



Temperature dependence of chemical exchange during seafloor weathering: Insights from the Troodos ophiolite

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Abstract

Chemical fluxes associated with low-temperature, off-axis, alteration of the upper oceanic crust (seafloor weathering) may play an important role in controlling the composition of the ocean and the long-term carbon cycle. However, it is challenging to quantify these fluxes and how they change with changing bottom water temperature over geological timescales. Here we study the exchange of major elements associated with seafloor weathering in the exceptionally preserved Cretaceous Troodos ophiolite and compare them to less well constrained data from drill cores from the modern ocean basins. Calcite O-isotope thermometry from four traverses through the lavas reveals a well-ventilated region at the top of the lava pile where alteration occurred at near-constant temperatures similar to that of bottom water. The lithological makeup of the crust appears to control the thickness of this region, with increased abundance of sheet flows marking the base of this zone. Maintaining low-temperatures in the well-ventilated region requires large fluid fluxes facilitated by the high permeability of the pillow lavas. Large-scale addition of CO₂ to the crust only occurs in this well-ventilated region indicating that the alkalinity producing reactions required for calcite precipitation occurred at temperatures sensitive to bottom water temperature. Comparison of whole rock geochemical data for samples from the well-ventilated zone with volcanic glass compositions shows that the changes in rock compositions due to seafloor weathering are large (roughly −4 wt% SiO₂, +0.5 to 1 wt% MgO, −5 wt% CaO, −0.5 wt% Na₂O, +3.5 wt% K₂O and +2.5 wt% CO₂). Comparison to data from drill cores from modern oceanic crust that was also altered under well-ventilated conditions suggests that bottom water temperature plays a major role in controlling the chemical exchange between the ocean and lavas; parameterizations for the associated fluxes are provided. Because elemental exchange between the ocean and oceanic crust globally depends strongly on bottom water temperature, fixed values for these fluxes cannot be used in models of the evolution of seawater chemistry or subduction fluxes.

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1. INTRODUCTION

The major ion composition of seawater is important in many aspects of the Earth system from its effect on the carbon cycle (via alkalinity) to modulating biomineralization (e.g. Sundquist, 1991; Porter, 2010). Additionally, because the major ion composition of the ocean reflects the integrated fluxes of ions into and out of the ocean, an under-

standing of the controls on ocean chemistry, along with a history of ocean chemistry, may allow the history of these fluxes to be unraveled (e.g. Spencer and Hardie, 1990; Demicco et al., 2005). Of the fluxes that play important roles in controlling ocean chemistry, perhaps the least well-constrained is that associated with low-temperature alteration of the upper oceanic crust (lavas) in off-axis hydrothermal systems. These systems operate across the abyssal plains starting close to the ridge axis and continuing until sufficient sediment accumulates to inhibit significant fluid exchange between the oceans and crustal aquifer

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(i.e. the high porosity and permeability lavas; Stein and Stein, 1992; Mottl and Wheat, 1994; Hasterok, 2013).

Chemical fluxes associated with off-axis hydrothermal circulation can potentially be determined either from changes in fluid or rock compositions. If the average change in fluid composition in the crust and the total fluid flux could be determined, the chemical flux could be calculated. However, finding fluids venting off-axis is incredibly challenging and estimates of aquifer fluid compositions from sediment pore fluid profiles suggest local hydrological conditions lead to massive variability in fluid composition; this makes using these to determine global average aquifer fluid compositions very difficult. Alternatively, if the average change in rock composition induced by off-axis fluid-rock reaction can be determined, and the total mass of rock altered in such systems estimated, the chemical flux can be calculated. It is this latter approach we use here, and we discuss the system from the perspective of the time-integrated change in rock composition that, via mass balance, reflect time integrated fluxes into the ocean.

The principal controls on the time-integrated chemical fluxes associated with off-axis hydrothermal systems are the temperature of fluid-rock reaction, the fluid flux and the composition of seawater. The temperature of fluid-rock reaction depends both on bottom water temperature (Brady and Gislason, 1997; Gillis and Coogan, 2011; Coogan and Dosso, 2015; Krissansen-Totton and Catling, 2017) and the extent of heating of the fluid in the aquifer; the latter depends on hydrological conditions, in particular sediment cover and aquifer permeability (Fisher and Becker, 2000; Anderson et al., 2012; Winslow et al., 2016). The same hydrological parameters control the fluid flux. Bottom water chemistry is potentially important in controlling chemical exchange between the crust and ocean because, in general, fluid fluxes are sufficiently large that the fluid composition for many species is similar to seawater even after fluid-rock reaction (Wheat et al., 2017; Coogan and Gillis, 2018). Unraveling the relative roles of the different factors in controlling chemical fluxes between the ocean and crustal aquifer in off-axis hydrothermal systems is complex because of the heterogeneity of hydrological conditions, limited sampling, and generally poor recovery of modern lavas through drilling. However, if we are to understand the role of off-axis hydrothermal systems in the evolution of seawater chemistry it is important to attempt to quantify the chemical fluxes as functions of the controlling parameters.

The Troodos ophiolite, Cyprus, is perhaps the only ophiolite that preserves a typical low-temperature alteration history analogous to that found in drill cores from the modern oceanic crust, being largely unaffected by obduction related overprinting or rapid early sedimentation (Gillis and Robinson, 1988; Gillis and Robinson, 1990). It also allows us to explore the controls on chemical exchange during low-temperature alteration of the lavas in ways that are not possible in drill cores from the modern ocean basins due to good exposure over a wide area. Additionally, there is a high-recovery (>90%) drill core through the lavas (Gibson et al., 1991). The Troodos ophiolite was formed (91.6 ± 1.4 Ma; Mukasa and Ludden, 1987) and altered

(Gallahan and Duncan, 1994; Booij et al., 1995) in the late Cretaceous, a time of high global temperatures (Cramer et al., 2011; Friedrich et al., 2012). Because surface temperature and bottom water temperature are strongly correlated (Krissansen-Totton and Catling, 2017) this ophiolite provides a good example of crust altered under warm bottom water conditions. The sedimentary record preserves a history of initially very slow, deep marine, sedimentation (Bear, 1960) with shallow marine conditions (e.g. reefal limestones) appearing in the Miocene (e.g. Follows and Robertson, 1990) as the ophiolite was uplifted (i.e. well after low-temperature alteration occurred). The spreading rate the ophiolite formed at is uncertain, but the continuous sheeted dike complex, general lack of crustal dismemberment, and limited topography on the lava-sediment boundary suggest either an intermediate- to fast-spreading rate or a magma supply rate consistent with these spreading rates.

Here we use whole rock geochemistry and carbonate mineral O-isotope thermometry (henceforth referred to as calcite although definitive mineralogical tests have not been performed on all samples) to investigate chemical exchange between the ocean and upper oceanic crust in off-axis hydrothermal systems. We show that the region of the crust in which the aquifer was well-ventilated, such that the aquifer temperature was similar to that of bottom water, depends largely on the lithological characteristics of the crust. Using samples from this well-ventilated area, and assuming that cogenetic volcanic glasses define fresh-rock compositions, we quantify the magnitude of chemical exchange during low-temperature alteration in this setting. Comparison of these chemical changes to those from drill cores in modern ocean crust that also underwent alteration under well-ventilated conditions suggests that bottom water temperature plays a key role in controlling major element fluxes between the ocean and upper oceanic crust. This dependence of the extent of alteration of the well-ventilated region on bottom water temperature means that use of the term “seafloor weathering” to describe this process is appropriate in that it makes the link between the control of climate, through bottom water temperature, and the extent of chemical exchange.

2. ANALYTICAL METHODS

Whole rock samples were crushed in an agate planetary mill and major element compositions were determined by XRF, and LECO for C, at Acme labs, Vancouver (Supplementary Table S1; previously reported in Coogan et al., 2017). Additionally, major element compositions measured on the CY1/1A drill cores as part of the Cyprus Crustal Study Project are also used (Gibson et al., 1991). Many of these drill core samples were analysed in up to four different laboratories and the averages of the data reported for these samples were calculated and used. The portion of the calcium hosted in silicate phases (CaO_{sil}) was determined by correcting for Ca in calcite by assuming all C exists as CaCO_3 (i.e. $\text{CaO}_{\text{sil}} = \text{CaO} - (56/44) * \text{CO}_2$). Chromium and Ni were analysed by solution ICP-MS. After dissolution using a HF:HNO₃ mixture samples were analysed on a Thermo X-Series ICP-MS at the University of Victoria

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