Magnesium isotope geochemistry in arc volcanism

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Incorporation of subducted slab in arc volcanism plays an important role in producing the geochemical and isotopic variations in arc lavas. The mechanism and process by which the slab materials are incorporated, however, are still uncertain. Here, we report, to our knowledge, the first set of Mg isotopic data for a suite of arc lava samples from Martinique Island in the Lesser Antilles arc, which displays one of the most extreme geochemical and isotopic ranges, although the origin of this variability is still highly debated. We find the δ^{26}Mg of the Martinique Island lavas varies from -0.25 to -0.10, in contrast to the narrow range that characterizes the mantle (-0.25 ± 0.04, 2 SD). These high δ^{26}Mg values suggest the incorporation of isotopically heavy Mg from the subducted slab. The large contrast in MgO content between peridotite, basalt, and sediment makes direct mixing between sediment and peridotite, or assimilation by arc crust sediment, unlikely to be the main mechanism to modify Mg isotopes. Instead, the heavy Mg isotopic signature of the Martinique arc lavas requires that the overall composition of the mantle wedge is buffered and modified by the preferential addition of heavy Mg isotopes from fluids released from the altered subducted slab during fluid–mantle interaction. This, in turn, suggests transfer of a large amount of fluid-mobile elements from the subducting slab to the mantle wedge and makes Mg isotopes an excellent tracer of deep fluid migration.

magnesium isotopes | arc magmatism | mantle wedge | Lesser Antilles arc | Martinique Island

Arc volcanism records the elemental cycling between the subducting slab and subarc mantle. Of particular interest is the mechanism by which the subducted material is incorporated into the arc lava. Except for the rare case where arc lava is the direct melting product of a subducted slab (1), most scenarios suggest that mantle wedge is the major magma source that melts after being modified by fluids or melts derived from the subducted basalt and sediment (2, 3). In addition, processes such as polybaric crystallization and crustal assimilation can also modify the composition of arc magmas on their way to the surface. These different processes have different implications on subduction dynamics and elemental cycling, but, in many cases, they are difficult to distinguish. One of the best examples comes from studies of island arc lavas from the Lesser Antilles arc (Fig. 1). Geochemical and Sr, Nd, Pb, Hf, and Li isotopic studies suggest that the Lesser Antilles arc lavas incorporated a variable but to some extent significant amount of subducted sediments (4–8). However, the exact mechanism by which the sediment was incorporated into the lavas is still highly debated and involves various processes such as crustal contamination, subarc mantle metasomatism by fluids released from the slab, or melts derived by partial melting of the subducted sediments (4–17).

Magnesium isotopes have the potential to provide new and independent constraints on both source composition and processes operating during the formation of arc magmas, not only because Mg is a major element in all magmas but also because surficial and low-temperature processes fractionate Mg isotopes whereas high-temperature magmatic processes do not (18, 19) (Fig. 2). Subducted marine sediments and altered basalts have isotopic compositions different from those of the normal mantle as sampled by global peridotite xenoliths (Fig. 2); however, they

generally have low Mg concentrations (18–25, *). In comparison, altered abyssal peridotites have Mg concentrations similar to the normal mantle whereas their Mg isotopic compositions are heavier because of the impact of hydrothermal circulation during accretion and residence in the deep ocean (Fig. 2).[†,‡] Finally, although the mechanism is still not well understood, studies of a few arc peridotites show that they also have slightly heavier Mg isotopic composition than the normal mantle (Fig. 2). Given these observed ranges, Mg isotopes may help in understanding the relative contributions of crustal and mantle components to arc magmatism, but no systematic study of either continental or island arc lavas has been carried out yet.

Here, we report Mg isotopic data for 27 arc lavas and 17 subducting forearc sediment samples. The lava samples are from the Martinique Island and cover most of the chemical and isotopic variations in the Lesser Antilles arc (4, 5) (Fig. 1). The sediment samples are from Deep Sea Drilling Project (DSDP) sites 543 and 144 (NE and SE of Martinique Island, respectively); they cover the whole compositional spectrum of subducting sediments and range in lithology from chalky ooze to terrigenous and pelagic deposits (6, 7).

The sediments display a large range of δ^{26}Mg (-0.76 to $+0.52$) with an average of -0.10 ± 0.61 (2 SD) (Table S1). This large variation is mainly controlled by sediment mineralogy, with carbonate-rich samples at site 144 generally having light Mg isotopic compositions, whereas clays, the dominant type of sediments at site 543, have heavy isotopic compositions (Fig. 2). This mineralogical control is also evident in studies of loess, shale, mudrock, and carbonates as well as leaching experiments that show preferential enrichment of light Mg isotopes in carbonates over silicates (26, 27).

Significance

Arc lavas from Martinique have nonmidocean ridge basalt Mg isotopic composition, which is consistent with the incorporation of subducted Mg. This is, to our knowledge, the first report of mantle-derived lavas with Mg isotopic composition heavier than oceanic basalts, heretofore shown to be isotopically homogenous. More importantly, our results provide insight into the strongly debated origins of Martinique arc lavas and suggest that contributions of Mg from fluids supplied by the subducted slab may play a significant control in the Mg isotopic systematics of arc lavas.
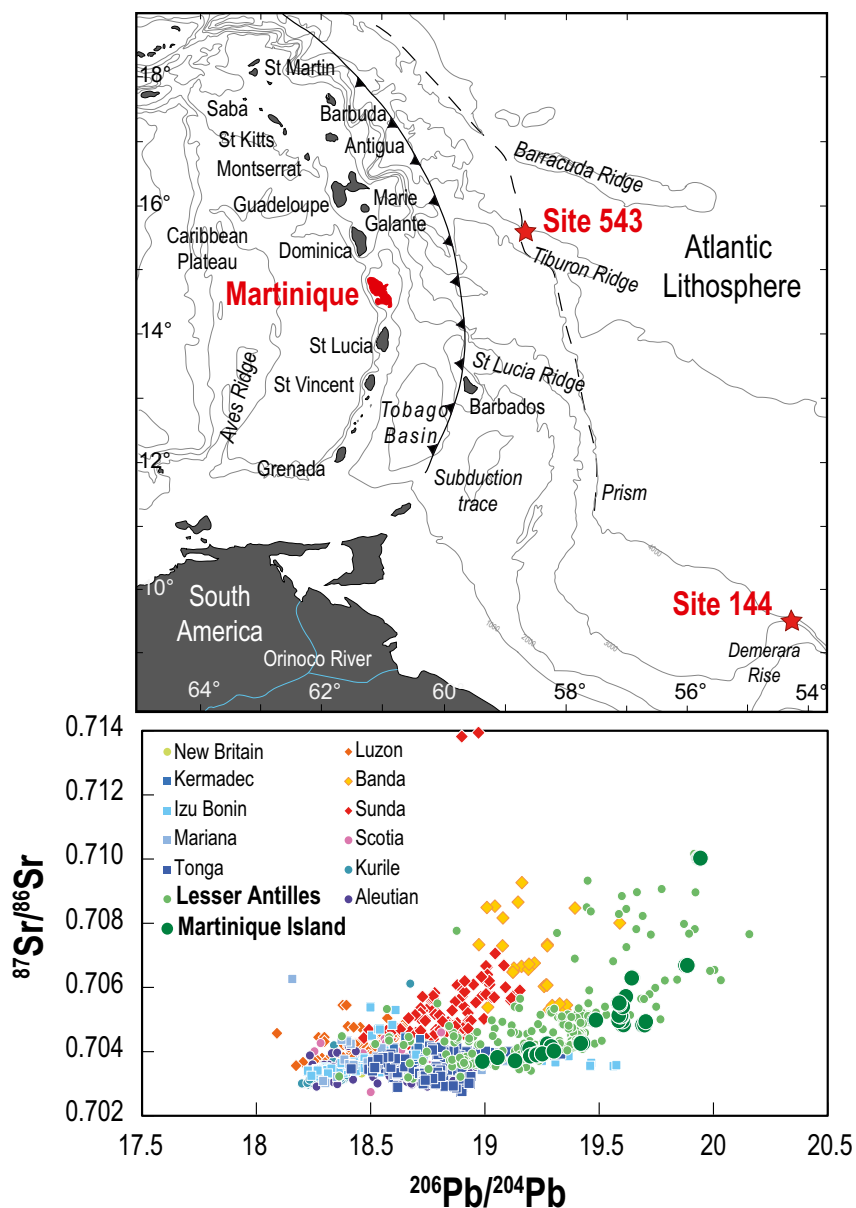
Fig. 1. (*Upper*) Geological map of the Lesser Antilles island arc and the two DSDP sites (sites 144 and 543). (*Lower*) Comparison of Martinique Island basalts with other Lesser Antilles and worldwide island arcs in $^{87}Sr/^{86}Sr$ versus $^{206}Pb/^{204}Pb$ isotopic space (data compiled by Geochemistry of Rocks of the Oceans and Continents database). Modified from ref. 4 with permission from Elsevier; www.sciencedirect.com/science/journal/0012821X.

The $\delta^{26}Mg$ values of Martinique lavas define a smaller range from −0.25 to −0.10, and are, on average (−0.18 ± 0.07, 2 SD) (Fig. 2), higher than midocean ridge basalt (MORB) ($\delta^{26}Mg$ = −0.25 ± 0.06, 2 SD) and mantle peridotite ($\delta^{26}Mg$ = −0.25 ± 0.04 2 SD) (18, 23, 28, 29). This difference indicates that the source of Martinique lavas is different from that of MORB, which could be related to a diversity of processes that include seawater alteration for submarine lavas, melting of a mantle source with different $\delta^{26}Mg$, or crustal contamination during magma ascent.

Chemical weathering and seawater alteration can potentially modify the Mg isotopic composition of arc basalts, and can shift their $\delta^{26}Mg$ to higher values if clays are the dominant alteration products (24). However, the analyzed lava samples are all fresh [loss on ignition (LOI) < 2% with one exception; Table S2], and most erupted above sea level (4, 5). A previous Li isotopic study on the same suite of samples has shown that only the three samples that erupted as submarine lava have high δ^7Li due to

interaction with isotopically heavy seawater (8). These three samples, however, have Mg isotopic compositions similar to the other samples. In addition, $\delta^{26}Mg$ of Martinique arc lavas does not correlate with their LOI. Therefore, different from Li isotopes, interaction with seawater has little effect on the $\delta^{26}Mg$. The different behavior between Li and Mg isotopes likely reflects the higher concentration of Mg over Li in basalts, which results in an easier isotopic fractionation of Li than Mg during weathering and alteration.

Partial melting of a peridotite source and fractional crystallization of olivine, pyroxene, and plagioclase can be ruled out too, as these processes do not fractionate Mg isotopes (18, 28–30). Nonetheless, arc lavas could potentially be isotopically heavier than MORB if they were produced by partial melting of a subducted oceanic crust, with garnet as a residual phase, e.g., adakite (1). This is because garnet has much lower $\delta^{26}Mg$ relative to coexisting pyroxene, as was observed in cratonic and
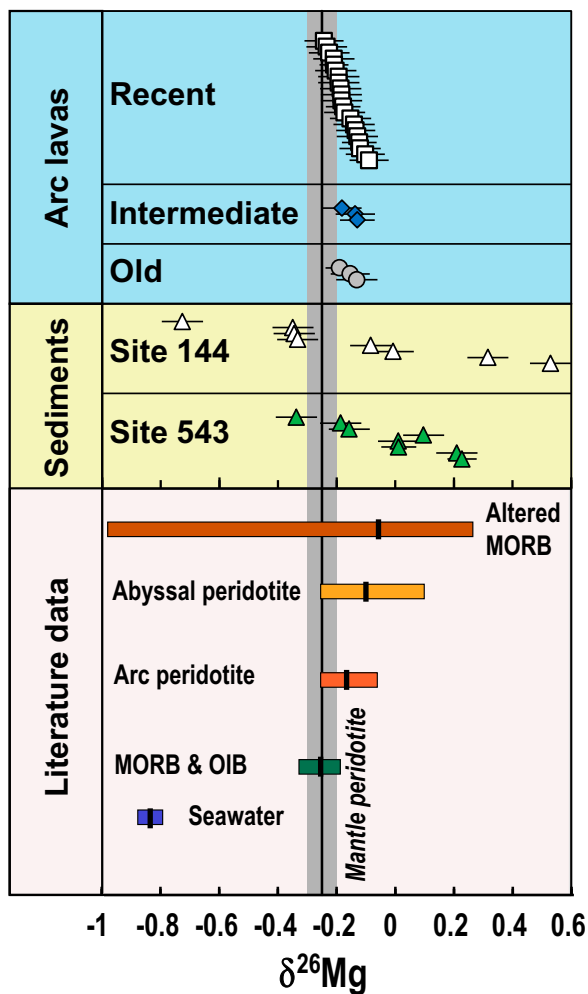
Fig. 2. Magnesium isotopic composition of Martinique arc lavas and subducting forearc sediments (sites 144 and 543). (Data are reported in Tables S1 and S2.) Data sources for the other reservoirs are from several references (18, 22–24, 36, 42, 43,*,†,‡). The vertical solid line and gray bar represent the average $\delta^{26}Mg$ and 2 SD of normal mantle, as sampled by global peridotite xenoliths (−0.25 ± 0.04) (18). The short bold black vertical lines represent the mean $\delta^{26}Mg$ value of each individual reservoir.

orogenic eclogites (31–33). However, this cannot be the cause of the high $\delta^{26}Mg$ values of Martinique lavas, because their chemical compositions are inconsistent with derivation from slab melting, i.e., adakite (4–7).

The forearc sediments that enter the Lesser Antilles Trench have, on average, a heavy Mg isotopic composition (−0.10 ± 0.61, 2 SD) (Fig. 2); they could thus be a potential source for the heavy Mg isotopic compositions of the Martinique lavas. Equivalent sediments in arc crust through which the Martinique lavas erupted could provide such a source, as well, if they were assimilated into the lavas. Furthermore, due to the lack of Mg isotope fractionation during prograde metamorphism (31, 33, 34), the metamorphic counterparts of the subducting sediments should preserve their original Mg isotopic signature. Previous isotopic studies of Martinique lavas show that the sedimentary input increases with age from old to intermediate lavas whereas it is much more variable in the recent lavas (4). However, Mg isotopic compositions of the Martinique lavas do not correlate with either age or any radiogenic isotopic system (Fig. 3), suggesting that the presence of heavy Mg is not caused by sediment addition to the subarc mantle source or directly to the lavas

themselves. Furthermore, neither binary mixing between subarc mantle peridotite and sediments nor assimilation and fractional crystallization of arc magma can explain the data (Fig. 3). In all modeled mixing arrays, the amount of sediments required to account for the $\delta^{26}Mg$ measured in the lavas is unrealistically high (>50%) due to the generally much lower Mg concentration in sediment (2–3%) compared with basalt (8%) and peridotite (48%) (25). Presence of such a large amount of sediment in a source producing basalts and andesites is impossible from a major element point of view. The opposite is true for elements such as Nd, Sr, Pb, or Li, which are drastically more enriched in sediment than in peridotite. In other words, a small addition of sedimentary materials into a peridotite or basalt can change their Nd, Sr, Pb, or Li isotopic compositions significantly, whereas a very large amount of sediment is required to change their Mg isotopic composition. The fact that $\delta^{26}Mg$ varies little in Martinique arc lavas, whereas their Nd, Sr, and Pb isotopes change significantly, implies that (*i*) the peridotite in the mantle wedge has an unusual Mg isotopic composition and (*ii*) the impact of sedimentary material, if any, is invisible from the Mg isotope perspective because of the large concentration contrast.

Our conclusions above are consistent with the few available Mg isotopic data for arc peridotites. Thus far, the only arc peridotites analyzed for Mg isotopes come from Avacha Volcano in Kamchatka, and they represent fragments of the subarc mantle that has been metasomatized by fluids released from the subducting Pacific plate (35). Their $\delta^{26}Mg$ values vary from −0.25 to −0.06 with an average of −0.18 ± 0.10 (2 SD) (36), overlapping the Martinique arc lava range but significantly different from values reported for normal mantle peridotites (18) (Fig. 2). Although the mechanism responsible for the heavy Mg isotopic composition of these arc peridotites is uncertain, it is possible that fluids coming from the subducted slab could modify the peridotite present in the mantle wedge.

The few available $\delta^{26}Mg$ data on altered MORB and abyssal peridotite are shown in Fig. 2. Abyssal peridotites are of particular interest because they have high Mg concentrations and their Mg isotopic compositions are on average heavier than fresh MORB (−0.25 to 0.10, with an average of −0.12 ± 0.14, 2 SD).[†,‡] The most likely explanation for their high $\delta^{26}Mg$ is that they were altered to various degrees by hydrothermal circulation during and after emplacement onto the seafloor.[†,‡] As subducted altered basalts and abyssal peridotites contain large amounts of fluids (their LOI is in the order of 10 wt.%), they can be the source of vast volumes of Mg-rich fluids (37, 38) released to the mantle wedge during dehydration of the subducted slab. These fluids infiltrate the mantle wedge through fluid−peridotite interactions and modify its Mg isotopic composition toward a heavy value, which comprises the source of the arc lavas.

Our interpretation that fluid−peridotite interactions in the subarc mantle have shifted mantle wedge and Martinique lavas to heavy Mg isotopic composition is also consistent with other independent observations. Dehydration of the subducting slab has been called on to explain the high concentration of fluid-mobile elements in Martinique lava samples (5, 39) and, more generally, in a large number of island arcs (40). In addition, the Sr isotopic composition of Martinique lavas remains quite low, at about 0.7035, indicating that the leached material is basaltic or peridotitic rather than sedimentary (see ref. 5 for more details). It could, however, be argued that the large amount of fluids needed to modify the $\delta^{26}Mg$ of the mantle wedge should also impact other geochemical parameters. For example, ratios of mobile to immobile trace elements should differ from normal mantle melts. This is indeed the case for Martinique lavas that have generally high Ba/Th and Pb/Ce ratios (85.3 ± 48.2 and 0.23 ±
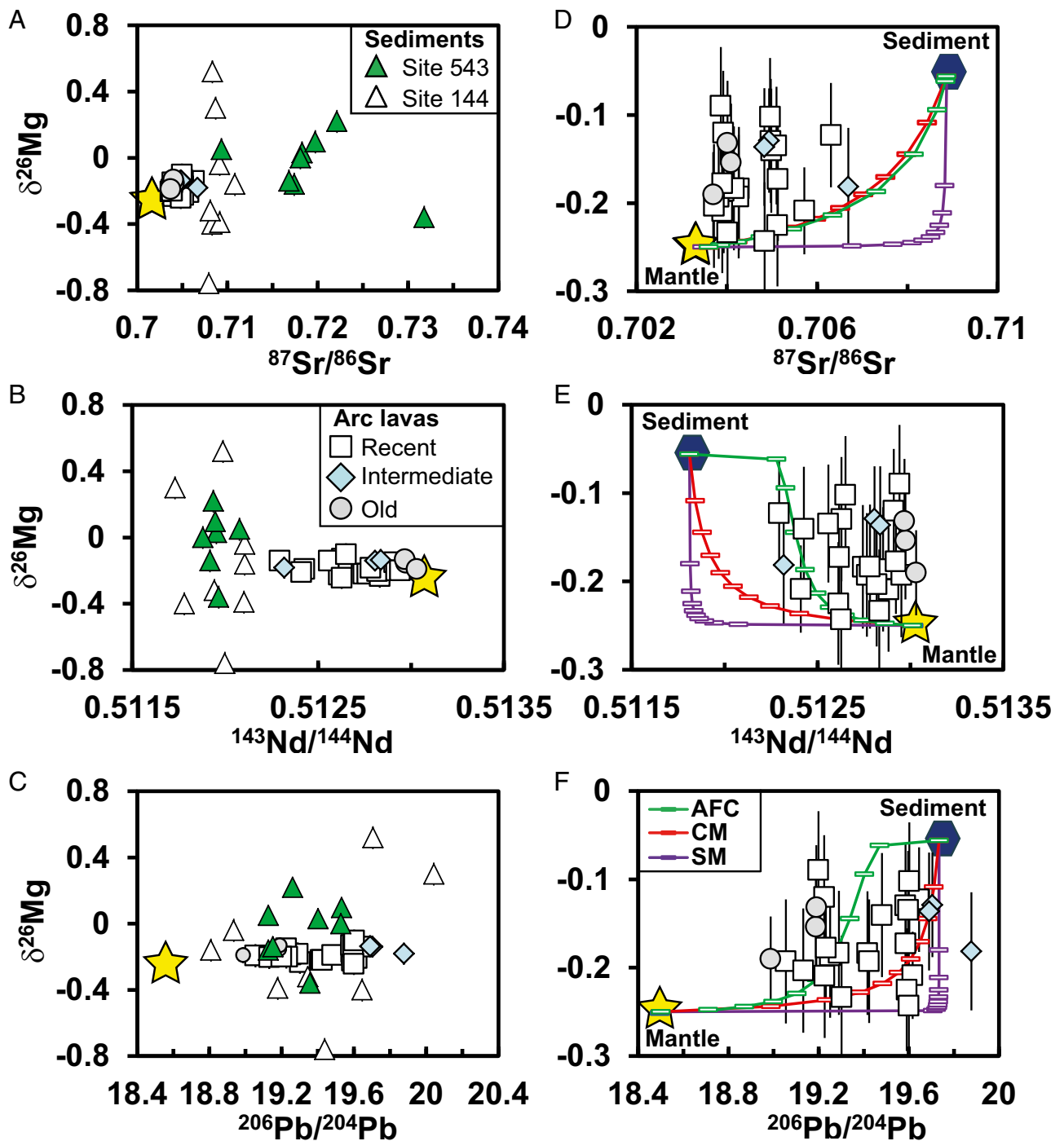
Fig. 3. Variations of Mg isotopic composition with Sr, Nd, and Pb isotopic compositions of Martinique arc lavas and subducting sediments from the Lesser Antilles arc (Tables S1 and S2). *A–C* include data of both Martinique arc lavas and subducting sediments, and *D–F* focus on the range observed in the lavas. The yellow star represents the hypothesized composition of the normal mantle. The hexagon represents an estimate of the average composition of subducting sediments (Tables S1–S3). *D–F* show mixing curves corresponding to three different types of mixing: Source mixing (SM) is depleted mantle–sediment mixing in the subarc mantle; crust mixing (CM) is primitive magma–sediment mixing; and AFC is assimilation and fractional crystallization. The ticks on modeling curves represent 10% increments during fractional crystallization or mixing. These mixing curves clearly do not fit the data measured on the Martinique arc lavas. *A* and *D* are $^{87}Sr/^{86}Sr$, *B* and *E* are $^{143}Nd/^{144}Nd$, and *C* and *F* are $^{206}Pb/^{204}Pb$. See Table S3 for modeling details.

0.06, 1 SD, respectively) relative to the values of MORB (71.93 ± 8.32 and 0.0402 ± 0.0016, 95% confidence, respectively) (41). However, no clear correlation exists between $\delta^{26}Mg$ and Ba/Th or Pb/Ce ratios in our dataset, mainly reflecting the combined effects of low concentrations of incompatible trace elements in the dehydrated subducted slab and the residual mineralogy that

could retain some trace elements during melting and fractionation processes.

Our study shows, for the first time to our knowledge, that the Mg isotopic composition of some arc lavas differs from that of MORB. Although both crustal assimilation during magma ascent and sediment addition in the mantle wedge likely occur
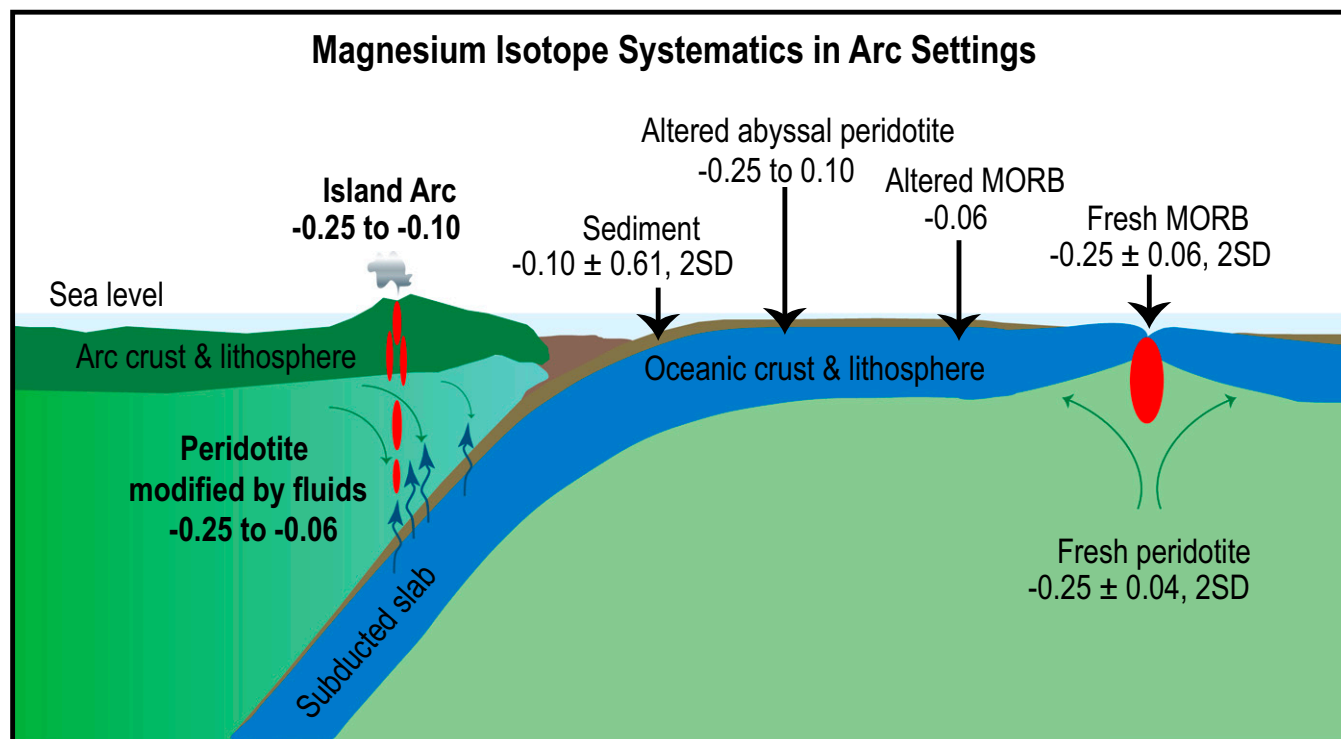
Fig. 4. Sketch of a subduction zone displaying the Mg isotopic compositions of the main components involved in arc magmatism. Seafloor alteration produces isotopically heterogeneous marine sediments, altered MORB, and altered abyssal peridotites. Subduction transports these components into the mantle. Fluids released from these components inherit the isotopically heterogeneous Mg, infiltrate the mantle wedge, and modify its Mg isotopic composition by fluid–rock interactions. Partial melting of this isotopically heterogeneous mantle wedge produced the heterogeneous island arc lavas. Data sources are the same as in Fig. 2.

(4–17), neither of them can be the major process responsible for the Mg isotopic variation in Martinique lavas due to the large concentration contrast between sediment and arc lava or peridotite. Instead, the difference between Martinique lavas and MORB likely results from massive flux of dehydration fluids that leave the subducted oceanic plate to invade the mantle wedge and change its overall isotopic composition (Fig. 4). By combining Mg isotopes with radiogenic isotopes such as Sr, Nd, or Pb, a better picture of the processes occurring during arc genesis can be obtained: In contrast to trace elements that track down the presence of enriched sedimentary materials, a major element such as Mg may help pinpoint the role of subducted products in the overall composition of arc magmas.

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