

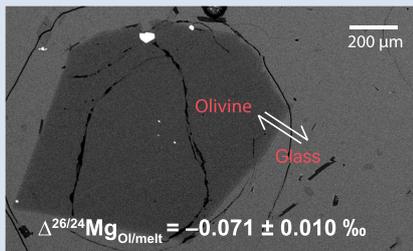
# Equilibrium olivine-melt Mg isotopic fractionation explains high $\delta^{26}\text{Mg}$ values in arc lavas

X.-N. Liu<sup>1\*</sup>, R.C. Hin<sup>1</sup>, C.D. Coath<sup>1</sup>, M. van Soest<sup>2</sup>, E. Melekhova<sup>3</sup>, T. Elliott<sup>1</sup>



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## Abstract



We determined equilibrium Mg isotope fractionation between olivine and melt ( $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ ) in five, naturally quenched, olivine-glass pairs that were selected to show clear textural and chemical evidence of equilibration. We employed a high-precision, critical mixture double-spiking approach to obtain a weighted mean of  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}} = -0.071 \pm 0.010$  ‰, for values corrected to a common olivine-glass temperature of 1438 K. As function of temperature, the fractionation can be expressed as  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}} = (-1.46 \pm 0.26) \times 10^5/T^2$ . The samples analysed have variable  $\text{H}_2\text{O}$  content from 0.1 to  $\sim 1.2$  wt. %, yet no discernible difference in  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  was evident. We have used this  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  to revisit the puzzling issue of elevated Mg isotope ratios in arc lavas. In new Mg isotope data on sample

suites from the Lesser Antilles and Mariana arcs, we show that primitive samples have MORB-like Mg isotope ratios while the evolved samples tend to have isotopically heavier compositions. The magnitude of this variability is well explained by olivine fractionation during magmatic differentiation as calculated with our new equilibrium  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ .

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## Introduction

A burgeoning literature in the mass-dependent variability of major rock forming elements in magmatic samples have the potential to provide novel constraints on source mineralogy and melting processes (e.g., Teng *et al.*, 2017; Soderman *et al.*, 2022), but in many cases the key parameter of isotopic fractionation, *i.e.* the fractionation factor between solid and melt, is insufficiently well constrained to make the most of the observations. In large part, mineral-melt fractionation factors have been determined by the magnitude (or absence) of isotopic variability in sample suites that show well behaved differentiation trends. Although valuable, this strategy convolves the natural complexity of magmatic fractionation with the determination of fractionation factors. A more direct method is to measure the isotope ratios of coexisting equilibrated mineral-melt pairs. This poses the difficulty of obtaining precise measurements on small samples that are demonstrably in equilibrium.

As the third most abundant element in the silicate Earth, there is much interest in the Mg isotopic systematics of magmatic rocks for improving our understanding of igneous processes and broader planetary evolution (e.g., Teng *et al.*, 2010; Hin *et al.*, 2017; Teng, 2017). In interpreting the relatively small isotopic variations in Mg, it is critical to determine a precise fractionation factor between olivine, the major mineral host of Mg, and melt. This parameter is expressed as  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ , defined as  $\delta^{26}\text{Mg}_{\text{Ol}} - \delta^{26}\text{Mg}_{\text{melt}}$ , where  $\delta^{26}\text{Mg}$  is the relative difference in  $^{26}\text{Mg}/^{24}\text{Mg}$  between sample and DSM-3 reference standard. Once determined,

$\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  can be used in conjunction with inter-mineral fractionation factors to model Mg isotopic variability in magmatic processes. While inter-mineral fractionations can be determined observationally or by *ab initio* methods, numerical modelling of isotopic exchange between mineral and melt structure is not straight-forward, which has motivated our empirical approach.

There have been two previous attempts to measure  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ . The absence of systematic Mg isotopic variation in a suite of whole rocks with variable MgO contents from Kilauea Iki (Teng *et al.*, 2007) is often cited as evidence for the absence of Mg isotope fractionation during crystallisation. Yet, strictly, this study placed a maximum bound on Mg isotope fractionation during differentiation, namely  $|\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}| \leq 0.07$  ‰. Schiller *et al.* (2017) analysed the Mg isotopic compositions of olivines and rapidly cooled groundmass (dominantly intergrown plagioclase and clinopyroxene) from an angrite meteorite (NWA1670). These authors also reprocessed Mg isotope measurements of olivine and groundmass from Teng *et al.* (2011) to calculate a fractionation factor. Yet, the latter had been originally used to illustrate the effects of diffusive fractionation of Mg isotopes in the chemical potential gradient of zoned minerals. Although Schiller *et al.* (2017) reported those samples closest to elemental Fe-Mg equilibrium, these samples evidently do not constitute an equilibrium assemblage necessary for reliable determination of a fractionation factor. Equally, the bulk olivine phenocrysts in NWA1670 are not in equilibrium with the groundmass, given their Mg/(Mg + Fe) decrease from  $\sim 0.9$  in their core to  $\sim 0.6$  in their rim (Jambon *et al.*, 2008).

1. School of Earth Science, University of Bristol, Wills Memorial Building, Bristol BS8 1RJ, UK  
2. School of Earth and Space Exploration, ISTB4, Arizona State University, Tempe 85287, AZ, USA  
3. Department of Earth Sciences, University of Oxford, South Parks Road, Oxford OX1 3AN, UK

\* Corresponding author (email: [x17349@bristol.ac.uk](mailto:x17349@bristol.ac.uk))

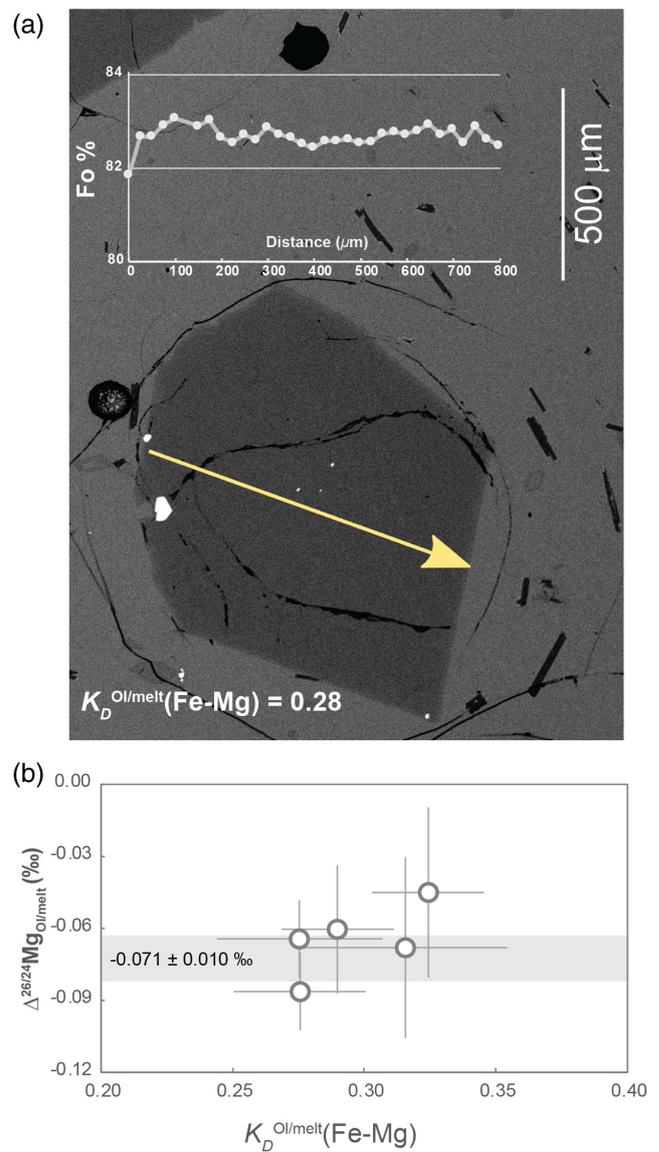


Here, we employ the high precision attainable using critical mixture double-spiking (Coath *et al.*, 2017; Hin *et al.*, 2017) to determine  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  for five carefully selected, equilibrated olivine-glass pairs from ocean island and mid-ocean ridge basalts. Using this value, we then explore the subtly elevated  $\delta^{26}\text{Mg}$  in arc lavas (Teng *et al.*, 2016) with new analyses of a suite of samples from the Lesser Antilles, as well as a set of archetypical mafic samples from the Mariana arc.

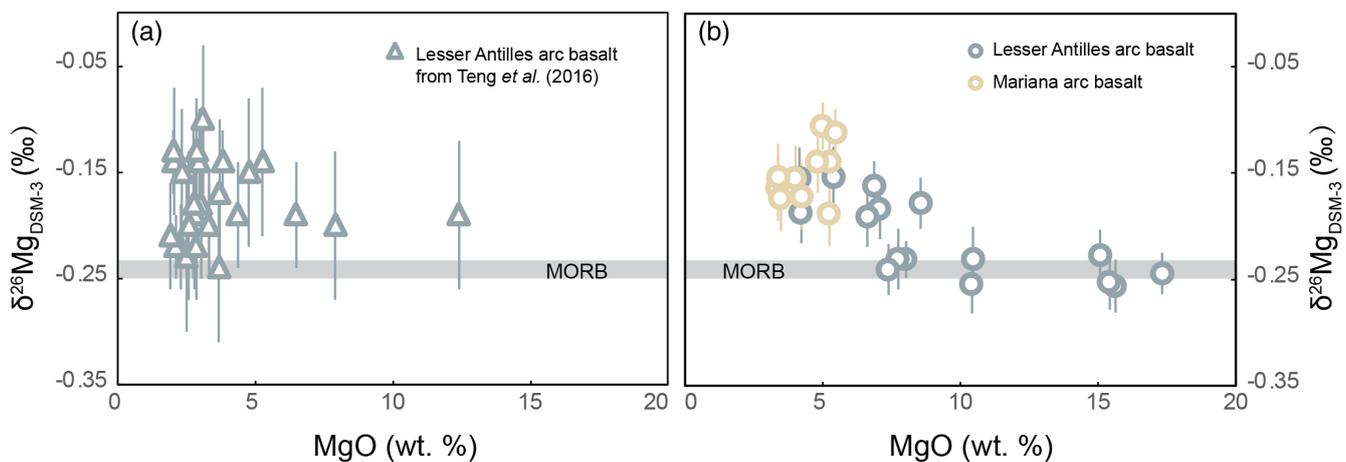
### Olivine-melt fractionation factor

We selected samples with petrographic, equilibrium olivine textures in naturally quenched glass from Kilauea (Hawaii), submarine eruptions from Pitcairn and mid-ocean ridge basalts from the Pacific and Indian oceans (for details, see section 1.1 of the Supplementary Information, SI). Prior work on the Hawaii and Pitcairn samples (Jeffcoate *et al.*, 2007) showed an absence of Li isotopic fractionation across the olivine phenocrysts, documenting an absence of late-stage diffusive fractionation. From these potentially suitable samples we then selected individual olivine crystals with a variability in Fo < 1 % across the full cross-sectional electron microprobe profile of the crystal (Fig. 1a). We rejected any olivine which had an olivine-glass Fe-Mg exchange coefficient outside the range of equilibrium values (Ulmer, 1989), namely  $K_D^{\text{Ol/melt}}(\text{Fe-Mg})$  from 0.28 to 0.32 (Fig. 1b).

We analysed micro-drilled spots (cones of 100  $\mu\text{m}$  depth and largest diameter) in these olivines and hand-picked coexisting glass and processed the samples for Mg isotope analysis by critical mixture double-spiking as reported in Hin *et al.* (2017). This method yields a long-term reproducibility of 0.027 ‰ ( $\delta^{26}\text{Mg}$ , 2 s.d.) based on repeated BHVO-2 analyses (see SI section 2.3). We used the pooled  $\delta^{26}\text{Mg}$  of each phase to yield Mg isotope differences between olivine and melt (*i.e.* glass) for each of the five basalt samples (Table S-1). Olivine thermometry (Putirka, 2005) indicates that equilibration temperatures of the five samples varied between 1379 and 1481 K (see SI section 1.2). Using a  $1/T^2$  scaling, we corrected the isotope differences to a single, average temperature of 1438 K, yielding  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  between  $-0.045 \pm 0.036$  ‰ and  $-0.086 \pm 0.016$  ‰ (Fig. 1b, Table S-2). These values are consistent with each other and yield a weighted mean of  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}} = -0.071 \pm 0.010$  ‰ or  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}} = (-1.46 \pm 0.26) \times 10^3/T^2$  ( $T$  in Kelvin). For the first time, we thus show that olivine in equilibrium with melt is significantly enriched in light Mg isotopes and we recommend that our olivine-melt fractionation factor should



**Figure 1** (a) Backscattered electron microscope image of an olivine-glass pair (PN3-10), together with electron microprobe traverse of olivine Fo content. (b) Measured  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ , corrected to a common temperature (1438 K), plotted against Fe-Mg distribution coefficients.



**Figure 2**  $\delta^{26}\text{Mg}$  against MgO for (a) Lesser Antilles lavas from Teng *et al.* (2016) and (b) Lesser Antilles and Mariana lavas from this study. MORB reference value is  $-0.24 \pm 0.01$  ‰ (2 s.e.); see Supplementary Information section 3.4.

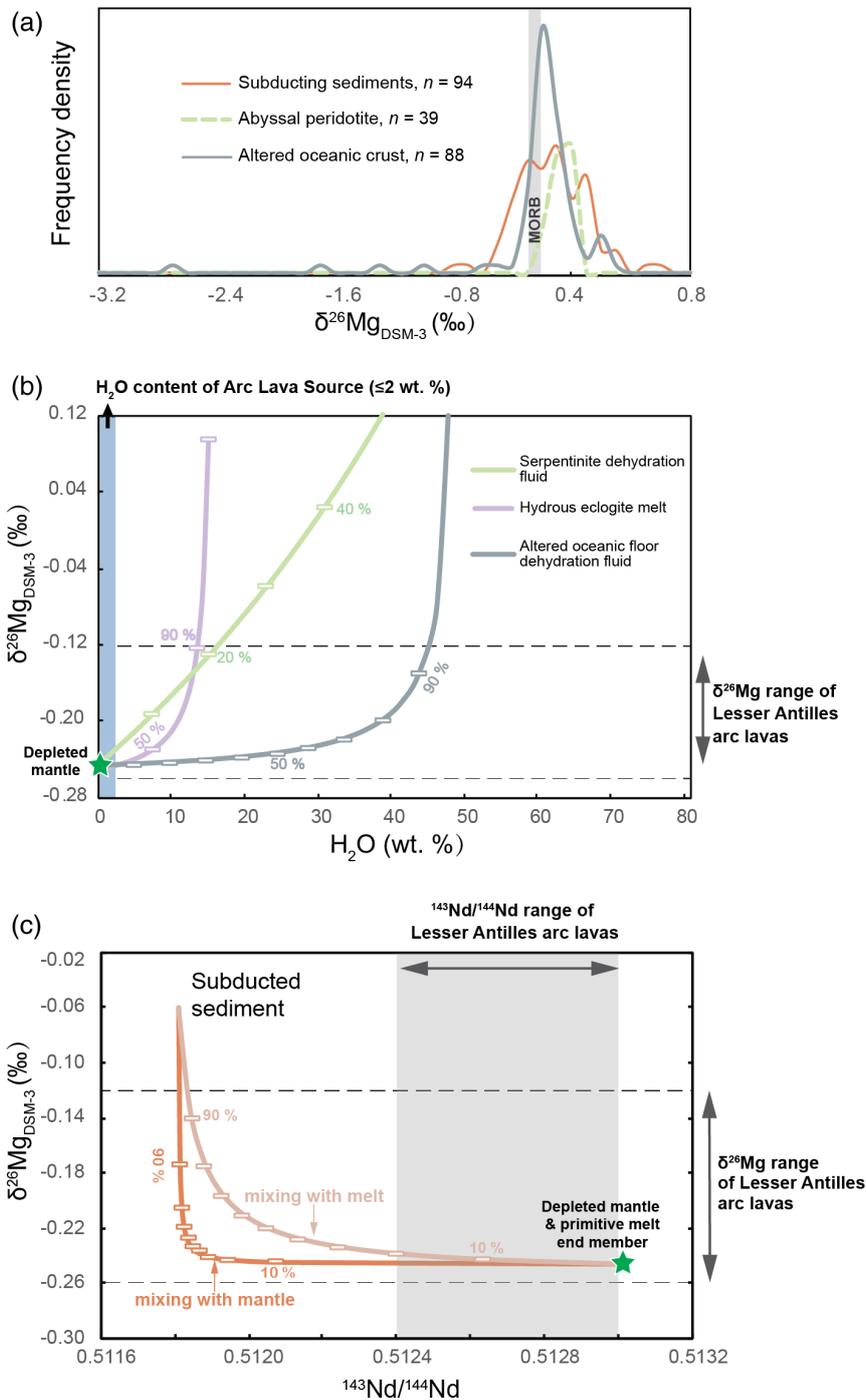
be considered when modelling Mg isotope fractionation in magmatic process. We also tried to experimentally determine  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  but this attempt unfortunately failed, most likely due to thermal diffusion (see SI section 1.3).

The Hawaiian and Pitcairn samples have water contents of ~0.1–0.4 wt. % (e.g., Hauri, 2002) and ~1.2 wt. % (Aubaud et al., 2006), respectively. MORB samples generally have low water contents around 0.1 to 0.2 wt. % (e.g., Sobolev et al., 1996). Water decreases the Mg–O coordination number of silicate melt (Mookherjee et al., 2008), which may lead to a preference for heavier isotopes. Nonetheless, the  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  determined

from Hawaiian, Pitcairn and the three MORB olvine-glass pairs are all within the analytical error (Fig. 1b). The absence of a discernible effect on the fractionation factor for water contents up to 1.2 wt. % leads us to assume that water has a limited effect on  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ .

### Elevated $\delta^{26}\text{Mg}$ in arc lavas

Teng et al. (2016) analysed arc lavas from Martinique, Lesser Antilles, and reported  $\delta^{26}\text{Mg}$  slightly higher than MORB



**Figure 3** (a) Frequency density plot of  $\delta^{26}\text{Mg}$  of subducted materials. (b) Modelled  $\delta^{26}\text{Mg}$  against  $\text{H}_2\text{O}$  from mixing depleted mantle with dehydration fluids and hydrous melt. (c) Modelled  $\delta^{26}\text{Mg}$  against  $^{143}\text{Nd}/^{144}\text{Nd}$  from mixing depleted mantle with subducted sediments (See SI section 3.5 for more details).

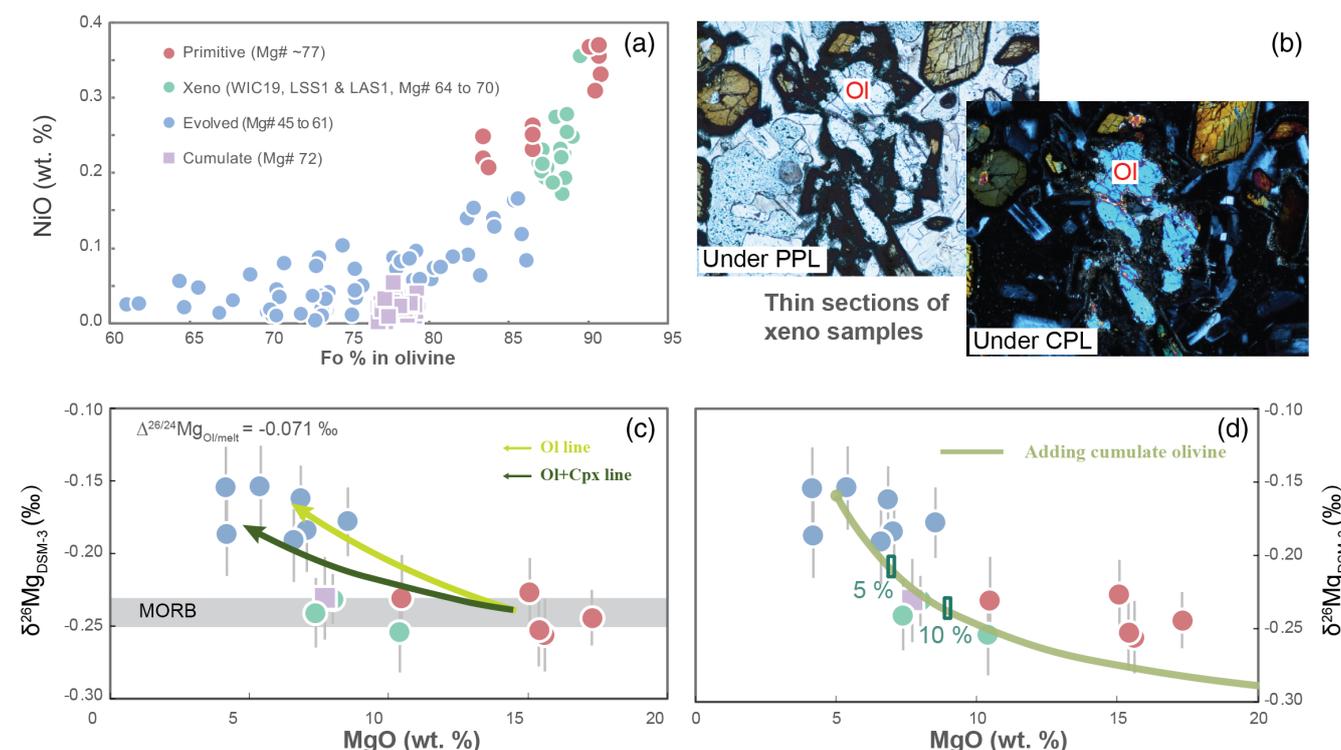
(Fig. 2a), which they attributed to subduction zone processes. As discussed by Teng *et al.* (2016), however, it is not clear that subduction zone components have the leverage to sufficiently perturb the  $\delta^{26}\text{Mg}$  of the mantle wedge, given mixing constraints from other elemental and isotopic tracers. To further explore this intriguing phenomenon, we have made high precision, critical mixture double-spiked analyses of Lesser Antilles samples from a wider range of islands, including rare primitive lavas from the southern arc. We also analysed a well characterised set of samples from the Mariana arc (see SI sections 3.2, 3.3). Our new measurements show the Lesser Antilles lavas have MORB-like  $\delta^{26}\text{Mg}$  in the most MgO-rich samples, while more evolved lavas have  $\delta^{26}\text{Mg}$  up to 0.12 ‰ higher (Figs. 2b, S-5). All the Mariana arc lavas plot together with the less mafic Lesser Antilles samples (Fig. 2b, Table S-5). To better understand the causes of the higher  $\delta^{26}\text{Mg}$  in some arc lavas, we initially concentrate on the Lesser Antilles example, for which we have samples with a wider compositional range.

First, we reconsider the possible role of subduction components (Fig. 3a). The results of binary mixing calculations (see SI section 3.5 for details) between the sub-arc mantle and potential subduction inputs are illustrated in Figure 3b, c. Fluids released from subducted oceanic crust and serpentinite are naturally water-rich, over 50 wt. % and 80 wt. %  $\text{H}_2\text{O}$  respectively, while the MgO concentrations of these fluids are generally low, less than 1 wt. % in oceanic crust dehydration fluid and about 6 wt. % in serpentinite dehydration fluid (Manning, 2004; Kessel *et al.*, 2005; Scambelluri *et al.*, 2015). Hydrous melts generated from eclogite are again enriched in water (over 15 wt. %) but also low in Mg (less than 2.5 wt. %) (Gervasoni *et al.*, 2017). The amount of these slab-derived fluid phases added to the mantle wedge is constrained by a maximum 2 wt. %  $\text{H}_2\text{O}$  in the mantle source (assuming at most 10 wt. %  $\text{H}_2\text{O}$  in primitive arc magmas and 20 % partial melting degree). As a result, the Mg

isotopic perturbation of the arc lava source is inappreciably influenced by plausible contributions of slab-derived fluids (Fig. 3b). The subducting sediments are isotopically heavier than MORB (Fig. 3a), but a contribution over 90 wt. % sediment would be required in the source of the Lesser Antilles lavas to reproduce the highest  $\delta^{26}\text{Mg}$ . Such an amount of sediment is equally inconsistent with the  $^{143}\text{Nd}/^{144}\text{Nd}$  of the arc lavas (Fig. 3c).

Our high precision analyses allow some structure to be discerned in the variability of  $\delta^{26}\text{Mg}$  in the Lesser Antilles samples, which can have been caused by neither weathering nor variable melting depths (see SI section 3.6). The most striking feature is a systematic relationship between  $\delta^{26}\text{Mg}$  and MgO content (Fig. 2b), which implies a role for magmatic differentiation in fractionating Mg isotopes. When dealing with the effects of differentiation in bulk samples, however, it is important to evaluate crystal accumulation. This is especially marked for Mg given the high MgO contents of olivine. Therefore, we analysed the compositions of olivine crystals in our Lesser Antilles samples. Three samples (WIC19, LSS1 & LAS1) have unexpectedly Fo-rich, xenocrystic olivines compared with their bulk Mg# (Fig. 4a, SI section 3.7). Moreover, disequilibrium textures between olivine crystals and groundmass are observed in the thin sections of these samples (Fig. 4b).

We have divided Lesser Antilles arc samples into four groups (Fig. 4a, c, d). Five samples with high MgO are dubbed “primitive”, as detailed petrological experiments have identified such compositions are plausibly in equilibrium with the mantle wedge (see SI section 3.2). They have  $\delta^{26}\text{Mg}$  values within error of MORB (Fig. 4c). Seven samples are grouped as “evolved” and display elevated  $\delta^{26}\text{Mg}$  values. The three samples that contain Fo-rich olivine xenocrysts are named “xeno” and these also have MORB-like  $\delta^{26}\text{Mg}$  values. A single troctolite sample, LAE3, is labelled “cumulate”. The Mariana samples are all geochemically



**Figure 4** (a) Electron microprobe analyses of Lesser Antilles olivines plotted in groups according to bulk Mg# of host magmas. (b) Thin-section images of disequilibrium olivines in “xeno” group samples. (c) Modelled  $\delta^{26}\text{Mg}$  evolution during two main differentiation paths. (d) Modelled effect on  $\delta^{26}\text{Mg}$  of cumulate olivine addition to an “evolved” sample composition.

similar to the “evolved” group and also have elevated  $\delta^{26}\text{Mg}$  values.

Unlike in the Marianas, the magma differentiation paths of Lesser Antilles arc lavas are well constrained (see [SI section 3.8](#)). Two liquid lines of descent have been identified: one involves co-crystallisation of olivine and clinopyroxene at high pressure, while at low pressures, olivine is the only liquidus phase (e.g., [Stamper et al., 2014](#)). We model the variations in  $\delta^{26}\text{Mg}$  that result from these two differentiation trends using our new  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  and  $\Delta^{26/24}\text{Mg}_{\text{Cpx/melt}}$  (derived by combining  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  with literature  $\Delta^{26/24}\text{Mg}_{\text{Cpx/Ol}}$  [SI section 3.9](#)). Both 20 % olivine fractionation (Ol line) and the co-crystallisation of 20 % olivine and 20 % clinopyroxene (Ol + Cpx line) reproduce the elevated  $\delta^{26}\text{Mg}$  of many evolved lavas ([Fig. 4c](#)), as a result of olivine crystallisation. The lower  $\delta^{26}\text{Mg}$  data of the “xeno” samples are well reproduced by olivine accumulation ([Fig. 4d](#)) in more evolved samples. The composition of the troctolite cumulate (LAE3) is consistent with its crystallisation from a melt with elevated  $\delta^{26}\text{Mg} \approx -0.16\text{‰}$  given our  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$ . We infer that the relatively low MgO and high  $\delta^{26}\text{Mg}$  of the Mariana samples ([Fig. 2b](#)) reflect a differentiation process similar to that experienced by the Lesser Antilles “evolved” samples.

In conclusion, we have replicated elevated  $\delta^{26}\text{Mg}$  in Lesser Antilles arc lavas and further shown this to be a common characteristic in Mariana arc lavas. However, our more precise analyses reveal that the elevated  $\delta^{26}\text{Mg}$  is only evident in more evolved, basaltic andesite compositions. Using the equilibrium  $\Delta^{26/24}\text{Mg}_{\text{Ol/melt}}$  we determined, we can model the increase in  $\delta^{26}\text{Mg}$  from MORB-like values in primitive arc lavas as a natural consequence of magmatic differentiation. This illustrates the importance of a well determined solid-melt fractionation factor in interpreting subtle differences in stable isotope ratios.

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## Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2226>.



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