

Volcanic fronts form as a consequence of serpentinite dehydration in the forearc mantle wedge

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ABSTRACT

The release of fluids from subducting slabs is considered to result in partial melting of the mantle wedge and arc magmatism. By contrast, we propose that the breakdown of serpentinites, which acted as a sink for water and fluid-soluble elements released from underlying slab in the mantle wedge, most likely leads to arc magmatism at volcanic fronts. Serpentinites exhumed from mantle wedges in Himalayas, Cuba, and the Alps are enriched in elements that are fluid soluble at low temperatures, such as As, Sb, and Sr. The downward movement of the serpentinite layer by mantle flow transports these elements to deeper, hotter levels in the mantle. Eventual dehydration of serpentinite discharges water and fluid-soluble elements, leading to partial melting of the overlying mantle wedge, thus accounting for the observed enrichment of these elements in magmas at the volcanic front.

Keywords: subduction zones, volcanic arcs, chalcophile elements, arsenic, antimony, mantle wedge, geochemical cycles.

INTRODUCTION

Active arc volcanoes are typically arranged parallel to the trench, where the subducting slab has reached a depth of ~80–140 km. The intervening forearc region, >100 km wide, is essentially barren of volcanoes (e.g., Gill, 1981; Tatsumi, 1986). The occurrence of the volcanic front is generally attributed to the rapid release of water from subducting slabs as they are metamorphosed to eclogite facies (Tatsumi, 1986; Peacock, 1993; Peacock and Wang, 1999). Water released from subducting slabs migrates upward, leading to the partial melting that drives arc volcanism.

Slabs and overlying sediments continuously release water during subduction (e.g., Schmidt and Poli, 1998; Mibe et al., 1999). Why does this not lead to partial melting and volcanism in the forearc, given that the temperature of the mantle in this region is considered to be >1200 °C (e.g., Furukawa, 1993; Peacock and Wang, 1999) and hence sufficient for partial melting? In addition, the formation of eclogite, causing water release, occurs at a shallower depth than that corresponding to the position of the volcanic front in most subduction zones (Fukao et al., 1983). Furthermore, volcanic-front magmas are enriched in As, Sb, Pb, and B (Ryan et al., 1995; Noll et al., 1996; Leeman, 1996), elements that are mobile even at surface temperatures (Rose et al., 1979; Smedley and Kinniburgh, 2002). How can these mobile elements be retained in descending slabs until they are transformed to eclogite? This paper presents evidence for the enrichment of fluid-soluble elements in serpentinites in the forearc mantle, an elemental assemblage similar to that of volcanic-front magmas. We propose that the generation of these magmas is related to the hydration and subsequent dehydration of peridotites in mantle wedges.

SAMPLES

The serpentinite samples for this study are all associated with eclogites and were exhumed from mantle wedges together with the eclogites (Guillot et al., 2001). Samples were collected from the Ladakh area of the northwest Himalayas, associated with the eclogitic Tso Moriri unit. This unit represents the subducted margin of the In-

dian continent, which reached a depth of ~100 km and a temperature of ~600 °C before it was exhumed together with the serpentinites (Guillot et al., 2001). Thus, the serpentinites represent hydrated mantle peridotites overlying the subduction zone.

Samples from the Alps and Cuba are associated with omphacitic eclogites, a metamorphic product of subducted oceanic crust. The samples from the Alps were collected near Mont Viso in northwestern Italy, and the Cuban samples came from Zaza in central Cuba, part of the Caribbean eclogites (Mann and Gordon, 1996). The serpentinite samples are similar in refractory major element abundance to the Himalayan samples. They also formed by hydration of the mantle wedge and were exhumed together with the eclogites (Schwartz et al., 2001). Voluminous oceanic peridotites in these areas were not used in the study. (See Tables 1 and 2.)¹

CHEMICAL COMPOSITION OF SERPENTINITES

Serpentinites consist mainly of antigorite and minor chrysotile, chlorite, talc, and Mg-rich olivine, as well as primary chromite. All of our serpentinite samples have a similar bulk chemical composition, with high concentrations of compatible elements; the serpentinites also show a gradual decrease in concentrations from highly compatible Mg to moderately compatible Zr (in order starting from the right side of Fig. 1). Such a pattern confirms our earlier interpretation that the serpentinites were derived from a refractory mantle wedge (Guillot et al., 2001).

A refractory mantle should contain low concentrations of Pb, As, Sb, and light rare earth elements (LREEs) because they are preferentially removed from the mantle during partial melting. However, our serpentinite samples, particularly the Himalayan samples, show an extreme enrichment in As (to 130 ppm), Sb (to 12 ppm), Pb (to 23 ppm), Sr (to 56 ppm), and LREEs (to 0.54 ppm Ce) (Fig. 1). The actual degree of enrichment was much greater than that shown in the diagram because their concentrations in a refractory mantle are deduced to be about one-third (Noll et al., 1996) of the primitive-mantle values that are used for normalization (Fig. 1).

CAUSE FOR THE ENRICHMENT OF SELECTED ELEMENTS

A common character among the enriched elements is their high solubility in aqueous fluids (e.g., Rose et al., 1979; Smedley and Kinniburgh, 2002), suggesting that the serpentinites incorporated these elements during hydration of mantle peridotites. It is not surprising that the Himalayan samples show a high enrichment of these elements (Fig. 2), because the subducted slab is mostly composed of clastic sediments derived from Archean granitoids in the northern margin of the Indian continent. Granitic rocks contain higher concentrations of Sb, As, Pb, and Sr than oceanic crust (Rose et al., 1979). This is also supported by very low ϵ_{Nd} values for the serpentinites, as low as -20 (Guillot et al., 2001), similar to Archean granitic rocks.

Physical mixing of crustal rocks with serpentinites may have

¹GSA Data Repository item 2003076, Tables 1 and 2, is available from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, editing@geosociety.org, or at www.geosociety.org/pubs/ft2003.htm.

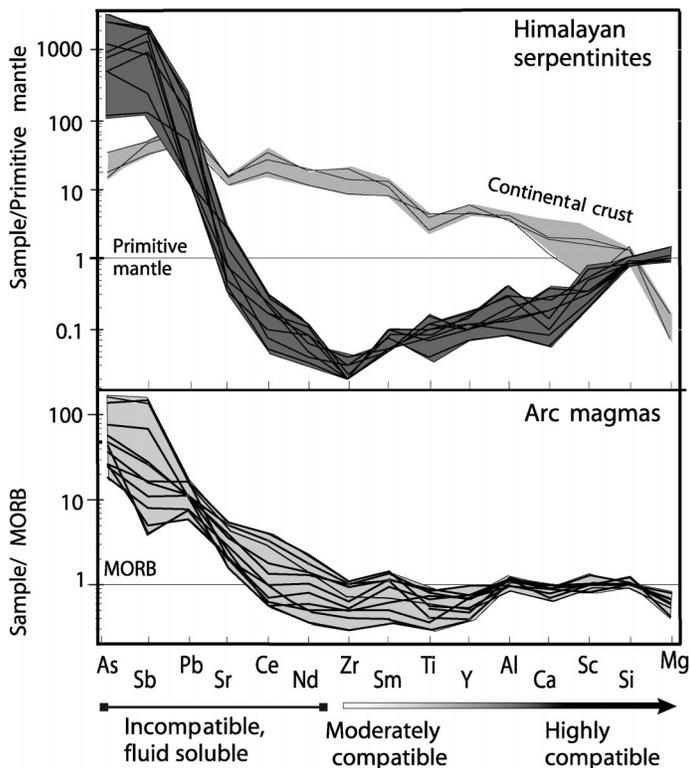


Figure 1. Composition of serpentinites from Himalayas compared to those of continental crust and arc magmas. As and Sb are similar in compatibility to light rare earth elements (Noll et al., 1996), but they are placed on left side of diagram to clarify their fluid-soluble enrichment. Elements from Zr to right are placed in order of their increasing compatibility with mantle minerals. Serpentinites and continental crust are normalized to primitive mantle (McDonough and Sun, 1995), and arc magmas are normalized to mid-oceanic-ridge basalts (MORBs; Noll et al., 1996). Arc magmas include data from Pinatubo, Philippines (Bernard et al., 1997), Esan (Noll et al., 1996), basalt rock standards JB-2 and JB-3 from Geological Survey of Japan (Ando et al., 1989), volcanic rocks in Hishikari area in southern Kyushu (see text footnote 1) and Miyake-jima volcano, Japan (see text footnote 1), Telica in Nicaragua (Lefebure, 1986; Noll et al., 1996), and Medvhizya and Brat volcanoes in Kuril arc, Russia (Volynets, 1994; Noll et al., 1996). Data for bulk continental crust and upper and lower crust are from Taylor and McLennan (1995).

caused such an enrichment, because tectonic mélanges are common along subduction zones. However, this possibility is rejected because of low SiO₂ and the compositional pattern of serpentinites (Fig. 1), and thus there is no evidence for mechanical mixing with crustal rocks.

As, Sb, and Pb are chalcophile, suggesting that sulfides may have some role in the enrichment. All serpentinite samples, however, contain <0.01 wt% S, and other chalcophile elements such as Cd are not enriched. In addition, Sr and Ce, elements unrelated to sulfides, are also enriched. Therefore, S is not a factor in the observed enrichment.

TIMING AND TEMPERATURE OF ENRICHMENT

The elements enriched in serpentinites are all mobile in low-temperature aqueous fluids, on the basis of their enrichment in groundwaters and organic-rich sediments (e.g., Rose et al., 1979) and low-temperature (100 °C) thermal springs (Weissberg et al., 1979). As and Sb are easily reduced from As⁵⁺ and Sb⁵⁺ to soluble As³⁺ and Sb³⁺ during burial of sediments, and Pb is released from feldspars during diagenesis (Hanor, 1979). Mississippi Valley-type deposits, a major source of Pb, formed at <150 °C (e.g., Hanor, 1979). These data indicate that As, Sb, and Pb could be mobile and thus discharged from subducting slabs at low temperatures during the early stage of subduction.

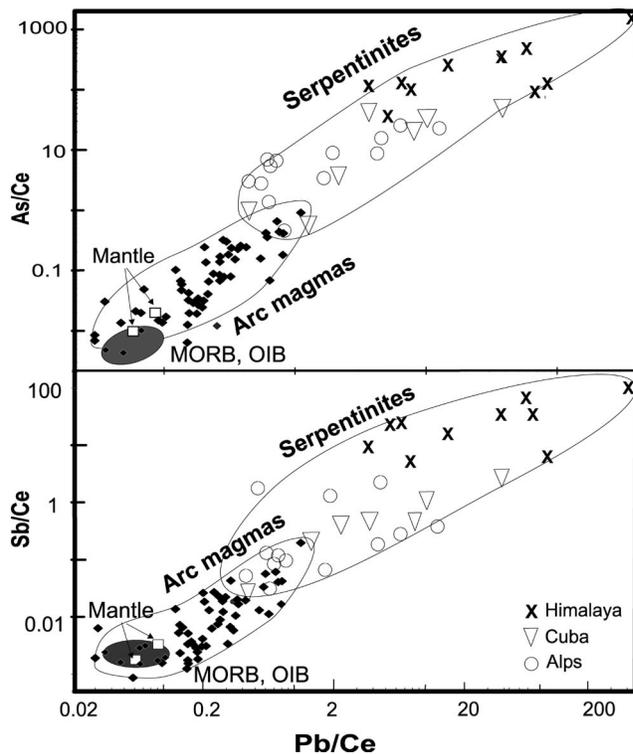


Figure 2. As/Ce, Sb/Ce, and Pb/Ce concentration ratios of serpentinites from Himalayas (X), Alps (open circles), and Cuba (open triangles). Ratios are compared to those of arc magmas (small solid circles), mantle (open squares for primitive mantle and refractory mantle), oceanic-island basalts (OIBs), and mid-oceanic-ridge basalts (MORBs). Ce is mobile in fluids (see Fig. 1), but it was selected for comparison because heavy rare earth elements are not commonly determined for many volcanic rocks. Mantle sample with higher As/Ce, Sb/Ce, and Pb/Ce ratios corresponds to primitive mantle (McDonough and Sun, 1995), whereas sample with low ratios is depleted mantle (Noll et al., 1996). Data sources for arc magmas are listed in Figure 1 caption; other sources are mantle (McDonough and Sun, 1995) and MORB and OIB (Noll et al., 1996).

Such mobility is supported by the composition of subducted sediments that now form the Catalina schist. The contents of As and Sb sharply decrease in epidote-blueschist facies and epidote-amphibolite facies rocks compared to lawsonite-albite facies rocks (Bebout et al., 1999), suggesting that As and Sb were released during early metamorphism at temperatures <300 °C.

Sn and Mo have a compatibility similar to that of LREEs, As, and Sb (Jochum et al., 1993). However, Sn and Mo are not mobile in low-temperature fluids, as demonstrated by the high-temperature (>500 °C) formation of most Sn and Mo deposits. Therefore, the enrichment of As, Sb, and Pb without significant enrichment of Sn and Mo in our serpentinite samples suggests that the incorporation of As, Sb, and Pb and the associated hydration of serpentinites likely took place at low to moderate temperatures, <400 °C.

Our proposed model is supported by recent geophysical data suggesting the occurrence of a serpentinite layer at the base of the forearc mantle wedge (Kamiya and Kobayashi, 2000; Bostock et al., 2002). Direct evidence for the presence of serpentinites is provided by serpentinite diapirs in forearc regions, such as the Mariana arc (Fryer, 1996). A study of B isotopes in these serpentinites confirms that the mantle peridotites hydrated at relatively low temperatures, <300 °C (Benton et al., 2001).

COMPOSITION OF ARC MAGMAS

Arc magmas are characterized by high concentrations of large ion lithophile elements (e.g., Gill, 1981). The enriched elements comprise

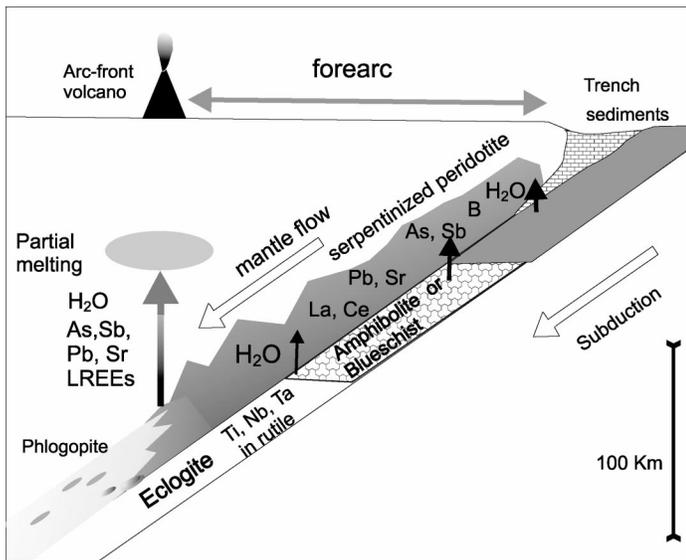


Figure 3. Mantle peridotites overlying subduction plane are continuously hydrated (as shown in gray shading) during dehydration of slabs, becoming enriched in As, Sb, Pb, Sr, and light rare earth elements (LREEs). Serpentinities are transported downward by movement of subducting slab. When they reach ~100 km depth, serpentine minerals break down, releasing most of their H₂O, ~13 wt%. Upward migration of H₂O leads to partial melting that generates arc magmas rich in fluid-soluble elements that resided in temporary serpentinite reservoir.

not only lithophile elements, but also fluid-soluble chalcophile elements such as Pb, As, and Sb (Ryan et al., 1995; Noll et al., 1996; Leeman, 1996). The As/Ce, Sb/Ce, and Pb/Ce ratios of arc magmas are much higher than those of mid-oceanic-ridge basalts (MORBs) and oceanic-island basalts (OIBs) (Fig. 2), illustrating the enrichment of As, Sb, and Pb in the mantle source region of arc magmas. The extent of enrichment at the source is evaluated by comparing arc magmas with MORB (Fig. 1), because the comparison of magmas eliminates the effects of melt formation. This approach shows that the source is enriched in the following order: As, Sb, Pb, and Ce. The enrichment of the source mantle is similar to that of the serpentinites (Fig. 1), suggesting a link between serpentinite composition and generation of arc magmas.

PROPOSED MODEL

Altered and hydrated oceanic basalts and sediments contain high concentrations of As, Sb, and Pb (Plank and Ludden, 1992; Jochum and Verma, 1996). Water and these soluble elements are expelled from slabs during the early stages of subduction. These components migrate upward and hydrate the mantle peridotite to form a serpentinite layer along and just above the subduction zone (Fig. 3). The dissolved elements become highly concentrated as the water is consumed by hydration and are eventually fixed in the serpentinites when the water is totally consumed.

Serpentinities contain >13 wt% H₂O in mineral structures, providing a sink for the flux of water during subduction dehydration. Volume increase during peridotite hydration also prevents water from permeating into the interior of the mantle wedge. Therefore, serpentinite layers act as a barrier, keeping the interior of the mantle wedge dry and free from partial melting, even though it is sufficiently hot (e.g., Furukawa, 1993) to partially melt if hydrated.

The serpentinite layers are dragged downward by mantle flow (e.g., Furukawa, 1993), allowing trace elements that are mobile at low temperatures to be transferred to the deep mantle wedge. When the temperature reaches ~650 °C, serpentinites decompose to forsterite and

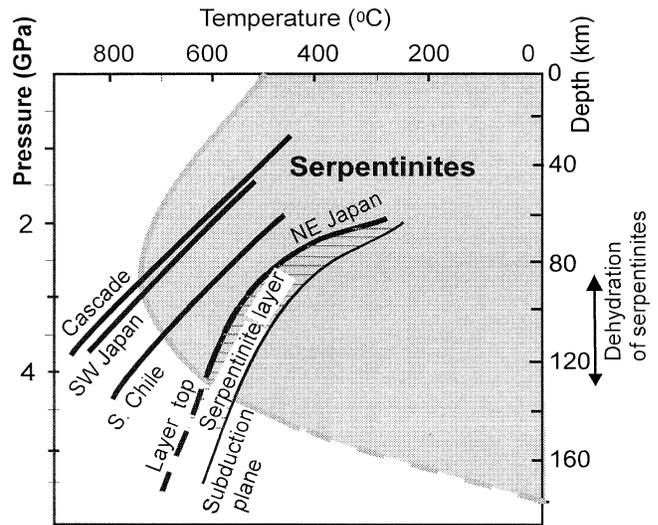


Figure 4. Stability of serpentinites along pressure-temperature (*P-T*) paths of subduction zones (thick solid curves). *P-T* condition at top of 10-km-thick serpentinites is schematically shown on right side of *P-T* paths for northeastern Japan. Stability of serpentinites is constructed from experimental data (Ulmer and Trommsdorff, 1995; Wunder et al., 2001) combined with invariant points for dehydration reactions determined by Peacock (2001). Data sources for *P-T* paths: Fukao et al. (1983), Furukawa (1993), and Peacock and Wang (1999).

enstatite (Ulmer and Trommsdorff, 1995; Wunder et al., 2001). This temperature corresponds to ~90–110 km depth in many arcs, such as Cascadia, southwestern Japan, and southern Chile (Fig. 4).

Our model also explains magmatism related to subduction of old slabs, which are cold. Their subduction planes may not reach 650 °C, even at 130 km depth. However, in these cases the serpentinite layers are thick because of an extended period of water supply. For example, the thickness of the serpentinite layer in northeast Japan, the longest-lasting subduction zone, is estimated to be >10 km (e.g., Kamiya and Kobayashi, 2000), and the top of this layer is much hotter than the base. Serpentinities ~10 km above the subduction plane in northeastern Japan at 130 km depth start dehydrating at 600–650 °C (Peacock, 2001; Fig. 4), thus driving partial melting above this region.

The breakdown of serpentinites releases a large amount of water as well as elements that are not compatible with olivine. These components migrate upward and are incorporated by the partial melt that generates arc magmas.

The widely held view of the formation of arc magmas involves a rapid release of water from subducting slabs during the transition from amphibolite or blueschist to eclogite (e.g., Tatsumi, 1986, 1989; Peacock, 1993; Peacock and Wang, 1999). The water forms amphiboles in the overlying mantle wedge, and their breakdown leads to partial melting. Several workers have noted the problems associated with this model, including the shallow depth for eclogite formation (Fukao et al., 1983; Schmidt and Poli, 1998; Mibe et al., 1999). The discrepancy in depth between that of eclogite formation and frontal-arc magma generation is >30 km in northeastern Japan (Fukao et al., 1983). Such discrepancy may be attributed to downward flow of amphibole peridotite, but this interpretation implies a kinetic delay of dehydration by >600 k.y., even using a steep subduction angle of 30° and a fast subduction rate of 10 km/yr. This time interval seems to be unreasonably long for the dehydration of amphiboles.

Other explanations for the lack of magmatism in forearc regions include mechanical difficulties involved with the ascent of magma and aqueous fluids (Mibe et al., 1999; Schmidt and Poli, 1998). Experiments indicate no upward movement of aqueous fluids at low pres-

tures, <2 GPa, in the mantle (Mibe et al., 1999). However, the static experimental conditions are quite different from the natural environment, where fluid flow is commonly driven by gradients in fluid pressure and under conditions of high stress. Abundant evidence indicates that fluids move along fault and shear zones at lower-crustal to mantle depths (e.g., Cathles, 1990). In addition, the occurrence of serpentinites in the forearc mantle (Fryer, 1996) attests to the movement of aqueous fluids under relatively low pressures in the mantle.

Our model is consistent with the across-arc geochemical variations of magmas. The enrichment of As, Sb, and Pb is strongest at the volcanic front and decreases sharply with increasing distance from the volcanic front (Ryan et al., 1995; Noll et al., 1996). Serpentinites incorporate these elements at low temperatures, but retain them only as long as serpentinites are stable. Once the serpentinites are destabilized, they are discharged with aqueous fluids, dissipated upward into the mantle, and partially incorporated in melt.

In addition, our model also accounts for the different behavior observed among high field strength elements (HFSEs; elements with large valance and small ionic radii) in subduction zones. Low concentrations of HFSEs are considered to be the key characteristic feature of arc magmas (Gill, 1981), because these elements are generally thought to be incorporated into rutile in subducting slabs and thus are not available for arc magmas. Sb is a HFSE with an ionic radius similar to that of Nb and thus should be low in arc magmas, but the two are decoupled (the so-called "Sb paradox"; Zack et al., 2002). The concentration of Sb is high in arc magmas, particularly in frontal-arc magmas (Noll et al., 1996). Our model suggests that Sb is released from the subducting slabs and transferred to the overlying mantle at low temperatures, leading to its eventual enrichment in frontal-arc magmas (Fig. 3). However, Nb and Ta are insoluble at low temperatures and remain in the slabs, to be incorporated into metamorphic rutile. Thus, the different behavior of HFSEs in low-temperature fluids can explain their decoupling and disparate fates during subduction.

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