

Re and Os isotopes of the central Oregon Cascades and along the arc indicate variable homogenization and mafic growth in the deep crust

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Abstract

The rhenium–osmium isotopic system has the potential to track the maturation of arc crust at lower crustal levels because the $^{187}\text{Os}/^{188}\text{Os}$ in magmas is sensitive to the age and composition of crust with which they interact. We here present Re and Os isotopic data for a suite of samples from the extensional central Oregon Cascade arc that includes diverse, primitive (high Mg) basalts and low- and medium-K basaltic andesites from North Sister and Little Brother volcanoes. The basaltic andesites contain higher $^{187}\text{Os}/^{188}\text{Os}$ than the two most common basalt types, calc-alkaline basalt and low-K tholeiites (0.17–0.19 vs. 0.14–0.15), indicating interaction with mafic crust. The evolution of $^{187}\text{Os}/^{188}\text{Os}$ in the deep crust depends on Re partitioning that in turn depends on sulfide and magnetite stability and oxygen fugacity. We estimate bulk mineral–melt partition coefficients for Re to be ~ 4 at $f\text{O}_2 \approx \text{QFM}$ and ~ 10 at $f\text{O}_2 \approx +1$ to $+3 \Delta\text{QFM}$ in keeping with near liquidus phase equilibria at deep crustal conditions (Mercer and Johnston, 2008) and on $f\text{O}_2$ of central Oregon primitive magmas (Rowe et al., 2009). Modeling fractional crystallization of the central Oregon primitive basalts indicates that the deep arc crust is likely to have $^{187}\text{Re}/^{188}\text{Os}$ similar to the range measured in North Sister basaltic andesites (~ 55 – 260). The age of the mafic crustal contaminant in the central Oregon Cascades is then estimated to be 9–29 Ma and is younger than the start of Cascade arc volcanism at ~ 45 Ma in central Oregon. The range in $^{187}\text{Os}/^{188}\text{Os}$ at the Cascade volcanic centers Mt. Lassen and Mt. Adams is greater than is observed for central Oregon arc magmas. We estimate a range in mafic crustal contaminant ages of 8–340 Ma for Mt. Lassen and 0–70 Ma for Mt. Adams and are older than suggested by previous studies (Hart et al., 2002, 2003). Mafic contaminant age ranges reflect the degree to which the crust is homogenized, or the extent to which the composition of the crust is a product of recent arc magmatism vs. pre-existing protoliths. Intra-arc extension in central Oregon facilitates homogenization and leads to assimilation of relatively young arc crust. Where extension is less narrowly focused, the range in Re and Os isotopes is greater and reflects isotopic heterogeneities, including variably aged crust at Mt. Lassen or variable enrichment of Re and Os in the lithosphere beneath Mt. Adams.

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1. INTRODUCTION

Mass and thermal fluxes from the mantle transform volcanic arc crust by gradually altering its structure, bulk composition, and mineral stability fields. Mature volcanic arcs are underpinned by ultramafic to gabbroic cumulates formed by differentiation of mantle-derived hydrous basalts (e.g., DeBari and Coleman, 1989; Holbrook et al., 1999; Müntener and Ulmer, 2006). As arcs mature, the character of the deep crust changes as diverse and variably slab-influenced mantle

melts overprint the crustal protolith. The rate and stage of maturation depends on the age and thermal history of the arc as well as the age, composition, and tectonic regime of the upper plate. These factors can vary greatly along the length of arcs, particularly in settings where the crust may consist of a patchwork of accreted terranes.

The influence of variable continental crust is reflected, for example, by Ce/Yb among andesites in the Southern Volcanic Zone of the Andean arc. Here, Ce/Yb correlates with crustal thickness and increasing garnet stability (Hildreth and Moorbath, 1988). The influence of the deep crust on magma compositions however may be cryptic where protracted injection of mantle melts reduces elemental and isotopic contrasts with the mantle.

The potential utility of the Re–Os isotopic system for unraveling crustal contributions to arc magmas is supported by typically higher $^{187}\text{Os}/^{188}\text{Os}$ in intermediate to silicic arc magmas than in associated primitive basalts (e.g., Lassiter and Luhr, 2001; Chesley et al., 2002; Hart et al., 2002). In particular, Re–Os systematics may shed light on the nature of the deep arc crust because $^{187}\text{Os}/^{188}\text{Os}$ is strongly influenced by assimilation of mafic lithologies. The parent isotope ^{187}Re ($t_{1/2} = 4.16 \pm 0.11 \times 10^{10}$ years; Shirey and Walker, 1998) behaves incompatibly during mantle melting and consequently partitions into the crust. Os, on the other hand, is highly compatible during crystallization of ultramafic minerals and typically correlates with indices of fractionation, such as Ni or MgO. The deep crust evolves to higher $^{187}\text{Os}/^{188}\text{Os}$ with time as it consists of gabbroic to ultramafic intrusions formed from mantle-derived, high Re/Os basaltic magmas and residual granulites after partial melt extraction. As primary basalts differentiate to more evolved compositions in the deep crust, fractional crystallization of mafic minerals strips the initial Os, and the $^{187}\text{Os}/^{188}\text{Os}$ is swamped by Os derived by assimilation of partial melts of the mafic crust (e.g., Asmerom and Walker, 1998; Lassiter and Luhr, 2001; Chesley et al., 2002). Variations in $^{187}\text{Os}/^{188}\text{Os}$ in magmas can thus give insight to the nature and age of the mafic crustal contaminants and how they might vary along strike of a volcanic arc. The Cascade arc is ideal to exploit the strength of the Re–Os isotope system because the continental crust is not evolved with respect to Nd and Sr isotope systematics, making it otherwise difficult to constrain the crustal contributions to magma compositions. In addition, the arc is segmented into discrete tectonic regimes and (or) crustal terranes (Couch and Riddihough, 1989; Wells et al., 1998; Schmidt et al., 2008).

In this paper, we present Re–Os isotopic data for the basaltic andesites that make up North Sister Volcano of the Three Sisters Volcanic Cluster and for nearby basalts of the central Oregon Cascade arc. Extension in this part of the arc has led to numerous volcanic vents (Guffanti and Weaver, 1988) and a preponderance of mafic magmatism, including primitive basalts ($>7\%$ MgO) that represent the primary inputs to the deep arc (Sherrod and Smith, 1990; Conrey et al., 1997; Schmidt et al., 2008; Rowe et al., 2009). Over the ~ 400 k.y. history of North Sister Volcano, it produced low-K basaltic andesitic magmas that systematically decrease in $^{87}\text{Sr}/^{86}\text{Sr}$ (from 0.70369 to

0.70356) and increase in $^{143}\text{Nd}/^{144}\text{Nd}$ (from 0.51285 to 0.51292). Continuous recharge by low-K mantle-derived basalts and interaction with deep crustal mafic lithologies are necessary to sustain the North Sister magma system (Schmidt and Grunder, 2009, 2011).

The Re–Os systematics of central Oregon Cascade magmas are then compared to Re and Os isotopic data for other parts of the arc, including Mt. Lassen in northern California and Mt. Adams in southern Washington (Borg et al., 2000; Hart et al., 2002, 2003; Jicha et al., 2009a). The overall goal of this work is to model the age of the mafic contaminant for the three centers. By comparing contaminant ages along the length of the Cascade arc, we attempt to understand the timeframes involved in maturation of the deep arc crust as a consequence of variable basaltic flux rates and age and composition of the upper plate.

1.1. The Cascade Arc

1.1.1. Tectonic framework

Cascadia is considered to represent a “hot” global end-member in the spectrum of subduction zones because the down going slab is young (~ 14 – 21 Ma beneath arc crest; Green and Harry, 1999) and relatively aseismic. Varying crustal domains and tectonic regimes in the upper plate (Fig. 1) correlate with many aspects of subduction, including slip along the interface with the down going plate (Brudzinski and Allen, 2007).

In northern California and southern Oregon, terranes of the Klamath block include Neoproterozoic basement, the Paleozoic Trinity ultramafic complex, and Paleozoic to Mesozoic marine arc-related volcanic, plutonic, and sedimentary rocks accreted to North America by the Late Triassic (Irwin, 1981; Snoke and Barnes, 2006). The forearc of central Oregon to southern Washington lies within the Columbia Embayment, a region defined by positive Bouguer anomalies and whose boundary delineates the Early Eocene margin of North America (Couch and Riddihough, 1989). The largest, most coherent crustal block of the Columbia Embayment is the Siletz Terrane, an Early Paleogene oceanic plateau that varies in thickness from 10 to 15 km in southern Washington to 30 km in Oregon (Stanley et al., 1990; Trehu et al., 1994; Parsons et al., 1999) and likely underlies the Quaternary arc in southern Washington and northern Oregon (Trehu et al., 1994; Parsons et al., 1999). Siletz crust is compositionally distinct in terms of its radiogenic isotopes, and its influence has been charted by stepwise variations in Pb-isotopic ratios in sulfide ore deposits (Church et al., 1986) and Sr- and Nd-isotopic ratios in primitive arc basalts (Schmidt et al., 2008). Farther north along the subduction zone, the North Cascades continental crust consists of Paleozoic to Mesozoic oceanic and arc-related rocks that were accreted during the Middle Cretaceous (Brown, 1985; Whitney and McGroder, 1989; Barnes et al., 1992).

Paleomagnetic and geodetic surveys indicate the Oregon and California Coast Range has rotated counterclockwise with attendant extension in the arc where it borders the Basin and Range Province, from northern California and propagating to the intra-arc High Cascade graben of Ore-

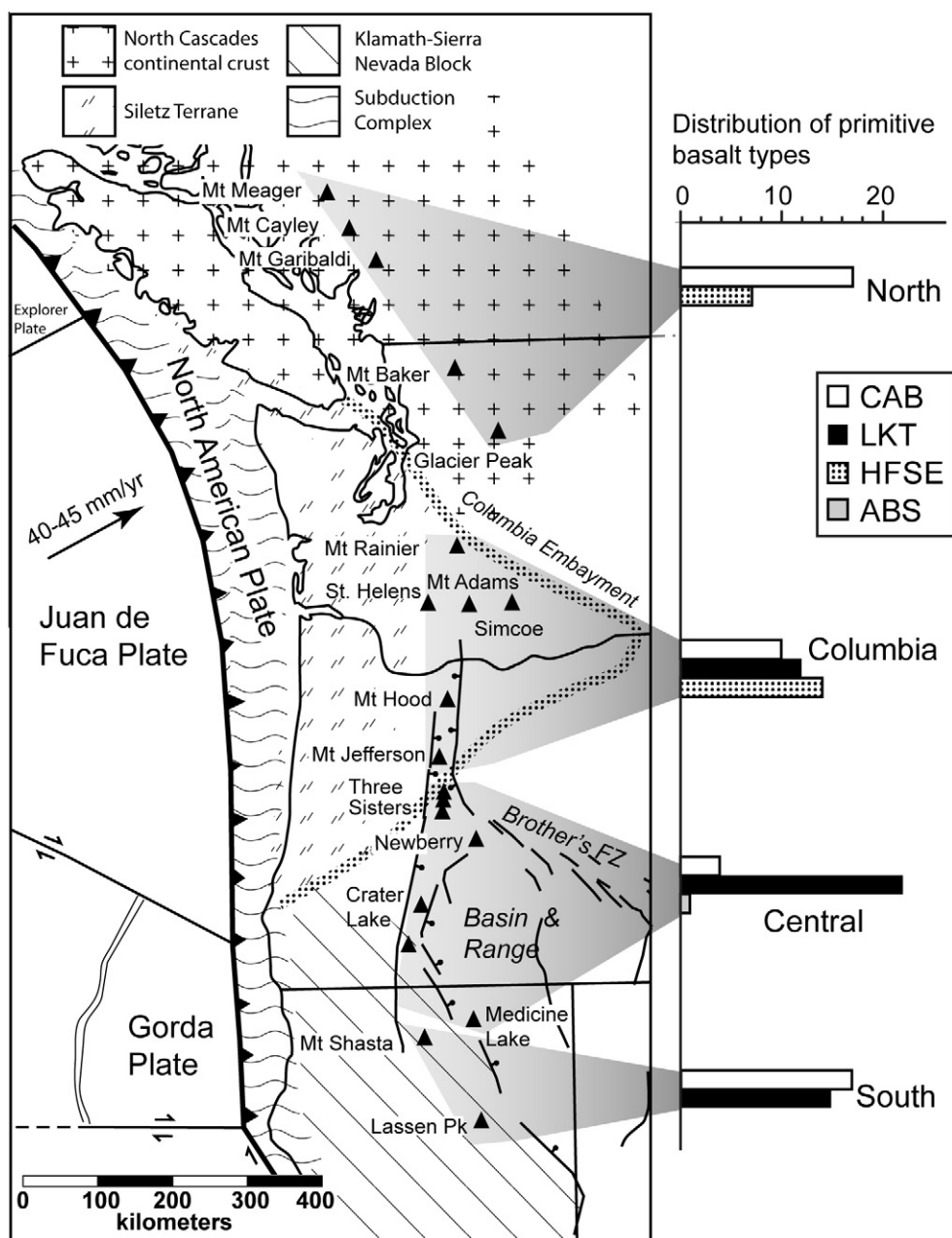


Fig. 1. Map of the Cascadia Subduction zone with important terranes and tectonic provinces indicated (Couch and Riddihough, 1989; Trehu et al., 1994; Wells et al., 1998). The Three Sisters Volcanic Cluster includes North Sister on the north side and Little Brother Volcano on the western flank of North Sister. Gray fields correlate to arc segments defined by $^{87}\text{Sr}/^{86}\text{Sr}$ and histograms indicate occurrence of primitive basalt types (Schmidt et al., 2008).

gon and southern Washington (Hughes and Taylor, 1986; Wells et al., 1998; Wells and Simpson, 2001). The Three Sisters Volcanic Cluster lies where the northwestern margin of the Basin and Range Province defined by the dextral Brother's Fault Zone merges with the High Cascade graben (Fig. 1). To the north, rotation of the Siletz block has contributed to compression of the arc (Wells et al., 1998).

Differences in extensional vs. compressional regime correspond with differences in volcanism along the Cascade arc. In northern California, extension combined with high flux rates from the mantle has led to the construction of

large stratovolcanoes surrounded by productive mafic fields. Extension of the Oregon arc has allowed anomalously abundant of mafic magmas to reach the surface (Sherrod and Smith, 1990). The southern Washington centers Mount St. Helens, Indian Heaven volcanic field, Mt. Adams, and Simcoe volcanic field make up a notable east-striking line of vents that run contrary to the regional fabric to the south. Farther north in Washington and British Columbia, the arc is undergoing compression and distributed stratovolcanoes erupt mainly intermediate to silicic magmas (Hildreth, 2007).

1.1.2. Petrologic framework

An array of primitive basaltic magmas (here defined as containing $>7\%$ MgO) is found in the Cascade Arc (e.g., Bacon et al., 1997; Conrey et al., 1997; Leeman et al., 2005; Schmidt et al., 2008). The composition and distribution of the various basalt types reflect the tectonic framework and mantle source composition (Fig. 1). As these mantle melts rise and differentiate, they modify the arc crust (e.g., Hildreth and Moor bath, 1988; Annen et al., 2006). Basaltic andesites are more abundant than basalts in the Cascades (Sherrod and Smith, 1990) and vary in composition from low- to high-K (0.56–2.2 wt% K_2O).

Four primitive basalt types, abbreviated CAB, LKT, HFSE-rich, and ABS are defined by major and trace element abundances (Table 2 in Schmidt et al., 2008; Electronic Supplement I). The CAB are large ion lithophile element (LILE)-enriched calc-alkaline basalt (also called arc basalt by Bacon et al., 1997). The LKT are incompatible element-poor low K tholeiites (also called high alumina olivine tholeiites, HAOT, by some workers; Bacon et al., 1997; Grove et al., 2002). The HFSE-rich are high field strength element-enriched; these are also called ocean island basalt, OIB, within plate basalts, WIP, or enriched intraplate basalt, EIB by other workers (Leeman et al., 1990; Bacon et al., 1997; Hildreth, 2007; Rowe et al., 2009). The ABS are rare, alkali-super enriched absarokite (Conrey et al., 1997). Mg-rich basaltic andesites and andesites (MgO >7.5 wt%) are considered primary melts by Baker et al. (1994) and Grove et al. (2002), but not by Streck et al. (2007). For this reason, the isotopic survey presented here does not focus on Mg-rich basaltic andesites and andesites.

CABs, distinguished by their lower FeO^*/MgO and higher alkali concentrations than tholeiitic basalts, typify subduction zones and carry the integrated geochemical signals of processes that involve dehydration of the subducting plate (Nye and Reid, 1986). The CABs are the most widely distributed basalt type along the entire Cascade arc crest. Concentrations of LILEs vary among the CABs and tend to be lowest in southern and central Oregon (0.5–0.9 wt% K_2O) and highest in southern Washington (up to 2.2 wt% K_2O). LKT magmas with their slight enrichments in fluid-mobile elements and near-flat chondrite-normalized rare earth element (REE) patterns are thought to form by decompression-induced melting of a subduction fluid-modified depleted mantle (Bacon et al., 1997; Conrey et al., 1997). LKTs are particularly abundant in parts of the Cascade arc that are undergoing extension, that is, from California to southern Washington. HFSE-rich basalts have high concentrations of Nb (>20 ppm), Ce/Yb (14–50), and low Al_2O_3/TiO_2 and likely reflect derivation from an enriched mantle source. The distribution of HFSE-rich basalts is geographically restricted to the northern half of the arc, particularly where it lies within the Columbia embayment in southern Washington (Fig. 1B; Schmidt et al., 2008).

On the basis of primitive basalt occurrence and stepwise distributions in Sr and Nd isotopic ratios, Schmidt et al. (2008) defined four segments of the Cascade arc. The isotopic segments are: (1) the dominantly calc-alkaline North

Segment from Mt. Meager to Glacier Peak ($^{87}Sr/^{86}Sr = 0.7030$ – 0.7036); (2) the Columbia Segment from Mt. Rainier to Mt. Jefferson where more primitive basalts tend to be of the HFSE-rich type (enriched in Nb; $^{87}Sr/^{86}Sr = 0.7028$ – 0.7037); (3) the extensional Central Segment from the Three Sisters to Medicine Lake, where primitive basalts have more restricted compositions ($^{87}Sr/^{86}Sr = 0.7034$ – 0.7038) and LKTs dominate, and (4) the South Segment from Mt. Shasta to Mt. Lassen, where high Ba/Ce (6.6–22.9) and $^{87}Sr/^{86}Sr$ (0.7030–0.7044) suggest fluid enrichment.

2. SAMPLES AND ANALYTICAL TECHNIQUES

The range of basaltic andesites is represented by four samples of crystal-poor, low-K basaltic andesite (~ 0.6 wt% K_2O) from North Sister Volcano (~ 400 – 55 ka), and one medium-K basaltic andesite (~ 1.0 wt% K_2O) from the nearby Little Brother Volcano (~ 153 ka; Schmidt and Gruner, 2009). The central Oregon primitive magmas were sampled from Brunhes age monogenetic centers (Conrey et al., 2004) to represent diverse mantle sources. Whole rock compositions determined by X-ray Fluorescence (XRF) and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) for these samples are provided in Table 1 (Schmidt and Gruner, 2011). The primitive basalts contain >7 wt% MgO and have a range in K_2O concentrations (0.4–3.1 wt%). The basalts from Cayuse Crater, Foley Ridge, and Quartzville belong to the CAB, LKT, and ABS types and are of the Central Segment of Schmidt et al. (2008). Also, one sample with both CAB and HFSE-rich basalt affinities (CAB/HFSE) from the Wizard Falls scoria cone was collected ~ 40 km north of the other samples (closer to Mt. Jefferson) and belongs to the Columbia Segment. The mineral, elemental, and Sr, Nd, and O isotopic compositions of these samples have been characterized (Rowe et al., 2006, 2009; Schmidt et al., 2008; Schmidt and Gruner, 2011).

Osmium and rhenium isotopes were analyzed at the University of Arizona in 2005 using the techniques of Chesley et al. (2002). Whole rock samples were wrapped in newspaper during hammering and chips were taken from the center of the rock to avoid metal contamination by the sledge and metal plate. Hammered chips were then powdered using an alumina jaw crusher and shatterbox. Between 7 and 15 g. of powdered sample were dissolved and spiked with ^{185}Re and ^{190}Os . Equilibration with the spike was achieved by using a modified Carius tube technique (Shirey and Walker, 1995). Using a modified organic separation method (Cohen and Waters, 1996 and Chesley and Ruiz, 1998), Os was separated and purified with $\sim 90\%$ recovery. Measured Os total process blanks are fairly stable and range from 0.5 to 1.2 pg over several years. A mean of 0.08 pg Os blank with a measured $^{187}Os/^{188}Os = 0.175 \pm 0.015$ was used for correction. Re blanks were 4–10 pg. Loading blanks are 0.010–0.050 pg. for Os and 0.040–0.150 pg Re. Two sigma errors and percent blank corrections are presented in Table 2. Purified Os and Re were analyzed using BaOH and BaSO₄ as emission enhancers on platinum filaments, respectively. Samples were analyzed utilizing negative thermal ionization

Table 1
Central Oregon Cascade elemental and Sr, Nd, and Pb isotopic compositions.

Sample #	CC02-1	FLR03-1	QV03-1	WF02 1	NS02-29	NS02-46	NS02-66	NS02-111 ^a	LB02-2
Loc.	Cayuse Crater	Foley Ridge	Quartz-ville	Wizard Falls	North Sister	North Sister	North Sister	North Sister	Little Brother
Magma type	CAB	LKT	ABS	CAB-HFSE	Low-K BA	Low-K BA	Low-K BA	Low-K BA	Med-K BA
<i>wt%</i>									
SiO ₂	51.94	49.08	50.06	49.09	53.4	53.8	52.5	54.7	52.8
Al ₂ O ₃	16.53	16.94	13.64	15.91	18.8	19.7	18.4	18.7	17.7
TiO ₂	1.053	1.510	1.581	1.588	1.08	1.16	1.20	1.03	1.41
FeO ^{ab}	7.85	9.57	6.53	9.78	7.6	7.4	8.1	7.1	8.5
MnO	0.150	0.175	0.105	0.179	0.13	0.13	0.14	0.13	0.15
CaO	9.15	9.83	10.14	9.44	9.1	9.1	9.1	8.5	8.7
MgO	8.62	9.03	9.86	10.00	5.4	4.0	5.9	5.1	5.6
K ₂ O	0.65	0.40	3.11	0.39	0.66	0.70	0.66	0.78	0.98
Na ₂ O	3.13	3.07	2.88	2.91	3.7	4.0	3.7	3.8	3.7
P ₂ O ₅	0.221	0.332	1.173	0.436	0.19	0.20	0.26	0.21	0.47
<i>ppm</i>									
Ni	153	176	256	235	75	35	98	83	71
Cr	411	359	436	349	56	30	54	71	97
V	185	229	140	184	186	199	179	167	195
Ba	224	231	2242	222	248	250	274	299	410
Th	1.27	0.98	7.97	1.23	0.87	0.86	1.00	3	1.71
Nb	6.67	7.39	7.23	10.04	5.0	5.0	7.1	6	13.1
Y	20.49	27.82	20.60	27.49	20.0	20.6	22.1	18	27.1
Hf	2.41	3.00	8.31	3.69	2.50	2.57	2.82		3.84
Ta	0.42	0.51	0.37	0.63	0.31	0.86	0.44		0.79
U	0.44	0.23	2.18	0.33	0.33	0.34	0.39		0.58
Cu	58	59	69	38	78	86	73	65	47
Zn	74	76	98	86	71	69	75	73	89
Pb	2.76	2.27	13.63	2.06	3.03	3.05	3.20	1	4.23
Rb	10.9	4.3	26.9	2.3	7.2	7.1	8.9	9	12.8
Cs	0.39	0.20	0.20	0.07	0.24	0.25	0.30		0.30
Sr	431	398	3124	701	582	609	579	596	647
Sc	31.3	35.7	23.5	35.4	27	26	27	21	25
Zr	93	116	336	156	93	95	111	111	155
La	10.0	11.8	83.9	17.4	9.7	9.7	11.6	17	20.1
Ce	21.31	25.95	184.60	39.03	21	21	25	14	42.3
Pr	2.76	3.58	24.35	5.13	2.7	2.8	3.2		5.4
Nd	12.48	16.37	100.09	22.54	12.6	13.4	14.6		23.7
Sm	3.39	4.58	17.49	5.41	3.6	3.7	4.0		5.8
Eu	1.23	1.64	4.59	1.84	1.29	1.33	1.42		1.85
Gd	3.66	4.95	10.35	5.37	3.7	3.9	4.0		5.5
Tb	0.61	0.82	1.11	0.85	0.61	0.64	0.66		0.86
Dy	3.77	5.18	4.85	5.22	3.7	3.8	4.1		5.1
Ho	0.78	1.05	0.74	1.03	0.74	0.79	0.85		1.04
Er	2.10	2.88	1.64	2.78	2.0	2.1	2.2		2.8
Tm	0.30	0.40	0.20	0.40	0.28	0.30	0.32		0.39
Yb	1.84	2.56	1.07	2.47	1.72	1.80	1.88		2.39
Lu	0.29	0.40	0.16	0.39	0.27	0.29	0.30		0.37
⁸⁷ Sr/ ⁸⁶ Sr	0.70354	0.70356	0.70382	0.70317	0.70357	0.70356	0.70369	0.70358	0.70376
¹⁴³ Nd/ ¹⁴⁴ Nd	0.51294	0.51292	0.51290	0.51296	0.51290	0.51292	0.51285	0.51287	0.51287
²⁰⁶ Pb/ ²⁰⁴ Pb	18.949	18.926	18.884	18.873	18.868	18.826	18.913	18.913	18.940
²⁰⁷ Pb/ ²⁰⁴ Pb	15.623	15.591	15.552	15.595	15.596	15.585	15.957	15.604	15.604
²⁰⁸ Pb/ ²⁰⁴ Pb	38.623	38.559	38.428	38.530	38.506	38.451	38.569	38.580	38.589

XRF major element analyses were normalized to 100%. Whole-rock powders were analyzed at the Washington State University GeoAnalytical lab by XRF only for NS-02-111^a and by XRF and ICP-MS for all other samples. Major and trace element and Sr and Nd isotopic compositions for the primitive basalts (CAB, LKT, ABS, CAB-HFSE) basaltic andesite analyses are from Schmidt et al. (2008) and Schmidt and Gruner (2011), respectively. ^bTotal Fe is expressed at FeO*.

mass spectrometry (NTIMS) on a Micromass Sector 54 mass spectrometer. Data were reduced using the methodology and spreadsheets developed at the Carnegie Institute,

Department of Terrestrial Magnetism (e.g., Pearson et al., 1995). We have been significantly concerned about low level samples with regard to reproducibility, sample heterogene-

ity and the possible “nugget affect.” In order to test our analytical technique, we have repeatedly measured two very low level samples of arc basalt from Western Mexico (KR-411 and KR-415), which contain Os concentrations of 0.003 and 0.0003 ppb, respectively. The samples were quite reproducible and have $^{187}\text{Os}/^{188}\text{Os} = 0.4064 \pm 0.0008$ ($n = 3$) and 0.3413 ± 0.012 ($n = 5$), respectively.

Table 1 also includes Pb isotopic data that were analyzed at the University of Colorado. Pb isotopic analyses were four-collector static mode measurements. Sixteen measurements of SRM-981 during the study period yielded $^{208}\text{Pb}/^{204}\text{Pb} = 36.56 \pm 0.03$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.449 \pm 0.008$, $^{206}\text{Pb}/^{204}\text{Pb} = 16.905 \pm 0.007$ (2σ). Measured Pb isotope ratios were corrected to SRM-981 values ($^{208}\text{Pb}/^{204}\text{Pb} = 36.721$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.491$, $^{206}\text{Pb}/^{204}\text{Pb} = 16.937$). Total procedural blanks averaged ~ 1 ng for Pb during the analytical period.

3. RESULTS

Os behaves as a compatible element among the central Oregon arc magmas; Os concentrations are higher (0.05–0.13 ppb) in the high-Ni primitive basalts than in the basaltic andesites with less than 100 ppm Ni (0.004–0.013 ppb Os; Fig. 2A). In contrast, Re behaves inversely with respect to Ni (Fig. 2B); the highest Re concentration is found in the alkali-poor LKT and the lowest Re in the alkali-rich ABS. Re best correlates with V (Fig. 2C), suggesting that oxygen fugacity and/or oxide minerals affect Re concentrations. Although there is a weak correlation between Re and TiO_2 for some of the central Oregon magmas (CAB, LKT, and low- and medium-K basaltic andesites), the ABS and CAB/HFSE samples fall off the trajectory with lower Re at high TiO_2 (~ 1.58 wt%; Fig. 2D). Zn and Re are similarly volatile and high Zn in the low-Re ABS sample is evidence against volatile loss (Fig. 2F; Richter et al., 2008).

The range in $^{187}\text{Os}/^{188}\text{Os}$ for central Oregon Cascade magmas is relatively small, but important distinctions are identified between the different magma groups. Basaltic andesites from both North Sister (low K) and Little Brother (medium K) have higher $^{187}\text{Os}/^{188}\text{Os}$ (0.17–0.19) than determined for regional primitive basalts (0.14–0.16; Fig. 3A).

As such, the $^{187}\text{Os}/^{188}\text{Os}$ increases with decreasing compatible elements Ni and Os (Fig. 3A and B). The lowest $^{187}\text{Os}/^{188}\text{Os}$ is found in the LKT and CAB basalt types that predominate in this part of the arc (0.14–0.15). These basalts contain higher Os isotopic ratios than typical mid-ocean ridge basalts (~ 0.125 ; Snow and Reisberg, 1995; Brandon et al., 2000). Interestingly, the alkali-rich ABS and relatively Nb-rich CAB/HFSE have higher $^{187}\text{Os}/^{188}\text{Os}$ and Ni than the more common basalt types, suggesting derivation from a separate, more enriched source (Fig. 3A).

Os isotopes are decoupled from other isotopic systems (Fig. 3D and E) and track different processes. Unlike for Nd- and Sr-isotope ratios, there is no apparent temporal trend in Re–Os isotopes for North Sister magmas (cf. Schmidt and Grunder, 2011). The Little Brother medium-K basaltic andesite is isotopically distinct from those from North Sister, having slightly lower $^{187}\text{Os}/^{188}\text{Os}$ and higher $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{206}\text{Pb}/^{204}\text{Pb}$. The differences in Os and Pb isotopic ratios between the North Sister magmas and the primitive LKT and CAB basalts suggest involvement of another (high $^{187}\text{Os}/^{188}\text{Os}$ and low $^{206}\text{Pb}/^{204}\text{Pb}$) component that is not detected by the Sr and Nd isotopic systems.

4. DISCUSSION

4.1. Comparison with other Cascade arc magmas

Re and Os data have been published for lavas from Mt. Lassen and Mt. Adams, and harzburgitic xenoliths from Simcoe Volcanic Field (Brandon et al., 1996, 1999; W.K. Hart et al., 1997; G.L. Hart et al., 2002, 2003; Borg et al., 2000; Jicha et al., 2009a). Central Oregon lavas contain concentrations of Re and Os comparable to other arc lavas (Electronic Supplement II). Os ranges to particularly low concentrations in Mt. Adams magmas, leading to higher Re/Os than found elsewhere in the Cascade arc (up to 219 in basaltic lavas; Fig. 4; Hart et al., 2003; Jicha et al., 2009a).

Central Oregon Cascade basalts and basaltic andesites encompass a more narrow range in $^{187}\text{Os}/^{188}\text{Os}$ (0.14–0.19) than has been determined for other parts of the Cascade arc (0.13–0.63; Fig. 5A; W.K. Hart et al., 1997; G.L. Hart et al., 2002, 2003; Borg et al., 2000; Jicha et al., 2009a,b).

Table 2
Os isotopic analyses for Central Oregon Cascade magmas.

Sample #	Magma type	Re (ppb)	Os (ppb)	$^{187}\text{Re}/^{188}\text{Os}$	$^{187}\text{Os}/^{188}\text{Os}$	2σ Error	Blank correction (%)
CC-02-1	CAB	0.074	0.1131	3.15	0.1414	0.0008	0.17
CC-02-1 ^a	CAB	0.076	Undspk ^b	Undspk ^b	0.1416	0.0002	0.13
FLR-03-01	LKT	0.259	0.0553	22.67	0.1459	0.0003	0.31
QV-03-1	ABS	0.007	0.0465	0.750	0.1588	0.0013	0.34
WF-02-1	CAB-HFSE	0.125	0.1311	4.63	0.1590	0.0004	0.11
NS-02-29	Low-K BA	0.138	0.0114	59.01	0.1732	0.0005	1.03
NS-02-46	Low-K BA	0.206	0.0038	262.7	0.1822	0.0036	2.46
NS-02-66	Low-K BA	0.186	0.0096	94.75	Overspk ^c		1.85
NS-02-111	Low-K BA	0.145	0.0127	55.51	0.1858	0.0007	1.16
LB-02-2	Med-K BA	0.255	0.0128	96.41	0.1662	0.0013	0.89

^a Duplicate analysis.

^b Underspike with ^{185}Re and ^{190}Os occurred during sample processing.

^c Overspike with ^{185}Re and ^{190}Os occurred during sample processing.

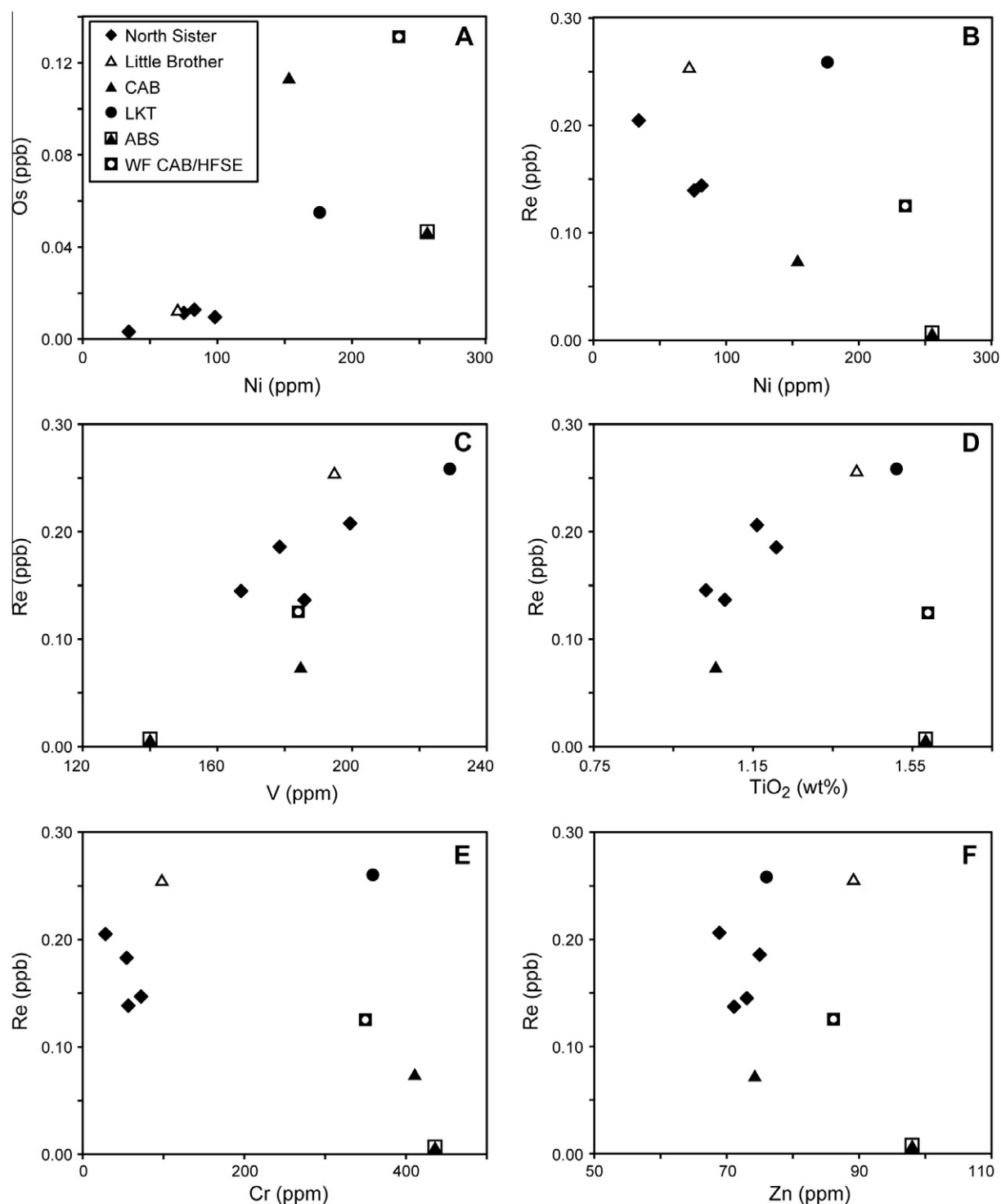


Fig. 2. Re and Os concentration variation diagrams for the central Oregon Cascade arc magmas (Tables 1 and 3; Schmidt and Gruner, 2011). North Sister and Little Brother are low-K and medium-K basaltic andesites, respectively. (A) Os vs. Ni; (B) Re vs. Ni; (C) Re vs. V; (D) Re vs. TiO₂; (E) Re vs. Cr; (F) Re vs. Zn.

The range is also very narrow when compared to other volcanic arcs (Fig. 5B). Re and Os compositions for more silicic magmas from Central Oregon were not determined and so the true range may be greater than indicated by our small sampling. However, the range of the primitive

basalts at Mt. Adams is eight times the range of all central Oregon magmas analyzed (0.17–0.56; Hart et al., 2003; Jicha et al., 2009a). Metasomatized harzburgite xenoliths from Simcoe Volcanic Field may represent the mantle lithosphere of the southern Washington Cascades backarc

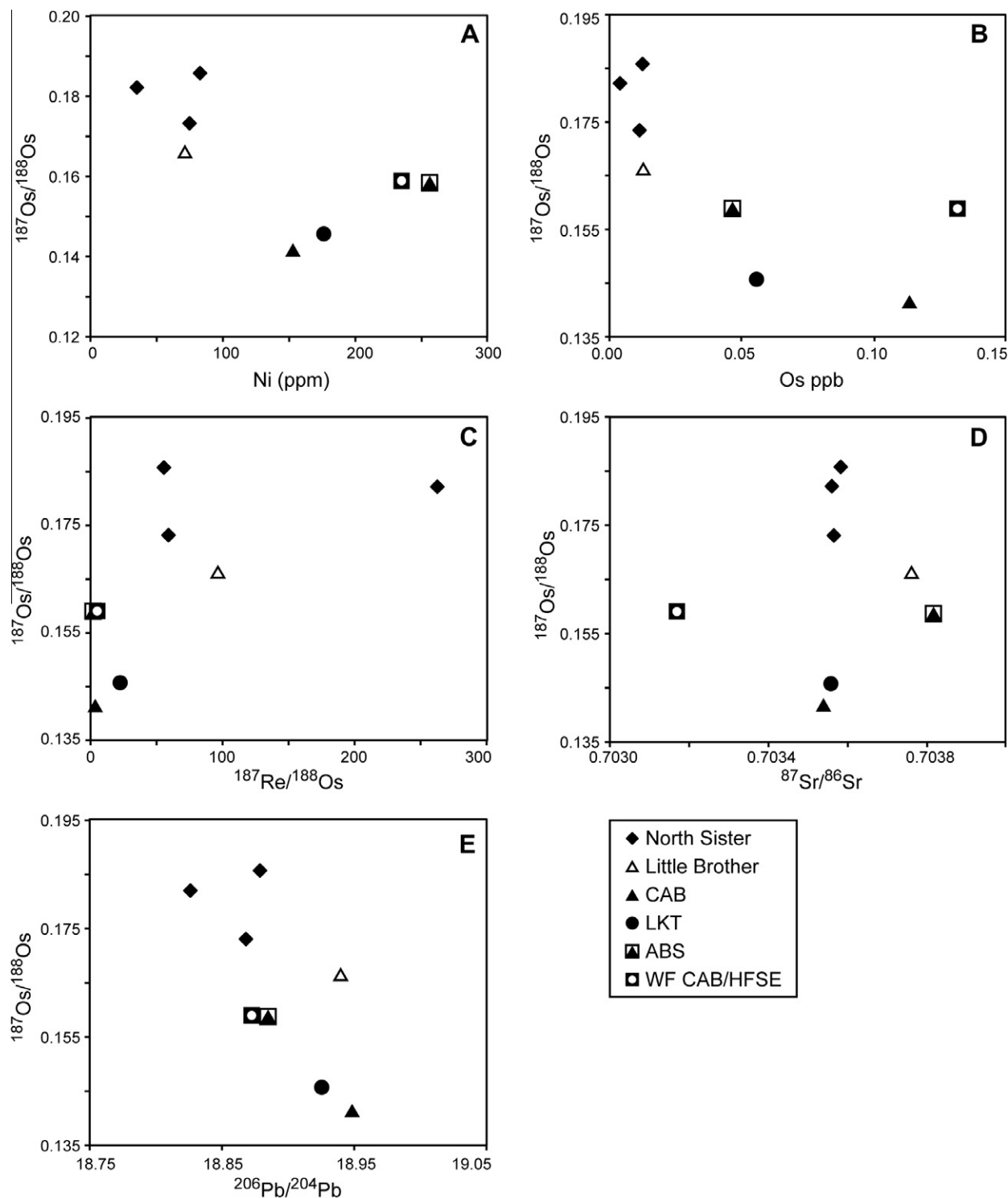


Fig. 3. Variations in Os and Re isotopic compositions for the central Oregon Cascade arc magmas (Tables 1 and 3; Schmidt and Grunder, 2011). (A) $^{187}\text{Os}/^{188}\text{Os}$ vs. Ni; (B) $^{187}\text{Os}/^{188}\text{Os}$ vs. Os concentration; (C) $^{187}\text{Os}/^{188}\text{Os}$ vs. $^{187}\text{Re}/^{188}\text{Os}$; (D) $^{187}\text{Os}/^{188}\text{Os}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$; (E) $^{187}\text{Os}/^{188}\text{Os}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$.

(Brandon et al., 1996, 1999) and contain lower $^{187}\text{Os}/^{188}\text{Os}$ (0.12–0.15) than has been observed in most arc lavas.

While having restricted Os-isotope ratios compared to other Cascade suites, the central Oregon basalt and basaltic andesite lavas generally lie in the region of overlap between

the Mt. Lassen and Mt. Adams suites, with closer affinity to the mafic suite of Mt. Lassen (Fig. 6). For example, with respect to Ba/Ce, a proxy for subduction fluid enrichment of the mantle source, and with respect to Nd-isotopes (Fig. 6C and D), the central Oregon suite allies with Mt. Lassen, ex-

cept for the HFSE basalt, which has affinity with HFSE basalts from Mt. Adams. Lassen has no HFSE basalt.

In isotope–isotope variation diagrams, the majority of primitive basalts including the central Oregon samples, fall in a band defined by the cluster of HFSE data from Mt. Adams and the CAB of Mt. Lassen. Four out of the 19 data do not conform: three from Mt. Adams (two LKT, one CAB) and one LKT from Mt. Lassen. The intersection of the $^{207}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ isotopic array for Simcoe xenoliths (Brandon et al., 1999) ranges to more radiogenic values than any of the arc lavas, but overlaps that of HFSE basalts and most other basalts from Mt. Adams (Fig. 6F). CABs are generally displaced to higher $^{207}\text{Pb}/^{204}\text{Pb}$ than LKT and HFSE and the basalt array is steeper (higher $^{207}\text{Pb}/^{204}\text{Pb}$ at given $^{206}\text{Pb}/^{204}\text{Pb}$) than the xenolith array. Coupled with higher Ba/Ce for CAB in general, this supports the interpretation that the CAB Pb source is linked to a subducted sediment component, either modern or ancient (Bacon et al., 1997).

The restricted range in Os isotope ratios for the central Oregon part of the arc resembles the narrow range observed in Sr isotope ratios (Schmidt et al., 2008). The Sr-isotope ratios of silicic volcanic rocks are also low (0.7032–0.7038; Hildreth, 2007; Schmidt et al., 2008) and almost completely overlap with the range among mafic rocks (0.70315–0.70385). In other segments of the arc, the range of Sr-isotope data among silicic rocks overlaps the high end of the range defined by mafic rocks (Hildreth, 2007) indicating a crustal addition to silicic magmas. In the Oregon segment, any crustal interaction in the genesis of silicic magmas would have to be with an isotopically primitive crust.

A subtle indication of the influence of a mafic crust on mafic magmatism is the slight displacement of basaltic andesites to higher $^{187}\text{Os}/^{188}\text{Os}$ relative to ambient basalts in the basalt band (Fig. 6). Also, detailed modeling of iso-

topic and elemental variation in the recharge-dominated North Sister Volcano (Schmidt and Grunder, 2011) calls for involvement of interaction with deep crustal mafic lithologies. We therefore explore crustal effects on Re–Os systematics.

4.2. The evolution of Re and Os in arc crust

In order for intermediate and silicic magmas to attain higher Os isotopic ratios than associated primitive magmas, the crust must evolve to comparably high $^{187}\text{Os}/^{188}\text{Os}$ over timescales less than the age of the crust itself. In central Oregon, this is supported by a lack of correlation between osmium and strontium isotopes (Fig. 3D) and a slight inverse correlation between osmium and lead isotopes (Fig. 3E). The Cascade arc crust is largely composed of accreted oceanic terranes with igneous lower to mid-crust. Juvenile basaltic crust refers to the newly added component (e.g., mafic plutons) since the inception of arc magmatism. We suggest that at first, the Cascade crust contained $^{187}\text{Os}/^{188}\text{Os}$ values that were comparable to regional mantle-derived melts. The goal of this discussion is to constrain how long it takes for ^{187}Os to build up in the crust by decay of ^{187}Re , and how timescales for this process vary along the length of the Cascade arc.

A global correlation between Re and oxidation state in primitive arc basalts suggests that diminished stability of sulfide minerals under oxidizing conditions ($f\text{O}_2 > \Delta\text{QFM}$) controls the transport of Re in the mantle wedge (Fig. 7A; Richter et al., 2008). As molecular water in arc melts correlates with oxidation state (e.g., Kelley and Cottrell, 2009), partitioning of Re into the crust by partial melting of the mantle is likely enhanced at water-rich and low sulfide conditions. Metasomatism by slab-derived fluids is unlikely to significantly influence the $^{187}\text{Os}/^{188}\text{Os}$ of the mantle wedge (Chesley et al., 2004) and is not supported by the Cascades basalt dataset (i.e., no correlation between $^{187}\text{Os}/^{188}\text{Os}$ and Ba/Ce, a proxy for slab fluid; Fig. 6C).

The rate of $^{187}\text{Os}/^{188}\text{Os}$ growth in deep arc crust depends on the process(es) and rate of ^{187}Re and Re/Os enrichment in mafic arc crust and is critical for assessing its age. Two limiting case models may be used to describe Re/Os enrichment: (1) continuous or (2) single-stage enrichment. The continuous enrichment model was developed by Hart et al. (2002) for Lassen arc magmas and suggests that Re/Os of the crust may be as high as 2500 via assimilation and fractional crystallization (AFC) processes (such as developed by dePaolo, 1981). In the continuous enrichment case, the lower arc crust may acquire high $^{187}\text{Os}/^{188}\text{Os}$ after only a few million years. Such high Re/Os values are rarely observed in evolved arc magma suites (e.g., Alves et al., 2002; Chesley et al., 2002). Whole rock Re/Os for central Oregon arc magmas are much lower than the extreme values suggested for Lassen (Fig. 4). The highest Re/Os recorded for magmas in the Cascade arc is found at Mt. Adams where primitive LKT magmas range to 219 (Hart et al., 2004; Jicha et al., 2009a,b). In addition, Trans-Mexican arc mafic granulite xenoliths that represent samples of the lower crust contain Re/Os < 80 (Valencia, 2005). These

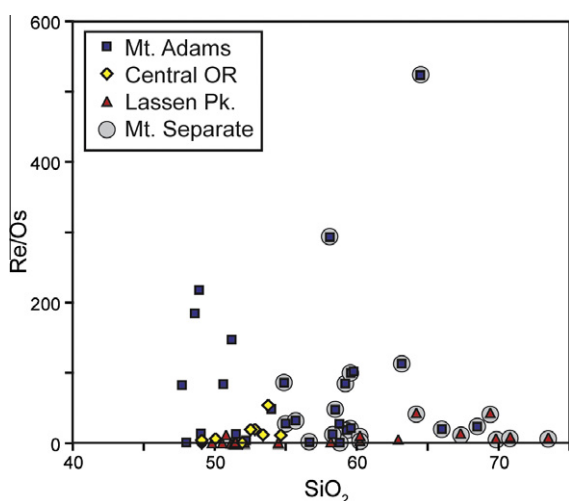


Fig. 4. Plot of Re/Os vs. SiO_2 for the Cascade arc magmas. Samples where Re and Os were determined for separated magnetite are indicated. Data are from this study, Bacon et al. (1997), Borg et al. (2000), W.K. Hart et al. (1997), G.L. Hart et al. (2002, 2003), and Jicha et al. (2009a,b).

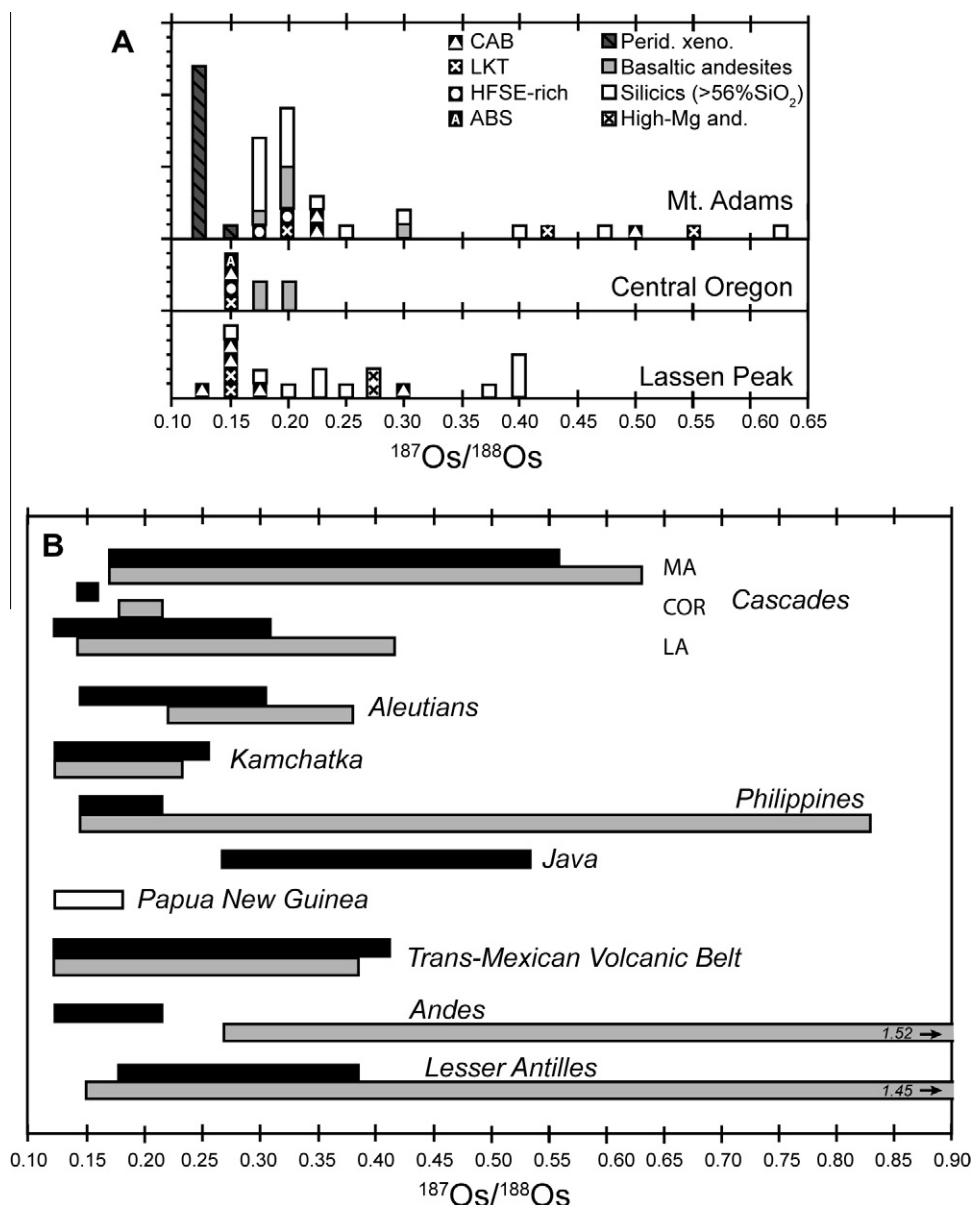


Fig. 5. (A) Histogram demonstrating the range in $^{187}\text{Os}/^{188}\text{Os}$ data along the Cascade arc. Data are from this study; Borg et al. (2000), Brandon et al. (1996, 1999), W.K. Hart et al. (1997), G.L. Hart et al. (2002, 2003) and Jicha et al. (2009a,b). (B) Ranges in $^{187}\text{Os}/^{188}\text{Os}$ in global arc magmas. Black and white bars represent ranges in basaltic (<52 wt% SiO₂) and more silicic magmas respectively. Magmas from Papua New Guinea are of unknown composition. Data are from Lassiter and Luhr (2001) and Alves et al., 2002.

real world cases suggest that arc lower crust is not continuously enriched in Re/Os and single stage enrichment is a more likely limiting case.

In single stage Re/Os enrichment, Os isotopic ratios reflect a single event of mantle separation and crystallization (e.g., Esperança et al., 1997). In reality, the mafic lower arc crust is likely constructed of many mafic and ultramafic intrusions that each represents single mantle separation and crystallization events. By preferring the single stage enrichment, we acknowledge that our construct integrates multiple intrusive events.

In subsequent sections, we develop a model to calculate the age of the mafic crustal contaminant by (1) discussing

the important mineral players as a function of oxygen fugacity and assigning bulk partition coefficients for Re and Os; (2) estimating the representative $^{187}\text{Re}/^{188}\text{Os}$ values for juvenile mafic crust by modeling fractional crystallization of central Oregon primary basaltic melts; (3) using a representative range in $^{187}\text{Re}/^{188}\text{Os}$ to calculate the age of the mafic crustal contaminant in central Oregon; and (4) applying this model to Re–Os isotopic data from elsewhere along the Cascade volcanic arc. The mafic contaminant ages are then used as a proxy for timescales of homogenization of lower arc crust as a function of the character (composition and tectonic regime) of the overlying plate.

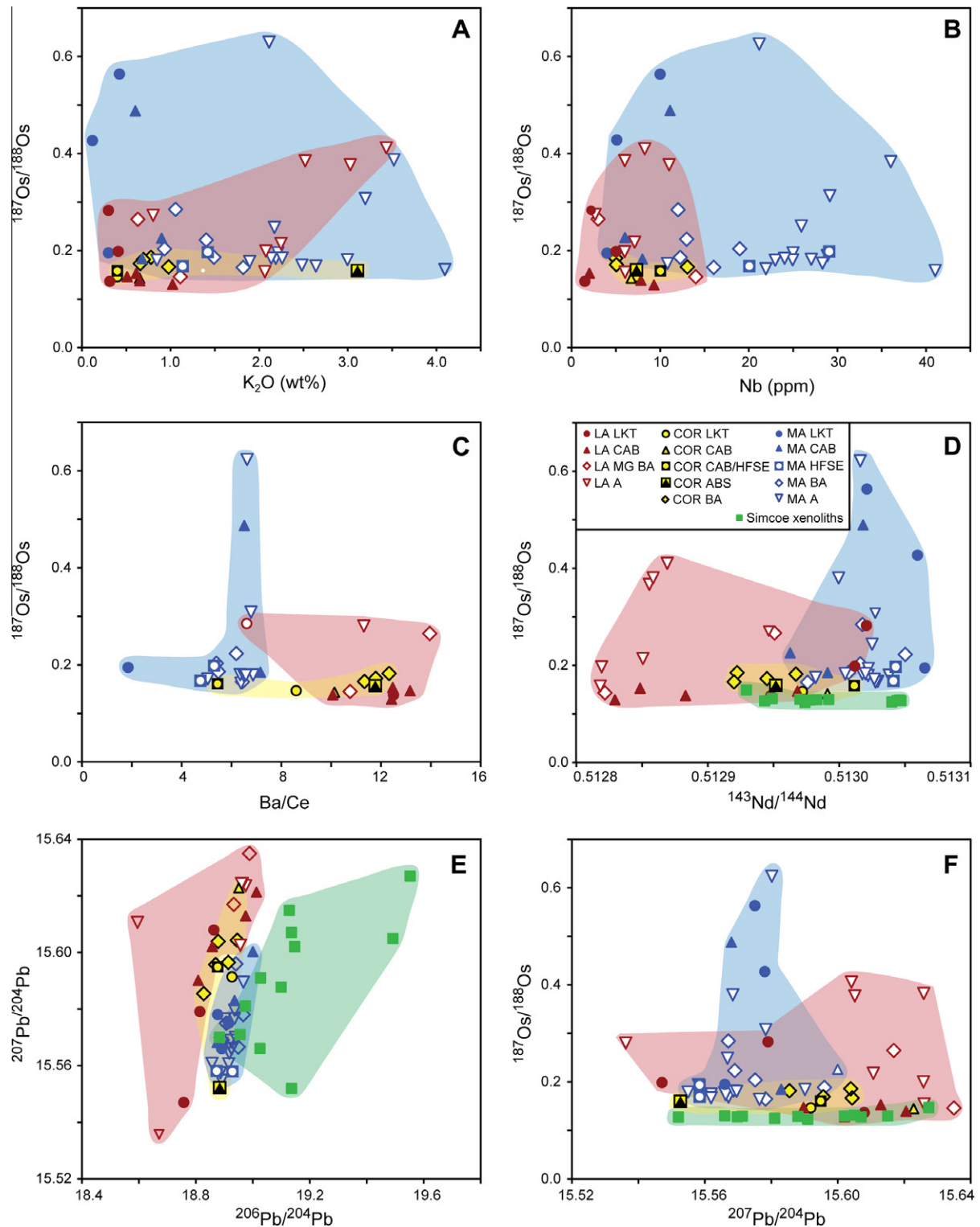


Fig. 6. Isotopic variation diagrams for Cascade arc magmas (Lassen LA, central Oregon COR, and Mt. Adams MA) and peridotite xenoliths from Simcoe Volcano. (A) $^{187}\text{Os}/^{188}\text{Os}$ vs. wt% K_2O ; (B) $^{187}\text{Os}/^{188}\text{Os}$ vs. ppm Nb as a proxy for source enrichment; (C) $^{187}\text{Os}/^{188}\text{Os}$ vs. Ba/Ce as a proxy for subduction fluid addition; (D) $^{187}\text{Os}/^{188}\text{Os}$ vs. $^{143}\text{Nd}/^{144}\text{Nd}$; (E) $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$; (F) $^{187}\text{Os}/^{188}\text{Os}$ vs. $^{207}\text{Pb}/^{204}\text{Pb}$. Data are from this study, Brandon et al. (1996, 1999), Bacon et al. (1997), W.K. Hart et al. (1997), G.L. Hart et al. (2002, 2003), Borg et al. (2000), and Jicha et al. (2009a,b).

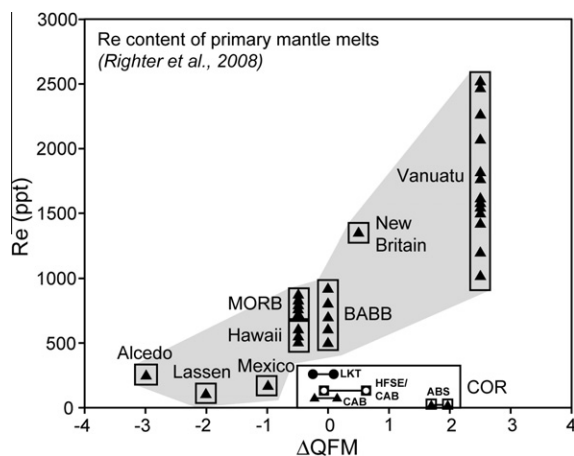


Fig. 7. Re contents in global primary mantle melts as a function of oxygen fugacity (ΔQFM) including Central Oregon Cascade (COR) magmas. The ranges in range in fO_2 for central Oregon Cascade magmas are based on olivine-hosted melt inclusions for the central Oregon Cascade arc magmas by Rowe et al. (2009). Graph is adapted from Richter et al. (2008).

4.2.1. Re and Os partitioning in arc magmas

Relative to other primary magmas from diverse tectonic settings, the Central Oregon Cascades magmas have low Re at $fO_2 \sim QFM$ and $QFM + 2$ and deviate from the global positive correlation of Re with fO_2 (Fig. 7; Richter et al., 2008). We interpret this as reflecting mineral fractionation of Re in the mantle or deep crust. Likely candidates that may affect Re and Os concentrations include garnets, sulfides, and (or) spinels (Table 3). The stability of sulfides and spinels depends on both fO_2 and water content (Mallmann and O'Neill, 2007), which can vary greatly in volcanic arcs (e.g., Wallace, 2005). That fO_2 plays an important role in partitioning of Re is further suggested by a general correlation with V concentration in Central Oregon Cascades magmas (Fig. 2c). Among the phases likely to retain Re, we argue for a leading role for spinel, particularly magnetite $\pm$ trace sulfides, over garnet.

Residual garnet is unlikely because rare earth element patterns are too flat and Ce/Yb are too low, except for the unusual ABS from the Cascade forearc, which has steep REE patterns (Ce/Yb = 173; Fig. 3). Even in that case, at least 37% residual garnet would be required in the residuum, if garnet is the only phase retaining Re (partition coefficient $D_{Re}^{gt-melt} = 2.7$; Richter et al., 1998) to account for its elevated $^{187}Os/^{188}Os$.

The presence of sulfides strongly influences the partitioning of Os in mantle-derived melts ($D_{Os}^{sulfide-melt} = 4.8 \times 10^4$; Roy-Barman et al., 1998). How Re partitions between sulfide and silicate melts greatly depends on magmatic fO_2 and fS_2 , but not on pressure (Fonseca et al., 2007). Reported $D_{Re}^{sulfide-melt}$ values vary over several orders of magnitude from $\sim 10^{-4}$ at high fO_2 and low fS_2 ($QFM + 2$, $\log fS_2 \approx -2$ to -4 ; Fonseca et al., 2007) to $3.3\text{--}5.2 \times 10^4$ at low fO_2 ($QFM - 2.2$, $\log fS_2 \approx -2$; Sattari et al., 2002). For central Oregon Cascade arc magmas with $fO_2 \sim QFM$ (LKT, CAB, and CAB/HFSE; Rowe et al., 2009), the cal-

culated $\log fS_2$ is -1.4 to 0.9 (using Eq. (7) of Wallace and Carmichael, 1992), and the corresponding $D_{Re}^{sulfide-melt}$ is ~ 1 (Fonseca et al., 2007). Trace sulfides may be present in these Central Oregon primitive magmas and affect Os concentrations, although they have not been observed in thin sections (Rowe et al., 2009). For the more oxidized ABS ($QFM + 2$), sulfides are not predicted to be stable, and the dominant S species would instead be sulfate (Carroll and Webster, 1994). Re and Os are apparently incompatible in sulfates (Richter et al., 2008).

Among the spinels, Re is not compatible in chromite and chromian spinels ($D_{Re}^{chromite-melt} \approx 0.1$ at $fO_2 \approx QFM$; Sattari et al., 2002; Brenan et al., 2003; Richter et al., 2004), and Re is somewhat compatible in Mg-Al spinels ($D_{Re}^{sp/melt} \approx 1$; Mallmann and O'Neill, 2007), but it partitions strongly into magnetite (Chesley and Ruiz, 1998; Richter et al., 1998). Magnetite-melt partition coefficients for Re in basalts vary from 40 at $fO_2 \approx QFM$ to 100 at $fO_2 \approx QFM + 2$ (Richter et al., 1998). The compatibility of Re in magnetite is commonly exploited to find measurable concentrations of Re and Os in evolved lava samples (e.g., Chesley and Ruiz, 1998). Re/Os of separated magnetite from Mt. Lassen and Mt. Adams range up to 523 (Fig. 4; Borg et al., 1997; Jicha et al., 2009a). How Re partitions into hercynite has not been documented.

In the central Oregon primitive basalts, chromite and chromian spinel are common inclusions in olivine phenocrysts, but magnetite (as part of a complete solid solution with chromite) is only found included in olivine in the high fO_2 , low-Re ABS sample QV-03-1 (Table 2). Low FeO* and V in the ABS sample further support the presence of magnetite (Fig. 2C; Rowe et al., 2006). Magnetite and titanomagnetite also occur as late-forming microphenocrysts in North Sister and Little Brother magmas (Schmidt and Gruner, 2009). At higher pressures, phase equilibria experiments by Mercer and Johnston (2008) on a North Sister basaltic andesite composition found hercynite as a near-liquidus phase at $[H_2O] \geq 5$ wt% and pressures ≥ 5 kbar. These experiments agree with thermodynamic calculations using the MELTS program of Ghiorso and Sack (1995) that suggest the near liquidus spinel for North Sister basaltic andesitic melts is hercynite (Al > Cr) in solid solution with $\sim 10\%$ magnetite at 10 kbar and 2–3.5% H_2O . For a primitive LKT near Medicine Lake, Sisson and Grove (1993) found 5–10% magnetite in the total fractionating assemblage at the Ni–NiO buffer and water saturation at 2 kbar.

Fractional crystallization of an assemblage containing 10% magnetite (in keeping with near liquidus assemblages for Ghiorso and Sack, 1995) leads to a D_{bulk} for Re of ~ 4 at $fO_2 \approx QFM$ and ~ 10 at $fO_2 \approx +1$ to $+3$ (Richter et al., 2008). Although dependent on fO_2 , bulk partition coefficients for Re are near the estimated D_{bulk} of ~ 7 for Os in the lower crust (Esperança et al., 1997; Saal et al., 1998). The $D_{bulk}^{Re}/D_{bulk}^{Os}$ for a (sulfide-undersaturated) basalt in the lower Cascade arc crust is 1 ± 0.5 , depending on the fO_2 conditions. If trace sulfides ($\sim 0.03\%$) are present in magmas with $fO_2 \approx QFM$, the change in D_{bulk} for Re is not significant, but the D_{bulk} for Os increases to 21.4 (Table 3).

Table 3
Mineral–silicate melt partition coefficients.

	pl	gt	ol	cpx	opx	mt	sulf	$D_{\text{bulk, sulfide-free}}$	$D_{\text{bulk, w/0.03\% sulfide}}$
D_{Ni}	0.06	5	15	5	7.3	15	270	7.0	7.1
D_{Os}^a							48,000	7	21.4
$D_{\text{Re}}(\text{QFM})$	0	2.7	<i>negl.</i>	0.2	<i>negl.</i>	40	1	4	4
$D_{\text{Re}}(\text{QFM} + 1 \text{ to } +3)^b$						100		10	–

^a The mineral–silicate melt partition coefficients for Os are not well known, but the D_{bulk} for Os is considered to be ~ 7 after Saal et al. (1998) for modeling purposes.

^b The D_{Re} for most minerals at $f\text{O}_2$ QFM + 1 to + 3 is equivalent to D_{Re} at QFM, except for magnetite. Partition coefficients are from Richter et al. (1998,2004), Roy-Barman et al. (1998), Fonseca et al. (2007), Rajamani and Naldrett (1979), and from compilation in Schmidt and Grunder (2011). Bulk D 's for Ni were calculated assuming 40% plagioclase, 30% olivine, 20% clinopyroxene, 10% magnetite, and $\pm 0.03\%$ sulfide. At $f\text{O}_2 \approx \text{QFM} + 2$, the D_{bulk} for Re increases to ~ 10 (Richter et al., 1998) and sulfide saturation is unlikely (Carroll and Webster, 1994).

4.2.2. Re/Os of mafic arc crust

As mantle-derived materials intrude and transform arc crust, the bulk composition of juvenile mafic arc crust is tied to the differentiation of primary basaltic magmas. To illustrate the evolution of Re and Os concentrations in primary basaltic magmas, Rayleigh fractional crystallization (RFC) models are presented for primitive central Oregon magmas (LKT, CAB, and ABS) in Fig. 8. In RFC, the concentration of element i in the liquid melt, C_L^i is given by:

$$C_L^i = C_0^i \times F^{(D-1)}, \quad (1)$$

where C_0^i is the initial concentration, F is the melt fraction, and D is the bulk partition coefficient (Table 3). Although primitive, the CAB and LKT likely experienced some degree of fractional crystallization. To find initial Os and Re concentrations, the C_0^{Ni} of primary arc mantle melts are assumed to be 300 ppm (in line with primary Aleutian CAB of Nye and Reid, 1986) and F was calculated for C_L^{Ni} (Table 1). The corresponding C_0^{Os} and C_0^{Re} were then calculated for C_L^{Os} and C_L^{Re} (Table 2) at F . Initial Os and Re concentrations found by this method are 0.33 and 0.33 ppb for the LKT and 1.08 and 0.1 ppb for the CAB, respectively (Fig. 8).

An important constraint for discriminating between models is that $^{187}\text{Re}/^{188}\text{Os}$ must be large enough for mafic crust to evolve to high $^{187}\text{Os}/^{188}\text{Os}$ over a geologically short period (i.e., younger than the age of the crust itself). Magnetite-bearing assemblages at $f\text{O}_2 \approx \text{QFM}$ (Fig. 7) in equilibrium with the CAB or LKT melts initially have Re/Os ratios that are roughly equivalent to those of the basaltic melts (Fig. 8C and D). As fractional crystallization of the LKT and CAB progresses, the Re/Os of the melt (C_{liq}) and sulfide-free crystalline assemblage (C_{xls}) increases. But unless crystallization progresses to degrees greater than 50% for the LKT or 70% for the CAB, magnetite fractionation is insufficient to account for enrichments of $^{187}\text{Os}/^{188}\text{Os}$ in arc crust over short time periods. Higher Re/Os may be attained if a sulfide phase is present, even in minute quantities (here 0.03%). In which case, Os is depleted from the melt and Re/Os attains high values (>50) after ~ 10 – 30% crystallization. This demonstrates that internal magmatic differentiation cannot achieve high enough Re/Os to produce the radiogenic $^{187}\text{Os}/^{188}\text{Os}$ in the time allowed.

The central Oregon Cascade basaltic andesites contain Re and Os concentrations that are consistent with fractional crystallization of a trace sulfide-bearing assemblage from the LKT composition (Fig. 8A and B). Elevated Os isotopic ratios of the basaltic andesites relative to the basalts do not support a simple fractional crystallization model, but the fit of the elemental concentrations provides independent support for the potential realism of these models. Indeed, these models suggest that the range in $^{187}\text{Re}/^{188}\text{Os}$ (56–263; Table 1) found in the North Sister basaltic andesites are reasonable estimates for the range in juvenile mafic arc crust if LKT magmas are the predominant input from the mantle.

Under conditions that are likely to affect the ABS magma during fractional crystallization (relatively high $f\text{O}_2$ of +2 ΔQFM , low Re concentration), $D_{\text{bulk}}^{\text{Re}} \approx 10$ and exceeds $D_{\text{bulk}}^{\text{Os}}$ (Fig. 8B). Consequently, at all degrees of fractional crystallization, the ABS melt contains Re/Os < 1 . The Ni is quite high in the ABS (256 ppm, Table 1) and this may in part reflect accumulated olivine. Magnetite fractionation alone cannot account for the low Re. The ABS source must contain a residual phase that strongly retains Re during partial melting, such as magnetite and (or) garnet (like serpentinized garnet peridotite).

4.2.3. Constraining the age of central Oregon Cascade deep crust

In order to constrain the age of the central Oregon Cascade mafic lower crust, we approximate its $^{187}\text{Os}/^{188}\text{Os}$ to be equivalent to that of basaltic andesitic magmas from North Sister Volcano. Higher $^{187}\text{Os}/^{188}\text{Os}$ in the low-K basaltic andesites from North Sister than in the medium-K basaltic andesite from Little Brother suggests a greater influence by mafic crust (Fig. 3). Our previous work (Schmidt and Grunder, 2011) demonstrates that the low-K basaltic andesites interacted with a gabbroic crust at depths of ~ 40 km. As a magma body differentiates by assimilation and fractional crystallization (A + FC) processes (Appendix 1, Schmidt and Grunder, 2011), the concentrations of trace element i changes as a function of its concentration in the assimilating melt (C_a^i) the melt fraction (F), and the ratio r (rate of assimilation/rate of crystallization) according to the equation:

$$C_L^i = \frac{(C_0^i F^{D-1}) + (C_a^i r(1 - F))}{(1 + r(1 - F))}. \quad (2)$$

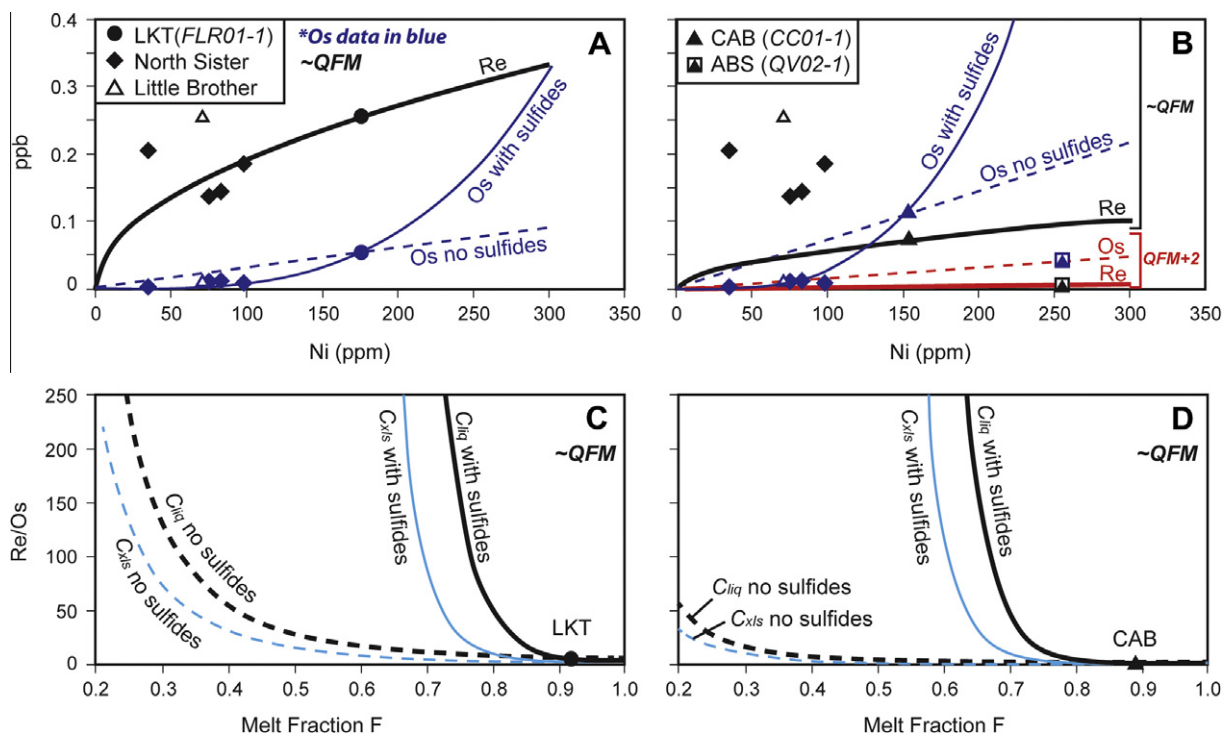


Fig. 8. Rayleigh fractional crystallization models (Eq. (1)) for Ni, Re, and Os concentrations for the primitive Cascade LKT and CAB at f_{O_2} of approximately QFM. Curves were calculated with or without a minute quantity (0.03%) of sulfide phase present using bulk partition coefficients in Table 3. Plots A and B are Re and Os vs. Ni concentration in the fractionating melt for the LKT and CAB, respectively. Central Oregon Cascade compositional data are represented in black for Re and blue for Os. Plot B includes fractionation models for Re, Os, and Ni concentration for the ABS at QFM + 2. Plots C and D are Re/Os vs. melt fraction F for the liquid (C_{liq}) and crystalline product (C_{xls}) for the LKT and CAB, respectively.

The A + FC formulation differs from AFC of dePaolo (1981) where C_L/C_0 is asymptotic, approaching infinity near $r = 1$. Several workers have presented diverse cases where $r \geq 1$ (e.g., Grove et al., 1988; Reiners et al., 1995) and in the hot, hydrous deep arc crust, $r \geq 1$ may be the more likely case (Annen et al., 2006). Once we calculate the C_L^{Os} at 1% increments, we can find the $^{187}Os/^{188}Os$ of the melt R_L^{Os} using a modified version of the general 2-component isotopic mixing equation:

$$R_L^{Os} = \frac{(R_{L*}^{Os} \times C_{L*}^{Os} \times (1 - 0.01r)) + (R_a^{Os} \times C_a^{Os} \times 0.01r)}{(C_{L*}^{Os} \times (1 - 0.01r)) + (C_a^{Os} \times 0.01r)} \quad (3)$$

Here, (R_{L*}^{Os}) and (C_{L*}^{Os}) are the Os isotopic ratio and concentration of the melt in the previous 1% increment, and (R_a^{Os}) and (C_a^{Os}) are the Os isotopic ratio and concentration of the assimilated melt. In this way, the differentiating magma body acquires the $^{187}Os/^{188}Os$ of the assimilated country rock as illustrated by A + FC curves presented in a plot of $^{187}Os/^{188}Os$ vs. Ni (Fig. 9). Although the rate of change in $^{187}Os/^{188}Os$ depends on the ratio r and (C_a^{Os}) , the differentiating magma approaches $^{187}Os/^{188}Os$ of the assimilated wallrock by the time Ni falls to concentrations in North Sister basaltic andesites (30–100 ppm). If radiogenic Os in evolved magmas is predominantly derived by assimilation of grain boundary phases (e.g., Peucker-Ehrenbrink et al.,

2012), then the differentiating magma body would acquire crustal values even more rapidly than is illustrated in Fig. 9.

The decay of ^{187}Re to ^{187}Os is predictable (half-life, $t_{1/2} = 4.16 \pm 0.11 \times 10^{10}$ years; decay constant, $\lambda (^{187}Re) = 1.666 \pm 0.04 \times 10^{-11} \text{ year}^{-1}$; Shirey and Walker, 1998). By assuming that the juvenile mafic crust $(^{187}Os/^{188}Os)_{\text{initial}}$ is like the regional primitive LKT basalt (as in Schmidt and Grunder, 2011), and $(^{187}Re/^{188}Os)$ and $(^{187}Os/^{188}Os)_{\text{final}}$ are like the North Sister basaltic andesites (Table 3), we estimate the range in age of the crust (t) with the radioactive decay equation:

$$t = \frac{1}{\lambda} \ln \left(\frac{(^{187}Os/^{188}Os)_{\text{final}} - (^{187}Os/^{188}Os)_{\text{initial}}}{(^{187}Re/^{188}Os)} + 1 \right) \quad (4)$$

Fig. 10 depicts how the determined age of the mafic contaminant varies as a function of its $^{187}Re/^{188}Os$. We interpret the range in age of the mafic contaminant to be where the time curve intersects the range in North Sister $^{187}Re/^{188}Os$, or ~9–29 Ma. The age range may extend to 6–43 Ma if the intersection of the upper and lower limit decay curves are also considered, but we prefer the more narrow age range given the limited variability of the North Sister magmas (Schmidt and Grunder, 2011).

As an alternate method to solve for t , we use the Re and Os ratios of individual lava samples (Table 2) to yield sequential ages of ~43, ~28, and ~9 Ma. The age of the

mafic contaminant *apparently* decreases over the history of North Sister Volcano (500–50 ka; Schmidt and Grunder, 2009). While an interesting outcome, this exercise involves a small set of compositionally restricted magmas erupted in short succession (no more than ~200 k.y.) and the crustal contaminant apparently did not change much in composition over that time (Schmidt and Grunder, 2011).

The estimated age range for the mafic crustal contaminant of ~9–29 Ma age is younger than the age of the Siletz Terrane (~50–60 Ma; Duncan, 1982) as well as the onset of Cascade arc volcanism in this part of the arc (45 Ma; du Bray and John, 2011). Clockwise rotation of the Cascadia forearc since docking of the Siletz Terrane against North America ~51 Ma (Wells and Simpson, 2001) allows space for crustal growth by intraplating of magma in the arc since its inception. Geophysical observations such as an apparent lack of crustal thickening (Trehu et al., 1994) and slow shear wave velocities (Porritt et al., 2011), suggest that spreading and basaltic dike injection are the main contributors to crustal growth. A pulse of LKT magmatism at ~7–8 Ma is interpreted to reflect renewed vigor in intra-arc rifting (Sherrod and Smith, 1990; Conrey et al., 2004) and postdates the interpreted age range. As crustal extension and magmatism are coupled in the Central Oregon arc, the lower crust is homogenized to isotopically primitive compositions by crystallization and assimilation of arc-related gabbroic to ultramafic intrusions. This interpretation is consistent with other observations of the North Sister system, such as high Al_2O_3 contents (18–20 wt% Al_2O_3) and the highest $^{87}\text{Sr}/^{86}\text{Sr}$ found among earliest magmas, that suggest melting and assimilation of older plagioclase-bearing basement (Schmidt and Grunder, 2011).

In the forearc, high $f\text{O}_2$ and low Re of the unusual ABS (Fig. 7) may suggest interaction with serpentinized garnet-

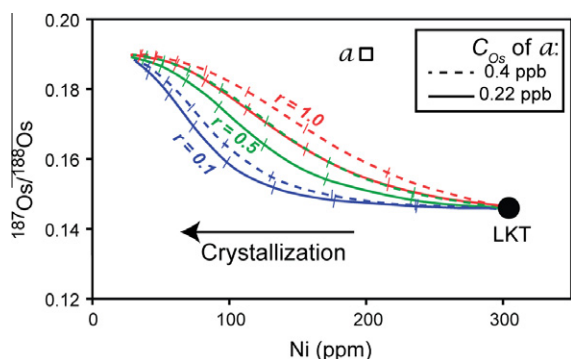


Fig. 9. Assimilation + fractional crystallization (A + FC) magma differentiation model (Eqs. (2) and (3); Schmidt and Grunder, 2011) for a primary LKT magma presented on a plot of $^{187}\text{Os}/^{188}\text{Os}$ vs. Ni concentration in ppm. Tick marks indicate 5% crystallization increments. Bulk partition coefficients for a trace sulfide-bearing assemblage are as in as in Fig. 8 and Table 3. Curves represent models at varying assimilation/crystallization (r ; $r = 0.1, 0.5$ and 1) and C_{Os} of the assimilated melt, a . The $^{187}\text{Os}/^{188}\text{Os}$ of the assimilant for this model is 0.19. The varying C_{Os} of 0.4 and 0.22 ppb represent 10% batch melts of accumulated crystals by 10% and 20% fractional crystallization, respectively of a mantle derived basalt with 1 ppb Os. The $^{187}\text{Os}/^{188}\text{Os}$ of the assimilant for this model is 0.19.

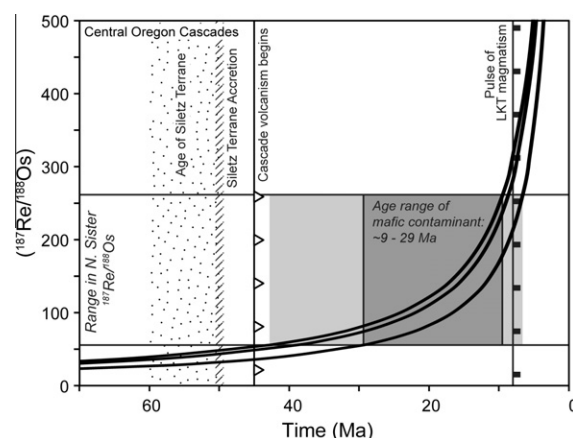


Fig. 10. Age of the crustal contaminant for North Sister basaltic andesite plotted as a function of $^{187}\text{Re}/^{188}\text{Os}$. The range in North Sister $^{187}\text{Re}/^{188}\text{Os}$ is indicated and likely represents the initial $^{187}\text{Re}/^{188}\text{Os}$ of juvenile mafic crust as discussed in Section 4.2.3. The intersection of the range in North Sister $^{187}\text{Re}/^{188}\text{Os}$ and the three decay curves (dark gray) gives an interpreted age range of the mafic lower crust of ~9–29 Ma. See text for discussion. Significant events in the history of the Cascade arc are indicated, including the Age of the Siletz Terrane (50–60 Ma), the age of Siletz accretion (~50 Ma; Duncan, 1982), the inception of Cascade volcanism in central Oregon (45 Ma; du Bray and John, 2011), and initial pulse of LKT magmatism related to intra-arc rifting (~8 Ma; Conrey et al., 2004). These calculations demonstrate that the age of the lower crust is younger than the Siletz Terrane and the onset of Cascade volcanism.

bearing peridotite. The deep arc mafic intrusions are therefore narrowly focused beneath the arc axis and High Cascade Graben structure and are consistent with a steep, negative heat flow gradient toward the western Cascades (Blackwell et al., 1982; Ingebritsen and Mariner, 2010).

4.2.4. Lassen Volcanic Field

The ages estimated for the crustal contaminant at North Sister contrasts with ages that have been determined by other workers for crustal contaminants at other Cascade volcanic systems. At Lassen, Hart et al. (2002) suggested that very radiogenic $^{187}\text{Os}/^{188}\text{Os}$ (up to 0.4127) could evolve after only 5–10 Ma through continuous Re/Os enrichment (Hart et al., 2002).

Applying Re–Os isotopic values of andesite to rhyodacite magmas from Lassen Volcanic Field (Table 4; Electronic Suppl. II) to Eq. (1) yields age range estimates for the mafic contaminant at Lassen by single-stage Re/Os enrichment. (Mg-rich basaltic andesite and andesites were not included in these calculations.) Apparent clustering of decay curves (Fig. 11) suggests three overlapping, yet distinct ranges in age describe the crustal contaminant at Lassen: 8–35 Ma, 21–135 Ma, and 68–340 Ma. The first range is comparable to the age range for the crustal contaminant in the central Oregon Cascades (Fig. 11) and likely represents recent additions from the mantle (i.e., since arc inception in the late Eocene). The later ranges are much larger and may represent mixtures of different age crust, including the modern arc component, Mesozoic plutons (141–

170 Ma; Barnes et al., 1992), Early Silurian plutons (431–404 Ma; Wallin and Metcalf, 1998), and Trinity Peridotite (472 ± 32 Ma; Jacobson et al., 1984). Importantly, these calculations demonstrate that it is not necessary to invoke very high Re/Os in the crustal contaminant to account for the radiogenic $^{187}\text{Os}/^{188}\text{Os}$ in evolved Lassen volcanic rocks.

The Lassen volcanic field also sits in an extensional setting, however the range in magma compositions is greater than found in the Central Segment, and includes some of the highest Ba/Ce, $^{87}\text{Sr}/^{86}\text{Sr}$, and $^{207}\text{Pb}/^{204}\text{Pb}$ of the Cascade arc (Fig. 6). Other workers have suggested that isotopic diversity of Lassen is at least in part caused by excess slab fluids brought about by subduction of the Mendocino Fracture Zone and/or the deformed Gorda Plate (e.g., Borg et al., 1997). The wide Re–Os age ranges presented here (~ 8 –340 Ma) suggest that the northern California crust with its patchwork of blocks with varied age, origin, and isotopic characteristics also contributed to the diversity observed in Lassen magmas.

4.2.5. Mt. Adams

The range in $^{187}\text{Os}/^{188}\text{Os}$ of from Mt. Adams is much greater than elsewhere along the arc (Fig. 5). $^{187}\text{Os}/^{188}\text{Os}$ as high as 0.56 is found in otherwise depleted, primitive LKTs and low as low as 0.17 has been determined for some of the more evolved magmas (Fig. 5; Hart et al., 2003). The high degree of variability in $^{187}\text{Os}/^{188}\text{Os}$ has been attributed to the assimilation of young mafic crust (~ 9 and 2 Ma) by Hart et al. (2003) by continuous Re/Os enrichment. Although Re/Os of Mt. Adams magmas ranges to higher values than have been found at other Cascade volcanic systems (Fig. 4), it is insufficient to create such large increases in $^{187}\text{Os}/^{188}\text{Os}$ over such short time periods by single-stage Re/Os enrichment.

To account for the variability seen in Mt. Adams magmas, age curves were calculated using three $^{187}\text{Os}/^{188}\text{Os}_{\text{initial}}$ values, including a low $^{187}\text{Os}/^{188}\text{Os}$ harzburgitic xenolith from Simcoe Volcanic Field, and two primitive basalts from

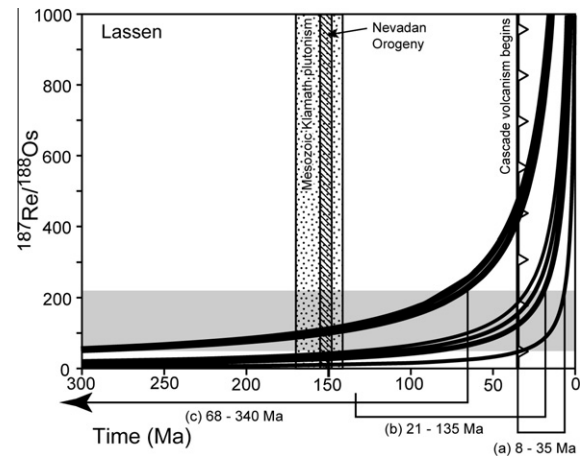


Fig. 11. Age curves for the crustal component in andesite to rhyodacite composition magmas from Mt. Lassen as a function of $^{187}\text{Re}/^{188}\text{Os}$. The age of the contaminant is determined at Lassen $^{187}\text{Re}/^{188}\text{Os}$ (gray field, Table 4). Clustering of the curves and suggests three overlapping, but discrete age ranges: 8.7–38 Ma, 23–150 Ma, and 75–360 Ma. Also shown is the range in age of Mesozoic Klamath plutonism that is coincident with, but occurred before and after the Nevadan Orogeny (Barnes et al., 1992) and the start of Cascade volcanism ~ 35 Ma in this part of the arc (Ewing, 1980; du Bray and John, 2011).

Mt. Adams (LKT and HFSE; Table 4; Electronic Suppl. II). Possible age ranges for the crustal contaminant for basaltic andesitic to silicic composition magmas (Fig. 12) were determined over the range $^{187}\text{Re}/^{188}\text{Os}$ in Mt. Adams basaltic andesites (140–430; excepting one analysis with anomalously low Re/Os; Hart et al., 2003; Jicha et al., 2009a). Modeled age ranges are 18–70 Ma, 0–64 Ma, and 0–61 Ma for the harzburgite, HFSE-rich basalt, and LKT starting compositions, respectively.

Zero to negative ages (Fig. 12B and C) are the result of $^{187}\text{Os}/^{188}\text{Os}_{\text{initial}}$ being higher than some of the more evolved magmas. Therefore, parental magmas with lower

Table 4
Mafic contaminant model age parameters.

	Central Oregon	Lassen	Mt. Adams
$(^{187}\text{Os}/^{188}\text{Os})_{\text{initial}}$	0.1459 ¹	0.12845 ³	A: 0.1232 ⁶ B: 0.1681 ⁷ C: 0.1951 ⁸
Range in $(^{187}\text{Re}/^{188}\text{Os})$	55.5–262.7 ²	50–218 ⁴	140–430 ⁹
Determined age range(s)	~ 9 –29 Ma	(a) ~ 8 –35 Ma ⁵ (b) ~ 21 –135 Ma (c) ~ 68 –340 Ma	A: ~ 18 –70 Ma B: ~ 0 –64 Ma C: ~ 0 –61 Ma

¹ Primitive LKT #FLR-03-01 from Central Oregon.

² Range in North Sister basaltic andesite (Table 2).

³ Primitive CAB #LC85-671 (Borg et al., 1997, 2000).

⁴ Range in Lassen andesites and dacites (Hart et al., 2002).

⁵ Three age ranges for Lassen interpreted from clustering in Fig. 11.

⁶ Harzburgitic xenolith from Simcoe Volcanic Field (Brandon et al., 1999).

⁷ Primitive HFSE-rich basalt from Mt. Adams #bsb (Hart et al., 2003; Jicha et al., 2009a,b).

⁸ Primitive LKT from Mt. Adams #bsc (Hart et al., 2003; Jicha et al., 2009a,b).

⁹ The range in $^{187}\text{Re}/^{188}\text{Os}$ was determined as $\sim 5X$ the reported Re/Os of Mt. Adams basaltic andesites, excepting #bdw-W, a sample with very low Re (Hart et al., 2003; Jicha et al., 2009a,b).

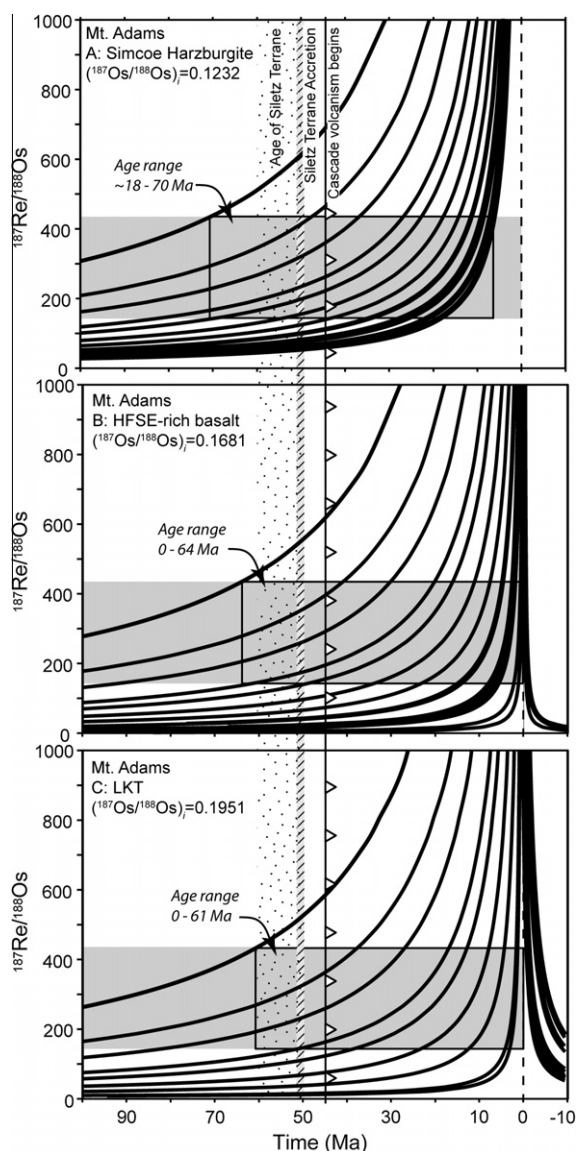


Fig. 12. Age curves for the crustal component in andesite to rhyodacite composition magmas from Mt. Adams as a function of $^{187}\text{Re}/^{188}\text{Os}$ at different $^{187}\text{Os}/^{188}\text{Os}$ starting compositions including A: Simcoe harzburgitic xenolith that represent depleted back arc lithospheric mantle (Brandon et al., 1999), and two Mt. Adams primitive basalts B: HFSE-rich basalt, and C: LKT (Hart et al., 2003; Jicha et al., 2009a,b). The age range of the contaminant is determined at Adams $^{187}\text{Re}/^{188}\text{Os}$ (gray field, Table 4). In all cases, the age ranges extend beyond the age of the Siletz Terrane. In this part of the arc, Cascades volcanism began ~ 45 Ma (du Bray and John, 2011). Also, the basaltic starting compositions have higher $^{187}\text{Os}/^{188}\text{Os}$ than some intermediate magmas. Together, these are strong indications that the lithospheric mantle and lower crust are heterogeneous with regard to Re–Os composition and age.

$^{187}\text{Os}/^{188}\text{Os}$ than the primitive LKT and HFSE-rich basalts (like the Simcoe harzburgite; Fig. 12A) must contribute to the Mt. Adams magma system, despite not being among the existing dataset. Also, in all cases, the magmas with the most radiogenic $^{187}\text{Os}/^{188}\text{Os}$ yield ages greater than the age of the Siletz Terrane (Duncan, 1982) and the onset

of Cascade volcanism in this part of the arc (du Bray and John, 2011).

The old age ranges suggest the existence of a crustal (or lithospheric) contaminant that is anomalously enriched in $^{187}\text{Os}/^{188}\text{Os}$. Although older (>60 Ma) basement is possible, it has not been identified by other surveys, and it seems more likely that the lithosphere beneath Mt. Adams contains heterogeneities with respect to Re and Os. The Re–Os heterogeneities may originate in the enriched Siletz Terrane and may be related the high proportion of HFSE-rich basalts in the vicinity. A variably enriched lithosphere beneath Mt. Adams is also supported by relatively high degree variability and distinct ranges in other isotopic systems, including Sr, Nd, Pb, (Fig. 7), Th, and U relative to other parts of the arc (Bacon et al., 1997; Hildreth, 2007; Jicha et al., 2009a,b). Alternatively, metasomatism may be suggested by Simcoe harzburgite xenoliths (Brandon et al., 1996, 1999), which contain trace sulfide blebs in association with textures indicative of metasomatism, such as annealed cracks and fluid inclusions (Ertan and Leeman, 1999).

Lithospheric source variability imparts a stronger signal to Mt. Adams magmas than seen elsewhere in the Cascade arc. While enriched Siletzia lithosphere is certainly important, upper plate tectonics likely also plays a role. In contrast to the southern half of the arc, extension is relatively recent at Mt. Adams (since ~ 1 Ma; Conrey et al., 2004). As such, there is less room for magmas to enter the deep crust. The lesser amounts of extension in combination with a variably enriched lithosphere apparently limits the degree of homogenization of the lower crust, or the degree to which its bulk composition reflects younger mantle-derived basalts rather than pre-existing lithologies.

5. CONCLUSIONS

Examination of Re and Os isotopes in diverse basaltic rocks from the central Oregon Cascades reveals a narrow range of $^{187}\text{Os}/^{188}\text{Os}$ (0.141–0.159). These basalts are intermediate in Nd-, Pb- and Os-isotopic character between primitive HFSE-rich basalts in the Mt. Adams region to the north and primitive calc-alkaline basalts in the Mt. Lassen region to the south.

Higher $^{187}\text{Os}/^{188}\text{Os}$ in central Oregon Cascades basaltic andesitic rocks than the primitive basalts suggests interaction with mafic crust with similarly high $^{187}\text{Os}/^{188}\text{Os}$. Partitioning of Re/Os in primitive magmas is relevant to assessing the age and character of the crustal contaminant. Low Re in the most oxidized central Oregon Cascades ABS is consistent with fractionation of Re via magnetite and may suggest a serpentinized source. For magmas with $f\text{O}_2 \sim \text{QFM}$, Re/Os fractionation into the crust likely involves magnetite and trace sulfides.

Age ranges modeled for mafic magmas to evolve to the $^{187}\text{Os}/^{188}\text{Os}$ of more intermediate magmas reflect how much the composition of the crust is a product of recent arc magmatism vs. pre-existing protoliths. The range of 9–29 Ma for the central Oregon Cascades is younger than the inception of arc volcanism and indicates extensive mafic additions to the crust facilitated by intra-arc extension. To the

south, Paleozoic basement and modern arc contributions are reflected in the wide 8–340 Ma age range for Lassen magmas. In contrast, the 0–70 Ma range for Mt. Adams is both too young and too old to be reasonable and are attributed variable Re/Os enrichments of the Siletz Terrane and the Columbia Embayment lithosphere.

The Re and Os isotopic system provides insight to the remaking of mafic arc crust over time. Mafic arc crust matures through the intrusion and crystallization of mantle derived basaltic magmas and as these magmas assimilate pre-existing lithologies. As seen in the central Oregon Cascades, extensional tectonics leads to a faster rate of maturation and more homogeneous magma compositions. Where extension is less narrowly focused, the range in Re and Os isotopes is greater and reflects crustal heterogeneities, including variably aged lithologies beneath the Lassen volcanic field or heterogeneously enriched Re and Os lithosphere of Mt. Adams.

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APPENDIX A. SUPPLEMENTARY DATA

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.gca.2013.02.003>.

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