Temporal-compositional trends in intraplate basalt eruptions: Implications for mantle heterogeneity and melting processes

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[1] Because primitive basaltic magmas travel from their mantle sources to the surface without significant cooling and crystallization en route, compositional variations within them provide direct information on the sources and processes of partial melting in the mantle. Most studies of basalt magmagenesis have focused on chemical variations in magmas erupted from multiple vents in regional complexes or on long timescale ($10^5$–$10^6$ years) variations in magmas from single shield volcanoes; few studies have aimed to measure changes in melt chemistry with time in individual eruptions. Available observations of individual basaltic eruption sequences, however, show remarkably similar temporal-compositional trends in the form of decreasing incompatible elements and MgO, with increasing SiO$_2$, as the eruption (or sequence of related, closely-spaced eruptions) continued. Both trace element and isotopic variations indicate that this does not reflect fractional crystallization nor can it be explained simply by varying extents of partial melting of a single source. Instead, these trends are consistent with systematically changing proportions of mixing between melts produced by different degrees of partial melting of two compositionally distinct but apparently common sources in the mantle. One potential explanation for this is that a relatively deep mantle source melts to large degrees, producing melts that ascend through and mix with small-degree melts of the lithosphere. The small-degree lithospheric melts are then progressively exhausted during melt migration and eruption, so that late-erupting melts contain the smallest proportion of the shallow, small-degree melt. This cannot explain Os isotopic correlations with melt composition and radiogenic Os isotope signatures of the late-erupting melts in several eruption sequences, however. An alternative scenario is that systematic changes in erupted melt chemistry with time reflect sequential eruption of melt from increasing depths in mantle melting regions that are compositionally zoned because of different solidi and melt productivities of distinct peridotite and pyroxenite lithologic domains. In either case, compositional variation in many eruptions of primitive, intraplate basalts is dominated by mixing of melts from distinct lithologies, and temporal-compositional changes in such eruptions reflect the distribution and composition of melt with depth in mantle melting and melt migration regions.

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1. Introduction

1.1. Mantle Mafic Heterogeneities and Sources of Basalts

[3] Direct observation of mantle-derived rocks in massifs and xenoliths provides clear evidence that the upper mantle is lithologically heterogeneous. Although mantle-derived basalts typically show compositional variation over a wide range of scales, the potential relationships between mantle lithologic heterogeneity and compositional variation in basalts are poorly understood at best. One of the potentially most important types of mantle lithologic heterogeneity is pyroxene-rich mafic rock. While most observable mantle samples are dominated by olivine-rich peridotite, they also contain some proportion of centimeter to meter scale veins or lenses of pyroxene-rich rock (pyroxenite or eclogite) [Polve and Allègre, 1980; Reisberg and Zindler, 1986; Nixon, 1987; Suen and Frey, 1987; Wilshire et al., 1991; Mukasa et al., 1991; Pearson et al., 1993; Hirschmann and Stolper, 1996]. Mantle pyroxenites probably originate from either migrating melts [Frey and Prinz, 1978; Frey, 1980; Irving, 1980; Frey and Roden, 1987; Kelemen et al., 1995; Kelemen et al., 1997] or recycling of subducted basaltic components [e.g., as suggested by Hauri, 1996; Lassiter and Hauri, 1998], although other origins are also possible including delamination of continental lithosphere and metamorphic segregation [Hirschmann and Stolper, 1996]. Although the exact origins and abundance of pyroxenite in the asthenospheric mantle are not well known, it is estimated that the upper mantle may contain 2–5% pyroxenite [Hirschmann and Stolper, 1996]. The ubiquity of pyroxenite in studied upper mantle sections, combined with its potential for trace element and isotopic characteristics different from that of typical peridotite, have motivated many previous studies to invoke a pyroxenite contribution to basalt genesis [Zindler et al., 1984; Allègre and Turcotte, 1986; Prinzhofer et al., 1989; Leeman and Harry, 1993; Hirschmann and Stolper, 1996; Phipps Morgan, 1999; Lundstrom et al., 2000; Carlson and Nowell, 2001, Phipps Morgan, 2001]. At least in part because mantle pyroxenites may relate the origin of distinctive ocean-island basalt geochemical signatures to the fate of recycled oceanic crust [e.g., Hofmann and White, 1982; Hofmann, 1997; Sobolev et al., 2000], distinguishing their potential signature in basalts is an important goal.

[3] Another important difference between pyroxenite and peridotite mantle lithologies is contrasting melting behavior. Pyroxenite melting experiments with appropriate compositions and conditions for basalt genesis are rare, and generalizations to mantle melting are difficult due to a large compositional variability in observed mantle pyroxenites and their constituent phases [e.g., Hirschmann and Stolper, 1996; van Westrenen et al., 2001]. Nevertheless, available evidence indicates that typical mantle pyroxenites have lower temperature solidi than peridotites (e.g., by 150–200°C at 2–3 GPa) and are capable of producing large melt fractions at depths where peridotite is either solid or only slightly melted [Hirschmann and Stolper, 1996; M. Pertermann, and M. M. Hirschman, Partial melting experiments on a MORB-like pyroxenite between 2 and 3 GPa: Constraints on the presence of pyroxenites in basalt source regions, submitted to Journal of Geophysical Research, 2000, hereinafter referred to as Pertermann and Hirschman, submitted manuscript, 2000]. Because primitive alkalic magmas are thought to be derived from relatively low integrated degrees of melting, they have been the target of recent attempts to identify geochemical signatures of pyroxenite sources [Lassiter et al., 2000; Carlson and Nowell, 2001]. In particular these studies have noted extremely radiogenic Os isotopic compositions in primitive alkalic magmas. Since nearly all mantle peridotite has relatively low $^{187}$Os/$^{188}$Os (<0.13) [Roy-Barman and Allègre, 1994; Shirey and Walker, 1998; Lassiter et al., 2000; Brandon et al., 2000; Meisel et al., 2001], these radiogenic Os signatures are difficult to explain unless their source contains a significant proportion of mafic mantle lithologies, such as pyroxenite.

1.2. Potential Importance of Temporal-Compositional Trends in Basaltic Eruptions

[4] Like most basalt geochemistry studies, previous attempts to distinguish signatures of mantle...
lithologic heterogeneity in erupted basalts have focused on lavas from multiple volcanoes in large-scale regional vent complexes or on long timescale \((10^5–10^6 \text{ years})\) variations in magmas from single shields. Several considerations of the expected melting behavior of a heterogeneous mantle suggest that sampling on smaller timescale and length scales may be a better strategy, however. First, given the range of plausible origins and compositions of mantle lithologic heterogeneities, clear mixing relationships and distinctions between discrete mantle components may be more difficult to observe in melts from multiple melting regions with varying component compositions and proportions. Different melting behaviors of distinct lithologic domains (e.g., different melt fractions and therefore compositions at a given set of conditions) may also be difficult to distinguish in basalts produced from a wider range of mantle melting conditions and integrated degrees. Second, because primitive basalts commonly ascend from their sources fairly rapidly (of the order of years; McKenzie [2000]), they may avoid shallow-level homogenization processes. Because of this, time series analyses of primitive basaltic eruptions that tap heterogeneous mantle regions could potentially provide information on the compositional structure of melting regions or the physical mechanisms by which component signatures are mixed.

[5] Unfortunately, few studies have attempted to measure compositional variation on the scale of individual basaltic eruptions, and even fewer have attempted to associate changes in melt chemistry with eruption sequence. Systematic temporal-compositional changes are frequently observed in silicic or intermediate-composition eruptions and are attributable to crystallization or magma mixing processes in shallow-level crustal magma chambers. Primitive mantle-derived magmas, however, do not reside for long time periods in crustal magma chambers, and compositional variations in these eruptions must reflect either mantle source heterogeneity or mixing during melt migration between source and eruption. This study focuses on interpretation and modeling of data from studies of chemical variations in samples from individual, generally primitive, basaltic eruptions and for which temporal control on eruption sequence is available. The observation that all of these eruption sequences show similar temporal-compositional trends is potentially important itself and may provide important constraints on the dynamics of melt migration during source-to-surface transport. The similarity between these trends and those expected from theoretical consideration of heterogeneous mantle models also provides support for a significant influence of lithologic heterogeneity on basalt geochemistry in general.

2. Eruption Sequences

[6] Several studies have shown that individual eruptions or series of cogenetic eruptions closely spaced in time \((<10^2 \text{ years})\) of basaltic magma show compositional variation that is related to eruption sequence. I have compiled compositional data for intraplate basalts for which (1) the relative time of eruption of at least some of the different flows or flow units can be determined, (2) the magmas are relatively primitive (i.e., compositions of most elements are not strongly affected by fractional crystallization), or their compositions can be fairly confidently corrected for the effects of fractional crystallization, and (3) their eruptions were from relatively short-lived monogenetic or short-duration polycyclic \((<\text{about }10^2 \text{ years})\) vents. The eruption sequences are from a variety of intraplate volcanic settings, and include (1) Plio-Pleistocene rejuvenated-stage alkalic lavas from Kaua’i, Hawai’i [Feigenson, 1984; Clague and Dalrymple, 1988; Maaløe et al., 1992; Reiners and Nelson, 1998; Reiners et al., 1999], (2) the 1730–1736 alkalic through tholeiitic eruption on Lanzarote in the Canary Islands [Carracedo and Badiola, 1991; Carracedo et al., 1992; Sigmarsson et al., 1998; Thomas et al., 1999], (3) Quaternary alkalic basalts erupted from the Fort Selkirk volcanic complex, Yukon, Canada [Francis and Ludden, 1990], and (5) tholeiitic basalts of the historic Mauna Ulu and Pu’u O’o eruptions of Kilauea volcano, Hawaii [Wright et al., 1975; Hofmann et al., 1984; Garcia et al., 1992,
1996, 2000]. Several of the most salient characteristics of each of these eruption sequences are described below; further details and discussion of previous interpretations related to the eruption sequences are presented in Appendix A.

[7] The Koloa volcanics eruption sequences included here are nephelinitic and alkalic basaltic lava sequences sampled by subsurface drilling in the Lihue basin [Reiners and Nelson, 1998; Reiners et al., 1999]. Each sequence probably represents cogenetic magmas erupted from individual vents that were probably active of the order of years. In each sequence, incompatible trace elements such as LREE, Th, Zr, P, and Sr decrease with stratigraphic height (increasing time in eruption sequence). In the nephelinite sequence, MgO also decreases with time, as SiO₂ and Al₂O₃ increase. Aside from P₂O₅, major elements in the alkalic basalt sequence do not show systematic trends with time. Samples from Koloa volcanics in general show weak but statistically significant correlations between major and trace elements and Sr and Nd isotopic compositions (Figure 2). Lassiter et al. [2000] also showed that both the Koloa volcanics and their analogue on Koʻolau Volcano on the island of Oahu (Honolulu volcanics; Clague and Frey [1982]) display good correlations between major and trace element compositions and Os isotopic compositions, with incompatible-element depleted and SiO₂-enriched samples having high ¹⁸⁷Os/¹⁸⁸Os (Figure 3).

[8] The Lanzarote 1730–1736 eruption produced 3–5 km³ of magma that systematically changed from nephelinite to olivine tholeiite during the course of the eruption [Carracedo and Badiola, 1991; Carracedo et al., 1992]. Incompatible trace elements systematically decreased in successively erupted melt batches (Figure 1), as did K₂O, P₂O₅, and MgO (14–8 wt %), while SiO₂ and Al₂O₃ increased (41–51 wt % and 10.7–13.7 wt %, respectively). Sigmarsson et al. [1998] documented a strong correlation between ⁸⁷Sr/⁸⁶Sr and ²³⁰Th/²³⁸U during the eruption [Sigmarsson et al., 1998; Thomas et al., 1999], while (²²⁶Ra/²³⁰Th) increased with time [Sigmarsson et al., 1998]. Published Os isotope data show that ¹⁸⁷Os/¹⁸⁸Os also displays a large increase with time during the eruption, from 0.129 to 0.157 [Widom et al., 1999], although another data set shows a more complex temporal progression in the latest stages of the eruption (J. Lassiter, unpublished data, 2001).

[9] Pisgah and Amboy craters are Quaternary cinder cones that each erupted a single sequence of alkalic basalts over a period of 10–20 years [Glazner et al., 1991]. Each sequence shows systematically decreasing MgO (~8–5 wt %) and incompatible elements and increasing SiO₂ (~46–51%) and Al₂O₃ (~15–17 wt %) (Figure 1). The ⁸⁷Sr/⁸⁶Sr and εNd are higher and lower, respectively, in these sequences than in others compiled here and show good correlations with major and trace element composition (Figure 2). The MgO of some of the Pisgah and Amboy crater lavas are lower than other magmas compiled here, but no attempt was made to correct for fractional crystallization because compatible and incompatible elements are positively correlated in these lavas, inconsistent with the expected effects of crystallization.

[10] The Fort Selkirk volcanic complex comprises five different vents and eruption sequences [Francis and Ludden, 1990]. There are systematic upsection decreases in whole-rock alkalinity in the four longest sequences (no change in the shortest one), with compositions ranging from basanite through low-Mg alkali olivine basalt (AOB). Although detailed stratigraphic information is not provided by Francis and Ludden [1990], temporal trends within most of these sequences include decreasing incompatible elements, MgO, and alkalinity, and increasing SiO₂ with time (e.g., Figure 3 in Francis and Ludden [1990, pp. 375–376]). The ⁸⁷Sr/⁸⁶Sr and εNd are well correlated with trace element and major element compositions in the Fort Selkirk lavas (Figure 2).

[11] Although both of the historic Kilauea eruptions, from Mauna Ulu and Puʻu Oʻo, are much more compositionally uniform than other eruption sequences compiled here, they both show temporal-compositional changes [Garcia et al., 1992, 1996, 2000] that are very similar to other sequences. Mauna Ulu (1969–1974) produced ~ 0.16 km³ of
Concentrations of elements and oxides with time in eruption sequences, normalized to the maximum value during each eruption (left to right is old to young in each panel). (a) $P_2O_5/P_2O_5^{\text{max}}$, (b) $Rb/Rb^{\text{max}}$ and $Th/Th^{\text{max}}$, (C) $SiO_2/SiO_2^{\text{max}}$. $Th$ is used for the Koloa volcanics sequences rather than $Rb$, owing to low-$T$ alteration effects on $Rb$ in these samples [Reiners and Nelson, 1998]. Within each eruption sequence there is an overall decrease in most incompatible elements and an increase in $SiO_2$ from the earliest to latest erupting lavas. In most sequences these trends are also accompanied by decreasing $MgO$ and $K_2O$ and increasing $Al_2O_3$. Detailed temporal control is not published for the Fort Selkirk magmas, but similar decreases in incompatible elements and $MgO$ and increases in $SiO_2$ are shown and described for these eruption sequences of Francis and Ludden [1990, Figure 3 and pp. 375–376]. Lavas from Amboy crater are also not shown, owing to uncertainties in stratigraphic control [Glazner et al., 1991]. Dashed lines across Pu‘u O‘o compositions represent the initiations of episodes 11 and 30, which have been interpreted as the end of petrographic and compositional (respectively) evidence of mixing between mafic, mantle-derived, and shallow, fractionated, magma [Garcia et al., 1992]. Pu‘u O’o trend is shown through September 1998 (most recent reported data of Garcia et al. [2000]).
basalt almost entirely from a single vent; only the chemically well-characterized 1969–1971 phase is considered here [Wright et al., 1975; Hofmann et al., 1984]. Although isotopic data are not available for this eruption, fractionation-corrected lava compositions display decreasing incompatible trace elements and P2O5 and K2O and increasing SiO2 and Al2O3, with time (Figure 1). The Pu‘u O‘o eruption began in 1983 and continues to the present, having erupted more than ~2 km2 of basalt (only published compositions (through September 1998) are shown here). Fractional crystallization of olivine has affected most Kilauea lavas, although 67% of Pu‘u O‘o samples have MgO >8 wt % and 93% have MgO > 7 wt % [Garcia et al., 1992, 1996, 2000]. Compositions shown here have been corrected for olivine (and a small amount of Cr-spinel) fractionation, to constant MgO of 10 wt % [Wright, 1971; Garcia et al., 2000]. Early-erupted Pu‘u O‘o basalts have fractionated beyond olivine control and are thought to have mixed with fractionated magma residing in the rift zone prior to eruption [e.g., Garcia et al., 1992]. The overall temporal-compositional trends cannot be attributed to this mixing process, however, and must reflect changing mantle melting or source controls [Garcia et al., 1992, 1996, 2000]. Fractionation-corrected lava compositions from Pu‘u O‘o display trends of decreasing incompatible trace elements and P2O5, K2O, and CaO, and increasing SiO2 with time (Figure 1). Pu‘u O‘o magmas show very little Sr, Nd, or Pb isotopic variation throughout the sequence, especially after the first 2 years of eruption [Garcia et al., 1992, 1996, 2000], and what little variation present is only weakly, if at all, correlated with other compositional characteristics.

3. Common Temporal-Compositional Trends

[12] Despite a wide range of geologic settings and magma types in each of the above eruption sequen-
ces, they share several common temporal-compositional trends. The most robust of these is an overall decrease in incompatible trace element concentrations (those with bulk crystal/melt distribution coefficients about $\sim 0.1$) from beginning to end of each eruption (Figure 1). A wide range of incompatible elements shows these trends in nearly all the sequences (e.g., La, Th, Rb, P, Sr, Ba, Nb, Zr, K, etc.). Decreases of greater magnitude are generally associated with more incompatible elements (e.g., in Lanzarote a factor of 2–4 for Th, La, Rb, Ba, K; a factor of 1.5–2 for Zr, Y), so that ratios of highly to moderately incompatible elements show similar decreases with time (e.g., Ba/Zr, Rb/Y, Nb/Zr, La/Sr, etc.). Compatible trace elements such as Ni and Cr show decreases with time in some of the eruption sequences, including Pisgah (Ni from $\sim 130$ to $\sim 50$ ppm), Lanzarote (Ni from $\sim 390$ to $\sim 240$ ppm), and Koloa nephelinites (Ni from $\sim 370$ to $\sim 200$ ppm). General lithologic differences in compatible trace element contents among Ft. Selkirk magmas suggest the same relationship of decreasing Ni and Cr during eruption sequences as well.

There are also common temporal-compositional trends in some of the major elements. Similar to all other highly incompatible elements, both K$_2$O and P$_2$O$_5$ show clear, generally large magnitude decreases with time in all the eruption sequences. In the Lanzarote eruption, both of these oxides decrease by $\sim 70\%$ from beginning to end of eruption. TiO$_2$ also decreases with time ($\sim 10$–$30\%$) in most of the sequences (but not Pu’u O’o or Koloa nephelinites). In all sequences except the Koloa alkalic basalts, SiO$_2$ contents gradually increase from beginning to end of eruption. SiO$_2$ increases vary from as little as $\sim 1\%$, in the Kilauea eruptions, to as much as $\sim 20\%$, in the Lanzarote eruption ($\sim 41$–$51\%$). In all but the Pu’u O’o eruption and the Koloa alkalic basalts, Al$_2$O$_3$ contents also increase with time, with increases ranging from $\sim 2\%$ (Mauna Ulu) to $\sim 20\%$ (Koloa nephelinites and Lanzarote). MgO decreases with time in all of the eruption sequences except for the Koloa volcanics alkalic basalts (although this is not possible to judge for the Kilauea sequences which are corrected to 10 wt % MgO). MgO decreases from $\sim 14$ wt % to

Figure 3. Os isotopic compositions versus SiO$_2$ and Zr for the posterosional Koloa volcanics from the Lihue basin, Kaua’i [Lassiter et al., 2000; Reiners and Nelson, 1998], and 1730-36 Lanzarote eruption (Os data from Widom et al. [1999]; concentration data from K. Hoernle (personal communication, 2000)). Posterosional lavas from Ko’olau volcano on Oahu (the Honolulu volcanics) show the same trends as the Kaua’i posterosionals [Lassiter et al., 2000]. Nearly all mantle peridotite samples have $^{187}\text{Os}/^{188}\text{Os}$ less than 0.130, while most pyroxenites have considerably higher values, typically 0.140–0.180 and higher [Lassiter et al., 2000]; the high $^{187}\text{Os}/^{188}\text{Os}$ of posterosional magmas suggest a large fraction is derived from pyroxenite. Positive correlations between $^{187}\text{Os}/^{188}\text{Os}$ and SiO$_2$ and Al$_2$O$_3$, and inverse correlations between $^{187}\text{Os}/^{188}\text{Os}$ and incompatible elements are consistent with a large degree of melting for the radiogenic Os source and a small degree of melting for the unradiogenic Os source.
8–9 wt % in Koloa nephelinites and Lanzarote and from ~8 to 6 wt % in the Pisgah and Amboy sequences. Interestingly, while K₂O shows clear trends, there are no discernable temporal trends in Na₂O in any of the sequences (though this is difficult to determine with certainty for the Ft. Selkirk lavas because of a lack of published stratigraphic information). CaO also does not show any consistent trend with time among the eruption sequences.

[14] In the Lanzarote and most of the Ft. Selkirk eruption sequences, lithologic changes accompany major and trace element changes with time. In both cases, rock types shift to progressively less alkaline lithologies. Lanzarote lavas change from nephelinite to tholeiite [Carracedo and Badiola, 1991; Carracedo et al., 1992], and Ft. Selkirk lavas change from nephelinite to basanite, basanite to alkaline basalt, and alkaline basalt to hypersthene-normative alkaline basalt [Francis and Ludden, 1990]. These nominal lithologic changes are simply normative consequences of relatively extreme major element changes with time that are exhibited to lesser degrees by nearly all the eruption sequences.

[15] Most of the eruption sequences (Pisgah, Amboy, Koloa, Ft. Selkirk, and Lanzarote if Sigmarsson et al.’s [1998] data are considered) show at least weak correlations between Sr and Nd isotopic compositions and major and trace element compositions (Figure 2), indicating more than one mantle source for the magmas, with different proportions from each source in melts of different major and trace element composition. In at least the cases of Lanzarote and the Koloa volcanics, Os isotopes show more consistent relationships with eruption sequence and magma composition than Sr and Nd. Specifically, higher 187Os/188Os is associated with higher SiO₂ and lower incompatible element concentration in both the Koloa and Lanzarote magmas, and this high 187Os/188Os signature is more prevalent in later erupting lavas in Lanzarote (Figure 3).

[16] Two separate studies have also shown that U series isotopes in the Lanzarote sequence show systematic temporal trends that are correlated with magma composition. As the eruption progressed, (²³⁰Th/²³⁵U) decreased (~1.4 to ~1.2) and (²²⁶Ra/²³⁰Th) increased (~1.1 to ~1.6), as SiO₂ increased and incompatible elements decreased [Sigmarsson et al., 1998; Thomas et al., 1999]. At present there are no other available data on U series isotopic disequilibria in other basaltic eruption sequences with temporal control. Given their young ages, these could be measured in the Kilauea sequences, and it is possible that at least some (²³⁰Th/²³⁸U) disequilibria may still be present in Pisgah/Amboy or Ft. Selkirk sequences, depending on their eruption ages.

4. Possible Origins of the Compositional Variation

[17] Production of remarkably similar temporal-compositional trends in these sequences (as well as others not discussed here but which show similar patterns possibly over different time scales [Segalstad, 1979; Maaløe et al., 1992; Wu, 1989; Anthony et al., 1989; Neumann et al., 1988; Thompson et al., 1972; Wood, 1976, 1979]), from a range of unrelated phenomena would be highly coincidental. I suggest that these trends are typical of monogenetic eruptions of primitive basalts such as these and that future studies of similar eruptions will also show trends of decreasing incompatible elements, increasing SiO₂, decreasing alkalinity, etc., with time, which cannot be attributed to crystallization or shallow- (crustal-) level processes. Although several potential origins for these temporal-compositional trends can be envisioned, a number of possibilities can be ruled out fairly simply, at least as a general origin for all the sequences discussed here.

4.1. Crystallization

[18] The largely primitive (or fractionation-corrected) compositions of these magmas preclude a significant role for fractional crystallization in generating the temporal-compositional trends of most elements within each eruption. The large ranges in incompatible elements in most of the sequences would require extents of fractionation large enough to produce severe depletions in
compatible elements and other compositional changes that are not observed. Also, MgO is positively correlated with incompatible elements in nearly all sequences, the opposite of what would be expected from fractional crystallization of olivine or pyroxene. Because the early stages of the Pu‘u O‘o eruption show petrographic and compositional features attributed to mixing with shallow, differentiated magma [Garcia et al., 1992, 1996, 2000], crystallization has been indirectly implicated as a partial source of chemical variation in one of the Kilauea eruptions. The overall nearly 20-year trend in this eruption cannot be attributed to this, however, and similar trends in other eruption sequences of clearly non-differentiated magmas suggest a common process is responsible.

4.2. Crustal Contamination

Several factors also make crustal contamination an unlikely explanation for compositional variation in most of these eruption sequences, including the massive amounts of crust that would be required to explain isotopic and trace element shifts (e.g., 80%) in some cases. Additionally, many of these lavas contain mantle xenoliths, indicating a rapid transit through the lithosphere. Rapid translithospheric melt velocities are also required by strong (226Ra/230Th) disequilibria, at least in the case of the Lanzarote eruption (minimum of ~0.5 km/yr). As discussed in the appendix, Glazner et al. [1991] advocated a model for temporal-compositional trends in the Pisgah and Amboy sequences that involved progressive addition of partial melt derived from mafic crust to erupting melt batches with time, although the crustal partial melt would have to have had very low incompatible element abundances. These sequences lack xenoliths and do not have U series disequilibria constraints, so it is difficult to rule out crustal residence times sufficient to produce contamination. However, as suggested by Glazner et al. [1991], it is also possible that this mafic source was located in the mantle rather than the crust. A higher-pressure garnet-pyroxenite mode and composition (rather than gabbroic crust) would also be easier to reconcile with the low incompatible elements required for this end-member.

4.3. Partial Melting of Single Source

Varying degrees of partial melting of a single mantle source has been proposed for the origin of compositional variation in some of the eruption sequences [Hofmann et al., 1984; Thomas et al., 1999]. Both trace element and isotopic variations preclude this for all suites except possibly the Mauna Ulu suite (which does not yet have isotopic data) for the following reasons. First, although some eruption sequences do not show significant radiogenic isotope variation (e.g., Pu‘u O‘o), all other eruption sequences display correlations between radiogenic isotopes and at least some trace and major element concentrations (Figure 2). This requires at least two compositionally distinct sources of melt in each sequence or a range of melt sources with a correlation between isotopic composition and either bulk composition or degree of melting. In the case of the Lanzarote eruption, only the Sigmarsson et al. [1998] data show Sr isotope variation being correlated with magma composition, but there is clearly significant Os isotope variation in this suite that is correlated with magma composition as well (Figure 3).

Partial melting of a single mantle source is also difficult to reconcile with trace element relationships in all of the eruption sequences (Figure 4). Model partial melting trends for typical mantle conditions are shown in Figure 5, along with fields of eruption sequence data shown in Figure 4. Single-source batch, fractional, and dynamic melting models of garnet or spinel peridotite sources with any reasonable range of modes, initial trace element compositions, and bulk distribution coefficients predict trends of highly versus moderately incompatible elements that are significantly curved, especially at degrees of melting below ~5–10%. This is because the rate of depletion of the highly incompatible element with progressive melting is higher than that of the moderately incompatible element. As is common in many mafic alkalic basalts, however, trace element variations in these eruption sequences do not show any systematic curvature and instead, where recognizable trends
emerge from scatter, they are quasi-linear (Figure 4 and 5; Reiners [1998]; Kamber and Collerson [2000]). At relatively high degrees of melting, where melting trends show less curvature, the model trends are generally offset to lower abundances of the highly incompatible element than the eruption sequence trends. The only way to force single-source partial melting trends through most of the quasi-linear eruption sequences trends in Figure 4 and 5 is to invoke an extremely incompatible element enriched source ($\sim 5-12$ times chondrites, with higher factors for more incompatible elements), a large range in extent of partial melting ($\sim 2-20\%$), and a mode with very little clinopyroxene or garnet ($<5-8\%$ of both). Similar models of high degrees of melting of an (usually recently) incompatible-element enriched mantle source have been widely invoked for the origin of many oceanic mantle-derived alkalic basalts in the past [e.g., Clague and Frey, 1982; Roden and Murthy, 1985; Frey and Roden, 1987; Clague and Dalrymple, 1988; Reiners and Nelson, 1998; Frey et al., 2000].

### 4.4. Melt Mixing

[22] Linear trace element trends, such as those in Figure 4, and common correlations between element concentrations and isotopic compositions strongly suggest a dominant role for binary melt mixing in producing the observed chemical variation [Francis and Ludden, 1990; Glazner et al., 1991; Sigmarsson et al., 1998; Thomas et al., 1999; Reiners and Nelson, 1998]. The origin of the hypothetical end-member melts in each of these studies has varied widely, however, from melts of lithologically distinct mantle domains, to melts from different depths in an upwelling mantle column, to melts from both mantle and crustal sources. In order to investigate the plausibility of a common binary melt mixing model for variation in all the eruption sequences, below I summarize the general compositional characteristics of the hypothetical end-member melts that would be required, without assigning genetic significance to them.

[23] The simplest melt mixing scenario would require two distinct end-member melts whose mixing form the trace element arrays in Figure 4 and whose mixing proportions change during eruption sequence, producing the temporal-compositional trends in Figure 1. Figure 4 suggests that one of the most striking differences between these end-member melts is contrasting incompatible element concentrations, as suggested by some of original studies [Francis and Ludden, 1990; Sigmarsson et al., 1998; Thomas et al., 1999].

**Figure 4.** Highly versus moderately incompatible trace elements ((top) Nd versus La (ppm); (bottom) Zr versus Nb (ppm)) for eruption sequences shown in Figure 1 (symbols and data as shown in Figure 1). Also shown are data from Oslo rift lavas (crosses) from Anthony et al. [1989], which show an overall temporal-compositional trend in incompatible and some major elements similar to those shown in Figure 1. Highly and moderately incompatible trace elements from these eruption sequences generally show quasi-linear trends suggesting a mixing origin for compositional variation, as suggested by some of original studies [Francis and Ludden, 1990; Sigmarsson et al., 1998; Thomas et al., 1999].
Figure 5. Partial melting models, with fields of data shown in Figure 4 (lighter grey field is Pu‘u O‘o eruption sequence). Partial melting of any single source predicts curved trends for highly versus moderately incompatible elements such as these. In contrast, data from these eruption sequences and other similar magmas show quasi-linear trends, suggesting mixing. Solid curved lines are nonmodal batch partial melting trends for pyroxenite (red) and peridotite (blue), for degree of melting (∘) from 0–15% (upper right to lower left). Melting trends for three different peridotite modes are shown, with the following modal compositions of olivine:orthopyroxene:clinopyroxene:garnet: 0.50:0.25:0.20:0.05 (peridotite 1); 0.58:0.27:0.13:0.02 (peridotite 2); 0.65:0.28:0.05:0.02 (peridotite 3). Pyroxenite mode is assumed to be 0.75:0.25 (clinopyroxene:garnet). Melting of peridotite with both primitive and depleted mantle compositions are shown (McDonough and Sun, 1995; Geochemical Earth Reference Model (GERM) website available at http://www-ep.es.lnl.gov/germ/germ-home.html), and pyroxenite is assumed to have the composition of average mantle pyroxenite in the dataset of Hirschmann and Stolper [1996] (excluding eclogites and the REE-enriched samples of Griffin et al. [1988]). This yields a pyroxenite composition of 0.844 ppm La, 2.47 ppm Nd, and 19.0 ppm Zr. Nb of pyroxenite was assumed to be 0.80 ppm (intermediate between GERM values for primitive mantle (0.66 ppm), cumulate pyroxenites (0.135 ppm), depleted mantle (0.112 ppm), and MORB (3 ppm)). The trace element partial melting trends for the lithologies and compositions shown here do not change appreciably with different types of melting (batch, fractional, dynamic) or with different solid/melt distribution coefficients. Dashed green lines are contours of constant degrees of partial melting for the peridotitic sources; % partial melting is noted next to each contour. Percent partial melting of the pyroxenite source is noted next to tick marks on melting trend. Dashed grey line is trend of mixing between 0.2 and 15% peridotite 3 melt. Mixing between 0.2% peridotite melt and high degree pyroxenite melt (> about 5%) would be essentially collinear. Peridotite solid/melt distribution coefficients (K∫’s) for cpx and garnet are from Salters and Longhi [1999], except La which was not given by that study, and was assumed here to be 0.015 in cpx and 0.02 in garnet. Peridotite K∫’s for olivine and opx are assumed to be 0 and 0.001, respectively. Pyroxenite K∫’s for Nb and Zr in cpx and garnet are from Zack et al. [1997] (which, for cpx are the same as those in Hart and Dunn [1993]). Pyroxenite K∫’s for La and Nd in all phases are averages of those of McKenzie and O’Nions [1991] and Frey et al. [1978].
require long-term source characteristics opposite of their melt composition characteristics). In detail, each eruption sequence does not span the same range of compositions, so these high and low incompatible element end-member melts may have different specific compositions for each sequence. In addition, trace element arrays of each eruption sequence in Figure 4 generally have similar intercepts but slightly different slopes (e.g., the Pu’u O’o sequence has a distinctly lower slope), suggesting that the composition of the melt 1 end-member may vary more widely than that of melt 2.

[24] Taken as a whole, the eruption sequences shown here require the early-erupting melt batches (dominated by the melt 1 end-member) to have relatively low SiO$_2$ (< about 41–48 wt %) and Al$_2$O$_3$ (< about 10–12 wt %), and high K$_2$O (> about 0.5–2 wt %), MgO (> about 8 wt %), TiO$_2$ (> about 2.5–3 wt %), and P$_2$O$_5$ (> about 0.3–0.9 wt %). The late-erupting melt batches (dominated by the melt 2 component) are characterized by relatively high SiO$_2$ (> about 45–52 wt %) and Al$_2$O$_3$ (> about 12–16.5 wt %), and low K$_2$O (< about 0.3–1.0 wt %), MgO (< about 8 wt %), TiO$_2$ (< about 1.6–2.2 wt %), and P$_2$O$_5$ (< about 0.2–0.3 wt %).

[25] Figure 3 shows that while most of the eruption sequences display correlations between Sr and Nd isotopic compositions and major and trace element compositions, these correlations are not consistent among sequences and some, such as Pu’u O’o, do not display significant correlations. In the context of a binary melt mixing scenario, this means that the melt 1 and melt 2 sources often possess different Sr and Nd isotopic compositions, but the contrast in Sr and Nd isotopic compositions of the two sources is variable in sense as well as in magnitude.

[26] In contrast to Sr and Nd, the sequences that provide constraints on the Os isotopic compositions of the melt end-members (Koloa and Lanzarote) clearly suggest a significant difference between $^{187}$Os/$^{188}$Os of the melt 1 and melt 2 sources (Figure 3). Melt 1, the early-erupting, incompatible-element enriched component, has low $^{187}$Os/$^{188}$Os (< about 0.13), while melt 2, the late-erupting, incompatible-element depleted component, has high $^{187}$Os/$^{188}$Os (>0.16) [Widom et al., 1999; Lassiter et al., 2000]. Given that $^{187}$Os/$^{188}$Os of a primary melt is a measure of the time-integrated Re/Os of its source and that Re is moderately incompatible while Os is highly compatible, it is somewhat surprising that the incompatible-element enriched component (melt 1) would have the low $^{187}$Os/$^{188}$Os, while the incompatible-element depleted component (melt 2) has high $^{187}$Os/$^{188}$Os.

[27] Only one eruption sequence (Lanzarote) constrains the potential contrasts between U series disequilibria signatures of the melt 1 and melt 2 components. If the characteristics of this sequence are applicable to the others, it requires the early-erupting melt 1 to have relatively high ($^{230}$Th/$^{238}$U) and low ($^{226}$Ra/$^{230}$Th) and melt 2 to have the opposite characteristics [Sigmarsson et al., 1998; Thomas et al., 1999]. Both activity ratios of each of the end-members would still be greater than unity, however.

4.5. Melt Mixing: Trace Element Constraints on End-Member Melt Component Sources

[28] Simple trace element modeling of mantle melting broadly constrains some essential characteristics of potential sources of end-member melt components. Figure 5 shows partial melting trends of peridotite and pyroxenite sources with various modes and compositions appropriate for the upper mantle. An important first-order observation from trace element model results such as those shown in Figure 5 is that despite lower concentrations of incompatible elements in peridotite, low-degree melts of peridotite can have higher incompatible element concentrations than pyroxenite-derived melts. This is because of the relatively high clino-pyroxene/melt and garnet/melt distribution coefficients for moderately incompatible elements. Using an average mantle pyroxenite composition for pyroxenite (Figure 5) and primitive mantle for peridotite, peridotite melts formed by up to 2% melting will have higher La, Nd, and Zr concentrations than pyroxenite melts of any extent of melting. One reason this is significant is that many
studies have assumed that pyroxenite-derived melts will generally be incompatible-element enriched relative to peridotite-derived melts. As Figure 5 shows, this is not necessarily true especially if these sources are melting in the same region, as pyroxenite melts to larger degrees than peridotite at the same conditions [Hirschmann and Stolper, 1996; Pertermann and Hirschman, submitted manuscript, 2000]. Figure 5 also shows a general extension of this observation: for a given source composition, sources with lower abundances of clinopyroxene and garnet (mineralogically infertile sources) produce low-degree melts with higher incompatible element concentrations. Mineralogically infertile (clinopyroxene-depleted) peridotite often has similar or higher incompatible element concentrations than fertile peridotite [Frey and Prinz, 1978; Galer and O’Nions, 1989], which suggests that low-degree, incompatible-element enriched melts may have mineralogically infertile peridotite sources (e.g., harzburgite).

In the context of a binary melt mixing scenario involving fixed composition end-members, Figure 5 shows that the end-member component with high incompatible elements (melt 1) is not consistent with melts of typical depleted mantle at any degree of melting. It is, however, consistent with compositions of small-degree melts (0.1–0.5%, but ~2–5% for Pu’u O’o) of peridotite with higher incompatible element concentrations (e.g., primitive mantle) provided the peridotite mode is mineralogically infertile. Larger degree melts could also serve as this end-member if the initial source had higher incompatible element concentrations. In contrast to melt 1, the low incompatible elements of the inferred melt 2 component require it to be a large degree melt. If the melt 2 source is peridotite, the degree of melting must be >15% and in some cases >20–25%. If the melt 2 source is pyroxenite, however, degrees of melting of 5–10% or greater produce melts with appropriate trace element compositions.

In summary, compositional characteristics of the basaltic eruption sequences point to a common origin of binary melt mixing as the dominant control on compositional variation in each sequence. This is supported by isotopic-concentration correlations suggesting isotopically distinct sources for each end-member melt. Simple trace element modeling indicates that the end-member melt 1 and 2 components are most likely derived by strongly contrasting degrees of melting and that the low melting degree component (melt 1) is from a peridotite source with low modal abundances of clinopyroxene and garnet but a composition more enriched in incompatible elements than typical depleted mantle. The melt 2 component is most likely derived from large degrees of melting of a separate source, which may be peridotite or another lithology than produces melts with relatively low incompatible element concentrations. Before further discussing the potential origins of the end-member melts, however, it is useful to return to the question of the significance of the temporal-compositional trends in each eruption sequence.

### 5. Significance of Temporal-Compositional Trends

Even allowing for reasonable ranges in compositions of different mantle lithologies, the large difference in incompatible element contents of the hypothetical melt end-members 1 and 2 requires that an important distinction between them is degree of melting. Thus any explanation for the temporal-compositional trends must provide a mechanism for progressive eruption of melt batches formed by an increasing proportion of the high-degree melting component. The nature and locations of each of the sources and the reason for their apparently distinct melting behavior is more difficult to constrain without some speculation, but two possibilities can be easily imagined.

#### 5.1. Melt Mixing During (Translithospheric?) Migration/Ascent

One possible scenario is that one mantle source is “original” and relatively deep and melts to large degrees, while the other is shallow and produces only small-degree melts that mix with ascending primary melts. In this case, the shallow, small-degree melts (melt 1) is progressively exhausted during melt migration and eruption so that late-erupting melts contain the largest proportion of the
large-degree, primary melt (melt 2). Assuming typical peridotitic sources, most of the eruption sequence arrays in Figure 4 can be explained by mixing 0.2% (melt 1) and 15% (melt 2) melts in proportions ranging from maximum and minimum ratios of 50:50 to 10:90, respectively. The Pu’u O’o sequence would require a larger degree of melting for the (melt 1) low-degree end-member (~2–5%), and changing melt 1:melt 2 proportions with time from about 50:50 to 25:75, respectively.

This scenario is similar in some respects to petrogenetic models for the volcanics of Grande Comore, in the Indian Ocean [Class and Goldstein, 1997; Class et al., 1998; Bourdon et al., 1998; Claude-Ivanaj et al., 1998]. Primitive magmas from La Grille Volcano on Grand Comore have been shown to be small-degree (1–5%) partial melts of amphibole-bearing peridotite, whereas magmas from an adjacent volcano are primarily larger degree melts of an isotopically distinct source but containing some of the La Grille component. Importantly, trace element signatures (e.g., low K/Nb, Ba/Th) require the small-degree La Grille melts to be formed in the presence of residual amphibole [Späth et al., 1996], essentially limiting it to P-T conditions found in the lithosphere [Class and Goldstein, 1997; Class et al., 1998]. Class and Goldstein’s [1997] model for binary mixing trends of Grand Comore lavas therefore involved ascent of large-degree asthenospheric melts through channels in amphibole-bearing (metasomatized) lithosphere, which produced small-degree partial melts of the lithosphere that mixed with migrating asthenospheric melts. The mechanism of component mixing is constrained to be melt-melt mixing by both trace element and isotopic (especially Os) variations [Class and Goldstein, 1997] that are inconsistent with predicted effects of melt-solid reaction [Hauri, 1997; Reiners, 1998] or solid-solid mixing [Kogiso et al., 2000], similar to variations in these sequences. These small-degree lithospheric melts would be expected to have high incompatible element signatures (especially in the case of metasomatized lithosphere) that could strongly affect compositions of the mixed melts. This petrogenetic model was supported by recent U-Th-Pa-Ra isotope results, including relative (230Th) and (226Ra) excesses requiring an amphibole-bearing source [Bourdon et al., 1998; Claude-Ivanaj et al., 1998].

The Grand Comore model is important in the context of the temporal-compositional trends of this study because is conceivable that during melt migration and ascent, amphibole and easily fused components in lithospheric channels could become progressively exhausted. Thus initial melt batches to migrate through lithospheric channels would have the highest incompatible element concentrations and, if amphibole were present in the lithosphere, the strongest amphibole signature. This could potentially impart inverse temporal-compositional trends in (230Th/238U) and (226Ra/230Th) as observed in the Lanzarote eruption [Sigmarsson et al., 1998]. K/Nb and Ba/Th also show weak increases with time in the Lanzarote sequence, which could be due to residual amphibole. The presence of a residual K-bearing phase in the end-member melt with high incompatible element concentrations (melt 1) has also been implicated in the case of the Ft. Selkirk and Koloa magmas [Clague and Frey, 1982; Francis and Ludden, 1990, 1995; Reiners and Nelson, 1998].

Other features of the temporal-compositional trends may also be consistent with a lithospheric origin for the early-erupting, incompatible-element enriched melt component. If the lithospheric source is peridotite metasomatized by a carbonate-rich component, as envisioned in some of the above studies and suggested in other similar melt-mixing models [Kamber and Collerson, 2000], it may be expected to produce melts with low SiO2 and Al2O3, and high MgO [Hirose, 1997]. Os isotopic compositions of lithospheric peridotite, metasomatized or not, would likely also be unradiogenic [Lassiter et al., 2000], similar to the early-erupting melts with low 187Os/188Os in these sequences.

Several features of the temporal-compositional trends of the eruption sequences, however, are difficult to reconcile with this model. First, nearly all the eruption sequences show decreasing MgO with time, pointing to an approximate MgO content of the melt 2 component of ~8 wt % or less. If melt 2 were distinguished from melt 1 primarily by larger degrees of melting at greater depths, it would be
expected to have MgO considerably higher than 8 wt %. For example, Walter [1998] shows MgO of ~17–25 wt % at 15% melting and 3–7 GPa, and Kushiro [1996] shows MgO of ~12–18 wt % at 3 GPa. Only if peridotite melted to less than ~5% at relatively low pressures (1 GPa) would primary melt be expected to have MgO ~8 wt % [Baker and Stolper, 1994; Baker et al., 1995; Hirschmann et al., 1998a, 1998b]. It may be possible, however, that limited crystal fractionation of the melt 2 component could occur at depth or during migration; this is consistent with both low MgO and compatible trace element contents of this end-member.

[37] Perhaps more problematic for this scenario is the Os isotopic composition of the melt 2 end-member indicated by the large-degree, late-erupting melts in both the Koloa and Lanzarote eruptions. In both cases, the melt 2 component has \(^{187}\text{Os}/^{188}\text{Os}\) greater than 0.16 (Figure 3), much greater than the maximum \(^{187}\text{Os}/^{188}\text{Os}\) of 0.13 observed for nearly all mantle peridotites [Roy-Barman and Allègre, 1994; Shirey and Walker, 1998; Lassiter et al., 2000; Brandon et al., 2000; Meisel et al., 2001]. The late-erupting, radiogenic Os melts in the Lanzarote eruption are not obviously contaminated by disaggregated xenoliths, as the early-erupting melts with unradiogenic Os may have been, so these radiogenic Os signatures are likely a primary feature of the magma. As suggested by Lassiter et al. [2000], radiogenic Os signatures of the magnitude of the melt 2 end-member are unlikely to be explained by melting of any typical peridotite but are easily explained by melting of mantle pyroxenite. If the Koloa and Lanzarote eruption sequences are similar to the others in trending to radiogenic Os (>0.13) with later-erupted melts, then an origin involving simply mixing asthenospheric peridotite melts with melts of overlying lithosphere cannot adequately explain the temporal-compositional trends. In some respects this bears on a larger issue of Os isotopes and peridotite sources in mantle petrogenesis, as in section 5.2.

5.2. Lithologically Heterogeneous Source

[38] Another scenario that could potentially produce temporal-compositional trends of the type observed in these eruptions sequences is that the melt 1 and melt 2 sources are derived from lithologically distinct domains that are mixed on a meter to kilometer scale. As discussed in Section 5.1, mixed peridotite-pyroxenite sources could theoretically produce end-member melts with strongly varying melt fractions and contrasting major and trace element compositions. Differing time-integrated parent/daughter ratios in peridotite and pyroxenite would also be expected to lead to contrasting radiogenic isotopic compositions, especially for the Re/Os system [Lassiter et al., 2000]. As shown by Hirschmann and Stolper’s [1996] compilation, however, Sm/Nd of average mantle pyroxenite and depleted mantle are not significantly different, and many pyroxenites have Sm/Nd higher than depleted mantle, so Nd isotopic compositions of pyroxenite and peridotite (assumed to have depleted mantle characteristics) may not be consistently distinct. The Rb/Sr system is probably similar in this regard. Other first-order characteristics of large-degree pyroxenite-derived melts that would distinguish them from small-degree peridotite-derived melts include (1) higher SiO\(_2\) and Al\(_2\)O\(_3\) and lower MgO [Hirschmann and Stolper, 1996; Pertermann and Hirschman, submitted manuscript, 2000], (2) higher \(^{187}\text{Os}/^{188}\text{Os}\), and possibly other radiogenic isotope differences [Lassiter et al., 2000], and (3) lower concentrations of incompatible trace elements (Figure 5). All of these are characteristics of the late-erupting melt batches of the eruption sequences shown in this study. Figure 5 also shows that mixtures of small-degree peridotite melts and large-degree pyroxenite melts can explain the overall trace element variation in eruption sequences. If a mixed pyroxenite-peridotite source can explain the overall chemical variation in the eruption sequences, then what mechanism would produce an increasing proportion of pyroxenite-derived melt with time in each eruption sequence? This may reflect the compositional distribution of melt in a region of upwelling, melting mantle.

[39] In an upwelling melting column of mixed peridotite and pyroxenite domains, the distribution and composition of melt vary as a function of depth. For typical mantle potential temperatures,
pyroxenite crosses its solidus at depths $\sim 35-50$ km deeper than peridotite [Hirschmann and Stolper, 1996; Pertermann and Hirschman, submitted manuscript, 2000] and may be largely molten (>50%) at the depth of incipient peridotite melting (Figure 6). Provided the pyroxenite domains are at least 1–10 m in size, they will not diffusively equilibrate with surrounding peridotite, and they will retain distinct geochemical signatures [Kogiso et al., 2000]. With continued upwelling, peridotite crosses its solidus and begins to melt, but because peridotite adiabatic melt productivity is extremely low ($dF/dP \sim 2\%/$GPa) near the solidus [Asimow et al., 1997; Hirschmann et al., 1999], it may not melt to fractions greater than 2% for another 30–45 km of upwelling. In typical intraplate settings in which the lithosphere is 30–90 km thick, this may limit the extent of peridotite melting to small degrees and restrict it to a relatively thin layer overlying a thick pyroxenite melt zone. Qualitatively this would lead to a mantle-melting column with a deep zone of largely melted pyroxenite, overlain by a zone of both large-degree pyroxenite melt and small-degree peridotite melt (Figure 6). If eruption from this melting region extracts incompletely mixed melts from increasing depths, it will result in a temporal-compositional trend of decreasing proportion of small-degree peridotite melt and increasing proportion of large-degree pyroxenite melt. This could produce decreasing incompatible element concentrations, changes in major elements that reflect partly an increasing degree of melting and partly a shift from peridotite to pyroxenite, and changes in isotopic compositions of elements, such as Os, that differ between pyroxenite and peridotite.

The first-order trace element predictions of this scenario can be simply modeled by assuming a mixed-source mantle with 99.5% peridotite and 0.5% pyroxenite, melt productivities of the different lithologies, and other parameters the same as in Figure 5. Intraplate basalts of this study were erupted over fairly old lithosphere (Cretaceous or older) that may be 50–100 km thick. Pyroxenite was assumed to begin melting at 150 km, 35 km deeper than peridotite, and to melt at a rate of $dF/dz = 0.4\%$/km upon ascent, close to the near-solidus adiabatic melt productivity of Pertermann and Hirschman (submitted manuscript, 2000). Peridotite was assumed to cross the solidus at 115 km depth and melt at an increasing rate at shallower depths ($F = -2.5 \ln P + 3.1$, where $F$ is percent melt fraction and $P$ is in GPa). The main results of this model are not changed significantly if the depths of melting are shifted (causing more melting to occur in the spinel stability field) nor are general results sensitive to melting region geometry.

Several salient features of the model are shown in Figure 6. Figure 6c shows volume of melt as a function of depth, for different final depths of melting. The lower, pyroxenite-only melt zone is distinguished from the upper, mixed pyroxenite-peridotite melt zone by a distinct increase in melt volume at the depth of peridotite solidus intersection (115 km). The final depth of upwelling and melting (and therefore distribution and composition of melt as a function of depth) can be considered a function of different lithospheric thicknesses at constant mantle potential temperature or, if melting depths are shifted, different mantle temperatures. In regions of relatively thick lithosphere (or cold mantle), the final depth of upwelling and melting will be deep, the upper zone of mixed peridotite-pyroxenite melt will be thin, and peridotite may melt to only a small degree (“cold colors” in Figure 6c). In regions of relatively thin lithosphere (or warm mantle), the final depth of upwelling and melting will be shallow, the zone of mixed peridotite-pyroxenite melt will be thicker, and the degree of melting of each lithology will be more similar (“hot colors” in Figure 6c). Assuming a source with only 0.5% solid pyroxenite and final melting depths of $\sim 60–100$ km, the volumes of melt present below and above the depth of peridotite solidus intersection will be similar within about a factor of five (Figure 6c). Concentrations of incompatible elements in melt in this column are highest near the depth of peridotite solidus intersection and decrease both below and above this depth (Figure 6d).

Figure 7 shows trace element compositions of melts formed in the mixed-source melting column depicted in Figure 6. If incremental melt batches at each depth in the column do not mix prior to eruption but are sequentially extracted from pro-
Figure 6. Schematic characteristics of mixed pyroxenite-peridotite source melting model. (a) Cartoon of upwelling mantle melting column [after Hirschmann and Stolper, 1996]. Pyroxenite and peridotite solidi are assumed to be at 150 and 115 km, respectively (Pertermann and Hirschman, submitted manuscript, 2000) estimate a 35–50 km gap between the peridotite and pyroxenite solidi at 2–3 GPa). The lower part of the column (red) comprises pyroxenite melt only, while the upper part (blue) includes both pyroxenite and peridotite melt. In this particular cartoon, upwelling and melting are assumed to cease at 75 km, which could be due to an overlying lithosphere of that thickness, but this final depth of melting (and therefore the thickness of the upper, mixed peridotite-pyroxenite melt zone) is varied in the model. (b) Assumed adiabatic melt productivities for each source: 13%/GPa for pyroxenite, and $F = -2.5 \ln (P) + 3.1$ (where $P$ is GPa and $F$ is % melt fraction), for peridotite. The pyroxenite productivity is the near-solidus productivity for Pertermann and Hirschman’s (submitted manuscript, 2000) G2 pyroxenite, and the $dF/dP$ of the peridotite is similar (e.g., low productivity at melt fractions less than about 2–5%) to that of Hirschmann et al. [1999, Figure 2]. Melting of both lithologies is assumed to cease at final depth of melting even though melt productivity curves in this cartoon are shown extrapolated to the 0 km depth. (c) Volume of melt, assuming axisymmetric cylindrical pyramidal melting region, at different depths in a mantle melting column comprising 0.5% pyroxenite and 99.5% peridotite. The change in shape of the melt volume curve at ~115 km represents the onset of peridotite melting at that depth. Melt volumes decrease with decreasing depth because of the triangular shape of the melting region. Final depths of melting between ~60–100 km produce volumes of melt above and below the peridotite solidus that vary by less than about a factor of five. (d) Concentration of La (ppm) in pyroxenite melt (red), peridotite melt (blue), and mixed melt (green), at each position in the melting column. The concentration of incompatible elements is highest in the melting column just above the peridotite solidus depth. This example is shown for upwelling and melting all the way to 0 km depth.
Figure 7. Partial melting and mixing model results, with fields of data shown in Figure 4 and 5. Solid curved lines are nonmodal batch partial melting trends for pyroxenite (red) and peridotite 3 of Figure 5 (blue), for melting ($F$) from 0–15% (upper right to lower left). Solid black lines are mixing trends between aggregate melt of pyroxenite-only melting zone (point P) and aggregate melt of upper, mixed peridotite-pyroxenite melting zone, for different final depths of melting. For point A, final depth of melting ($d_f$) is 109 km, where $F$ of peridotite ($F_{pr}$) is 0.11%, and $F$ of pyroxenite ($F_{px}$) is 14%. B: $d_f = 105$ km, $F_{pr} = 0.21\%$, $F_{px} = 18\%$; C: $d_f = 93$ km, $F_{pr} = 0.50\%$, $F_{px} = 22.5\%$; D: $d_f = 62$ km, $F_{pr} = 1.5\%$, $F_{px} = 35\%$; E: $d_f = 51$ km, $F_{pr} = 2.0\%$, $F_{px} = 39\%$; F: $d_f = 15$ km, $F_{pr} = 5.0\%$, $F_{px} = 53\%$. Mixing between upper and lower melting zone compositions produces linear trace element trends similar to those of most of the eruption sequences for final depths of upwelling of $\sim 60–100$ km, but Pu‘u O‘o requires shallower melting to generate mixing trend with lower slope. Squares along mixing lines represent composition of aggregate melt in entire melting column for each final depth of melting. Curved green line is trend of incremental melt compositions at each depth in melting column. In a column with a final depth of melting of 93 km, melt at the shallowest depth would have the composition at point c on the green line. Progressively deeper melts have compositions that fall along the green trend in a counterclockwise pattern, until reaching the pyroxenite melting curve. Other letters on green trend represent the compositions of shallowest melts in melting column with final depths corresponding to capital letters on mixing trends.
gressively greater depths, the sequence of erupted melt compositions falls along the green trend in Figure 7. The first melt to emerge would be largely peridotite-derived, and the temporal-compositional trend would follow a counterclockwise pattern as greater depths in the melting column are tapped. These strongly curved sequential trends do not resemble the observed quasi-linear trace element arrays of the eruption sequences, so it is unlikely that simply sequential tapping of greater depths can explain the temporal-compositional trends.

A slight variation on the sequential melt extraction scenario does, however, produce trace element trends similar to those of the eruption sequences. If incremental melts above and below the depth of the peridotite solidus are allowed to mix prior to extraction, aggregate melts from above and below this depth form end-members that could mix to form the eruption sequence arrays (Figure 7). With this scenario, the slope of the mixing line between aggregate melts produced below and above the depth of the peridotite solidus is controlled by the final depth of melting. Assuming model parameters similar to those in Figures 5 and 6, cessation of melting at depths of ~60–100 km, followed by mixing of aggregate melts from the upper and lower portions of the melt column, produces mixing trends similar to those of most of eruption sequences (except Pu‘u O‘o) in this study. For cessation of melting at 76 km, pyroxenite melts to a maximum $F$ of 29%, peridotite melts to 1.0%, and mixing between aggregate melts above and below the peridotite solidus depth produces mixing curve P-D in Figure 7. For cessation of melting at 103 km, pyroxenite melts to a maximum $F$ of 18%, peridotite melts to 0.2%, and the resulting mixing trend has a higher slope, P-B in Figure 7. Extending the depth of melting to ~15 km produces a maximum peridotite $F$ of 5% (and pyroxenite $F = 53$%), and the resulting mixing trend (P-F in Figure 7) has a much shallower slope similar to that of the Pu‘u O‘o eruption sequence.

The positions of the mixing trends, as well as the relative melt volumes above and below the depth of peridotite solidus intersection, are functions of not only the total height of the upwelling column but also the peridotite to pyroxenite proportions in the solid source and their relative melting behaviors. Increasing the proportion of pyroxenite in the solid source increases the amount of high-degree melt in the melt 1 component, decreasing its incompatible element concentrations and shortening the mixing trend to the low-incompatible element side. For the conditions shown in Figures 6 and 7 and a final depth of melting of 76 km, pyroxenite abundances of more than ~3% decrease La, Nd, Nb, and Zr concentrations in the melt 1 end-member to less than maximum concentrations in the eruption sequences. However, 3% pyroxenite in the source can be reconciled with the observed data if its melting rate is decreased. If the pyroxenite adiabatic melt productivity is only $dF/dz = 0.06$%/km, with 3% pyroxenite, the trace element results are essentially identical to the case of 0.4%/km for 0.5% pyroxenite, the case shown in Figure 7. Since the original assumption of a pyroxenite melting rate of 0.4%/km is close to the minimum (near-solidus) melting rate deduced for Pertermann and Hirschman’s (submitted manuscript, 2000) experimental composition, this suggests that solid proportions of significantly more than 1–2% of pyroxenite in the mantle source are not likely to reproduce the trace element compositions in this model. This maximum pyroxenite proportion agrees well with independent constraints (of <1–2% pyroxenite) from ocean crust thickness and mid-ocean ridge basalt rare-earth element constraints (Pertermann and Hirschman, submitted manuscript, 2000).

In cases of relatively thick mantle melting columns, caused by either (1) warm mantle intersecting peridotite and pyroxenite solidi at great depths or (2) shallow upwelling due to thin or eroded lithosphere, peridotite will melt to a larger maximum degree and melt in the upper part of the melting column will show strongly decreasing incompatible element concentrations with height (e.g., “hot” colors in Figure 6b). In this case, slightly more complicated eruption-sequence temporal-compositional trends might be expected, in which incompatible elements first increase (with melt extraction from the peridotite melting zone), then decrease, as mixing with the pyroxenite-only melt zone continues. This may be the case for the
historic Kilauea eruptions, in which the slope of the trace element trends suggests that on the whole, the degree of peridotite melting is relatively high (~2–5%, instead of 0.2–0.5% for the other eruption sequences). Interestingly, the temporal-compositional trends for both historic Kilauea eruption sequences are slightly more complex than other sequences, with some indices, such as CaO/Al₂O₃, showing an initial increase in the earliest erupted melts, followed by a steady decrease throughout the rest of the eruption (Figure 8).

6. Implications of Lithologic Heterogeneity Models

6.1. Major Elements

[46] As discussed in section 5, these eruption sequences require the end-member component in the early-erupting melt (melt 1) to have relatively low SiO₂ and Al₂O₃ and high K₂O, MgO, TiO₂, and P₂O₅. The late-erupting melt component (melt 2) possesses opposite characteristics. A model involving an origin in the metasomatized lithospheric mantle for melt 1 is broadly consistent with these major element constraints, especially if the source is carbonated [Hirose, 1997]. However, it is difficult to reconcile the low MgO and high SiO₂ of the melt 2 component with the expected characteristics of a large-degree melt of a relatively deep mantle source, unless the melt 2 end-member (but not melt 1) is somewhat differentiated by crystallization, to drive down MgO.

[47] In the context of the mixed peridotite-pyroxenite model, if peridotite was the dominant source of the melt 1 component, the low SiO₂ of this end-member again suggests that it may have been carbonated. For the other end-member, the melt 2 characteristics are easily reconciled with most of the recent experimental results for pyroxenite partial melting of Pertermann and Hirschmann [1999, submitted manuscript, 2000]. They show that through 60% melting at 3 GPa, pyroxenite produces melts with distinctly higher SiO₂ (51–57 wt %) and Al₂O₃ (14–17 wt %) and low CaO (8–10 wt %) and MgO (2–6 wt %). With the exception of TiO₂, all of the pyroxenite major element results of Pertermann and Hirschmann’s pyroxenite melting experiments are consistent with the composition of the melt 2 end-member required by these eruption sequences.

[48] It should be pointed out that both Sigmarsson et al. [1998] and Francis and Ludden [1990] also invoked a pyroxenite-derived melt as an end-member for their inferred melt mixing trends in the Lanzarote and Ft. Selkirk lavas, respectively. However, their models proposed essentially the opposite characteristics of the pyroxenitic melt as those proposed here: they favored a large pyroxenitic component in the early erupting melt batches, with low SiO₂ and Al₂O₃, high MgO and K₂O, high incompatible trace elements, and, in the case of Lanzarote, high (²³⁰Th/²³⁸U). Compositions of natural mantle pyroxenites and their component minerals vary widely [Hirschmann and Stolper, 1996; van Westrenen et al., 2001], which may mean compositional variations in natural pyroxenite melts are also large. However, several lines of evidence strongly suggest that pyroxenitic melts possess chemical characteristics that are, in general, the opposite of these, including (1) Pertermann and Hirschman’s (submitted manuscript, 2000) experimental major element results indicating relatively high SiO₂ and Al₂O₃ and low MgO and K₂O, (2) trace element modeling shown here suggesting low incompatible element concentrations of pyroxenite melts relative to peridotite melts, especially at the high melt fractions expected for pyroxenite, and (3) considerations of the relative U-Th partitioning behavior in garnets with compositions typical of pyroxenite and peridotite sources [Stracke et al., 1999; van Westrenen et al., 2001].

6.2. Radiogenic Isotopes

[49] Both of the models proposed here require very low degrees of melting (typically 0.1–0.5%; up to 5% for Pu’u O’o) for the early-erupting melt 1 end-member. Most U-series isotopic studies of primitive intraplate basalts require maximum porosities throughout much of the melting region to be 0.1–0.5% [Spiegelman and Elliot, 1993; Lundstrom et al., 1995; Sims et al., 1999; Spiegelman and Reynolds, 1999], or less (e.g., 0.001% [Turner et al., 2000]), so such small degrees of melting of at least some source component may be reasonable.
Because in general, \(^{230}\text{Th}/^{238}\text{U}\) decreases with increasing melt fraction (at least in a batch melting scenario), a progressive increase with time in a component created by larger degrees of melting would be expected to cause decreasing \(^{230}\text{Th}/^{238}\text{U}\) with time, as observed in Lanzarote 1730–1736, the only eruption sequence with available temporal-compositional U series constraints [Sigmarsson et al., 1998; Thomas et al., 1999]. The low \(^{230}\text{Th}/^{238}\text{U}\) of the melt 2 end-member could be from either large degrees of melting of peridotite or pyroxenite because available constraints on U and Th partitioning suggest that melts derived from mantle pyroxenite will have lower \(^{230}\text{Th}\) excesses than those derived from peridotite [Stracke et al., 1999; van Westrenen et al., 2001]. As discussed above, the trend of increasing \(^{226}\text{Ra}/^{230}\text{Th}\) with time in the Lanzarote eruption may reflect an amphibole signature in the shallow, melt 1 source, which may indicate a lithospheric source for this component.

[50] Isotopic compositions of Sr, Nd, and Hf are difficult to interpret in the context of either model proposed for the temporal-compositional trends. Although metasomatized mantle sources may have
elevated Rb/Sr and decreased Sm/Nd and Lu/Hf, as suggested by many previous studies, the metasomatic event may have occurred only shortly before (or may even be related to) the event that produced melting and mixing between the two end-member components. Isotopic compositions of these elements are also difficult to interpret in the context of potential pyroxenite contributions. Stracke et al. [1999] argued on the basis of combined ($^{230}$Th) excesses and inferred melt-source Sm/Nd and Lu/Hf fractionations ($\delta$$_{Sm/Nd}$ and $\delta$$_{Lu/Hf}$) that Hawaiian lavas show no evidence for a contribution from a pyroxenitic source. However, because of the highly incompatible nature of U and Th, if a significant contribution of aggregate melt is derived from very small degree melts of peridotite, the ($^{230}$Th/$^{238}$U) signature is likely to be dominated by the peridotite contribution. The same holds true, albeit to a lesser degree, for Nd and Hf isotopic compositions. Sm/Nd and Lu/Hf of mantle pyroxenites are also not systematically different from those of peridotite (or depleted mantle) [Hirschmann and Stolper, 1996] and not unless ancient recycled oceanic crust, specifically, is assumed to be the dominant type of pyroxenite component will it show a systematic difference in $^{143}$Nd/$^{144}$Nd and $^{176}$Hf/$^{177}$Hf. Finally, relationships between degree of melting and melt mixing proportions are critical to evaluating pyroxenite contributions from both isotopic and trace element perspectives. Stracke et al. outlined several possible types of mixing trends for peridotite and pyroxenite melts. However, based on the experimental results of Pertermann and Hirschmann [1999, submitted manuscript, 2000] and model parameters used here, melt-mixing trends for a mixed peridotite-pyroxenite source should be manifest as mixing between large degree pyroxenite melts ($\sim$20–60%) and small degree peridotite melts (0.1–5%).

[51] Radiogenic Os isotope ratios in late-erupting melts ($^{187}$Os/$^{188}$Os = 0.14–0.16) with relatively low incompatible elements provide potentially important constraints on both of the models proposed here. Os isotopic compositions of most basaltic magmas, including mid-ocean ridge basalts, are generally more radiogenic than those of potential peridotite sources [Roy-Barman and Allègre, 1994; Shirey and Walker, 1998; Lassiter et al., 2000; Brandon et al., 2000; Meisel et al., 2001]. This has led many studies to suggest that Os isotopic signatures in many types of mafic magmas have are strongly affected by, or even dominated by, shallow-level contamination (e.g., basalt, Mn crust, seawater) [e.g., Shirey and Walker, 1998; Lassiter and Luhr, 2001]. The highly compatible behavior of Os means even moderate amounts of fractional crystallization leaves residual melts susceptible to such contamination. Contamination associated with Os-removal by fractionation crystallization should lead to good correlations between 1/[Os] and $^{187}$Os/$^{188}$Os [Lassiter and Luhr, 2001], as is observed in many mafic magmas, including the Lanzarote eruption sequence melts. This is unlikely to explain the radiogenic Os in the Lanzarote melts, however, because in this case it is the early-erupting melts, with unradiogenic Os compositions (not the late-erupting, radiogenic ones) that are suspected to be contaminated by lithosphere [Widom et al., 1999; J. C. Lassiter, unpublished data, 2001]. In addition, the low Os (and MgO and Ni) melts have the lowest incompatible element concentrations, which would be expected to increase with fractionation. It is possible that these correlations (along with those of most other compositional indices in these eruption sequences) instead represent mixing between melts with contrasting Os concentrations and isotope ratios. If so, pyroxenite-derived melts represent a potential candidate the low-Os, high $^{187}$Os/$^{188}$Os end-member [e.g., Schiano et al., 1997].

6.3. Melt Transport Rates

[52] Both models discussed here imply that melt is extracted from deep melting regions (of the order of 100 km) within the timescale of single eruptions. These eruption sequences suggest that an appropriate timescale for observing temporal-compositional trends in primitive basalts is $\sim$6–20 years. If these eruptions tap melt from depths of $\sim$100 km ($\sim$3 GPa), this requires source-to-surface melt velocities of $\sim$5–17 km/yr or $\sim$0.05–0.2 cm/s. These rates are generally consistent with those derived by U/Th series disequilibria and xenolith size analyses [Sigmarsson et al., 1998; Turner et al., 2001; McKenzie,
and require fast extraction via channelization. Even if the explanations for the origin of the temporal-compositional trends proposed here are incorrect, changing composition of erupted melt with relatively fast migration rates raise the question of what spatial pattern of compositional heterogeneity is being reflected by temporal-compositional trends. While there is some independent evidence that progressively greater mantle depths are tapped with time in at least some basaltic eruptions [Putirka, 1997], it is possible that instead of progressively greater mantle depths or sequential melt batches through lithospheric melt channels, later-erupted melt batches may be derived from progressively greater lateral distances, or less permeable mantle domains, with time. However, if the perhaps simplest scenario of progressively greater mantle depths is assumed, then many primitive basaltic eruptions (excluding those that pool in magma chambers and aggregate melts from different depths) may provide a relatively simple and potentially powerful record of compositional variation in mantle melting columns, mapped into a time-series.

7. Binary Melt Mixing in Basalt Petrogenesis

Several recent studies, especially those focusing on compositional variations on relatively short timescales and length scales, have emphasized evidence for binary mixing as a dominant source of chemical variation in primary basalts. Such evidence has come from Pb isotope arrays [Saal et al., 1998; Abouchami et al., 2000], Zr-Nb systematics [Kamber and Collerson, 2000], multi-isotope topology [Phipps Morgan, 1999], U series disequilibria [Lundstrom et al., 2000; Condomines and Sigmarsson, 2000], and other isotopic and trace element systematics [Class and Goldstein, 1997]. As discussed in section 4.4 and in the appendix, some of the eruption sequences that are the focus of this study have also been interpreted in the context of binary mixing [Francis and Ludden, 1990; Sigmarsson et al., 1998; Thomas et al., 1999], although with considerably different scenarios than proposed here. In most of these cases the data can be interpreted in terms of melt-melt, but not melt-solid or solid-solid, mixing. Additional evidence for binary melt mixing comes from Os isotopes. Correlations between isotopic compositions of Os and other elements are usually sublinear in OIBs [Hauri and Hart, 1993; Hauri and Kurz, 1997]. Because of strongly contrasting Os concentrations in distinct mantle lithologies, mixing trends between isotopes of Os and other elements would generally be strongly curved if mixing occurred prior to melting [Class and Goldstein, 1997; Becker, 2000; Kogiso et al., 2000]. One possible inference from this is that binary melt mixing trends manifest on relatively short timescales and length scales in basaltic petrogenesis reflect the presence of two common, but physically and chemically distinct sources in the mantle, such as mafic lithologies embedded in peridotite matrix. One potential problem with this hypothesis, however, is the mechanism of extracting pyroxenite melts from the deeper portions of the melting column surrounded by largely unmelted peridotite. In these models, pyroxenite melts of about F = 20% must be able to escape surrounding peridotite and mix with overlying mixed peridotite-pyroxenite melts in order to generate temporal-compositional trends. Experimental results of Yaxley and Green [1998] demonstrated that pyroxenite melt reacts with solid peridotite to form pyroxene-rich peridotite, potentially limiting the extractability of pyroxenite melt from solid peridotite matrix. As suggested by Kogiso et al. [2000], however, a thin pyroxene-rich reaction zone may form a protective channel allowing further pyroxenite-derived melts to escape.

8. Conclusions

Systematic decreases in incompatible elements and changes in major elements and isotopic compositions during eruption sequences of primitive alkalic and alkalic-to-tholeiitic magmas from a wide variety of intraplate volcanoes indicate that systematically changing proportions of melts from two distinct mantle sources, with time, are a characteristic feature of this type of eruption. Trace element variations and other lines of evidence indicate that melts from each source are distinguished by a large difference in degree of melting, such that the proportion of the small-degree melt decreases with time during each eruption. An origin
involving migration of a large-degree sublithospheric melt mixing with small-degree lithospheric melt during migration and ascent cannot be excluded. Such a model would explain residual amphibole signatures and other trace element features of the small-degree melt component in some sequences and would imply that this component is exhausted in sequentially erupted melt batches by passage of progressively larger volumes of large-degree melt through the lithospheric channels. However, both major element and Os isotope characteristics of the late-erupting, large-degree melt component are difficult to reconcile with this model. Instead, they are consistent with a model in which the high-degree melt is derived from pyroxenite, and the low-degree melt is derived from peridotite, and melts are extracted from progressively greater depths in a compositionally zoned mantle melting column. Because pyroxenite begins melting at greater depths, and melts to a larger degree, than peridotite, early erupted, shallowly derived melts contain a large proportion of relatively low-degree peridotite melt, and later erupted melts derived from greater depths contain a larger proportion of high-degree pyroxenite melt.

If either of these models are generally applicable to alkalic and alkalic-to-tholeiitic magmas then a number of conceptual features of such magmatism need to be reconsidered. The common observation of quasi-linear correlations between incompatible elements of different compatibility is due to binary melt mixing, not melting of a highly enriched mantle source. This solves the long-standing problem of requiring highly incompatible element enriched mantle sources with strongly depleted Sr and Nd isotopic compositions. Also, most of the trace element variation in primitive basalts is produced by mantle source heterogeneity (and melt mixing), not a variation in the degree of melting. Even if the specific hypotheses proposed here for the origins of temporal-compositional trends in individual eruptions of primitive intraplate basalts are incorrect, the trends themselves indicate a systematic organization to distinct melt sources and mixing mechanisms in the mantle, and highlight the potential for new insights on basalt petrogenesis held by examining geochemical variations on smaller time- and length-scales (e.g., single eruptions) than typically studied.

Appendix A. Additional Information and Previous Work on Eruption Sequences

A.1. Koloa Volcanics, Kaua‘i, Hawai‘i

Rejuvenated, or posterosional, stage volcanics such as the Koloa volcanics are characteristic of the waning stages of magmatic activity on many Hawai‘ian islands [Clague, 1987]. Compositionally similar magmas are also erupted offshore of the islands, far from the inferred location of the center of the Hawai‘ian plume [Clague et al., 1990; Dixon et al., 1997; Frey et al., 2000]. The eruption sequences that are discussed here were sampled by subsurface drilling in the Lihue basin in eastern Kaua‘i [Reiners and Nelson, 1998; Reiners et al., 1999] and include a nephelinite flow sequence ~50-m thick and an alkalic basalt sequence ~40-m thick. Petrographic and chemical features within sequences and the absence of intercalated sediments between flows suggest that each sequence represents cogenetic magmas erupted from an individual vent, of the order of years. These eruption sequences are part of a longer, 300-m sequence of lava flows and other lithologies that document an overall, long-term temporal-compositional trend that is opposite to that observed within each eruption sequence. Through the entire stratigraphic pile, incompatible elements increase and rock types evolve from alkalic basalt to basanite to nephelinite with time [Reiners et al., 1999]. Previous interpretations for compositional variation in the Koloa eruption sequences offered by Reiners and Nelson [1998] included mixing of melts produced by very different degrees of melting (e.g., 0.1% and 2–15%), or variable degrees of melting of metasomatized sources, with a correlation between degrees of melting and metasomatism. The temporal-compositional trends were suggested to be the result of either (1) tapping of progressively greater depths of a melting region with a vertical zonation in degree of melting or (2) extraction of low-degree melts from surrounding mantle by early-ascending magma batches. Later work by Lassiter et al.
[2000], focusing on radiogenic Os isotope compositions and their correlations with other compositional features, suggested an origin for Hawai’ian rejuvenated-stage magmas involving melting of mafic mantle pyroxenite, although no explanation was offered for the temporal-compositional trends.

A.2. Lanzarote 1730–1736, Canary Islands

[57] The volcanologic and geochemical evolution of the 1730–1736 eruption on the island of Lanzarote in the Canary Islands has been studied by Carracedo and Badiola [1991], Carracedo et al. [1992], Sigmarsson et al. [1998], Thomas et al. [1999], and Widom et al., 1999, among others. While Sigmarsson et al. [1998] documented a strong correlation between $^{87}$Sr/$^{86}$Sr and other chemical indices and eruption sequence, Sr and Nd isotopic analyses of Thomas et al. [1999] do not show such correlations. While two data sets suggest that $^{187}$Os/$^{188}$Os generally increased significantly during the eruption, the unradiogenic Os isotope signatures in the early-erupting lavas may be at least partly a result of contamination by disaggregated lithospheric mantle xenoliths [Widom et al., 1999]. Previous interpretations for the origin of the extreme compositional variation in the Lanzarote 1730–1736 eruption have focused on chemical and isotopic trends that are inconsistent with a dominant role for variation in degree of partial melting, and instead support an origin by melt-mixing. Thomas et al. [1999] suggested mixing between melts formed by two different degrees of melting, within the same dynamic melting column. Sigmarsson et al. [1998] interpreted the compositional range as reflecting incomplete mixing of peridotite- and pyroxenite-derived melts, with a larger proportion of peridotite-derived melt in the latter stages of eruption. This latter interpretation requires pyroxenite-derived melts to be relatively low-SiO$_2$, high MgO, and alkali-rich, and to have relatively high $(^{230}$Th/$^{238}$U).

A.3. Pisgah and Amboy, Southern California

[58] Pisgah and Amboy craters are Quaternary expressions of the Mojave neovolcanic province of southern California. Stratigraphic relations between erupted units at each volcano were refined by Glazner et al. [1991], based on earlier work [Parker, 1959, 1963; Wise, 1969]. Temporal control is poor for most of the Amboy crater lavas. Nevertheless, robust observations reveal that each volcano erupted alkalic basalts with systematically changing major and trace element and isotopic compositions. Glazner et al. [1991] favored assimilation of partial melt of gabbroic crust into a primitive mantle-derived magma to explain the temporal-compositional trends at Pisgah and Amboy. However, to generate the observed compositional trends, the assimilated melt would have to have very low concentrations of incompatible elements (e.g., La < 15 ppm, Ba < 225 ppm, Zr < 150 ppm, Th < 2 ppm), which would require either a mafic crust much more depleted in incompatible elements than typical gabbro or high degrees of melting. An alternative scenario that can explain the low incompatible elements is that the mafic source was pyroxenitic rather than gabbroic and was located in the mantle rather than crust.

A.4. Fort Selkirk, Yukon

[59] The Fort Selkirk volcanic complex comprises lavas erupted from five different vents and eruption sequences. Francis and Ludden [1990] describe systematic upsection decreases in whole-rock alkalinity in the four longest sequences and no change in the shortest one. Two eruption sequences produced nephelinite followed by basanite, one produced basanite followed by alkali olivine basalt (AOB) followed by hypersthene-normative AOB, and one produced AOB followed by hypersthene-normative AOB followed by low-Mg AOB. Although detailed stratigraphic information is not provided by Francis and Ludden [1990], temporal trends within most of these sequences are apparent in figures of the paper [e.g., Francis and Ludden, 1990, Figure 3, pp. 375–376]. Francis and Ludden [1990] demonstrated that nesication–ither crystallization nor variable degrees of melting of a single mantle source could explain the compositional spectrum in the Ft. Selkirk lavas. Instead, they favored an origin by incomplete melt mixing, whereby the relatively SiO$_2$-rich end-member was derived from peridotite, and the alkalic end-member was
derived from amphibole-garnet-clinopyroxene mantle veins. Similar to the interpretation of Sigmarsson et al. [1998] for Lanzarote, this requires that melts of the mafic veins are relatively low-SiO$_2$, high-MgO, and alkali-rich.

A.5. Pu’u O’o and Mauna Ulu, Kilauea, Hawai’i

[60] Careful monitoring and analysis of the historic eruptions of Kilauea’s east rift zone, especially the Pu’u O’o/Kupaianaha eruption [Garcia et al., 1992, 1996, 2000], have provided unprecedentedly detailed and valuable constraints on temporal-compositional trends during individual basaltic eruptions. The Mauna Ulu eruption of 1969–1974 produced ~0.16 km$^3$ of basalt almost entirely from a single vent (Mauna Ulu-Alae area). The eruption is commonly broken into two phases separated by a 3.5-month gap in eruptive activity in 1971; only the chemically well-characterized 1969–1971 phase of the eruption is considered here [Wright et al., 1975; Hofmann et al., 1984]. The Pu’u O’o eruption began in 1983 and continues to the present, having erupted more than ~2 km$^3$ of basalt. Early-erupted Pu’u O’o basalts have fractionated beyond olivine control and are thought to have mixed with fractionated magma residing in the rift zone prior to eruption [e.g., Garcia et al., 1992]. The compositional effects of shallow-level mixing on erupted Pu’u O’o lavas probably extended through episodes 10 or 30 (dashed lines in Figure 1). Continued temporal-compositional trends in major and trace elements in subsequently erupted melts, however, cannot be attributed to this mixing process and must reflect changing mantle melting or source controls [Garcia et al., 1992, 1996, 2000]. Hofmann et al. [1984] interpreted systematically decreasing incompatible trace element concentrations with time in the 1969–1971 portion of the Mauna Ulu eruption to reflect an ~20% increase in the degree of melting of a mantle source. They reasoned that this may not necessarily represent a progressive increase in extent of melting of exactly the same mantle source, but instead might reflect a shift in the volume tapped by the eruption in a zoned source region, from regions with low to high melt fractions with time. Garcia et al. [1996] also originally proposed a possible origin for the Pu’u O’o compositional trends by progressive partial melting of a homogeneous mantle source. However, this was supplanted by later work [Garcia et al., 1996], which determined that the gradual shift in erupted melt compositions required the progressive influence (increasing by ~25%) of a second, more depleted, mantle source, to the melt supply region of Pu’u O’o over time.

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