficiency of 77–103% (Cl), and ~4–5% (F), implying near-total
83 return of Cl to surface reservoirs whereas most F is returned to the mantle. John et al. (2011)
84 reached similar conclusions based on Cl mass balance constraints regarding in- and out-fluxes
85 from the subducted slab perspective. Barnes et al. (2018) also concluded that F is efficiently
86 returned to the mantle, however their estimated Cl fluxes are more variable, indicating that some
87 Cl could be delivered back into the mantle by serpentinites (John et al. 2011; Kendrick et al.
88 2017). In a recent study, Pagé et al. (2018) found that Himalayan antigorite serpentinites host
89 significant quantities of F (50–650 $\mu\text{g/g}$) and are enriched in Cl (8.8–35 $\mu\text{g/g}$) with respect to
90 depleted mantle (11.0 $\mu\text{g/g}$ F and 0.51 $\mu\text{g/g}$ Cl, Salters and Stracke (2004)). Taken together,
91 these studies illustrate the challenge of estimating halogen input fluxes. A better understanding

92 of the inherent variability within different lithologies used in flux calculations could help to
93 refine such estimates.

94 In particular, the halogen content of eclogitized AOC remains largely unconstrained. Here
95 we discuss the F and Cl abundances of Raspas Complex eclogites, a Cretaceous eclogite suite
96 that we consider to be an ideal analog for subducted AOC. We combine in situ SIMS and EMP
97 analyses of minerals with pyrohydrolysis-derived bulk halogen abundances and discuss the
98 relative contribution of minerals, grain boundaries and fluid inclusions to the total budget of
99 halogens in subducted eclogites. Finally, we present new constraints on slab halogen fluxes and
100 discuss the implications for the signature of eclogitized AOC in the HIMU mantle source.

101

102

2. Geologic Context

103 An accreted parcel of oceanic lithosphere, the early Cretaceous Raspas Complex consists of
104 blueschist- and eclogite-facies rocks (Feininger 1980; 1987; Maruyama et al. 1996). The high-
105 pressure metamorphic suite is bound by the La Palma-El Guayabo Shear Zone to the north and
106 lower grade greenschists and amphibolites to the south (Gabriele 2002). While no pillow
107 structures or sheeted dikes were observed in the field, fluid-immobile REE and HFSE
108 abundances, Nb/Zr and Hf/Yb systematics, TiO₂/Yb ratios, and Nd isotope systematics provide a
109 well-reasoned rationale for inferred protoliths (John et al. 2010; Halama et al. 2011). Based on
110 these characteristics, the blueschists and eclogites were derived from seamount-like basalt and
111 normal mid-ocean ridge basalt (MORB) protoliths, respectively. Both underwent similar peak
112 metamorphic conditions (based on major element thermobarometry) of ~600°C to a maximum
113 burial depth of approximately 60 km (John et al. 2010). Raspas eclogites show evidence for low-
114 temperature seafloor alteration typical of basalts based on O isotopic compositions, and occur as

115 two geochemically distinct groups: a MORB-type group (this study) with LREE depleted
116 patterns, and a zoisite eclogite group characterized by significant incompatible trace-element
117 enrichments which are interpreted to have been derived from serpentinite-derived dehydration
118 fluids, and which also display a fluid component indicative of some minor sediment contribution
119 (Halama et al. 2011; Herms et al. 2012). Thus, the Raspas MORB-type group provides an
120 excellent analog to study the in situ distribution of halogens in eclogitized AOC processed
121 through subduction systems globally.

122 3. Sample Description

123 We chose a set of six minimally retrogressed MORB-type eclogites which have been
124 previously described by John et al. (2010). Samples consist of omphacite + garnet + rutile +
125 apatite ± amphibole ± phengite ± quartz with minor secondary titanite. Subhedral to euhedral
126 garnet porphyroblasts vary in size from <100 μm to 4mm, often hosting inclusions of rutile and
127 quartz. Some samples exhibit weakly deformed fabrics characterized by lineated omphacite. A
128 single sample, SEC42-06, shows a strongly foliated texture with alternating layers of nearly
129 monomineralic garnet and omphacite domains. Modal mineralogy varies significantly between
130 samples, from nearly bimineralic garnet + omphacite to ~30% modal amphibole; such variability
131 at similar P-T conditions is likely the result of bulk chemical compositional effects (Beinlich et
132 al. 2010; John et al. 2010; Rebay et al. 2010). Representative photomicrographs of sample
133 textures are presented in [Fig. 1 and S1](#). Petrographic observations reveal near-ubiquitous fluid
134 inclusions (FI). All samples contain variable quantities of FI hosted in omphacite and garnet,
135 typically located in mineral cores ([Fig. 1](#); see also Herms et al. (2012)). In SEC46-02,
136 monomineralic amphibole veinlets crosscut eclogite textures and contain secondary titanite
137 overgrowths rimming rutile. Directly adjacent to these veinlets, FI-rich omphacite appears

138 opaque in thin section (Fig. 1), which is unique to this sample and appears to reflect fluid
139 infiltration.

140

141

4. Methods

4.1 SIMS

143 SIMS analyses were conducted at the Northeast National Ion Microprobe Facility
144 (NENIMF) at the Woods Hole Oceanographic Institution using a Cameca IMS 1280. We follow
145 the methods outlined in Urann et al. (2017), adding OH and S measurements to the protocol.
146 150 μm polished thick-sections were used for all analyses. Samples were cleaned with ethanol
147 and deionized water to remove surface contamination, then placed in a vacuum oven to dry.
148 Samples were gold coated to ~ 160 nm thickness and placed into a vacuum chamber for storage
149 until analysis. Samples were loaded into the instrument sample chamber at least 12 hours prior to
150 analysis, to allow adequate pump down time and achieve chamber pressures of no more than
151 $\sim 5 \times 10^{-9}$ torr. Each spot location was chosen to avoid surface cracks, which are known to
152 influence analyses (Urann et al. 2017). A primary Cs^+ beam of 5.0–7.5 nA was sputtered through
153 the sample surface with a $30 \times 30 \mu\text{m}^2$ raster and a $400 \mu\text{m}$ field aperture, allowing only
154 transmission of ions from the innermost $5 \times 5 \mu\text{m}^2$ of the beam crater. Secondary magnet mass
155 calibration was done before each measurement with mass resolving power of > 6000 ($m/\Delta m$ at
156 10 % peak height). We measured $^{19}\text{F}/^{30}\text{Si}$, $^{16}\text{O}^1\text{H}/^{30}\text{Si}$, $^{32}\text{S}/^{30}\text{Si}$, and $^{35}\text{Cl}/^{30}\text{Si}$ ratios in glass
157 reference materials D51-3, D52-5, ALV519-4-1, 46D, 1649-3, 1654-3 6001, and AII107-D20 to
158 produce a calibration slope for each one-week analytical session (Fig. S2). Calibration slopes (m)
159 were obtained by plotting measured isotope ratios (x) against known reference material
160 concentrations (y) of the form $y = mx$ for each element of interest. Sample unknowns were then

161 calculated by multiplying measured ratios by m . Calibration slope uncertainties were assessed
162 utilizing a bootstrapping technique (5000 iterations) to derive confidence intervals.
163 Measurements where the internal precision (one standard deviation) was greater than 20% were
164 screened and excluded from the data set; these low precision measurements (less than ten in
165 total) were associated with elevated C (not reported) and Cl concentrations, and are likely the
166 result of analyses conducted on micro-fractures. After screening, analytical uncertainties over
167 five counting cycles (internal precision: typical standard error $\sim 1\%$ OH, $<1.5\%$ F, $<3\%$ Cl)
168 were combined with calibration slope uncertainties (accuracy: typical uncertainty 6% OH, 5% F,
169 $<10\%$ Cl, 95% confidence intervals) to yield no more than 8%, 6% and 16% total uncertainty
170 (2SE, 95% confidence intervals) for OH, F and Cl measurements, respectively. Since mineral-
171 specific calibrations are not well constrained and/or not available for halogens in most eclogitic
172 phases, we used a basaltic glass calibration to present a unified dataset that is self-consistent.
173 This may introduce crystal-specific matrix effects, although preliminary calibrations of F in
174 pyroxene and glass (Kumamoto et al. 2017) show slopes indistinguishable within uncertainty.
175 Continuous measurements of ALV519-4-1 were used throughout the session to monitor
176 instrument drift, which was found to be negligible. The OH, F, and Cl concentrations of Suprasil
177 and Herasil 102, both optical-quality glasses, along with Synthetic Forsterite, were measured
178 regularly in each session. There are no published values for the OH, F, and Cl content of
179 Suprasil, Herasil 102 and Synthetic Forsterite, but those samples are believed to have very low
180 OH, F, and Cl concentrations (less than $0.1\ \mu\text{g/g}$), respectively (E. Hauri, *pers. comm.*). To
181 quantify our maximum backgrounds, we assume that Suprasil contains no OH, Herasil contains
182 no F, and Synthetic Forsterite contains no Cl. Background OH, F, and Cl values were less than
183 $7.3\ \mu\text{g/g}$, $0.19\ \mu\text{g/g}$, and $0.15\ \mu\text{g/g}$, respectively, for all sessions. Individual mineral analyses

184 were not corrected for background when background values were less than propagated 2SE
185 uncertainty; otherwise, measurements are corrected to remove backgrounds (Table 1). Cl
186 measurement uncertainties on omphacite, garnet, and phengite are near parity with Synthetic
187 Forsterite Cl values. We therefore subtracted our background Cl values from measured values for
188 all bulk calculations.

189

190 **4.2 EPMA**

191 All mineral phases were analyzed using JEOL JXA-8200 electron microprobe (EMP) at
192 the Massachusetts Institute of Technology. A 1nA beam current and 15kV accelerating potential
193 were used for all analyses except apatite which utilized a 10 nA beam current. Beam diameter
194 was <1 micron for garnet and omphacite, and 10 μm for amphibole and phengite. Data reduction
195 was done using CITZAF software (Armstrong 1995). The counting times used for phase
196 analysis was 40s on peak, and 20s on background. With respect to halogen abundances in
197 amphibole and phengite, detection limits based on counting statistics were $\sim 470 \mu\text{g/g}$ for F and
198 $\sim 60 \mu\text{g/g}$ for Cl. For apatite, care was taken to address known issues pertaining to fluorine
199 excitation by electron microprobe analysis (Stormer and Pierson 1993). Peak searches were
200 conducted only on the standard, and background was only measured on the first point for each
201 grain. In addition, the LDE1 (W/Si) crystal was used for F measurements, which suppresses
202 elemental interferences. Counting times for F were abbreviated to 10s on peak and 40 s for P, Ca,
203 and Cl while background counts were 5s for F and 20s for Ca, P, and Cl. Analytical uncertainties
204 including background for F and Cl in apatite based on counting statistics were 3.4–4.4% and 7.2–
205 11.6% (1σ), respectively, at concentrations of 1.47–3.25 wt % F and 192–515 $\mu\text{g/g}$ Cl.

206

207 **4.3 Bulk halogen measurements**

208 Sample powders were washed in ultra-pure (18M Ω) deionized water and oven-dried
209 overnight to avoid surficial Cl contamination by adsorption. This may have removed a portion of
210 halogens along grain boundaries or in fluid inclusions; therefore we report Cl concentrations as
211 minimums. We note that fluid inclusion dimensions (\sim 1 μ m to 10 μ m, on average) are smaller
212 than estimated powder grain size (<100 μ m), and were likely retained, as demonstrated in a
213 subsequent section. Halogens were extracted from approximately 2 grams of sample powder by
214 pyrohydrolysis, in which a stream of water vapor captures volatiles as they are released from the
215 sample during fusion by a gas torch. The vapor is condensed to produce an aqueous solution
216 (Schnetger and Muramatsu 1996). From this solution, F and Cl concentrations are measured via
217 ion chromatography (IC) using a Dionex Integrion HPIC System at the University of Texas at
218 Austin. Detection limits of the analyzed solution are 0.05 μ g/g for both F and Cl. The halogen
219 content of the whole rock is then calculated using solution concentration, solution volume, and
220 the mass of the powder melted. Using the JB-2 basalt reference material (98.5 μ g/g F and 281
221 μ g/g Cl (Imai et al. 1995)), F and Cl yields are 75–83% and 68–95% (n = 5), respectively.
222 Samples were analyzed with triple replicates in two batches. Since JB-2 yields are constrained
223 within each contemporaneous batch, concentrations were yield-corrected by batch to the JB-2
224 reference material to more rigorously quantify absolute halogen concentrations. The yield
225 correction assumes that standard yields and sample yields are similar. Although not perfect, this
226 correction likely provides more accurate values. Both uncorrected and yield-corrected values are
227 presented in this study for comparison. Reproducibility for each sample (precision) was

228 combined with batch-specific JB-2 yield uncertainty (accuracy) to derive 1σ uncertainties.
229 Replicate measurements of JB-2 and uncertainties are given in Table S9.

230

231 **4.4 Modal mineralogy**

232 We calculated the modal abundances of each phase by inverting the bulk-rock major
233 element data of John et al. (2010) with EMP analyses of individual phases (Tables S1-S6).
234 Although EMPA indicates zonation in some phases (e.g. TiO_2 and MnO), changes in modal
235 proportions were negligible (often $<1\%$ difference) when varying mineral compositions were
236 used. We performed our calculations by taking the mean composition for each phase. SiO_2 ,
237 TiO_2 , Al_2O_3 , FeO , MnO , MgO , CaO , Na_2O , K_2O , and P_2O_5 were used in the minimization. P_2O_5
238 was assumed to reside entirely within apatite. Calculated modes agree well with independently
239 estimated mineral modes based on optical microscopy. Calculated mineral modes and
240 uncertainties are shown in Table 2.

241 All eclogite samples contain >35 modal % garnet. Omphacite modal proportions range
242 from 23–53 vol.%, whereas phengite, when present, makes up 2–4 vol% of the bulk-rock
243 modally. Samples contain varying modal proportions of amphibole, from 0.5 vol% to ~ 30 vol%.
244 Quartz, when present, is up to 8 vol% of the bulk rock whereas rutile (1–2 vol%) and apatite
245 (0.25–1.6 vol%) are omnipresent.

246

247

247 **5. Results**

248 **5.1 Major element variability by EMPA**

249 Major element profiles show garnet zonation with respect to end member components
250 almandine, pyrope, grossular, and spessartine (Table S1). Garnet porphyroblast cores are often

251 elevated in the spessartine component as well as TiO₂, with a gradual decrease towards the rim
252 consistent with prograde garnet growth.

253 Omphacite (Cpx) Mg# (molar Mg/(Mg+Fe)) varies from 0.68 to 0.79 (Table S2) and
254 correlates negatively with Cpx abundances, reflecting the influence of bulk-rock chemistry on
255 mineral modes. Omphacite EMPA profiles show some intra-mineral zonation; in particular,
256 SEC42-06 and SEC43-01 show increasing Si concentrations from core to rim, while SEC43-03
257 Si content decreases from core to rim. Mg# remain uniform within single grains.

258 Phengite was analyzed in samples SEC42-06, SEC43-01, and SEC47-01 (Table S5).
259 SEC42-06 is relatively homogeneous, with slight Na enrichment approaching the rim. A full rim-
260 to-rim profile in SEC43-01 shows strong zonation with increases in Na and Al from core to rim,
261 and decreases in Si, K, Fe and Mg (Fig. S3). SEC47-01 shows slight Si, K, and Mg rim
262 depletions. Both phengite profiles are consistent with continued grain growth or partial
263 recrystallization during decompression due to decreasing Si from core to rim (Massonne and
264 Schreyer 1987).

265 Amphibole occurs as several different species which likely reflect bulk compositional
266 variations; analyses are presented in Table S4, along with IMA 2012 recommended
267 nomenclature calculated using the routine of Locock (2014). SEC42-06, SEC43-01, and SEC43-
268 03 are Na-Ca subgroup katophorite, SEC46-01 and SEC46-02 are Ca subgroup pargasite, and
269 SEC47-01 is classified as a Na-Ca subgroup winchite. Nearly all EMP analyses show Cl
270 abundances at or below the detection limit of ~60 µg/g. However, F concentrations show
271 significant intra-grain variability, differing by as much as a factor of two from core to rim (e.g.
272 SEC42-06, 869–1834 µg/g).

273 Apatite halogen contents range from 1.47–3.25% wt% F and 0.019–0.051 wt% Cl (Table
274 S5). The hydroxyl content of the apatite are taken as the oxide sum subtracted from one hundred,
275 which classifies apatite in samples SEC43-03, SEC46-01, SEC46-02, and SEC47-01 as
276 hydroxylapatite and apatite in sample SEC43-01 and SEC42-06 as fluoroapatite. Apatite halogen
277 abundances are similar to apatite from other blueschist and eclogite metamorphic terranes with
278 low Cl abundances (Pagé et al. 2016; Hughes et al. 2018) whereas some HP terranes developed
279 from oceanic protoliths also contain Cl-rich apatite (John and Schenk 2003). Typical MORB
280 (0.164 wt% P₂O₅, Gale et al. (2013)) must undergo significant fractional crystallization (>70%)
281 before reaching apatite saturation (~0.7 wt% P₂O₅); indeed, apatite-saturated MORB are few
282 (Anderson and Greenland 1969; Watson 1979). Based on the bulk rock chemistry of Rapas
283 provided by John et al. (2010) these conditions are unlikely to have been met. Therefore
284 interstitial grains most likely formed as new growth during progressive metamorphism.

285

286 **5.2. Halogen and water variability**

287 Bulk rock pyrohydrolysis analyses are presented in Table 3. As noted previously, we
288 report both yield-corrected and uncorrected halogen abundances. Yield-corrected F and Cl
289 concentrations for all samples were 115–199 µg/g and 6.7–19.9 µg/g, respectively. In situ
290 halogen measurements are presented in Tables 1 and S5. Resolvable intra- and inter-grain
291 heterogeneity is observed for F and OH in all phases both in single-grain SIMS profiles (Table
292 S7) and in multiple grains from the same sample.

293 Halogen variability is notable in nominally anhydrous minerals (NAMs), particularly
294 garnet with both inter- and intra-grain heterogeneity observed (Figure S4, Tables 1 and S7).
295 Grain-averaged sample values range from 1.7 to 8.9 µg/g F and 60 to 201 µg/g OH, while Cl is

296 always at or near detection limits. Intra-sample garnet F and OH variability is up to 93% and
297 67%, respectively (1σ , Table 1). Variations from core to rim were not systematic, nor can they be
298 attributed to analytical uncertainty; in some cases cores were enriched in F or OH relative to
299 rims, while in other grains the case was reversed. Core and rim measurements for OH and F are
300 presented in Fig. 2, illustrating that while some intragrain variability is present, most grains plot
301 on or near 1:1 lines within uncertainty.

302 Omphacite has highly variable F abundances (6.5–54.1 $\mu\text{g/g}$) while OH range from 445
303 to 773 $\mu\text{g/g}$. Similar to garnet, Cl abundances are less than 0.1 $\mu\text{g/g}$ for five of the six samples.
304 However, SEC46-02 yields significantly higher mean Cl abundances of 1.05 $\mu\text{g/g}$. F and OH
305 variability within a single sample was less than 24% and 14%, respectively. A profile of Cpx
306 from SEC43-01 exemplifies core to rim variations, with F and OH concentrations of 42.1–69.7
307 $\mu\text{g/g}$ and 362–557 $\mu\text{g/g}$, respectively (Fig. S5). While these variations are discernable,
308 heterogeneity in this grain is 15% or less for all three species; core to rim variations are modest
309 and typically plot along a 1:1 line, within uncertainty (Fig. 2).

310 Amphibole occurs as both interstitial grains and monomineralic veinlets, with veins only
311 occurring in SEC46-02. Interstitial amphibole yields F and Cl abundances of 747–4727 $\mu\text{g/g}$ and
312 0.56–82.6 $\mu\text{g/g}$, respectively. Pargasite veins from SEC46-02 contain on average 563 $\mu\text{g/g}$ F and
313 11.7 $\mu\text{g/g}$ Cl. Single sample averaging produces variability of no more than 18% for F and 37%
314 for Cl (1σ).

315 Phengite from three samples contains 610–1822 $\mu\text{g/g}$ F and 1.01–1.98 $\mu\text{g/g}$ Cl. Two of
316 the three grains analyzed show no systematic core to rim halogen variations. Phengite is
317 relatively homogeneous in terms of F and Cl, with 1σ sample variations of less than 20%.
318 However, in a rim to rim transect of SEC43-01 phengite (Fig. S6), intra-grain variability is

319 evident. F concentrations show a systematic decrease from core to rim, paralleling major element
320 trends of decreasing K_2O , SiO_2 , and MgO . Cl is enriched near grain boundaries by
321 approximately 40% with respect to cores, following Al_2O_3 and Na_2O rim enrichments (Fig. S3
322 and S6). Finally, a single secondary titanite in SEC46-02 contains 1394 $\mu\text{g/g}$ F, <0.1 $\mu\text{g/g}$ Cl,
323 and 4010 $\mu\text{g/g}$ OH.

324

325

6. Discussion

6.1. Halogen redistribution during eclogitization

327 Halogen distribution during prograde metamorphism depends to a large extent on bulk-
328 rock chemistry, which dictates the equilibrium mineral assemblage and phase proportions at any
329 given pressure and temperature. Raspas eclogites show a strong positive correlation between the
330 proportion of omphacite in the bulk-rock and the F content of omphacite (Fig. S7), suggesting
331 that F is repartitioned during eclogitization and progressive omphacite growth. In addition, we
332 compare the modal proportion of nominally anhydrous minerals (omphacite + garnet + quartz) to
333 the volatile content of analyzed phases (Fig. S8). As the proportion of NAMs in the bulk
334 assemblage increases, the F content of each phase shows a commensurate increase. Indeed,
335 modal abundances of amphibole are negatively correlated with F concentrations in both
336 omphacite and apatite, again suggesting effective redistribution of F during prograde
337 metamorphism. Further, apatite Cl abundances increase as amphibole modes decrease and the
338 mineral assemblage becomes dominated by garnet and omphacite. In all cases concerning lattice-
339 hosted halogens, F is accommodated primarily in apatite (1.47–3.25 wt %), amphibole (563–
340 4727 $\mu\text{g/g}$), and phengite (610–1822 $\mu\text{g/g}$) with lesser amounts in omphacite (6.5–54.1 $\mu\text{g/g}$) and
341 garnet (1.7–8.9 $\mu\text{g/g}$). In addition, Cl is primarily hosted in apatite (192–515 $\mu\text{g/g}$), with lesser

342 amounts in amphibole (0.64–82.7 $\mu\text{g/g}$), phengite (1.2–2.1 $\mu\text{g/g}$), omphacite (<0.05–1 $\mu\text{g/g}$) and
343 garnet (<0.05 $\mu\text{g/g}$).

344 Our in situ measurements permit the calculation of distribution coefficients between
345 various phases of interest (Figs. 3 and 4, Table S8). Given the scarcity of data, these new values
346 provide important constraints on the distribution of F and Cl in eclogitic phases. A number of
347 observations can be made from the above distribution coefficients. First, F partitioning between
348 mineral pairs defines a narrow range and appears to be insensitive to concentrations, which in
349 some cases span nearly an order of magnitude. Second, as expected apatite will preferentially
350 host crystal lattice-bound F and Cl when present in eclogitic assemblages, similar to apatite's
351 halogen affinity in the presence of aqueous fluids (Kusebauch et al. 2015). Finally, F and Cl are
352 preferentially partitioned into amphibole over phengite in all samples by a factor of 2.4 and 20,
353 respectively. Interestingly, this observation is in contrast to blueschists studied by Pagé et al.
354 (2016) who found $D_F^{Amph-Phen}$ was always less than unity whereas $D_{Cl}^{Amph-Phen}$ was
355 approximately equal to one within error. Hence, blueschist to eclogite phase transformations may
356 elicit changes in the relative compatibilities of halogens between coexisting phases, likely as a
357 function of changing modal proportions, mineral compositions (e.g. glaucophane to katophorite),
358 and P-T conditions.

359

360 **6.2. Comparison between measured and reconstructed bulk halogen abundances**

361 Using mineral modes, in situ SIMS data, and EPMA halogen concentrations, we calculate
362 a reconstructed bulk-rock F and Cl content for each eclogite sample, which can then be
363 compared to bulk values measured by pyrohydrolysis (Table 3, Fig. 5). Both raw (not corrected

364 for yield) and yield-corrected pyrohydrolysis halogen abundances are given in Table 3. We use
365 yield-corrected concentrations in subsequent discussions.

366 Reconstructed concentrations and pyrohydrolysis measurements yield F concentrations of
367 168–415 $\mu\text{g/g}$ and 115–199 $\mu\text{g/g}$, respectively. Reconstructed F abundances are broadly
368 concordant with measured values; three of the six samples agree within uncertainty, whereas
369 three samples display higher reconstructed values (Fig. 6; Table 3). The relative congruence
370 between reconstructed and measured F values implies that F may be entirely hosted within the
371 crystal lattice of eclogitic phase assemblages. The discrepancy observed for three of the six
372 samples may be due to a combination of factors including uncertainties in pyrohydrolysis yields
373 (Table S9) that are typically larger than SIMS analyses, inter-grain halogen heterogeneity, or
374 uncertainties in the calculated modal proportions (Table 2). We note that calculated modal
375 proportions as derived from whole rock analysis were assessed, but found to be minor.

376 Bulk reconstructed and measured Cl concentrations for all samples are 0.8–8.2 $\mu\text{g/g}$ and
377 6.7–19.9 $\mu\text{g/g}$, respectively. Unlike F, Cl shows very large and systematic discrepancies between
378 reconstructed and measured values, with reconstructed values representing only 12–41% of the
379 pyrohydrolysis-derived bulk values. We discuss these drastic differences below, in terms of
380 uncertainties and possible means of reconciliation between the two methods. We consider three
381 possible processes to explain the difference: retrograde alteration, grain boundary hosted Cl, and
382 Cl-bearing fluid inclusions.

383 Bulk halogen analyses rely on the assumption that samples are perfectly preserved and
384 have not been retrogressed along their exhumation path. Even minute amounts of alteration (e.g.
385 chlorite or titanite) can drastically affect such bulk measurements (Debret et al. 2016). Most
386 Raspas eclogites appear to be pristine with little to no visible alteration or retrogression, with the

387 exception of SEC46-02 that contains titanite-on-rutile overgrowths and monomineralic
388 amphibole veinlets adjacent to FI-rich omphacite, indicating possible fluid assimilation. This
389 sample also displays elevated reconstructed (8.2 $\mu\text{g/g}$) and bulk (19.9 $\mu\text{g/g}$) Cl compared to the
390 rest of the suite. In light of this suspected fluid assimilation, and the large F incongruity ($\sim 100\%$)
391 between the two methods, we exclude this sample from subsequent bulk rock calculations. Minor
392 retrogression may have contributed to the discrepancy for SEC46-02, however all other samples
393 still show at least a factor of three difference between reconstructed and measured bulk Cl
394 concentrations. Therefore, a factor other than late alteration is likely at play.

395 To further explore this discrepancy, we conducted additional SIMS analyses on grain
396 boundaries of FI-free amphibole and garnet (Fig. S9). Both profiles across grain boundaries
397 display pronounced spikes in $^{35}\text{Cl}/^{30}\text{Si}$ signals within the immediate grain boundary at near-
398 constant ^{30}Si counts. These grain boundary measurements are equivalent to 7–18 $\mu\text{g/g}$ Cl. Yet,
399 the grain boundaries themselves are far smaller than the dimensions of our analysis spot (3.8x3.8
400 μm) and depending on their size, could contain > 1 wt% Cl assuming a grain boundary thickness
401 of 5 nm or less. As grain boundary porosity values in eclogites are poorly constrained, we used
402 equation 9.6 of Turcotte and Schubert (2002) to calculate grain boundary porosity values based
403 on a simplified grain geometry over a range of grain boundary thicknesses (1–25 nm) with
404 realistic grain sizes (500 μm) to yield values of 9×10^{-12} to 6×10^{-9} (Table S10). The Cl
405 contribution from grain boundaries alone (< 0.1 $\mu\text{g/g}$ Cl in all cases, or $< 2\%$ contribution
406 compared to pyrohydrolysis Cl abundances), given the above parameters, is unable to reconcile
407 the reconstructed versus measured Cl abundances.

408 Finally, we consider the possible role of fluid inclusions, as they could represent an
409 important host for Cl (Philippot and Selverstone 1991; Herms et al. 2012; Kendrick 2018). FI

410 (~1 μm to 10 μm on average) are found ubiquitously hosted in garnet (Fig. S1C) and omphacite
411 cores, both of which appear in textural equilibrium with adjacent phases and occur in all
412 samples. FI are far more abundant in SEC46-02, in particular where omphacite is in contact with
413 monomineralic paragonitic amphibole veinlets (Fig. 1B). FI appear cloudy in thin section owing to
414 their prolific abundance. These features likely reflect crystallization in the presence of fluids,
415 possibly during prograde conditions (e.g. John et al (2010) and Herms et al. (2012)); such
416 features have also been noted elsewhere in previous studies (e.g. Massonne et al. (2012), Zack et
417 al. (2001)). Although we cannot directly analyze fluid inclusions by SIMS, heterogeneous Cl
418 enrichments in SEC46-02 FI-rich omphacite (44–94 $\mu\text{g/g}$ Cl) indicate that each SIMS analysis
419 may incorporate some Cl-rich FI material. FI-free (optically clear) omphacite from the same
420 sample also contains an order of magnitude more Cl (1.05 $\mu\text{g/g}$) compared to omphacite from all
421 other samples (<0.1 $\mu\text{g/g}$), which is consistent with assimilation of pervasive fluids during
422 crystallization (Herms et al. 2012). FI may host significant quantities of Cl from hundreds of
423 $\mu\text{g/g}$ to >10 wt% NaCl equivalent (Philippot and Selverstone 1991; Philippot et al. 1998;
424 Scambelluri and Philippot 2001; Svensen et al. 2001; Kendrick et al. 2015; Kendrick 2018).
425 Herms et al. (2012) have shown that omphacite and garnet hosted FIs from Raspas contain ~2
426 wt% NaCl, congruent with the salinity of dehydration-related FI found in rocks that have
427 undergone blueschist-to-eclogite conversion (Gao and Klemd 2001). Assuming that FI contain
428 ~2 wt% NaCl and represent 0.05–0.1% of the total sample volume, one may reconcile the
429 discrepancy between in situ and bulk Cl abundances. Thus, pyrohydrolysis appears to
430 incorporate fluid inclusion contributions, whereas SIMS analyses do not. Our calculations imply
431 that FI host the majority (at least 80%) of Cl in Raspas eclogites; whether these fluid inclusions

432 are all primary in nature, and therefore should be considered in the halogen budget of recycled
433 eclogites, remains to be determined.

434 Combining SIMS and pyrohydrolysis techniques affords unique insights on the
435 distribution and abundance of halogens in subducted slabs. SIMS provides detailed constraints
436 on crystal lattice hosted halogen abundances, and is well suited to determine bulk F
437 concentrations, given the uncertainty that can be associated with pyrohydrolysis yields.
438 Minimally retrogressed Raspas eclogites analyzed by SIMS contain between 145 and 258 $\mu\text{g/g}$
439 F; F may be entirely hosted within the crystal lattice of eclogites, whereas only $\sim 20\%$ of Cl (at
440 most), on average, is crystallographically hosted when FI are present. However, SIMS cannot
441 accurately account for Cl abundances if primary fluid inclusions are present in the sample, which
442 dominate the Cl budget of eclogites. Assuming that fluid inclusions observed here are primary
443 features, pyrohydrolysis is the preferred method to measure the bulk Cl content of pristine FI-
444 bearing rocks; minimally retrogressed Raspas eclogites analyzed by pyrohydrolysis contain
445 between 6.7 and 11.3 $\mu\text{g/g}$ Cl.

446

447

7. Implications

448 Previous studies have suggested that amphibole is the primary carrier of halogens to the
449 upper mantle (Debret et al. 2016; Barnes et al. 2018). Our results suggest a more nuanced view
450 when one takes into account the contributions from coexisting phases and FI; amphibole's
451 contribution to mineral-hosted bulk F and Cl abundances range from 6–84% and 4–85%,
452 respectively (Fig. 6, S7). When amphibole modes are below $\sim 5\%$ (three of six samples), apatite
453 is the most important carrier for mineral-hosted halogens, containing $>80\%$ of mineral-hosted Cl
454 and 38–71% of mineral-hosted F. Apatite is thought to be stable to 7.5 GPa at 950° (Konzett and

455 Frost 2009), and will act as an important halogen carrier in eclogitized ocean crust. The
456 remaining F is distributed between amphibole (4–53%), phengite (6–32%), omphacite (4–16%),
457 and garnet (up to 2.2%). The above calculations consider only crystal matrix hosted halogen
458 abundances; our results further indicate that FI host the majority of Cl in the bulk rock (at least
459 80%, on average). The ultimate fate of Cl will be dictated by the redistribution of free fluid
460 phases during subsequent phase transformations, diffusive re-equilibration, and dynamic
461 recrystallization at higher pressures and temperatures.

462 Raspas reconstructed bulk F concentrations (145–258 $\mu\text{g/g}$) are similar to previous pre-
463 subduction AOC estimates of 216 $\mu\text{g/g}$ (Straub and Layne 2003), blueschist ($457 \pm 159 \mu\text{g/g}$ F)
464 and eclogite-facies ($283 \pm 146 \mu\text{g/g}$ F) AOC (Pagé et al. 2016; Hughes et al. 2018), as well as
465 mean N–MORB values of $183 \pm 107 \mu\text{g/g}$ calculated from recent work from Le Voyer et al.
466 (2018) (Fig. 7A). Therefore, F concentrations remain relatively unchanged during the
467 progression from nascent MORB, to AOC, and finally eclogitized AOC. Our results indicate that
468 F is efficiently returned (~95%, derived from SIMS) to the upper mantle during subduction
469 (Straub and Layne 2003; John et al. 2011; Pagé et al. 2016). Conversely, bulk pyrohydrolysis Cl
470 measurements (6.7–11.3 $\mu\text{g/g}$) are at least an order of magnitude lower than pre-subduction AOC
471 estimates (e.g. 207 $\mu\text{g/g}$ (Barnes and Cisneros 2012)). Yet, observed Raspas concentrations are
472 quite similar to recent blueschist and eclogite-facies AOC estimates by Hughes et al. (2018) of
473 3–23 $\mu\text{g/g}$ Cl and Pagé et al. (2018) of 12 $\mu\text{g/g}$ Cl, and well within the range observed in MORB
474 (excluding Cl/K >0.1 after Shimizu and Saal (2016) and references therein) (Fig. 7A).
475 Progression from initial MORB generation to seafloor alteration leads to an increase in Cl
476 content, while subsequent eclogitization leads to a decrease in Cl abundances, returning to values
477 similar to MORB. Using the pre-subduction AOC Cl bulk-rock estimate of Barnes and Cisneros

478 (2012) of 207 $\mu\text{g/g}$ Cl and data presented here, we calculate that the vast majority (>95%,
479 derived from bulk pyrohydrolysis) of subducted Cl is expelled from the slab by the time the
480 AOC parcel equilibrates at eclogite-facies conditions (Table 3), in agreement with previous
481 workers (Straub and Layne 2003; John et al. 2011; Kendrick et al. 2014; Pagé et al. 2016). What
482 little Cl remains will reside primarily in FI (at least 80%, on average). Thus, while most F is
483 retained in the subducting slab at eclogite-facies conditions, most Cl previously added through
484 hydrothermal alteration is expelled, fractionating F from Cl in eclogitized AOC. Yet, halogen
485 abundances in Raspas are similar to unaltered MORB (Fig. 7A), while ratios of F and Cl with
486 elements of similar compatibility during melting, Cl/K (0.0066 ± 0.0043), Cl/Nb (3.9 ± 1.0) and
487 F/Nd (15.5 ± 4.8), are indistinguishable within uncertainty from DMM ratios of Salters and
488 Stracke (2004) (Fig. 7B).

489 The HIMU (high U/Pb) mantle source is thought to contain ancient altered oceanic crust
490 based on Pb isotope systematics (and more recently Tl, see Shu et al. (2019)), yielding
491 enrichments in F/Nd, $^{35}\text{Cl}/^{37}\text{Cl}$, and often Cl/K and Cl/Nb ratios (Chase 1981; Hofmann and
492 White 1982; Zindler et al. 1982; Stroncik and Haase 2004; John et al. 2011; Cabral et al. 2014;
493 Jackson et al. 2015; Le Voyer et al. 2015; Kendrick et al. 2017; Rose-Koga et al. 2017; Hanyu et
494 al. 2019). Our data indicate that halogen enrichments, and elevated F/Nd and Cl/K ratios, in the
495 HIMU source cannot be produced by simple addition of recycled eclogitized AOC (Fig. 7B), but
496 rather require delivery by an additional lithology, e.g. serpentinite as proposed by Kendrick et al.
497 (2017), or an efficient fractionation mechanism to elevate halogen ratios during subsequent
498 processing (e.g. Niu and O'Hara (2003)). The broad spectrum of F/Nd and Cl/K ratios observed
499 in HIMU melts may then reflect variable contributions from subducted serpentinite (green line,
500 Fig. 7B). However, antigorite serpentinite breakdown by the reaction: antigorite = olivine +

501 orthopyroxene + H₂O requires geochemical transfer of halogens, likely into a fluid phase as both
502 F and Cl are highly incompatible in olivine and orthopyroxene (Dalou et al. 2014; Joachim et al.
503 2015; Beyer et al. 2016); their ultimate host within the upper mantle remains unknown. Direct
504 phase transformation from antigorite serpentinite to the dense hydrous magnesium silicate Phase
505 A may occur along cold geothermal gradients (Schmidt and Poli 1998), or halogens may become
506 more compatible in K-rich clinopyroxene at increasing pressures as K solubility in clinopyroxene
507 increases to ~1 wt. % K₂O at 100 kbar (Schmidt and Poli 1998). Thus, subduction proceeds in
508 returning to the upper mantle an eclogitized AOC parcel with halogen ratios quite similar to the
509 MORB from which it was derived.

510

511 **Figures**

512 See attached folder

513 **Figure Captions**

514 Figure 1. Representative photomicrographs of Raspas eclogite textures. A. SEC43-03 contains
515 garnet porphyroblasts within a matrix of omphacite and lesser quartz. B. Amphibole veinlet in
516 sample SEC46-02. Adjacent omphacite shows abundant fluid inclusions (yellow arrow). C.
517 SEC43-03 Megacrystic, inclusion-rich garnet showing sector zoning.

518

519 Figure 2. Core and rim measurements for OH (A. and B.) and F (C. and D.) in garnet and
520 omphacite. The majority of samples show core and rim values that fall on a 1:1 line, within
521 uncertainty, allowing a high degree of confidence in bulk rock calculations based on in situ
522 measurements. Error bars are 2SE.

523

524 Figure 3. Inter-mineral F partitioning between A. omphacite and garnet, B. omphacite and
525 apatite, C. omphacite and amphibole, and D. omphacite and phengite. White triangles are
526 literature data from Debret et al. (2016). Solid lines show constant inter-mineral partition
527 coefficients (labeled), while mean partition coefficients for each mineral pair are presented in the
528 bottom right panel corner.

529

530 Figure 4. Inter-mineral Cl partitioning between A. phengite and apatite, B. apatite and
531 amphibole, C. phengite and amphibole, and D. apatite Cl and garnet F concentrations. White
532 triangles are literature data from Debret (2016), green diamonds from Hughes et al. (2018) and

533 blue upside down triangles from Pagé et al. (2016). Solid lines denote constant inter-mineral
534 partition coefficients as in Fig. 3.

535

536 Figure 5. Bulk reconstructed and bulk measured (yield corrected) halogen abundances for F (A.)
537 and Cl (B.). Y error bars show analytical reproducibility propagated with yield precision. Yield-
538 uncorrected pyrohydrolysis values are shown in panels C and D, where positive Y error bars
539 incorporate yield uncertainty. Red dots denote SEC46-02, which was not included in bulk
540 calculations; see text for more information. Solid lines represent 1:1 lines; dashed lines plot
541 various slopes, labeled accordingly.

542

543 Figure 6. Pie diagrams show modal abundances (A. and B.), as well as the percent contribution
544 from each phase to mineral-hosted halogen abundances as derived from SIMS analyses for F (C.
545 and D.) and Cl (E. and F.) in two representative, yet mineralogically distinct, samples.

546

547 Figure 7. A. F and Cl abundances from this study in comparison to literature values for MORB
548 glasses from Le Voyer et al. (2018), HIMU OIB (Cabral et al. 2014; Jackson et al. 2015;
549 Kendrick et al. 2017), blueschist and eclogite (Debret et al. 2016; Pagé et al. 2016; Hughes et al.
550 2018), antigorite serpentinite (Pagé et al. 2018), and AOC estimates (Straub and Layne 2003;
551 Van den Bleeken and Koga 2015; Barnes et al. 2018; Kendrick 2019a). MORB with Cl/K values
552 in excess of 0.1 were screened out, due to possible seawater contamination after Shimizu and
553 Saal (2016) and references therein. Peridotite mantle plots off the scale and would be 1.4–31
554 $\mu\text{g/g}$ for F and from 0.14 to 0.38 $\mu\text{g/g}$ for Cl (Urann et al. 2017). B. F/Nd and Cl/K ratios of
555 Raspas samples (Nd and K from John et al. (2010)), MORB glasses from Le Voyer (2018), mean
556 antigorite serpentinite halogen concentrations from Page et al. (2018) with DMM K (60 $\mu\text{g/g}$)
557 and Nd (0.713 $\mu\text{g/g}$) values of Salters and Stracke (2004), and OIB MIs and glasses from the
558 literature (Cabral et al. 2014; Jackson et al. 2015; Kendrick et al. 2017). We note that our
559 eclogitized AOC samples indeed plot below the canonical F/Nd of 21, as posited by Rose-Koga
560 et al. (2012) for deeply subducted oceanic crust.

561 Figure S1. Representative full-section photomicrographs of Raspas samples analyzed in this
562 study. A. Prograde assemblage in SEC43-01 contains garnet porphyroblasts within a matrix of
563 omphacite and lesser quartz and phengite with minor apatite and rutile. The sample shows no
564 evidence for retrogression. B. SEC43-03 megacrystic garnet showing sector zoning with minor
565 quartz. C. Amphibole veinlet in sample SEC46-02, outlined in white dashed line. Adjacent
566 omphacite shows abundant fluid inclusions (dark patches).

567

568 Figure S2. Representative calibration slope used in this study. Calibration slope confidence
569 intervals were calculated with a non-linear bootstrapping technique using 5,000 iterations.

570 Figure S3. SEC43-01 phengite EMP profile from rim to rim shows Na_2O and Al_2O_3 increases
571 from core to rim, while K_2O , SiO_2 , and MgO all decrease toward the grain boundary.

572

573 Figure S4. SEC43-01 garnet core to rim transect. Zero values represent rim, 750 μm spot
574 location is the garnet core. Overall, OH and F show intragrain variability likely related to
575 partitioning and re-equilibration during garnet growth. The combination of internal and
576 propagated calibration slope uncertainty corresponds to the positive error bar on our analyses.

577 The negative error bars are larger for Cl because they incorporate the conservative uncertainty on
578 the maximum background.

579

580 Figure S5. SEC43-01 omphacite core (~1300 μm) to rim (zero μm) profile for OH, F, and Cl.
581 Significant variations occur on length scales of <100 μm , with apparent rim depletions in F and
582 OH.

583

584 Figure S6. SEC43-01 phengite rim-to-rim transect. F shows depletions toward the grain
585 boundaries, while Cl shows rim enrichments of approximately 40%.

586

587 Figure S7. Fluorine concentrations (black y-axis, yellow dots) and contributions to the bulk-rock
588 (blue y-axis, blue dots) plotted against modal proportion of each phase for A. omphacite, B.
589 amphibole, C. apatite, and D. phengite.

590

591 Figure S8. Fluorine and chlorine concentrations in each phase as a function of the modal
592 proportion of nominally anhydrous minerals (omphacite plus garnet). As the modal proportion of
593 NAMs increases, so does the halogen concentration in most phases.

594

595 Figure S9. Grain boundary transects using SIMS across amphibole (A.) and garnet (B.). While
596 there are likely unaccounted for matrix effects with this approach, both measurements on the
597 grain boundary qualitatively show pronounced spikes in Cl/Si ratios at near-constant constant Si
598 counts, indicating Cl enrichment within the grain boundary (C. and D.). Also shown are raw ^{30}Si
599 counts for comparison (panels E and F).

600

601

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607

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Figure 1.

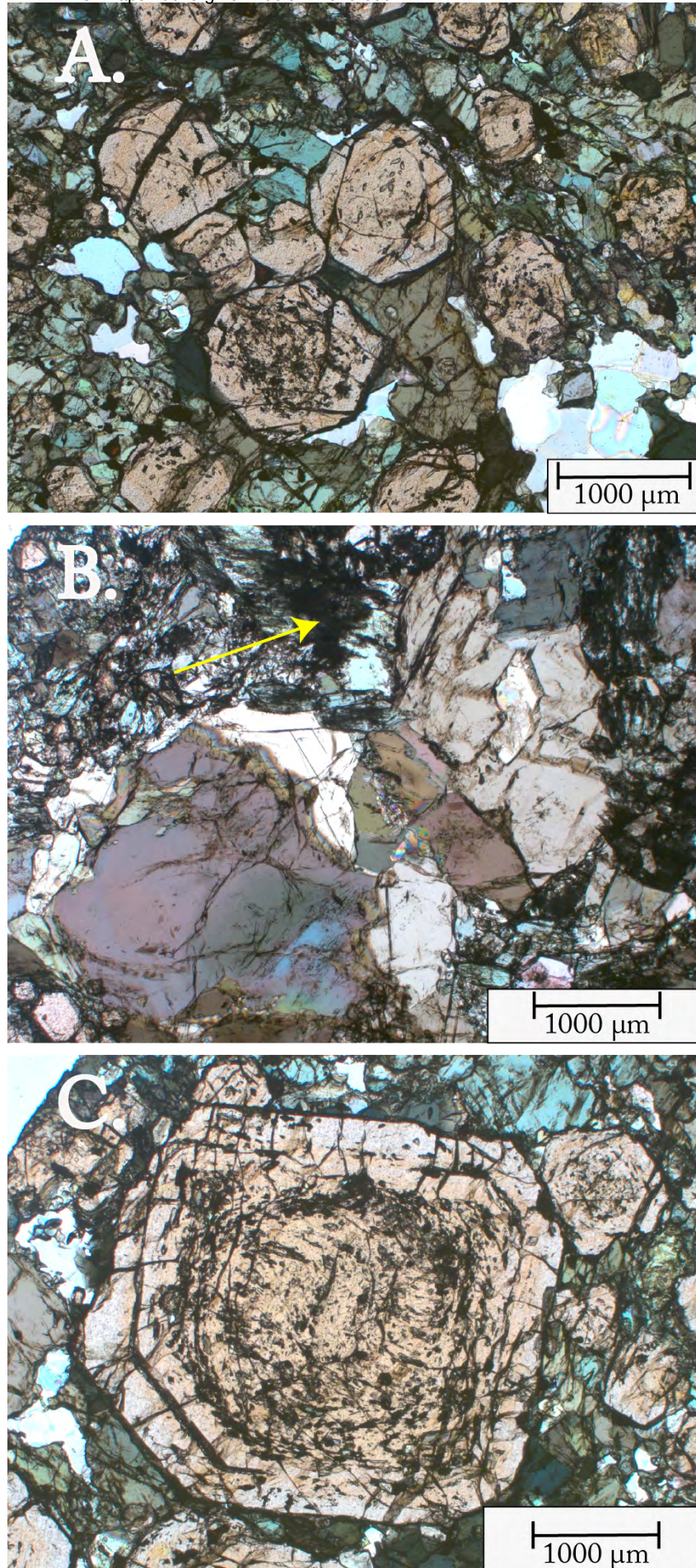


Figure 2.

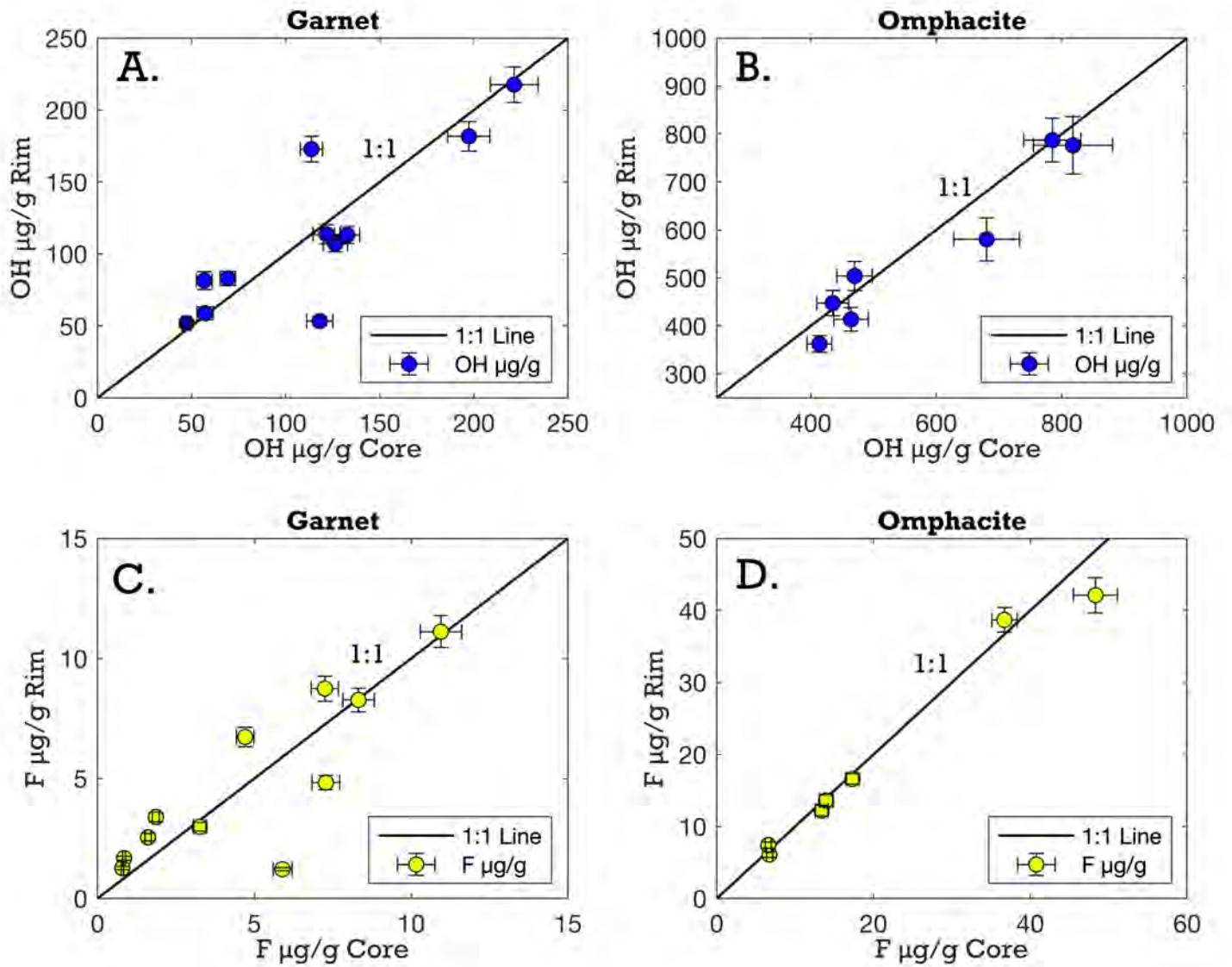


Figure 3.

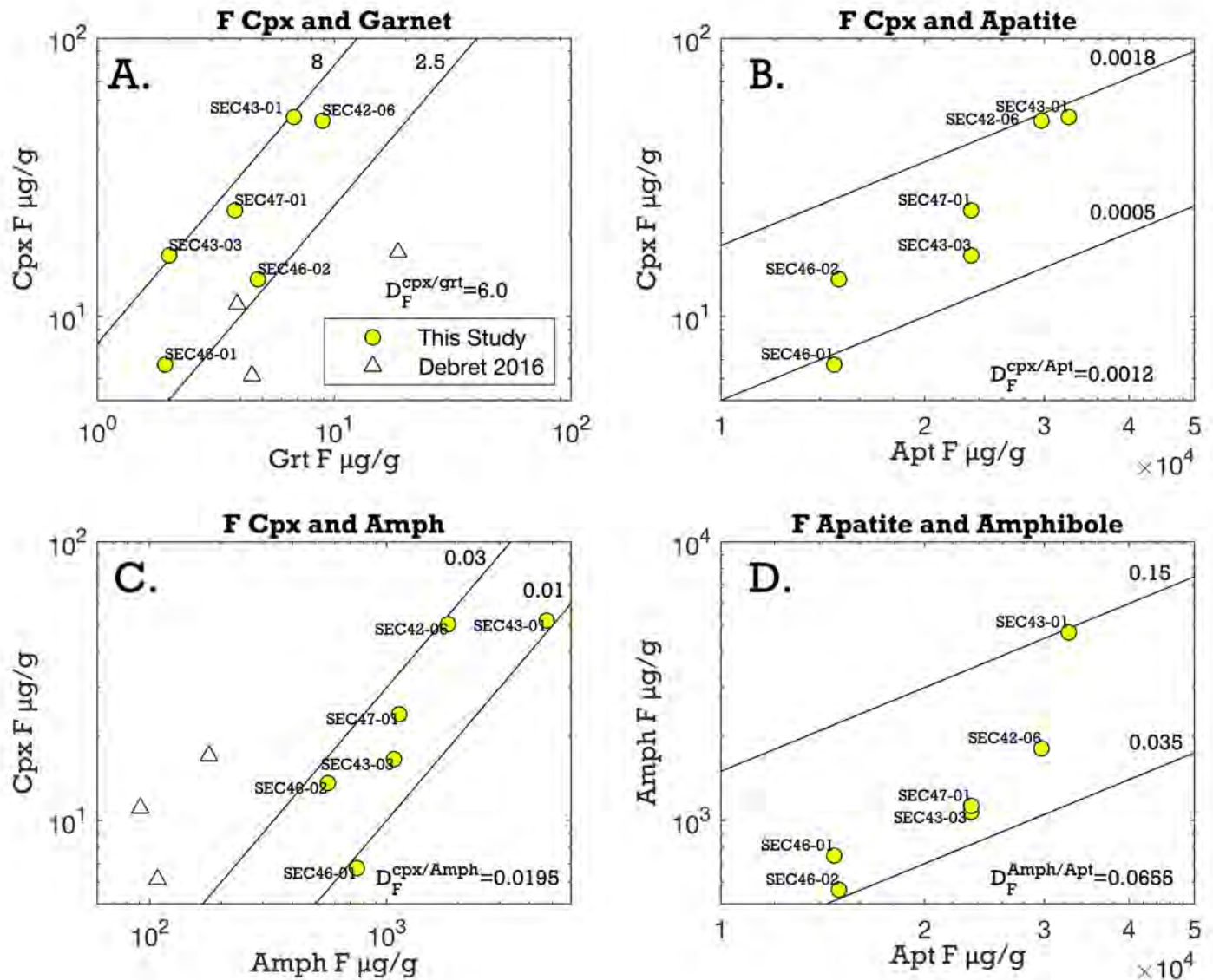


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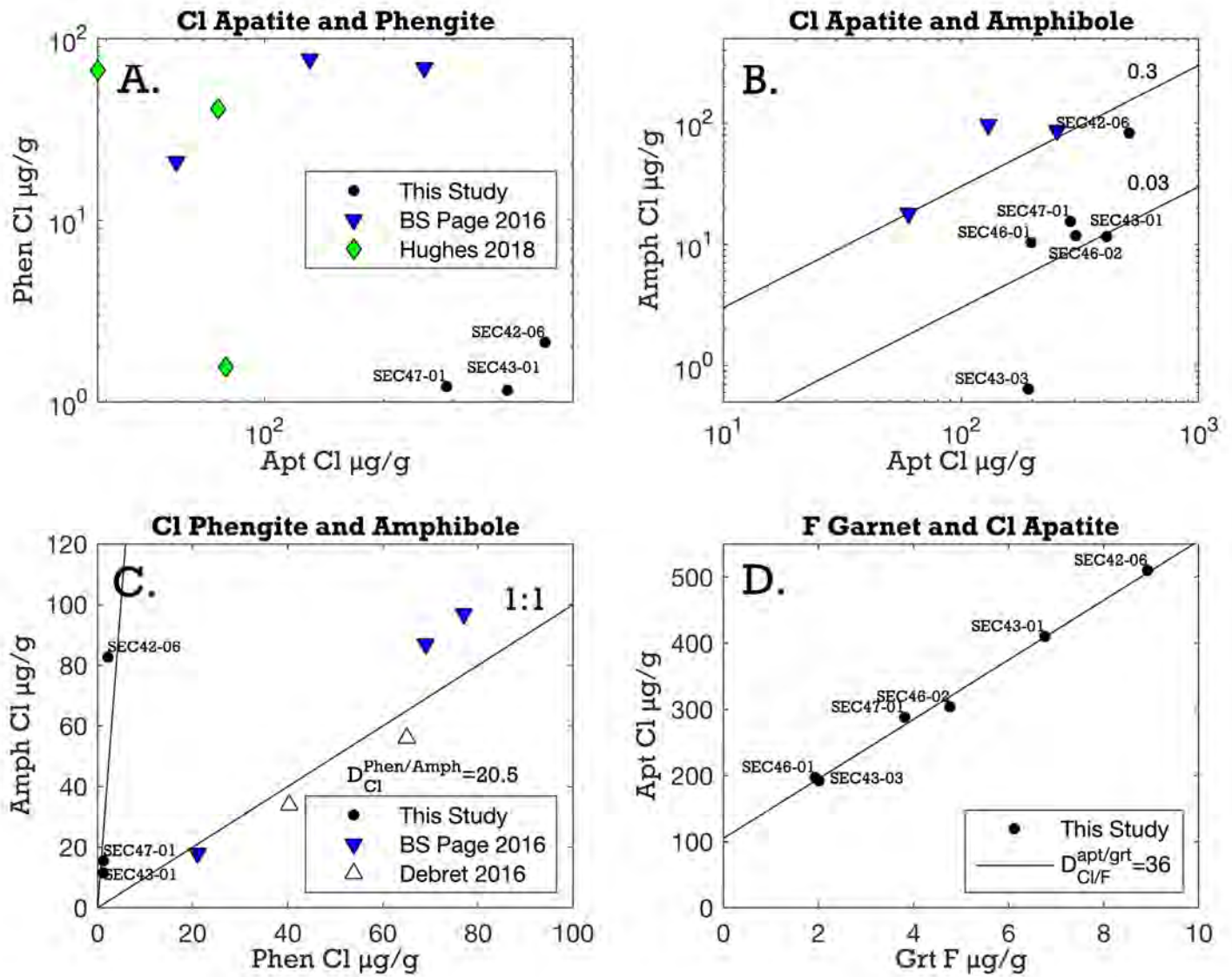


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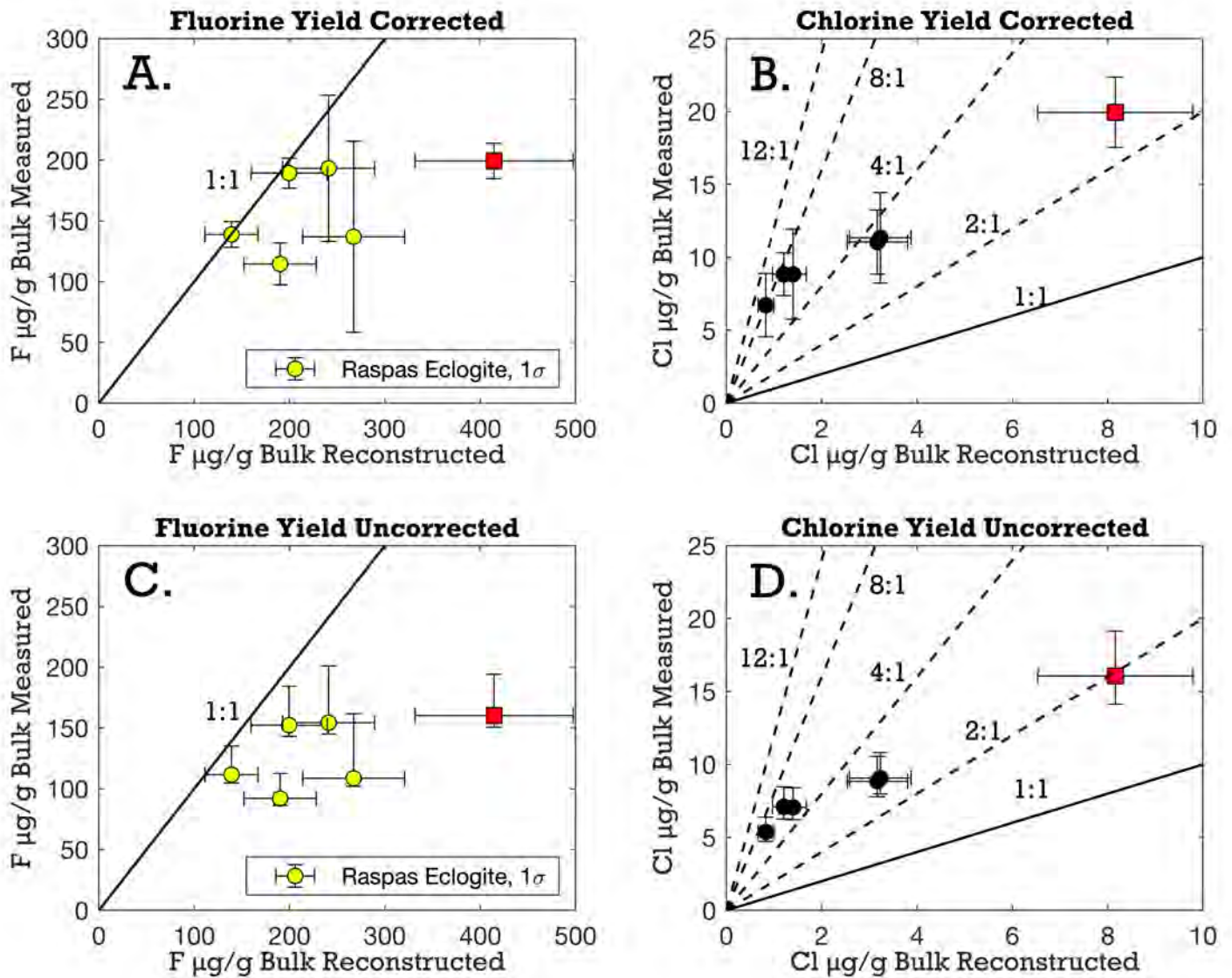


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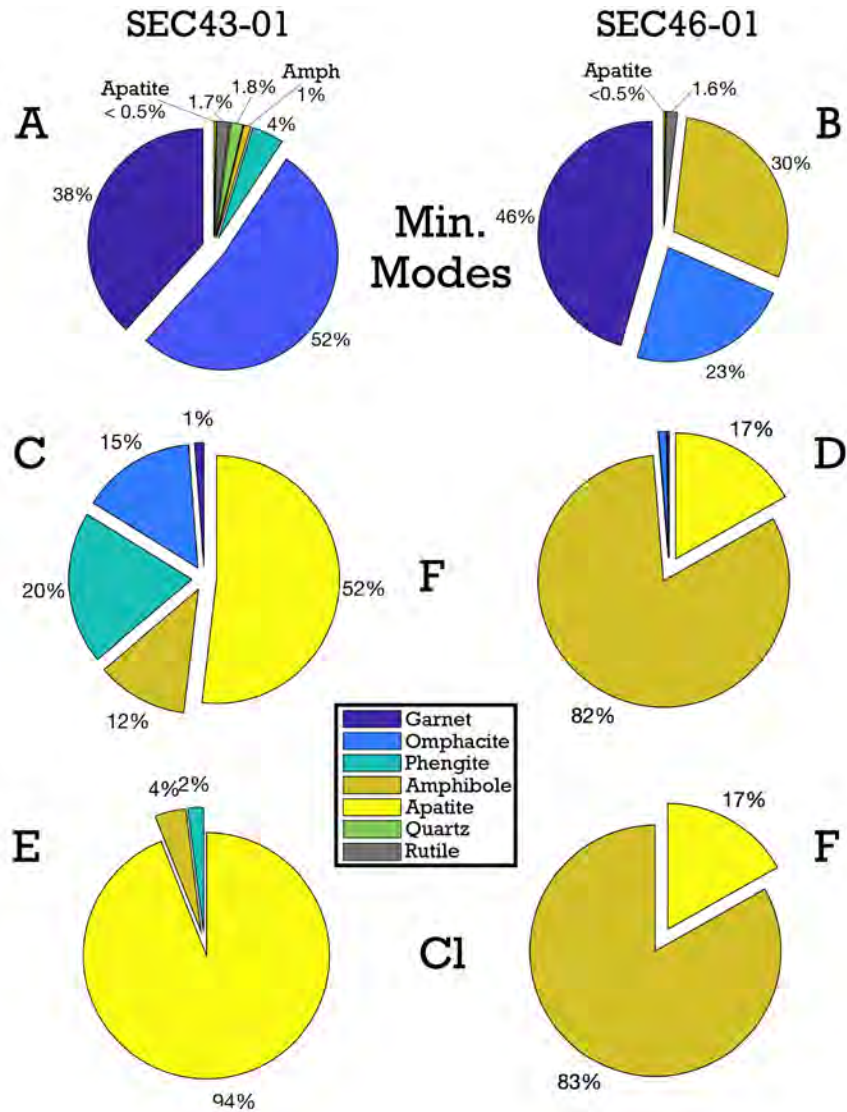


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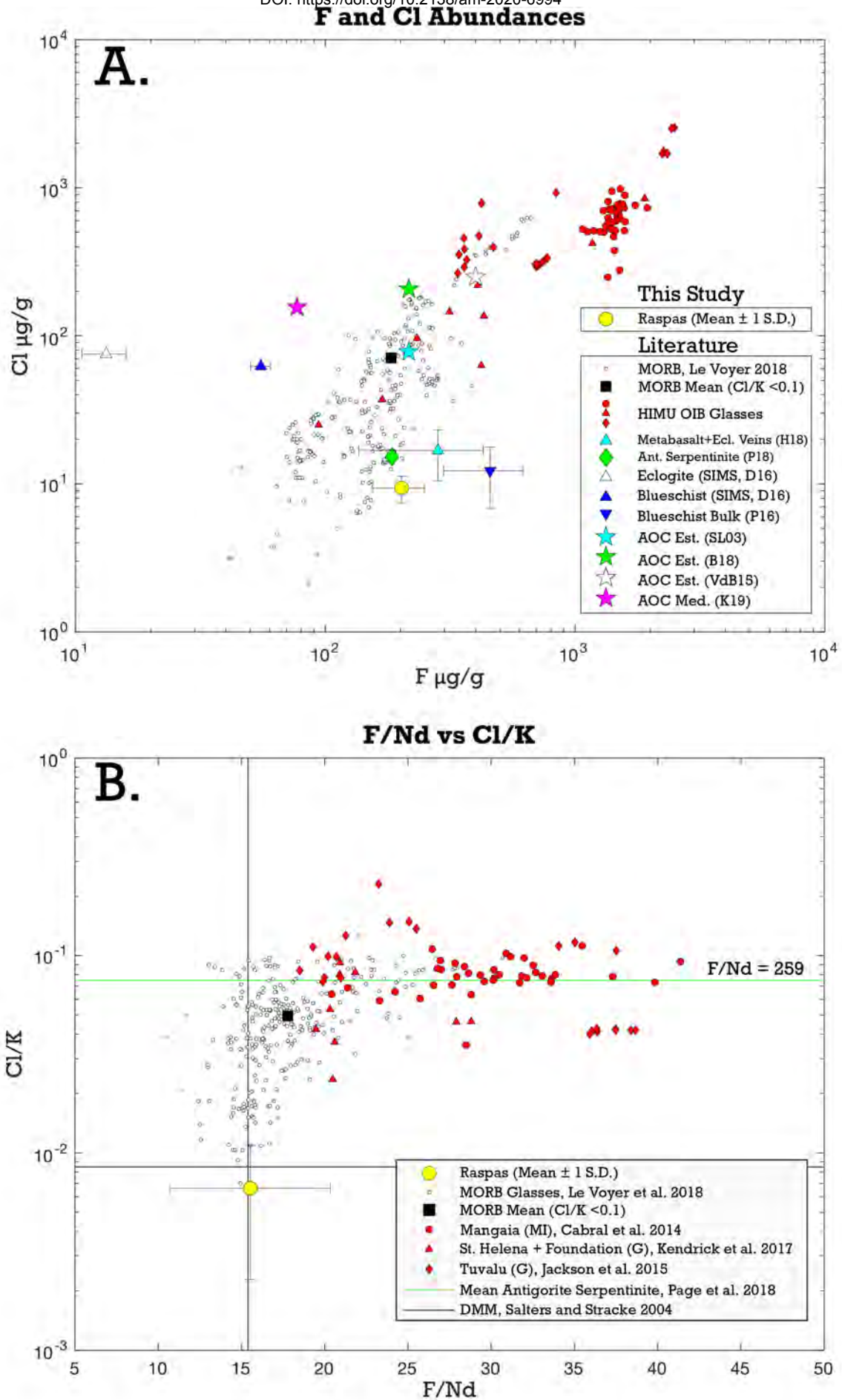


Figure S1.

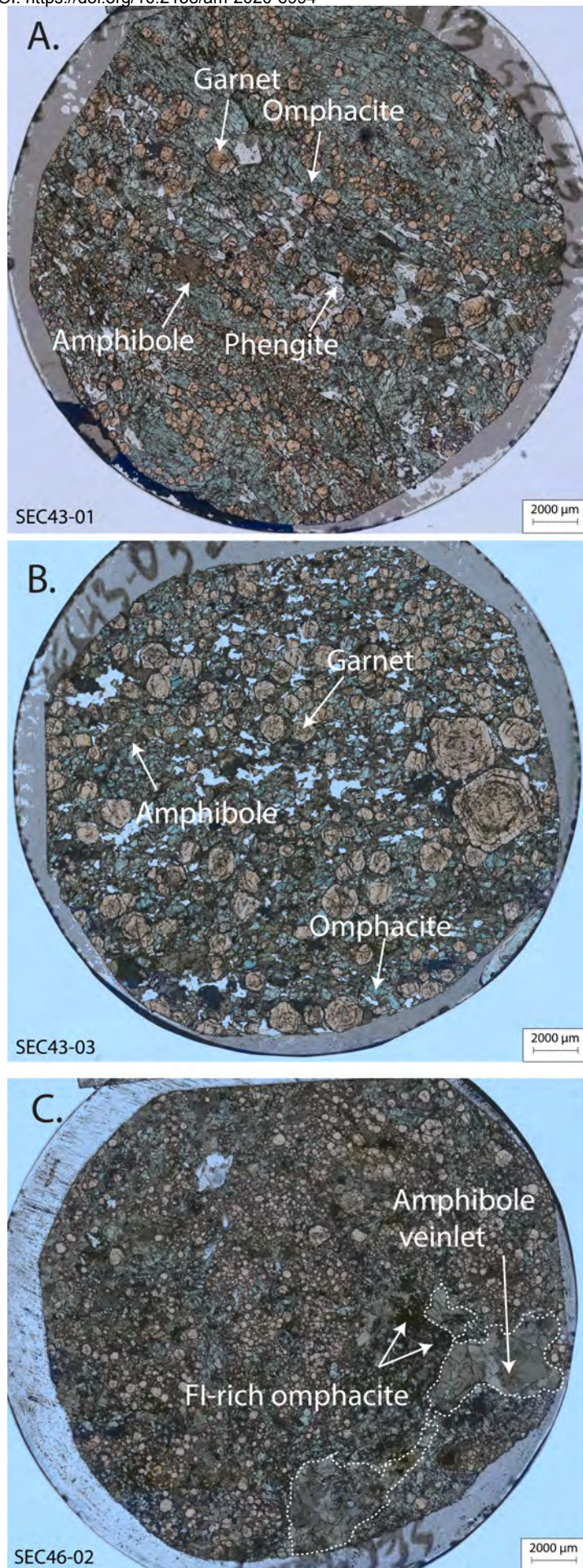


Figure S2.

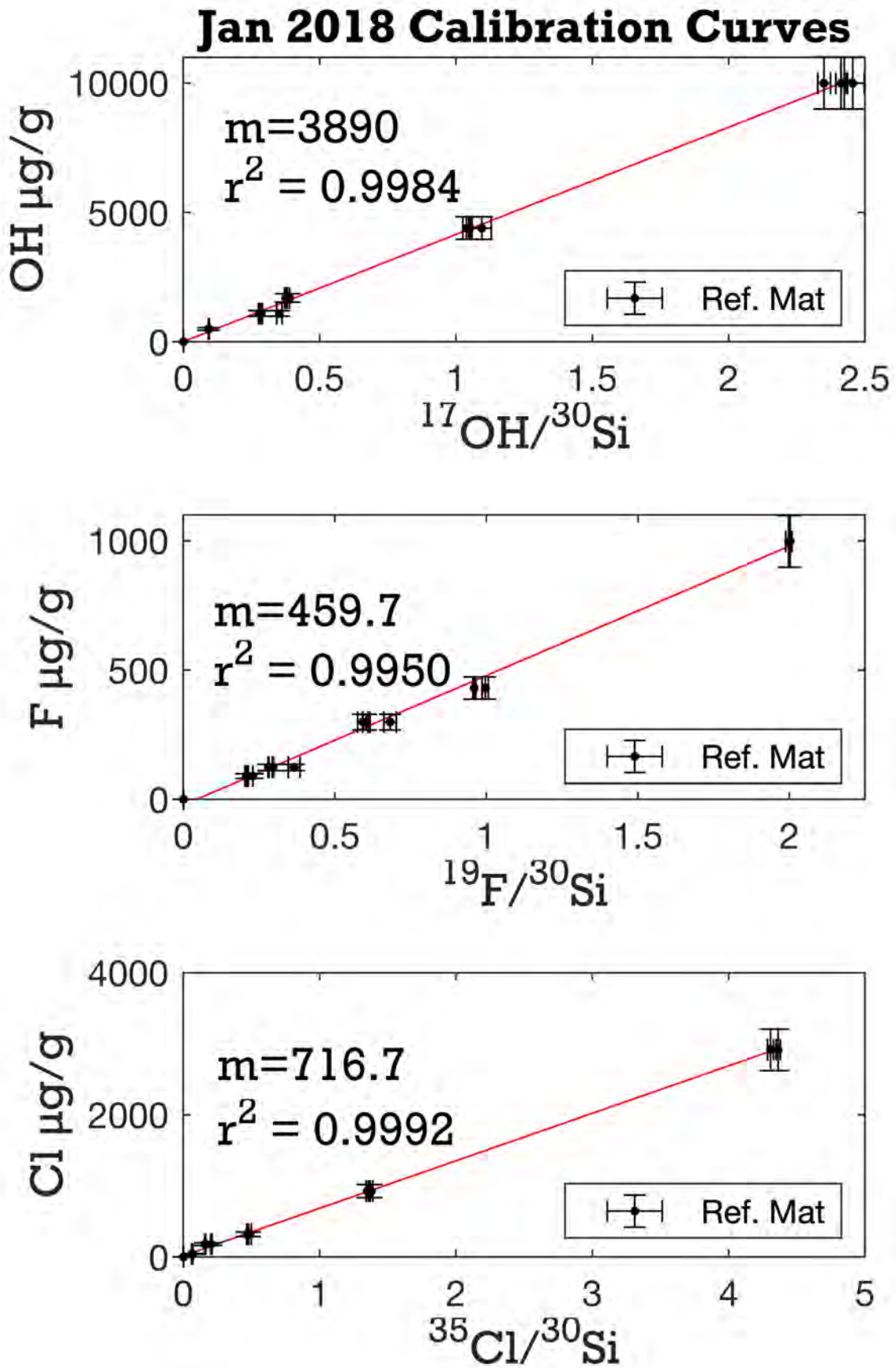


Figure S3.

SEC43-01 Phengite Major Element Zonation

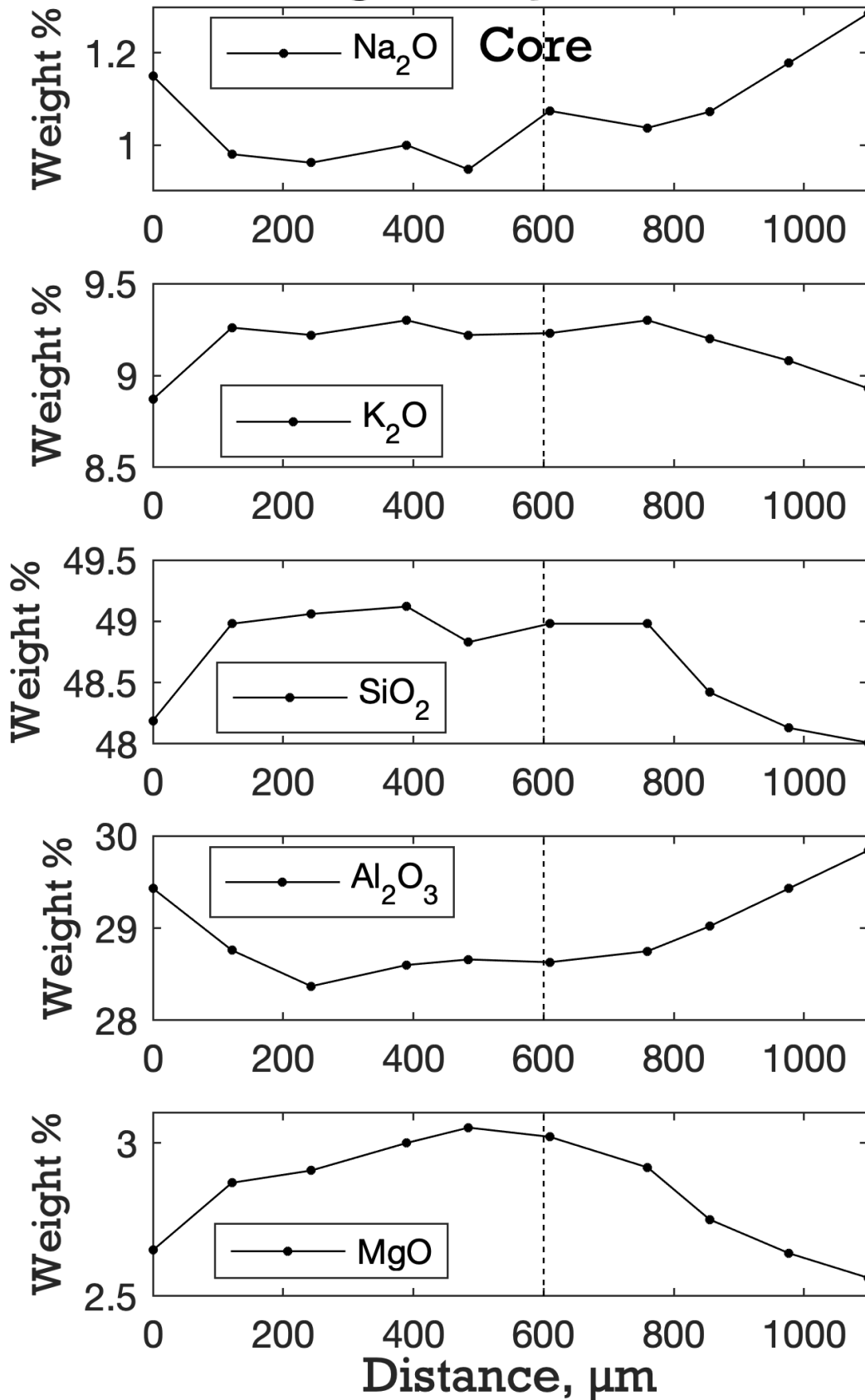


Figure S4.

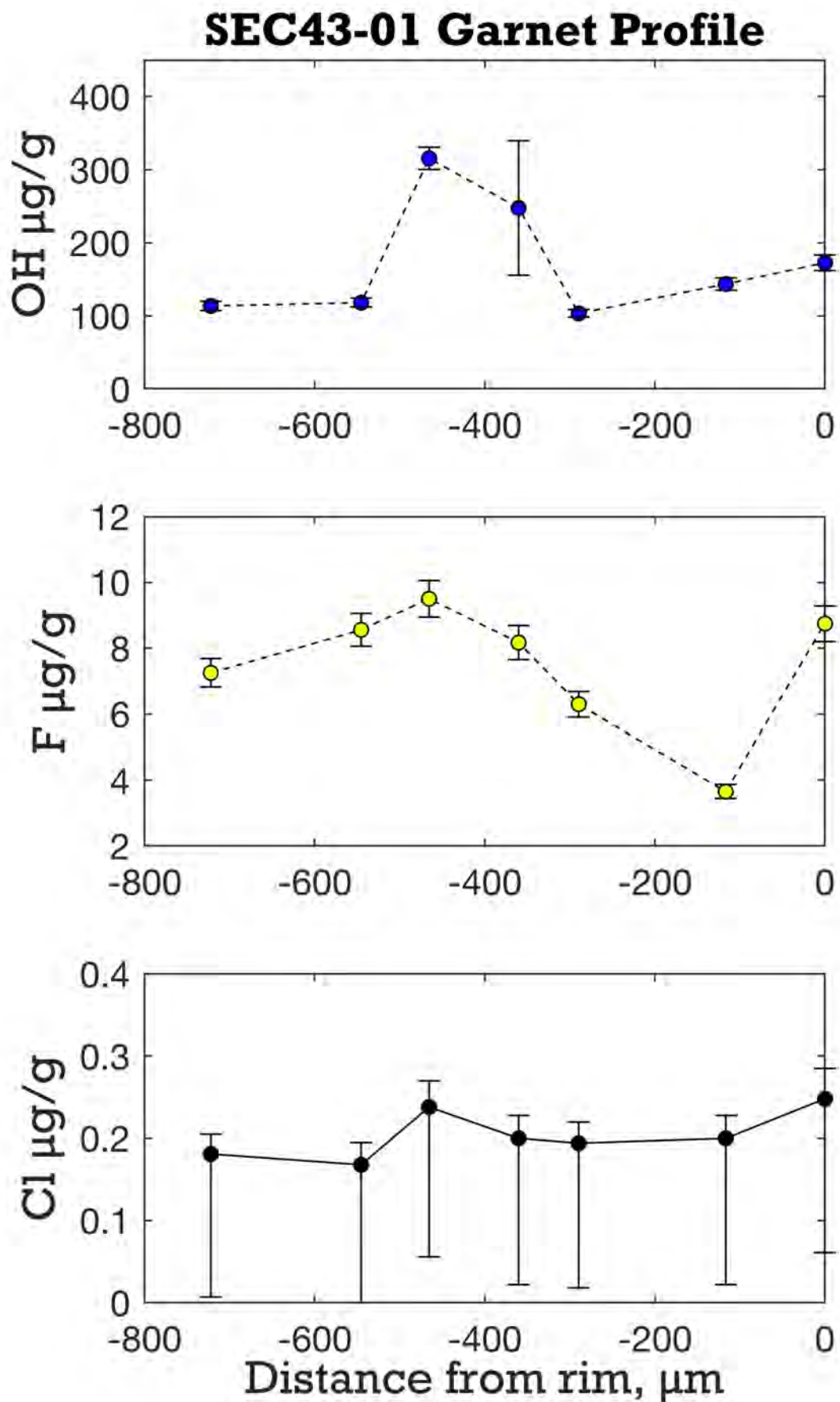


Figure S5.

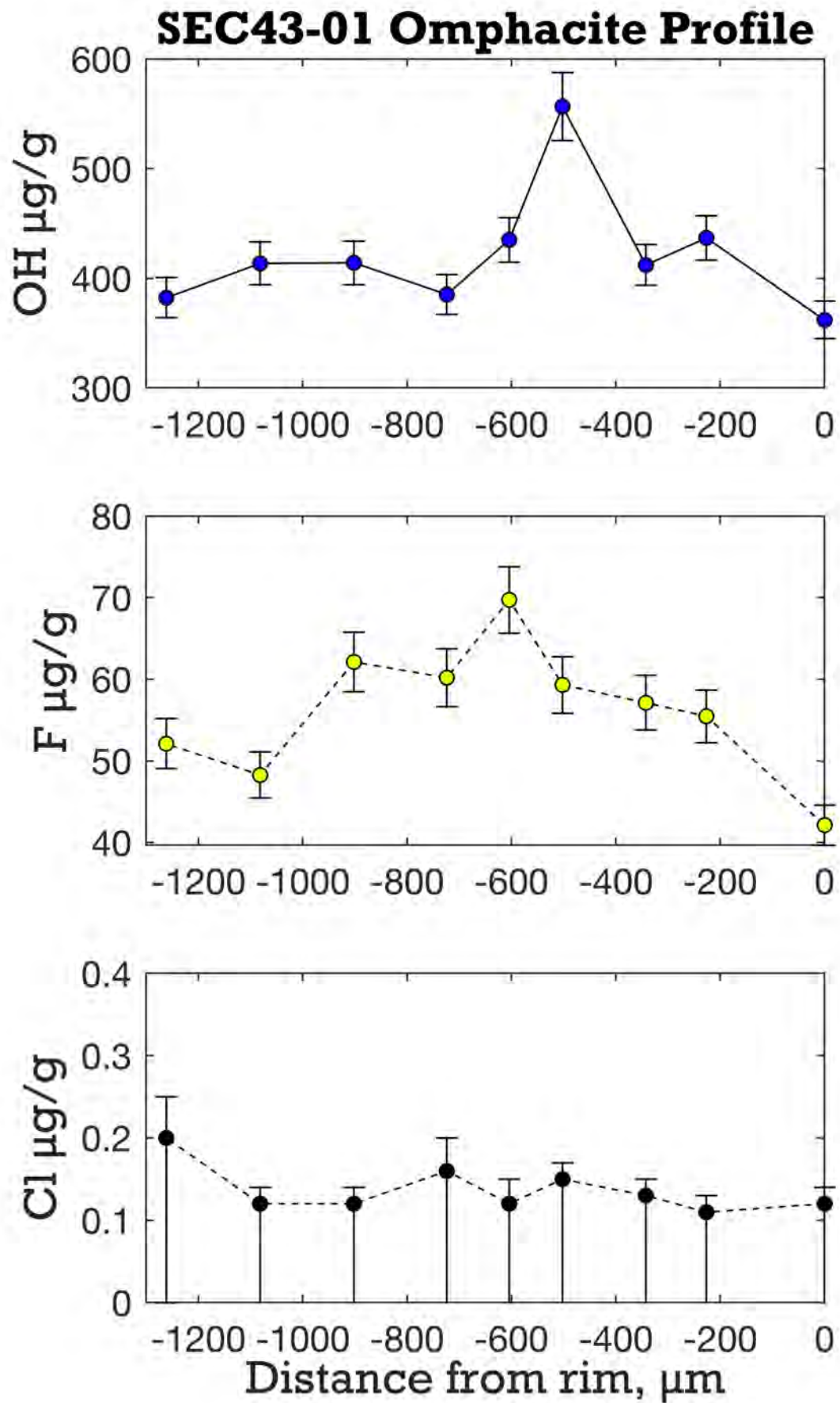


Figure S6.

SEC43-01 Phengite Halogen Zonation

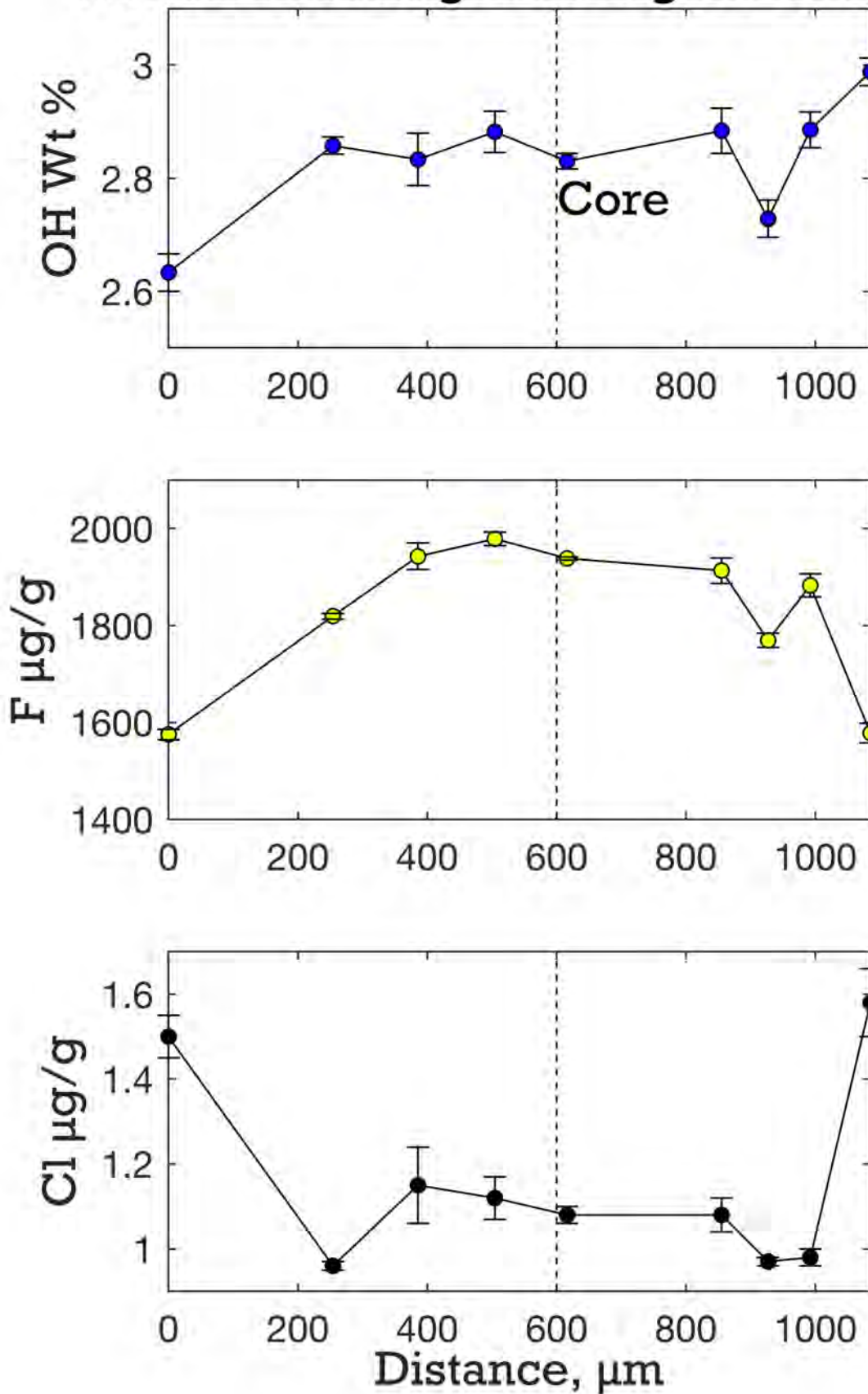


Figure S7.

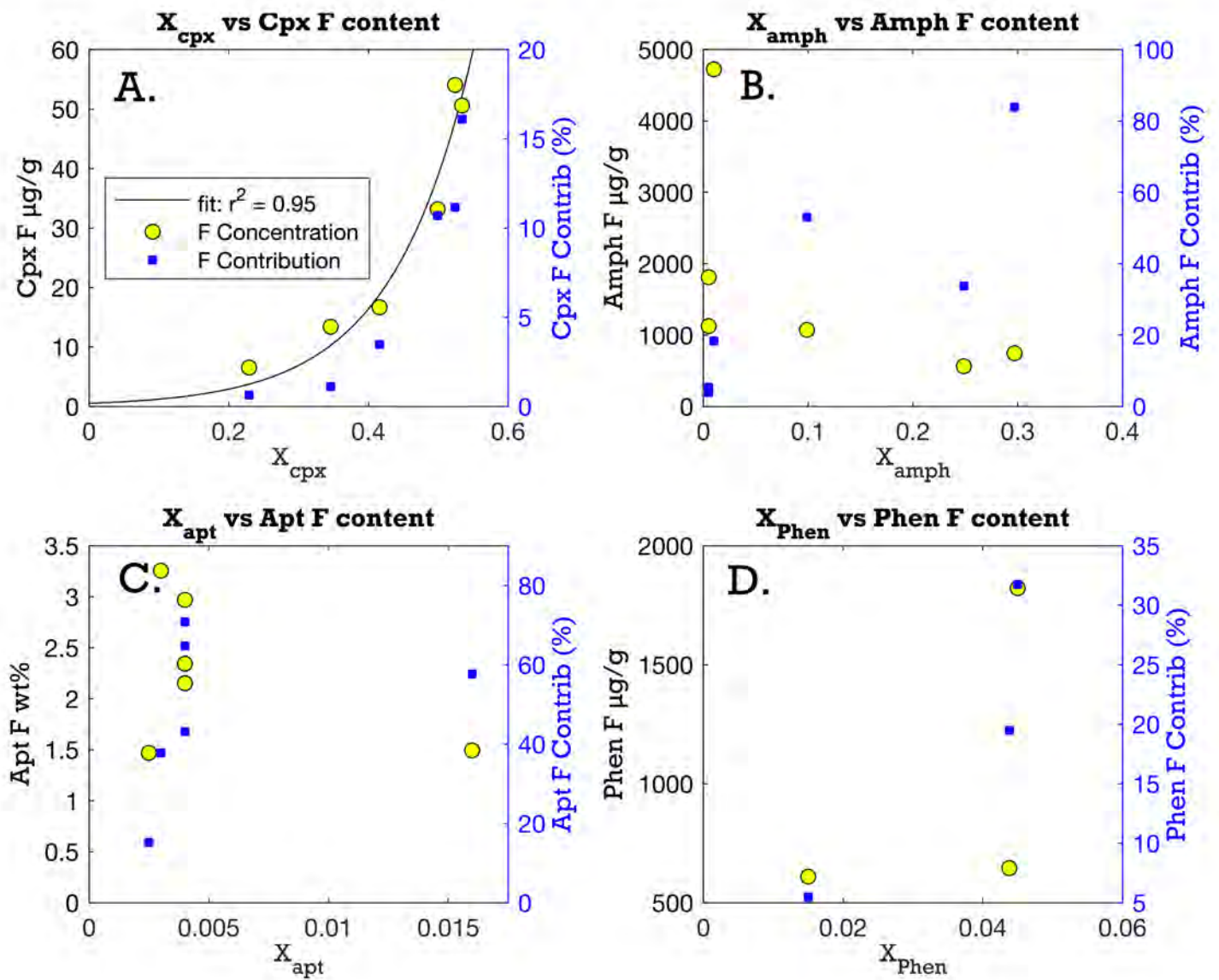


Figure S8.

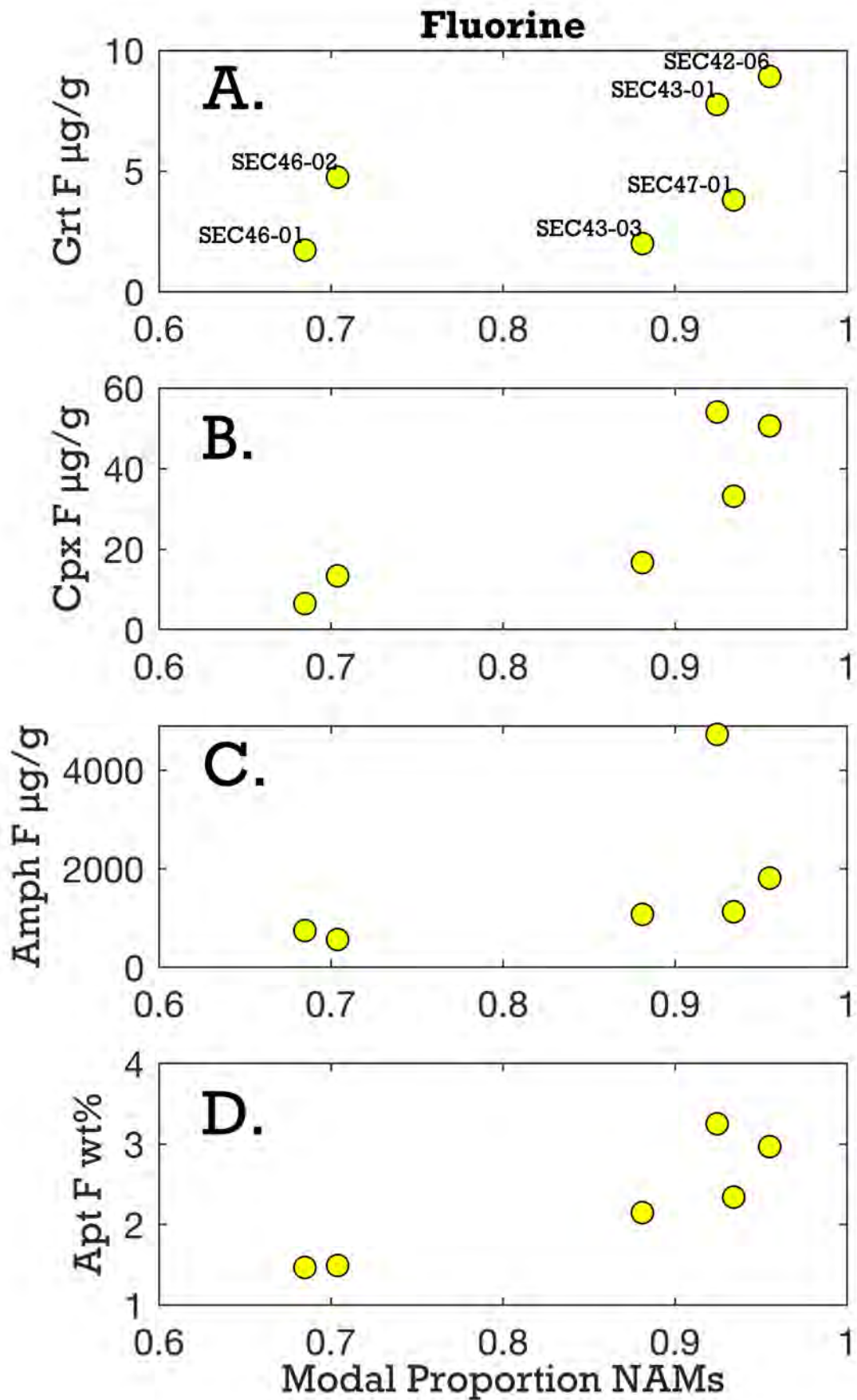


Figure S9.

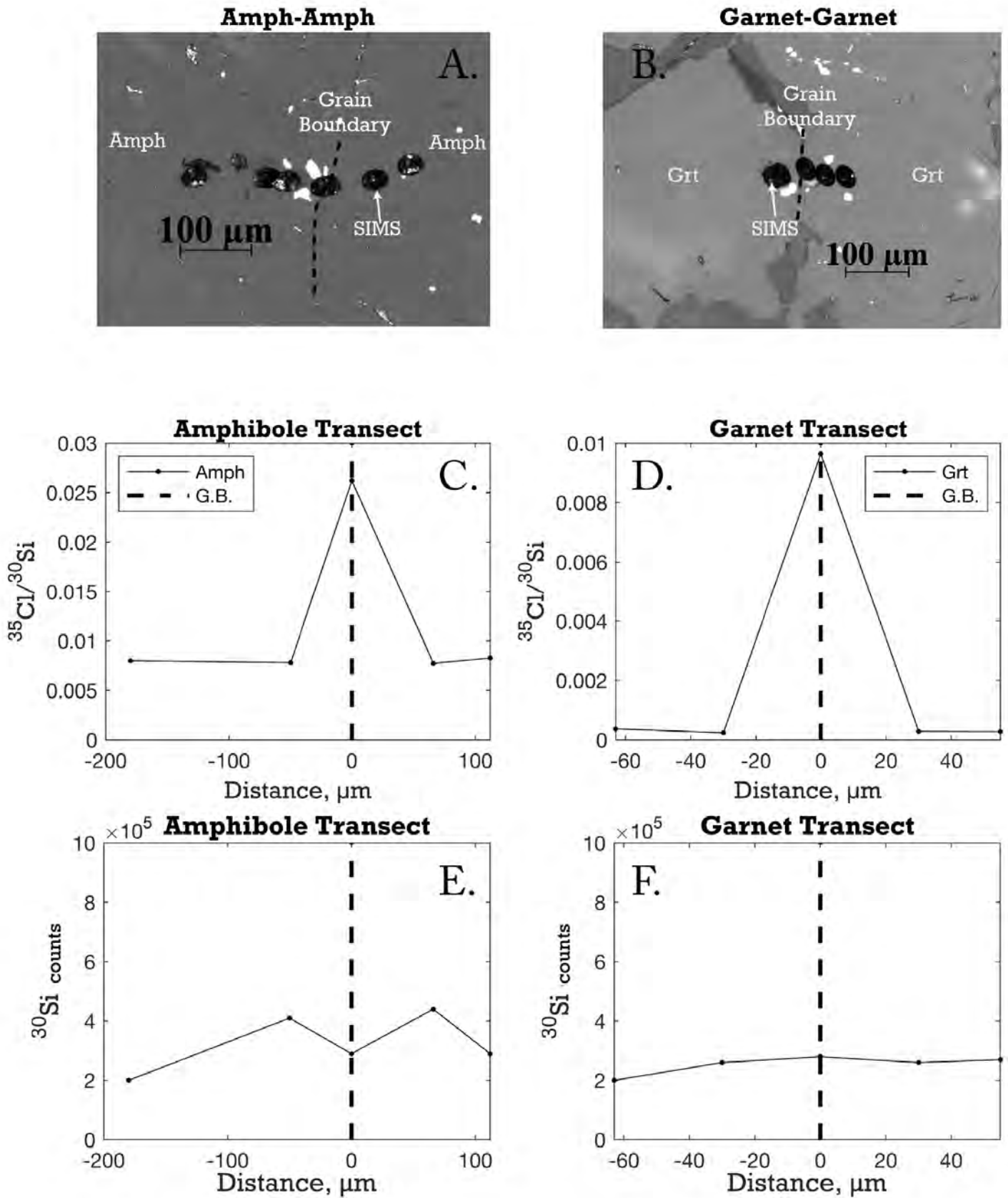


Table 1. Halogen abundances by phase (SIMS)

																Clinopyroxene				Intra-sample Variability			
Sample	Description	No. grains analyzed	No. Analyses	F (µg/g)	F Error % (2SE)		Cl (µg/g)	background corrected (µg/g)	Cl Error % (2SE)		OH (µg/g)	OH Error % (2SE)		S (µg/g)	S Error % (2SE)		F, 1 sigma	OH, 1 sigma	Cl, 1 sigma				
					Internal + Calibration Curve)	Internal + Calibration Curve)			Internal + Calibration Curve)	Internal + Calibration Curve)		Internal + Calibration Curve)	Internal + Calibration Curve)										
SEC 42-06	Eclogite	1	1	50.6	6.0%	0.18	0.03	15.7%	651	5.5%	<0.1	12.6%	-	-	-								
SEC 43-01	Eclogite	1	6	54.1	5.8%	0.13	0.00	14.6%	433	4.8%	<0.1	15.5%	15%	13%	-								
SEC 43-03	Eclogite	1	3	16.7	4.5%	0.08	0.00	11.2%	773	5.8%	<0.1	9.2%	3%	3%	-								
SEC46-01	Eclogite	2	5	6.53	5.8%	0.11	0.03	10.5%	446	5.9%	<0.1	8.4%	4%	5%	-								
SEC46-02	Eclogite	1	3	13.4	6.0%	1.17	1.05	9.7%	753	7.7%	<0.1	9.2%	5%	10%	25%								
SEC47-01	Eclogite	3	7	33.2	4.4%	0.07	0.00	12.4%	445	6.0%	<0.1	13.4%	24%	14%	-								

																Garnet			
Sample	Description	No. grains analyzed	No. Analyses	F (µg/g)	F Error % (2SE)		Cl (µg/g)	background corrected (µg/g)	Cl Error % (2SE)		OH (µg/g)	OH Error % (2SE)		S (µg/g)	S Error % (2SE)		F, 1 sigma	OH, 1 sigma	Cl, 1 sigma
					Internal + Calibration Curve)	Internal + Calibration Curve)			Internal + Calibration Curve)	Internal + Calibration Curve)		Internal + Calibration Curve)	Internal + Calibration Curve)						
SEC 42-06	Eclogite	2	5	8.92	6.0%	0.17	0.02	14.5%	113	5.1%	10.1	11.4%	24%	16%	-				
SEC 43-01	Eclogite	2	9	7.76	6.0%	0.18	0.03	14.0%	125	5.2%	10.8	16.1%	29%	67%	-				
SEC 43-03	Eclogite	2	6	2.01	6.1%	0.10	0.02	11.5%	201	5.7%	18.8	9.2%	42%	10%	-				
SEC46-01	Eclogite	2	7	1.73	5.1%	0.11	0.03	10.0%	85	5.9%	12.3	11.5%	93%	27%	-				
SEC46-02	Eclogite	2	6	4.76	6.1%	0.11	0.00	11.8%	60	7.6%	13.8	10.5%	35%	22%	-				
SEC47-01	Eclogite	3	3	3.81	5.3%	0.08	0.00	11.7%	80	6.2%	12.2	10.4%	12%	7%	-				

																Phengite			
Sample	Description	No. grains analyzed	No. Analyses	F (µg/g)	F Error % (2SE)		Cl (µg/g)	background corrected (µg/g)	Cl Error % (2SE)		OH (µg/g)	OH Error % (2SE)		S (µg/g)	S Error % (2SE)		F, 1 sigma	OH, 1 sigma	Cl, 1 sigma
					Internal + Calibration Curve)	Internal + Calibration Curve)			Internal + Calibration Curve)	Internal + Calibration Curve)		Internal + Calibration Curve)	Internal + Calibration Curve)						
SEC 42-06	Eclogite	1	2	610	5.9%	2.13	1.98	12.7%	22452	4.6%	0.65	8.4%	6%	1%	4%				
SEC 43-01	Eclogite	1	9	1822	5.9%	1.16	1.01	12.6%	28360	4.6%	0.54	7.6%	8%	4%	20%				
SEC 43-03	Eclogite																		
SEC46-01	Eclogite																		
SEC46-02	Eclogite																		
SEC47-01	Eclogite	2	6	645	4.4%	1.22	1.14	8.7%	26410	5.7%	0.48	5.7%	7%	9%	9%				

																Amphibole			
Sample	Description	No. grains analyzed	No. Analyses	F (µg/g)	F Error % (2SE)		Cl (µg/g)	background corrected (µg/g)	Cl Error % (2SE)		OH (µg/g)	OH Error % (2SE)		S (µg/g)	S Error % (2SE)		F, 1 sigma	OH, 1 sigma	Cl, 1 sigma
					Internal + Calibration Curve)	Internal + Calibration Curve)			Internal + Calibration Curve)	Internal + Calibration Curve)		Internal + Calibration Curve)	Internal + Calibration Curve)						
SEC 42-06	Eclogite	1	5	1812	4.7%	82.7	82.6	13.1%	18682	4.2%	27.9	11.0%	18%	5%	37%				
SEC 43-01	Eclogite	1	3	4727	5.9%	11.6	11.5	12.2%	20187	4.6%	18.0	5.9%	3%	3%	24%				
SEC 43-03	Eclogite	1	2	1073	4.6%	0.64	0.56	8.5%	17952	5.9%	27.0	4.4%							
SEC46-01	Eclogite	4	8	747	5.5%	10.3	10.3	8.7%	17292	5.9%	5.6	5.3%							
SEC46-02	Eclogite	2	6	563	6.2%	11.8	11.7	8.6%	23449	7.5%	5.9	6.1%							
SEC47-01	Eclogite	1	3	1127	5.0%	15.5	15.4	8.6%	15747	5.8%	3.0	5.2%							

																Titanite			
Sample	Description	No. grains analyzed	No. Analyses	F (µg/g)	F Error % (2SE)		Cl (µg/g)	background corrected (µg/g)	Cl Error % (2SE)		OH (µg/g)	OH Error % (2SE)		S (µg/g)	S Error % (2SE)				
					Internal + Calibration Curve)	Internal + Calibration Curve)			Internal + Calibration Curve)	Internal + Calibration Curve)									
SEC 46-02	Eclogite	1	3	1394	6.0%	0.11		9.7%	4010	7.4%	<0.1	9.3%							

Table 2. Calculated mineral modes

Est. Modal Abundances	Phase Fraction									NAMs (Grt+Cpx+Qtz)
	Garnet	Omphacite	Phengite	Amphibole	Quartz	Rutile	Titanite	Apatite	SUM	
SEC 42-06	0.421(32)	0.534(32)	0.015(2)	0.0050(5)	0	0.021(2)	0	0.0040(4)	1	0.96
SEC 43-01	0.384(30)	0.524(31)	0.045(4)	0.010(1)	0.017(2)	0.018(2)	0	0.0030(3)	1	0.92
SEC 43-03	0.378(28)	0.416(28)	0	0.099(10)	0.087(2)	0.016(2)	0	0.0040(4)	1	0.88
SEC46-01	0.456(28)	0.229(21)	0	0.297(25)	0	0.016(2)	0	0.0025(3)	1	0.68
SEC46-02	0.358(27)	0.346(27)	0	0.248(22)	0	0.010(1)	0.02(1)	0.016(2)	1	0.70
SEC47-01	0.406(31)	0.499(31)	0.044(4)	0.0050(5)	0.024	0.018(2)	0	0.0040(4)	1	0.93
Average Mineral Modes	0.41	0.43	0.011	0.107	0.023	0.000	0.018	0.006	1.000	

Values in parenthesis represent 1 sigma
 uncertainty

Table 3. Halogen bulk rock abundances, ratios

Sample	Uncorrected for Yield			Corrected for Yield				Uncertainty (Yield Corrected)						Notes	
	Calculated Bulk (WHOI)			Pyrohydrolysis (UTA)		Pyrohydrolysis (UTA)		1SD Propogated		Calculated/Measured Bulk		Cl/Nb ^{***}	Cl/K ^{**}		F/Nd ^{**}
	OH $\mu\text{g/g}$	F $\mu\text{g/g}$	Cl $\mu\text{g/g}$	F $\mu\text{g/g}$	Cl $\mu\text{g/g}$	F $\mu\text{g/g}$	Cl $\mu\text{g/g}$	F ($\mu\text{g/g}$)	Cl ($\mu\text{g/g}$)	F %	Cl %				
SEC42-06	901	168	2.5	92	8.9	115	11.0	15	2.2	146%	23%	3.03	0.0089	10.8	
SEC43-01	1783	258	1.4	109	7.1	137	8.9	68	3.1	189%	16%	4.34	0.0020	22.5	
SEC43-03	2342	200	0.8	152	5.4	189	6.7	11	2.2	105%	12%	2.80	0.0116	15.8	
SEC46-01	5123	241	3.2	154	9.1	193	11.3	52	3.1	125%	28%	5.18	0.0085	17.3	
SEC46-02	6836	415	8.2	160	16.1	199	19.9	12	2.4	208%	41%	4.68	0.0218	32.1	Evidence for fluid interaction, minor retrogression
SEC47-01	1663	145	1.3	112	7.1	139	8.8	9.1	1.5	104%	15%	4.23	0.0021	11.2	Table 1. Halogen abundances by phase (SIMS)
Average		237.7	2.9	129.8	8.9	162.0	11.1								
Standard Deviation		96.6	2.7	29.2	3.8	36.1	4.6								
Average*		202.3	1.85	123.7	7.5	154.5	9.4					3.9	0.0066	15.5	
Standard Deviation*		47.7	0.987	28.0	1.51	34.9	1.88					1.0	0.0043	4.8	
AOC (Barnes et al. 2018)		216	207												
Average Recycling Efficiency (to mantle)		93.7%	0.9%	57.3%	3.6%	71.5%	4.5%								

* Excludes SEC46-02 due to retrograde alteration

** Values used in bulk flux calculations

*** Cl and F values from yield corrected pyrohydrolysis, K, Nb, Nd using bulk rock chemistry from John et al. 2010

Supplementary Table 1. Guest major element compo

Sample	C2O3	Al2O3	CaO	MnO	Na2O	Weight % Oxide										Atoms per formula unit													
						SiO2	K2O	FeO	MgO	Total	Cr	Al	Ca	Mn	Na	Ti	Si	K	Fe	Mg	Total	X Aluminde	X Prope	X Grossular	X Spessartine	(Fe/Mg)			
Line 1 SEC42-06.grn.pdf#0	0	-1346	0.04	21.06	7.76	0.62	0.00	0.07	38.00	0.00	25.30	4.23	98.98	0.0024	0.0399	0.739	0.033	0.00	0.0044	2.993	0.0001	1.667	0.497	7.983	0.57	0.17	0.25	0.01	0.77
Line 2 SEC42-06.grn.pdf#1	144	-1202	0.02	22.07	8.48	0.52	0.02	0.03	37.75	0.00	26.38	4.58	98.86	0.0014	0.054	0.633	0.045	0.003	0.0018	2.981	0	1.742	0.539	7.903	0.39	0.18	0.21	0.01	0.76
Line 3 SEC42-06.grn.pdf#2	316	-1030	0.07	21.92	7.68	0.36	0.00	0.09	37.54	0.00	26.39	4.59	98.65	0.0023	0.048	0.612	0.0243	0	0.0054	2.976	0	1.763	0.518	7.9911	0.60	0.18	0.22	0.01	0.77
Line 4 SEC42-06.grn.pdf#3	440	-907	0.05	21.81	8.22	0.44	0.03	0.12	37.29	0.00	26.36	4.20	98.72	0.0032	0.042	0.61	0.0298	0.0049	0.0069	2.962	0	1.765	0.497	8.0188	0.39	0.17	0.21	0.01	0.78
Line 5 SEC42-06.grn.pdf#4	580	-786	0.04	21.48	8.54	0.35	0.00	0.15	36.81	0.00	26.35	4.36	97.87	0.0027	0.027	0.716	0.0237	0	0.0088	2.961	0	1.813	0.426	8.0062	0.60	0.14	0.22	0.01	0.81
Line 6 SEC42-06.grn.pdf#5	724	-622	0.07	21.30	8.56	0.77	0.00	0.10	36.54	0.00	26.74	3.32	97.61	0.0047	0.048	0.741	0.0528	0	0.0064	2.953	0	1.807	0.4	8.0129	0.60	0.13	0.25	0.02	0.82
Line 7 SEC42-06.grn.pdf#6	904	-442	0.05	21.80	8.90	0.80	0.00	0.07	36.82	0.00	25.48	4.04	97.73	0.0011	0.042	0.765	0.051	0	0.0041	2.956	0	1.71	0.484	8.0383	0.57	0.16	0.25	0.02	0.78
Line 8 SEC42-06.grn.pdf#7	1001	-345	0.10	21.53	8.89	0.54	0.06	0.07	36.85	0.00	25.25	4.39	97.67	0.0006	0.034	0.764	0.0454	0.009	0.004	2.955	0	1.693	0.525	8.0264	0.56	0.17	0.25	0.01	0.76
Line 9 SEC42-06.grn.pdf#8	1195	-151	0.13	21.31	9.04	0.72	0.00	0.10	36.09	0.00	26.14	3.54	96.91	0.0002	0.031	0.789	0.0495	0	0.0043	2.954	0.0003	1.781	0.43	8.0353	0.58	0.14	0.26	0.02	0.81
Line 10 SEC42-06.grn.pdf#9	1346	0	0.03	22.07	8.02	0.49	0.00	0.03	38.43	0.00	26.23	4.78	100.08	0.0002	0.026	0.669	0.0245	0.0004	0.0015	2.994	0.0001	1.709	0.555	7.9896	0.58	0.19	0.23	0.01	0.75
SEC42-06 Average			0.06	21.71	8.41	0.56	0.01	0.08	37.21	0.00	26.16	4.10	98.31	0.00	0.02	0.704	0.024	0.000	0.00	2.97	0.00	1.75	0.49	8.01	0.58	0.16	0.24	0.01	0.78
Line 1 SEC43-01.grn.pdf#0	0	-555	0.03	21.84	8.81	0.91	0.04	0.11	38.30	0.00	26.11	3.90	100.06	0.0018	0.015	0.738	0.0603	0.0067	0.0067	2.998	0	1.709	0.455	7.9906	0.58	0.15	0.25	0.02	0.79
Line 2 SEC43-01.grn.pdf#1	62	-493	0.14	21.86	8.74	0.81	0.04	0.10	38.05	0.01	25.88	4.27	100.00	0.0086	0.026	0.733	0.0538	0.0057	0.0059	2.978	0.0013	1.693	0.498	8.0033	0.57	0.17	0.25	0.02	0.77
Line 3 SEC43-01.grn.pdf#2	124	-431	0.09	22.02	8.42	0.71	0.02	0.02	38.19	0.00	25.92	4.30	99.68	0.0014	0.034	0.707	0.0409	0.003	0.0011	2.992	0	1.698	0.502	7.9894	0.57	0.17	0.24	0.02	0.77
Line 4 SEC43-01.grn.pdf#3	185	-370	0.06	21.92	7.77	0.60	0.00	0.08	38.23	0.00	26.48	4.56	99.70	0.0018	0.024	0.612	0.0399	0	0.0044	2.995	0	1.735	0.533	7.9972	0.59	0.18	0.22	0.01	0.76
Line 5 SEC43-01.grn.pdf#4	247	-308	0.08	22.02	7.54	0.61	0.05	0.07	38.29	0.01	26.36	4.23	100.36	0.0048	0.024	0.61	0.0403	0.0077	0.0043	2.985	0.0008	1.745	0.561	8.0029	0.59	0.19	0.21	0.01	0.76
Line 6 SEC43-01.grn.pdf#5	308	-247	0.04	22.00	6.93	0.59	0.12	0.05	38.53	0.01	26.71	5.01	100.01	0.0028	0.027	0.578	0.0297	0.018	0.0027	3.003	0.0014	1.741	0.585	7.9915	0.59	0.20	0.20	0.01	0.75
Line 7 SEC43-01.grn.pdf#6	370	-185	0.07	22.21	7.18	0.49	0.00	0.01	38.50	0.00	26.45	5.11	100.02	0.0041	0.037	0.599	0.0223	0.0007	0.0004	2.996	0.0004	1.721	0.593	7.984	0.58	0.20	0.20	0.01	0.74
Line 8 SEC43-01.grn.pdf#7	431	-124	0.02	22.49	7.74	0.44	0.01	0.01	38.70	0.00	26.45	5.26	100.13	0.0014	0.053	0.642	0.0289	0.0022	0.0005	2.996	0.0004	1.648	0.607	7.995	0.56	0.21	0.22	0.01	0.73
Line 9 SEC43-01.grn.pdf#8	493	-62	0.02	22.50	7.95	0.49	0.00	0.01	38.81	0.00	25.51	5.17	100.47	0.0005	0.047	0.658	0.0124	0.0004	0.0004	2.996	0.0001	1.647	0.595	7.9778	0.56	0.20	0.22	0.01	0.73
Line 10 SEC43-01.grn.pdf#9	555	0	0.02	22.79	8.53	0.52	0.06	0.02	39.07	0.04	24.75	4.97	100.77	0.0009	0.063	0.702	0.034	0.0096	0.0011	3	0.0038	1.589	0.569	7.9725	0.55	0.20	0.24	0.01	0.74
Line 1 SEC43-01.grn.pdf#0	0	-451	0.04	22.07	8.79	0.79	0.05	0.11	38.28	0.00	25.64	3.97	99.74	0.0023	0.017	0.717	0.0523	0.007	0.0048	2.997	0	1.679	0.463	7.9814	0.57	0.16	0.25	0.02	0.78
Line 2 SEC43-01.grn.pdf#1	49	-402	0.06	22.11	8.60	0.68	0.00	0.10	38.41	0.00	25.49	4.40	100.06	0.0015	0.033	0.718	0.048	0	0.0056	2.993	0	1.674	0.511	7.9829	0.57	0.17	0.24	0.02	0.77
Line 3 SEC43-01.grn.pdf#2	97	-353	0.06	22.15	8.16	0.62	0.00	0.09	38.53	0.00	25.93	4.47	100.01	0.0016	0.034	0.681	0.0411	0	0.0051	3.001	0	1.689	0.519	7.9738	0.58	0.18	0.23	0.01	0.76
Line 4 SEC43-01.grn.pdf#3	150	-301	0.09	22.25	8.58	0.59	0.00	0.03	38.22	0.00	25.56	4.48	99.80	0.0055	0.048	0.718	0.039	0	0.0019	2.984	0	1.669	0.522	7.9785	0.57	0.18	0.24	0.01	0.76
Line 5 SEC43-01.grn.pdf#4	200	-251	0.02	22.10	7.25	0.55	0.00	0.05	38.33	0.00	27.12	4.69	100.12	0.0014	0.033	0.606	0.0366	0	0.0031	2.991	0	1.77	0.455	7.9861	0.60	0.18	0.20	0.01	0.76
Line 6 SEC43-01.grn.pdf#5	250	-201	0.02	22.01	7.06	0.45	0.00	0.06	37.57	0.01	27.23	4.64	99.04	0.0012	0.051	0.598	0.0302	0	0.0033	2.97	0.0008	1.8	0.546	8.0005	0.61	0.18	0.20	0.01	0.77
Line 7 SEC43-01.grn.pdf#6	299	-152	0.07	22.29	6.80	0.35	0.04	0.05	38.46	0.01	27.26	5.01	100.34	0.0043	0.042	0.567	0.0228	0.0055	0.0031	2.989	0.001	1.772	0.518	7.9868	0.60	0.20	0.19	0.01	0.75
Line 8 SEC43-01.grn.pdf#7	349	-103	0.09	22.44	6.54	0.22	0.06	0.03	38.53	0.01	26.96	5.15	100.49	0.0013	0.046	0.569	0.0243	0	0.0027	2.993	0	1.746	0.596	7.9794	0.59	0.19	0.21	0.01	0.75
Line 9 SEC43-01.grn.pdf#8	401	-50	0.04	22.44	7.43	0.47	0.00	0.05	38.84	0.00	24.89	5.69	99.85	0.0024	0.046	0.616	0.0205	0	0.0028	3.005	0.0003	1.64	0.566	7.969	0.55	0.23	0.21	0.01	0.71
Line 10 SEC43-01.grn.pdf#9	451	0	0.04	22.27	8.06	0.53	0.00	0.02	38.48	0.00	25.30	4.86	99.57	0.0026	0.047	0.673	0.035	0	0.0014	3	0.0004	1.65	0.565	7.9745	0.56	0.19	0.23	0.01	0.74
SEC43-01 Average			0.05	22.19	7.86	0.58	0.02	0.05	38.42	0.01	26.08	4.74	99.99	0.00	0.04	0.66	0.04	0.00	0.00	2.99	0.00	1.70	0.55	7.99	0.58	0.19	0.22	0.01	0.76
Line 1 SEC43-01.Grt1	0	-502	0.00	22.21	9.32	1.13	0.01	0.07	38.46	0.01	24.72	4.96	100.90	0	0.2022	0.772	0.0742	0.0014	0.0042	2.971	0.001	1.597	0.572	8.0148	0.53	0.19	0.26	0.02	0.74
Line 2 SEC43-01.Grt1	36	-465	0.00	22.14	9.96	1.17	0.05	0.12	38.44	0.00	24.08	4.02	100.87	0	0.2016	0.824	0.0646	0.0022	0.0071	2.977	0	1.563	0.555	8.019	0.52	0.18	0.27	0.01	0.74
Line 3 SEC43-01.Grt1	72	-430	0.00	21.99	9.97	1.28	0.01	0.15	38.23	0.00	24.09	4.73	100.46	0	0.2012	0.829	0.0441	0.014	0.0085	2.968	0	1.564	0.55	8.017	0.52	0.18	0.27	0.01	0.74
Line 4 SEC43-01.Grt1	107	-395	0.00	21.76	10.41	1.30	0.04	0.14	38.00	0.00	23.27	4.55	99.97	0	0.1999	0.869	0.086	0.0061	0.0377	2.961	0	1.517	0.529	8.0048	0.51	0.18	0.29	0.01	0.74
Line 5 SEC43-01.Grt1	143	-358	0.00	21.86	10.28	1.33	0.05	0.16	38.27	0.00	23.72	4.65	100.32	0.0002	0.202	0.856	0.088	0.0076	0.0092	2.975	0	1.542	0.539	8.019	0.51	0.18	0.28	0.01	0.74
Line 6 SEC43-01.Grt1	179	-323	0.00	21.87	10.71	1.67	0.08	0.19	38.18	0.00	23.33	4.27	100.30	0	0.2007	0.893	0.11	0.0119	0.011	2.972	0	1.519	0.495	8.0189	0.50	0.16	0.30	0.04	0.75
Line 7 SEC43-01.Grt1	215	-287	0.00	21.48	10.98	2.43	0.05	0.12	37.65	0.00	24.00	3.32	100.03	0	0.1995	0.927	0.162	0.0074	0.0073	2.965	0	1.581	0.389	8.0337	0.52	0.13	0.30	0.05	0.80
Line 8 SEC43-01.Grt1	251	-251	0.00	21.04	10.15	2.12	0.00	0.16	37.00	0.01	23.98	4.06	98.31	0	0.1983	0.97	0.144	0	0.0093	2.959	0.0007	1.59	0.484	8.0401	0.51	0.16	0.28	0.05	0.77
Line 9 SEC43-01.Grt1	289	-214	0.02	22.24	11.27	2.22	0.01	0.11	38.25	0.01	24.03	4.60	100.11	0	0.2002	0.941	0.081												

SEC46-01 Average	0.02	22.50	8.09	0.87	0.03	0.06	38.55	0.00	24.61	5.99	100.72	0.00	2.04	0.67	0.06	0.00	0.00	2.97	0.00	1.58	0.69	8.01	0.53	0.23	0.22	0.02	0.70		
Line 1 SEC46-02_gr1	0	-216	0.03	22.16	10.31	0.71	0.06	0.13	38.48	0.00	24.07	4.87	100.82	0.0018	2.036	0.853	0.0484	0.0093	0.0077	2.97	0	1.554	0.56	8.0382	0.52	0.19	0.28	0.02	0.74
Line 2 SEC46-02_gr1	54	-162	0.02	22.29	10.56	0.65	0.04	0.14	38.77	0.00	23.51	5.04	101.03	0.0013	2.037	0.809	0.0424	0.0067	0.0084	2.977	0	1.51	0.576	8.0078	0.50	0.19	0.29	0.03	0.72
Line 3 SEC46-02_gr1	108	-108	0.05	22.39	10.48	0.67	0.05	0.09	38.42	0.00	23.82	4.88	100.85	0.0028	2.034	0.866	0.0436	0.0078	0.0054	2.962	0	1.536	0.561	8.0186	0.51	0.19	0.29	0.03	0.73
Line 4 SEC46-02_gr1	163	-53	0.02	22.60	9.58	0.57	0.04	0.11	38.78	0.00	24.21	5.41	101.31	0.0011	2.039	0.785	0.0367	0.0056	0.0062	2.968	0	1.55	0.617	8.0086	0.52	0.21	0.26	0.01	0.72
Line 5 SEC46-02_gr1	216	0	0.00	22.44	8.17	0.65	0.05	0.06	38.69	0.00	25.31	5.70	101.07	0	2.032	0.672	0.0425	0.0068	0.0037	2.973	0	1.626	0.653	8.009	0.54	0.22	0.22	0.01	0.71
Line 1 SEC46-02_gr2	0	-222	0.01	22.44	11.06	0.62	0.03	0.07	38.73	0.00	22.83	4.96	100.73	0.0005	2.033	0.911	0.0401	0.0039	0.0038	2.977	0	1.607	0.568	8.0043	0.49	0.19	0.31	0.01	0.72
Line 2 SEC46-02_gr2	23	-198	0.06	22.47	10.01	0.58	0.06	0.05	38.68	0.00	24.89	4.66	101.45	0.0014	2.034	0.824	0.0274	0.0089	0.0028	2.97	0	1.599	0.534	8.0135	0.53	0.18	0.28	0.01	0.75
Line 3 SEC46-02_gr2	48	-174	0.00	22.56	10.55	0.60	0.09	0.04	38.54	0.00	23.75	4.94	101.07	0.0003	2.044	0.868	0.039	0.0127	0.0022	2.961	0	1.527	0.566	8.0202	0.51	0.19	0.29	0.03	0.73
Line 4 SEC46-02_gr2	72	-150	0.01	22.43	10.37	0.60	0.00	0.04	38.51	0.00	24.63	4.57	101.16	0.0008	2.037	0.856	0.039	0	0.0021	2.967	0	1.587	0.524	8.0129	0.53	0.17	0.28	0.01	0.75
Line 5 SEC46-02_gr2	95	-127	0.03	22.33	9.73	0.70	0.03	0.01	38.31	0.00	26.11	4.21	101.47	0.002	2.033	0.806	0.0457	0.0051	0.0008	2.96	0	1.687	0.485	8.0246	0.56	0.16	0.27	0.02	0.78
Line 6 SEC46-02_gr2	122	-100	0.03	22.48	9.50	0.66	0.06	0.10	38.84	0.00	24.54	5.40	101.62	0.0016	2.025	0.778	0.043	0.0095	0.0039	2.969	0	1.569	0.635	8.016	0.52	0.20	0.26	0.01	0.72
Line 7 SEC46-02_gr2	146	-76	0.01	22.53	9.86	0.63	0.01	0.13	38.69	0.00	24.00	5.49	101.36	0.0007	2.033	0.808	0.0405	0.0021	0.0078	2.961	0	1.536	0.626	8.0152	0.51	0.21	0.27	0.01	0.71
Line 8 SEC46-02_gr2	171	-51	0.02	22.60	9.72	0.56	0.05	0.12	38.80	0.00	24.15	5.30	101.31	0.001	2.039	0.797	0.0363	0.007	0.0067	2.97	0	1.546	0.604	8.007	0.52	0.20	0.27	0.01	0.72
Line 9 SEC46-02_gr2	194	-28	0.01	22.39	9.73	0.59	0.01	0.10	38.64	0.00	24.83	4.95	101.24	0.0004	2.029	0.801	0.0381	0.0018	0.0055	2.971	0	1.596	0.567	8.0098	0.53	0.19	0.27	0.01	0.74
Line 10 SEC46-02_gr2	222	0	0.06	22.63	8.32	0.59	0.04	0.02	38.98	0.00	24.64	5.98	101.26	0.0014	2.038	0.681	0.038	0.006	0.0012	2.979	0	1.575	0.681	8.0026	0.53	0.23	0.23	0.01	0.70
SEC46-02 Average	0.02	22.45	9.86	0.62	0.04	0.08	38.66	0.00	24.35	5.09	101.18	0.00	2.03	0.81	0.04	0.01	0.00	2.97	0.00	1.56	0.58	8.01	0.52	0.19	0.27	0.01	0.73		
Line 1 SEC47-01_gr1	0	-146	0.00	22.07	9.78	1.01	0.08	0.12	38.40	0.00	25.80	4.20	101.46	0	2.031	0.81	0.0662	0.0116	0.0069	2.968	0	1.668	0.484	8.0258	0.55	0.16	0.27	0.02	0.78
Line 2 SEC47-01_gr1	37	-109	0.05	22.28	9.43	0.59	0.06	0.15	38.37	0.00	26.22	4.58	101.73	0.0012	2.022	0.778	0.0384	0.0089	0.0087	2.955	0.0003	1.609	0.526	8.0294	0.56	0.17	0.26	0.01	0.76
Line 3 SEC47-01_gr1	73	-73	0.03	22.25	8.06	0.55	0.00	0.12	38.55	0.00	26.99	5.00	101.56	0.0012	2.02	0.665	0.0361	0	0.0068	2.969	0	1.739	0.574	8.012	0.58	0.19	0.22	0.01	0.75
Line 4 SEC47-01_gr1	110	-36	0.03	22.38	7.59	0.64	0.00	0.11	38.43	0.00	27.12	5.18	101.48	0.001	2.033	0.626	0.0416	0	0.0065	2.962	0	1.748	0.595	8.0142	0.58	0.20	0.21	0.01	0.75
Line 5 SEC47-01_gr1	146	0	0.04	22.35	8.89	0.64	0.00	0.07	38.63	0.00	26.42	4.64	101.67	0.0012	2.026	0.733	0.0418	0	0.0038	2.971	0	1.7	0.532	8.0098	0.57	0.18	0.24	0.01	0.76
Line 1 SEC47-01_gr2	0	-112	0.01	22.17	9.72	0.64	0.09	0.14	38.41	0.00	26.12	4.26	101.55	0.0004	2.017	0.804	0.0417	0.011	0.0083	2.965	0	1.686	0.49	8.0254	0.56	0.16	0.27	0.01	0.77
Line 2 SEC47-01_gr2	28	-84	0.04	22.18	9.46	0.70	0.00	0.13	38.42	0.00	26.30	4.49	101.52	0.0012	2.017	0.782	0.0407	0.0004	0.0075	2.964	0	1.684	0.536	8.0188	0.56	0.17	0.26	0.02	0.77
Line 3 SEC47-01_gr2	56	-56	0.05	22.05	9.26	0.44	0.07	0.15	38.40	0.00	26.85	4.39	101.66	0.0011	2.007	0.766	0.0286	0.0098	0.009	2.965	0	1.713	0.585	8.0265	0.57	0.17	0.25	0.01	0.77
Line 4 SEC47-01_gr2	84	-28	0.02	22.42	8.13	0.41	0.07	0.08	38.47	0.00	27.12	4.85	101.57	0.0013	2.036	0.671	0.0268	0.0101	0.0045	2.964	0	1.748	0.556	8.0177	0.58	0.19	0.22	0.01	0.76
Line 5 SEC47-01_gr2	112	0	0.04	22.52	7.99	0.58	0.07	0.07	38.50	0.00	26.35	5.25	101.37	0.0017	2.043	0.659	0.0376	0.0097	0.0042	2.962	0	1.696	0.602	8.0162	0.57	0.20	0.22	0.01	0.74
Line 1 SEC47-01_gr3	0	-160	0.10	22.08	9.78	0.77	0.05	0.09	38.39	0.00	26.06	4.13	101.44	0.0006	2.013	0.81	0.0502	0.0069	0.0052	2.968	0	1.685	0.476	8.0203	0.56	0.16	0.27	0.02	0.78
Line 2 SEC47-01_gr3	48	-112	0.16	22.10	9.75	0.63	0.06	0.12	38.50	0.00	25.85	4.48	101.66	0.01	2.007	0.804	0.041	0.009	0.0071	2.966	0	1.665	0.514	8.0231	0.55	0.17	0.27	0.01	0.76
Line 3 SEC47-01_gr3	96	-74	0.09	22.10	8.86	0.66	0.01	0.16	38.25	0.00	26.91	4.35	101.40	0.0088	2.017	0.735	0.0414	0.0022	0.0091	2.962	0	1.743	0.502	8.0195	0.58	0.17	0.24	0.01	0.78
Line 4 SEC47-01_gr3	123	-36	0.03	22.40	7.98	0.57	0.01	0.11	38.39	0.00	27.23	4.80	101.52	0.0017	2.037	0.66	0.0373	0.0022	0.0063	2.962	0	1.757	0.512	8.0155	0.58	0.18	0.22	0.01	0.76
Line 5 SEC47-01_gr3	160	0	0.02	22.47	8.11	0.62	0.04	0.07	38.75	0.00	25.83	5.59	101.50	0.0014	2.01	0.666	0.0404	0.0053	0.0038	2.971	0	1.656	0.639	8.0129	0.55	0.21	0.22	0.01	0.72
SEC47-01 Average	0.05	22.25	8.85	0.63	0.04	0.11	38.46	0.00	26.46	4.68	101.54	0.00	2.02	0.73	0.04	0.01	0.01	2.96	0.00	1.71	0.54	8.02	0.57	0.18	0.24	0.01	0.76		

Supplementary Table 2. Osmphacite major element comp

Line	SiO2	TiO2	Weight Percent Oxide										Atoms per formula unit										Total	Total	Mg/(Mg+Fe)			
			K2O	Na2O	CaO	MgO	FeO	Total	Ti	Si	K	Na	Ca	Al	Fe	Fcd	Mg											
Line 1 SEC4-06_cpml1	0	-0.14	0.17	54.41	0.01	0.05	6.17	0.05	10.07	13.56	6.70	8.14	99.33	0.0046	1.976	0.0007	0.0036	0.434	0.0014	0.433	0.527	0.203	0.441	4.0203	45.00	37.66	17.34	0.68
Line 2 SEC4-06_cpml1	32	-282	0.19	54.26	0.00	0.04	6.56	0.07	9.99	14.99	6.66	8.10	99.33	0.0044	1.975	0.0003	0.0033	0.462	0.0019	0.428	0.526	0.203	0.439	4.0188	45.03	37.59	17.28	0.68
Line 3 SEC4-06_cpml1	64	-250	0.17	54.47	0.00	0.04	6.33	0.04	10.06	13.66	6.73	8.13	99.33	0.0046	1.974	0.0011	0.0033	0.443	0.0011	0.43	0.53	0.204	0.439	4.029	45.18	37.43	17.39	0.68
Line 4 SEC4-06_cpml1	108	-206	0.20	54.46	0.00	0.04	6.56	0.05	10.27	13.48	6.63	7.97	99.66	0.0055	1.972	0.0011	0.0036	0.438	0.0016	0.428	0.523	0.201	0.43	4.0322	45.32	37.26	17.42	0.68
Line 5 SEC4-06_cpml1	153	-161	0.17	54.36	0.00	0.02	6.37	0.07	9.90	13.54	6.65	8.09	99.67	0.0047	1.978	0.0001	0.0037	0.45	0.002	0.425	0.528	0.202	0.439	4.0296	45.17	37.55	17.28	0.68
Line 6 SEC4-06_cpml1	185	-129	0.16	54.77	0.00	0.02	6.35	0.06	10.07	13.40	6.88	8.03	99.74	0.0045	1.981	0.0007	0.0036	0.429	0.0016	0.429	0.519	0.208	0.433	4.0294	44.74	37.33	17.93	0.68
Line 7 SEC4-06_cpml1	224	-89	0.12	54.63	0.00	0.05	6.42	0.06	10.26	13.32	6.86	8.07	99.79	0.0033	1.975	0.0014	0.0033	0.437	0.0016	0.437	0.516	0.207	0.435	4.0266	44.56	37.56	17.88	0.68
Line 8 SEC4-06_cpml1	254	-60	0.14	54.56	0.00	0.02	6.48	0.02	10.42	13.15	6.70	7.95	99.63	0.0037	1.974	0.0007	0.0037	0.444	0.0012	0.444	0.521	0.203	0.429	4.0339	44.66	37.57	17.79	0.68
Line 9 SEC4-06_cpml1	293	-21	0.13	54.78	0.01	0.04	6.34	0.02	9.87	13.54	6.89	8.19	99.62	0.0036	1.98	0.0003	0.0033	0.444	0.0007	0.42	0.528	0.208	0.441	4.027	44.86	37.47	17.67	0.68
Line 10 SEC4-06_cpml1	314	0	0.13	54.83	0.00	0.02	6.11	0.03	9.88	13.46	6.95	8.22	99.74	0.0033	1.982	0.0001	0.0036	0.428	0.0009	0.425	0.521	0.21	0.443	4.0142	44.38	37.73	17.89	0.68
Line 1 SEC4-06_cpml2-000	0	-692	0.17	54.73	0.00	0.02	6.74	0.00	9.74	13.82	6.65	8.19	100.07	0.0047	1.977	0.0007	0.0037	0.472	0.0001	0.415	0.535	0.201	0.441	4.0466	45.45	37.47	17.08	0.69
Line 2 SEC4-06_cpml2-000	157	-358	0.14	54.85	0.00	0.00	6.29	0.03	10.05	13.67	6.74	8.11	99.47	0.0037	1.975	0.0011	0.0037	0.425	0.0002	0.425	0.531	0.205	0.433	4.0255	45.42	37.64	17.44	0.68
Line 3 SEC4-06_cpml2-000	443	-249	0.19	54.63	0.00	0.03	6.57	0.01	10.20	13.75	6.69	7.96	100.01	0.0052	1.972	0.0009	0.0036	0.434	0.0004	0.434	0.532	0.202	0.429	4.0356	45.74	36.89	17.37	0.68
Line 4 SEC4-06_cpml2-000	565	-127	0.18	54.35	0.00	0.03	6.51	0.03	10.17	13.68	6.79	8.06	99.64	0.005	1.965	0.001	0.0036	0.435	0.0008	0.435	0.532	0.206	0.436	4.0319	45.32	37.14	17.55	0.68
Line 5 SEC4-06_cpml2-000	692	0	0.15	54.66	0.00	0.02	6.51	0.06	10.20	13.51	6.81	7.93	99.86	0.0041	1.976	0.0001	0.0037	0.456	0.0018	0.435	0.523	0.206	0.427	4.0297	45.24	36.94	17.82	0.67
SEC4-06 Average			0.16	54.55	0.00	0.03	6.44	0.04	10.08	13.54	6.76	8.07	99.67	0.003	1.98	0.000	0.00	0.45	0.00	0.43	0.53	0.20	0.44	4.03	45.07	37.37	17.55	0.68
Line 1 4-01_Cpml1	0	-1944	0.12	54.95	0.00	0.02	6.13	0.05	10.37	13.99	6.49	8.89	99.50	0.0033	1.976	0.0005	0.0036	0.435	0.0015	0.439	0.539	0.15	0.476	4.0134	46.27	36.86	12.88	0.76
Line 2 4-01_Cpml1	211	-1734	0.15	54.82	0.00	0.02	6.09	0.12	10.22	14.16	5.04	8.73	99.34	0.004	1.977	0.0005	0.0036	0.435	0.0033	0.435	0.547	0.152	0.469	4.0138	46.43	40.15	13.01	0.76
Line 3 4-01_Cpml1	442	-1502	0.13	54.97	0.00	0.01	5.94	0.09	10.04	14.39	4.92	9.26	99.74	0.0034	1.974	0.0003	0.0036	0.434	0.0024	0.425	0.554	0.148	0.495	4.0161	46.28	41.35	12.36	0.77
Line 4 4-01_Cpml1	647	-1298	0.16	54.87	0.00	0.01	5.90	0.06	10.09	14.41	5.10	8.89	99.50	0.0044	1.976	0.0004	0.0036	0.438	0.0028	0.438	0.556	0.154	0.477	4.0096	46.84	40.19	12.97	0.76
Line 5 4-01_Cpml1	862	-1082	0.19	54.21	0.00	0.01	6.24	0.04	10.47	13.52	5.02	9.27	99.63	0.0033	1.975	0.0004	0.0036	0.435	0.0012	0.431	0.545	0.152	0.477	4.0221	45.83	38.42	12.85	0.76
Line 6 4-01_Cpml1	1078	-866	0.11	55.44	0.00	0.00	6.22	0.06	10.02	14.41	4.88	8.89	99.63	0.0033	1.984	0.0011	0.0036	0.431	0.0016	0.423	0.553	0.146	0.474	4.0127	45.14	40.41	12.45	0.76
Line 7 4-01_Cpml1	1293	-651	0.13	55.36	0.00	0.03	6.29	0.06	10.06	14.40	4.88	8.87	100.07	0.0034	1.982	0.0008	0.0036	0.436	0.0017	0.425	0.552	0.146	0.473	4.0199	45.14	40.39	12.47	0.76
Line 8 4-01_Cpml1	1532	-412	0.10	55.35	0.00	0.01	6.15	0.07	9.85	14.35	4.98	8.90	99.77	0.0027	1.987	0.0003	0.0036	0.428	0.0021	0.417	0.552	0.15	0.476	4.0352	46.86	40.41	12.73	0.76
Line 9 4-01_Cpml1	1748	-207	0.14	55.43	0.00	0.01	6.07	0.06	10.19	14.18	4.97	8.57	99.61	0.0037	1.99	0.0002	0.0036	0.431	0.0016	0.431	0.545	0.149	0.489	4.0034	47.27	39.81	12.92	0.75
Line 10 4-01_Cpml1	1930	0	0.14	55.32	0.00	0.01	6.20	0.07	10.28	14.28	5.04	8.61	99.65	0.0038	1.982	0.0003	0.0036	0.431	0.0021	0.434	0.548	0.151	0.46	4.0319	47.28	39.69	13.03	0.75
SEC4-01 Average			0.13	55.13	0.00	0.01	6.12	0.07	10.13	14.28	4.98	8.85	99.70	0.003	1.98	0.000	0.00	0.43	0.00	0.43	0.55	0.15	0.47	4.01	46.86	40.88	12.77	0.76
Line 1 SEC4-03_cpmlh1	0	-210	0.21	54.26	0.00	0.03	6.19	0.00	10.38	13.85	6.15	8.37	99.45	0.0059	1.965	0.0001	0.0036	0.434	0.0001	0.443	0.538	0.186	0.452	4.0251	45.75	38.44	15.82	0.71
Line 2 SEC4-03_cpmlh1	23	-187	0.20	54.38	0.00	0.03	6.21	0.03	10.42	13.82	6.18	8.43	99.45	0.0053	1.964	0.0011	0.0036	0.435	0.0008	0.444	0.535	0.187	0.454	4.0262	45.49	38.61	15.90	0.71
Line 3 SEC4-03_cpmlh1	47	-164	0.17	54.45	0.00	0.02	6.27	0.02	10.41	13.94	6.22	8.48	99.34	0.0045	1.962	0.0008	0.0036	0.438	0.0004	0.442	0.539	0.188	0.456	4.0308	45.56	38.75	15.89	0.71
Line 4 SEC4-03_cpmlh1	70	-141	0.21	54.35	0.00	0.04	6.08	0.02	10.46	13.96	6.18	8.41	99.74	0.0058	1.963	0.0002	0.0036	0.435	0.0007	0.445	0.54	0.187	0.454	4.0221	45.72	38.44	15.83	0.71
Line 5 SEC4-03_cpmlh1	94	-117	0.21	54.36	0.00	0.02	5.94	0.04	10.35	13.95	6.23	8.52	99.62	0.0056	1.965	0.0007	0.0036	0.436	0.0011	0.441	0.54	0.188	0.459	4.0329	45.77	38.64	15.84	0.71
Line 6 SEC4-03_cpmlh1	117	-94	0.19	54.22	0.00	0.03	6.29	0.06	10.30	13.94	6.17	8.41	99.61	0.0051	1.963	0.0011	0.0036	0.441	0.0017	0.439	0.541	0.187	0.454	4.0329	45.77	38.44	15.82	0.71
Line 7 SEC4-03_cpmlh1	138	-73	0.24	54.20	0.00	0.03	6.04	0.03	10.46	13.93	6.27	8.53	99.73	0.0065	1.958	0.0011	0.0036	0.445	0.0009	0.445	0.539	0.189	0.459	4.0251	45.41	38.67	15.92	0.71
Line 8 SEC4-03_cpmlh1	164	-47	0.19	54.21	0.00	0.01	6.24	0.03	10.47	14.09	6.23	8.56	100.03	0.0051	1.955	0.0004	0.0036	0.436	0.0009	0.445	0.545	0.188	0.46	4.0354	45.48	38.56	15.76	0.71
Line 9 SEC4-03_cpmlh1	187	-24	0.21	54.10	0.00	0.05	6.18	0.02	10.33	14.00	6.33	8.49	99.69	0.0057	1.958	0.0014	0.0036	0.434	0.0006	0.441	0.543	0.191	0.458	4.0328	45.55	38.42	16.02	0.71
Line 10 SEC4-03_cpmlh1	210	0	0.20	53.95	0.00	0.03	6.59	0.03	10.33	13.87	6.66	8.35	100.01	0.0056	1.953	0.0008	0.0036	0.442	0.0009	0.441	0.538	0.202	0.45	4.0353	45.21	37.82	16.97	

Supplementary Table 3. Phengite major

Line	Cumulative Dist μm	Dist to grain boundary (μm)	Weight %												Total	H ₂ O (100 minus sum)	F $\mu\text{g/g}$	Cl $\mu\text{g/g}$
			TiO ₂	F	Cl	MnO	Na ₂ O	Cr ₂ O ₃	SiO ₂	K ₂ O	FeO	MgO	Al ₂ O ₃	CaO				
Line 1 SEC42-06_Phen1	0	-123	1.04	0.00	0.00	0.00	1.27	0.00	47.68	8.89	2.60	2.45	29.31	0.01	93.26	6.74	-	-
Line 2 SEC42-06_Phen1	31	-92	0.97	0.00	0.00	0.00	1.08	0.03	47.59	8.87	2.54	2.44	29.35	0.00	92.88	7.12	-	-
Line 3 SEC42-06_Phen1	61	-62	1.05	0.00	0.01	0.00	1.23	0.05	47.54	8.95	2.53	2.39	29.45	0.00	93.19	6.81	-	-
Line 4 SEC42-06_Phen1	91	-32	0.99	0.00	0.01	0.00	1.18	0.02	47.87	8.87	2.50	2.47	29.56	0.00	93.47	6.53	-	82
Line 5 SEC42-06_Phen1	123	0	0.92	0.00	0.01	0.00	1.43	0.03	47.98	8.77	2.51	2.40	29.42	0.02	93.49	6.51	-	106
SEC42-06 Average			0.99	0.00	0.01	0.00	1.24	0.03	47.73	8.87	2.54	2.43	29.42	0.01	93.26	6.74		
Line 1 43-01_Phen1	0	-1098	0.92	0.00	0.01	0.01	1.15	0.06	48.19	8.87	1.72	2.65	29.43	0.01	93.02	6.98	-	71
Line 2 43-01_Phen1	121	-977	0.76	0.06	0.01	0.01	0.98	0.07	48.98	9.26	1.86	2.87	28.76	0.00	93.61	6.39	648	-
Line 3 43-01_Phen1	243	-855	0.74	0.02	0.00	0.00	0.96	0.05	49.06	9.22	1.87	2.91	28.37	0.00	93.20	6.80	-	-
Line 4 43-01_Phen1	390	-708	0.78	0.02	0.00	0.02	1.00	0.07	49.12	9.30	1.98	3.00	28.60	0.00	93.90	6.10	-	-
Line 5 43-01_Phen1	485	-613	0.68	0.07	0.01	0.00	0.95	0.11	48.83	9.22	1.87	3.05	28.66	0.00	93.45	6.55	668	102
Line 6 43-01_Phen1	609	-489	0.62	0.01	0.01	0.01	1.07	0.08	48.98	9.23	1.87	3.02	28.63	0.00	93.53	6.47	-	125
Line 7 43-01_Phen1	760	-338	0.81	0.00	0.01	0.01	1.04	0.04	48.98	9.30	1.99	2.92	28.75	0.01	93.84	6.16	-	63
Line 8 43-01_Phen1	855	-243	0.90	0.00	0.01	0.00	1.07	0.06	48.42	9.20	1.89	2.75	29.02	0.00	93.33	6.67	-	114
Line 9 43-01_Phen1	977	-121	0.96	0.04	0.00	0.00	1.18	0.05	48.13	9.08	1.90	2.64	29.43	0.00	93.41	6.59	-	-
Line 10 43-01_Phen1	1098	0	0.88	0.00	0.01	0.00	1.29	0.02	48.01	8.93	1.77	2.56	29.84	0.00	93.30	6.70	-	71
SEC43-01 Average			0.80	0.02	0.01	0.01	1.07	0.06	48.67	9.16	1.87	2.84	28.95	0.00	93.46			
Line 1 SEC47-01_Phen1	0	-1108	0.75	0.05	0.00	0.00	1.20	0.06	48.42	9.09	2.32	2.83	29.1	0	93.86	6.14	519	-
Line 2 sec47-01_Phen1	277	-831	0.73	0.00	0.00	0.00	1.06	0.05	48.4	9.18	2.36	2.92	28.9	0	93.58	6.42	-	-
Line 3 sec47-01_Phen1	553	-555	0.69	0.04	0.00	0.01	1.01	0.03	48.77	9.13	2.28	2.85	29.0	0.0054	93.78	6.22	434	-
Line 4 sec47-01_Phen1	863	-244	0.75	0.00	0.00	0.00	1.12	0.01	48.76	9.15	2.36	2.76	29.3	0.0107	94.25	5.75	-	-
Line 5 sec47-01_Phen1	1108	0	0.69	0.00	0.00	0.00	1.09	0.07	48.66	9.21	2.24	2.73	29.3	0	93.96	6.04	-	-
Line 1 SEC47-01_Phen2	0	-305	0.64	0.00	0.00	0.00	1.14	0.07	48.3	9.18	2.37	2.92	28.5	0	93.15	6.85	-	-
Line 2 SEC47-01_Phen2	29	-275	0.58	0.00	0.00	0.01	0.97	0.06	49.13	9.28	2.42	3.05	28.2	0.0061	93.67	6.33	-	-
Line 3 SEC47-01_Phen2	68	-237	0.62	0.00	0.00	0.00	1.03	0.05	48.94	9.34	2.46	3.11	28.1	0	93.59	6.41	-	-
Line 4 SEC47-01_Phen2	101	-204	0.63	0.00	0.00	0.01	1.04	0.04	49.08	9.32	2.31	3.01	28.4	0.0097	93.84	6.16	-	-
Line 5 SEC47-01_Phen2	136	-169	0.53	0.00	0.00	0.00	1.00	0.03	48.98	9.13	2.47	3.36	27.9	0.0286	93.44	6.56	-	-
Line 6 SEC47-01_Phen2	169	-135	0.57	0.00	0.00	0.01	0.94	0.06	49.15	9.41	2.47	3.04	28.3	0	93.91	6.09	-	-
Line 7 SEC47-01_Phen2	203	-102	0.59	0.00	0.00	0.00	0.89	0.06	49.15	9.38	2.43	3.05	28.3	0	93.81	6.19	-	-
Line 8 SEC47-01_Phen2	237	-68	0.58	0.00	0.00	0.00	0.94	0.06	49.23	9.47	2.53	2.98	28.1	0	93.89	6.11	-	-
Line 9 SEC47-01_Phen2	270	-34	0.53	0.00	0.00	0.02	1.02	0.07	48.15	8.98	3.94	2.74	28.3	0.04	93.82	6.18	-	-
Line 10 SEC47-01_Phen2	305	0	0.63	0.00	0.00	0.00	1.05	0.06	48.01	9.01	2.65	2.44	29.8	0.0097	93.64	6.36	-	-
SEC47-01 Average			0.63	0.01	0.00	0.00	1.03	0.05	48.74	9.22	2.51	2.92	28.62	0.01	93.75			

Supplementry Table 4. Amphibole major element composition

Line	Oxide (wt %)										Weight %										Atoms per formula unit										Formula
	TiO2	F	Cl	MeO	Na2O	CaO	SiO2	K2O	FeO	MgO	Al2O3	CrO	Total	H2O	Ti	F	Cl	Me	Na	Ca	Si	K	Fe	Mg	Al	Cr	Total	Species			
Line 1 SEC40-06 Amph1	0	0.370	0.019	0.018	0.000	0.008	44.17	0.70	13.73	10.06	14.82	0.72	97.33	2.67	0.076	0.086	0.001	0.020	1.190	0.005	6.513	0.132	1.035	2.212	2.577	1.377	15.886				
Line 2 SEC40-06 Amph1	112	-208	0.72	0.14	0.000	0.006	3.94	0.20	45.16	0.69	11.30	10.79	15.69	0.76	97.48	2.12	0.079	0.066	0.001	0.007	1.221	0.003	6.626	0.129	1.022	2.199	2.268	1.409	15.800		
Line 3 SEC40-06 Amph1	172	-197	0.60	0.12	0.001	0.004	4.28	0.03	44.28	0.41	12.70	10.30	14.36	0.43	95.75	4.25	0.067	0.054	0.004	0.005	1.236	0.003	6.593	0.115	1.582	2.287	2.521	1.345	15.811		
Line 4 SEC40-06 Amph1	173	-196	0.64	0.13	0.001	0.005	4.10	0.00	43.52	0.60	12.21	10.23	14.20	0.39	95.89	6.11	0.073	0.061	0.002	0.006	1.205	0.000	6.596	0.117	1.547	2.311	2.536	1.331	15.779		
Line 5 SEC40-06 Amph1	180	-190	0.63	0.17	0.001	0.007	4.17	0.03	44.76	0.63	13.22	10.38	14.33	0.62	97.09	2.91	0.072	0.079	0.004	0.009	1.192	0.004	6.563	0.118	1.628	2.279	2.526	1.360	15.831		
Line 6 SEC40-06 Amph1	218	-151	0.67	0.09	0.001	0.007	4.16	0.02	44.84	0.65	13.11	10.68	13.99	0.55	97.34	2.66	0.074	0.040	0.002	0.006	1.184	0.003	6.587	0.132	1.607	2.334	2.466	1.342	15.779		
Line 7 SEC40-06 Amph1	264	-106	0.71	0.30	0.001	0.004	4.33	0.04	44.57	0.67	13.06	10.34	14.03	0.51	97.41	2.99	0.078	0.047	0.002	0.005	1.230	0.005	6.532	0.126	1.605	2.239	2.996	1.337	15.82		
Line 8 SEC40-06 Amph1	312	-58	0.71	0.11	0.001	0.006	4.35	0.02	44.64	0.67	12.95	10.28	14.99	0.46	96.99	3.01	0.078	0.049	0.002	0.006	1.182	0.003	6.560	0.126	1.592	2.232	2.291	1.322	15.77		
Line 9 SEC40-06 Amph1	339	-31	0.67	0.11	-	0.006	4.33	0.04	45.05	0.63	13.08	10.40	14.43	0.54	97.34	2.88	0.074	0.051	0.000	0.007	1.233	0.005	6.612	0.118	1.605	2.275	2.497	1.312	15.79		
Line 10 SEC40-06 Amph1	370	0	0.33	0.10	0.000	0.004	4.24	0.05	47.93	0.34	12.82	11.43	11.94	0.19	95.43	2.57	0.037	0.044	0.001	0.005	1.194	0.006	6.977	0.064	1.556	1.473	2.044	1.324	15.66		
SEC40-06 Average		0.64	0.12	0.01	0.06	4.18	0.03	44.68	0.62	13.02	10.49	14.24	8.49	96.78	3.22	0.071	0.058	0.002	0.007	1.196	0.004	6.614	0.117	1.604	2.304	2.475	1.341	15.79 Na-Ca	hombite		
Line 1 SEC41-01 Amph1	0	-456	0.59	0.28	-	0.004	3.98	0.05	45.32	0.63	11.29	10.86	15.39	0.89	97.28	2.72	0.065	0.131	0.000	0.005	1.120	0.002	6.588	0.116	1.373	2.354	2.627	1.385	15.77		
Line 2 SEC41-01 Amph1	53	-405	0.60	0.23	0.000	0.005	4.06	0.05	44.60	0.64	11.33	11.24	15.18	0.56	96.44	3.36	0.066	0.108	0.001	0.006	1.155	0.006	6.538	0.119	1.389	2.456	2.621	1.361	15.83		
Line 3 SEC41-01 Amph1	202	-354	0.68	0.20	-	0.004	4.10	0.02	44.66	0.71	11.39	11.11	15.56	0.77	97.24	2.76	0.074	0.090	0.000	0.005	1.157	0.002	6.508	0.132	1.386	2.414	2.674	1.350	15.82		
Line 4 SEC41-01 Amph1	152	-304	0.66	0.13	0.000	0.004	4.13	0.06	44.62	0.69	11.23	11.12	15.55	0.63	96.87	3.13	0.072	0.060	0.001	0.005	1.170	0.007	6.517	0.129	1.371	2.420	2.677	1.350	15.78		
Line 5 SEC41-01 Amph1	203	-253	0.65	0.09	-	0.005	4.03	0.04	44.58	0.72	11.00	11.13	15.50	0.66	96.44	3.56	0.072	0.040	0.000	0.006	1.144	0.004	6.528	0.134	1.347	2.430	2.676	1.359	15.74		
Line 6 SEC41-01 Amph1	253	-202	0.69	0.18	-	0.005	4.10	0.04	44.24	0.72	10.95	11.12	15.33	0.53	96.57	3.70	0.077	0.086	0.000	0.006	1.167	0.005	6.580	0.135	1.343	2.433	2.693	1.368	15.81		
Line 7 SEC41-01 Amph1	304	-152	0.69	0.11	-	0.005	4.06	0.05	44.39	0.69	10.85	11.16	15.33	0.78	96.34	3.66	0.077	0.049	0.000	0.006	1.154	0.006	6.509	0.139	1.330	2.438	2.684	1.380	15.76		
Line 8 SEC41-01 Amph1	355	-101	0.67	0.20	0.000	0.005	4.26	0.00	44.55	0.71	11.09	11.20	15.36	0.66	96.77	3.23	0.074	0.094	0.001	0.007	1.208	0.001	6.521	0.133	1.337	2.443	2.659	1.358	15.81		
Line 9 SEC41-01 Amph1	405	-51	0.69	0.13	0.001	0.005	4.17	0.03	44.81	0.63	11.03	11.38	15.02	0.68	96.20	3.80	0.065	0.090	0.002	0.007	1.190	0.001	6.544	0.118	1.536	2.494	2.602	1.368	15.81		
Line 10 SEC41-01 Amph1	456	0	0.61	0.13	0.000	0.004	4.44	0.04	44.48	0.68	11.06	11.53	14.80	0.74	96.47	3.53	0.067	0.090	0.000	0.006	1.263	0.005	6.532	0.132	1.338	2.524	2.661	1.376	15.87		
SEC41-01 Average		0.64	0.17	0.00	0.04	4.13	0.03	44.60	0.67	11.12	11.19	15.34	8.71	96.45	3.35	0.071	0.078	0.000	0.006	1.173	0.004	6.529	0.126	1.361	2.441	2.648	1.367	15.80 Na-Ca	hombite		
Line 1 SEC41-01 Amph1	0	-440	0.58	0.12	-	0.002	4.98	0.02	45.88	0.28	10.94	12.49	14.29	0.63	97.61	2.39	0.063	0.054	0.000	0.002	1.229	0.002	6.622	0.130	2.087	2.400	2.332	1.570	15.769		
Line 2 SEC41-01 Amph1	35	-405	0.69	0.08	-	0.003	4.27	-	45.01	0.28	11.5	12.06	15.4	0.64	97.56	2.04	0.075	0.035	0.000	0.004	1.194	0.000	6.491	0.082	1.386	2.594	2.617	1.335	15.726		
Line 3 SEC41-01 Amph1	95	-409	0.70	0.06	-	0.002	4.07	0.03	45.33	0.31	11.39	11.86	15.22	0.41	97.80	2.20	0.076	0.027	0.000	0.003	1.250	0.003	6.540	0.058	1.374	2.530	2.389	1.300	15.795		
Line 4 SEC41-01 Amph1	137	-403	0.74	0.04	-	0.006	4.34	0.02	45.28	0.31	11.16	12.03	15.22	0.46	97.62	2.35	0.080	0.020	0.000	0.003	1.251	0.002	6.514	0.056	1.347	2.588	2.389	1.308	15.786		
Line 5 SEC41-01 Amph1	142	-408	0.61	0.08	-	0.006	3.63	0.03	46.47	0.31	11.42	12.72	13.12	0.15	97.78	2.22	0.066	0.035	0.000	0.007	1.009	0.004	6.728	0.057	1.377	2.373	2.229	1.413	15.679		
Line 6 SEC41-01 Amph1	227	-422	0.69	0.11	-	0.002	4.27	0.03	44.97	0.34	11.48	12.07	15.32	0.49	97.36	2.24	0.075	0.049	0.000	0.003	1.196	0.002	6.500	0.062	1.387	2.400	2.608	1.356	15.799		
Line 7 SEC41-01 Amph1	272	-367	0.72	0.08	-	0.005	4.22	0.03	45.18	0.37	11.3	12.09	15.21	0.49	97.59	2.03	0.078	0.035	0.000	0.006	1.264	0.003	6.513	0.087	1.362	2.597	2.502	1.324	15.831		
Line 8 SEC41-01 Amph1	320	-320	0.63	0.16	-	0.003	4.32	0.03	44.76	0.36	11.64	11.17	15.07	0.64	97.90	2.10	0.069	0.072	0.000	0.002	1.211	0.001	6.473	0.067	1.408	2.523	2.471	1.339	15.831		
Line 9 SEC41-01 Amph1	366	-274	0.75	0.05	-	0.003	4.38	-	44.97	0.36	11.79	12.08	15.2	0.47	96.88	1.92	0.082	0.025	0.000	0.003	1.226	0.000	6.489	0.066	1.423	2.599	2.586	1.309	15.804		
Line 10 SEC41-01 Amph1	411	-229	0.72	0.16	-	0.003	4.46	0.00	44.75	0.34	11.71	11.91	15.49	0.52	98.09	1.91	0.079	0.075	0.000	0.004	1.249	0.000	6.464	0.062	1.414	2.566	2.633	1.319	15.803		
Line 11 SEC41-01 Amph1	466	-174	0.64	0.01	0.000	0.006	4.35	0.00	43.87	0.37	12.3	11.54	15.81	0.4	97.46	2.14	0.070	0.086	0.000	0.003	1.230	0.000	6.395	0.069	1.481	2.527	2.764	1.343	15.821		
Line 12 SEC41-01 Amph1	506	-134	0.71	0.02	0.001	0.002	4.42	0.01	44.49	0.34	12.04	11.58	15.12	0.83	98.00	2.20	0.078	0.030	0.001	0.004	1.244	0.001	6.415	0.062	1.440	2.634	2.611	1.342	15.809		
Line 13 SEC41-01 Amph1	551	-89	0.69	0.09	0.000	0.005	4.24	0.03	44.25	0.34	12.44	11.42	15.79	0.54	97.88	2.12	0.075	0.042	0.001	0.007	1.193	0.003	6.424	0.064	1.530	2.472	2.302	1.328	15.818		
Line 14 SEC41-01 Amph1	596	-43	0.64	0.05	0.001	0.005	4.46	0.03	44.14	0.30	12.37	11.57	15.49	0.56	97.46	2.14	0.070	0.022	0.001	0.003											

Supplementary Table 5. Apatite Compositions (EMPA)

			Weight %									
	# grains	# measurements	P2O5	1 s.d. (%)	F	1 s.d. (%)	CaO	1 s.d. (%)	Cl	1 s.d. (%)	Total	OH
SEC 42-06	5	23	40.94	0.8%	2.97	7.7%	53.94	0.8%	0.051	16%	97.89	2.11
SEC 43-01	5	27	41.62	1.5%	3.25	3.2%	53.52	1.1%	0.041	75%	98.44	1.56
SEC 43-03	3	10	41.93	0.6%	2.15	4.4%	52.46	1.2%	0.019	54%	96.56	3.44
SEC46-01	3	9	42.02	0.6%	1.47	5.6%	52.19	0.5%	0.020	14%	95.70	4.30
SEC46-02	3	9	42.00	0.5%	1.49	6.6%	52.39	0.3%	0.030	13%	95.91	4.09
SEC47-01	3	9	41.92	0.7%	2.34	5.7%	51.64	0.3%	0.029	18%	95.93	4.07

Supplementary Table 6. Titanite major elem

Comment	No.	Weight %											
		TiO2	F	K2O	Cl	Na2O	Cr2O3	SiO2	CaO	MnO	MgO	Al2O3	FeO
Line 1 SEC46-02_titanite	46	36.97	0	0.012	0.0092	0.012	0.11	29.65	28.38	0.0454	0.01	1.84	0.20
Line 2 SEC46-02_titanite	47	36.78	0	0.014	0.0124	0.021	0.10	29.91	28.23	0.0285	0.00	1.85	0.22
Line 3 SEC46-02_titanite	48	36.48	0	0.014	0.0038	0.097	0.08	29.83	28.3	0.0310	0.00	2.07	0.20
Line 4 SEC46-02_titanite	49	36.96	0	0.012	0.0065	0.044	0.07	29.89	28.26	0.0060	0.00	1.79	0.21
Line 5 SEC46-02_titanite	50	36.91	0	0.015	0.0086	0.025	0.07	29.73	28.18	0.0320	0.00	1.78	0.28
Average		36.82	0	0.013	0.0081	0.040	0.09	29.802	28.27	0.0286	0.00	1.87	0.22

Total
97.24
97.17
97.11
97.25
97.03
97.16

Supplementary Table 7. Halogen intra-grain profiles (SIMS)

SEC43-01												
Garnet	SEC43-01_grt_1@44	SEC43-01_grt_07@50	SEC43-01_grt_3@46	SEC43-01_grt_06@49	SEC43-01_grt_04@47	SEC43-01_grt_5@48	SEC43-01_grt_2@45			Grain Mean	Grain S.D. (%)	
OH $\mu\text{g/g}$	114	118	316	248	104	144	173			174	46%	
OH Error % (2SE Int+Calib)	5.4%	5.0%	4.8%	37.1%	5.1%	5.8%	6.2%					
F $\mu\text{g/g}$	7.25	8.56	9.51	8.17	6.29	3.66	8.74			7.45	26%	
F Error % (2SE Int+Calib)	6.0%	5.8%	5.9%	6.3%	6.2%	5.9%	6.1%					
S $\mu\text{g/g}$	-	0.04	0.06	0.05	0.05	0.07	-					
C Error % (2SE Int+Calib)	-	13.8%	17.7%	17.1%	12.9%	16.0%	-					
Cl $\mu\text{g/g}$	0.18	0.17	0.24	0.20	0.19	0.20	0.25			0.20	14%	
Cl Error % (2SE Int+Calib)	13.0%	16.1%	13.4%	13.8%	13.5%	13.8%	14.8%					
Distance (μm)	722	546	465	360	290	117	0					
	Core						Rim					
Omphacite												
Omphacite	SEC43-01_omph_1@70	SEC43-01_omph_3@72	SEC43-01_omph_5@74	SEC43-01_omph_7@76	SEC43-01_omph_8@77	SEC43-01_omph_9@78	SEC43-01_omph_6@75	SEC43-01_omph_4@73	SEC43-01_omph_2@71			
OH $\mu\text{g/g}$	362	437	412	557	435	385	414	414	383	422	13%	
OH Error % (2SE Int+Calib)	4.7%	4.6%	4.6%	5.6%	4.7%	4.7%	4.8%	4.7%	4.8%			
F $\mu\text{g/g}$	42.1	55.5	57.1	59.3	69.7	60.2	62.1	48.3	52.1	56.3	14%	
F Error % (2SE Int+Calib)	5.8%	5.8%	5.8%	5.8%	5.8%	5.9%	5.9%	5.8%	5.9%			
S $\mu\text{g/g}$	0.03	0.03	-	0.03	0.02	-	0.04	-	-			
C Error % (2SE Int+Calib)	13.4%	16.0%	-	17.4%	13.7%	-	12.2%	-	-			
Cl $\mu\text{g/g}$	0.12	0.11	0.13	0.15	-	-	0.12	0.12	-	0.13	11%	
Cl Error % (2SE Int+Calib)	16.2%	15.1%	12.2%	14.7%	-	-	13.8%	13.5%	-			
Distance (μm)	0	227	343	502	604	724	902	1082	1262			
	Rim								Core			
Phengite												
Phengite	SEC43-01_phen_2@52	SEC43-01_phen_3@53	SEC43-01_phen_5@55	SEC43-01_phen_4@54	SEC43-01_phen_1@51	SEC43-01_phen_9@63	SEC43-01_phen_8@62	SEC43-01_phen_7@61	SEC43-01_phen_6@60			
OH $\mu\text{g/g}$	26335	28580	28337	28820	28304	28840	27286	28857	29884	28360	4%	
OH Error % (2SE Int+Calib)	4.6%	4.5%	4.7%	4.6%	4.5%	4.7%	4.6%	4.6%	4.6%			
F $\mu\text{g/g}$	1575	1819	1943	1979	1938	1913	1769	1883	1578	1822	8%	
F Error % (2SE Int+Calib)	5.8%	5.8%	6.0%	5.9%	5.8%	5.9%	5.9%	5.9%	5.9%			
S $\mu\text{g/g}$	0.90	0.53	0.42	0.43	0.45	0.58	0.52	0.50	0.51			
C Error % (2SE Int+Calib)	7.4%	7.4%	10.0%	6.9%	8.3%	6.7%	8.1%	7.0%	6.7%			
Cl $\mu\text{g/g}$	1.50	0.96	1.15	1.12	1.08	1.08	0.97	0.98	1.58	1.16	20%	
Cl Error % (2SE Int+Calib)	12.5%	12.1%	13.9%	12.6%	12.2%	12.6%	12.2%	12.2%	12.8%			
Distance (μm)	0	254	386	505	616	855	927	992	1086			
	Rim, to grt grain boundary				Core		Rim, to omphacite grain boundary					

Supplementary Table 9. Pyrohydrolysis raw data and reference material replicates

Analysis #	Uncorrected for Yield				Corrected for Yield				n	
	F (pp/g)	Cl (pp/g)	Average	Std. Dev.	F (pp/g)	Cl (pp/g)	Average	Std. Dev.		
SEC42-6	2	99.74	10.00	91.94	8.85	130.98	13.13	114.56	11.04	1
SEC42-6'	3	84.72	8.35	7.53	1.00	102.35	10.09	14.77	1.81	2
	4	91.34	8.20	8%	11%	110.35	9.90	12.89%	16%	2
SEC43-1	2	163.91	9.27	108.51	7.05	215.24	12.17	136.83	8.85	1
SEC43-1'	3	77.69	6.40	48.08	1.97	93.86	7.73	68.01	2.95	2
	4	83.92	5.49	44%	28%	101.39	6.64	49.71%	33.12%	2
SEC43-3	2	151.98	6.73	152.25	5.37	199.58	8.84	189.25	6.73	1
SEC43-3'	3	148.65	5.44	3.74	1.40	179.58	6.57	10.02	2.04	2
	4	156.12	3.94	2%	26%	188.61	4.76	5.3%	30.4%	2
SEC46-1	2	192.26	11.03	154.37	9.06	252.47	14.49	193.23	11.34	1
SEC46-1'	3	128.56	7.56	33.52	1.78	155.31	9.14	51.97	2.80	2
	4	142.30	8.59	22%	20%	171.91	10.38	26.9%	24.7%	2
SEC46-2	2	162.16	15.16	160.25	16.06	212.95	19.90	199.26	19.94	1
SEC46-2'	3	156.39	16.95	3.35	0.90	188.93	20.47	12.35	0.52	2
	4	162.22	16.09	2%	6%	195.98	19.44	6%	3%	2
SEC47-1	2	113.07	7.62	111.60	7.10	148.48	10.01	138.78	8.84	1
SEC47-1'	3	108.15	7.03	3.00	0.49	130.65	8.49	9.02	1.04	2
	4	113.58	6.65	3%	7%	137.22	8.03	6%	12%	2

*apostrophe (') indicates a duplicated sample

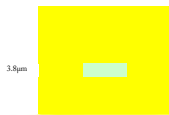
Sample	Uncorrected for Yield				Corrected for Yield by Batch							
	F (pp/g)	Cl (pp/g)	1SD Pos (yield uncert)		1SD		1SD Propagated		1SD Propagated			
SEC42-06	91.94	8.85	22.3%	23.0%	114.56	11.04	14.77	1.81	13.05%	19.91%	14.95	2.20
SEC43-01	108.51	7.05	49.0%	34.4%	136.83	8.85	68.01	2.95	49.75%	34.98%	68.07	3.09
SEC43-03	152.25	5.37	21.1%	32.8%	189.25	6.73	10.02	2.04	5.66%	32.39%	10.71	2.18
SEC46-01	154.37	9.06	30.2%	28.1%	193.23	11.34	51.97	2.80	26.91%	27.38%	52.00	3.10
SEC46-02	160.25	16.06	21.1%	20.8%	199.26	19.94	12.35	0.52	6.27%	12.08%	12.49	2.41
SEC47-01	111.60	7.10	21.1%	21.2%	138.78	8.84	9.02	1.04	6.57%	16.65%	9.11	1.47

Standards		
	F ppm	Cl ppm
JB-2-1	76.65	268.22
JB-2-2	73.70	215.25
JB-2-3	74.67	233.68
Batch 1 Mean	75.01	239.05
Batch 1 Std	1.50	26.89
Yield Batch 1	76.1%	85.1%
Yield Std (%)	2.0%	11.3%
JB-2-9	80.64	191.80
JB-2-11	82.04	208.73
JB-2-12	81.93	241.47
Batch 2 Mean	81.53	214.00
Batch 2 Std	0.78	25.25
Yield Batch 2	82.8%	76.2%
Yield Std (%)	1.0%	11.8%
Average (all)	78.27	226.53
Std. Dev. (all)	3.73	27.07
JB-2**	98.50	281.00
Yield (min)*	75%	68%
Yield (max)*	83%	95%
Yield (average)	79%	81%
STD ALL (average)	4%	5%

*means.html reports the F and Cl concentration of JB-2 as 98.5ppm and 281ppm, respectively.

Supplementary Table 10. Grain Boundary Calculations

Grain boundary calculations with simso spot size of 3.8µm*2



Area (µm)	Ratio	CL GB meas	Calc	CI GB
3µm	0.019	0.00131579	17.65	13414
3.8 µm	14.44	6.5	4940	
Simso 3.8µm	0.019	0.00131579	17.65	13414

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Flux in Porous Media

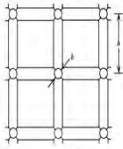


Figure 9.1 An idealized model of a porous medium. Circular tubes of diameter δ form a cubical matrix with dimensions b .

cube on each of these edges. Thus the equivalent of three tubes of diameter δ and length b lie within the cube. The porosity is therefore given by

$$\phi = 3\pi \left(\frac{\delta}{2}\right)^2 \frac{b}{b^3} = \frac{3\pi \delta^2}{4 b^2} \quad (9.6)$$

Tarcatte and Schubert 2002

Method 1	Simso Spot Size (µm)	Phi	Dolomite											
Est Phi GB	14.44	Phi	5.00E-04	5.00E-05	5.00E-06	5.00E-07	5.00E-08	5.00E-09	5.00E-10	5.00E-11	5.00E-12	5.00E-13		
Width			Increments of 10x											
	Grain bound. Area (µm)	Ratio	CL GB meas	Calc	CI GB	CI GB µg/g	CI GB µg/g	CI GB µg/g	CI GB µg/g	CI GB µg/g	CI GB µg/g	CI GB µg/g	CI GB µg/g	
	1.00E-03	0.0038	0.00026316	17.65	67070	33.54	3.35	0.34	0.03	0.00	0.00	0.00	0.00	
	5.00E-03	0.019	0.00131579	17.65	13414	6.71	0.67	0.07	0.01	0.00	0.00	0.00	0.00	
	2.50E-02	0.095	0.00657895	17.65	26828	1.34	0.13	0.01	0.00	0.00	0.00	0.00	0.00	
	1.25E-01	0.475	0.03289474	17.65	53656	0.27	0.03	0.00	0.00	0.00	0.00	0.00	0.00	
Increments of S_x	6.25E-01	2.375	0.16447368	17.65	107312	0.05	0.01	0.00	0.00	0.00	0.00	0.00	0.00	
	3.80E-00	14.44	1	17.65	17.65	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
	1.90E+01	72.2	5	17.65	3.53	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
	9.50E+01	361	25	17.65	0.706	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
	4.75E+02	1805	125	17.65	0.1412	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
delta	1.00E-03													
b	500 µm													
Phi	9.42E-12													
		%												
CI GB contribution (%)	#REF!													
	Delta µm	Phi												
Method 2	1mm	0.001	9.42E-12											
Calc Phi	5mm	0.005	2.36E-10											
from GB	25mm	0.025	5.89E-09											
	125mm	0.125	1.47E-07											
	625mm	0.625	3.68E-06											
	3.125µm	3.125	9.20E-05											
	15.625µm	15.625	2.30E-03											
	78.125µm	78.125	5.75E-02											
	390.625µm	390.625	1.44E-00											