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The nature of metasomatism in the sub-arc mantle wedge: evidence from Re–Os isotopes in Kamchatka peridotite xenoliths

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Abstract

We have performed Re–Os isotope measurements on a suite of 21 Kamchatka mantle xenoliths including 19 harzburgites and two lherzolites, from the northern arc front (Valovayam Volcano), the southern arc front (Avachinsky Volcano), and behind the arc front in the south (Bakening Volcano). Os and Re concentrations vary from 0.02 to 8.2 and 0.003 to 0.437 ppb, respectively, and ^{187}Re/^{188}Os varies from 0.004 to 3.811. ^{187}Os/^{188}Os ratios range from 0.1226 to 0.1566. Regional variations in Re–Os isotope signatures are apparent, with peridotites from Avachinsky exhibiting the least radiogenic Os isotope signatures and lowest Re/Os ratios, and those from Bakening the most radiogenic Os and highest Re/Os. Peridotites from Valovayam span the distinct compositional fields defined by the Avachinsky and Bakening peridotites. All of the Kamchatka peridotites are, however, characterized by radiogenic ^{187}Os/^{188}Os compared to non-arc continental peridotites with comparable Re abundances or Re/Os ratios.

The relatively radiogenic Os isotope signatures in the Kamchatka peridotites cannot easily be explained by contamination of the xenoliths by their host lavas, as this process would result in Re/Os ratios higher than observed in the xenoliths. In situ radiogenic ingrowth of high Re/Os mantle followed by recent Re depletion also cannot explain the observed radiogenic Os signatures in the Kamchatka peridotites, as the time required for radiogenic ingrowth would be significantly greater than the age of the lithospheric terranes that make up the respective regions of Kamchatka. The radiogenic Os isotope signatures in the Kamchatka peridotites are instead attributed to metasomatism of the Kamchatka sub-arc mantle wedge by radiogenic slab-derived fluids and melts. The regional variations in Re–Os isotope signatures are consistent with previous petrographic and geochemical studies of the Kamchatka mantle xenoliths that reveal multistage metasomatic histories resulting from interaction of the mantle wedge with a variety of slab-derived fluids and melts, including silicic slab-melt metasomatism associated with subduction of relatively hot, young (~ 15–25 Ma) oceanic crust in the northern arc front, hydrous slab-fluid metasomatism associated with subduction of colder, old (~ 100 Ma) oceanic crust in the southern arc front, and carbonate-rich slab-melt metasomatism in the southern segment behind the arc front, where the slab is deeper.

Positive correlations between ^{187}Os/^{188}Os, La/Sm, and Ru/Ir in Avachinsky harzburgites support a model in which high f_{O_2}, Cl-rich, hydrous slab fluids transport LREE, Ru, and radiogenic Os into the mantle wedge beneath the southern arc front. Re is either not transported, or is not retained in the mantle during fluid–mantle interaction. Relatively higher Re and more radiogenic Os (but low Os abundances) in the Valovayam and Bakening peridotites indicate that both scavenging of mantle Os as well as exchange with radiogenic slab-derived Os, and incorporation of Re, occurs during interaction of the mantle wedge with oxidized, adakitic, and carbonate-rich slab melts. Similar ranges of Re–Os isotope signatures in peridotites from

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Avachinsky, Japan and Lihir, and from Valovayam and the Cascades, respectively, suggest that the age (temperature) and depth of subducting oceanic crust influences the Re–Os composition of metasomatized sub-arc mantle.

Keywords: Kamchatka; Mantle; Subduction; Osmium; Isotope

1. Introduction

Convergent boundaries are the sites of significant mass exchange between the Earth's surface and interior, and play an important role in continental crust formation and the depletion and enrichment history of the Earth's mantle. Geochemical fluxes from the slab to the shallow mantle wedge during subduction control the composition of arc volcanism and newly generated continental crust, as well as the degree to which crustal material may be recycled into the Earth's deep mantle. Crustal material that escapes fluxing into the mantle wedge overlying subducting slabs may be an important component of geochemically enriched mantle plumes, which may ultimately resurface in the form of hotspot volcanism. The most direct method for investigating chemical fluxes at subduction zones is through petrographic and geochemical studies of mantle xenoliths from arc wedges. Important questions that have been relatively little explored, due largely to the paucity of mantle xenoliths in arc settings, include the nature and diversity of slab-derived metasomatic fluids and the relationship to age, temperature, and depth of the subducting slab. In addition, relatively little is known about the mechanisms of fluid interaction with the mantle and the geochemical behavior during subduction of some element groups such as the highly siderophile elements (HSE), including the platinum group elements (PGE).

The potential mobility of HSE in subduction zone environments has important implications regarding the siderophile element budget of the mantle. Understanding the mantle siderophile element budget is critical for assessing processes of accretion and core formation (e.g. Walter et al., 2000 and references therein), as well as evaluating the role of crustal recycling vs. core–mantle interaction in producing radiogenic Os isotope signatures in mantle plumes (Widom and Shirey, 1996; Brandon et al., 1998; Walker et al., 1999). In addition, the potential mobility of HSE in subduction zone environments has important implications regarding the formation of economic PGE ore deposits such as the major epithermal gold deposits associated with some volcanic arcs (McInnes et al., 1999). Recent studies of some metasomatized orogenic peridotites suggest that significant fractionations of PGE can occur during fluid–mantle interactions (Gueddari et al., 1996; Garuti et al., 1997a). However, other recent studies of continental peridotite xenoliths indicate that PGE are relatively immobile in both hydrous and carbonate-rich metasomatic fluids (e.g. Handler et al., 1997; Handler and Bennett, 1999; Lorand and Alard, 2001).

Re–Os isotope studies of mantle xenoliths from arc settings provide the opportunity to directly document the behavior of these HSE during slab fluid-induced metasomatism of the mantle wedge. Relatively radiogenic Os isotope signatures in mantle xenoliths from arc settings, including the Cascades, Canadian Cordillera, Japan, and Lihir, Papua New Guinea, have recently been documented, and attributed to the mobility of Os in slab fluids (Brandon et al., 1996; McInnes et al., 1999; Peslier et al., 2000). In this paper, we report on a study of Re–Os isotopes in mantle xenoliths from the Kamchatka arc, including metasomatic harzburgites and lherzolites. The geologic setting of Kamchatka, in which young (15–25 Ma), hot oceanic crust was subducted in the north and relatively old (100 Ma), cold oceanic crust is subducted in the south, offers a unique opportunity to investigate the range of metasomatic styles that result from slab dewatering and slab melting under different P–T regimes. The xenolith suite on which we report Re–Os data includes samples from both the northern and southern segments of the arc, as well as samples from the arc front and behind the arc front, thus providing a three-dimensional view of the metasomatized mantle wedge.

2. Geologic setting of the Kamchatka arc

The Kamchatka arc is located at the junction of the North American, Eurasian, and Pacific lithospheric plates, and records a protracted history of subduction, terrane accretion, and arc volcanism. The arc can be divided into northern and southern segments separated by crustal shear zones (Fig. 1). The southern segment is built upon Mesozoic–Tertiary accreted terranes and lower crustal high-grade metamorphic rocks associated with the northwestward subduction of cold Mesozoic Pacific lithosphere (Kepezhinskas et al., 1997).

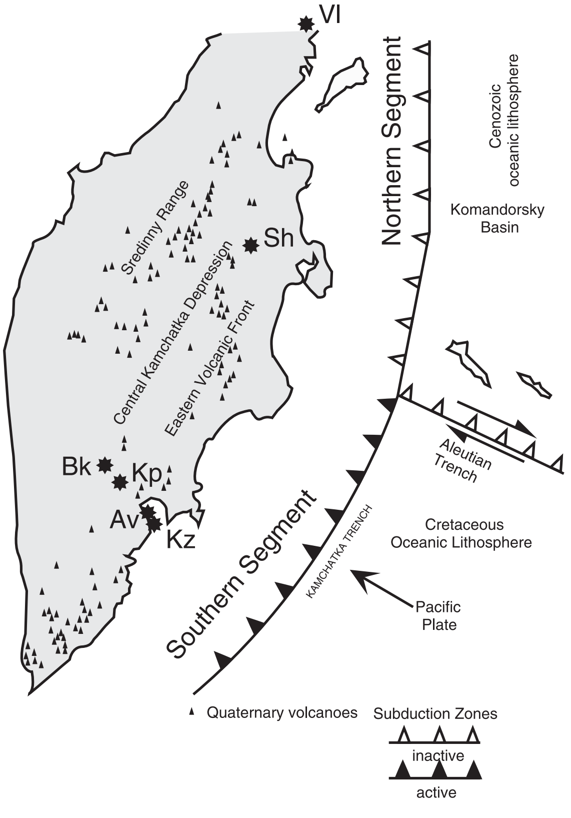


Fig. 1. Tectonic setting of Kamchatka arc, showing locations of volcanoes associated with mantle xenolith occurrences, and the positions of active and fossil subduction zones and trenches. Ages for the Komandorsky Basin crust and Pacific lithosphere are adopted from Baranov et al. (1991). The boundary between the northern and southern segments is manifested by major transcurrent faults and crustal discontinuities and is adopted from Hochstaedter et al. (1994). Volcano locations include Kozelsky (Kz), Avachinsky (Av), Kupol (Kp), Bakening (Bk), Shiveluch (Sh), and Valovayam (Vl). After Kepezhinskas et al. (1997).

The northern segment was formed in response to the westward subduction of young (15–25 Ma), hot oceanic crust of the Komandorsky Basin, which is believed to have formed as a result of back-transform spreading (Baranov et al., 1991; Hochstaedter et al., 1994). The northern segment is underlain by thickened (up to 18 km) oceanic crust (Kepezhinskas et al., 1993).

Ultramafic xenoliths of mantle origin have been found at six volcanic centers both across and along the arc (Fig. 1), including Kozelsky, Avachinsky, Kupol, Bakening, Shiveluch, and Valovayam. Detailed petrographic and geochemical studies of these xenolith suites reveal multistage metasomatic histories, which are interpreted to result from interaction of various fluids and melts liberated from the downgoing slab with the overlying mantle wedge beneath Kamchatka (Kepezhinskas et al., 1995, 1996; Kepezhinskas and Defant, 1996a,b).

3. Ultramafic xenolith suites in Kamchatka

Ultramafic xenoliths from Kamchatka exhibit porphyroclastic and protoclastic fabrics due to variable deformation under mantle conditions. Abundant creep-related deformational features, along with mineral defect structures (e.g. undulose olivine and kink-banded pyroxene), are consistent with their derivation from the mantle wedge beneath the Kamchatka arc (Kepezhinskas et al., 1995; Turner et al., 1996). Two major compositional groups are identified among Kamchatka ultramafic xenoliths: Cr-rich harzburgites, dunites, and rare lherzolites (group 1); and Cr-poor pyroxenites, wehrlites, and websterites (group 2). The first group exhibits variable extents of cryptic and modal metasomatism and is interpreted as variably metasomatized mantle wedge peridotite. The second group is thought to be formed as a result of extensive reaction of magmatic veins and dikes with mantle wedge peridotite under mantle conditions (Kepezhinskas et al., 1996). Pressure and temperature estimates for Kamchatka xenoliths based on mineral and fluid inclusion thermobarometry indicate their equilibration at 27–30 kbars (~ 100 km depth below the arc) with a temperature range of 830–1130 °C (Kepezhinskas et al., 2002). The Kamchatka mantle geotherm appears to be similar to the mean oceanic geotherm at 60.4 Ma (Turcotte and Schubert, 1982) and is lower than elevated Cenozoic geotherms in Eastern Australia (Sutherland et al., 1994) and Southwest Japan (Umino and Yoshizawa, 1996). These data are consistent with derivation of Kamchatka ultramafic xenoliths from relatively deep portions of thermally unperturbed mantle wedge beneath this convergent margin.

Cr-rich peridotite xenoliths (group 1) are systematically more depleted than the MORB mantle in both bulk chemical and mineral compositions. Modal mineralogy (low modal clinopyroxene content) and refractory mineral chemistry (high Cr/[Cr + Al] ratios of spinels and high Mg# of olivines and orthopyroxenes) indicate an overall depletion in basaltic components (Fig. 2). Relic (pre-metasomatic) clinopyroxenes, which occur as primary-textured porphyroclasts associated with deformed olivine and orthopyroxene, have low Ti, Na, Al, and high Mg and Cr concentrations coupled with rather uniform light rare earth element (LREE) depletions and negative high-field strength element (HFSE, e.g. Ti, Zr) anomalies on chondrite-normalized graphs. These compositions are similar to those of clinopyroxenes from depleted (MORB-type) residual mantle (Kepezhinskas and Defant, 1996a). Primary clinopyroxene geochemistry suggests that the mantle wedge was depleted with respect to HFSE and LREE throughout the 1000 km length of the Kamchatka arc; superimposed enrichment signatures in Kamchatka peridotite xenoliths have been attributed to subduction-related metasomatism (Kepezhinskas et al., 1995, 1996; Kepezhinskas and Defant, 1996a).

3.1. Evidence for mantle wedge metasomatism

Peridotite xenoliths from the northern segment of the Kamchatka arc contain abundant textural and chemical signatures of fluid-induced mantle metasomatism, including three textural groups of clinopyroxenes that exhibit progressive enrichment in Na and LREE. In addition, the occurrence in these xenoliths of felsic (dacitic) metasomatic veins that are compositionally similar to experimental melts derived via partial melting of metabasalt at 15–32 kbars (Fig. 3a), suggests the involvement of slab melts during metasomatism of the northern Kamchatka sub-arc mantle (Defant and Drummond, 1990; Kepezhinskas et al., 1996).

Harzburgites from the southern segment of the Kamchatka arc front display slight, but detectable LREE enrichments coupled with negative Th and Ta, and positive Ba and U anomalies on chondrite-

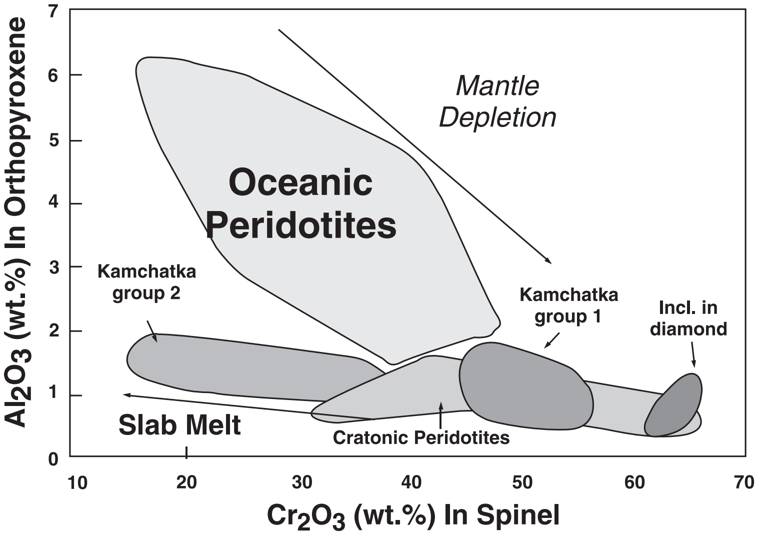


Fig. 2. Al_2O_3 in orthopyroxene vs. Cr_2O_3 in co-existing spinel for Kamchatka mantle xenoliths. Low Al_2O_3 and high Cr_2O_3 in group 1 peridotites reflect high degrees of melt depletion in the Kamchatka mantle wedge relative to oceanic peridotites (Bonatti and Michael, 1989), comparable to those recorded by diamond inclusions (Gurney and Zweistra, 1995) and cratonic peridotites (Rudnick et al., 1994). Higher Cr_2O_3 in group 2 peridotites and pyroxenites reflect subsequent slab melt–mantle reactions. After Kepezhinskas and Defant (1997).

normalized plots (Fig. 3b), consistent with metasomatism by hydrous fluids (Kepezhinskas and Defant, 1996a). Since residual mineral phases (relic, pre-metasomatic clinopyroxenes) in these xenoliths do not display any chemical enrichments, this subduction component likely resides along the grain interfaces and in fluid inclusions.

Peridotite xenoliths from Bakening Volcano (southern segment, behind the arc front) contain melt pockets with apatite, amphibole, and phlogopite, providing evidence for an additional metasomatic process. Clinopyroxenes associated with these glass pockets exhibit enrichments in Cr, Mg, Al, Ti, and Na and diverge from the typical metasomatic trends caused by silicate melts. Ion probe analyses of glasses reveal high Sr, Nb, Zr, and REE concentrations coupled with high La/Yb, Zr/Hf, Sr/Sm, and Nb/La, and low Ti/Eu ratios (Fig. 3c) similar to carbonate-rich melts (Rudnick et al., 1993). These glasses either directly represent carbonate-rich melt introduced into the lithospheric mantle or have originated through decomposition of primary mantle carbonates during xenolith transport. Elevated Na contents of clinopyroxenes and amphiboles suggest that the metasomatizing melt was more similar to a sodic carbonate-rich liquid rather than to a typical low-Na calcitic or dolomitic carbonatite.

The distinct styles of metasomatism documented in Kamchatka peridotite xenoliths have been related to the *P*–*T* conditions of the subducting slabs associated with the northern and southern segments of the Kamchatka arc (Kepezhinskas and Defant, 1996a). Fig. 4 illustrates the inferred relationship between type of metasomatic fluids/melts and the age and depth of the subducted oceanic crust. Silicic slab-melt metasomatism is associated with subduction of relatively hot, young (~ 15–25 Ma) oceanic crust in the northern arc front. Hydrous slab-fluid metasomatism is associated with subduction of colder, old (~ 100 Ma) oceanic crust in the southern arc front; in contrast, carbonate-rich slab-melt metasomatism occurs in the southern segment behind the

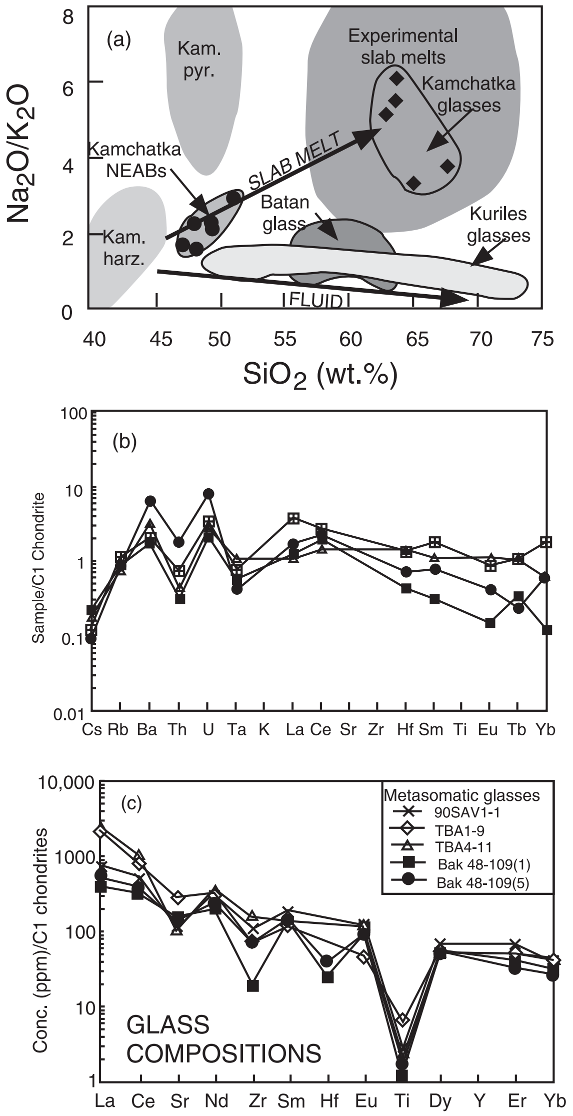


Fig. 3. (a) Na_2O/K_2O vs. SiO_2 for Kamchatka mantle xenoliths and associated glasses (Kepezhinskas et al., 1996), as well as glasses from mantle xenoliths from Batan (Schiano et al., 1995) and Kuriles (Volynets et al., 1990). Experimental slab melts (Rapp and Watson, 1995) and Kamchatka Nb-enriched arc basalts (NEABs; Kepezhinskas et al., 1996) are shown for comparison. After Kepezhinskas and Defant (1997). (b) Chondrite-normalized trace element diagram showing LREE enrichment and positive Ba and U anomalies in the Kamchatka southern arc front (Avachinsky) harzburgites, attributed to metasomatism by hydrous slab fluids. Chondrite-normalizing values from Sun and McDonough (1989). After Kepezhinskas and Defant (1996a). (c) Chondrite-normalized trace element patterns for xenolith glasses from behind the arc front in southern Kamchatka (Bakening volcano) compared to metasomatic glasses from the Samoa–Macdonald hot spot xenoliths. The latter are believed to represent carbonatite metasomatism in the oceanic upper mantle (Hauri et al., 1993). Chondrite-normalizing values are from Anders and Grevesse (1989). After Kepezhinskas and Defant (1996a).

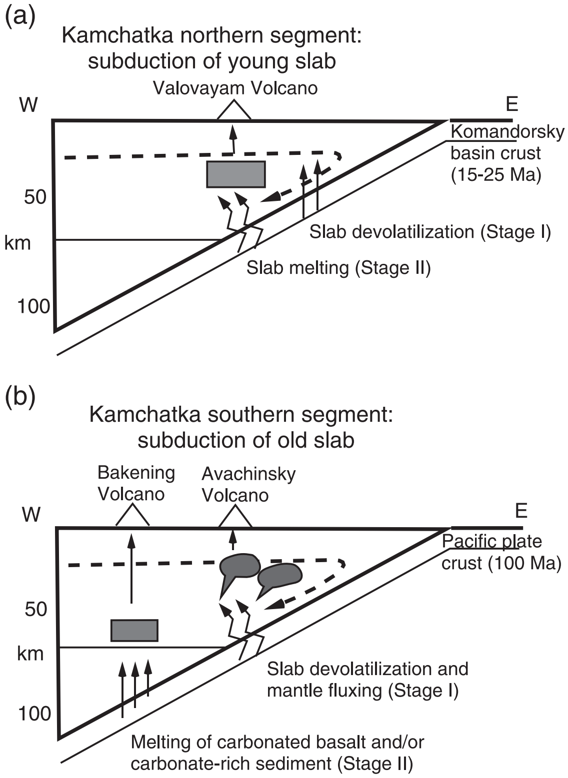


Fig. 4. A schematic diagram illustrating the relationship between styles of mantle metasomatism and age and depth of the subducting slabs in (a) the northern segment and (b) the southern segment of the Kamchatka arc. Silicic (adakitic) slab-melt metasomatism is associated with subduction of relatively hot, young (~ 15–25 Ma) oceanic crust in the northern arc front; and hydrous slab-fluid metasomatism is associated with subduction of colder, old (~ 100 Ma) oceanic crust in the southern arc front; in contrast, carbonate-rich slab-melt metasomatism occurs in the southern segment behind the arc front, where the slab is deeper. After Kepezhinskas and Defant (1996a).

arc front, where the slab is deeper (Kepezhinskas and Defant, 1996a).

4. Analytical techniques

We have analyzed a suite of 21 Kamchatka mantle peridotite xenoliths, including 19 harzburgites and two lherzolites, for Re and Os isotopes. The xenoliths are associated with volcanoes from the Kamchatka northern arc front (Valovayam Volcano), southern arc front (Avachinsky Volcano), and behind the arc front in the south (Bakening Volcano).

The xenolith samples were digested using the Carius tube method (Shirey and Walker, 1995), in which 1–2 g of sample powder were dissolved in concentrated inverse aqua regia in a sealed glass Carius tube by heating in an oven for 24–48 h at 240 °C. Following sample digestion, Re and Os were separated by an aqua regia–CCl_4 solvent extraction technique (Cohen and Waters, 1996), in which Os was extracted into the CCl_4 and Re remained in the aqua regia. Os was then back-extracted into HBr, dried down, and further purified by a microdistillation procedure (Roy-Barman, 1993; Roy-Barman and Allegre, 1995). Re was further purified using AG1-X8 anion exchange column chemistry similar to the methods described by Pearson et al. (1995a). Total processing blanks were approximately 2 pg for both Os and Re. The measured blank composition averaged $^{187}Os/^{188}Os$ of 0.170.

Re and Os isotopes were measured at the Department of Magnetism (DTM), Carnegie Institution of Washington. Measurements were made as the negative oxides ReO_4^- and OsO_3^- following existing techniques (Creaser et al., 1991; Volkening et al., 1991). The procedures for measuring Re and Os at DTM have been described in detail by Pearson et al. (1995a). In-run precision ranged from 0.07% to 1.6% 2σ for $^{187}Os/^{188}Os$ measurements, but were generally better than 0.5% 2σ. Precision for $^{187}Re/^{185}Re$ measurements were all better than 0.5% 2σ.

5. Results

Re–Os isotope data for the Kamchatka peridotites are reported in Table 1. The peridotite xenoliths exhibit extreme ranges in both Os and Re concentrations, with Os varying from 0.02 to 8.2 ppb, and Re from 0.003 to 1.036 ppb. Re/Os ratios exhibit a similarly large range, with $^{187}Re/^{188}Os$ varying from 0.004 to 187.9. One of the two lherzolite xenoliths marks the low end of the peridotite range in Os concentration, but the two lherzolites fall within the range of the 19 harzburgites in terms of Re abundances and Re/Os ratios.

$^{187}Os/^{188}Os$ ratios range from 0.1226 to 0.1566 in the harzburgites, and 0.1410–0.1498 in the lherzolites. No correlation of $^{187}Os/^{188}Os$ with Re abundances is observed in the peridotites, but the peridotites with the lowest Os abundances exhibit the most

Table 1
Kamchatka harzburgite and lherzolite Re–Os isotope data

Harzburgites		Os, ppb	$^{187}Os/^{188}Os$	2σ error	Re, ppb	$^{187}Re/^{188}Os$
Avachinsky	AVX8	1.033	0.1297	0.0021	0.014	0.067
	AVX14	2.013	0.1226	0.0004	0.003	0.006
	AVX33	3.960	0.1278	0.0003	0.014	0.018
	AVX33-1	2.390	0.1267	0.0002	0.012	0.024
	AVX34	1.133	0.1252	0.0004	0.137	0.582
	AVX45	8.205	0.1282	0.0001	0.007	0.004
	AVX48	2.020	0.1275	0.0002	0.012	0.028
	AVX49	1.773	0.1299	0.0001	0.003	0.009
	AVX50	4.845	0.1292	0.0002	0.009	0.009
	AVX51	2.678	0.1282	0.0001	0.015	0.027
	AVX53	0.869	0.1269	0.0001	0.006	0.036
Valovayam	8710/f	0.370	0.1428	0.0002	0.162	2.111
	Val32/15	1.302	0.1255	0.0001	0.014	0.053
Bakening	48-X	0.027	0.1566	0.0014	1.036	187.9
	48-31	1.917	0.1352	0.0001	0.149	0.374
	48-56	0.282	0.1444	0.0002	0.035	0.595
	48-93	0.558	0.1425	0.0001	0.032	0.274
	48-500	0.554	0.1409	0.0003	0.437	3.811
	48-504	0.171	0.1409	0.0003	0.026	0.741
Lherzolites						
Valovayam	8710-P	0.196	0.1410	0.0008	0.027	0.654
Bakening	BAK48-47	0.023	0.1498	0.0003	0.017	3.717
Host Lavas						
Valovayam	Val8710	0.116	0.1388	0.0004	0.203	8.491
	Val55	0.006	0.1679	0.0010	0.314	236.6
Bakening	BAK 48	0.360	0.1378	0.0007	0.074	0.997

radiogenic Os isotope signatures. As a group, the peridotites exhibit a broad positive correlation of $^{187}Os/^{188}Os$ with $^{187}Re/^{188}Os$. In detail, this correlation is dominated by the samples from Bakening and Valovayam, which are generally more radiogenic than the Avachinsky peridotites. No correlation of $^{187}Os/^{188}Os$ with $^{187}Re/^{188}Os$ is apparent within the Avachinsky peridotite suite. Although only two lherzolites were analyzed, there is no clear compositional distinction between the lherzolites and harzburgites in terms of their Re, Os, and $^{187}Os/^{188}Os$ characteristics.

6. Discussion

6.1. Re–Os systematics in Kamchatka peridotites

Kamchatka peridotite Re and Os concentrations span significant ranges that overlap and extend to lower values than most continental and oceanic mantle (Fig. 5a). The Avachinsky peridotites have Os concentrations generally in excess of 1 ppb and are comparable in Os concentrations to most subcontinental lithospheric mantle (SCLM). In contrast, the Avachinsky Re concentrations are generally significantly lower than SCLM. Low Re contents in mantle peridotites are indicative of melt depletion since Re is moderately incompatible during mantle melting (e.g. Morgan, 1986; Burnham et al., 1998; Shirey and Walker, 1998 and references therein), hence, the very low Re contents in Avachinsky mantle are consistent with the strong melt depletion that was recognized for the Kamchatka sub-arc mantle based on the low Al_2O_3 and high Cr_2O_3 contents in group 1 peridotite orthopyroxenes and spinels, respectively (Fig. 2).

In contrast to the Avachinsky peridotites, the peridotites from Valovayam and Bakening volcanoes tend towards lower Os and generally higher Re abundances. These samples overlap only minimally in Os concentration with SCLM from non-arc environments, but have comparable Re abundances (Fig. 5a). The relatively low Os concentrations in the Valovayam and Bakening peridotites compared to typical SCLM are not unusual in arc environments; both the Re and Os concentrations of the Valovayam and Bakening peridotites are similar to peridotites from the Cascades and Japan (Fig. 5b; Brandon et al., 1996, 1999). Low Os abundances are, however, not a characteristic of all arc mantle, as harzburgites from Avachinsky volcano as well as those from Lihir, Papua New Guinea (Fig. 5b; McInnes et al., 1999) do not exhibit anomalously low Os concentrations. Low Os concentrations in arc mantle peridotites relative to non-arc SCLM have previously been attributed to mobility of Os in the presence of oxidizing and Cl-rich slab fluids (Brandon et al., 1996), and might also explain the relatively low Os abundances in some of the Kamchatka peridotites. This is discussed in more detail in Section 6.3.

The $^{187}Os/^{188}Os$ signatures of the Kamchatka peridotites range from 0.1226 to 0.1566. For comparison, the range in $^{187}Os/^{188}Os$ for minimally altered oceanic abyssal peridotites is more limited, varying from 0.1221 to 0.1276 (Snow and Reisberg, 1995; Brandon et al., 2000). In contrast to the relatively limited range of Os isotope signatures of MORB mantle, $^{187}Os/^{188}Os$ signatures in peridotites from non-arc continental mantle are more variable, generally ranging from ~ 0.104 to 0.130 (Walker et al., 1989; Carlson and Irving, 1994; Pearson et al., 1995b; Reisberg and Lorand, 1995; Shirey and Walker, 1998; Meisel et al., 2001), although extremely rare occurrences of continental mantle xenoliths with $^{187}Os/^{188}Os$ signatures as high as 0.338 are known (Pearson et al., 1995a).

The Kamchatka peridotites extend to $^{187}Os/^{188}Os$ signatures significantly more radiogenic than MORB mantle and more radiogenic than typical non-arc continental mantle. The relatively radiogenic signatures of many Kamchatka peridotites become apparent when compared to non-arc continental peridotites on a plot of $^{187}Os/^{188}Os$ vs. Re abundance (Fig. 6a). The $^{187}Os/^{188}Os$ signatures of most non-arc continental peridotites exhibit an overall negative correlation of $^{187}Os/^{188}Os$ vs. Re, which is best explained by variable degrees of melt depletion followed by aging that results in more radiogenic present-day Os isotope signatures in more fertile (i.e. less Re-depleted) mantle. This phenomenon is exemplified by the negative trend of $^{187}Os/^{188}Os$ vs. Re exhibited by Pyrenean peridotites, which has been attributed to variable melt depletion in continental mantle >1.9 Ga ago (Reisberg and Lorand, 1995; Burnham et al., 1998). Fig. 6a illustrates clearly that many of the Kamchatka peridotites, especially those from Bakening and Valovayam, have $^{187}Os/^{188}Os$ signatures more radiogenic than most non-arc SCLM. In addition, given the extremely low Re abundances (i.e. severe melt depletion) of the Avachinsky peridotites, these samples also appear to have anomalously radiogenic $^{187}Os/^{188}Os$ signatures compared to most non-arc continental mantle with comparable Re depletion.

Elevated $^{187}Os/^{188}Os$ compared to SCLM at a given Re abundance is also observed in other arc systems (Lihir, Cascades, Japan, and the Canadian Cordillera), although only for low Re abundance samples (Fig. 6b). In Kamchatka, the elevated $^{187}Os/^{188}Os$ is even more pronounced at low Re abundances, and is also apparent in higher Re abundance peridotites. The Kam-

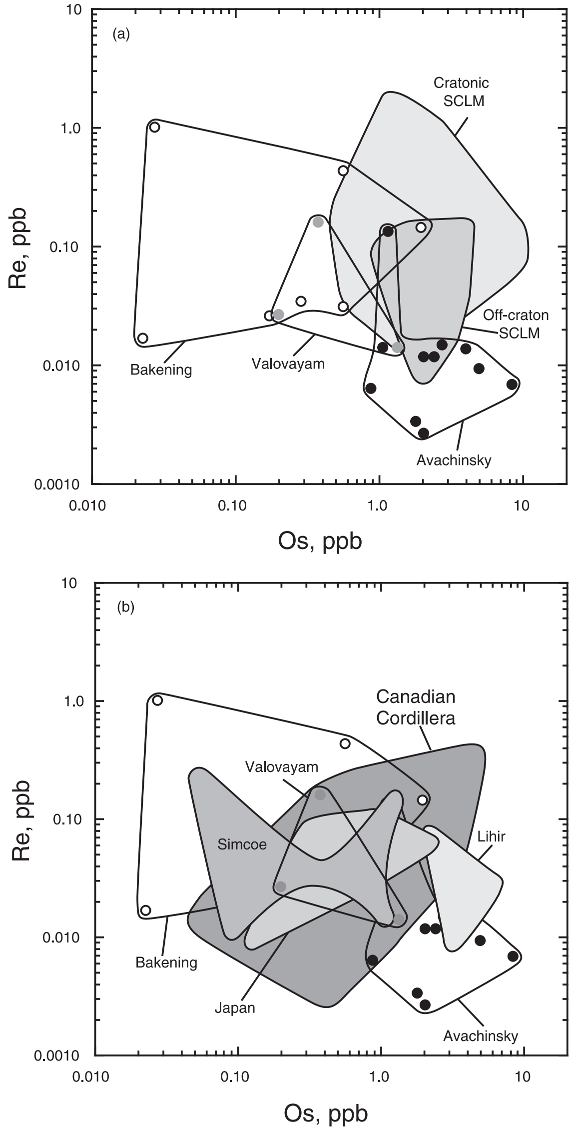


Fig. 5. Plot of Re concentration vs. Os concentration for the Kamchatka peridotites compared to cratonic and off-craton SCLM (a), and to other sub-arc mantle xenoliths (b). The Avachinsky harzburgites (black circles) have Os abundances similar to those found in peridotites from both cratonic and off-cratonic SCLM, and similar to Lihir arc peridotites, but have generally much lower Re contents, consistent with the Kamchatka mantle being extremely melt-depleted. The Valovayam (gray circles) and Bakening (unfilled circles) peridotites have higher Re contents, comparable to most SCLM, but their Os contents are generally lower than non-arc SCLM and are similar to those found in peridotites from other arcs, including Japan and Simcoe, Cascades. Data for cratonic SCLM from Carlson and Irving (1994), Pearson et al. (1995a), Pearson et al. (1995b), Olive et al. (1997), and Graham et al. (1999). Data for off-craton SCLM from Handler et al. (1997) and Meisel et al. (2001). Literature data for sub-arc mantle xenoliths from Brandon et al. (1996; Japan); Brandon et al. (1999; Simcoe, Cascades); Peslier et al. (2000; Canadian Cordillera); and McInnes et al. (1999; Lihir, Papua New Guinea).

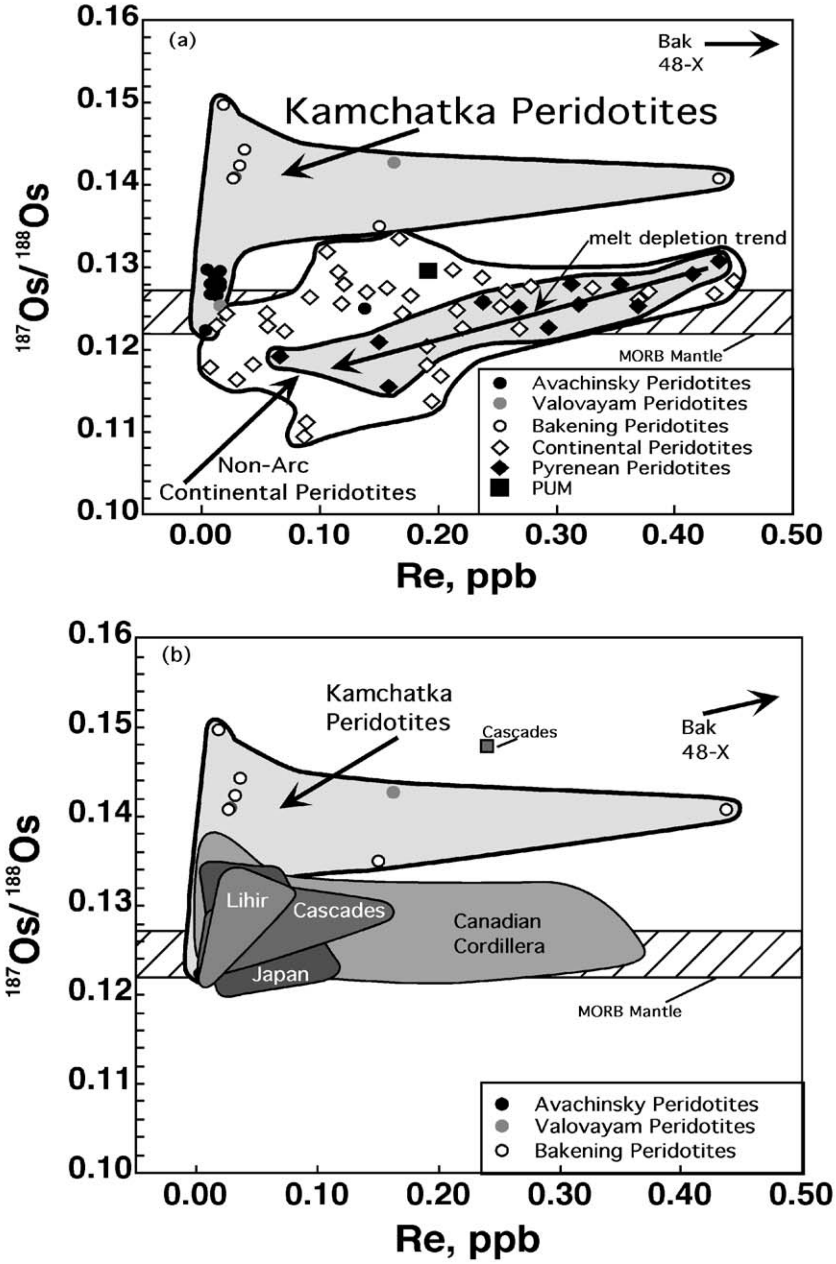


Fig. 6. Plot of ^{187}Os/^{188}Os vs. Re concentration for the Kamchatka peridotites compared to (a) non-arc continental peridotites and (b) other arc peridotites. The Re abundances of most of the Kamchatka peridotites are consistent with the extremely melt-depleted nature of the Kamchatka sub-arc mantle, with Re concentrations at the very low end of the range for strongly depleted mantle beneath ancient cratons. However, for a given Re abundance, the Kamchatka peridotites have ^{187}Os/^{188}Os signatures more radiogenic than any non-arc continental peridotites, and in many cases more radiogenic than estimates of hypothetical primitive upper mantle (PUM; Meisel et al., 2001). The radiogenic Os in the Kamchatka peridotites cannot be explained by ingrowth of ^{187}Os, due to the very low Re abundances and Re/Os ratios in most xenoliths, and is best explained by the addition of radiogenic slab-derived Os. Data for non-arc continental peridotite field and PUM from Meisel et al. (2001), and data for Pyrenean peridotites from Burnham et al. (1998). Data for arc peridotites from Brandon et al. (1996; Japan), Brandon et al. (1999; Simcoe, Cascades), McInnes et al. (1999; Lihir, Papua New Guinea), Peslier et al. (2000; Canadian Cordillera). Range in ^{187}Os/^{188}Os for MORB mantle field from Snow and Reisberg (1995) and Brandon et al. (2000).

chatka harzburgites and lherzolites exhibit the most radiogenic Os isotope signatures found so far in peridotites from arc systems (Fig. 6b), and therefore are important for better understanding Re–Os enrichment processes in subduction zone settings.

6.2. Origin of radiogenic Os isotope signatures in Kamchatka peridotites

Three possibilities might be invoked to explain the relatively radiogenic $^{187}Os/^{188}Os$ signatures of the Kamchatka peridotites compared to non-arc continental mantle: (1) contamination of the xenoliths by their host lavas; (2) radiogenic ingrowth of mantle with higher than chondritic Re–Os, followed in most cases by recent Re depletion; or (3) metasomatism of the Kamchatka sub-arc mantle by radiogenic slab-derived fluids.

6.2.1. Peridotite–host magma interaction

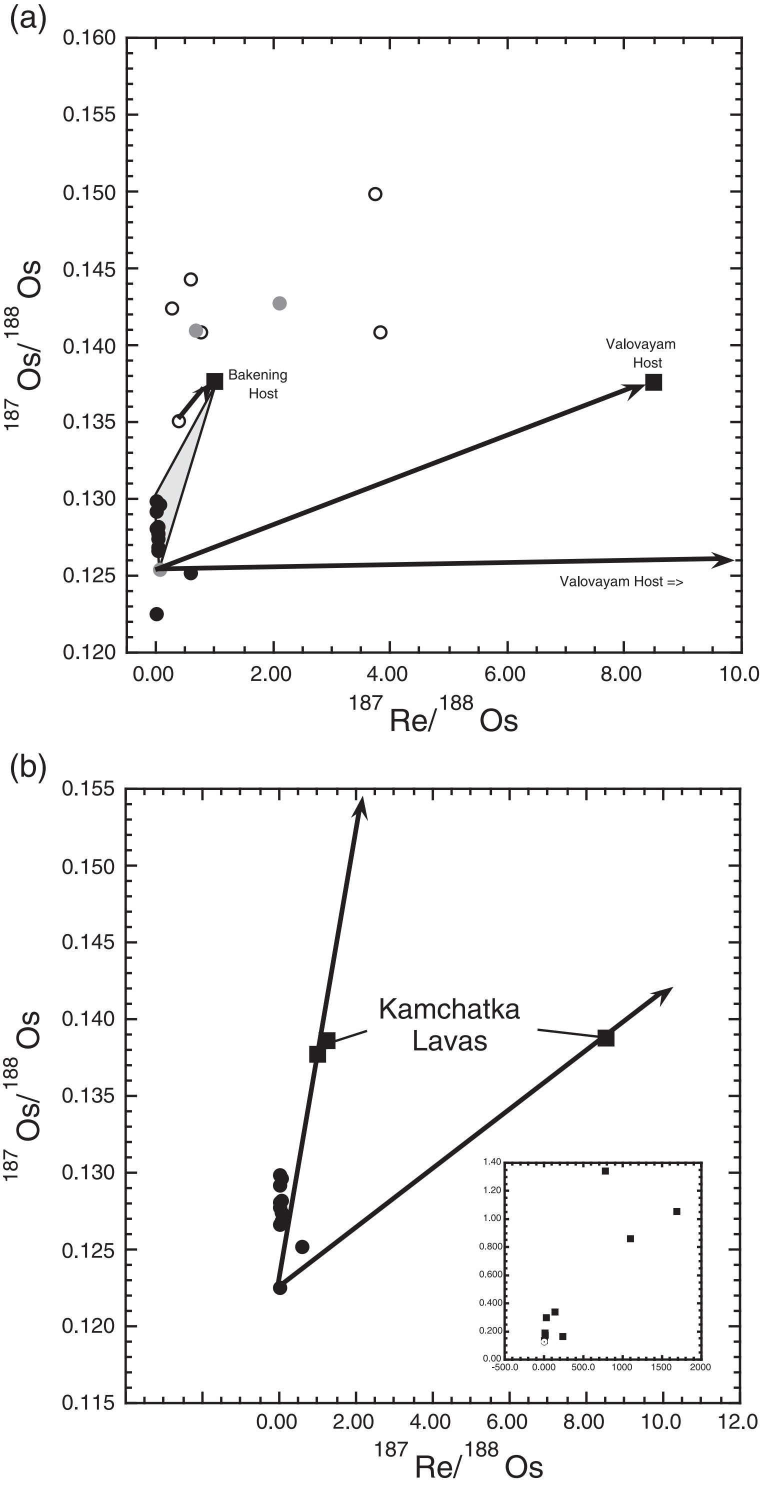
The possibility of mantle xenoliths acquiring radiogenic isotope signatures during interaction with their host magmas is always a concern. However, unlike the situation for the Sr, Nd, or Pb isotope systems, in which host magmas often have Sr, Nd, and Pb concentration orders of magnitude greater than the peridotite xenoliths that they host, Os concentrations in mantle peridotites are generally significantly greater than those of their host magmas, and thus the potential for contamination of the Os isotope signatures is minimized. Nevertheless, in situations where mantle xenoliths are characterized by unusually low Os abundances, or in which host magmas have unusually high or radiogenic Os isotopic compositions, the potential for contamination of the mantle xenoliths must be carefully evaluated.

The possibility that the radiogenic Os isotope signatures in the Kamchatka peridotites are due to interaction with host magmas can be tested based on the Re–Os isotope signatures of the host lavas. Although the host lava for the Avachinsky peridotites has not been analyzed, the host lavas for the Bakening and Valovayam mantle xenoliths have been analyzed (Widom et al., 2001). Fig. 7a illustrates the expected effects of bulk mixing of host lavas and mantle peridotites for the cases of Bakening and Valovayam. The host of the Bakening mantle xenoliths has a $^{187}Os/^{188}Os$ signature that is more radiogenic than one of the Bakening peridotites, but significantly less radiogenic than all of the others. Interaction of relatively unradiogenic mantle (similar to that of Avachinsky) with the Bakening host lava, thus, cannot explain the radiogenic signatures in most of the Bakening peridotites. Even the one Bakening peridotite (48–31) that is less radiogenic than the host is not easily explained by host magma–peridotite interaction, as such mixing scenarios would result in Re/Os ratios that are higher than that observed for this sample (Fig. 7a). The expected increase in Re–Os due to interaction of highly melt-depleted (low Re) peridotites with host magmas is even more pronounced in the case of Valovayam, where the host lavas for the xenoliths have very high $^{187}Re/^{188}Os$ ratios (Fig. 7a).

The ppb-level Os concentrations and, with one exception, extremely low Re concentrations (≤ 15 ppt) in the Avachinsky peridotites greatly limits the extent to which interaction with their host magma could have affected the Os isotope signatures of these peridotites. In fact, if we consider sample AXV14 to represent the "pre-metasomatic" mantle wedge Re–Os composition ($^{187}Os/^{188}Os = 0.1226$, $^{187}Re/^{188}Os = 0.006$), none of the 14 lavas from Kamchatka for which Re–Os data are available (Widom et al., 2001) can produce the range in $^{187}Os/^{188}Os$ of the other Avachinsky harzburgites by lava–peridotite mixing without producing a concomitant increase in Re–Os above what is observed (Fig. 7b).

The relatively radiogenic Os isotope signatures in Kamchatka peridotites thus cannot easily be attributed

Fig. 7. Plot of $^{187}Os/^{188}Os$ vs. $^{187}Re/^{188}Os$ for the Kamchatka peridotites and their host basalts. (a) Potential mixing relationships between the least radiogenic Bakening and Valovayam peridotites and their respective host lavas. The host lavas are not radiogenic enough to account for the most radiogenic signatures in the peridotites, and host basalt–peridotite mixing would generally produce Re/Os ratios in the peridotites that are higher than observed. (b) Potential mixing relationships between the least radiogenic Avachinsky peridotite and a range of lava compositions based on 14 Kamchatka lavas for which Re–Os isotope data are available (full range shown in inset; Widom et al., 2001). All but one of the Avachinsky peridotites have Re/Os ratios lower than expected if lava–peridotite mixing were the cause of the radiogenic Os isotope signatures in the peridotites. Peridotite xenolith symbols as in Fig. 5: Avachinsky harzburgites, black circles; Valovayam peridotites, gray circles; Bakening peridotites, unfilled circles. Kamchatka lavas shown as black squares.


(a)
0.160
0.155
0.150
0.145
0.140
0.135
0.130
0.125
0.120
^{187}Os/^{188}Os
Bakening Host
Valovayam Host
Valovayam Host =>
0.00
2.00
4.00
6.00
8.00
10.0
^{187}Re/^{188}Os
(b)
0.155
0.150
0.145
0.140
0.135
0.130
0.125
0.120
0.115
^{187}Os/^{188}Os
Kamchatka Lavas
1.40
1.20
1.00
0.800
0.600
0.400
0.200
0.00
-500.0
0.000
500.0
1000
1500
2000
0.00
2.00
4.00
6.00
8.00
10.0
12.0
^{187}Re/^{188}Os

to interaction between the host lavas and normal (i.e. unmetasomatized) depleted mantle. This conclusion is consistent with previous arguments against contamination of the mantle xenoliths by their host lavas based on lithophile element abundance patterns, which require reaction of the xenoliths with fluids and melts completely unlike the respective host basalts (Kepezhinskas et al., 1996, 1997).

6.2.2. Radiogenic ingrowth

The Bakening peridotites and two of the three Valovayam peridotites have $^{187}Os/^{188}Os$ signatures that are significantly more radiogenic than almost any non-arc continental mantle (Fig. 8), and more radiogenic than estimates (Meisel et al., 2001) for primitive upper mantle (PUM; Fig. 8). Hence, regardless of the melting history of this mantle, it would have had to evolve with a long-term history of suprachondritic Re/Os ratios in order to generate the present-day $^{187}Os/^{188}Os$ ratios by in situ ingrowth of ^{187}Os. Some of these samples have $^{187}Re/^{188}Os$ ratios that are close to or higher than PUM (Fig. 8), and could theoretically have developed their radiogenic Os isotope signature by in situ radiogenic ingrowth. Calculations using the Bakening peridotite with the most radiogenic $^{187}Os/^{188}Os$ (0.1566) and the highest

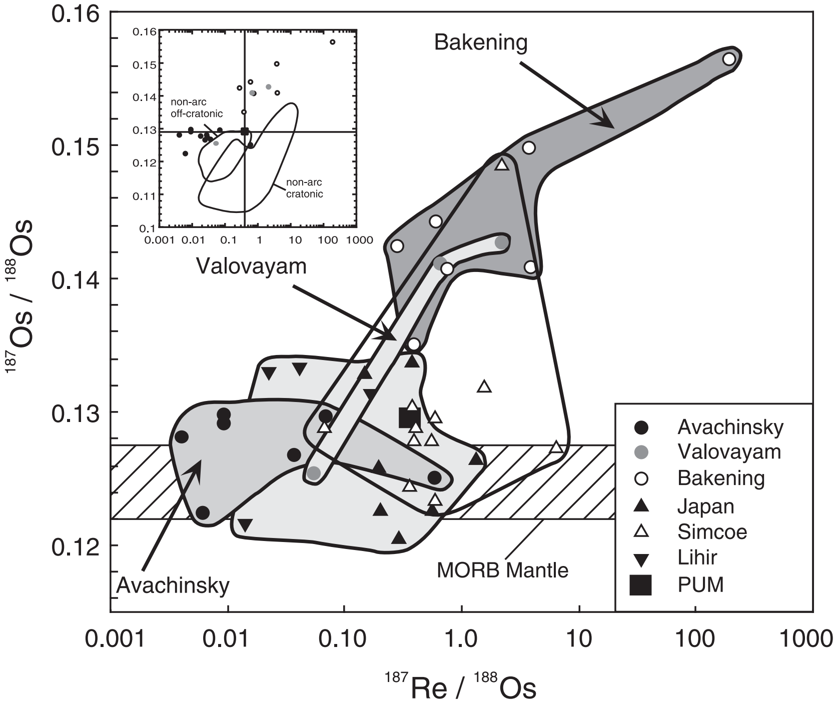


Fig. 8. Plot of $^{187}Os/^{188}Os$ vs. $^{187}Re/^{188}Os$ for the Kamchatka peridotites. The Avachinsky harzburgites, which have experienced hydrous slab-fluid metasomatism, have less radiogenic Os isotope signatures and no apparent slab-derived Re. In contrast, the Valovayam and Bakening peridotites, which experienced metasomatism by siliceous slab melts and carbonate melts, respectively, have more radiogenic Os and have inherited significant slab-derived Re. The Avachinsky peridotites are similar in Re–Os isotopic composition to peridotites from Japan and Lihir, arcs that are similar to the southern Kamchatka arc front with subduction of cold, old (~ 100–175 Ma) oceanic crust at depths of ~ 100–150 km. The Valovayam peridotites span a similar range in Re–Os isotopic composition to peridotites from Simcoe, Cascades, which is similar to the Kamchatka northern arc front with subduction of hot, young (~ 25 Ma) oceanic crust. The Bakening peridotites, which have experienced carbonate-rich slab-melt metasomatism, extend to Os isotopic compositions and Re/Os ratios higher than any other arc peridotites. Inset shows composition of Kamchatka peridotites compared to non-arc cratonic and off-craton peridotites and to PUM. Data for cratonic SCLM from Carlson and Irving (1994), Pearson et al. (1995a), Pearson et al. (1995b), Olive et al. (1997), and Graham et al. (1999). Data for off-craton SCLM from Handler et al. (1997) and Meisel et al. (2001). Literature data for sub-arc mantle xenoliths from Brandon et al. (1996; Japan), Brandon et al. (1999; Simcoe, Cascades), and McInnes et al. (1999; Lihir, Papua New Guinea). PUM value from Meisel et al. (2001); range in $^{187}Os/^{188}Os$ for MORB mantle field from Snow and Reisberg (1995) and Brandon et al. (2000).

^{187}Re/^{188}Os (188) indicate that this mantle would have had to be isolated from the convecting mantle for only ~ 10 Ma to develop the observed Os isotope signature. However, the next most radiogenic Bakening peridotite with a ^{187}Os/^{188}Os ratio of 0.1498 and a ^{187}Re/^{188}Os ratio of 3.7 would require an ingrowth period of ~ 420 Ma, significantly longer than the age of the Mesozoic–Tertiary accreted terranes that comprise the southern Kamchatka lithosphere (Kepezhinskas et al., 1997). A similar result is obtained for the most radiogenic of the Valovayam peridotites, which would require >550 Ma of in situ ingrowth to explain the radiogenic Os isotope signature with its Re/Os ratio that is only slightly higher than PUM. The northern Kamchatka crustal terranes are, however, comparatively young (pre-Neogene) oceanic and volcanic arc terranes (Hochstaedter et al., 1994). Although one might argue that these mantle domains could have evolved previously with significantly higher Re/Os, and only recently experienced Re depletion due to melt extraction, most continental mantle has Re/Os ratios similar to or significantly lower than that of the radiogenic Bakening and Valovayam peridotites (Fig. 8 inset). It seems unlikely, therefore, that the radiogenic Os isotope signatures in Bakening and Valovayam peridotites are due solely to in situ radiogenic ingrowth. Alternatively, slab-fluid metasomatism might explain the radiogenic Os signatures in these peridotites. The implications of this are considered in Section 6.2.3.

The Avachinsky peridotites extend to Os isotope signatures only slightly above PUM. However, three samples that have ^{187}Os/^{188}Os signatures equal to or higher than PUM have ^{187}Re/^{188}Os ratios significantly below PUM (Fig. 8), and thus have “future ages” that require either a previous history with suprachondritic Re/Os ratios followed by recent melt extraction, or open system addition of radiogenic Os. If one makes the assumption that the Avachinsky mantle evolved until recently with a ^{187}Re/^{188}Os ratio of ~ 10 (the highest value observed in any but the rarest example of continental mantle; Fig. 8 inset), then ingrowth over only ~ 10 Ma would be required to produce the most radiogenic Os isotope signatures in the Avachinsky peridotites. This possibility cannot be ruled out. On the other hand, a rough positive correlation of ^{187}Os/^{188}Os with degree of LREE enrichment (La/Sm) in the Avachinsky harzburgites, and very radiogenic Sr in a clinopyroxene separate from Avachinsky harzburgite AVX50 (one of the most radiogenic in Os), support the possibility that the radiogenic Os in these samples is slab-derived (Fig. 9).

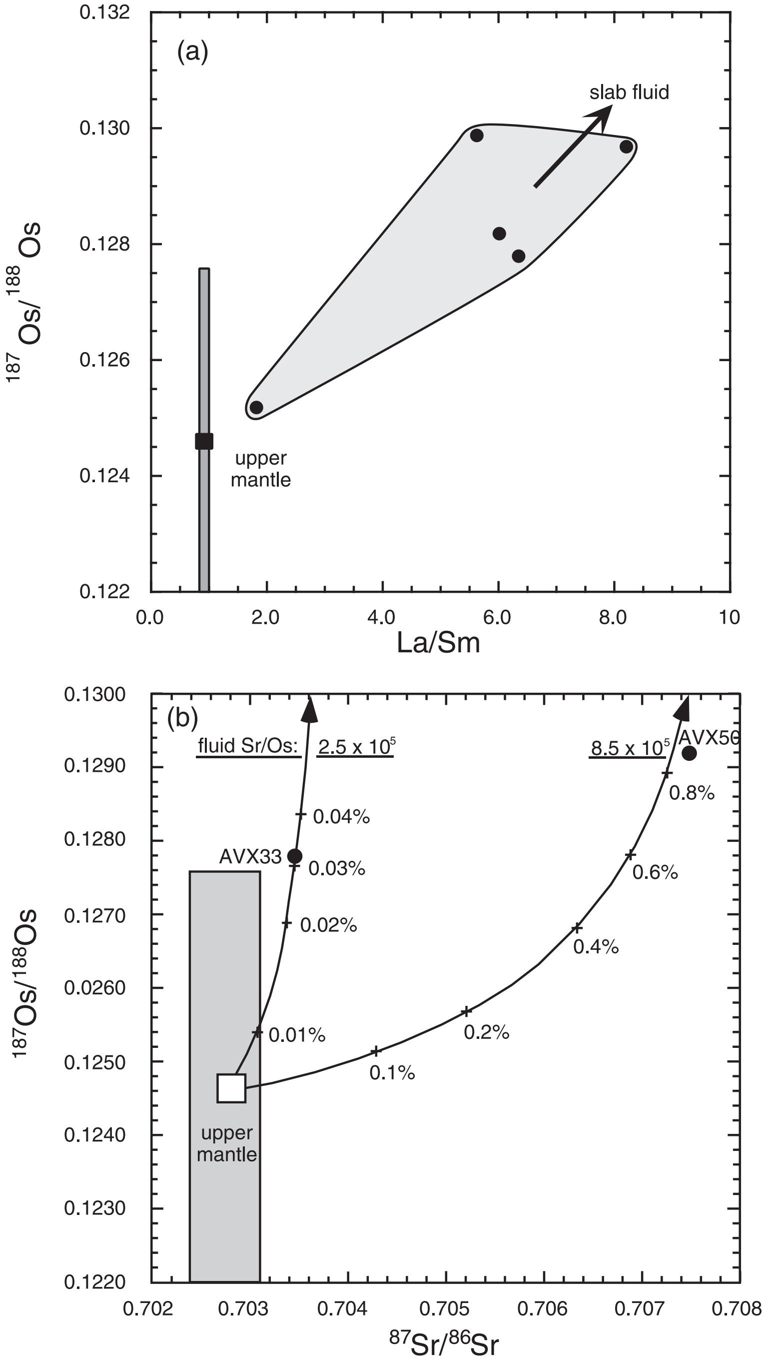
Given the difficulty in explaining the radiogenic Bakening and Valovayam peridotites by either interaction with their host basalts or in situ radioactive ingrowth, and given the abundant petrographic and geochemical evidence for slab-fluid metasomatism in all of the Kamchatka arc xenoliths (described previously), we consider it likely that all of the Kamchatka peridotites have been metasomatized by slab-derived fluids with radiogenic crustal Os. The implications of this model are discussed below.

6.2.3. Slab-derived fluids as a source of radiogenic Os in the Kamchatka peridotites

To the extent that Os might be liberated during slab dewatering, one would expect the slab fluids to have radiogenic Os isotope signatures characteristic of the subducting oceanic crust plus or minus any contributing subducted sediment. Terrigenous and pelagic sediments are generally characterized by highly radiogenic Os isotope signatures (^{187}Os/^{188}Os ≈ 1.0), due to the time-integrated high Re/Os ratios of the continental crust, and the derivation of seawater Os from radiogenic continental protoliths (Esser and Turekian, 1993; Ravizza et al., 1996; Burton et al., 1999a; Sharma et al., 1999).

Hydrothermally altered oceanic lithosphere also can be highly radiogenic in Os, due both to aging of the high Re/Os basaltic and gabbroic crustal components, and to interaction of the crust and uppermost oceanic mantle with radiogenic seawater (Hart et al., 1999; Standish et al., 2002). Analyses of MORB-related gabbros from ODP hole 735B (Hart et al., 1999; Blusztajn et al., 2000), Hawaii (Lassiter and Hauri, 1998), and the Canary Islands (Widom and Neumann, unpublished data) provide estimates of the composition of lower oceanic crust over a broad geographic and crustal age range (Indian Ocean, 11–12 Ma; Pacific Ocean, ~ 100 Ma and Atlantic Ocean, ~ 160–180 Ma, respectively). ^{187}Os/^{188}Os ratios in these samples range from 0.140 to 0.467, with Os concentrations ranging from 0.5 to 838 ppt. An important observation from the combined MORB gabbro data sets is that the ^{187}Os/^{188}Os signatures are essentially independent of the age of the oceanic crust.

For example, gabbros with 0.03–0.40 ppb Os from both the 11–12 Ma Indian Ocean crust and the 160–180 Ma Atlantic Ocean crust have essentially identical ranges in $^{187}Os/^{188}Os$ from ~ 0.14 to 0.24, and similar $^{187}Os/^{188}Os$ for a given Os abundance. A negative correlation of $^{187}Os/^{188}Os$ with Os abundance (Hart et al., 1999) and a positive correlation of $^{187}Os/^{188}Os$ with seawater alteration indices such as



Rb/Cs ratios (Blusztajn et al., 2000) suggest that seawater alteration exerts a stronger control on the Os isotope signature of oceanic lower crust than does age. Many Re–Os isotope analyses are available for MORB upper oceanic crust, but they generally show low Os concentrations, high Re/Os ratios, and radiogenic Os signatures (Roy-Barman and Allegre, 1994; Schiano et al., 1997; Roy-Barman et al., 1998), similar to those of the gabbroic lower crust.

Slab-derived fluids associated with subduction in central and southern Kamchatka (e.g. beneath Avachinsky and Bakening) have been proposed, based on U-series disequilibria and Sr and Pb isotope systematics, to be dominated by altered oceanic crust, with minimal sediment contribution (Turner et al., 1998). A best estimate of a likely sediment-free slab composition might be taken as the average of the gabbroic strip samples from Hart et al. (1999), which have an Os concentration of ~ 10 ppt, and $^{187}Os/^{188}Os$ of ~ 0.2. However, the possible range of compositions, based on the general correlation of $^{187}Os/^{188}Os$ with Os concentration, is much larger (~ 350 ppt Os and $^{187}Os/^{188}Os \approx 0.14$ to ~ 5 ppt Os and $^{187}Os/^{188}Os \approx 0.44$).

The concentration of Os that may be liberated in a slab-derived fluid is more difficult to constrain. The increased volatility of PGE with increase in fluid f_{O_2} and f_{HCl} (Wood, 1987; Xiong and Wood, 2000) has been used in support of the idea that oxidizing and Cl-rich slab fluids should be effective transport agents of the volatile element Os (Brandon et al., 1996). On the other hand, a recent study of Os systematics in blueschists and eclogites suggests that Os is not significantly mobilized from the slab during subduction (Becker, 2000). Our data, however, suggest that Os must be highly mobile in slab-derived fluids, in order that radiogenic Os isotope signatures be imprinted on mantle peridotites with high Os abundances. For comparison, Os isotope signatures in mantle peridotites from the Cascades and Japan, which have similar Os isotope signatures but significantly lower Os concentrations than peridotites from Avachinsky (Figs. 5b and 8), have been explained by bulk mixing of mantle and 5–30% slab component (assuming the slab fluid composition is approximately the composition of the subducted slab component; Brandon et al., 1996, 1999). Similar calculations using the above range of slab compositions would require a mixture with 80–90% slab component to generate the most radiogenic Avachinsky peridotite ($^{187}Os/^{188}Os = 0.1299$) from the least radiogenic peridotite ($^{187}Os/^{188}Os = 0.1226$), due to the high Os abundance (2 ppb) in the starting peridotite. Similar results are obtained for most of the Bakening peridotites, which have lower Os abundances but more radiogenic Os isotope signatures. Even the lowest Os peridotite from Bakening (0.027 ppb Os) would require a 40–70% slab component to reach the observed Os isotope signature (0.1566), assuming bulk mixing of potential slab compositions based on the gabbroic strip samples from Hart et al. (1999), as described previously.

More likely scenarios than the simple mixing models described above involve the generation of a slab-derived fluid that is either strongly enriched in Os relative to the subducting slab materials, or which interacts with the overlying mantle wedge at very high fluid/rock ratios, possibly stabilizing an Os-rich phase (Borg et al., 2000). Previous estimates of the Sr/Os of slab-derived fluids required to produce observed Sr–Os isotope variations in arc lavas from Lassen (Borg et al., 2000) and Grenada (Woodland et al., 2002) are 7.5×10^5 and 1.25×10^5, respectively, which are similar to, or significantly lower than, subducting-altered

Fig. 9. (a) Plot of $^{187}Os/^{188}Os$ vs. La/Sm for Avachinsky harzburgites. The rough positive correlation of $^{187}Os/^{188}Os$ with degree of LREE enrichment (La/Sm) is consistent with a slab-derived origin for both the LREE enrichment and the radiogenic Os in these samples. (b) Plot of $^{187}Os/^{188}Os$ vs. $^{87}Sr/^{86}Sr$ for two Avachinsky harzburgites ($^{87}Sr/^{86}Sr$ from Jon Davidson, unpublished data). Mixing between average depleted MORB mantle (Snow and Reisberg, 1995) and slab fluids with Sr/Os ratios ranging from 2.5×10^5 to 8.5×10^5, similar to those proposed for Lassen and Grenada (Borg et al., 2000; Woodland et al., 2002), can reproduce the Sr–Os isotope signatures in the two Avachinsky peridotites with less than 1% of the slab-fluid component. Model parameters are as follows: DMM: Os = 2 ppb (Shirey and Walker, 1998), $^{187}Os/^{188}Os = 0.1246$ (Snow and Reisberg, 1995), Sr = 9 ppm (Sun and McDonough, 1989), $^{87}Sr/^{86}Sr = 0.7028$ (Ito et al., 1987); Slab fluid: Sr = 2933 ppm (Tatsumi and Kogiso, 1997); for AVX33 and AVX50, respectively, Sr/Os = 2.5×10^5 and 8.5×10^5, $^{187}Os/^{188}Os = 0.25$ and 0.44 (Hart et al., 1999), and $^{87}Sr/^{86}Sr = 0.704$ and 0.709. The upper mantle Os isotopic composition is based on abyssal peridotite data (Snow and Reisberg, 1995; Brandon et al., 2000), the La/Sm ratio (~ 0.95) is based on the average for MORB (Sun and McDonough, 1989), and the upper mantle $^{87}Sr/^{86}Sr$ isotopic composition is based on the range for Pacific N-MORB (Ito et al., 1987).

oceanic crust (2.5×10^5–3.1×10^8; Hart et al., 1999). Fig. 9b illustrates potential mixing relationships that could generate the observed Sr–Os isotope relationships for the two Kamchatka harzburgites for which data are available, assuming that the Kamchatka unmetasomatized mantle is similar to typical depleted Pacific MORB mantle. In the illustrated models, slab fluids are assumed to have 2933 ppm Sr (~ 27-fold enrichment in Sr relative to subducting oceanic crust; Tatsumi and Kogiso, 1997), ^{87}Sr/^{86}Sr = 0.704 and 0.709, and ^{187}Os/^{188}Os of 0.25 and 0.44 (the radiogenic end of values found in altered oceanic gabbros; Hart et al., 1999). Slab fluids with Sr/Os ratios ranging from 2.5×10^5 to 8.5×10^5, similar to those proposed for Lassen and Grenada, can reproduce the Sr–Os isotope signatures in the two Avachinsky peridotites with less than 1% of the slab-fluid component (Fig. 9b). Although such models are non-unique, it is difficult to reproduce the Kamchatka data with fluid Sr/Os ratios significantly greater than the low end of the range observed in altered oceanic crust (e.g. 10^5–10^6), suggesting that Os may be comparably mobile to Sr in slab-derived fluids.

It is important to note that if mixing of slab fluids with mantle wedge is thought of as a strict two-component mixing process, one would expect to see a linear correlation of ^{187}Os/^{188}Os with 1/Os. Although such a correlation is observed in harzburgites from the Canadian Cordillera (Peslier et al., 2000), it is not observed in any other arc peridotite suites including the Kamchatka peridotites. The lack of linear ^{187}Os/^{188}Os vs. 1/Os correlations in most arc peridotite suites suggests that slab fluid enrichment is generally a more complex process than simple two-component mixing. Several possible mechanisms might be invoked to explain the generally negative but non-linear correlation of ^{187}Os/^{188}Os vs. 1/Os observed in the Kamchatka peridotite suite. For example, if the pristine (unmetasomatized) mantle wedge were characterized by initially variable Os abundances, then fluxing of the wedge with a homogeneous radiogenic slab fluid would yield more radiogenic signatures in those samples with lower Os abundances, but the mixing process would not be a strictly two-component process. Another possibility is that primary Os within the mantle wedge is mobilized during slab-fluid infiltration. For example, scavenging of primary mantle wedge Os combined with isotopic exchange between mantle and slab-fluid Os also could result in mantle with lower than normal Os abundances and negative but non-linear trends of ^{187}Os/^{188}Os vs. 1/Os. Support for this latter process is discussed below.

Relatively low Os abundances in some mantle peridotites have led to the suggestion that in addition to transporting radiogenic Os, oxidizing slab fluids may destabilize sulfides in the mantle wedge, thus, ultimately decreasing the Os abundance of slab-fluid metasomatized mantle (Brandon et al., 1996). However, there is alternative evidence for the formation of secondary sulfides during some arc-related mantle metasomatic processes. Composite inclusions consisting of glass plus water and glass plus sulfides in metasomatic minerals from Luzon arc harzburgites have been interpreted as evidence for mantle metasomatism by a hydrous and S-rich silicate melt (Batan Island; Schiano et al., 1995). The formation of metasomatic sulfides is thus a potential mechanism for enrichment of sub-arc mantle in radiogenic slab-derived Os (Borg et al., 2000), due to very high partition coefficients for Os in sulfides (e.g. Hart and Ravizza, 1996; Burton et al., 1999b). In addition to the possibility of stabilizing metasomatic sulfides with slab Os isotope signatures, the Os isotopic composition of pre-existing interstitial mantle sulfides could be overprinted (Burton et al., 1999b) during equilibration with slab-derived fluids, based on the low closure temperatures for Os in at least some sulfide phases (Brenan et al., 2000). This process may be similar to that attributed to resetting of abyssal peridotite Os isotope signatures during hydrothermal seafloor alteration (Hart et al., 1999; Blusztajn et al., 2000).

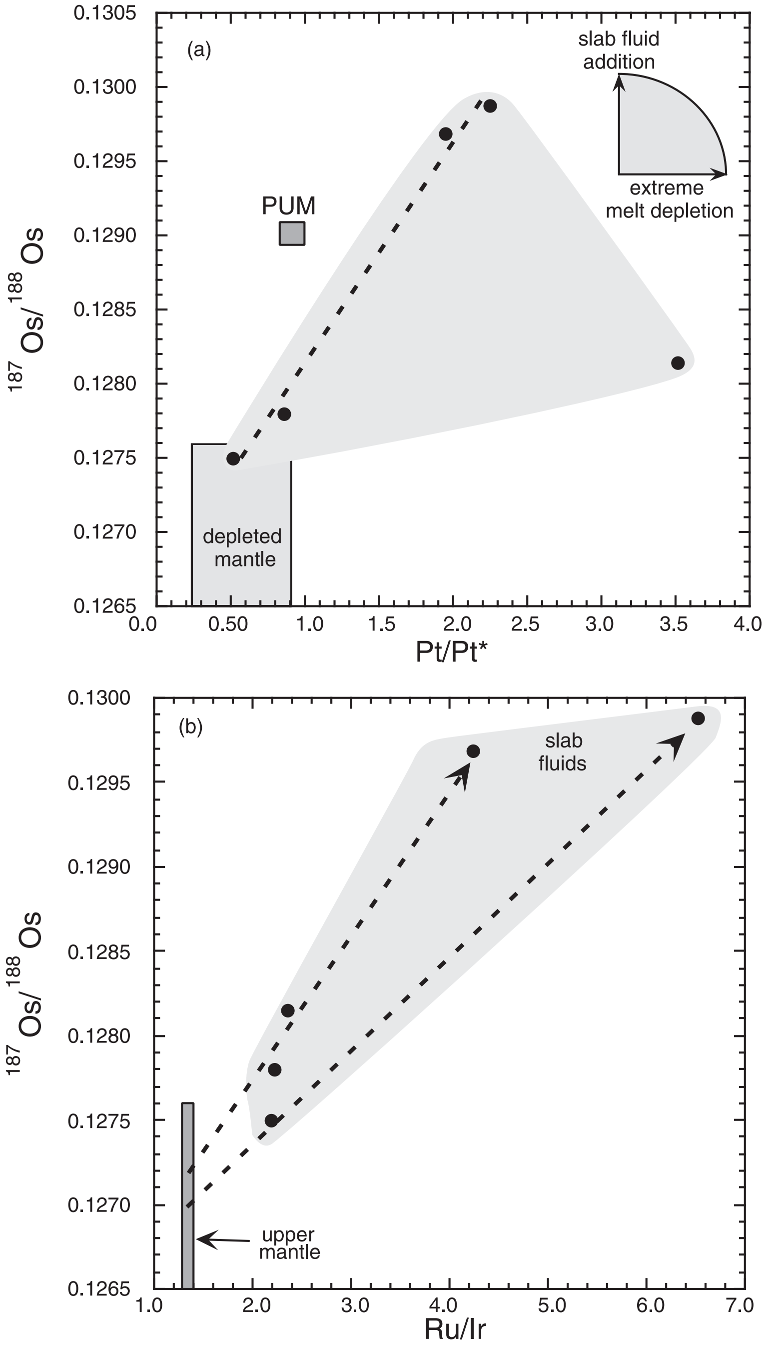
In the case of the Kamchatka mantle peridotites, however, sulfides are extremely rare (Kepezhinskas et al., 2002), so the extent to which the above processes are responsible for the radiogenic Os isotope signatures is unclear. An alternative possibility is that breakdown of sulfides in the presence of oxidizing slab fluids, combined with fluid-induced melting, may have caused Os to partition into residual phases such as PGE and/or Pt alloys (e.g. Borisov and Walker, 2000 and references therein). Four of five Avachinsky harzburgites for which PGE abundance data are available (Kepezhinskas et al., 2002) display a strong positive correlation between ^{187}Os/^{188}Os and Pt/Pt*, a measure of Pt enrichment relative to other Pd-group PGE ($Pt/Pt^* = [Pt_N]/\sqrt{\{[Rh_N] \times [Pd_N]\}}$ where N denotes chondrite normalized abundances; Garuti et al., 1997b). This correlation, extending to $^{187}Os/^{188}Os$ signatures more radiogenic than PUM and suprachondritic Pt/Pt* (Fig. 10a), may suggest a link between introduction of radiogenic Os and Pt enrichment. The Pt enrichment has been attributed to stabilization of Fe–Pt alloys due to extreme melt depletion of the Kamchatka sub-arc mantle (Kepezhinskas et al., 2002), which could have been triggered by slab-fluid fluxing of the mantle wedge, thus, simultaneously introducing radiogenic slab-derived Os. Concentration of Pt as Pt–Fe and Pt–Ir alloys, along with Ir–Os alloys, is known to occur during dunite formation in the absence of a sulfide liquid, and may be related to development of high f_{O_2} conditions (Amosse et al., 1990; Garuti et al., 1997a). A positive correlation between $^{187}Os/^{188}Os$ and Ru/Ir in the Avachinsky peridotites (Fig. 10b) is also consistent with the known volatility of Os and Ru (but not other PGEs) in the presence of high f_{O_2} and Cl-rich fluids (Wood, 1987), and further supports the hypothesis that the radiogenic Os is imparted to the mantle wedge by slab-fluid metasomatism.

6.3. Intra-arc variations in slab-derived Os and Re

One of the more striking results of the Re–Os isotope systematics among the Kamchatka peridotites is the intra-arc variation in isotopic signatures (Fig. 8). The Avachinsky peridotites generally have the least radiogenic Os and lowest Re/Os, while the Bakening peridotites have the most radiogenic Os and highest Re/Os. The Valovayam peridotites span the distinct compositional fields defined by Bakening and Avachinsky. As a group, there is a broad positive correlation of $^{187}Os/^{188}Os$ with $^{187}Re/^{188}Os$, but that is defined primarily by the Valovayam and Bakening samples, and is not apparent in the Avachinsky peridotites (Fig. 8). The origin of the radiogenic Os in these samples was discussed earlier and it was argued that slab-derived Os, as opposed to aging of high Re/Os mantle or peridotite–host lava contamination, is the primary contributing factor for the radiogenic Os signatures. The implication of the positive correlation between $^{187}Os/^{188}Os$ and $^{187}Re/^{188}Os$ in the Valovayam and Bakening samples is thus that some Re was introduced into the mantle wedge beneath these volcanoes during slab-fluid metasomatism. Mobility of Re in slab-derived fluids is consistent with low Re abundances relative to normal MORB in some eclogites and blueschists (Becker, 2000). However, other eclogites are known to have high Re abundances and high Re/Os ratios, indicative of retention of Re in the slab during subduction (Pearson et al., 1995c; Ruiz et al., 1998; Woodland et al., 2002). The extremely low Re abundances in the Avachinsky peridotites (<15 ppt in all but one) also suggest that slab fluid-derived Os can be imparted to the mantle wedge without any significant addition of Re in some situations.

The differences in metasomatic addition of slab-derived Re and Os between Avachinsky, Valovayam, and Bakening are presumably related to the distinct nature of the metasomatizing fluids/melts affecting the mantle wedge beneath the respective volcanoes, which is in turn related to the variable $P-T$ conditions of the subducting slabs (Kepezhinskas and Defant, 1996a). In detail, the differences in the Re–Os isotope characteristics of the metasomatic products could be due to differences in the Re and Os abundances of the metasomatic fluids/melts, which might be controlled either by variable solubility of Re and Os in the compositionally distinct fluids/melts or by differences in the extent to which Re- and Os-bearing phases are accessed during slab dewatering vs. melting at a range of $P-T$ conditions. Alternatively, the Re–Os isotope characteristics of the metasomatic products could be controlled primarily by differences in the nature of the fluid/melt–mantle reactions that occur during mantle wedge metasomatism.

Hydrous slab-fluid metasomatism in the southern arc front beneath Avachinsky has apparently resulted in an enrichment in radiogenic slab-derived Os with no significant enrichment in Re. A similar situation is indicated by the Re–Os systematics of mantle xenoliths from Japan and Lihir, which are compositionally very similar to those from Avachinsky (Fig. 8). All three of these cases involve subduction of old oceanic crust (100–175 Ma), with comparable depths to the slabs for at least Japan and Avachinsky (~ 100–150 km; Gill, 1981; Xiong and Wood, 1999; depth to Lihir slab is unknown). The solubility of both Re and Os have been shown to be high in Cl-rich fluids (Xiong and Wood, 1999, 2000), and thus both elements might be expected to be effectively transported in hydrous slab fluids. Although the solubility of Os increases strongly with increasing f_{O_2}, and the solubility of Re is relatively insensitive to increasing f_{O_2}, the existing experimental results suggest the solubility of Re should be higher than Os for a fluid with a given chlorinity and f_{O_2} (Xiong and Wood, 1999, 2000). Thus, it seems likely that the



budget of slab-derived Re and Os in hydrous metasomatized peridotites is controlled primarily by fluid–mantle reactions rather than the relative compatibility of the respective elements in hydrous fluids generated during slab dewatering. One possibility is that hydrous fluid-fluxing of the mantle wedge is accompanied by fluid-induced melting, which could favor the incorporation of slab-derived Os over Re, since the former is compatible and the latter incompatible during mantle melting. Preferential retention of Os relative to Re might be facilitated by stabilization of Pt–Ir and Ir–Os alloys during extreme melt depletion, as discussed previously.

The Valovayam peridotites are inferred to have been metasomatized by adakitic slab melts (Fig. 3a), formed during the subduction of hot, young (~ 15–25 Ma) oceanic crust beneath northern Kamchatka. Although one of the Valovayam peridotites overlaps those of Avachinsky in terms of ^{187}Os/^{188}Os–^{187}Re/^{188}Os, the other two have much more radiogenic ^{187}Os/^{188}Os, and ^{187}Re/^{188}Os higher than PUM (Fig. 8). The higher Re/Os in the Valovayam peridotites relative to the Avachinsky peridotites is due in part to lower Os abundances and in part to higher Re abundances in the former (Fig. 5). A similar range in Re–Os abundances and isotopic compositions characterizes mantle peridotites from the Cascades (Fig. 8; Brandon et al., 1999), which like northern Kamchatka, involve subduction of young (~ 25 Ma) oceanic crust and the production of adakites (slab melts; Defant and Drummond, 1990, 1993). These data suggest that both slab-derived Re and Os are imparted to the mantle wedge by slab melts, but that there is a net loss of Os from affected regions of the mantle wedge, possibly due to scavenging of Os by strongly oxidized slab melts (Brandon et al., 1999). The experimentally determined solubility of Os in high f_{O_2} silicate melts supports the suggestion that significant amounts of Os, in addition to Re, might be transported in oxidized slab melts (Borisov and Walker, 2000). However, silicate melts are also believed to be efficient oxidizing agents in the mantle (Amundsen and Neumann, 1992; Brandon and Draper, 1996; Frost and Ballhaus, 1998), particularly when reacting with highly depleted mantle wedge which has a limited buffering capacity (Parkinson and Arculus, 1999), and could result in significant Os loss. In contrast, hydrous fluids are believed to be significantly less effective oxidizing agents in the mantle based on thermodynamic considerations (Frost and Ballhaus, 1998) as well as the lack of correlation between increased mantle f_{O_2} and hydrous phases (Amundsen and Neumann, 1992). Mobilization and net loss of Os from localized regions within the mantle wedge by interaction with oxidized slab melts but not hydrous fluids may explain the generally lower Os abundances in Valovayam relative to Avachinsky peridotites.

The Bakening peridotites exhibit the most radiogenic Os signatures and the highest Re/Os ratios found in Kamchatka or any other reported arc peridotite suite. As with the Valovayam peridotites, the high Re/Os ratios in Bakening peridotites are due both to anomalously low Os and high Re compared to the hydrous fluid-metasomatized Avachinsky peridotites. Bakening volcano resides behind the Kamchatka arc front, where the slab is at a depth of ~ 200 km (Gorbatov et al., 1997, 1999), hence, deeper than the slabs associated with the other arc xenolith suites discussed including Avachinsky volcano (~ 100 km above the slab at the Kamchatka arc front; Gorbatov et al., 1997, 1999). Given the evidence in the Bakening xenoliths for metasomatism by carbonate-rich melts (Fig. 3c; Kepezhinskas and Defant, 1996a), these data may suggest that deeply derived, carbonate-rich slab

Fig. 10. Plot of ^{187}Os/^{188}Os vs. (a) Pt/Pt* and (b) Ru/Ir for Avachinsky harzburgites. Pt/Pt* is defined as $[\mathrm{Pt_N}]/\sqrt{\{[\mathrm{Rh_N}] \times [\mathrm{Pd_N}]\}}$, where N denotes chondrite-normalized abundances (Garuti et al., 1997b). Four of the Avachinsky harzburgites display a strong positive correlation of ^{187}Os/^{188}Os with Pt/Pt*, extending to ^{187}Os/^{188}Os signatures more radiogenic than PUM and suprachondritic Pt/Pt*. The Pt enrichment has been attributed to stabilization of Fe–Pt alloys due to extreme melt depletion of the Kamchatka sub-arc mantle (Kepezhinskas et al., 2002), which could have been triggered by slab-fluid fluxing of the mantle wedge, thus simultaneously introducing radiogenic slab-derived Os. The positive correlation between ^{187}Os/^{188}Os and Ru/Ir in the Avachinsky peridotites is consistent with the known volatility of Os and Ru (but not other PGE) in the presence of high f_{O_2} and Cl-rich fluids (Wood, 1987), and supports the hypothesis that radiogenic Os is imparted to the mantle wedge by hydrous slab-fluid metasomatism. The depleted mantle range for Pt/Pt* is from Garuti et al. (1997a,b). The upper mantle Os isotopic composition is based on abyssal peridotite data (Snow and Reisberg, 1995; Brandon et al., 2000). The upper mantle Ru/Ir ratio (~ 1.27–1.42) is based on average carbonaceous chondrites (Jochum, 1996) and upper mantle peridotites (Chou et al., 1983; Barnes et al., 1988). PUM value from Meisel et al. (2001).

melts impart stronger metasomatic signatures in the Re–Os isotope system than either hydrous slab fluids or adakitic slab melts.

Unfortunately, there are no experimental data with which to directly evaluate the solubilities of Re and Os in carbonate-rich melts. The few existing Re–Os isotope analyses of carbonatites (from both continental and oceanic settings; Shank et al., 1992; Pearson et al., 1995b; Widom et al., 1999) indicate that they are characterized by extremely low Os abundances (<15 ppt), and relatively high Re abundances (400 ppt–2.4 ppb). It is likely, however, that these are not good analogues for deep slab-derived carbonate melts, as they may be generated by a distinctly different process (e.g. Yaxley and Green, 1996), and they may represent relatively evolved melts that have lost Os through fractionation processes. PGE studies of some continental xenolith suites in which carbonatite metasomatism has been inferred also indicate that PGEs and Re are not significantly mobilized during subcontinental carbonatite-type metasomatism (Handler et al., 1997; Handler and Bennett, 1999; Lorand and Alard, 2001). However, Wood (1987) suggests that in the C–O–H–Cl system, for a given high f_{O_2}, Cl-rich fluid, decreasing f_{H_2O} (thus increasing CO_2 and CO) causes increased solubility of PGEs. Hence, it might be expected that melts derived from carbonate-rich altered oceanic crust would be rich in PGE, explaining the significant radiogenic slab-derived Os in the Bakening peridotites.

Os abundances in the Bakening peridotites are generally significantly lower than those in Avachinsky and Valovayam (Fig. 5a), suggesting that carbonate-rich slab-melt metasomatism involves mobilization of pre-existing mantle wedge Os combined with exchange with radiogenic slab-derived Os. This may be attributed to highly oxidizing conditions in the mantle wedge resulting from carbonate-rich slab-melt metasomatism, similar to the process proposed above for adakitic melt metasomatism beneath Valovayam. Oxidation of arc mantle wedge in response to carbonatitic slab-melt metasomatism has been proposed based on high Ti in phlogopite and high Fe^{3+}/Fe^{2+} in clinopyroxene in mantle xenoliths from Simberi Island, Papua New Guinea (McInnes and Cameron, 1994). High f_{O_2} contents have also been documented in strongly melt-depleted mantle xenoliths from Tanzania that show chemical evidence for carbonatite melt metasomatism (Canil et al., 1994). These xenoliths have higher f_{O_2} than is observed in other continental mantle xenoliths that have been modally metasomatized by hydrous silicate melts and fluids, indicating that carbonatitic melts may be more effective oxidizing agents than other types of fluids and melts (Canil et al., 1994). Metasomatism and oxidation of the mantle wedge by carbonate-rich slab melts could therefore explain the lower Os abundances in most Bakening peridotites relative to both Avachinsky and Valovayam peridotites. Concomitant isotopic exchange with, and scavenging of Os by, highly oxidized and radiogenic carbonate-rich slab melts would produce both the radiogenic Os and relatively low Os abundances observed in the Bakening peridotites.

7. Conclusions

The Kamchatka peridotite mantle xenoliths exhibit a large range in Os and Re abundances, Re/Os ratios, and $^{187}Os/^{188}Os$ isotope signatures that overlap with and extend beyond the ranges found in most non-arc continental peridotites. Regional variations in the Kamchatka peridotite Re–Os isotope signatures are attributed to compositionally distinct slab fluids generated in the variable P–T regimes beneath the Kamchatka arc. The Re–Os isotope results are consistent with previous petrographic and trace element studies that documented distinct styles of metasomatism in Kamchatka mantle xenoliths: silicic slab-melt metasomatism associated with subduction of relatively hot, young (~ 15–25 Ma) oceanic crust in the northern arc front, hydrous slab-fluid metasomatism associated with subduction of colder, old (~ 100 Ma) oceanic crust in the southern arc front, and carbonate-rich slab-melt metasomatism in the southern segment behind the arc front, where the slab is deeper.

Harzburgites from Avachinsky have Os abundances comparable to typical non-arc continental mantle, but extremely low Re abundances due to severe melt depletion of the Kamchatka arc mantle wedge. Os isotope signatures are similar to or less radiogenic than PUM, but are more radiogenic than most non-arc continental mantle at comparable Re abundances. Scattered but positive correlations between $^{187}Os/^{188}Os$, La/Sm, and Ru/Ir in Avachinsky harzburgites support a model in which high f_{O_2}, Cl-rich, hydrous slab fluids transport LREE, Ru, and radiogenic Os into the mantle wedge beneath the southern arc front. Re is either not transported, or more likely is simply not retained, during fluid–mantle interaction. The Re–Os isotope compositions of the Avachinsky harzburgites are similar to those from Japan and Lihir, suggesting similar metasomatic processes in the arc mantle above relatively cold, old (~ 100–175 Ma) subducting slabs at depths of ~ 100–150 km.

Peridotites from Valovayam overlap with and extend to more radiogenic Os isotope compositions, and generally have lower Os abundances and higher Re abundances than those from Avachinsky. The higher Re and more radiogenic Os isotope signatures are attributed to significant transport of slab-derived Re and Os into the mantle wedge by highly oxidized, adakitic slab melts. The relatively radiogenic but low Os abundances in the Valovayam peridotites compared to Avachinsky peridotites are suggestive of both scavenging of mantle Os as well as exchange with radiogenic slab-derived Os during interaction of the mantle wedge with oxidized slab melts. Similar Re–Os isotope compositions are found in mantle peridotites from the Cascades. Both the Cascades arc and northern Kamchatka involve subduction of hot, young (~ 25 Ma) oceanic crust and are characterized by the generation of adakitic slab melts.

Bakening peridotites exhibit the most radiogenic Os and highest Re/Os ratios found so far in arc mantle peridotites. These results suggest that carbonate-rich slab melts can be important transport agents of both Re and Os into the sub-arc mantle wedge, despite previous studies based on non-arc peridotites that have concluded that carbonate-rich fluid metasomatism does not significantly affect PGE mantle abundances. Os abundances in Bakening peridotites extend to lower values than Avachinsky and Valovayam peridotites, again suggesting that scavenging of mantle Os by oxidizing, carbonate-rich slab melts causes mobilization of Os from localized regions within the mantle wedge, in addition to overprinting by radiogenic slab-derived Os.

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