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Fluid- and melt-related enrichment in the subarc mantle: Evidence from Nb/Ta variations in island-arc basalts

A. J. Stolz*

K. P. Jochum

B. Spettel

A. W. Hofmann

Max-Planck-Institut für Chemie, Abt. Geochemie, Postfach 3060, 55020 Mainz, Germany

ABSTRACT

The single most distinctive feature of volcanic rocks from convergent-margin settings is a marked depletion of the high field strength elements (HFSE) Nb, Ta, and Ti relative to large ion lithophile and light rare earth elements when compared with basalts from mid-oceanic ridges (MORB) and the oceanic islands. A major impediment to a better understanding of this problem has been a lack of high-quality data for the HFSE (particularly Nb and Ta) that occur in very low concentrations in most volcanic rocks from convergent-margin settings. We report new analyses of Nb and Ta for a suite of island-arc volcanic rocks as well as some sea-floor sediments. Our data show that Nb/Ta values for relatively depleted island-arc volcanic rocks are similar to MORB and essentially chondritic (Nb/Ta ~ 17), whereas more potassic arc volcanics have substantially higher Nb/Ta values (up to 33). We interpret these high values as due to modification of the subarc mantle source by silicic melts derived from the subducting slab, whereas enrichment of the source regions of the less potassic arc rocks involved a slab-derived fluid.

INTRODUCTION

Possible explanations for the relative high field strength element (HFSE) depletion in subduction-related volcanic rocks include previous melt depletion of the peridotitic mantle wedge (Green, 1972; Pearce, 1983), depletion by reaction with depleted mantle material, which fractionates the large ion lithophile elements (LILE) from the HFSEs due to differing bulk partition coefficients (Kelemen et al., 1990), and the presence of residual Ti-rich phases either within the mantle wedge (Morris and Hart, 1983) or the subducted slab (Saunders et al., 1980, 1991). Previous studies of Nb/Ta variation in mid-ocean ridge basalt (MORB), komatiite, depleted mantle xenoliths (Jochum et al., 1989), and chondritic meteorites (Jochum et al., 1986) suggested that the Nb/Ta of the Earth's mantle has a chondritic value of about 17.5 and that there is no significant fractionation of these elements, at least at relatively large degrees of partial melting. However, experimental studies (Green and Pearson, 1987; Green et al., 1989; Green, 1995) of the partitioning of these elements between various melt compositions and potential residual phases in the mantle and subducted oceanic lithosphere suggest that in certain circumstances this pair of geochemically similar elements may be fractionated. In this paper we explore the potential constraints Nb/Ta variations may provide on the source components of island-arc volcanics.

Nb/Ta FRACTIONATION IN SUBARC MANTLE SOURCE REGIONS

Most models for the origin of island-arc volcanics invoke at least two principal source components, typically an HFSE-depleted peridotite in the mantle wedge above the subducting oceanic crust, and either an LILE-enriched fluid or melt released during dehy-

dration or partial melting of the subducting slab (Fig. 1; Nicholls and Ringwood, 1973; Saunders et al., 1980). However, the criteria for distinguishing these contrasting styles of source enrichment are not well known. The addition of LILE via a fluid to a previously depleted mantle peridotite is consistent with the relatively high solubilities of these elements in hydrous fluids (Tatsumi et al., 1986) and the generally low concentrations of HFSE and rare earth elements (REE) in island-arc basalts compared with MORB and ocean-island basalt (OIB). However, this mechanism does not explain the observation that the more potassic island-arc volcanic rocks also appear to require some addition of HFSEs (Zr, Nb, Ta) and light (L) REEs to a depleted mantle source prior to melting (Stolz et al., 1990). The addition of significant amounts of HFSEs via a fluid phase is generally considered unlikely due to the low solubilities of these elements in hydrous fluids, hence an alternative mechanism is required to account for their source characteristics (Stolz et al., 1995).

For this study we analyzed a suite of island-arc volcanic rocks and some modern sea-floor sediments by a combination of spark source mass spectrometry (SSMS; Jochum et al., 1990) and instrumental neutron activation (INAA). All samples were crushed in agate, because tungsten carbide crushing vessels are a well-known source of Ta contamination. The Nb and Ta analyses by SSMS have a precision of 5% to 7% (Jochum et al., 1990), and the INAA Ta analyses have a precision better than 7%. Our island-arc volcanic samples are predominantly from the Sunda arc and range in composition from low-K tholeiite, through medium-K and high-K calcalkaline types, to the more potassic trachybasalts and leucite basanites (Stolz et al., 1990). In addition, we have included some relatively

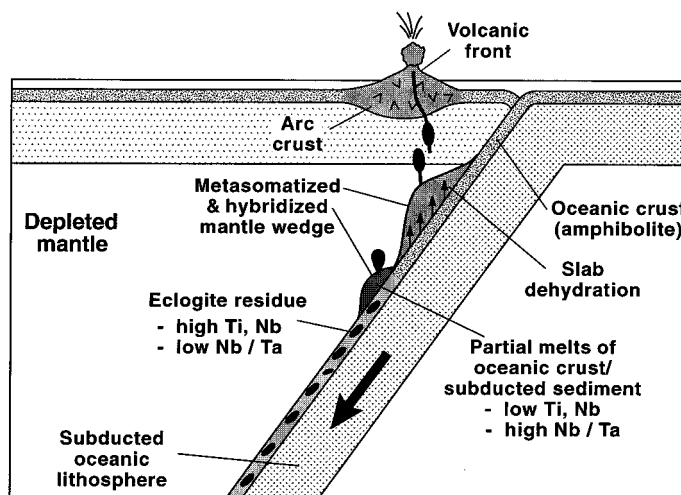


Figure 1. Diagrammatic model showing different styles of source modification for subduction-related magmas. Metasomatism of mantle wedge due to dehydration of subducting slab is principal mechanism for large ion lithophile element enrichment of source at relatively shallow levels, whereas hybridization of depleted mantle peridotite by silicic melts derived from slab is inferred to be more important at greater depths.

*Present address: Department of Geology, University of Tasmania, G.P.O. Box 252C, Hobart, Tasmania 7001, Australia.

TABLE 1. TRACE ELEMENT DATA AND Nb/Ta VALUES FOR ISLAND-ARC VOLCANIC ROCKS AND SEDIMENTS

	SiO ₂ (%)	Zr (ppm)	Nb (ppm)	Ta (ppm)	Nb/Ta
Island-arc volcanic rocks					
32NG0754 Bismark Arc	48.10	24.2	0.663	0.037	17.9
T113 Tonga	52.84	13.6	0.736	0.043	17.1
89UH5B	48.17	31.1	0.934	0.052	18.0
85UH197	49.97	30.7	1.178	0.070	16.8
67238	52.90	78.6	4.89	0.250	19.6
LBK86/2	53.74	114	5.57	0.310	18.0
66554	45.19	100	8.95	0.270	33.1
66556	47.06	109	8.12	0.290	28.0
66575	45.89	111	6.69	0.220	30.4
67132	47.76	195	18.7	0.750	24.9
67130	47.68	195	19.6	0.660	29.7
Pelagic sediments					
GS7605/48			10.8	0.620	17.4
GS7605/58			19.2	1.24	15.5
GS7605/61			31.4	1.91	16.4

Note: Nb in arc volcanic rocks was analyzed by SSMS and Ta by INAA (see text for details). Nb and Ta in sediment samples analyzed by SSMS. Additional data for the sediment samples are given in Ben Othman et al. (1989).

depleted tholeiitic samples from the Bismark (Johnson et al., 1985) and Tongan (Ewart et al., 1977; Ewart and Hawkesworth, 1987) arcs as representatives of more-depleted island-arc volcanic compositions. The volcanic rocks mostly have SiO₂ < 53 wt%, TiO₂ < 1 wt%, MgO > 5.5 wt%, and Nb concentrations of 0.66 to 19.6 ppm and Ta in the range 0.037 to 0.75 ppm (Table 1). A plot of Nb versus Nb/Ta (Fig. 2) shows that our tholeiitic and low to medium K calc-alkaline samples from island arcs have essentially constant Nb/Ta values with an average of 17 ± 1, which is similar to the chondritic value and the average for MORB (~17; Stolz et al., 1995). In contrast, relatively HFSE-enriched calc-alkaline and K-rich alkaline rocks from the Sunda arc have remarkably high Nb/Ta values (up to 33). The leucite basanites (i.e., 67130, 67132) represent near-primary magma compositions with 100 Mg/(Mg + Fe²⁺) = 68, and they have minor Cr-spinel and Ti-poor magnetite microphenocrysts associated with olivine, clinopyroxene, and leucite phenocrysts. There is no evidence in these rocks of ilmenite or sphene that may have fractionated the HFSE during low-pressure crystallization, which indicates that substantial fractionation of Nb and Ta has either occurred in their source or been imposed on their source.

DISCUSSION

There are several possible explanations for the very high Nb/Ta values in the more potassic arc volcanic samples. These include equilibration in their source region with a residual phase that resulted in the Nb/Ta fractionation, or derivation from a depleted mantle source that had been enriched by a component characterized by a high Nb/Ta. This component could be a silicate or carbonatite melt, or possibly a fluid.

Residual Phase Control

The experimental data on the solubility of Ti in basaltic melts argue against the presence of residual Ti-rich phases such as rutile, ilmenite, and perovskite in the mantle wedge (Ryerson and Watson,

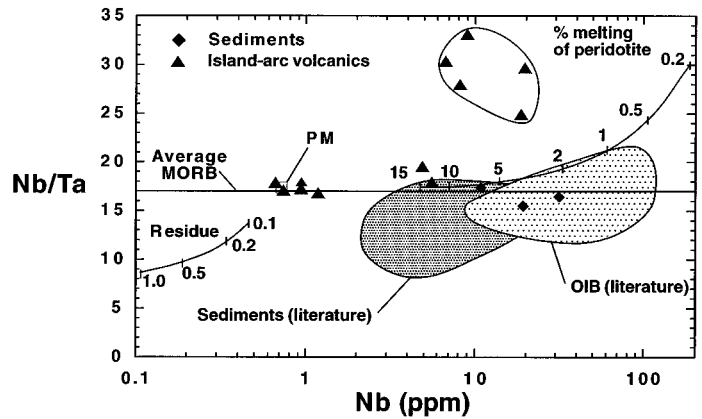


Figure 2. Plot of Nb vs. Nb/Ta comparing island-arc basalts and oceanic sediments analyzed in this study with literature data for sediments (Govindaraju, 1994; Vroon et al., 1995), oceanic-island basalts (OIB; Clague and Frey, 1982; Frey and Clague, 1983; Palacz and Saunders, 1986; Weaver et al., 1987; Chauvel et al., 1992), average mid oceanic-ridge basalt (MORB; Sun and McDonough, 1989), and primitive mantle (PM). Also shown is calculated variation of Nb with Nb/Ta in melt and residue at variable degrees of batch melting of peridotitic source with primitive mantle Nb (0.713 ppm) and Ta (0.041 ppm) concentrations. Peridotite melting models are based on starting composition of olivine (0.60), orthopyroxene (0.25), clinopyroxene (0.10), and garnet (0.05) with all phases residual up to 15% melting. D_{Nb} and D_{Ta} values used for garnet/melt (0.02 and 0.06) and clinopyroxene/melt (0.005 and 0.013) are from Green et al. (1989).

1987), although it has been suggested that under the unusual conditions relating to the origin of K-rich arc volcanic rocks (i.e., high pressure, fO_2 , alkalis, and aH_2O) such a phase may be stabilized in the residue (Foley and Wheller, 1990). High-pressure (up to 7 GPa) experiments on a lamproite from the Leucite Hills demonstrated the stability of an unusual K-Ti silicate phase (Mitchell, 1995), although these lamproites have substantially higher TiO₂ than the subduction-related potassic volcanic rocks from the Sunda arc, and the Ti-rich phase only occurred at near-solidus temperatures, which suggests that it may not remain in the residue during melting.

Other possible residual phases in the source regions of potassic alkaline arc rocks include phlogopite, amphibole, garnet, clinopyroxene, orthopyroxene, and olivine. The latter two phases from ultramafic xenoliths have such low concentrations of Nb and Ta (Ionov et al., 1995) that they are unlikely to influence the Nb/Ta values in derivative melts. Experimental data for the partitioning of Nb and Ta between phlogopite and basanitic melt (Adam et al., 1993) indicate that phlogopite has a negligible capacity to fractionate Nb/Ta, and although pargasitic amphibole can fractionate Nb/Ta to relatively high values in carbonatitic compositions, it has a less pronounced or opposite effect for basanitic compositions (Green, 1995). Of considerable interest are very high Nb/Ta values in pargasitic amphiboles and micas recently reported for metasomatized ultramafic xenoliths from Siberia and Mongolia (Ionov and Hofmann, 1995). These were interpreted as products of reaction between depleted mantle and a silica-rich fluid from a subducted slab, and they clearly demonstrate the strong dependence of Nb and Ta partitioning on fluid or melt composition. Richteritic amphibole is also a potential phase in the source regions of K-rich volcanic rocks, but its ability to buffer Nb/Ta values is unknown.

Experimental partition coefficient data for clinopyroxene and garnet with basaltic melts (Green et al., 1989) indicate that they may be important in fractionating Nb and Ta during mantle melting. These effects have been modeled assuming primitive mantle Nb and Ta concentrations, and the calculated likely maximum variation of Nb concentration with Nb/Ta in the melt and residue at various

degrees of batch melting is shown in Figure 2. At very low degrees of melting (<1%) Nb/Ta values may be similar to the leucites, but calculated Nb concentrations (>100 ppm at 0.5% melting) are much higher, and Zr/Nb values (~5) are much lower than for the leucites. However, if an HFSE-depleted mantle source is used as the starting composition, the calculated Nb/Ta values for the melt are much lower (<14) due to the significant reduction of Nb/Ta in the residue after removal of even small melt fractions (Fig. 2). The above common residual phases clearly have the capacity to fractionate Nb/Ta values in very small degree equilibrium basic melts and their residues if the experimentally determined D_{Nb} and D_{Ta} (mineral-melt partition coefficient) values are appropriate. However, the high Nb/Ta values and other trace element characteristics of the K-rich arc rocks cannot be explained by a simple control involving residual phases in their mantle sources.

Fluid- or Melt-Related Source Enrichment?

Recent experimental determination of the partitioning of Nb and Ta between rutile and hydrous fluid (Brenan et al., 1994) demonstrated a strong preference of Nb and Ta for rutile compared with the coexisting hydrous fluid. The very high measured D_{Nb} and D_{Ta} values for rutile/fluid (>100) suggest that negligible transfer of Nb and Ta will occur from the slab to the mantle wedge via a hydrous fluid in the presence of rutile. Moreover, as D_{Nb}/D_{Ta} for rutile/fluid >1 in most experiments, any enrichment in HFSE via a hydrous fluid would be expected to result in a decreased rather than increased Nb/Ta in the mantle wedge. Carbonatite melts have been proposed as an important agent for modifying the subcontinental lithospheric mantle (Green and Wallace, 1988; Yaxley et al., 1991) and also in subduction zones (Sweeney et al., 1992). Experimental data (Green, 1995) suggest that Nb and Ta may be significantly fractionated in primary carbonatite melts that have equilibrated with pargasite (cf. Sweeney et al., 1995). However, carbonatites typically have very low K and Rb and hence are not likely to be important in modifying the sources of these K-rich magmas. Simple bulk addition of a sedimentary component also cannot produce the high Nb/Ta values of the K-rich rocks: our data and those in the literature (Fig. 2; Govindaraju, 1994; Vroon et al., 1995) indicate that Nb/Ta values for pelagic or terrigenous sediments typically are similar to, or lower than, the chondritic value.

A more likely scenario is that silicic melts derived either from the altered oceanic crust or subducted pelagic or terrigenous sediment may hybridize the depleted mantle peridotite above the subducting slab, resulting in a phlogopite-bearing harzburgite/lherzolite or phlogopite orthopyroxene/websterite (Wyllie and Sekine, 1982), depending on the degree of clinopyroxene depletion in the source and the amount of silicic melt involved. Because of the lower Ti solubility in more siliceous melts, it is reasonable to expect a phase such as rutile or ilmenite to persist in the residue that equilibrated with these silicic melts (Ryerson and Watson, 1987). $D_{Nb/Ta}$ (i.e., the mineral-melt partition coefficient for Nb divided by the partition coefficient for Ta) is ~0.6 for rutile/andesitic melt (Green and Pearson, 1987), but is unknown for more silicic melt compositions. Batch melting calculations utilizing an N-MORB (normal-type mid-oceanic ridge basalt) starting composition (Nb = 2.33, Ta = 0.132, Nb/Ta = 17.7; Sun and McDonough, 1989) and melt and phase proportions based on recent melting experiments (Sen and Dunn, 1994) suggest that an increase in Nb/Ta to about 25 is possible with ~1 wt% rutile in the residue. Higher values would require larger amounts of rutile in the residue or a smaller value of $D_{Nb/Ta}$ for rutile/melt. It could be argued that as the D_{Nb} and D_{Ta} for rutile/melt are very high, there will be insufficient Nb and Ta added to the mantle via a melt phase to significantly modify the mantle Nb/Ta. However, the calculated Nb concentration in a 10% silicic melt of

MORB is ~2.8 ppm. When added to a previously depleted mantle source with <0.1 ppm Nb, the Nb/Ta value of the silicic melt will dominate the hybrid source.

Nb/Ta variations during partial melting of pelitic sediment are also controlled by residual rutile unless biotite is present in the residue. If the low-pressure phenocryst/matrix partition coefficient data for biotite/silicic melt (Nash and Crecraft, 1985) are applicable at elevated pressures, it may be possible to generate substantially lower Nb/Ta values (~8) in the melt. Such low values are clearly inappropriate for the Indonesian K-rich rocks, but may explain the low average Nb/Ta value (~10) reported for leucite basanites from Vulsini volcano, Italy, which also display substantial isotopic evidence for an older crustal-derived component (Rogers et al., 1985).

Nb/Ta values lower than the chondritic value (10–17) have also recently been reported for MORB and volcanic rocks from the Mariana arc (Plank et al., 1994; Plank and White, 1995), as well as volcanic rocks from the Bismark arc (Eggs et al., 1994). In these cases the lower Nb/Ta values seem to correlate with lower HFSE concentrations in the volcanic rocks. Clearly, previous melt depletion of a mantle source region has a significant effect on the absolute abundances of HFSE in island-arc volcanic rocks, and may result in fractionation of Nb and Ta (cf. Fig. 2). However, the essentially constant and chondritic Nb/Ta values in our tholeiitic and calc-alkaline samples suggest they were derived from a moderately depleted mantle wedge (with unfractionated Nb/Ta) that had been modified by the addition of an LILE-enriched and HFSE-poor fluid derived from the subducted slab.

The conclusion from this study that both fluid and melt-related source enrichment occurs in the subarc mantle is supported by other trace element data for low-K to K-rich alkaline volcanic rocks from Indonesia. Edwards et al. (1993) showed that tholeiitic and medium-K calc-alkaline volcanic rocks have elevated B/Be values, whereas shoshonitic and K-rich alkaline rocks (which have an average Nb/Ta ~24; Edwards et al., 1991) have consistently lower B/Be values. They interpreted the high B/Be values as evidence that the tholeiitic and calc-alkaline volcanic rocks were derived from mantle modified by a fluid flux from the subducted slab, whereas they suggested the source regions of the K-rich alkaline rocks had not been modified by recent subduction. In our model the low B/Be values of the K-rich alkaline rocks would reflect extensive recycling of the B via hydrous fluids at relatively shallow levels prior to partial melting of the slab to give the silicic melts that hybridized the mantle source of the K-rich arc magmas.

CONCLUSIONS

This study provides substantial support for a model in which the mantle source regions of K-rich arc magmas are hybridized by silicic melts derived from the subducting slab in contrast to fluid metasomatism in the source regions of relatively low-K arc-volcanic rocks. Additional high-quality Nb and Ta analyses of island-arc volcanics of varying compositions from different arcs will permit a more detailed assessment of the role of residual phases in subducted crust, particularly with respect to modification of the mantle source regions of island-arc volcanic rocks.

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