Re-Os systematics of high-Mg andesites and basalts from the Setouchi volcanic belt, SW Japan: Implications for interaction between wedge mantle and slab-derived melt

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High-Mg andesites (HMAs) and basalts in the Setouchi volcanic belt exhibit large variations in both Os abundance and Os isotopic composition. For HMAs, $^{187}Os/^{188}Os$ ratios and Os concentrations range from 0.1718 to 0.2041 and from 8.1 to 11.5 ppt, respectively. For basalts, $^{187}Os/^{188}Os$ ratios and Os concentration range from 0.1556 to 0.1769 and from 31.0 to 53.7 ppt, respectively. The measured $^{187}Os/^{188}Os$ ratios are higher than that of the primitive upper mantle estimate. The petrological and geochemical characteristics of HMAs do not support a scenario whereby HMA magmas are derived from basalt magmas via assimilation-fractional crystallization processes within the crust. Instead, the observed elevated Os isotopic compositions may be explained by mixing between the wedge mantle and partial melts from subducted sediments, rather than slab-derived fluids.

INTRODUCTION

The unique geochemical characteristics of subduction zone magmas have long been attributed to recycling of components from the subducting slab into the overlying mantle wedge source. However, the relative importance of different mechanisms by which this material transfer may occur remains unclear, as do the factors controlling which mechanisms dominate. Melting of the descending lithospheric slab may have occurred during subduction in Archean time due to steep mantle geothermal gradients. Since first proposed by Nicholls and Ringwood (1973) many petrologists have thus favored a mechanism involving slab melting and reaction of these melts with the overlying mantle wedge (e.g., Kay, 1978; Sekine and Wyllie, 1982; Brophy and Marsh, 1986; Martin, 1987; Defant and Drummond, 1990; Keleman, 1995). Alternatively, fluids produced by dehydration reactions in the subducting lithosphere may cause partial melting and metasomatism in the mantle wedge (e.g., Coats, 1962). In most modern subduction zones, these aqueous fluids are the most probable cause of melting of the overlying mantle wedge (e.g., Tatsumi and Eggins, 1995), as melting of the oceanic crust is not likely to occur (Peacock, 1990). Whether direct slab melting or slab dehydration

occurs, Os isotopes are a potentially useful tracer for detecting subduction components in arc magmas because crustal materials, including oceanic crust and sediments, have much higher Re/Os and $^{187}Os/^{188}Os$ ratios (Esser and Turekian, 1993; Ravizza, 1993; Peucker-Ehrenbrink *et al.*, 1995) than upper mantle peridotite (e.g., Meisel *et al.*, 2001).

The behavior of Re and Os in the subduction environment is poorly constrained. Peridotite xenoliths with higher Os isotopic compositions (up to 0.156) than the hypothetical Primitive Upper Mantle (PUM) Os isotopic ratio of 0.1296 (Meisel *et al.*, 2001) have been found in arc and back-arc settings (Brandon *et al.*, 1996, 1999; McInnes *et al.*, 1999; Becker *et al.*, 2001; Widom *et al.*, 2003). Most of the researchers proposed that these high $^{187}Os/^{188}Os$ ratios may result, at least in part, from the contribution of slab-derived components, while some argued it possibly attributed to crustal contamination. Several studies have recently reported that primitive arc lavas possess elevated Os isotope ratios of 0.128–3.70 (Alves *et al.*, 1999, 2002; Borg *et al.*, 2000; Lassiter and Luhr, 2001; Suzuki and Tatsumi, 2001; Hart *et al.*, 2002; Woodland *et al.*, 2002). Alves *et al.* (1999, 2002) and Borg *et al.* (2000) suggested that these radiogenic $^{187}Os/^{188}Os$ ratios reflect the variable input of slab-derived materials. Fluids have been shown experimentally to be able to transport Os (e.g., Suzuki *et al.*, 2003) and Brandon *et al.* (1996) suggested that high f_{H2O} and f_{HCl} in a slab-derived fluid may increase Os mobility in subduction zone envi-

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ronments. However, Becker (2000) considered that Os is not significantly lost from the subducting slab via dehydration processes, on the basis of similar Os abundances in eclogites and blueschists to those in MORB.

Contribution to the sub-arc mantle by overlying crust with highly radiogenic Os isotopes could also produce high $^{187}Os/^{188}Os$ ratios (Hart *et al.*, 2002). Lassiter and Luhr (2001) have suggested that late stage contamination of primitive arc magmas by crustal material during assimilation/fractional crystallization (AFC) processes is the dominant route by which the Os isotopic signatures of the Mexican primitive arc volcanic rocks were generated. Borg *et al.* (2000) also attempted AFC modeling for the Cascade arc lavas, but they could not explain correlations between Os and other isotopes and therefore favored the hypothesis that slab-related material input contributed to the correlations.

High magnesian andesites (HMAs) can occur in subduction zone settings. The high Mg/Fe ratios of HMAs provide evidence for their equilibration with upper mantle peridotite (Sato, 1977; Kuroda *et al.*, 1978; Jenner, 1981; Crawford *et al.*, 1989). Melting experiments in both synthetic peridotite systems (e.g., Kushiro, 1969, 1972) and using natural HMAs (Tatsumi, 1981, 1982; Umino and Kushiro, 1989; van Der Laan *et al.*, 1989) confirmed that hydrous melting of peridotite can yield HMAs. Such hydrous upper mantle conditions can be attained through addition of fluids released during dehydration of the subducting slab (Crawford *et al.*, 1989; Tatsumi and Maruyama, 1989). Alternatively, interaction between slab-derived hydrous silicic partial melts and overlying mantle wedge peridotite has also attracted recent attention as a mechanism for HMA formation (Kay, 1978; Pearce *et al.*, 1992; Yogodzinski *et al.*, 1994; Kelemen, 1995; Hanyu *et al.*, 2002; Tatsumi and Hanyu, 2003). Magmatism in the Setouchi volcanic belt (SVB) may provide an excellent example for such unusual tectonic conditions, as the lithosphere being subducted beneath the region was created at 27–15 Ma (Okino *et al.*, 1994; Tamaki *et al.*, 1999), immediately prior to the Setouchi magmatism at 13 Ma. As such, the subducting oceanic crust may have been sufficiently hot for partial melting to have occurred (Furukawa and Tatsumi, 1999; Peacock and Wang, 1999). In addition, opening of the backarc basin behind the SW Japan arc at 28–14 Ma (Kaneoka *et al.*, 1992; Tada and Tamaki, 1992; Tamaki *et al.*, 1992) may have resulted in unusually high temperature conditions in the mantle wedge (Tatsumi and Maruyama, 1989; Kimura *et al.*, 2003).

Shimoda *et al.* (1998) reported Pb-Nd-Sr isotopic compositions for HMAs and basalts from the SVB and demonstrated a significant contribution from a subducted sediment component to the HMA magma source. Furthermore,
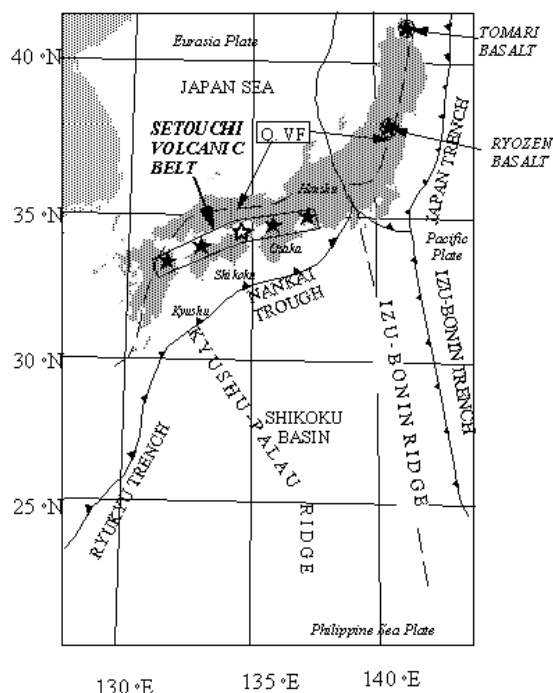

Fig. 1. A tectonic map of the SW Japan arc (modified from Shimoda et al., 1998). The Setouchi volcanic belt is located in the forearc region of the SW Japan arc, i. e., trench-side of the Quaternary volcanic front (Q. VF). The central three subprovinces of the volcanic belt in Shikoku and Osaka are characterized by the occurrence of HMAs. The samples analyzed in this study were collected from the area shown by an open star in the SVB.

the results of mixing calculations using an inferred original mantle composition and local oceanic sediments led Shimoda *et al.* (1998) to conclude that a sediment-derived melt, rather than aqueous fluids or oceanic crust-derived melts, could have served as a plausible metasomatic agent for the HMA magma source. This was further supported by the results of geochemical modeling of partial melting of sediments and altered MORB and melt-solid interactions (Tatsumi, 2001; Tatsumi and Hanyu, 2003). A sediment-melt produced at 1050°C and 1.0 GPa changes its composition from rhyolitic to andesitic as it dissolves olivine and clinopyroxene from peridotite in the mantle wedge and crystallizes orthopyroxene to generate HMA (Tatsumi, 2001; Tatsumi and Hanyu, 2003).

In this study, we report Re and Os concentrations and Os isotopes for Setouchi HMAs and basalts from SW Japan. Ryozen and Tomari basalts from NE Japan are also analyzed for comparison. We then examine the contribution of slab-derived melts to the genesis of HMA and basalt magmas in the SVB.

Table 1. Major element compositions of samples

Sample Rock type	Setouchi						Miocene NE Japan	
	SD SYB basalt	SD424 basalt	SD-261 HMA	SD407 HMA	SD411 HMA	JA-2 HMA	RZ-5 basalt	TM-0 basalt
SiO_2	49.20	48.79	55.73	56.99	55.80	56.18	48.46	49.24
TiO_2	1.01	0.97	0.71	0.60	0.61	0.67	0.62	0.5
Al_2O_3	15.31	13.46	15.44	15.63	15.41	15.32	14.35	14.1
Fe_2O_3*	2.77	2.27	2.27	2.01			2.53	9.04
FeO	6.28	6.76	4.13	4.27			6.76	
MnO	0.16	0.18	0.13	0.14	0.14	0.11	0.18	0.17
MgO	11.55	10.59	7.21	7.75	7.77	7.68	13.62	12.31
CaO	8.81	10.09	6.99	6.95	6.92	6.48	10.19	10.29
Na_2O	2.57	2.46	2.82	2.88	2.64	3.08	1.77	1.9
K_2O	1.27	1.35	2.21	2.35	2.42	1.80	0.25	0.63
P_2O_5	0.22	0.23	0.14	0.15	0.16	0.15	0.09	0.1
Total	99.15	97.15	97.78	99.72	98.71	98.42	98.82	98.28
FeO*/MgO	0.78	0.78	0.90	0.81	0.79	0.81	0.93	0.93
$^{87}Sr/^{86}Sr$	0.70439	0.70408	0.70490	0.70502	0.70491	0.70621		
ε_{Nd}	+2.1	+3.7	+1.2	+1.7	+1.5	−1.7		
$^{206}Pb/^{204}Pb$	18.314	18.298	18.369	18.361	18.367	18.394		

Data are from Shimoda et al. (1998).

GEOLOGICAL SETTING AND SAMPLES

The Philippine Sea plate is actively subducting beneath the Eurasian plate to form the arc-trench system of SW Japan (Fig. 1). Although the clearly defined seismicity and active volcanism in Kyushu does not continue as far as Honshu and Shikoku, the leading edge of the subducting slab is likely to have reached the upper mantle beneath the Quaternary volcanic front in SW Honshu (Fig. 1) (Nakanishi, 1980). The SVB, characterized by the occurrence of unusual HMAs, is located in the present forearc region. This is ~80 km south of the Quaternary volcanic front and extends east-westwards for about 600 km along the SW Japan arc. Five major volcanic regions exist (Fig. 1), namely Shitara, Osaka, NE and SW Shikoku, and E Kyushu. New K-Ar age data for Setouchi lavas (Tatsumi *et al.*, 2001) confirm an earlier suggestion (Tatsumi, 1983a) that the Setouchi magmatism took place within a short period of time, at 13.2 ± 0.4 Ma. This is almost synchronous with the timing of a 40–50° clockwise rotation of the arc sliver of SW Japan, and the major episode of rifting of the Japan Sea backarc basin at 16–14 Ma (Otofuji *et al.*, 1991). The southward drift of the SW Japan arc, caused by the clockwise rotation of the arc sliver, most likely resulted in the subduction of very young (15–27 Ma; Okino *et al.*, 1994; Tamaki *et al.*, 1999) oceanic crust of the Shikoku Basin beneath the Japan arc lithosphere (Tatsumi and Maruyama, 1989).

The samples studied here were collected from the central part of the SVB (Fig. 1). They include phenocryst-poor basalts to HMAs (sanukitoids, Tatsumi and Ishizaka, 1981). The HMAs are characterized by high MgO contents (ca. 7 %) and MgO/FeO* ratios (>1). High Mg contents may result from processes other than the accumulation of mafic phenocrysts such as olivine (Tatsumi and Ishizaka, 1982). The reasons are twofold. First, these rocks are not porphyritic and contain <10% phenocrysts of olivine and clinopyroxene. Second, the olivine phenocrysts have a limited Fo content of 88–90 and are in equilibrium with the bulk composition in terms of Mg-Fe partitioning. The primitive nature of HMAs has been confirmed by melting experiments of a HMA (SD261). The andesite melt coexists with olivine, clinopyroxene and orthopyroxene at 1.5 GPa and 1030°C and at 1.0 GPa and 1070°C in the presence of 20% (water-saturated) and 7% H_2O (water-undersaturated), respectively (Tatsumi, 1982). Presence of a point of multiple saturation with lherzolitic phases (olivine, clinopyroxene and orthopyroxene) strongly suggests that the HMA is produced by partial melting of mantle peridotite. The basalt sample (SDSYB) with a high MgO content (ca. 12%, Table 1) is also primitive and may represent the composition of mantle-derived primary magma, as inferred by both petrological and experimental studies (Tatsumi and Ishizaka, 1982; Tatsumi, 1982).

We also analyzed two Miocene basalt samples (RZ-5 and TM-0) from the NE Japan arc in order to compare the unusual Setouchi magmatism caused by subduction

Table 2. Re-Os isotopic compositions of Setouchi HMAs and basalts

	Re conc (ppt)	Os conc (ppt)	Measured $^{187}Os/^{188}Os$	Age-corrected $^{187}Os/^{188}Os$
Setouchi HMA				
SD261	126.4 ± 2.3	8.651 ± 0.056	0.2041 ± 0.0026	0.1875
SD407	45.7 ± 2.4	11.529 ± 0.054	0.1718 ± 0.0031	0.1673
SD411	29.2 ± 0.8	8.102 ± 0.053	0.2040 ± 0.0026	0.1999
JA-2	46.7 ± 1.6	11.4 ± 1.5	0.2092 ± 0.078	0.2046
Setouchi basalt				
SDSYB	39.0 ± 3.3	31.02 ± 0.20	0.1769 ± 0.0006	0.1755
SD424	73.8 ± 0.8	53.72 ± 0.58	0.1556 ± 0.0013	0.1540
NE Japan arc basalt				
Ryozen (RZ-5)	103.8 ± 1.2	148.3 ± 6.7	0.1370 ± 0.0011	0.1357
Tomari (TM-0)	96.2 ± 1.7	56.6 ± 0.7	0.1399 ± 0.0011	0.1378

Errors are $2\sigma_m$. Age correction for Os isotopic compositions were made using 13, 22, 15 Ma as ages of the Setouchi, Ryozen, and Tomari lavas, respectively. JA-2 data are the average of four analyses (Hattori et al., 2003).

of the hot and young lithosphere with the more typical NE Japan subduction zone magmatism. These basalts were selected for their unfractionated characteristics, high MgO, Ni, Cr contents (Table 1) and MgO/FeO* ratios. They are thus expected to be less affected by crustal contamination and may have Os isotopic compositions that reflect the source mantle.

ANALYTICAL METHODS AND RESULTS

The Re-Os analytical procedure used here has been described elsewhere (Suzuki and Tatsumi, 2001; Suzuki and Honda, 2003: Kato *et al.*, 2005) and utilizes a Carius tube digestion (Shirey and Walker, 1995) combined with bromine-extraction of Os (Birck *et al.*, 1997). Osmium and Re contents of the samples were determined by isotopic dilution method. Approximately 2.5 g of the finely pulverized rock powders were weighed, spiked with ^{190}Os and ^{185}Re and digested in inverse *aqua regia* in a sealed Carius tube at 220°C for at least 24 hours. After cooling, the tube was opened, ultrasonicated and the decomposed sample and solution were transferred into a 60-ml PFA vessel. Liquid bromine (3 ml), concentrated HNO_3 (3 ml) and 40% CrO_3 solution in 8N HNO_3 (4 ml) were added. The vessel was then placed on a hotplate and heated to about 60°C. Bromine boiling under the aqueous phase evaporated, scavenged volatile OsO_4 at the top of the vessel and flowed back to the bottom. The Os-bearing liquid bromine at the bottom was then pipetted into another container with 1 ml water. This bromine extraction step was then repeated. The water in the vial containing the pipetted bromine was discarded, and HBr (0.5 ml) was added to the liquid Br_2, which was then gently evaporated. The residue including Os was further purified by micro-distillation (Roy-Barman, 1993). Rhenium was separated from the residual solution using anion exchange chromatography.

All measurements in the present study were performed in negative ion detection mode (Volkening *et al.*, 1991, Creaser *et al.*, 1991) on a Finnigan® MAT 262 mass spectrometer equipped with a gas leak valve and an ion counting multiplier, at Kyoto University, Beppu. From the measured oxide ratios of Os and Re, atomic ratios of Os and Re were calculated after correcting for oxide interferences. Oxide corrections were made using $^{17}O/^{16}O$ = 0.00037 and $^{18}O/^{16}O$ = 0.002047 (Nier, 1950). Instrumental mass fractionation for Os was corrected against stable $^{192}Os/^{188}Os$ ratio of 3.0827. Total procedural blank was ~7 pg for Re, and ~2 pg for Os with $^{187}Os/^{188}Os$ of ~0.298. Precision of $^{187}Os/^{188}Os$ measurements, based on analysis of an in-house standard over a period of several months, was better than 0.4% (2 S.D.). Standard errors (2 S.D.) associated with the measured $^{187}Os/^{188}Os$ are given in Table 2.

Re-Os isotopic compositions of the Setouchi primitive HMAs and basalts as well as the Ryozen and Tomari basalts are presented in Table 2. The Os isotope ratios of the volcanic rocks are age-corrected to account for growth of radiogenic ^{187}Os since time of eruption. Osmium concentrations and isotopic compositions are highly variable in Setouchi HMAs, with Os concentration ranging from 8.1 to 11.9 ppt and age-corrected $^{187}Os/^{188}Os$ varying from 0.1718 to 0.2041. The Setouchi basalts have lower Os isotopic compositions (0.1556–0.1769) and higher Os concentration (31.0–53.7 ppt) than those in the HMAs. On the other hand, $^{187}Os/^{188}Os$ ratios of the basalts from the NE Japan arc are 0.1370 and 0.1399. These are considerably lower than those of Setouchi lavas and in the

Table 3. Results of mass-balance calculations

	SiO$_2$	Al$_2$O$_3$	FeO*	MgO	CaO	Na$_2$O	Fraction
granite							
basalt	51.1	15.9	9.1	12.0	9.2	2.7	68.5
granite	74.7	15.8	2.3	0.4	3.3	3.5	33.3
olivine	40.0	0.0	10.0	50.0	0.0	0.0	−1.7
clinopyroxene	52.5	5.0	3.0	15.5	23.0	1.0	−0.1
HMA	59.1	16.4	6.5	7.6	7.4	3.0	
calculated	59.1	16.2	6.8	7.5	7.4	3.0	$R^2 = 0.183$
gneiss							
basalt	51.1	15.9	9.1	12.0	9.2	2.7	39.9
gneiss	66.8	17.9	5.4	0.7	5.3	3.9	53.0
olivine	40.0	0.0	10.0	50.0	0.0	0.0	3.3
clinopyroxene	52.5	5.0	3.0	15.5	23.0	1.0	3.7
HMA	59.1	16.4	6.5	7.6	7.4	3.0	
calculated	59.1	16.0	6.9	7.4	7.4	3.2	$R^2 = 0.380$
gabbro							
basalt	51.1	15.9	9.1	12.0	9.2	2.7	−184.3
gabbro	55.9	18.7	8.9	4.1	8.9	3.5	234.7
olivine	40.0	0.0	10.0	50.0	0.0	0.0	34.2
clinopyroxene	52.5	5.0	3.0	15.5	23.0	1.0	15.4
HMA	59.1	16.4	6.5	7.6	7.4	3.0	
calculated	58.8	15.3	8.0	7.0	7.4	3.5	$R^2 = 4.05$
inferred lower crust							
basalt	51.1	15.9	9.1	12.0	9.2	2.7	−301.2
lower crust	54.3	17.2	8.7	7.4	9.8	2.7	371.5
olivine	40.0	0.0	10.0	50.0	0.0	0.0	33.3
clinopyroxene	52.5	5.0	3.0	15.5	23.0	1.0	−3.6
HMA	59.1	16.4	6.5	7.6	7.4	3.0	
calculated	59.1	15.9	8.2	7.3	7.7	1.9	$R^2 = 4.57$

Major elements data for granite, gneiss and gabbro are from Tatsumi et al. (2002); Data for average lower crust are from Rudnick and Fountain (1995); Data for basalt are from Shimoda et al. (1998).

range measured for arc and back-arc ultramafic xenoliths from Ichinomegata, Japan (Brandon *et al.*, 1996), Simcoe, USA (Brandon *et al.*, 1996, 1999), Lihir Island, Papua New Guinea (McInnes *et al.*, 1999) and Kamchatka (Widom *et al.*, 2003).

DISCUSSION

Shalow-level crustal assmilation

One possible mechanism for producing the ^{187}Os/^{188}Os variation observed for the Setouchi rocks is greater degrees of shallow-level crustal assimilation in HMAs than basalts, as crustal material generally possesses higher ^{187}Os/^{188}Os (e.g., upper crust; 1.0–1.4, Peucker-Ehrenbrink and Jahn, 2001; Hattori *et al.*, 2003). Ruiz *et al.* (1998) showed that crustal contamination can produce high ^{187}Os/^{188}Os ratios in arc magmas, on the basis of a positive correlation between Os isotope ratios and crustal thickness in the Chilean Southern Volcanic Zone. Lassiter and Luhr (2001) suggested that an inverse correlation of

^{187}Os/^{188}Os ratios with Os abundance in the Mexican arc lavas can also be explained by assimilation of lower crustal material, with ^{187}Os/^{188}Os ranging from 0.5 to 2.2 and Os concentrations between 20 and 160 ppt, during fractional crystallization. Hart *et al.* (2002) also interpreted the radiogenic Os isotope compositions of calc-alkaline volcanic rocks from the Lassen volcanic region of the Cascade arc to reflect significant interaction with lower crustal material.

The petrological characteristics of the Setouchi HMAs (Tatsumi, 1982; Tatsumi and Ishizaka, 1982; Tatsumi *et al.*, 2002), such as limited Mg# (= $100 \times$ Mg/(Mg + Fe)) and high NiO contents in olivine phenocrysts (88–90, 0.40 wt%, respectively), high Cr/(Cr + Al + Fe^{3+}) in chromite inclusions (~0.65) may imply lesser role of crustal contamination. However, it is still important to assess the likelihood of such a process using the geochemical data available.

In order to evaluate the effect of AFC processes, four crustal components were examined (Table 3). Those were

granite and gneiss, from basement rocks on Shodo-Shima Island (Tatsumi *et al*., 2002), representative of the upper crust. A gabbro composition from Shodo-Shima Island (Tatsumi *et al*., 2002) and an average lower crust composition (Rudnick and Fountain, 1995) were also used as representative of the lower crust. Because olivine and clinopyroxene are present as phenocryst phases in both the basalts and the HMAs, a simple least-square mixing calculation using the compositions of basalt, HMA, the crustal materials, and phenocryst phases may provide reasonable estimates for degrees of assimilation and crystallization. Although partial, rather than bulk assimilation may be the case during such a process, it is difficult to estimate the fraction of melting phases and also lack of fundamental data of Os for calculation such as partition coefficients into the rock constituting minerals and mobilities from the subducted material into fluid hinders further calculation based on partial assimilation. The present modeling thus assumes the assimilation of bulk crustal rocks. The results of mixing calculations (Table 4) indicate that an HMA magma composition can be reproduced from a parental basalt magma through crustal AFC processes by assimilating granitic upper crust with crystallizing olivine and clinopyroxene. It seems unlikely that only a small degree of crystallization of phenocrysts (<2 wt%) would be the consequence of a large degree of assimilation of granite (>30 wt%) does not seem likely. Further examination, however, based on trace element and isotopic compositions is obviously required. The modeling results with the other three crustal components are not appropriate for the following reasons: (1) a parental basalt magma requires olivine and clinopyroxene accumulation which is not observed in petrography of the HMA and (2) the sum of squares of residuals (R^2) obtained for the mixing calculations is far beyond the acceptable limit (~1.5; Mann, 1983).

The results of trace element and isotope model calculations for granite AFC are listed in Table 4. Potassium, Ba, Rb, Zr and isotopic compositions for Sr, Pb and Os yield results inconsistent with the data observed for Setouchi volcanic rocks. This suggests that the HMA magmas were not produced from the basalt magmas through crustal AFC processes. Alves *et al*. (2002) reported that Os initial isotope ratios are positively and linearly correlated in $^{187}Os/^{188}Os$ ratio vs. 1/[Os] space in arc lavas from 10 different subduction zones including Lesser Antilles, Java and Izu Bonin. They claimed that it is hard to reconcile a single shallow-level process of crustal assimilation as being responsible for these correlations because of the variability of the crustal components. Alternatively, they introduced variability of subducted materials that may account for the origin of the linear arrays found in arc level or volcano level variations. Our crustal AFC calculation results also suggest

Table 4. Results of mixing calculations

	basalt	granite	calculated	HMA
inconsistent				
K_2O (wt%)	1.20	2.95	1.78	2.25
Ba (ppm)	219	1052	497	195
Rb (ppm)	33	83	50	114
Zr (ppm)	99	176	125	80
Os (ppt)	31	30	31	8.65
Re (ppt)	39	200	93	126
$^{87}Sr/^{86}Sr$	0.7044	0.7079	0.7055	0.7049
$^{206}Pb/^{204}Pb$	18.32	18.54	18.42	18.37
$^{208}Pb/^{204}Pb$	38.47	38.83	38.64	38.56
$^{187}Os/^{186}Os$	0.1755	1.0000	0.6301	0.1875
consistent				
P_2O_5	0.27	0.07	0.20	0.17
Nb (ppm)	4	7	5	5
Ni (ppm)	207	0	138	148
Sr (ppm)	277	257	270	267
Nd (ppm)	14	23	17	16
Pb (ppm)	8	15	10	17
Th (ppm)	4	12	7	5
Y (ppm)	18	16	17	15
$^{143}Nd/^{144}Nd$	0.5127	0.5124	0.5126	0.5127
$^{207}Pb/^{204}Pb$	15.57	15.61	15.59	15.58

Data sources are from Tatsumi et al. (2002), Shimoda et al. (1998), and this study.

derivation of the isotopic variation is likely to be caused by mantle processes rather than crustal processes.

Slab-fluids or slab-melts?

The alternative possible cause of the elevated Os isotopic compositions of the Setouchi basalts and HMAs (Table 2) is an input of radiogenic Os from a slab-component. It is widely accepted that the chemistry of the mantle wedge overlying a subducted slab varies considerably compared to other portions of the upper mantle. This can be caused by metasomatic overprinting by subduction components. Elevated Os isotopic ratios relative to primitive upper mantle can be found in peridotite xenoliths from arc and back-arc environments (Brandon *et al*., 1996, 1999; McInnes *et al*., 1999; Widom *et al*., 2003), indicating that at least a part of the wedge mantle is metasomatized by slab-derived radiogenic Os. Identification of such subduction components (slab-fluid or slab-melt) has thus been a principal target of petrological and geochemical studies of subduction zone magmatism.

In order to evaluate the signature of the slab-derived component, mixing calculations between wedge mantle and hypothetical slab-melts and fluids have been conducted based on Os vs. Sr, Nd and Pb isotopes. Tatsumi and Hanyu (2003) presented sophisticated geochemical models to account for various processes; mixing between altered oceanic crust and sediments melts/fluids to pro-
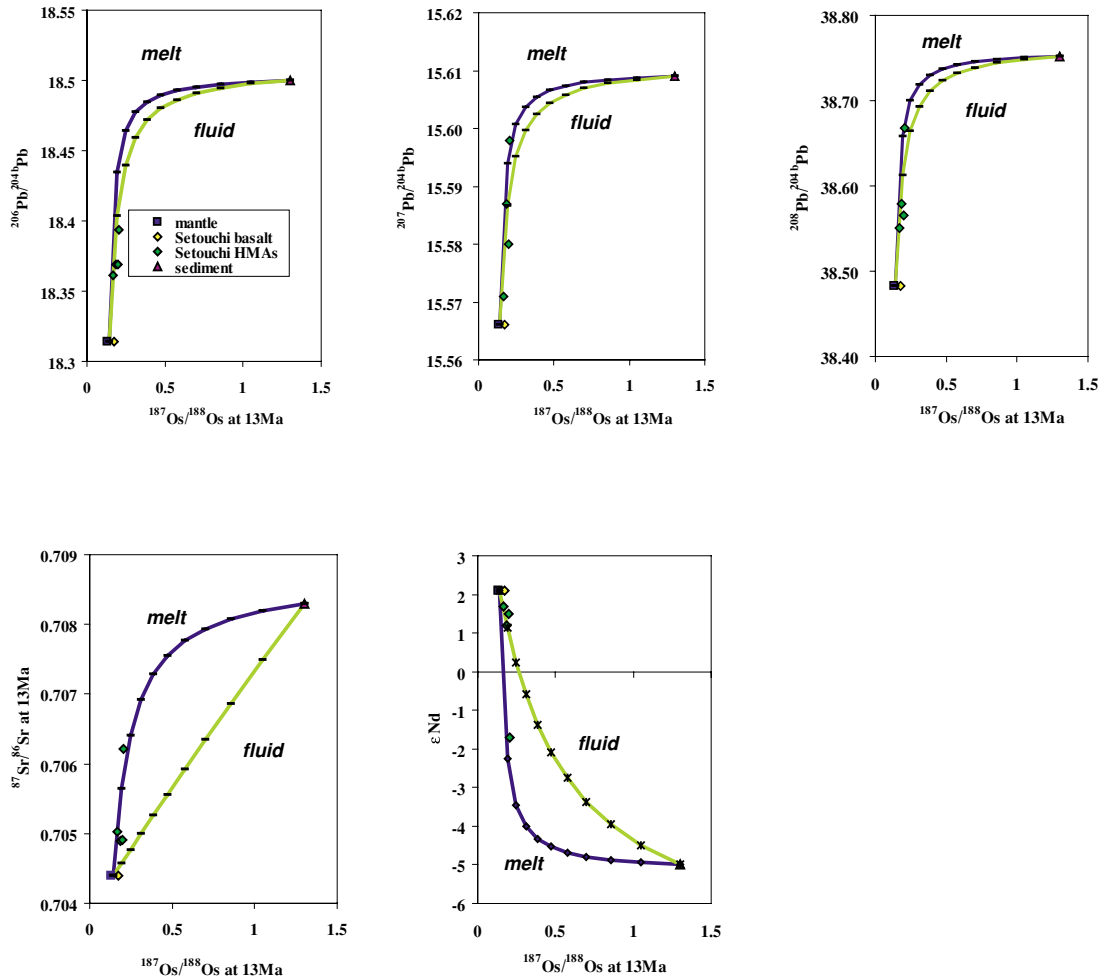
Fig. 2. Results of mixing calculations. For chemical compositions of the assumed end-members, see text. The Sr, Nd and Pb isotopic compositions of the inferred mantle used in the calculations are the data for the primitive Setouchi basalt, SDSYB (Table 1). The $^{87}Sr/^{86}Sr$, ε_{Nd} and $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ isotopic compositions of the sediment are 0.70829, –5.0, 18.500, 15.609 and 38.752, respectively. These are the data obtained for terrigenous sediment from the Nankai Trough (Fig. 1) (Shimoda et al., 1998). The isotopic compositions of the Setouchi HMAs can explained by the addition of several percentages of sediment melt to an original mantle, but not by addition of fluid.

duce the metasomatic agent, and interaction of this melts/ fluids and original wedge mantle. We, however, prefer to employ the simple mixing calculation used by Shimoda et al. (1998). This is because we are currently lacking sufficient fundamental data on Os behavior during various geological processes, such as its distribution coefficients during partial melting and dehydration processes. Our calculations employ the following assumptions proposed by Shimoda et al. (1998) for the chemical compositions of Sr, Nd and Pb end-members: (1) total melting of terrigenous sediments (Sr, Nd and Pb abundance of 180, 30, 20 ppm, respectively), (2) the chemical composition of the fluid was based on the experimental results of Aizawa et al. (1999) (Sr, Nd and Pb abundance of 18, 3, 10 ppm, respectively), and (3) the composition of the

inferred mantle wedge was calculated using chemical compositions of the primitive basalt, SYSYB, by assuming appropriate partition coefficients (Green, 1994), and 15% batch partial melting of lherzolite with 55% olivine, 30% orthopyroxene and 15% clinopyroxene (Shimoda et al., 1998). We assume that the mantle possesses a $^{187}Os/$ ^{188}Os of 0.14 and an Os concentration of 2.5 ppb, and that slab-derived components, both fluid and melt have similar Os isotopic compositions of average upper continental crust ($^{187}Os/^{188}Os = 1.3$; Peucker-Ehrenbrink and Jahn, 2001; Hattori et al., 2003). For the Os contents of both melt and fluid from sediment, we used a ratio of Os concentration between mantle and slab-components of 2.5:1, following the assumptions of Brandon et al. (1996, 1999). The subducted oceanic crust is likely to contrib-

ute little to the melt or fluid generation. One of the reasons is that the Os concentration of MORB (<5 ppt, e.g., Escrig et al., 2005) is much lower than the typical marine sediments (30–1000 ppt, e.g., Peucker-Ehrenbrink et al., 1995: Cohen et al., 1999; Dalai et al., 2005). The other reason is that the subducted Shikoku Basin is too young (15–27 Ma; Okino et al., 1994; Tamaki et al., 1999) to have the elevated Os isotopic composition. The age of the Shikoku Basin lithosphere was 2–14 Ma when the SVB was formed at 13 Ma. The oceanic crust with typical $^{187}Re/^{188}Os$ ratio of 1000 (e.g., Escrig et al., 2005) has $^{187}Os/^{188}Os$ of 0.16–0.36 at 2–14 Ma. The oceanic crust with such low $^{187}Os/^{188}Os$ and low Os concentration is less likely to play an important role in producing the Os concentration and isotopic compositions of the Setouchi HMAs.

The results of the mixing calculations are shown in Fig. 2. The addition of several percent of a sediment-derived melt reproduces the Os-Sr-Nd-Pb isotope compositions of the HMA magma source. Despite the obvious success in reproducing the Os-Pb isotope relationship, the sediment-derived fluid is an unlikely end-member, as mixing between mantle and sediment-derived fluid cannot account for their Os vs. Sr or Nd isotope data. Therefore, a slab-derived melt may have played a major role in the Os-Sr-Nd-Pb characteristics of Setouchi HMAs.

We assumed the assimilated mantle possesses a $^{187}Os/^{188}Os$ ratio of 0.14, which is higher than that of hypothetical primitive upper mantle (0.1296, Meisel et al., 2001), but is in the range of peridotite xenoliths from arc and back-arc settings. Though Chesley et al. (2004) suggested metasomatism of the upper mantle by slab-derived material does not elevate Os isotopic compositions of large-scale mantle, their calculation does not necessarily rule out the likely elevation of wedge mantle Os isotopic compositions on a localized scale. Lassiter and Luhr (2001) proposed that Os isotopic and abundance signatures of Mexican volcanic lavas were caused by upper crustal assimilation, assuming a $^{187}Os/^{188}Os$ ratio of 0.135 for the initial mantle melt. This Os isotopic ratio is still higher than that of typical upper mantle, reflecting the input of slab-derived materials (Lassiter and Luhr, 2001). Meisel et al. (2001) also mentioned the possible contamination of mantle wedge by a crustal component resulting in higher $^{187}Os/^{188}Os$ ratios in the mantle wedge than in typical upper mantle. Moreover, it is also possible that mantle lithosphere beneath SW Japan has been affected by a slab component, since the arc has been subjected to subduction magmatism since the Cretaceous (e.g., Kagami et al., 1992). We have little knowledge about the mantle lithosphere beneath the arc; however, Os isotope ratios reported in N-NE China and NE Japan (Ichinomegata) range from 0.117 to 0.133 (Wu et al., 2003; Gao et al., 2002; Brandon et al., 1996). Although petrographically

the most primitive basalt, SDSYB (c.f., Table 1, 2, Tatsumi, 1982; Tatsumi and Ishizaka, 1982) should possess Os isotopic compositions similar to that of primitive upper mantle (~0.13), this basalt yields a high $^{187}Os/^{188}Os$ ratio of 0.175. This result indicates the primitive basalt SDSYB also contains slab-derived components, as suggested by Shimoda et al. (1998) and Tatsumi (2001).

The Os isotope ratios of the primitive basalts from NE Japan (0.136–0.138) are within the range of Os isotopic compositions of arc peridotites (0.120–0.156, Brandon et al., 1996, 1999; McInnes et al., 1999; Becker et al., 2001; Widom et al., 2003) and are much lower than the Setouchi HMAs and basalts (0.167–0.204). Though we cannot reach a conclusive interpretation because of the severely limited number of Os isotopic data sets, the difference in the Os isotope ratios may reflect differences in tectonic settings. Widom et al. (2003) reported the less radiogenic $^{187}Os/^{188}Os$ ratios of harzburgites from Avachinsky which is associated with relatively cold, old (~100–175 Ma) subducting slabs. Relatively more radiogenic Os are from peridotite xenoliths from the Kamchatka volcanoes associated with hot and young (~25 Ma) slab. The latter case is characterized by adakitic slab melts. They suggested multistage metasomatism resulting from the interaction of the mantle wedge with a variety of slab-derived fluids and melts. They suggest the presence of hydrous slab-fluid metasomatism associated with cold and old oceanic crust and silicic slab-melt metasomatism associated with subduction of young oceanic crust. Our SBV and NE Japan results compare well with the result of Widom et al. (2003). The SVB volcanism involves the subduction of young (2–14 Ma) slab and the volcanic rocks yield relatively radiogenic Os isotopic compositions, which is likely to be caused by contemporaneous slab-derived melts. On the other hand, the basalts from NE Japan with less radiogenic Os isotopic compositions are possibly associated with slab-derived fluid.

CONCLUSIONS

Primitive Setouchi high-Mg andesites and basalts possess elevated Os isotopic signatures. Geochemical modeling suggests that the HMAs are not produced by AFC processes within the crust, but are mantle-derived. Osmium vs. Sr, Nd and Pb isotopic trends can be explained by melting of subducting sediments and subsequent melt-mantle interaction. In contrast, the Os isotopic ratios of primitive basalts from NE Japan, yielding lower Os isotopic ratios, are within the range of arc peridotites, and may reflect a different mechanism of Os transport (slab-derived fluids?) in this region where an older, cooler slab is being subducted.

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