Bimodal Alteration of the Oceanic Crust Revealed by Halogen and Noble Gas Systematics in the Oman Ophiolite

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Abstract  Hydrothermal alteration of oceanic crust represents a globally significant geochemical exchange between the crust and oceans, introducing volatile elements into the oceanic lithosphere and removing latent heat from crystallization. The extent to which hydrothermal systems penetrate the lower oceanic crust is debated and is significant for competing models of lower oceanic crust formation. In this study, we apply halogen and noble gas geochemistry in order to better understand the fluid sources and conditions during a hydrothermal alteration in a suite of gabbros sampled from drill cores in the mid- (hole GT3A) and lower- (hole GT1A) crust of the Oman Ophiolite. Low Br/Cl in mid-crustal samples is controlled by amphibole formed at relatively high fluid/rock ratios. Lower crustal samples, meanwhile, have Br/Cl ranging from seawater to much higher values (<5 x 10^-3). These require alteration at highly variable fluid/rock ratios from ~1 to <0.01 in the lower crust. Noble gas isotopes also suggest alteration at relatively high and low fluid/rock ratios in the mid- and lower-crust, respectively. Indicators of alteration and proxies for fluid/rock ratios (loss on ignition, Pb/ Ce, Br/Cl, 40Ar/36Ar) vary systematically with distance from major fault zones in the lower crust with the strongest correlation in the least altered minerals. Together with age constraints derived from K-Ar decay, these data indicate faults were major conduits, supplying seawater to the lower crust roughly contemporaneous with the formation of the ophiolite, and commencing at the magmatic stage. These results suggest that hydrothermal alteration of the lower crust is primarily controlled by major fault structures.

Plain Language Summary  After a new oceanic crust is formed by volcanic activity at mid-ocean ridges, it cools and reacts with circulating seawater. However, there is controversy over whether water penetrates to the lower oceanic crust, or whether this is cooled by conduction alone. This is important for theories of how the lower oceanic crust is formed. Investigating the lower crust is difficult since few samples have been recovered by deep seafloor drilling. However, ophiolites—sections of oceanic crust, uplifted onto continental margins during tectonic collisions—offer a potential window into processes in the deep oceanic crust. In this study, we use halogens and noble gases as chemical fingerprints for the origin of water in rocks from the Oman Ophiolite. We show that rocks from the mid-oceanic crust and from the lower-oceanic crust interacted with water under markedly different conditions and that the supply of water to the lower crust was very variable. Rocks close to large fractures cutting through the lower crust were altered by seawater ~96 Myr ago. Further from these fractures, compositions are increasingly different from seawater and water was scarce. This provides important evidence that large fractures delivered water to the lower crust and may be important for cooling new oceanic crust globally.

1. Introduction

Hydrothermal alteration of the oceanic crust and its eventual subduction into the mantle exerts a key control on the long-term evolution of the surface and interior chemistry of Earth. Subduction of volatile elements to the deep mantle may effectively remove them from the surface and sequester them at depth. This has important implications for Earth’s deep carbon cycle and the long-term salinity of the oceans (Jarrard, 2003; Kelemen & Manning, 2015; Kendrick et al., 2013).

The nature of alteration in the oceanic crust is intimately linked to models of lower crustal formation. Competing endmember models involving either: in-situ crystallization of the lower oceanic crust (the so-called “sheeted sills” model; e.g., Kelemen et al., 1997); or subsiding crystal mushes/cumulates from an axial magma chamber...
(the “gabbro glacier”; e.g., Phipps Morgan & Chen, 1993) have been longstanding topics of discussion, with the former requiring deep hydrothermal circulation to remove latent heat from the lower crust. Definitive support for either model is lacking, with many thermal models inconclusive (e.g., Maclellan et al., 2005) and suggestions of near-conductive cooling from several geospeedometry studies (Coogan et al., 2002; Faak et al., 2015; Faak & Gillis, 2016) contrasted by other geochemical proxies suggesting rapid cooling (e.g., Sun & Lissenberg, 2018; Van Tongeren et al., 2008) and by numerical modeling which reproduces coexisting shallow and deep hydrothermal circulation systems (Hasenclever et al., 2014). In recent years, increasing attention has focused on the role of major crustal faults as potential pathways for fluid delivery to the lower oceanic crust as a potential avenue to reconciling these conflicting observations (Abily et al., 2011; Coogan et al., 2006; Harris et al., 2017; Zihlmann et al., 2018).

Understanding how hydrous minerals, temperature, and fluid supply interact at depth to produce the volatile inventory of the lower crust, however, is hampered by the lack of samples recovered from in-situ oceanic crust. Scientific drilling has, to date, only penetrated the upper ∼2 km of oceanic crust. While samples recovered by dredging at transform zones or drilling of exhumed oceanic core complexes recover deeper material, the extent to which the volatile characteristics of these are modified by emplacement to shallower seafloor situations remains unclear (e.g., Kendrick, 2019b).

Ophiolites offer a complementary view to what has been achieved through oceanic drilling. They can preserve complete sections of oceanic lithosphere and allow for relatively easy sampling and excellent field control on sample context, compared, for example, to dredged samples.

In this study, we analyze halogen and noble gas abundances in a suite of gabbros from the upper/mid- and lower crust of the Oman Ophiolite, in order to critically evaluate the nature of hydrothermal circulation in the altered oceanic crust (AOC) and better quantify its volatile inventory. The Oman Ophiolite offers an ideal natural laboratory for this study. It is widely thought to offer a close analog to fast-spreading oceanic crust (Coogan, 2003; MacLeod & Yaouancq, 2000; Manning et al., 2000; McClain, 2003; Nicolas et al., 1994), which dominates the mass flux of global subduction zones (Van Keken et al., 2011). Furthermore, we are able to take advantage of recent scientific drilling (see Section 1.2) which avoids subaerial weathering processes and preservation biases that may affect material sampled at the surface. We show that the styles and extent of hydrothermal alteration are markedly different in upper/mid versus lower crust in Oman, attesting to a homogeneous and heterogeneous fluid supply, respectively. Using coupled analyses of halogen abundances, noble gas isotopes, and the K-Ar system, we are able to demonstrate that the bulk of the alteration was by hydrous fluids derived from seawater at ∼96 Ma. The extent of alteration and composition of fluids in the lower Oman crust is closely related to their proximity to major fault zones with progressive fractionation of fluids away from these zones of recharge during alteration at very low fluid/rock ratios. This provides strong chemical evidence for hydrothermal flow in the lower crust, commencing at the magmatic stage, along major permeable structures.

1.1. Geological Context

The Oman Ophiolite is the largest known ophiolite (and among the best-studied) in the world, encompassing 20,000 km² of exposed crustal and mantle lithologies, transtended by normal faults, but otherwise maintaining a complete oceanic crustal and mantle sequence (Figures 1a–1c). From top to bottom the ophiolite comprises sediment-draped pillow lavas (0.5–2.0 km); a sheeted-dyke complex (1–1.5 km); crustal gabbros and ultramafic cumulates (0.5–6.5 km); and mantle harzburgites (8–12 km). The combined mantle and crust section is inferred to have been 15–20 km thick prior to obduction (Boudier & Coleman, 1981; Hacker et al., 1996; Manghnani & Coleman, 1981). Multiple geochronological methods give ages that converge on a range of 95–96 Ma for emplacement of the ophiolite onto the Arabian continental margin during Tethys Ocean closure (Rioux et al., 2013), only shortly after, or even synchronous with, the final stages of crustal crystallization (Hacker et al., 1996; Warren et al., 2005).

Based on a range of petrological, geochemical, and geochronological criteria there is broad consensus that the ophiolite formed in a supra-subduction zone setting and not at a true mid-ocean ridge (Alabaster et al., 1982; Gnos, 1998; Ishikawa et al., 2002; MacLeod et al., 2013; Rioux et al., 2013). That said, there are outstanding objections to significant subduction-related magmatism having affected the ophiolite. These include the contention that the ophiolite formed at a normal mid-ocean ridge and was emplaced by decollement of one limb over the
other at the spreading center (e.g., Arai & Miura, 2015; Boudier et al., 1985, 1988; Hacker et al., 1996). Lavas with clear island-arc tholeiite affinity are best preserved in the northernmost sections of the ophiolite, and though recent studies have argued for a later phase of arc-related intrusions in northern and central blocks (Goodenough et al., 2010; Styles et al., 2006), these appear to diminish southwards and are largely absent in the southernmost Wadi Tayin block (Goodenough et al., 2014).

Figure 1. Location map of the Oman Ophiolite, igneous stratigraphy of cores and sample locations. (a) Regional setting of the Oman ophiolite. (b) Geological map of the Semail and Wadi Tayin blocks showing the locations of holes GT1A and GT3A. (c) Generalised schematic log of the full ophiolite succession in Oman indicating the approximate original depths of holes GT1A and GT3A in the oceanic crust. (d) Igneous stratigraphic logs of cores from Hole GT3A and Hole GT1A. (e) Downhole plots of loss on ignition (LOI) from shipboard sampling (see Section 1.2). (f) Sample locations and lithologies for this study (a-c adapted from Kelemen et al. (2020) and Goodenough et al. (2014), petrological and LOI data from Kelemen et al. (2020)).
Evidence for hydrothermal alteration of the lower Oman crust has been explored in a number of studies resulting in a wide range of estimates for fluid/rock ratios from >400 to <0.5 (Bosch et al., 2004; Gregory & Taylor, 1981; McCulloch et al., 1981; Zihlmann et al., 2018) and in calculated cooling rates which range from close to conductive (Coogan et al., 2002) to those showing evidence for enhanced heat removal (Coogan et al., 2006). Recent work indicates that zones of enhanced permeability such as synmagmatic faults may be important conduits for both heat and fluid fluxes in the lower crust in Oman (Abily et al., 2011; Coogan et al., 2006; Zihlmann et al., 2018) and in the modern oceanic crust (e.g., Harris et al., 2017). However, conclusive evidence for a seawater origin of fluids and the timing of enhanced alteration has remained elusive.

1.2. The Oman Drilling Project

The Oman Ophiolite was recently sampled by diamond wireline coring as part of the Oman Drilling Project (International Continental Drilling Program Expedition 5057; Kelemen et al., 2020) yielding nine new cores that collectively span the full ophiolite succession from the upper crust to the lithospheric mantle and totaling 3,221 m of the core. This study focuses on two of the three crustal cores. The GT3A (drilled in Wadi Abdah in the Semail massif) core samples the sheeted-dyke to gabbro transition in the upper to the mid crust, while GT1A (drilled in Wadi Gideah in the southernmost Wadi Tayin massif) samples the lower crustal layered gabbros. The GT3A core is therefore approximately equivalent to the depths reached in IODP Hole 1256D in the Pacific ocean crust basement (Expedition 335 Scientists, 2012) whereas GT1A represents material from much deeper in the oceanic crust than has been drilled in situ to date (Figure 1c). The two southernmost massifs of the ophiolite are unaffected or little affected by later subduction-related magmatism and therefore offer a relatively close analog to crust formed at a normal mid-ocean ridge (Goodenough et al., 2014). Both cores extend from the surface to ~400 m depth with almost 100% recovery, including many soft mineralogical features typically weathered away in the arid desert environment at the surface. Note that where reference is made to shallower or deeper cores this is in reference to their original depth in the oceanic crust, which is estimated to have been ~2 and ~5 km for GT3A and GT1A, respectively (Kelemen et al., 2014).

The cores were curated and logged aboard the IODP drilling vessel Chikyu in Shimizu, Japan, during the summer of 2017. They are therefore well-characterized, and associated with a full suite of petrological, structural, and alteration logs, as well as spaced sampling for geochemistry, mineralogy, and paleomagnetic properties, while onboard. The full results of these analyses have been published as part of the proceedings of the project (Kelemen et al., 2020), and only a summary of pertinent features is given here.

The two cores show notable differences in a variety of characteristics. The petrological logs (Figure 1d) and geochemistry clearly indicate that the shallower GT3A core is compositionally more evolved overall than GT1A, with the former containing a variety of oxide gabbros, diorites, and tonalities intercalated with the gabbros, whereas the latter section is dominated by olivine gabbros and gabbros (Figure 1d).

The loss on ignition (LOI) for all shipboard samples correlates very strongly with H$_2$O contents determined by combustion in a CHNS Analyzer (ThermoFisher FLASH 2000), while CO$_2$ and H$_2$O show little or no correlation (Kelemen et al., 2020). This indicates that water dominates the volatile inventory and suggests that either the bulk of CO$_2$ is unrelated to hydrothermal alteration or that the addition of CO$_2$ is decoupled from that of water during hydration. The mean LOI in both holes is similar (~2%); however, GT3A shows a more restricted range of values while GT1A is more variably hydrated (Figure 1e). This is consistent with petrographic observations that very fresh gabbros, with few visible signs of secondary alteration, are common in GT1A (e.g., Figure S1e in Supporting Information S1).

The density of calcite veins decreases markedly downhole and roughly correlates with the percentage of CaCO$_3$ calculated from whole-rock geochemistry (Kelemen et al., 2020). There are two broad zones of intense calcite veining in the upper 100 m of GT3A; less abundant and more evenly distributed veins occur from 100-200 m and almost none are present below 200 m depth. Similar patterns are seen in the distribution of calcite veins in hole GT1A with abundant calcite veins recorded in the upper 50 m of the core, and sparse veins present between 100 and 350 m depth. This is taken as evidence that these veins are related to meteoric rather than seafloor processes.
1.3. Sample Selection and Preparation

Although Hole GT3A is dominantly formed of basalt and diabase dykes, especially in its upper 250 m, samples for this study were chosen exclusively from gabbro units in both cores (Figure 1f). The reasons for this were two-fold. Firstly, gabbros form the majority of the thickness of oceanic crust and so are likely to dominate the volatile inventory. Secondly, the use of samples from gabbro units only makes the results more comparable across the two cores, allowing differences to be more easily attributed to variations in the process rather than lithology. Samples were chosen to encompass the range of lithologies and alteration styles in each core (Figure 1 and Figure S1 in Supporting Information S1). Selected lithologies included gabbro, olivine gabbro, oxide-gabbro, oxide-olivine gabbro, troctolitic gabbro, and vari-textured gabbro. The extent of alteration spans a range from being almost fresh to being intensely altered. Full petrological details, together with photomicrographs illustrating key features, are provided in Text S1 and Figure S1 in Supporting Information S1.

2. Methods

Twelve samples from GT3A and GT1A were sawn into two adjacent slabs with one prepared for bulk geochemical analysis, and the other as a thin section for petrography and in-situ analyses. In addition to whole rock aliquots, five samples were hand-picked to obtain separates of their major constituent minerals resulting in a total of 28 whole rock and mineral separate samples. All analyses were undertaken at the University of Manchester. For all samples, whole-rocks and mineral separate alike, the proportions of constituent minerals were determined by powder X-Ray diffraction (XRD). Halogen abundances (Cl, Br, I) and heavy noble gas isotopes (Ar, Kr, Xe) were determined by neutron-irradiation noble gas mass-spectrometry (NI-NG-MS; Kendrick, 2012; Ruzié-Hamilton et al., 2016) using an ARGUS VI noble gas mass spectrometer following irradiation for 24 hr at the University of Missouri Research Reactor (MURR). All samples were analyzed by laser heating (giving bulk halogen abundances) while a subset of 12 samples was analyzed by crushing (releasing volatiles trapped in fluid inclusions or along grain boundaries; Wieler, 2013). A subset of 8 unirradiated whole-rock samples was analyzed for natural noble gas abundances and isotopes (He, Ne, Ar, Kr, Xe) by laser heating using a HELIX-MC multi-collector noble gas mass spectrometer. In-situ major elements (Si, Al, Fe, Ca, Na, Mg, K, Ti, Mn, and P) and Cl were measured in alteration minerals (amphibole, iddingsite, clays) in two samples by electron probe microanalysis (EPMA) using a Cameca SX100. Bulk water content was analyzed for all whole rock samples by gravimetry following ignition for 1 hr at 1,000°C. For full details of preparation and analytical procedures see Text S2 in Supporting Information S1. Analytical methods for the dataset of 140 shipboard samples from GT1A and GT3A are detailed in full by Kelemen et al. (2020), and a brief summary is given in Text S2 in Supporting Information S1.

Throughout this manuscript, unless otherwise stated, halogen and K abundance ratios are reported as wt./wt. and their concentrations as weight %, ppm, or ppb as appropriate; noble gas elemental ratios are reported as mol/mol and concentrations as cm³/g at standard temperature and pressure (STP). All errors are given to 1 S.D. unless otherwise stated. All of the data underpinning this manuscript (including XRD, EPMA, halogen, noble gas, and LOI datasets) are presented in Supplementary Tables S1-S8 and are available via Mendeley Data at https://doi.org/10.17632/v9gmsj5m59.2.

3. Results

3.1. Mineralogy of Primary and Alteration Phases

XRD spectra reveal a variety of fresh and alteration minerals in the whole rock samples, with clear differences between the two cores (Table S1). In GT1A gabbros, fresh minerals include augite and calcic plagioclase as major species and olivine as a minor constituent. Clinoclase was present in all samples, and lizardite in most, usually in association with olivine. Prehnite, epidote, and zeolites were only detected in one sample (GT1A 95z2). Amphibole was detected in four of the GT1A samples and, in those, ranges from 4-16 wt.% of the whole rock. Results from GT3A gabbros were similar but augite rarely exhibits a major peak suggesting it has undergone greater alteration. Clinoclase is always present as a minor species (3%–16%) and amphibole as a major species (18%–30%). Prehnite was detected in two samples (107z3 and 134z2, 5%–10%) and epidote in three samples (107z3, 134z3, 139z2; 15%–30%). No samples from GT3A showed resolvable zeolite peaks.
Where minerals were not fully identifiable by petrographic characteristics alone, they were identified or further subdivided on the basis of their chemistry and stoichiometry. Minerals investigated by EPMA included amphibole, thomsonite, iddingsite, prehnite, and albite. The full dataset is presented in Table S2. Figure S2 in Supporting Information S1 summarizes the Cl contents of each mineral determined in situ by EPMA and in bulk separates by the noble gas method together with literature data from Atlantis Bank gabbros (Kendrick, 2019b).

The amphibole is mostly magnesio-hornblende but extends to more Si-rich actinolitic and tremolitic compositions (Table S2). Iddingsite shows a continuous range of compositions from near-stoichiometric saponite toward lizardite, consistent with a microcrystalline mixture of the two. Some studies have considered saponite and iddingsite as separate minerals (Kendrick et al., 2019b). However, given the continuous range of compositions and the petrographic similarity of each of the grains analyzed we refer to all here as iddingsite.

3.2. Halogens and Noble Gases

3.2.1. EPMA Chlorine Determinations

Halogen contents of mineral phases from GT1A and GT3A are compared in Figure S2 in Supporting Information S1 and given in Table S3. The Cl contents of the potential halogen host phases show a wide range of concentrations from below detection limits (mean DI$_{Cl}$ = 170 ppm) to 4,000 ppm (Figure S2 in Supporting Information S1). Chlorine is relatively abundant in only three of the minerals investigated in this study: amphibole, iddingsite, and chloride, which show mean Cl contents of 1,210 ppm, 610, and ∼180 ppm, respectively. By contrast prehnite, thomsonite and albite all showed low Cl concentrations with the majority of EPMA spots giving results that were below detection limits. In the case of chloride, only 15% of the analyses reveal Cl above detection limits and the median Cl of all analyses (including those below detection limits) is somewhat less than the detection limit (80 ppm cf. ∼170 ppm). Using the approach of setting data below detection to half the detection limit gives a mean chloride Cl concentration of 180 ppm which we take as a best-guess estimate.

The findings above are consistent with published mineral data from Atlantis Bank gabbros, which showed similar ranges of Cl concentrations in all of the minerals investigated (Kendrick, 2019b; Figure S2 in Supporting Information S1). The biggest contrast between the Atlantis Bank data and the present dataset is in the Cl contents of iddingsite (means: CI$_{Atlantic}$ = 1,079 ± 861 ppm, CI$_{Uman}$ = 596 ± 374 ppm) and serpentine (means: CI$_{Atlantic}$ = 725 ± 236 ppm, CI$_{Uman}$ = 571 ± 123 ppm), with Atlantis Bank data extending to slightly higher concentrations for each. Where both in-situ EPMA data and bulk mineral separate data (by the noble gas method, see Section 3.2.2) were collected for our sample set (e.g., amphibole, albite), the resultant mean values are similar (Figure S2 in Supporting Information S1). The only exception is serpentine, which shows significantly lower concentrations in the bulk separates than in the in-situ analyses (CI$_{bulk}$ = 237 ± 137 ppm, CI$_{EPMA}$ = 571 ± 123 ppm). This may reflect the serpentine chips used for bulk analysis containing some proportion of other, lower Cl minerals, rather than being pure serpentine separate.

3.2.2. Halogens by Noble Gas Method (NI-NGMS)

3.2.2.1. Concentrations

Measured whole rock Cl concentrations (by heating) are similar in both cores, with mean concentrations of 195 and 211 ppm in GT1A and GT3A, respectively (Figure S3b in Supporting Information S1; Table S5). However, the range of Cl concentrations is larger and somewhat bimodal in GT1A (23–550 ppm) than GT3A (105–354 ppm) mirroring the pattern of LOI in the two holes (Figure 1). A similar pattern is seen in Br (Br$_{GT1A}$ = 431 ± 407 ppb, n = 6; Br$_{GT3A}$ = 201 ± 111 ppb, n = 7) and I concentrations (I$_{GT1A}$ = 6.4 ± 5.4, I$_{GT3A}$ = 14.4 ± 7.3). These concentrations are comparable to, but at the lower end of, the range determined from basalts and gabbros in IODP cores (15–1,620 ppm Cl, 44–2,720 ppb Br, 0.5–2,486 ppb I; Chavrit et al., 2016; Kendrick, 2019a, 2019b).

In GT3A, amphibole shows the highest halogen concentrations of the mineral separates (mean Cl = 742 ± 279 ppm, n = 2) while clinozoisite and plagioclase are characterized by lower concentrations (28–133 ppm Cl). A sulfide separate revealed a relatively low Cl content of ∼12 ppm. Bromine is likewise high in amphibole (572 ± 547 ppb, n = 2) and lower in clinozoisite and plagioclase (109–415 ppb); however, the highest Br abundance was in a clinopyroxene separate from sample 107z3 (1,574 ppb). Iodine is low (relative to the global altered oceanic crust) in all mineral separates (0.2–13.6 ppb) with the exception of plagioclase separate from sample S6z1, which has 64 ppb (Table S5).
In GT1A, the highest halogen concentrations are in serpentine separates (mean Cl = 237 ± 137 ppm) with plagioclase and clinopyroxene separates showing relatively low concentrations (3–139 ppm Cl). Bromine and I are both highest in serpentine (Br = 738 ± 402 ppb; I = 11.0 ± 1.5 ppb, n = 2) and relatively low in plagioclase and clinopyroxene (Br = 11–236 ppb, I = 0.1–4 ppb). Clinopyroxene shows a wider range of concentrations than plagioclase for both Br and I. Plagioclase and pyroxene separates from the same sample show very similar abundances to one another for Cl, Br, and I (49±4 and 143±2, Table S5).

In comparison to the heat data, the crush data show lower concentrations of Cl, ranging from 6 to 119 ppm. Where both heat and crush data are available for the same sample, the crush released Cl was between 5% and 63% of the total Cl release by heating, averaging 24%. In every case, the proportion of Br and I released by crushing was proportionally higher than that for Cl, averaging 76% and 77% of the heat totals, respectively (Table S5). In one sample, the amounts of Br and I released by crushing were higher than that released by heating. This likely reflects a degree of heterogeneity between sample aliquots.

### 3.2.2.2. Ratios

The I/Cl ratios from heating analyses show a limited and similar range in whole rock data from both holes (I/Cl = 1.46 × 10⁻³ to 1.43 × 10⁻⁴) and are orders of magnitudes lower than the most I-rich oceanic crust samples in the literature (e.g., Chavrit et al., 2016; I/Clmax = 0.043) but do overlap the lowermost I/Cl range of the literature AOC data (Figure 2a).

In contrast, heating analysis whole-rock Br/Cl ratios show significant differences between the two holes. GT1A shows uniformly higher ratios (GT1A: [2.66 ± 0.79] × 10⁻³, GT3A: [1.00 ± 0.39] × 10⁻³). The bimodal distribution of Br/Cl ratios mirrors the global dataset of heavy halogens in the altered oceanic crust (Chavrit et al., 2016; Kendrick, 2019a, 2019b) which cluster as two groups, each extending to higher I/Cl but with markedly different ranges of Br/Cl values (Figure 2a).

The GT3A clinzoisite and plagioclase separates show much higher I/Cl and Br/Cl than their respective whole rock values, while amphibole separates (and the sulfide separate) plot at similar or slightly lower Br/Cl values. In contrast, mineral separates from GT1A mineral separates have similar halogen ratios to their respective whole rock values (Figure 2a).

In all cases, the crush analyses of a given sample show higher Br/Cl and I/Cl than the corresponding heated analyses (Figures 2a and 3b).

### 3.2.3. Noble Gases

The mean abundances of non-radiogenic noble gases are very similar in both holes (³⁶Armean GT1A = 3.56 × 10⁻⁹, GT3A = 5.68 × 10⁻⁹ cm⁻³ g⁻¹) although, as with Cl, there is a much wider range in abundance in GT1A samples compared to GT3A. The Kr/Ar and Xe/Ar ratios (measured in irradiated samples) are similar in cores from both holes and range from slightly above air values (³⁶Kr/³⁶Ar = 0.21, ¹²³Xe/³⁶Ar = 7.84 × 10⁻⁴) to above air-saturated water (ASW; ³⁶Kr/³⁶Ar = 0.037, ¹²³Xe/³⁶Ar = 2.34 × 10⁻³).

Values of ⁴He/³²He in GT3A are air-like and range from 0.7 to 1.2 Rs (where Rs is the atmospheric ratio of 1.38 × 10⁻⁶). The GT1A samples reveal more mantle-like ratios (⁴He/³²He of 6.2–8.7 Rs), although with smaller overall ⁴He release (mean 0.34 × 10⁻⁶ cm⁻³ g⁻¹) than GT3A (2.23 × 10⁻⁶ cm⁻³ g⁻¹). Corrections for atmospheric ⁴He, assuming all Ne derives from the air (⁴He_c = ⁴He - ²¹Ne × [⁴He/²¹Ne]air), were negligible and altered ⁴He/³²He ratios by less than 1.5%. Since Ne data was not available for all samples, ³²He/⁴He data are reported uncorrected. Values of ⁴He/³⁶Ar range from 1.28 × 10⁻⁹ to 2.70 × 10⁻⁹ with similar averages for both holes (⁴He/³⁶ArGT1A = 1.46 ± 1.29 × 10⁻³, ⁴He/³⁶ArGT3A = 1.04 ± 0.51 × 10⁻³). Even the lowest values are several orders of magnitude above atmospheric ⁴He/³⁶Ar (2.3 × 10⁻⁷), indicating that mantle-derived ⁴He is present in all samples. The neon release was relatively minor (6–15 × 10⁻⁹ cm⁻³ g⁻¹) and isotopically within analytical error of air (Table S6). The ⁴He/²¹Ne ratios range from 768 to 1905, elevated with respect to air (0.29) or ASW (0.41 at 25°C) but markedly lower than the highest values recorded in mid-ocean ridge basalts (MORB; ⁴He/²¹Ne <30,000; Raquin et al. (2008)). Xenon isotopic ratios are within error of their atmospheric values.

Irradiated whole rock and mineral separates from GT3A samples show a strong correlation between ³⁶Ar/³⁶Ar (≡K/³⁶Ar) and ⁴₀Ar/³⁶Ar, plotting close to a 96 Ma isochron (Figure S4 in Supporting Information S1) with an
4. Discussion

4.1. Halogen Siting and Mass Balance

Combining the mineral abundances determined by XRD with their Cl contents determined in situ by EPMA allows us to assess the relative importance of each mineral to the whole rock Cl budget. The concentration of Cl in a given sample (whole rock or mineral separate) may be estimated by summing the products of mineral abundances (from XRD) and mineral Cl concentrations (from EPMA; 1,210 ppm Cl in amphibole, 610 ppm in lizardite, and 180 ppm in chlorite) and assuming a constant 35 ppm Cl (the mean crush release) in fluid inclusions or other sites. These estimated Cl concentrations show a fair degree of correlation with measured (NI-NGMS) concentrations ($R^2 = 0.70, 0.45$ for whole rock only; Figure S3a in Supporting Information S1) indicating that variable age determination is not possible.
combinations of amphibole, chlorite, and serpentine/iddingsite can broadly explain the halogen budget of altered gabbros from both cores. The scatter observed (Figure S3a in Supporting Information S1), particularly for whole rock samples, likely reflects variation in mineral Cl content from sample to sample, as well as the limitations of determinating accurate mineral proportions by bulk powder XRD.

Figure 3. Comparison of crushing and heating analyses (a) Br/Cl versus Cl/36Ar (both mol/mol) for all whole rock and mineral separates. Tie lines are plotted connecting heating data (filled symbols) and crushing data (open symbols) from the same sample and appear to define conjugate compositions which span the Cl/36Ar composition of seawater. Within the datasets for either hole, there is no correlation between Br/Cl and Cl/36Ar. (b) Br/Cl versus I/Cl plotted on linear scales with tie lines connecting crushing and heating analyses as in the previous panel. Crush data (open symbols) are systematically offset to higher Br/Cl and I/Cl ratios relative to heat data (filled symbols) likely reflecting fluid-mineral partitioning. Estimating the original fluid composition as the mid-point of these tie-lines (see Text S3 in Supporting Information S1) suggests markedly different fields of alteration fluid compositions for GT1A and GT3A.
As expected from their high Cl contents, amphibole and iddingsite are the most important Cl hosts in the gabbros (Figure 2b). In GT3A samples, amphibole is the most important Cl-bearing phase, while in GT1A it is iddingsite and/or lizardite when present. Amphibole nonetheless appears to contribute significant Cl in several GT1A samples.

The proportion of whole rock Cl released by crushing varies from 18% to 42% (mean 25%) of the total released by heating indicating that a modest but not insubstantial proportion of the Cl is hosted in sites other than within mineral lattices, for example, fluid inclusions or grain boundaries. Amphibole and serpentine separates have relatively low crush-released Cl (5%–13%) but clinozoisite has a relatively high proportion of 63%.

In comparison, the proportions of Br and I released during crushing are significantly higher, ranging from averaging 77% of total Br and 78% of the total I, and reflected by systematically higher Br/Cl and I/Cl ratios during crushing relative to heating the same samples (Figures 2a and 3b). This suggests the hosting of the heavier halogens is more complex and a much larger proportion of these elements may be hosted in fluid inclusions or along mineral grain boundaries.

4.2. Fluid Compositions and Sources of Halogens

The sources of fluids during hydrothermal alteration offer important insights into the circulation and alteration conditions at depth in the oceanic crust. Sources of fluids and volatiles may potentially include seawater-derived fluids or brines, recycled magmatic volatiles, and sedimentary pore fluids. The halogen and noble gas geochemistry of such fluids provide key insights into their provenance (e.g., Carter et al., 2021).

To constrain the original composition of the fluids which altered the gabbros, it is necessary to account for any modification of fluid signatures during incorporation of halogens in alteration minerals. The preceding discussion indicates that amphibole is the dominant control on the Cl budget of all GT3A samples and many GT1A samples too (Figure 2b). Amphibole is known to incorporate Cl preferentially over Br and I, resulting in very low mineral Br/Cl and I/Cl ratios (Kendrick et al., 2013, 2015). This effect is clearly seen in the amphibole separates and the whole rock samples from GT3A, all of which plot to low Br/Cl values (<1.5 x 10^-3; Figures 2a and 2c).

The composition of the fluid prior to its involvement in hydrothermal reactions and potential modification by the formation of halogen-bearing minerals can be constrained by two methods.

First, we can plot the composition of our samples against the proportion of Cl hosted in amphibole (calculated using XRD-determined mineral proportions of amphibole, lizardite, and chlorite, assuming 1,210, 610, and 180 ppm Cl, respectively, and normalized to the total Cl of the sample). This results in distinct vectors of amphibole addition for different starting fluid compositions (Figure 2d). Plotted in this manner, the compositions of GT3A amphibole separates are consistent with formation by seawater, resulting in Br/Cl = 0.34 x 10^-3 – 1.0 x 10^-3, close to the 0.35 x 10^-3 expected from the estimated ratio of mineral-fluid partition coefficients D_{Br}/D_{Cl} = 0.1 (Figure 2a; Kendrick et al., 2015). Furthermore, the majority of the GT3A data are bounded by mixing lines between amphibole and seawater or MORB (Br/Cl = 3.5 x 10^-3 and 2.5 x 10^-3, respectively).

In contrast to the GT3A data, many of the GT1A whole rock samples lie to higher Br/Cl, despite having a significant proportion of their halogen budget in amphibole (Figure 2d). Bromine/Cl is high, in many cases approaching the seawater ratio, in all amphibole-bearing GT1A samples and is markedly different from GT3A samples which have similar proportions of amphibole (e.g., GT3A 146z4: amphibole XRD = 19%, Br/Cl = 5.2 x 10^-4; GT1A 95z2: amphibole XRD = 16.2%, Br/Cl = 4.1 x 10^-3). There is no evidence that contributions from (higher Br/Cl) lizardite, iddingsite, or chlorite are masking a signature of low Br/Cl amphibole, raising the whole rock Br/Cl. Rather, the highest Br/Cl ratio is from a sample without lizardite or iddingsite and with a low abundance of chlorite (95z2; Table S1). These features suggest that, compared to GT3A, amphibole in GT1A either formed from a higher Br/Cl fluid or under different conditions (or both).

A second approach to constraining the original composition of the altering fluid is to consider crush and heat analyses of the same samples. Crushing releases volatiles from fluid inclusions and grain boundaries while heating releases volatiles from throughout the sample mineral matrix. Fluid inclusions in hydrous phases such as amphibole are likely to have been trapped during hydrothermal formation and therefore the
composition of their trapped fluid will have been modified by preferential incorporation of Cl in amphibole relative to Br and I. Crush and heat data, therefore, represent complementary compositions which may be used to constrain the original fluid composition via application of the lever rule and assuming that fluid and mineral host are in equilibrium (e.g., Kendrick et al., 2015). There are uncertainties associated with the exact salinity, fluid/rock ratio (during alteration/entrapment), and degree of post-entrapment re-equilibration of the trapped fluid; however, these effects tend to oppose one another. We, therefore, take the mid-point of the tie lines connecting crush and heat analyses as the best estimate of the original fluid composition (see Text S3 in Supporting Information S1 for further detail).

Application of this approach to the GT3A amphibole and clinozoisite analyses suggests original fluid compositions close to the seawater-sedimentary pore fluid trend (Br/Cl = 3.0–4.2 × 10⁻³, I/Cl = 1.7 - 8.9 × 10⁻⁵; Figure 3b). Pairs of crush and heat data from whole rock GT3A samples show similar systematics with crush data always offset to higher Br/Cl and I/Cl. Although the assumption that fluid inclusions and mineral matrices are in equilibrium and derive from a single fluid is less reliable for whole rock samples composed of multiple minerals, the same lever rule approach suggests similar fluid compositions close to the seawater–sedimentary pore fluid trend (Figure 3b).

Compared to the GT3A data, the fluid compositions estimated from GT1A crush and heat analyses are markedly different and extend from seawater-like compositions to much higher Br/Cl (Figure 3b). While some of the alteration in GT1A (e.g., 16z3 WR) appears to have been by seawater (Br/Cl = 3.4 × 10⁻³), a fluid with Br/Cl = 5.5–6.8 × 10⁻³ and I/Cl = 0.8–2.5 × 10⁻⁴ was responsible for the alteration of at least some of the GT1A samples (49z4 WR, 95z2 WR; Figure 3b).

There is therefore good evidence for heterogeneous sources of fluids/volatiles in lower crustal GT1A gabbros, sometimes within the same sample (e.g., sample 16z3). In comparison, the composition and halogen host of GT3A samples is much more homogeneous across the samples studied, suggesting less variability in fluid supply and/or sources during alteration.

Modest enrichments in I/Cl and Br/Cl (especially in clinozoisite and plagioclase separates from GT3A sample 107z3) likely reflect the incorporation of a small proportion of sedimentary pore fluid during alteration. The distribution of both the GT1A and GT3A data are well fit by mixing curves between the various fluid endmembers discussed above and a common sedimentary component (Figure 2a, see also Text S4 in Supporting Information S1).

4.3. Contrasting Modes of Alteration in Mid and Lower Crustal Gabbros

4.3.1. Br-Cl Fractionation During Alteration at Low Fluid/Rock Ratios

A common feature of the preceding discussion is the contrast in the volatile signatures and mineralogy of the two holes. The key difference is that the Br/Cl is clustering at two separate Br/Cl ratios (Figure 2a) requiring either the Br/Cl of the altering fluid to have been different in each case, or for the fluid/rock ratios to have been much lower during alteration of the deeper GT1A gabbros. In this section, we examine the potential causes of these differences.

Variation in Br/Cl has been suggested to occur as a result of phase separation during hydrothermal alteration, fractionating Br from Cl in the resulting brine/vapor. However, vent fluid Br/Cl is generally indistinguishable from seawater (e.g., Campbell & Edmond, 1989) with the exception of transient events possibly related to the magmatic activity (Berndt & Seyfried, 1997; Oosting & Von Damm, 1996). Furthermore, experimental evidence for the direction of this fractionation is contradictory, suggesting that phase separation may produce brines with high (Liebscher et al., 2006), low (Berndt & Seyfried, 1999; Foustoukos & Seyfried, 2007), or similar (Berndt & Seyfried, 1997) Br/Cl to initial fluid compositions.

Furthermore, there is limited evidence of the involvement of high salinity brines in our dataset. The Cl/³⁶Ar ratios provide a strong constraint on this since Cl partitions preferentially into the brine and Ar into the vapor during phase separation. Differential partitioning into minerals may subsequently alter the original fluid ratio; however, pairs of data from heating and crushing the same sample bracket the composition of seawater (Cl/³⁶Ar = 1 × 10⁻⁷ mol/mol) with heating data offset to higher Cl/³⁶Ar and crush data to lower values (Figure 3a). This strongly suggests they form conjugate compositions due to differential incorporation of Cl and Ar in mineral matrices.
Contrasting Ar and He isotope ratios between whole rock GT1A and GT3A samples (a) He-U-Th isochron plot showing initial $^{3}$He/$^{4}$He ratios of $\sim$8 $R_\alpha$ and 1–2 $R_\alpha$ for GT1A and GT3A samples, respectively. Isochrons (dotted lines) are plotted for initial $^{3}$He/$^{4}$He ratios of 1, 2, 4, and 8 $R_\alpha$ (where $R_\alpha = 1.38 \times 10^{-6}$). He isotopes measured on unirradiated samples (Table S6), U abundance measured by NI-NG-MS on irradiated samples (Table S5), and Th calculated assuming Th/U = 0.5. Isochrons were calculated using the equation of Moreira et al. (2003). (b)$^{40}$Ar/$^{36}$Ar$_{web}$ versus $^{3}$He/$^{4}$He plotted with a mixing curve calculated between MORB and air saturated water. Water/rock ratios are plotted at regular intervals for reference.

Figure 4. Contrastingly, Ar and He isotope ratios between whole rock GT1A and GT3A samples (a) He-U-Th isochron plot showing initial $^{3}$He/$^{4}$He ratios of $\sim$8 $R_\alpha$ and 1–2 $R_\alpha$ for GT1A and GT3A samples, respectively. Isochrons (dotted lines) are plotted for initial $^{3}$He/$^{4}$He ratios of 1, 2, 4, and 8 $R_\alpha$ (where $R_\alpha = 1.38 \times 10^{-6}$). He isotopes measured on unirradiated samples (Table S6), U abundance measured by NI-NG-MS on irradiated samples (Table S5), and Th calculated assuming Th/U = 0.5. Isochrons were calculated using the equation of Moreira et al. (2003). (b)$^{40}$Ar/$^{36}$Ar$_{web}$ versus $^{3}$He/$^{4}$He plotted with a mixing curve calculated between MORB and air saturated water. Water/rock ratios are plotted at regular intervals for reference.

Another mechanism by which Br/Cl may vary is via the formation of amphibole. Since amphibole forms with systematically lower Br/Cl than the fluid, there is a coexistent enrichment of Br in the residual fluid. At high fluid/rock ratios this has little to no effect on the bulk fluid compositions; however, at low fluid/rock ratios, progressive removal of Cl relative to Br can result in large fractionations. Since Cl and H$_2$O are likely incorporated into amphibole in similar (or slightly lower) proportions to their ratio in the fluid (Chan et al., 2016; Kendrick et al., 2015; Sato et al., 2005), this mechanism should not result in any major change in salinity.

This scenario implies that high Br/Cl ratios in GT1A gabros derive from seawater-derived fluids which have been fractionated by amphibole-forming reactions at very low fluid/rock ratios. Assuming a value for $D_{Cl}$ of $10^{-3}$–$10^{-5}$ (Fabbri et al., 2013) and $D_{Br}/D_{Cl} = 0.1$ (Kendrick et al., 2015) producing the highest Br/Cl ratios observed in GT1A samples (Br/Cl = $5 \times 10^{-3}$) via a Rayleigh fractionation model implies fluid/rock ratios of between $10^{-2}$ and $10^{-3}$. These are somewhat lower than suggested by Bosch et al. (2004) who estimated alteration at fluid/rock ratios of $\sim$3–5 in the lower crust but are broadly comparable to, though still somewhat lower than, estimates from the least altered gabbros from Oman (e0.3, Gregory & Taylor, 1981; 0.5, McCulloch et al., 1981) and Hess Deep (0.2–1; Lecluse & Reynard, 1996). Equally, the coexistence of such high Br/Cl ratios with apparently seawater-like compositions (see above) suggests variability in fluid/rock ratios during alteration of the GT1A gabbros. This implies that very low ($<<1$) fluid/rock ratios are unlikely to be representative of all alteration of the lower crust and that records of such conditions may only be preserved where subsequent alteration has not overprinted them.

4.3.2. Noble Gas Evidence for Low Fluid/Rock Ratios During Lower Crustal Alteration

Support for a distinction between the two cores can also be drawn from considering noble gas isotopes. Helium and Ar isotopes are unaffected by fluid-mineral interactions and reflect only mixing of different volatiles sources and radiogenic ingrowth ($^{4}$He from $^{235,238}$U, $^{232}$Th; $^{40}$Ar from $^{40}$K). Argon isotope ratios have been corrected for radiogenic ingrowth of $^{40}$Ar to 96 Ma, the age of the ophiolite (Rioux et al., 2013). Correcting He isotopes is somewhat less precise than with Ar-Ar since U (measured by NI-NG-MS; Table S5) and He were analyzed on different sample aliquots. However, when plotted on a U-Th-He isochron diagram with 96 Ma isochrons of varying initial $^{3}$He/$^{4}$He (calculated using the equation of Moreira et al., 2003), all GT3A samples plot between initial $^{3}$He/$^{4}$He ratios of 1–2 $R_\alpha$ whereas all GT1A samples plot to within an error of the 8 $R_\alpha$ isochron (Figure 4a).

The $^{3}$He/$^{4}$He and $^{40}$Ar/$^{36}$Ar ratios are markedly different between the two holes (Figure 4b). Several GT1A samples show some excess (mantle-derived) $^{40}$Ar and all samples analyzed have $^{3}$He/$^{4}$He ratios close to the MORB value of 8 $R_\alpha$. By contrast, the GT3A data are uniform in having decay-corrected $^{40}$Ar/$^{36}$Ar very close to the atmospheric ratio (298.56). Moreover, their $^{3}$He/$^{4}$He ratios are all low (<1.2 $R_\alpha$) and closely grouped near the atmospheric ratio. The $^{40}$Ar/$^{36}$Ar ratios of both holes are fairly similar ($^{40}$Ar/$^{36}$Ar = 1.28 $\times 10^{-4}$ to 2.70 $\times 10^{-3}$), markedly lower than the ratio in MORB (0.7; Moreira et al., 1998), and approaching the atmospheric ratio (6.76 $\times 10^{-8}$). These features suggest both that the $^{3}$He preserved in the rocks is ultimately mantle-derived and (released by heating) and fluid inclusions/grain boundaries (released by crushing), and that the original fluids had a Cl/$^{36}$Ar consistent with the salinity of seawater. Additionally, there is no systematic difference in the Cl/$^{36}$Ar ratios of samples from GT1A and GT3A and no correlation between Cl/$^{36}$Ar and Br/Cl to suggest that phase separation is responsible for the differences in Br/Cl seen between the two holes.
that this has been mixed with a large proportion of atmospheric noble gases. Similarly, $^4\text{He}/^2\text{He}$ ratios in the gabbros range from 768 to 2,480, intermediate between the ASW ($\approx$0.41 at $25^\circ$C) and MORB ($>$30,000, Raquin et al., 2008) values. Simple mixing between ASW and MORB endmembers suggests that 92%–97% of the noble gases are atmospheric.

The noble gas systematics are best explained by variable mixing between magmatic noble gases (with MORB-like composition) and seawater (with atmospheric isotopic composition). Vent fluids typically have $^4\text{He}/^3\text{He}$ similar to local basalts (e.g., Jean-Baptiste et al., 2004; Jenkins et al., 1978; Rudnicki & Elderfield, 1992), suggesting that magmatic and seawater volatiles commonly form a mixed reservoir during hydrothermal circulation. The contrast in noble gas isotopes between the two holes is therefore notable since this implies a difference in the proportions of magmatic and seawater-derived volatiles in the system and that variations in isotopic ratios may reflect variation in fluid/rock ratios.

The preservation of mantle-derived $^{40}\text{Ar}/^{36}\text{Ar}$ in GT1A is not restricted only to very fresh samples or fresh minerals with two samples having $^{40}\text{Ar}/^{36}\text{Ar} = 330$ to 420 and LOI between 1.5 and 3.8 wt.%. By contrast, all samples from GT3A and many from GT1A show nearly atmospheric initial $^{40}\text{Ar}/^{36}\text{Ar}$ ratios ($\approx$300). These are best explained by the loss of magmatic volatiles to the fluid at high fluid/rock ratios such that magmatic Ar is completely diluted by seawater-derived signatures.

The preservation of mantle-like He isotopes in GT1A and not in GT3A likewise suggests fluid circulation characterized by relatively low and high fluid/rock ratios respectively. The occurrence of mantle-like He with air-like Ar isotopes in GT1A samples strongly indicates alteration by fluid bearing a mixture of magmatic and seawater-derived volatiles. Mixing curves between MORB and air-saturated water indicate that $^3\text{He}/^4\text{He}$ ratios $>7.5$ R in GT1A rocks can only be preserved for fluid/rock ratios $<0.6$ (Figure 4b). Meanwhile, $^3\text{He}/^4\text{He}$ ratios $<1.2$ R in GT3A rocks imply fluid/rock ratios $>120$ (Figure 4b). The occurrence of air-like He isotopes in the GT3A rocks is somewhat surprising since vent fluid He isotopes are typically mantle-like suggesting that magmatic He dominates during focused on-axis hydrothermal circulation (e.g., Jean-Baptiste et al., 2004). One explanation for these data might be prolonged off-axis alteration by seawater. In that case, the large fluid/rock ratio calculated above might reflect time-integrated ratios for the entire sequence of alteration rather than the instantaneous mixing of magmatic and seawater volatiles immediately following magmatic emplacement.

### 4.3.3. Magmatic Assimilation of Fractionated Seawater Derived Fluids

Several altered GT1A whole rock samples have high $^{40}\text{Ar}/^{36}\text{Ar}$ and $^4\text{He}/^3\text{He}$ ratios indicative of magmatic contributions to alteration fluids. Similar excesses of $^{40}\text{Ar}$ are present in fresh mineral separates from several GT1A samples too. High $^{40}\text{Ar}/^{36}\text{Ar}_{96\text{Ma}}$ (i.e., corrected for radiogenic ingrowth from $^{40}\text{K}$ over 96 Myr) shows a strong association with fresh minerals: clinopyroxene and plagioclase (Figure 5). There is a general trend of increasing Br/Cl and $^{40}\text{Ar}/^{36}\text{Ar}_{96\text{Ma}}$ in the GT1A samples (Figure 5). The highest Br/Cl ratios are in fresh clinopyroxene and plagioclase separates and are significantly higher than the maximum GT1A whole rock ratios. This implies that the magmatic component with mantle-derived $^{40}\text{Ar}$ also had high Br/Cl. The highest Br/Cl ratios (Br/Cl $<5 \times 10^{-3}$) are well above those measured in fresh MORB, unaffected by seawater assimilation (Figure 5; Br/Cl = $2.9 \pm 0.7 \times 10^{-3}$; Kendrick et al., 2017), and also above the seawater value (Br/Cl = $3.4 \times 10^{-3}$). This suggests that the primary magmatic component in these minerals incorporated a high Br/Cl component.

One candidate for such a component might be slab-derived fluids in subduction zone magmas. The Oman Ophiolite is widely believed to have formed in a supra-subduction zone setting (e.g., Searle & Cox, 1999) and shows evidence for a subduction zone influence in many of its lava compositions (MacLeod et al., 2013; Pearce et al., 1981). However, halogens in a wide range of back-arc basin basalts are characterized by elevated I/Cl or correlated excesses of I/Cl and Br/Cl over MORB values (Kendrick et al., 2014). In contrast, the fresh minerals from GT1A show no excess I/Cl relative to more altered samples or MORB compositions. Instead, they are
elevated in Br/Cl with no discernible change in I/Cl ratios. Production of such high Br/Cl ratios without a con-
comitant increase in I/Cl is often attributed to the assimilation of fractionated seawater derived fluids and/or 
brines (Kendrick et al., 2014, 2015). At low fluid/rock ratios these may develop high Br/Cl through reactions to 
form low Br/Cl amphibole, as discussed in the preceding section. Calculated two-stage mixing curves, first for 
assemblage of a high Br/Cl seawater-derived fluid by a MORB-like magma, and subsequent minor alteration 
toward the composition of altered GT1A whole rocks reproduces the range of compositions seen (Figure 5).

4.4. Delivery of Fluid to Lower Crust Along Major Faults

A problem with models requiring hydrothermal circulation in the lower oceanic crust is the requirement for 
pathways of fluid delivery. Large faults have been documented in the Oman Ophiolite (e.g., Abily et al., 2011; 
Zihlmann et al., 2018). However, there remains controversy over whether these and their associated alteration, 
represent oceanic crustal structures or late features related to ophiolite emplacement. A major fault zone outcrops 
close to the site of the GT1A drill site and has been demonstrated to record alteration at high fluid/rock ratios 
(>400; Zihlmann et al., 2018). Intersecting such faults at depth was one of the key motivations of this drill site 
(Kelemen et al., 2020) and, although the exact intersection of this structure with the GT1A core is unclear, seven 
major fault zones between 2 and 22m thick were logged in the GT1A core (Kelemen et al., 2020). It is therefore 
worth considering the evidence from halogens and noble gases, as well as the shipboard geochemistry dataset, for 
these structures representing zones of fluid supply to the lower Oman crust in hole GT1A.

A marked feature of the shipboard GT1A data is a relationship between LOI and distance to the nearest major 
fault zone (Figure 6a; Kelemen et al., 2020) with the most hydrated samples occurring nearest to the faults. 
Furthermore, ratios of fluid mobile elements to immobile elements of similar incompatibility during magmatic 
processes (Pb/Ce, Rb/Ce) show a similar relationship with distance from major fault zones (Figures 6b and 6c). 
Close to the faults, ratios are highly variable, in some cases elevated by a factor of 10 with respect to fresh gab-
bros (defined as the mean and standard deviation of GT1A gabbros with LOI <1 wt.%) and minor depletions are 
also more common. With increasing distance from the fault zones, the range of Rb/Ce and Pb/Ce systematically 
decreases and the mean composition tends toward that of fresh gabros. We note that several of the highest LOI 
and trace element ratios are from fault zone fragments or vein halos and may not be representative of the gen-
eral degree of alteration at their position. However, these trends are nonetheless apparent in the distribution of 
(olivine-)gabbro data alone.

The relationships in Figure 6 strongly suggest that the major fault zones were important conduits for fluid supply. 
It seems improbable that pre-existing variations in hydration could have exerted a major influence on the location 
of later faulting. Rather, the simplest interpretation of the data is that these fault zones provided the major path-
way for fluids during alteration. The average degree of alteration appears to have diminished steadily away from 
these faults, potentially reflecting decreasing porosity in subordinate fracture networks.

Previous work has estimated very high fluid/rock ratios (>400) in highly altered lithologies within the core of 
one such fault zone exposed at the surface near the GT1A drill site (Zihlmann et al., 2018). This is potentially at 
 odds with the evidence from halogens and noble gases discussed in previous sessions which suggest alteration at 
low fluid/rock ratios in GT1A cores. Evidence for the latter includes high Br/Cl ratios ascribed to fractionation of 
seawater by amphibole-forming reactions, mantle-like 3He/4He ratio, and mantle-derived 40Ar, ascribed here 
to incomplete overprinting of magmatic volatiles by circulating seawater. However, in each of these cases, GT1A 
samples showing evidence for low fluid/rock ratios contrast with samples from the same core apparently altered 
by unfractiated seawater with atmospheric noble gas ratios. This apparent contradiction can be reconciled by 
a model in which fluid/rock ratios during alteration are strongly influenced by the presence of large faults. Fluid/ 
rock ratios would be high in and around these zones such that seawater-like volatiles would be expected near to 
the faults and increasingly fractionated fluids (with greater proportions of magmatic volatiles) would be seen far 
from the faults (Figure 7).

The halogen and noble gas data support such a model. Looking at all sample fractions including whole rock and 
mineral separates (so as to explore the full range of potential fluid compositions experienced by the rocks from 
magmatic to low temperature stages) we observe that, generally, the lowest and most seawater-like Br/Cl and 
40Ar/36Ar ratios are present in samples from close to faults zones (16z2, 40z1, 107z3; Figures 6e and 6f). While 
some samples close to faults have Br/Cl less than seawater, this is likely to be due to the presence of low Br/Cl
amphibole (Figures 2b–2d). Moreover, age-corrected $^{40}$Ar/$^{36}$Ar ratios are air-like, precluding alteration having taken place more recently than $\sim$96 Ma in all but one sample which shows $^{40}$Ar/$^{36}$Ar $< 300$ (indicating incorporation of air-like Ar more recently than 96 Ma; possibly related to its olivine-rich composition and a high potential for further hydration). By contrast, the highest Br/Cl and $^{40}$Ar/$^{36}$Ar ratios are seen in samples furthest

Figure 6. Graphs of geochemical variation related to distance from major fault zones (FZ) in hole GT1A. (a) Loss on Ignition versus distance to FZ for all shipboard samples (b) Pb/Ce versus distance to FZ for all shipboard samples (Kelemen et al., 2020). The mean composition of fresh GT1A gabbros with LOI < 1 wt.% ($n = 11$) is plotted as a dashed line (±1 standard deviation as dotted lines) and seawater (Li, 1991) as a blue line. (c) Rb/Ce versus distance to FZ for all shipboard samples with lines for the mean composition of fresh GT1A gabbros plotted as in the previous panel. Note that seawater composition is elevated and plots beyond the range of the y axis. (d) Br/Cl versus distance to FZ for all GT1A samples in this study, showing increasing maximum Br/Cl with distance to FZ in both heated and crushed data. Also plotted is the result of linear regression and its 1 sigma confidence envelope calculated for the heat data. (e) $^{40}$Ar/$^{36}$Ar$_{96Ma}$ versus distance to FZ for all GT1A samples showing increasing maximum $^{40}$Ar/$^{36}$Ar with distance. Linear regression is plotted as in the previous panel. Crush data are not plotted due to large uncertainties in the age correction. One sample shows age-corrected $^{40}$Ar/$^{36}$Ar $< 300$ suggesting it has been affected by some meteoric alteration more recently than $\sim$96 Ma.
from the faults (49z4, 95z2, 143z2; Figures 6e and 6f). In addition, while clinopyroxene and plagioclase from sample 49z4 show the highest Br/Cl, the whole rock sample shows broadly seawater-like Br/Cl. This suggests that fluids and fluid/rock ratios may have varied throughout alteration. These data strongly suggest that at the time of ophiolite formation (∼96 Ma), seawater was the major fluid driving alteration close to the faults and that more diverse fluids were more common in gabbro domains distant from these faults. Away from these faults, fluid/rock ratios likely dropped markedly but could nonetheless be highly variable (plausibly controlled by smaller scale, syn-magmatic structures; Figure 7).

4.5. Halogen Composition of the Oceanic Crust

There is a broad consensus between currently available studies that bulk Cl contents increase with depth in the upper oceanic crust through greenschist to amphibolite facies reaching a maximum of ∼1,400 ppm at ∼1.5 km depth (Barnes & Cisneros, 2012; Chavrit et al., 2016; Kendrick, 2019a, 2019b; Sano et al., 2008; Zhang et al., 2017). However, the nature of bulk Cl distribution below this depth is more contentious. Chavrit et al. (2016) assumed a homogeneous depth distribution below 800 ms depth resulting in a bulk lower oceanic crust Cl concentration of 284 ppm. By contrast, Kendrick (2019b) argues for a rapid asymptote with depth to very low background concentrations of 15–50 ppm Cl. Our results indicate that the answer appears to be somewhere in between, with mean Cl concentrations for GT1A of 193 ppm (though with a large range from 24 to 532 ppm resulting in a standard deviation of 217 ppm). Because of the low-temperature overprint demonstrated to have affected the Atlantis Bank gabbros during core-complex exhumation (Kendrick, 2019b) and the inherent uncertainties of extrapolating from the upper 1.5 km to the full thickness of oceanic crust (Chavrit et al., 2016), we suggest that our data represents a more accurate estimate of the halogen composition of the lower oceanic crust. Based on the average lower oceanic crust Br/Cl and I/Cl ratios of 2.7 ± 0.8 × 10⁻³ and 52.8 ± 45.6 × 10⁻⁶, respectively, 425 ± 400 ppb Br and 6.4 ± 5.4 ppb are available to subduct. Combining these values with literature data for the upper oceanic crust (Barnes & Cisneros, 2012; Chavrit et al., 2016; Kendrick, 2019a, 2019b; Sano et al., 2008; Zhang et al., 2017; Table S8), the average halogen composition of altered oceanic crust is approximately 233 ppm Cl (range 28–810; interquartile range 71–354), 403 ppb Br (range 49–1,320; IQR 96–641) and 8.4 ppb I (range 1.1–31.3; IQR 1.6–12).
5. Conclusions

Through a detailed study of halogen and noble gas geochemistry in altered gabbros cored from the sheeted-dyke to gabbro transition (GT3) and lower crustal layered gabbros (GT1) of the Oman Ophiolite, we have been able to critically assess and contrast the fluid sources, processes, and conditions during alteration of mid- and lower-oceanic crust.

Mass balance of halogen-bearing phases indicates that the CI abundances are well-explained by the relative proportions of halogen-bearing hydrous alteration minerals (amphibole ≥ iddingsite/lizardite >> chlorite) and are largely derived from hydrothermal alteration in most samples. Relative to Cl, a greater proportion of Br and I appear to be sited in fluid inclusions or other non-matrix sites.

Halogen abundance ratios provide strong evidence that seawater was the main source of fluids during an alteration with a minor contribution from sedimentary pore fluid apparent in some samples. Low Br/Cl ratios of GT3A whole rock samples are explained by the presence of 20–30 wt.% of low Br/Cl amphibole. Several amphibole-bearing GT1A samples have Br/Cl significantly above seawater and must have interacted with a Br-enriched fluid. This appears unlikely to relate to phase separation and instead enrichment of Br in the fluid through preferential removal of Cl during precipitation of amphibole is the best explanation of the data. Modeling this process as a Rayleigh fractionation suggests fluid/rock ratios <<1 in the earliest stages of (amphibolite facies) alteration.

Noble gas isotopes support relatively low fluid/rock ratios in GT1A (<0.6) and high ratios in GT3A (≤100) resulting in air-like (decay-corrected) He and Ar isotope ratios in GT3A samples (3He/4He = 1–2 R⊙, 40Ar/36Arstr ∼300) and preservation of mantle-like 3He/4He (6–8 R⊙) and remnant mantle-derived 40Ar excesses in GT1A (40Ar/36Arstr < 400). At the same time, seawater-like compositions are inferred for altering fluids for some GT1A samples suggesting variable fluid/rock ratios and fluid supply.

A negative correlation between the distance to major fault zones in GT1A and the degree of hydration; the relative abundance of fluid mobile elements (Pb/Ce, Rb/Ce); and positive correlations with Br/Cl; and 40Ar/36Arstr strongly indicates these structures were important zones of fluid supply during alteration. Away from these zones, fluid/rock ratios are inferred to have dropped rapidly, resulting in fractionated fluids with a great proportion of inherited magmatic volatiles. The fact that the correlations in Br/Cl and 40Ar/36Arstr are strongest in the freshest minerals (clinopyroxene and plagioclase) suggests these dynamics were operating from the magmatic stage or very shortly after. Furthermore, the seawater halogen abundances of samples near these zones and their age-corrected 40Ar/36Ar ratios indicate alteration by seawater contemporaneous with the formation of the ophiolite at Ma and preclude a significant role for more recent meteoric processes.

Altogether, the data suggest a highly heterogeneous delivery of water to the lower crust commencing at the magmatic stage and likely mediated by large faults, away from which fluid supply may be extremely limited. In contrast, the mid-crustal gabbros appear to have been altered under more homogenous conditions at relatively higher fluid/rock ratios. These results, therefore, contribute to a developing picture of discrete and possibly co-existing upper and lower crustal systems of hydrothermal circulation in the oceanic crust more generally.

Appendix A: Phase 1

Appendix A1 Onsite Drilling Team

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Appendix A3 ChikyuOman 2017 Description Team

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Data Availability Statement

In accordance with the AGU’s Data Policy, data for this work, consisting of Supplementary Tables S1-S8 (including full XRD, EPMA, halogen, and noble gas datasets), have been deposited in a open access Mendeley Data repository available at https://doi.org/10.17632/v9gmsj5m39.2. The files associated with this dataset are licensed under a Creative Commons Attribution 4.0 International license.

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