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Magnesium isotopic composition of altered oceanic crust and the global Mg cycle

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Abstract

To investigate the behavior of Mg isotopes during low-temperature alteration of oceanic crust and to further understand its role in the global Mg cycle, we measured the Mg isotopic compositions ($^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$) of a set of samples of altered oceanic crust (AOC) recovered from the Ocean Drilling Program Hole 801C, the reference site for old crust (~170 Ma) subducting in the Pacific. The measured $\delta^{26}Mg$ values range from −1.70‰ to 0.21‰, deviating from that of pristine oceanic basalts (−0.25 ± 0.07‰). Composite samples of volcanoclastic breccia that have experienced relatively intense alteration have larger variation in $\delta^{26}Mg$ values (−1.01‰ to 0.15‰) than composite samples of massive basaltic flows (−0.53‰ to −0.04‰), indicating significant Mg isotope fractionation during low-temperature alteration of the oceanic crust. Moreover, the upper off-axis basement has on average lower $\delta^{26}Mg$ values (−1.70‰ to −0.04‰) than the lower on-axis basement (−0.16‰ to 0.21‰). These findings, combined with the co-variations between MgO content and FeO^*/CaO ratio and between $\delta^{26}Mg$ and FeO^*/CaO ratio, suggest that formation of Mg-bearing minerals (i.e., saponite and calcite) during low-temperature alteration of the oceanic crust accounts for the highly variable $\delta^{26}Mg$ of AOC. Early formation of saponite under anoxic condition preferentially takes up heavy Mg isotopes and accounts for Mg enrichment and relatively high $\delta^{26}Mg$ in the on-axis basement. Subsequent precipitation of carbonates results in the dilution of Mg and relatively low $\delta^{26}Mg$ in the off-axis basement. In addition, accumulation of carbonate-rich interflow sediments in the upper basement may contribute further to the low $\delta^{26}Mg$. A weighted average $\delta^{26}Mg$ value of 0.00 ± 0.09‰ is estimated for the AOC at Site 801, implying that low-temperature alteration of oceanic crust drives the ocean to a lighter Mg isotopic composition, and thus requires additional carbonate precipitation to maintain a steady-state Mg isotopic composition of seawater. A mass balance calculation suggests that the Mg output flux due to low-temperature alteration of the oceanic crust equals ~12% of the annual Mg riverine input, indicating that AOC is a significant sink of Mg in seawater. Our study further highlights that recycling of AOC with highly variable

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δ^{26}Mg along with overlying marine sediments into the mantle through subduction may generate Mg isotopic heterogeneity in the mantle at small scales.

Keywords: Magnesium isotopes; Oceanic crust; Low-temperature alteration; Mg cycle; Mantle heterogeneity

1. INTRODUCTION

Oceanic crust undergoes extensive seawater alteration prior to subduction into trenches. This process not only modifies compositions of seawater and the upper oceanic crust (Hart et al., 1974; Seyfried et al., 1984; Elderfield et al., 1999; Chan et al., 2002), but also regulates long-term climate change by uptake of carbon (Staudigel et al., 1989, 1996; Alt and Teagle, 1999; Coogan and Gillis, 2013). Ultimately, the altered oceanic crust (AOC) is recycled into the mantle by subduction, making an important contribution to mantle heterogeneity (Hofmann, 1997; Kelley et al., 2005; Chauvel et al., 2008). Thus, knowledge of the composition of AOC is pivotal to understanding the cycling of elements and its impact on the composition of the Earth's mantle.

As a major element in both the silicate Earth and hydrosphere, Mg is critical in deciphering the chemical evolution of the oceans and change of the global climate, which is borne out by the co-variation between seawater Mg/Ca and climate throughout Earth's history (Hardie, 1996; Stanley and Hardie, 1998; Demicco et al., 2005). In general, Mg is transferred from continents to the oceans via river and groundwater discharge, and is sequestered mainly by carbonate deposition and alteration of the oceanic crust on the ridge axis and the ridge flank (Drever, 1974; Elderfield and Schultz, 1996). Based on heat flow anomalies and compositions of hot springs, between 10% and 40% of the riverine input of Mg is estimated to be taken up by hydrothermal alteration of the oceanic crust along the axis (Mottl and Wheat, 1994). A higher uptake flux of Mg is expected during low-temperature alteration of the oceanic crust on the ridge flank since approximately two-thirds of the hydrothermal heat is lost there (Mottl, 2003). However, accurate assessment of the Mg uptake flux associated with low-temperature alteration of the oceanic crust is still not clear due to the large variation in alteration temperature and the small change in composition of the ridge flank fluids (Staudigel, 2014). This hinders our understanding of the controlling factors on the cycling of Mg in the oceans and the linkage between climate and seawater chemistry.

Magnesium isotopes may provide a potential constraint on the geochemical cycling of Mg in the oceans and the magnitude of Mg flux into the oceanic crust during low-temperature alteration, because the main inputs and sinks of seawater Mg have distinctive isotopic compositions. The current riverine input has an average δ^{26}Mg value of −1.09‰ (Tipper et al., 2006), which is slightly lighter than modern seawater (-0.83 ± 0.1‰, Foster et al., 2010; Ling et al., 2011). The δ^{26}Mg of the total high-temperature hydrothermal Mg sink should be equal to that of seawater since removal of Mg is quantitative in high-temperature hydrothermal systems (Tipper et al., 2006; Higgins and Schrag, 2010). By assuming a steady-state Mg isotopic composition for the oceans, the isotopically light riverine input has to be balanced by the combined removal of Mg through deposition of carbonates and low-temperature alteration. Thus, the behavior of Mg isotopes during low-temperature alteration of the oceanic crust is crucial to understanding Mg cycle in the oceans. However, this is still poorly known.

Knowledge of the Mg isotopic composition of the AOC can also provide valuable insight into the role of subduction in creating chemical heterogeneity in the mantle. Previous Mg isotopic measurements of global peridotite xenoliths and oceanic basalts led to the inference of a homogenous Mg isotopic composition of the mantle (-0.25 ± 0.07‰, Teng et al., 2010a). However, other mantle and mantle-derived rocks, including metasomatized mantle xenoliths (Pogge von Strandmann et al., 2011; Xiao et al., 2013; Hu et al., 2016b), continental basalts (Yang et al., 2012; Huang et al., 2015b; Liu et al., 2015; Tian et al., 2016; Li et al., 2017; Su et al., 2017) and cratonic eclogites (Wang et al., 2012, 2015; Huang et al., 2016) commonly have relatively low δ^{26}Mg values (-0.38 ± 0.23‰). These distinct Mg isotopic signatures are proposed to derive from AOC and marine sediment subduction into the mantle. This interpretation was supported by a first-order mass balance calculation showing that the bulk AOC has lighter Mg isotopic compositions than the unaltered oceanic crust (Teng et al., 2010b). However, recent studies of altered oceanic basalts recovered from IODP Sites 1256 (Huang et al., 2015a), U1365, U1367, and U1368 (Zhong et al., 2017) support a bulk AOC with Mg isotopic composition similar to or slightly heavier than that of the mantle. The contradiction between the mass balance approach and actual measurements of the Mg isotopic composition of the AOC has become a stumbling block in our understanding of the global Mg cycle.

In order to provide clarity to this conundrum, we present new Mg isotopic measurements of AOC samples recovered from the Ocean Drilling Program (ODP) Hole 801C outboard of the Mariana trench in the western Pacific (Fig. 1). This site offers some benefits in evaluating the elemental and isotopic effects of low temperature seafloor alteration and the potential influence of AOC subduction on mantle geochemistry. The reasons are twofold: (1) this site is in the oldest oceanic crust of the Pacific (~170 Ma, Koppers et al., 2003) and has undergone a full history of low-temperature alteration (Alt and Teagle, 2003); (2) this site lies in the fast spreading crust of the subducting Pacific