Subseafloor Cross-Hole Tracer Experiment Reveals Hydrologic Properties, Heterogeneities, and Reactions in Slow-Spreading Oceanic Crust

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Abstract The permeability, connectivity, and reactivity of fluid reservoirs in oceanic crust are poorly constrained, yet these reservoirs are pathways for about a quarter of the Earth's heat loss, and seawater-rock exchange within them impact ocean chemical cycles. We present results from the second ever cross-hole tracer experiment within oceanic crust and the first conducted during a single expedition and in slow-spreading crust west of the Mid-Atlantic Ridge at North Pond. Here we employed boreholes that were drilled by the Integrated Ocean Drilling Program (Sites U1382 and U1383) that were instrumented and sealed. A cesium salt solution and bottom seawater tracer experiment provided a measure of the minimum Darcy fluid velocity (2 to 41 m/day) within the upper volcanic crust, constraining the minimum permeability of $10^{-11}$ to $10^{-9}$ m$^2$. We also document chemical heterogeneities in crustal fluid compositions, rebound from drilling disturbances, and nitrification within the basaltic crust, based on systematic differences in borehole fluid compositions over a 5-year period. These results also show heterogeneous fluid compositions with depth in the borehole, indicating that hydrothermal circulation is not vigorous enough to homogenize the fluid composition in the upper permeable basaltic basement, at least not on the time scale of 5 years. Our work verifies the potential for future manipulative experiments to characterize hydrologic, biogeochemical, and microbial process within the upper basaltic crust.

Plain Language Summary Seawater flows within the oceanic crust, much like groundwater flows though permeable aquifers within continental crust. As seawater flows through the oceanic crust it dissolves some minerals while precipitating others. These biogeochemical reactions coupled with subsurface seawater flow affect the distribution of heat, solutes, and microbial populations within the oceanic crust, and such reactions and transport can impact oceanic processes. To assess hydrologic and biogeochemical processes that occur within the oceanic crust west of the Mid-Atlantic Ridge, we conducted the second ever borehole-to-borehole tracer experiment within oceanic crust; however, similar experiments are commonplace in continental settings. On the basis of this tracer experiment and prior sampling of these boreholes, we determined that the upper several hundred meters of volcanic oceanic crust are highly permeable and support microbial nitrification; however, fluids within the upper oceanic crust are not well mixed at this location. Our results provide the foundation to conduct future manipulative experiments within the oceanic crust to characterize hydrologic and biogeochemical processes within this poorly constrained, yet globally significant, environment.

1. Introduction

A quarter of Earth’s heat loss is attributed to hydrothermal circulation within the oceanic crust (e.g., Lister, 1972; Parsons & Sclater, 1977), with major effects on crustal and ocean chemistry (e.g., Elderfield & Schultz, 1996; Mottl & Wheat, 1994). In off-axis settings, this circulation occurs mainly in the upper several hundred meters of the volcanic crust (Layer 2A), which has a permeability of $10^{-11}$ to $10^{-9}$ m$^2$. We also document chemical heterogeneities in crustal fluid compositions, rebound from drilling disturbances, and nitrification within the basaltic crust, based on systematic differences in borehole fluid compositions over a 5-year period. These results also show heterogeneous fluid compositions with depth in the borehole, indicating that hydrothermal circulation is not vigorous enough to homogenize the fluid composition in the upper permeable basaltic basement, at least not on the time scale of 5 years. Our work verifies the potential for future manipulative experiments to characterize hydrologic, biogeochemical, and microbial process within the upper basaltic crust.
seamounts with basaltic exposures, and at sites where formation fluids discharge, primarily through smaller basalt exposures (e.g., Fisher et al., 2003). Likewise, chemical data have been used to indicate the presence of crustal circulation, but in most cases, there is insufficient data to assess the direction of flow (Baker et al., 1991; Elderfield et al., 1999; Mewes et al., 2016; Mottl and Wheat 2004; You et al., 2003; Ziebis et al., 2012). On the basis of these chemical and thermal data, models have been devised to estimate rates of flow within the upper volcanic crust, making assumptions of flow geometry (Lauer et al., 2018; Orcutt et al., 2013; Wheat and Fisher, 2007 and 2008; Winslow et al., 2016). To date, the only crustal location with a clearly defined subsurface flow direction is in 3.5 Ma crust on the eastern flank of the Juan de Fuca Ridge (JFR). Here, systematic variations in the chemical composition of formation fluids from boreholes, springs, and pore water data defined the direction of flow (Hulme & Wheat, 2019; Wheat et al., 2000). That flow direction was recently confirmed with the first cross-hole tracer experiment ever conducted in basaltic oceanic crust in which a chemical tracer was introduced into one borehole and observed in another (Neira et al., 2016).

The success of this initial cross-hole tracer experiment in basaltic crust provided the foundation to attempt a second cross-hole tracer experiment, the focus of this paper. Our study utilized borehole observatories at North Pond, a sediment-filled (~10 km by 15 km) depression that overlies 8 Ma crust west of the slow-spreading Mid-Atlantic Ridge (Figure 1). Here subsurface fluid flow through the upper basaltic crust is thought to either recharge and discharge convectively via exposed basement or through thin sediment or be redistributed laterally by flow in upper basement beneath the sediment pond (Langseth et al., 1992; Schmidt-Schierhorn et al., 2012; Villinger et al., 2019).

We present data from three boreholes drilled in 2011 during Integrated Ocean Drilling Program Expedition 336. Each borehole was cased through the sediment section and cemented into upper volcanic basement, eliminating the hydrologic connection between the basaltic formation and sediment (Edwards, Bach, et al., 2012). After the cement solidified additional drilling operations occurred followed by the installation of sensors and samplers. On a return expedition in 2017 (AT39-01) with the remotely operated vehicle (ROV) Jason II, a tracer was injected into one of the boreholes, and borehole fluids were subsequently collected. On the basis of systematic variations in fluid compositions during this and prior expeditions, our observations provide an indication of the length of time for the crust to rebound from drilling disturbances, a measure of subsurface reactions, and constraints for hydrologic characteristics in the permeable upper basaltic basement within slow-spreading crust. This experiment also serves as the foundation for future in situ hydrologic/microbial experiments in cool (<10 °C), young oceanic crust.

2. Methods

2.1. Seafloor Borehole Systems and Operations

During Integrated Ocean Drilling Program Expedition 336 in September–November 2011, three boreholes at two sites were drilled in North Pond for long-term observatories (Edwards, Bach, et al., 2012) (Figure 1). Within tens of meters of Deep Sea Drilling Project Hole 395A, Hole U1382A was drilled through 90 m of sediment into volcanic basement. Steel casing (102 m in length) was deployed in the borehole and cemented to stabilize the sediment column and upper basaltic basement. The cement was drilled out, and the hole was cored to 210 m below seafloor (bsf). An instrumented borehole system (Circulation Obviation Retrofit Kit, CORK; Davis et al., 1992) was deployed with fiberglass, perforated fiberglass, and coated, perforated steel casing (Edwards, Bach, et al., 2012). Perforations covered the depth range of 146 to 188 m bsf to provide a path for formation fluids to flow into the CORK casing structure where continuous fluid samplers (OsmoSampler; Jannasch et al., 2004; Wheat et al., 2011) and other experiments were deployed (Edwards, Bach, et al., 2012). This zone can also be sampled via umbilicals, two of which were armored 0.5-inch inner diameter Tefzel tubing that terminated within the single hydrologic zone (160 m bsf) and at a Jannasch handle connector on the wellhead (Wheat et al., 2011). Here we define a hydrologic zone by the length of open borehole below the packer, noting that within such a zone there is a complex network of permeable structures within the basaltic matrix. Data presented herein were from samples that were collected after purging the umbilical during R/V Atlantis cruise (AT39-01) in 2017. Such fluids were pumped from the Tefzel umbilical into a series of acid-washed containers (Cowen et al., 2012).

Site U1383 was positioned 6.1 km from Hole U1382A, near the northeast edge of the sediment pond (Figure 1). Hole U1383B was drilled through 52.8 m of sediment, cased and cemented to 53.8 m bsf to
stabilize the formation, and then drilled to a depth of 89.8 m bsf before being abandoned after the drill bit was destroyed (Edwards, Bach, et al., 2012) (Figure 2 and supporting information Figure S1). About 6 months after drilling, a CORK‐Lite was deployed in Hole U1383B with continuous fluid samplers (OsmoSampler), experiments, and temperature and pressure sensors during RV Maria S. Merian cruise MSM20‐5 (Wheat et al., 2012). OsmoSampler intakes were positioned at a depth of 60.6 m bsf, below the protection of the casing in the open portion of the borehole that defines a single hydrologic zone in the upper 4.2–37 m of basaltic crust.

Hole U1383C was then positioned 25 m from Hole U1383B and drilled through 38.3 m of sediment into volcanic crust (Figures 2 and S1) (Edwards, Bach, et al., 2012). The borehole was cased and cemented to 60.4 m bsf, and the cement was drilled out. Coring continued to a depth of 331.5 m bsf before installation of a three‐zone CORK of similar construction to that in Hole U1382A. The CORK system included sections of fiberglass and steel casing that reached a depth of 247.6 m bsf. Three sections of casing were perforated: shallow 76.3–129.4 m bsf, middle 145.9–181.1 m bsf, and deep 203.7–247.6 m bsf. Umbilicals were attached to the outside of the casing with terminations at depths of 100, 162, and 203 m bsf. An armored 0.5‐inch inner diameter Tefzel tubing was installed at each depth and terminated at the wellhead with a Jannasch handle (Edwards, Bach, et al., 2012). These Tefzel umbilicals hold 12.7, 20.5, and 25.7 L, respectively. Inflatable and swellable packers were deployed between the umbilical terminations, partitioning the borehole into three hydrologic zones, 26.5–103.9, 103.9–158.1, and 158.1–293.2 m bsf. The upper interval was chosen based on its phric or sparsely phric basalt morphology, in contrast to aphyric basalt in deeper portions of the borehole. The middle zone was chosen to isolate a more conductive layer, based on gamma ray data from logging results (Edwards, Bach, et al., 2012). Continuous fluid samplers (OsmoSampler; Jannasch et al.,
Figure 2. Model of tracer (yellow) and bottom seawater (blue) flow within the volcanic crust with respect to whether the top seals on the boreholes were open or closed. Umbilicals are represented by blue, red, and green lines in Hole U1383C. The black line in Hole U1383B represents the intake for the OsmoSamplers. Black stars highlight where the intakes for the two downhole OmoSamplers were positioned. (a) A Cs tracer is injected into Hole U1383B and (b) reaches Hole U1383C after the borehole seal is removed. (c) The borehole seal is removed at Hole U1383B, allowing seawater to flow into the basaltic formation. (d) Hole U1383C was sealed and (e) several days later bottom seawater impacted the fluid composition in Hole U1383C. (f) An alternative explanation for the Cs data is that flow was dominantly horizontal in the uppermost basaltic basement and reached the upper interval in Hole U1383C before mixing vertically within the annulus of the borehole. This scenario could explain the tracer data if the seals (packers) were ineffective; however, it is not consistent with the other chemical data or with the increasing value of positive formation pressure with depth, indicating that the three zones were sealed by packers (Figure S6). The image is not to scale.
2004: Wheat et al., 2011) and experiments were deployed within these perforated zones on a sensor string within the CORK casing (Edwards, Bach, et al., 2012). The sensor string included seals within the CORK casing to prevent fluid communication among the hydrologic zones.

2.2. Tracer Injection
On expedition AT39-01, 15.6 L of a 1.68 mol/kg CsCl tracer solution was injected into Hole U1383B (11,400 L), assuming that it would be observed in at least one of the three hydrologic levels in Hole U1383C (Figure 2a, Text S2, and Figure S2). This tracer solution was chosen because (1) it was a dense solution that should have descended within the borehole; (2) the difference in concentration between the well-mixed tracer concentration and seawater (2.1 nmol Cs/kg) was about 6 orders of magnitude; (3) Cs concentrations in seawater could be measured with an inductively coupled plasma mass spectrometer (ICPMS) with a precision of typically better than 5% of the seawater value, and thus even small amounts of the tracer could be detected in borehole fluids; and (4) even though dissolved Cs will react with basalt at low temperatures, removal of Cs into secondary minerals should be minimal for the 2-week duration of this experiment (Fisher et al., 2011; Solomon et al., 2009; Wheat et al., 2010).

2.3. Discrete and Continuous Samples
Discrete fluids samples were collected in 2012, 2014, and 2017 from each of the crustal intervals using an in situ pumping and collection system mounted on the ROV that included an in-line Aanderaa Optode (dissolved oxygen; DO) (Cowen et al., 2012) (Text S2). The umbilical was typically purged long enough to flush the umbilical volume more than once prior to sample collection into 15-L acid-washed bags (Figure S3). Upon recovery of the ROV, fluids were filtered into hot acid-washed (10% HCl) high-density polyethylene bottles. Some aliquots were frozen, and others were acidified with distilled HCl (4 mL HCl per liter of sample).

Fluid samples also were collected from downhole continuous fluid samplers with in situ acidification (OsmoSamplers) that were recovered in 2017 (Wheat et al., 2011). A second type of continuous fluid sampler employed a reverse osmosis (RO) cellulose acetate membrane and was deployed for several days in 2017 during which the intake was placed in the top of Hole U1383C when the hole was temporarily unsealed (Table 1). This membrane, and RO membranes in general, has a pump rate that decreases with time and temperature but does not change with the resistance induced by a sample coil (Text S2 and Figures S4 and S5). Given the known change in the pump rate with time and temperature, we designed the deployment of the RO sampler to mark changes in operations. This was accomplished by placing the intake into a bottle with a loose fit during the deployment. This bottle was filled with a diluted solution of CsCl; thus, changes in Cs and the major ions in seawater mark the timing when the sampler was deployed in the ocean. The Cs data also indicated the timing when the sample intake was removed from the loose fit bottle and collected bottom seawater prior to being placed in the top of the open borehole and again when the intake was returned to the bottle prior to recovery.

2.4. Analytical Techniques
Discrete and continuous fluid samples were analyzed for pH, alkalinity, and nitrate at sea (Gordon et al., 1994; Wheat et al., 2017) (Text S2). Ashore, acidified fluids were analyzed using standard techniques with an ICPMS and inductively coupled plasma optical emission spectrometer (ICPOES). Unacidified aliquots were titrated with AgNO₃ for chlorinity. Frozen samples were analyzed for nitrate, nitrite, phosphate, ammonium, and dissolved silica using an automated colorimetric system (Gordon et al., 1994). Oxygen respiration rates were determined during a 1.5-year period by measuring DO in filtered and unfiltered discrete aliquots stored at 4 °C in glass serum bottles and in the dark (Text S2).

2.5. Pressure and Temperature Measurements
Each of the boreholes also was instrumented with pressure and temperature sensors. Pressure was measured using a Paroscientific Model 887000-2 absolute gauge with temperature-compensated pressure resolution of ~2 Pa (0.2 mm water) (Edwards, Bach, et al., 2012). Autonomous temperature logging instruments included Antares Datensysteme GmbH (model 1857) and Onset Computer Corporation (model U12-015-3) (Edwards, Bach, et al., 2012). We present pressure and temperature data obtained in 2012 and 2017.
3. Results and Implications

3.1. Cross-Hole Tracer Experiment

In 2017 we conducted a cross-hole tracer experiment beginning with the injection of a salt-saturated Cs tracer solution into Hole U1383B (Figure 2a and Tables 1 and S1). This solution was denser than seawater. Fluid samples from OsmoSamplers that were in the open borehole at Hole U1383B and recovered 12 days later show a peak concentration of 410 μmol Cs/kg, but more typical anomalies were 7 to 20 nmol Cs/kg, compared to bottom seawater with ~2.1 nmol Cs/kg (Table S2). The timing of this peak was difficult to discern because of difficulties in the recovery (Text S2) and for long-term (5-year) deployments in general.

Although the timing was not certain, the Cs tracer reached the open borehole at a depth of 60.5 m bsf in Hole U1383B within a day(s) of the injection. Cesium concentrations were measured in discrete samples extracted from Holes U1383C and U1382A at each depth horizon before the seals to the boreholes were removed (Figures 2a and 3a and Tables 2 and S3). Each of these samples had seawater Cs concentrations. Thus, the Cs tracer solution did not reach the umbilicals at Hole U1383C within 5 days of the injection, even after the removal of ~300 L of formation fluid from each umbilical during sample collection (300 L represents a 7-m length of borehole or less than 13% of the volume of each hydrologic zone.). Furthermore, the downhole OsmoSampler from the upper hydrologic horizon was recovered from Hole U1383C when the top plug was recovered; however, there was no trace of excess Cs even 6 days after the tracer was injected (Table S4). This downhole OsmoSampler was positioned in the shallow interval (76.3–129.4 m bsf), with the intake at a depth of 102 m bsf.

Because Hole U1383C was overpressured (Figure S6), having a slightly positive pressure differential with respect to seafloor hydrostatic at the time of the tracer injection experiment, formation fluids could freely discharge from the open borehole (Figure 2b). Discharging formation fluids would have entered through the upper perforated section of the shallow interval (76.3–129.4 m bsf) (Figure S1). About 11.3 days after injection of the tracer, a RO OsmoSampler was deployed with the intake in the discharging fluids from Hole U1383C (Figure 3b and Table S5).

| Table 1 | Operations Related to IODP Holes 1383B and C in Reference to the Time of Tracer Injection |
|---------|---------------------------------------------------------------------------------------------|
| U1383B  | U1383C                                                                                      |
| Tracer Injected |                                                                                           |
| Deep umbilical sampled | 0.00 | 230 |
| Deep umbilical sampled | 1.10 | 70 |
| Shallow umbilical sampled | 2.88 | 70 |
| Deep umbilical sampled | 2.95 | 260 |
| Middle umbilical sampled | 3.71 | 70 |
| Top plug removed—shallow downhole | 6.10 | — |
| OsmoSamplers recovered | 11.31 | — |
| Downhole OsmoSamplers recovered | 12.04 | — |
| Top plug deployed | 12.07 | — |
| RO Osmo—recovered | 12.79 | — |
| RO Osmo—moved off wellhead | 14.04 | — |
| RO Osmo—on ROV platform | 14.11 | 210 |
| RO Osmo—on well head | 14.26 | 110 |
| Shallow umbilical sampled | 14.31 | 100 |

Note. A more detailed list of operations is presented in supporting information Table S1.
Within hours of this deployment the top plug from Hole U1383B was removed. Because Hole U1383B was slightly underpressured relative to hydrostatic (Figure S6), cold, dense bottom seawater could then descend into the formation (Figure 2c). This would likely increase the rate at which formation fluids flowed toward the open borehole at Hole U1383C.

The temporal record of Cs concentrations in the RO OsmoSampler is complex because of the experimental design (Figure 3b). Initially the intake for the RO OsmoSampler was placed in a container with dilute CsCl. Thus, fluids collected in the sample coil prior to deployment had high Cs concentrations but lacked seawater salts (Table S5). Once deployed, Cs concentration decreased toward seawater values. When the intake was

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**Figure 3.** Cs concentrations from discrete samples collected from umbilicals that were part of the CORK observatories (Holes U1382A and U1383C [black squares]) and a reverse osmosis (RO) OsmoSampler (red open squares) that was deployed when the top plug was removed from Hole U1383C are plotted as a function of time from the injection of a CsCl solution in Hole U1383B. (a) Cs concentrations for discrete samples are displayed with respect to times when the seals were removed and replaced on Holes U1383B and U1383C. (b) A RO OsmoSampler was deployed on the wellhead of Hole U1383C after the top plug was removed. Spikes in Cs concentrations result from Cs in the fluid reservoir that held the intake prior to and after deployment on the wellhead. (c) Data from the RO OsmoSampler that highlights the presence of Cs tracer in formation fluids that discharged from Hole U1383C with respect to the typical analytical range for bottom seawater (green lines).
removed from the container with the tracer, a Cs concentration spike resulted prior to returning to the seawater value (2.1 nmol/kg) (Figure 3c). Within the next day Cs tracer was observed in the RO OsmoSampler, indicating that the tracer made the 25 m lateral transit in less than 11.8 days. When the intake was removed from the wellhead the Cs concentration dropped to the seawater value then rose when the intake was repositioned within the container that still contained residue Cs (Figure 3c).

At the end of the experiment the top plugs were deployed to seal Holes U1383C and B and U1382A. Then a second set of discrete fluid samples was collected from the umbilicals at Holes U1383C and U1382A (Figure 3a). Discrete samples from each of the three depth horizons from Hole U1383C after the borehole was sealed had Cs concentrations in the range of 38.0–49.7 nmol Cs/kg, again confirming tracer flowed from Hole U1383B to Hole U1383C (Figure 2e and Table 2). In contrast, no excess Cs was measured in the discrete samples from Hole U1382A.

### 3.2. Time Series Data Reveal the Influx of Bottom Seawater

Discrete fluid samples were collected from umbilicals at Holes U1383C and B and U1382A. Then a second set of discrete fluid samples was collected from the umbilicals at Holes U1383C and U1382A (Figure 3a). Discrete samples from each of the three depth horizons from Hole U1383C after the borehole was sealed had Cs concentrations in the range of 38.0–49.7 nmol Cs/kg, again confirming tracer flowed from Hole U1383B to Hole U1383C (Figure 2e and Table 2). In contrast, no excess Cs was measured in the discrete samples from Hole U1382A.

### Table 2

*Concentrations of Selected Ions in Fluids Collected From the North Pond CORKs*

| Sample     | Dive          | Time (years) | pH | Alk | Oxygen | Nitrate | Si | V | U |
|------------|---------------|--------------|----|-----|--------|---------|----|---|---|
|            |               |              |    |     |        |         |    |   |   |
| 2012 BW    | 0             | —            | —  | —   | —      | —       | —  | — | — |
| 2014 BW    | 0             | 7.92         | 2.32| —   | 250    | —       | —  | — | — |
| 2017 BW    | 0             | 7.92         | 2.33| —   | 250    | 21.8    | 55 | 32| 14.6|
| 2014       | 1383C deep    | J2-623/630   | —  | —   | —      | 213     | —  | — | — |
| 1383C middle| J2-629/630    | —            | —  | —   | —      | 213     | —  | — | — |
| 1383C Shallow| J2-625/629   | 0.5          | —  | —   | —      | 216     | —  | — | — |
| 1382A      | J2-628       | —            | —  | —   | 244    | —       | —  | — | — |
| 2017       | 1383C deep    | J2-766       | 2.5| 8.10| 2.42   | 187     | 22.9| 146| 52| 12.5|
| 1383C middle| 2.5          |              |    |    |        |         |    |   |   |
| 1383C Shallow| J2-764/771   | 2.5          | 8.19| 2.44| 202    | 22.3    | 158| 53| 12.2|
| 1382A      | J2-768       | 2.5          | 7.95| 2.32| 233    | 22.5    | 73 | 27| 13.9|
| 2017       | 1383C deep    | J2-1026      | 6  | 8.06| 2.36   | 173     | 22.7| 159| 54| 13.0|
| 1383C middle| J2-1029      | 6            | 8.13| 2.45| 205    | 22.9    | 160| 63| 13.1|
| 1383C Shallow| J2-1028     | 6            | 8.07| 2.42| 198    | 22.8    | 151| 48| 12.5|
| 1382A      | J2-1027      | 6            | 7.91| 2.34| 228    | 22.3    | 70 | 29| 14.7|
| 1383C deep  | J2-1035      | 6            | 8.07| 2.35| 215    | 22.0    | 132| 45| 14.1|
| 1383C middle| J2-1035      | 6            | 8.01| 2.39| 219    | 22.0    | 130| 37| 13.4|
| 1383C Shallow| J2-1035     | 6            | 8.06| 2.35| 205    | 22.1    | 141| 37| 12.9|
| 1382A      | J2-1035      | 6            | 7.90| 2.31| 229    | 22.0    | 74 | 18| 13.8|

*Note. Additional analyses are listed in supporting information Table S3. Meyer et al. (2016). Tulley et al. (2017). Samples collected near the end of operations in 2017.*
U1383B had a greater impact with depth, consistent with higher Cs concentrations with depth (Figure 3a and Table 2). Furthermore, at the end of the experiment the DO concentration in the deeper interval in Hole U1383C was higher than the value in the shallow interval. The DO concentration was also higher in the middle zone than the shallow zone before and after the intrusion of bottom seawater. This consistency of heterogeneity of formation fluids, both prior to and after the intrusion of bottom seawater provides the first evidence to definitively document chemical heterogeneity in the upper basaltic crust, at least on the time scale of years. Thus, in this setting, convection was not vigorous enough on this spatial and temporal scale to homogenize the formation fluid composition in upper permeable basaltic crust. These data also provide a second measure of fluid flow in upper basaltic basement.

### 3.3. Hydrologic Implications for Upper Basaltic Crust

Our observations of the Cs tracer and bottom seawater allow us to estimate the minimum linear Darcy flow velocities in upper basement between Holes U1383B and U1383C. First, if the tracer descended Hole U1383B within hours after injection, and ascended Hole U1383C within hours, then the Darcy lateral flow velocity would be 2.1 m/day (25 m/11.8 days), based on the data from the RO OsmoSampler that was deployed in Hole U1383C (Figure 3c). This rate is similar to the linear velocity that was determined with tracers on the eastern flank of the JFR (2–3 m/day; Neira et al., 2016).

Taking into account the hydrologic setting in North Pond leads to a second estimate for the Darcy velocity. Prior to tracer injection, Hole U1383B was slightly underpressured or hydrostatic and Hole U1383C was overpressured. Thus, the lateral pressure gradient would have been counter to flow from Hole U1383B to Hole U1383C. However, once the seal at Hole U1383C was removed, formation fluid would have discharged, causing a subsurface “chimney” effect that would draw fluids from the formation. If we use the time of 6.1
3.4. Reactions Within the Basaltic Aquifer

In general, solute compositions provide a measure to assess disturbances caused by drilling operation. Such disturbances are documented in physical, chemical, and microbial conditions in the formation and can persist for months to years (Bullard, 1947; Jungbluth et al., 2016; Wheat et al., 2003). Similar changes in the chemical composition at North Pond resulting from drilling disturbances have been observed (Figure 4). For example, samples collected in 2012, about 6 months after drilling ceased, recovered particle-laden seawater descending in Hole U1383B. If tracer and seawater DO traveled fluids from Hole U1383C could result, in part, from continued inflow of bottom seawater after drilling operations ceased. Thus, microbial data from 2012 represent a mixture of community sources (bottom and surface (from drilling) seawater and formation fluids) (Meyer et al., 2016; Tully et al., 2018; Walter et al., 2018) and should be viewed in this context. Comparison of DO data from days after Cs injection when the top plug was removed from Hole U1383C as the start of flow between the two boreholes, then the estimated lateral Darcy flow velocity would be 4.4 m/day (25 m/(11.8–6.1 days)).

A third estimate for the minimum linear Darcy flow velocity between Holes U1383B and U1383C is based on the arrival of DO after Hole U1383B was open for 2.8 days prior to sample collection at Hole U1383C. Each of the three samples from Hole U1383C had elevated Cs concentrations and fluid from the deepest umbilical had the highest Cs concentration (Figure 3 and Table 2). This suggests a vertical component to fluid transport within the volcanic crust as tracer migrated more than a hundred meters (Figure 2e). This may indicate that vertical flow in the upper permeable layer is favored under conditions of a cold plume (from seawater descending in Hole U1383B). If tracer and seawater DO traveled from the base of Hole U1383B (89.8 m bsf) to the deepest umbilical (203 m bsf) in less than 2.8 days, the minimum estimated Darcy flow velocity would be 41 m/day. This value is a minimum because no samples were collected within the first 2.8 days after the seal was removed from Hole U1383B.

Using these three estimated values for the direct Darcy fluid velocity and estimated pressure differentials in the basaltic crust between Holes U1383B and U1383C, we apply Darcy’s Law to estimate upper crustal permeability at Hole U1383 (Figure 5). The simplified Darcy’s Law relates flow velocity (m/s) to the product of permeability (m²) and pressure gradient (Pa/m) divided by fluid viscosity (Pa·s). Temperature loggers from the shallow basement sections indicate temperatures of ~5 °C in both holes, corresponding to a viscosity of 1.62 × 10⁻³ Pa·s. The pressure difference could be as much as 10 kPa, but as low as 1 kPa (Figure S6). Figure 5 illustrates the range of permeability values that would be consistent with the differential pressures and the three estimates of the minimum Darcy flow velocities, resulting in a range of reasonable permeabilities from 10⁻¹¹ to 10⁻⁹ m². This range is significantly more permeable than values obtained with flow measurements and packers that interrogated longer depth intervals in uppermost basaltic basement in nearby Hole 395A (10⁻¹⁴ m²; Becker, 1990). It is also significantly more permeable than packer and flow measurements in similarly aged 6.9 Ma shallow basaltic crust on the Costa Rica Rift (10⁻¹⁴ m²; Becker, 1996), but upper basement there may be more sealed because of warmer temperatures due to a thick sediment cover, and more altered formation fluid. Our range of permeabilities is consistent with values obtained in uppermost basement in 0.9–3.4 Ma crust on the flank of the JFR, especially in two boreholes that were drilled into 0.9 and 1.3 Ma crust (10⁻¹² to 10⁻¹⁰ m²; Becker & Fisher, 2000; Becker & Davis, 2003). Our permeability range is also consistent with large-scale (>km), average upper crustal permeabilities (10⁻¹⁰ to 10⁻⁹ m²) that are consistent with the basement pore pressure response to seafloor tidal loading in young crust (Davis et al., 2000) and required to simulate subsurface fluid flow within a young ridge flank (Davis et al., 2004; Lauer et al., 2018).
Figure 6. The minimum percent DO that is consumed by nitrification as a function of time since deployment. The ratio of dissolved nitrate produced to DO consumed is based on Ziebis et al. (2012).

2014 and 2017 suggest that the formation fluid may have continued to rebound from drilling disturbances; however, other solute data suggest that the boreholes had chemically rebounded by 2014.

Another significant observation from the time series data is the increase in the nitrate concentrations in borehole fluids relative to bottom seawater (Figure 4 and Table 2). This increase has two potential sources, diffusion of nitrate from sediment pore waters into formation fluids or nitrification within the basaltic crust. A diffusive flux of nitrate from the sediment pore waters is not significant in this environment, given that the DO flux into the sediment is an order of magnitude greater than the diffusive flux of nitrate from the sediment and that transport-reaction models indicate that most of the DO consumption occurs in basaltic crust and diffusional losses to the sediment are minimal (Edwards, Bach, et al., 2012; Orcutt et al., 2013). Thus, observed increases in nitrate concentrations stem primarily from nitrification within the basaltic basement, not from diffusive fluxes from sediment pore waters.

On the basis of dissolved organic carbon (DOC), DO, and nitrate concentrations in crustal fluids, we estimate nitrogen cycling in this system as follows. Fourteen sediment cores from North Pond provide a consistent trend of nitrification in the sediment within North Pond, where one mole of nitrate was produced for every 13 moles of DO consumed (Ziebis et al., 2012). Applying the same ratio to crustal fluids, nitrification would consume about 10–60% of the DO loss in formation fluids with oxidation of basaltic minerals responsible for the remaining DO loss (Figure 6). Note that little of this DO loss is expected from the physical CORK installation, because the installation is comprised of fiberglass components with some steel components that were coated prior to deployment (Edwards, Bach, et al., 2012).

This nitrification result differs from results in similar thermal and hydrologic ridge flank hydrothermal settings, which do not indicate nitrogen cycling within upper basaltic basement (McManus et al., 2019; Wheat et al., 2017). Assuming that nitrification is due to heterotrophic microbial processes, a source of carbon is required. A potential source of carbon is pelagic sediments that are interspersed within the basaltic crust in this and other slow-spreading crust (Edwards, Bach, et al., 2012). However, the more likely source is DOC. DOC in borehole samples from North Pond collected in 2014 show a loss of 11 to 23 μM (Table 2). Therefore, if the change in DOC results in the consumption of DO by denitrification within the basaltic crust, which would require the removal of additional DO. Alternatively, if nitrification in crustal fluids is based on autotrophic microbial processes, for instance by ammonia-oxidizing archaea which have been documented in this environment (Tully et al., 2018), then the change in nitrate would not be linked to DOC-based processes. However, borehole fluids are typically depleted (<0.1 μM; Table S3) in ammonium. Such low ammonium levels cannot support the level of observed nitrate production. Thus, ammonia-oxidizing archaea are not the primary mechanism for generating nitrate in this system; however, the data set confirms that at least nitrification must occur in some form.

Changes in the alkalinity and dissolved silica data also reflect reactions within the basaltic crust. For example, we observed an increase in dissolved inorganic carbon (Meyer et al., 2016) and alkalinity (Figure 4) concentrations relative to bottom seawater. Such increases are, in part, a product of microbial production, but most of the increase stems from the dissolution of basaltic glass (McManus et al., 2019). Likewise, the increase in dissolved silicon results from seawater-basaltic reactions (Figure 4) (Wheat et al., 2017).

In contrast to the composition of crustal fluids from Hole U1383C, which reflect subsurface reaction processes, the chemical compositions of formation fluids from Hole U1382A are similar to bottom seawater values for most of the major, minor and trace ions. For example, the alkalinity at Hole U1382A is indistinguishable from bottom seawater (Figure 4). These results also are consistent with temperature data at a
depth of 153 mbsf (2.4 °C), almost identical to bottom seawater. However, some solutes (DO, V, Ba, and U) and nutrient (nitrate, dissolved silicon) concentrations are significantly different (Tables 2 and S3). A measurable change in DO and nitrate concentrations is consistent with nitrification. Nitrification affects fluid composition from Hole U1382A with a greater impact on the DO concentration loss. Here the ratio of DO consumed to nitrate produced is similar to the ratio measured in the sediment pore waters (Ziebis et al., 2012) (Figure 6). Considering that (1) Hole 395A is 56 m from Hole U1382A, (2) Hole 395A has a history of bottom seawater recharge (Becker et al., 1998; Gable et al., 1992; Gieskes & Magenheim, 1992; McDuff, 1984), and (3) umbilicals were severed during the CORK deployment at Hole 395A (Edwards, Bach, et al., 2012), bottom seawater may continue to flow into basement at Hole 395A and transfer to Hole U1382A, affecting its thermal and chemical composition, and by supposition the structure of the microbial community.

4. Refining Characteristics of Fluid Circulation Within Ridge Flank Hydrothermal Systems

Fluid compositions from North Pond exhibit similarities with formation fluids from Dorado Outcrop, a ridge flank hydrothermal system in the eastern Pacific (McManus et al., 2019; Wheat et al., 2017). Both have elevated alkalinities and dissolved inorganic carbon that stem, in part, from the dissolution of basaltic glass (McManus et al., 2019). However, formation fluids from North Pond contain dissolved nitrate concentrations greater than that of bottom seawater, consistent with nitrification, whereas fluids from Dorado Outcrop do not exhibit this pattern (Wheat et al., 2017). This difference may stem from different hydrologic conditions. Zhao et al. (2019) report nitrification rates of $10^{-5}$ to $10^{-7}$ mol·m$^{-3}$·year$^{-1}$ in sediment from North Pond. To assess if this rate, based on data from the sediment column, may be applicable to the basaltic basement, we conducted an oxygen respiration experiment on discrete borehole samples at 4 °C and in the dark (Table S6). Although the change in the DO concentrations with time were near the limit of detection for the method used, the estimated rate of biological oxygen consumption ranged from 0.4–6 nmol·L$^{-1}$·day$^{-1}$ ($10^{-4}$ to $10^{-3}$ mol·DO·m$^{-3}$·year$^{-1}$), several orders of magnitude greater than the nitrification rate (Zhao et al., 2019). Assuming heterotrophy and this nitrification rate, 100 to 10,000 years would be required to accumulate a 1 μM nitrate anomaly in the formation fluid. This amount of time is consistent with conclusions from other work at North Pond (Walter et al., 2018), but orders of magnitude greater than the time required to reduce DO by 50 μM (20–400 years). These rates are still at least an order of magnitude greater than the residence time of years calculated for the ridge flank hydrothermal system associated with Dorado Outcrop (Wheat & Fisher, 2008).

Likewise, North Pond hydrologic data are similar to and different from other ridge flank hydrothermal systems. As noted above the minimal permeability range of $10^{-11}$ to $10^{-9}$ m$^2$ for uppermost basement is consistent with measurements in younger crust on the JFR, but not with 6.9 Ma crust on the Costa Rica Rift where the crust may be more sealed because of the thicker, more continuous sediment cover increased the basement temperature and extent of crustal alteration (Becker, 1996; Becker, et al., 2004 and 2013; Becker and Fisher, 2000 and 2008). In the more discontinuously sedimented, slow-spread North Pond environment, crustal faults and fractures may remain open longer leading to higher permeabilities. On a regional scale, subsurface fluid flow beneath North Pond is likely distinct from ridge flank hydrothermal systems on medium to fast spreading crust, because of the numerous and close proximity of exposed volcanic structures (Figure 7). These topographic highs have a north-south fabric consistent with the trend in active spreading to the east. Subsurface flow probably follows this trend as it does on the eastern flank of the JFR where off-axis abyssal hill-related faulting dictates subsurface flow patterns (Fisher et al., 2003; Hulme & Wheat, 2019; Karsten et al., 1998; Wheat et al., 2000). In contrast, subsurface flow at Dorado Outcrop is guided by seamounts without a defined trend associated with crustal accretion (Wheat et al., 2019).

This new data set from North Pond represents the first tracer-based hydrologic study of a low temperature ridge flank hydrothermal system on slow-spread crust and the first to be conducted during a single expedition. We deployed a CsCl tracer and used a natural tracer (bottom seawater) to elucidate hydrologic and biogeochemical processes within the upper permeable portion of the volcanic crust. Our results indicate that fluid transport in the upper oceanic crust is horizontal and vertical, highlighting the extent of fluid connectivity and partitioning in the oceanic crust on the scale of tens to hundreds of meters. Our data are also...
consistent with convection that is not vigorous enough to homogenize the upper permeable volcanic crust in this setting at least over a 6-year period. Such a result is in contrast to the modeled vigor of circulation in medium to fast spreading crust (Davis et al., 2004).

Given the vastness of the oceanic crust, surveying the entire ocean seafloor is not fiscally feasible, thus we need to utilize seafloor boreholes in characteristic settings, such as North Pond, to elucidate hydrologic, biogeochemical, and microbial process that are scalable to the global network of oceanic environments (Edwards et al., 2012). Our results illustrate that such boreholes hold the potential for manipulative experiments during a single expedition to further constrain subsurface hydrologic and biogeochemical processes, using, for example, isotopically labeled elements or compounds to surmise microbial rates of consumption and metabolic pathways.

Data Availability Statement

All of the data used in this research are freely available and may be downloaded through the links detailed in the supporting information and at the MGDS IEDA database. The MGDS IEDA database includes the pressure data (at http://www.marine-geo.org/tools/search/entry.php?id=AT39-01 and http://www.marine-geo.org/tools/search/entry.php?id=MSM-37).

Author Contributions

Each author, except C. P., participated in seagoing operations that included sensor, sampler, and tracer operations and data and sample recovery. K. B. and H. V. collected the pressure and temperature data. B. N. O. conducted the oxygen consumption experiment. T. F. C. P., and C. G. W designed the tracer experiment. C. P., T. F., and A. H., conducted the chemical analyses. C. G. W. wrote the initial manuscript with major revisions by K. B. and B. N. O. All authors contributed to the ideas and commented on the manuscript.

Competing interests:

The authors declare no competing interests.
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