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Letters

Mechanisms of magnesium isotope fractionation in volcanic soil weathering sequences, Guadeloupe

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ABSTRACT

Magnesium (Mg) stable isotopes are increasingly used as a weathering proxy in soils and rivers, but the impact of the mineralogy of secondary phases on isotope fractionation remains obscure. A better understanding of the behaviour of Mg isotopes during weathering processes is a mandatory step toward deployment of this new tracer for understanding chemical fluxes exported from the critical zone. Here we investigate isotopic variations in $\delta^{26}\text{Mg}$ in bulk soils and clay fractions relative to their parent andesite in three soil weathering sequences from Guadeloupe formed under contrasting climatic conditions. Soils formed in drier conditions (low precipitation) contain smectite, whereas soils formed under wet conditions (high rainfall) are characterized by halloysite and Fe-oxides or kaolinite.

All clay fractions have Mg isotopic compositions ($\delta^{26}\text{Mg} - 0.41\text{‰}$ to -0.10‰) similar to or heavier than their parent andesite ($\delta^{26}\text{Mg} - 0.47\text{‰}$) supporting the preferential incorporation of heavy Mg isotopes in secondary Mg-bearing clay minerals with the first direct measurements on clay fractions. Soils with lighter Mg isotope compositions have greater quantities of exchangeable Mg. The data support a contribution from sea spray to the exchangeable Mg pool correlated to the soil weathering degree. This study highlights for the first time that the soil $\delta^{26}\text{Mg}$ not only depend on $\delta^{26}\text{Mg}$ of the parent rock, and on any fractionation that might occur, but also on the Mg retention on the exchange complex, which could in turn be controlled by external inputs such as sea spray.

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1. Introduction

The chemical weathering of continental Ca–Mg silicate rocks influences global climate by consuming atmospheric CO_2 (Bernier, 1995). Chemical weathering of the host regolith dominantly occurs at the soil–rock interface (White et al., 1998; Brantley, 2010). Therefore, investigating the processes responsible for elemental transfers in the critical zone is fundamental for improving estimates for continent derived input fluxes to the ocean and thus the long-term atmospheric C-budget.

Magnesium (Mg) is the eighth most abundant element in the continental crust (Taylor and McLennan, 1985) and the fourth most abundant species in seawater (Millero, 1974). It is transferred from the continents to the oceans via rivers. During weathering reactions,

Mg derived from dissolution of primary minerals may be incorporated into secondary minerals (Borchardt, 1989). Magnesium is also retained on the soil exchange complex, the loss from which constitutes an important source of cations to rivers (Markewitz et al., 2001), representing up to 30% of the annual cation export (Miller et al., 1993). Crucially, Mg is also an essential plant nutrient used for the synthesis of chlorophyll (Epstein and Bloom, 2005), and is released back to the soil via litterfall degradation.

The recent advent of high-precision measurement of stable Mg isotopes offers an unprecedented opportunity to obtain new insights into the sources and processes that control the release of chemical elements from the critical zone and therefore, for better understanding of riverine fluxes (Gaillardet et al., 2010). Magnesium isotopes (^{24}Mg , ^{25}Mg , ^{26}Mg) are fractionated during uptake by plants (Black et al., 2008; Bolou-Bi et al., 2010), and through incorporation into secondary silicate minerals. One current view is that heavy Mg isotopes are incorporated into secondary silicate phases, producing lighter riverine signatures in silicate catchments (De Villiers et al., 2005; Brenot et al., 2008; Tipper et al., 2008a; Wimpenny et al., 2011). This is based on isotope measurements of bulk soils that were found to be

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isotopically heavier than their parent silicate bedrock (Teng et al., 2010; Tipper et al. 2006a), or of detrital smectite Mg heavier than the bulk silicate Earth (Tipper et al., 2010). In contrast, it has also been suggested from an experimental study that fluids can become heavier following precipitation of secondary phases (Wimpenny et al., 2010), although the Mg isotope composition of the secondary phase was not obtained in latter study, leaving some uncertainty. Also, one soil derived from basalt is reported to be isotopically lighter than the basalt (Pogge Von Strandmann et al., 2008). Thus far, the mechanisms controlling Mg retention in the soil and the secondary phases driving the isotope fractionation have not been identified. Here, a direct comparison between Mg isotope compositions in secondary clay minerals and their parent silicate material should provide new insights to better constrain the behaviour of Mg isotopes in weathering processes.

Weathering sequences of soils developed on volcanic ash in humid tropical regions are well suited for studying how the degree of weathering affects soil constituents and nutrient distribution (Chadwick et al. 2003, 1999). Volcanic arc settings such as Guadeloupe, characterized by high precipitation rates, are amongst the fastest eroding regions on Earth (Gaillardet et al., 2011). Better understanding silicate weathering processes in these regions is a mandatory step to refine estimates of their CO₂ consumption rates and the impact of weathering on the global C budget. In soils from Guadeloupe, secondary Mg-bearing phases such as smectite and hydroxy-Al interlayered smectite/hydroxy-Al interlayered vermiculite (HIS/HIV) have been identified (Colmet-Daage and Lagache, 1965) and were formed from a typical local andesite (Samper et al., 2007; Sak et al., 2010). Here we investigate Mg isotope variations in bulk soils and clay fractions relative to the parent andesite in three soil weathering sequences from Guadeloupe in order to provide a direct *in situ* measurement of the fractionation as a function of the degree and detailed processes of weathering.

2. Environmental setting

The Guadeloupe archipelago is located in the French Indies of the Caribbean (16°N, 61°W). The two main islands of Guadeloupe are Basse-Terre and Grande-Terre. Basse-Terre is a volcanic island dominated by the volcano La Soufrière (1467 m above sea level (asl)). Volcanic ash soils cover extensive areas on the slopes surrounding the volcano, developed from andesitic ash deposits of Pliocene to Holocene age, dominated by plagioclase, pyroxene and ferromagnesian volcanic glasses (Dagain et al., 1981; Ndayiragije, 1996; Ndayiragije and Delvaux, 2003). Grande Terre comprises a coral basement covered by volcanic pyroclasts contemporary to the volcanic deposits from the north of Basse-Terre.

Contrasting climatic conditions characterise the western (Ws), eastern (Es) and northern (Ns) slopes of la Soufrière volcano (Fig. 1). The eastern slopes are exposed to rain-bearing Northeast trades from the Atlantic Ocean, with a mean annual rainfall (MAR) exceeding 5000 mm at 500 m asl, and decreasing to 2500 mm downslope at sea level (Chaperon et al., 1985). The western slopes are drier with MAR decreasing from 3200 mm at 450 m asl to 850 mm at sea level. On the northeastern slopes, annual rainfall decreases from 4000 mm on the northeastern slope tops of Basse-Terre to 1200 mm on the eastern coast of Grande-Terre.

The age of the soils increases from Ws, which has young soils containing a reserve of weatherable primary minerals, to older soils in Es (10³–10⁴ yr), to the oldest soils from Ns where weathering has exhausted the pool of primary minerals (10⁵ yr) (Cabidoche et al., 2009).

The distribution of soils in Guadeloupe with contrasting degree of weathering and clay mineralogy is related to climate and to the

age of the soils as chrono-climo-topo-sequences (Colmet-Daage and Lagache, 1965; Ndayiragije, 1996; Ndayiragije and Delvaux, 2003, 2004). This is typical in the South of Basse-Terre for soils from Ws and Es (Fig. 2). The Ws soil pattern developed in dry conditions involves a weathering sequence Andosol–Cambisol corresponding to the mineralogical sequence ash→allophane→halloysite/smectite (Colmet-Daage and Lagache, 1965; Pineros-Garcet, 1994). The Es soil pattern developed in wetter conditions at the same elevation and produced a weathering sequence Perhydrated Andosol–Andosol–Nitisol, corresponding to the mineralogical sequence ash→gibbsite, allophane→halloysite, Fe-oxide (Colmet-Daage and Lagache, 1965; Ndayiragije, 1996; Ndayiragije and Delvaux, 2003, 2004). In the North of Basse-Terre and Grande Terre, the Ns soil pattern produced a weathering climo-sequence Ferralsol–Vertisol from wet to dry conditions yielding the mineralogical sequence kaolinite, Fe-oxide→smectite.

3. Methods

3.1. Sampling

Soil samples representative of the contrasting chrono-climo-topo-sequences in Guadeloupe were collected during fieldwork in January 2006 (Henriet et al., 2008a, 2008b). A total of seven sites from three soil weathering sequences were selected, including Andosol–Cambisol (Ws), Perhydrated Andosol–Andosol–Nitisol (Es), Ferralsol–Vertisol (Ns) (Figs. 1 and 2). The topsoil (0–20 cm) was collected in each site. A fresh rock sample representative of the protolith andesite was collected in November 2009 from the Piton Tarade, the local basement of the La Soufrière volcano (altitude 1090 m asl) (Fig. 1).

The soils were sampled from sites that have been used for intensive banana cropping for a long time (Colmet-Daage and Lagache, 1965; Ndayiragije, 1996; Dorel et al., 2000). The agricultural system is based on monoculture with successive cropping cycles of 7–9 months. At harvesting time, leaves and pseudostems are cut and left on the soil surface, which significantly limits the mineral export by the vegetation. Before plantation and in a non-recurring process, lime (coral limestone or dolomite stone, 1T/ha; Sansoulet, 2007) is applied to the banana fields for fertilisation. To avoid a direct bias from this initial lime treatment we have chosen sampling sites that are located on ~10 yr old plantations, where lime vanished over the years, as supported by similar pH values in the sampled soils (soil pH_{H2O}=6.2±0.5; Henriet et al., 2008b) relative to pH values measured in uncultivated soils from the same areas in Guadeloupe (soil pH_{H2O}=5.5±0.6; Pineros-Garcet, 1994; Ndayiragije, 1996).

3.2. Soil characterisation

The soils samples were characterised in a previous study (Table 1; Henriet et al., 2008b). Briefly, soil samples were air-dried and sieved at 2 mm to recover the fraction representative for the bulk soil (<2 mm). The recovery of clay fractions (<2 μm) was achieved after sonication and sieving at 50 μm to remove sand fractions, and prolonged dispersion with Na⁺-resin (Rouiller et al., 1972). Clay (<2 μm) was separated from silt (2–50 μm) by successive 24 h cycles of decanting and pipetting in deionised water. Clay fraction mineralogy was assessed by XRD after K⁺ and Mg²⁺ saturation, ethylene glycol solvation and thermal treatments at 300 and 550 °C (Robert and Tessier, 1974).

Bulk soils, clay fractions and the andesite sample were analysed for major elements by ICP-AES (Université catholique de Louvain, Belgium) after Li borate fusion at 1000 °C (Chao and

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