



Massive magnesium depletion and isotope fractionation in weathered basalts

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Abstract

Magnesium isotopes have been shown to fractionate significantly during continental weathering, however, the degree and direction of fractionation varies from one study to the next, and the main factors that control Mg isotope fractionation during weathering have yet to be delineated. Here, we report Mg contents and isotopic compositions for two ~10 m deep drill cores through bauxites developed on Columbia River Basalts (CRBs). Samples from these two drill cores have very low MgO contents (0.12–0.25 wt.%) and variable but high $\delta^{26}\text{Mg}$ values (from -0.1 to up to $+1.8$, the heaviest isotopic composition ever reported for Mg) relative to the average values of fresh CRBs ($\delta^{26}\text{Mg} = -0.23 \pm 0.07$ and $\text{MgO} = 5.9$ wt.%). These features reflect the near complete loss of Mg in the isotopically heavy regolith. The most highly weathered bauxites at the tops of the profiles have the lowest $\delta^{26}\text{Mg}$ values, reflecting the addition of isotopically light eolian dust. Excluding these samples, $\delta^{26}\text{Mg}$ in bauxites displays a weak, positive correlation with gibbsite abundance, suggesting that gibbsite preferentially retains ^{26}Mg in the bauxites. The integrated Mg isotopic fractionation factors between bauxite and fluid ($\Delta^{26}\text{Mg}_{\text{bauxite-fluid}}$), inferred from Rayleigh fractionation calculations, vary from 0.05‰ to 0.4‰, which is similar to the range inferred from previous studies of igneous rock weathering profiles, though none of these previous studies found the extremely heavy $\delta^{26}\text{Mg}$ and massive Mg depletion seen here. Collectively, studies of weathering profiles suggest loss of isotopically light Mg from the continents, which must contribute to the very light $\delta^{26}\text{Mg}$ seen in river water and seawater.

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1. INTRODUCTION

Non-traditional stable isotopes are increasingly used as tracers of chemical weathering (e.g., Li, Si, Fe). Magnesium isotopes have several advantages in this regard. First,

magnesium is a major element in the continental crust (e.g., Taylor and McLennan, 1995; Rudnick and Gao, 2003), is water-soluble, and is systematically transported from the continental crust into the oceans via chemical weathering and river transport. It has therefore been suggested that chemical weathering may help to explain the non-basaltic composition of the bulk continental crust (e.g., Albarède, 1998; Lee et al., 2008). Magnesium has only one redox state (+2), and is thus insensitive to changes in oxygen fugacity (cf. Fe, Cu, Mo, etc.). In addition, there is no significant fractionation of Mg isotope in bulk igneous rocks due to crystal fractionation (e.g., Liu et al., 2010;

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Teng et al., 2007, 2010b). Magnesium is a bio-nutrient (Epstein and Bloom, 2005 and references therein), and is susceptible to isotopic fractionation in plants (e.g., Black et al., 2008), though previous studies of the role of plants in Mg isotopic fractionation in soils conclude that they have a very small influence (<1% of Mg, by mass) (Tipper et al., 2010; Bolou-Bi et al., 2012; Opfergelt et al., 2012). Most importantly, Mg isotopes in both soils and river waters are greatly fractionated from the mantle value, indicating that they are significantly influenced by chemical weathering (e.g., Tipper et al., 2006a, 2008, 2010, 2012a,b; Pogge von Strandmann et al., 2008; Jacobson et al., 2010; Teng et al., 2010a; Wimpenny et al., 2010; Huang et al., 2012; Opfergelt et al., 2012).

Nevertheless, the direction of fractionation varies from study to study and the main factors that control Mg isotope fractionation during chemical weathering are under debate. In a study of a weathering profile developed on a meta-diabase, Teng et al. (2010a) suggested that Mg isotope fractionation follows simple Rayleigh fractionation and is controlled by the formation of secondary minerals, producing regoliths enriched in heavy Mg isotopes by up to 0.65‰. Similarly, the experimental study of Ryu et al. (2011) suggested that preferential dissolution of isotopically light minerals, such as chlorite ($\delta^{26}\text{Mg}$ is as low as -1.82), may lead to isotopically light Mg in solution ($\delta^{26}\text{Mg}$ down to -1.59) and isotopically heavy Mg in the regolith. Finally, Huang et al. (2012) studied a saprolite profile developed on basalt in Southern China, suggesting the same sense of Mg isotopic fractionation and found that $\delta^{26}\text{Mg}$ correlates with the abundance of kaolin minerals, which was attributed to adsorption of heavy Mg isotopes by these clays.

By contrast, Pogge von Strandmann et al. (2008) analyzed Icelandic rivers and found a very large range in $\delta^{26}\text{Mg}$ (-0.96 to 0.64), suggesting that some secondary minerals take up heavy Mg isotopes, whereas others, such as allophane that formed under glaciers, preferentially incorporate light Mg isotopes, leaving residual water to be isotopically heavy. In another experimental study, Wimpenny et al. (2010) found that light Mg isotopes are preferentially incorporated into water due to dissolution of certain minerals, such as forsterite, and then precipitation of certain secondary minerals (chrysotile) that preferentially remove light Mg isotopes from solution, leaving the residual water with a heavy Mg isotopic composition. Note that the Mg isotope behavior in the experimental results of Wimpenny et al. (2010) is the opposite to most natural observations (except for the previously described allophane-bearing soils from Iceland of Pogge von Strandmann et al., 2008), where waters tend to be isotopically light and regoliths heavy.

Some of the confusion related to the direction of Mg isotopic fractionation in topsoils (no more than 2 m from the surface) may be explained by the formation of soil exchange complexes, which consist of charged secondary clays and organic compounds that retain Mg, as well as other exchangeable cations (Opfergelt et al., 2012). Thus, in some cases the observed low $\delta^{26}\text{Mg}$ in topsoils (compared to the unweathered parental rock) may be due to preferential retention of light Mg isotopes by these exchange complexes (Opfergelt et al., 2012, 2014).

Here we report Mg isotope compositions for two sets of bauxite drill cores (starting from ~ 2 m below the surface), fresh parental basalts, as well as two eolian deposits in order to determine the factors controlling Mg isotope fractionation during basalt weathering, especially under intense weathering conditions. Our findings have implications for understanding the Mg isotopic compositions of rivers and seawater and help to illuminate the role of chemical weathering in changing the bulk composition of the continental crust.

2. SAMPLES

The bauxites studied here are the same as those investigated in a companion Li isotope study (Liu et al., 2013). These bauxites developed on Columbia River Basalts (CRBs), flood basalts that erupted between 17 Ma and 6 Ma in the Pacific Northwest of the United States, that crop out both west and east of the Cascade mountains. The bauxites were recovered in two drill cores that penetrated highly weathered CRBs from the western, wet side of the Cascades, where present-day mean annual temperature (MAT) and mean annual precipitation (MAP) are ~ 11 °C and ~ 1100 mm, respectively (<http://www.wrcc.dri.edu>). Paleo-environmental evidence suggests that in the middle to late Miocene, around the time that the CRBs erupted, climate in the inland Pacific Northwest was warmer and wetter than today (Takeuchi et al., 2010). The samples in this study include: (1) fresh parental basalts from two flows of the Columbia River Basalts upon which the bauxites developed (sampled from outcrops): the Pomona Member of the Saddle Mountains Basalt and the Sentinel Bluffs Member of the Grande Ronde Basalt; (2) two ~ 2 –10 m drill cores through bauxites developed on these two flows; (3) two samples of recent eolian deposits (Portland Hills Silt and Palouse Formation), which may represent eolian additions to the weathering profiles.

Mineralogy, elemental geochemistry as well as Li and Nd isotopes were previously reported in Liu et al. (2013), and therefore only a brief summary is provided below. Both of the drill core bauxites contain gibbsite, hematite, \pm halloysite, kaolinite, goethite, and maghemite; quartz, which is not a primary mineral of the basalts, occurs at the top of both drill cores and its abundance decreases progressively with depth (Table A.1). Major and trace elements are depleted in the bauxites compared to the parental basalts. Extreme weathering recorded by these bauxites is also seen in the extreme CIA values (defined in molar ratio as $\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO}^* + \text{Na}_2\text{O} + \text{K}_2\text{O}) \times 100$, where CaO^* is the CaO that is not from carbonate or phosphate (Nesbitt and Young, 1982)), ranging from 98.5 to 99.9, while fresh CRBs have values between 37 and 40 (Liu et al., 2013). Significant Li depletion is seen in the bauxites and $\delta^7\text{Li}$ values in both bauxite profiles decrease systematically towards the surface, and are systematically lower than the average of the fresh basalt parents. Similarly, [Nd] is significantly depleted relative to fresh basalts; $^{143}\text{Nd}/^{144}\text{Nd}$ in the bauxites overlap those of the fresh basalts at depth, but trend towards less radiogenic values towards the surface. Liu et al. (2013) quantitatively demonstrated the importance

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