

Nd and Sr isotopes in ultrapotassic volcanic rocks from the Leucite Hills, Wyoming

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Abstract. Nd and Sr isotopic compositions and Rb, Sr, Sm and Nd concentrations are reported for madupites, wyomingites and orendites from the Pleistocene volcanic field of the Leucite Hills, Wyoming. All Leucite Hills rocks have negative ϵNd signatures, indicating derivation or contribution from an old light rare earth element (LREE) enriched source. In this respect they are similar to all occurrences of high potassium magmas so far investigated. But Sr isotopic variations are comparatively small and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are unusually low for high-K magmas (0.7053–0.7061, one sample excluded). These values suggest that the light REE enrichment of the source was not accompanied by a strong increase in Rb/Sr.

Wyomingites and orendites are isotopically indistinguishable which is consistent with chemical and petrographic evidence for their derivation from a common magma series depending on emplacement conditions. Basic to ultrabasic madupites and more silicic wyomingites/orendites are distinct in their Nd isotopic variations (madupites: $\epsilon\text{Nd} = -10.5$ to -12.3 ; wyomingites/orendites: $\epsilon\text{Nd} = -13.7$ to -17.0) despite similar Sm/Nd ratios and complete overlap in $^{87}\text{Sr}/^{86}\text{Sr}$.

Selective or bulk assimilation of crustal material is unlikely to have significantly affected the Nd and Sr isotopic compositions of the magmas. The measured isotopic ratios are considered to reflect source values. The distinct isotopic characteristics of madupite and wyomingite/orendite magmas preclude their derivation by fractional crystallization, from a common primary magma, by liquid immiscibility or by partial melting of a homogeneous source. Two isotopically distinct, LREE enriched and slightly heterogeneous sources are required. Heterogeneities were most pronounced between magma sources from each volcanic centre (butte or mesa). The relationship between the madupite and wyomingite/orendite sources and their evolution is discussed on the basis of two simple alternative sets of models: (1) a two-stage evolution model with an old enrichment event (a metasomatic event?) perhaps taking place during the stabilization of the Wyoming Craton 3.2 to 2.5 Gyr ago but not later than 1.2 Gyr ago or (2) a mixing model involving mixing between one end-member with ϵNd near zero and another end-member with a strong negative ϵNd signature.

Introduction

Interest in ultra-potassic volcanic rocks has been stimulated by the recognition that some members of this very heterogeneous group are diamond host rocks. Although diamondiferous lamproites from Prairie Creek, Arkansas and West Kimberley, Australia have been confused with kimberlites, they are, according to Scott-Smith and Skinner (1984) and Jaques et al. (1984), an extension of the ultra-potassic volcanic rock suite and are distinct and separate from kimberlites. Low $^{143}\text{Nd}/^{144}\text{Nd}$ and high $^{87}\text{Sr}/^{86}\text{Sr}$ of these magmas indicate that they are derived from, or at least contain a contribution from, an old source region enriched in Rb/Sr and the light rare earth elements (LREE) (Nixon et al. 1982; McCulloch et al. 1983). These isotopic features are unlikely to be a result of contamination of the magmas but are considered to reflect source characteristics (McCulloch et al. 1983). The presence of diamonds and mantle xenoliths seems to be compelling evidence for the derivation of these magmas from the upper mantle, hence the important implications of these findings for the use of radiogenic isotopes as petrogenetic tracers: low $^{143}\text{Nd}/^{144}\text{Nd}$ and high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios may no longer be regarded as reliable isotopic signatures of continental crustal material.

By comparing the Sr, Nd, Pb and oxygen isotope interrelations for the Italian and Virunga potassic volcanic rocks Vollmer (1983 and in preparation) has suggested that the old LREE enriched source component observed for these two occurrences (Hawkesworth and Vollmer 1979; Vollmer and Norry 1983a) is likely to be continental crustal material, possibly previously metasomatised. Characteristically, mantle nodules have not been found in these volcanics. These apparently conflicting inferences may be reconciled if this very heterogeneous group of rocks has diverse origins, or by the generation of anomalous mantle sources by recycling of continental crustal material into the mantle.

The importance of the Leucite Hills ultrapotassic rocks to this problem lies in their similar major element chemistry to the Australian lamproites (Sahama 1974). However, except for one harzburgite nodule (Barton and van Bergen 1981), no clearly mantle-derived nodules have been found in the Leucite Hills. We undertook an Nd and Sr isotope and trace element study of the Leucite Hills to further characterize the different magma types and their source regions and to evaluate petrogenetic models. The Nd and Sr isotopic compositions reported here point again to an old LREE enriched source or source component which is thus a common feature of all basic potassic magmas investigated

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so far. The origin of this enriched source however, still remains uncertain and the existence of ancient large-scale LREE enriched regions in the mantle which are not directly or indirectly due to involvement of continental crustal material remains a distinct possibility (see also Smith 1983).

Regional geology

The Leucite Hills comprise some 22 exposures of volcanic and subvolcanic rocks over an area of 2,500 km² in southwestern Wyoming (Fig. 1). An age of 1 Myr is suggested for the volcanism from a mica K–Ar age determination (McDowell 1966). The volcanic rocks overlay, and are erupted through, Paleozoic to Eocene sediments (Malahoff and Moberly 1968). The basement is generally believed to be part of the Archean Wyoming craton which seems to have formed and stabilized in the period 3,200–2,500 Myr B.P. (Peterman 1979). Some 200 km south of the Leucite Hills, there is a major transcontinental suture between the Archean Craton and Proterozoic basement (~1,800 Myr). The suture may have been the locus of both post Archean continental rifting and subsequent continent-continent collision and accretion in the 1.7 to 1.8 Gyr period (Warner 1978a, b; Condie 1982).

The volcanic field lies on the NE flank of the Rock Springs uplift, one of the Late Cretaceous to Paleocene Laramide uplifts. The Wind River uplift to the north of the Leucite Hills has been investigated in detail by Smithson et al. (1979) from deep reflection seismic and gravity data. This uplift, as are most others, is an asymmetrical anticline whose steep limb is cut by a reverse fault. There was at least 26 km of horizontal and 14 km of vertical movement along the thrust. A positive Bouguer gravity anomaly above the uplift might be caused by uplift of dense lower crustal material or the mantle. The Mohorovicic discontinuity does not appear as a strong or continuous event in the reflection data and does not seem to be displaced by faulting. The crust in the Wind River uplift began to deform by large-scale folding and then broke and faulted as a rigid slab.

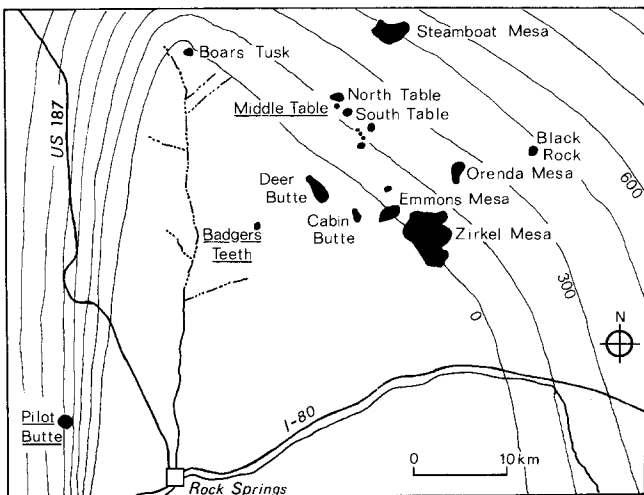


Fig. 1. Map of the Leucite Hills, Wyoming. Potassic alkaline volcanic rocks in black; the three important madupite exposures are underlined. Contours of thickness (in meters) of Late Cretaceous and Early Paleocene sediments deposited above the Late Cretaceous Mesaverde Formation outline the Laramide Rock Springs Uplift (after Curry 1973)

The Rock Springs uplift is upwarped but does not seem to be uplifted to the extent that faulting occurred.

Petrology and geochemistry

Cross (1897) first described the three major types of lava from the Leucite Hills: *Madupite* is composed of microphe-nocrysts of diopside enclosed by poikilitic phlogopite phenocrysts and set in a matrix of leucite, diopside, apatite and glass. *Wyomingite* has phlogopite phenocrysts in a groundmass of leucite, diopside, apatite and glass. *Orendite* again has phlogopite phenocrysts, but a groundmass of san-idine, leucite, diopside and apatite. Often wyomingite forms the chilled margins of orenditic lavas. In terms of bulk chemistry wyomingite and orendite are indistinguishable (Table 1). Differences in their mineralogy are probably related to depth of crystallization (Kuehner et al. 1981), cooling rates or P_{H₂O} (Ogden 1979).

Chemically, madupite and orendite/wyomingite are ultra-potassic basic lavas. Madupite is lower in SiO₂, Al₂O₃

Table 1. Average composition and standard deviation of madupite, wyomingite and orendite. Number of samples in parenthesis. Major elements from Ogden (1979). Madupite trace element data are for Badger Teeth and Pilot Butte only; Middle Table has intermediate values between these and wy-or

| Oxide | Orendite (28) | Wyomingite (23) | Madupite (12) |
|--------------------------------|------------------|--------------------|--|
| SiO ₂ | 53.0 ± 1.7 | 52.4 ± 2.8 | 45.5 ± 1.9 |
| TiO ₂ | 2.6 ± 0.2 | 2.5 ± 0.1 | 2.4 ± 0.1 |
| Al ₂ O ₃ | 9.8 ± 0.7 | 9.7 ± 0.9 | 8.4 ± 0.7 |
| Fe ₂ O ₃ | 4.5 ± 0.4 | 4.7 ± 0.6 | 6.3 ± 0.4 |
| MnO | 0.060 ± 0.009 | 0.064 ± 0.022 | 0.110 ± 0.015 |
| MgO | 7.3 ± 1.7 | 7.2 ± 1.5 | 9.7 ± 0.8 |
| CaO | 4.0 ± 0.8 | 4.6 ± 2.0 | 10.1 ± 1.6 |
| Na ₂ O | 1.2 ± 0.2 | 1.5 ± 1.1 | 0.9 ± 0.1 |
| K ₂ O | 10.9 ± 1.0 | 10.2 ± 2.4 | 6.9 ± 1.3 |
| P ₂ O ₅ | 1.8 ± 0.6 | 1.7 ± 0.5 | 2.4 ± 0.6 |
| 100 Mg | 76.3 | 75.2 | 75.3 |
| Mg+Fe | | | |
| Sc | 11.9 ± 1.7 (29) | | 19.9 ± 0.8 (6) |
| V | 86 ± 11 (28) | | 110 (Badger Teeth) 50 (Pilot Butte) |
| Cr | 350 ± 90 (29) | | 511 ± 48 (6) |
| Co | 20.4 ± 3.6 (29) | | 28 ± 2 (6) |
| Ni | 256 ± 56 (32) | | 192 ± 17 (5) |
| Rb | 279 ± 40 (36) | | 210 ± 23 (9) |
| Sr | 2500 ± 430 (36) | | 5100 ± 903 (9) |
| Y ^a | 16.5 | | 19 |
| Zr ^a | 1270 | | 1210 |
| Nb ^a | 50 | | 116 |
| Cs | 2.05 ± 0.28 (19) | | 2.96 ± 0.43 (6) |
| Ba | 5730 ± 1300 (36) | | 7350 ± 2400 (9) |
| La | 157 ± 41 (29) | | 402 ± 24 (6) |
| Ce | 354 ± 84 (29) | | 850 ± 78 (6) |
| Nd | 131 ± 29 (29) | | 292 ± 33 (6) |
| Sm | 17.4 ± 3.4 (29) | | 39 ± 2 (6) |
| Eu | 4.2 ± 0.7 (29) | | 8.0 ± 1.1 (6) |
| Tb | 1.8 ± 0.5 (29) | | 3.5 ± 0.8 (6) |
| Yb | 1.3 ± 0.2 (29) | | 1.6 ± 0.2 (6) |
| Pb ^a | 27 | | 57 |

^a From Kuehner et al. (1981). All other trace element data from Table 3 and unpublished data (Isotope laboratory, Graduate School of Oceanography, URI). Ni: analyst G. Pajari. Oxydes in wt %, trace elements in ppm

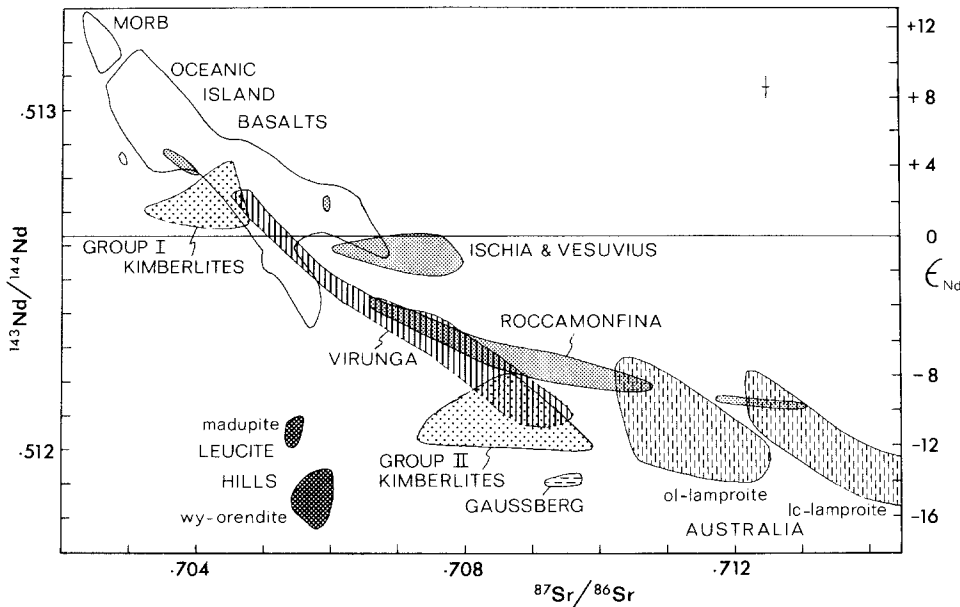


Fig. 2. Nd and Sr isotopic compositions for Leucite Hills lamproites compared with potassic rocks from Italy (Hawkesworth and Vollmer 1979), Virunga (Vollmer and Norry 1983a, b), West Kimberley, Australia (McCulloch et al. 1983) and Gaussberg (Collerson and McCulloch 1983). Also shown are fields for southern African Cretaceous kimberlites (Smith 1983) and oceanic basalts (see White and Hofmann (1982) for references). Madupite and wyomingite/orendite form two distinct groups distinguished by ϵ_{Nd} . The cross indicates typical analytical uncertainty

and K_2O and richer in MgO, CaO and Fe relative to wyomingite/orendite (Table 1). In common with other ultra-potassic lavas, madupite and wyomingite/orendite are strongly enriched in the light REE and other large ionic lithophile (LIL) elements as well as fairly enriched in Ni, Cr and Sc (Table 1 and Fig. 3).

The rocks from Middle Table are difficult to classify. Greenish layers of 'madupite' alternate here on a cm scale with brownish layers of 'wyomingite' (Ogden 1979). However, from a strict interpretation of the mineralogical definition of rock types both varieties are madupites, although the madupite texture in the 'wyomingite' layer is very difficult to discern and is seen only on a few of the phlogopites and there only in very thin rims. There is no chemical difference between the 'madupite' and 'wyomingite' layers (Tables 2 and 3) and we will group both mineralogical varieties together with the madupites from Pilot Butte and Badger Teeth. There are however distinct chemical differences to these latter madupites: the Middle Table madupites may be regarded as transitional between madupite and orendite end-member compositions.¹

Additional information on the mineralogy and petrology can be found in Carmichael (1967), Ogden (1979), Barton (1979), Kuehner et al. (1981) and Barton and van Bergen (1981). Melting experiments on some of these lavas at pressures up to 30 kbars under dry or H_2O -saturated conditions are reported by Barton and Hamilton (1978, 1979). Basic xenoliths have been described by Barton and van Bergen (1981); except for one phlogopite-harzburgite nodule, ultramafic xenoliths seem to be absent in Leucite Hills magmas. Rounded sediment inclusions are often frequent and range from 0.1 to 10 cm in diameter.

Analytical procedures

On the basis of detailed field work and petrologic investigation by Ogden (1979) representative samples from all major exposures

¹ Kuehner et al. (1981) refer to the madupites from Badger Teeth (=Twin Rock) as being transitional. These rocks are distinct in their trace element signature to madupites from Pilot Butte but may not be regarded as being transitional between madupite and wyomingite/orendite

were taken. The rocks are fresh; vesicular carbonates and zeolites are observed only in madupites from Badger Teeth and are probably due to subsolidus alteration.

The frequent sediment xenoliths were avoided with care during crushing. Millimeter sized chips were handpicked and only those showing no sign of discolouration due to minute inclusions were selected. The chips were ultrasonically cleaned in distilled water before grinding in an agate mortar. This procedure was checked using a sample with abundant small inclusions (L79-P1). A second fraction (L79-P1C) was prepared from this sample by repeating the handpicking, discarding half of L79-P1. Both fractions were found to be within the analytical uncertainty for all trace elements and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, and we are confident that the final samples generally represent the lava compositions. However, some modification of the magma chemistry by complete resorption of xenoliths is difficult to exclude and we will discuss the problem of contamination in some detail later.

The isotope ratios of Sr were obtained from 100 mg samples spiked with ^{84}Sr and those of Nd on unspiked samples. Sr and Nd were isolated using essentially the same method described by Hart and Brooks (1974). Nd was separated from other rare earth elements on a second column (HDEP coated on teflon powder) using a reverse phase chromatographic technique (Winchester 1963; Schilling 1966; Richard et al. 1976). The isobaric contribution of ^{144}Sm to ^{144}Nd is usually less than 10 ppm. Total procedural blanks were about 0.3 ng Sr and 0.2 ng Nd and are negligible for these rocks. Isotopic analyses were performed on the URI 30.5 cm, 60° VG MM30 mass-spectrometer. Sr was loaded with H_3PO_4 on a single Ta-filament. Nd samples were loaded as chloride on a triple filament assembly using Ta side filaments and a Re centre filament. Nd was measured as metal. The $^{143}\text{Nd}/^{144}\text{Nd}$ ratio is normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Prior to November 1980 measurements of standards in our laboratory were the following (σ : standard deviation; n: number of repeated analyses): SRM987: $^{87}\text{Sr}/^{86}\text{Sr} = 0.710281 \pm 41$ (2σ , $n = 26$), BCR-1: $^{143}\text{Nd}/^{144}\text{Nd} = 0.512701 \pm 26$ (2σ , $n = 4$), Johnson Mathey (obtained from Lamont): $^{143}\text{Nd}/^{144}\text{Nd} = 0.511183 \pm 30$ (2σ , $n = 7$). Measurements made after November 1980, after changing the $10^{11} \Omega$ resistor (and sequence of mass scanning for Nd) were: SRM987: $^{87}\text{Sr}/^{86}\text{Sr} = 0.710247 \pm 24$ (2σ , $n = 5$), E&A standard: $^{87}\text{Sr}/^{86}\text{Sr} = 0.708022 \pm 23$ (2σ , $n = 7$), BCR-1: $^{143}\text{Nd}/^{144}\text{Nd} = 0.512569 \pm 24$ (2σ , $n = 5$), Johnson Mathey: $^{143}\text{Nd}/^{144}\text{Nd} = 0.511069 \pm 22$ (2σ , $n = 7$). Shift in machine bias is within the errors quoted identical for both Nd standards.

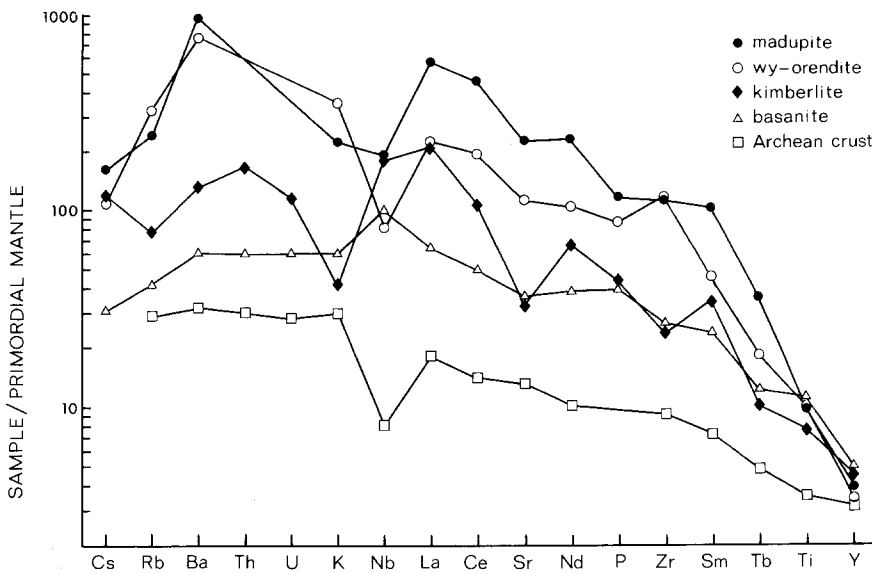


Fig. 3. *hyg* element abundance patterns normalized to estimated primordial mantle abundances (Wood et al. 1979) for average madupite, wyomingite/orendite, kimberlite (Wedepohl and Muramatsu 1979) and a basanite (sample 2679: Frey et al. 1978). Note the extreme enrichment of *hyg* elements in Leucite Hills magmas even when compared with kimberlites, the close similarity between madupite and orendite and the disproportionally low abundance of Nb in Leucite Hills magmas relative to other *hyg* elements. Average Archean crustal abundances are shown for comparison (Taylor and McLennan 1981)

Table 2. Nd isotope data for Leucite Hills ultra-potassic rocks

| Locality | Sample | Na ^a | Sm ^a | Sm/Nd | ¹⁴³ Nd/ ¹⁴⁴ Nd ^b | εNd ^d |
|----------------------------|-------------------|-----------------|-----------------|-------|---|------------------|
| | | (ppm) | | | | |
| <i>Madupite</i> | | | | | | |
| Badger Teeth | L79-BT3 | 271 | 37.9 | 0.140 | 0.51201 ± 3 ^e | -12.3 |
| | L79-BT4 | 295 | 39.1 | 0.133 | 0.51202 ± 2 | -12.1 |
| Middle Table | L79-MT2#2W | | 25.1 | | 0.51210 ± 2 | -10.5 |
| | L79-MT2#2M | 221 | 22.9 | 0.104 | 0.51210 ± 3 | -10.5 |
| | L79-MT3 | 192 | 24.9 | 0.130 | 0.51205 ± 3 | -11.5 |
| Pilot Butte | L79-P1C | 266 | 38.9 | 0.146 | 0.51205 ± 3 | -11.5 |
| | L79-P4 | | | | 0.51208 ± 2 | -10.9 |
| <i>Wyomingite-orendite</i> | | | | | | |
| Boars Tusk | L79-B1 | 117 | 15.3 | 0.131 | 0.51194 ± 3 | -13.7 |
| | L79-B2C | | | | 0.51194 ± 3 | -13.7 |
| | LH-1 ^c | 131 | 16.2 | 0.124 | 0.51192 ± 5 | -14.0 |
| Deer Butte | L79-D3#2 | 199 | 24.2 | 0.122 | 0.51184 ± 2 | -15.6 |
| Orenda Mesa | L79-S3 | | | | 0.51185 ± 2 | -15.4 |
| South Table | L79-ST2 | 124 | 15.0 | 0.121 | 0.51177 ± 2 | -17.0 |
| | L79-ST4 | | | | 0.51181 ± 2 | -16.2 |
| Steamboat Mesa | L79-SM3 | 97 | 15.0 | 0.155 | 0.51184 ± 2 | -15.6 |
| | L79-SM4 | 98 | 14.7 | 0.150 | 0.51185 ± 2 | -15.4 |
| Zirkel Mesa | L79-Z4 | 153 | 19.0 | 0.124 | 0.51178 ± 2 | -16.8 |
| | L79-Z6 | | | | 0.51185 ± 3 | -15.4 |
| | | | | | 0.51183 ± 2 | -15.8 |

^a Neutron activation analyses

^b Normalized to ¹⁴⁶Nd/¹⁴⁴Nd = 0.7219 and relative to BCR-1: 0.51265

^c Sm, Nd conc. from Kay and Gast (1973), Nd isot. comp. from De Paolo and Wasserburg (1977)

^d εNd = $\left[\frac{^{143}\text{Nd}/^{144}\text{Nd}_{\text{sample}}}{^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}}} - 1 \right] \cdot 10^4$ where ¹⁴³Nd/¹⁴⁴Nd_{CHUR} = 0.51264

^e Errors reported are on the fifth decimal and represent 2σ_m

Results

The Nd and Sr isotope data and parent-daughter element abundances are listed in Tables 2 and 3. With one exception, a ⁸⁷Sr/⁸⁶Sr ratio of 0.70779 ± 5 for sample L79-B1 from Boars Tusk, the range in ⁸⁷Sr/⁸⁶Sr is remarkably small

for potassic rocks (wyomingites and orendites: 0.7053–0.7061; madupites: 0.7054–0.7057). Sr isotope data for the Leucite Hills reported by Powell and Bell (1970) are similar but have a slightly larger range which is probably due to the comparatively large analytical uncertainty in this early study.

Table 3. Sr isotope data for Leucite Hills ultra-potassic rocks

| Locality | Sample | Rb ^a | Sr ^a | ⁸⁷ Sr/ ⁸⁶ Sr ^b |
|----------------------------|-------------------------|-----------------|-----------------|---|
| | | (ppm) | | |
| <i>Madupite</i> | | | | |
| Badger Teeth | L79-BT3 | 215 | 6060 | 0.70539 ± 3 ^f |
| | L79-BT4 | 195 | 5780 | 0.70541 ± 4 |
| Middle Table | L79-MT1 | 297 | 4050 | 0.70557 ± 4 |
| | L79-MT2W ^c | 256 | 2530 | 0.70560 ± 4 |
| | L79-MT2M ^d | 255 | 2550 | 0.70563 ± 5 |
| | L79-MT2#2W ^c | 264 | 2570 | 0.70560 ± 3 |
| | L79-MT2#2M ^d | 261 | 2650 | 0.70560 ± 5 |
| | L79-MT3 | 286 | 4040 | 0.70556 ± 5 |
| Pilot Butte | L79-MT4 | 312 | 2450 | 0.70564 ± 6 |
| | L79-P1 | 234 | 3960 | 0.70544 ± 4 |
| | L79-P1C | 234 | 3980 | 0.70550 ± 5 |
| | L79-P2 | 214 | 4110 | 0.70554 ± 5 |
| | L79-P3 | 219 | 5110 | 0.70549 ± 5 |
| | L79-P4 | 164 | 6300 | 0.70541 ± 4 |
| | L79-P5 | 204 | 4810 | 0.70546 ± 5 |
| L79-P7 | 234 | 4610 | 0.70541 ± 5 | |
| <i>Wyomingite-orendite</i> | | | | |
| Black Rock | L79-BR2 | 323 | 2390 | 0.70552 ± 5 |
| | L79-BR3 | 214 | 2770 | 0.70530 ± 6 |
| | L79-BR5 | 352 | 2300 | 0.70541 ± 5 |
| Boars Tusk | L79-B1 | 130 | 3020 | 0.70779 ± 5 |
| | L79-B2C | 236 | 2420 | 0.70601 ± 4 |
| | L79-B3 | 275 | 2550 | 0.70609 ± 5 |
| | LH-1 ^e | 460 | 2700 | 0.70567 ± 5 |
| Cabin Butte | L79-C1 | 302 | 2810 | 0.70547 ± 3 |
| | L79-C2 | 289 | 2910 | 0.70550 ± 4 |
| Deer Butte | L79-D1 | 265 | 2620 | 0.70579 ± 3 |
| | L79-D2 | 245 | 3190 | 0.70537 ± 5 |
| | L79-D3 | 260 | 3410 | 0.70543 ± 4 |
| | L79-D3#2 | 265 | 3630 | 0.70545 ± 5 |
| Emmons Mesa | L79-E1 | 277 | 2370 | 0.70582 ± 4 |
| | L79-E2 | 277 | 2190 | 0.70585 ± 6 |
| | L79-E3 | 272 | 2380 | 0.70581 ± 3 |
| North Table | L79-NT1 | 292 | 1850 | 0.70579 ± 3 |
| Orenda Mesa | L79-S1 | 254 | 2550 | 0.70556 ± 4 |
| | L79-S2 | 258 | 2590 | 0.70574 ± 5 |
| | L79-S3 | 259 | 2470 | 0.70571 ± 4 |
| | L79-S4 | 258 | 2580 | 0.70564 ± 4 |
| | L79-S5 | 269 | 2460 | 0.70573 ± 4 |
| | L79-S6 | 266 | 2470 | 0.70564 ± 4 |
| South Table | L79-ST1 | 267 | 2190 | 0.70572 ± 5 |
| | L79-ST2 | 263 | 2300 | 0.70579 ± 4 |
| | L79-ST4 | 260 | 2310 | 0.70571 ± 5 |
| Steamboat Mesa | L79-SM1 | 317 | 1810 | 0.70597 ± 3 |
| | L79-SM2 | 321 | 2090 | 0.70581 ± 5 |
| | L79-SM3 | 323 | 1970 | 0.70594 ± 4 |
| | L79-SM4 | 297 | 2060 | 0.70602 ± 4 |
| | L79-SM5 | 318 | 2000 | 0.70596 ± 6 |

^a Isotope dilution analyses^b Normalized to ⁸⁶Sr/⁸⁸Sr=0.1194 and relative to Eimer & Amend: ⁸⁷Sr/⁸⁶Sr=0.70800 and SRM987: 0.71022^c 'wyomingite'-layer, see text for discussion^d 'madupite'-layer, see text for discussion^e Rb, Sr concentrations: Carmichael (1967); Sr isotopic composition from DePaolo and Wasserburg (1977)^f Errors reported are on the fifth decimal and represent 2σ_m**Table 3** (continued)

| Locality | Sample | Rb ^a | Sr ^a | ⁸⁷ Sr/ ⁸⁶ Sr ^b |
|-------------|--------|-----------------|-----------------|---|
| | | (ppm) | | |
| Zirkel Mesa | L79-Z1 | 261 | 2020 | 0.70574 ± 4 |
| | L79-Z2 | 312 | 2100 | 0.70578 ± 4 |
| | L79-Z3 | 322 | 2270 | 0.70571 ± 6 |
| | L79-Z4 | 336 | 2810 | 0.70578 ± 4 |
| | L79-Z5 | 302 | 3080 | 0.70570 ± 4 |
| | L79-Z6 | 286 | 3060 | 0.70568 ± 6 |

Madupite and wyomingite/orendite are readily distinguishable in the Nd–Sr isotope diagram (Fig. 2) where they plot as two relatively tight and distinct clusters. The Nd isotopic compositions of 7 madupites from the three major occurrences range from εNd = −12.3 to −10.5, while 11 wyomingites and orendites vary from εNd = −17.0 to −13.7 (see Table 2 for definition of εNd). Wyomingites and orendites are indistinguishable in this diagram. The lack of any systematic Nd or Sr isotopic difference between wyomingite and orendite supports the contentions of Ogden (1979) and Kuehner et al. (1981) that both rock types are derived from the same magma series and probably reflect variable conditions of magma emplacement and extrusion. Their relation will not be discussed further.

With one exception (Zirkel Mesa), samples from the same locality have identical ¹⁴³Nd/¹⁴⁴Nd ratios within the analytical error, which suggests homogeneity of the Nd isotopic composition for most occurrences. ⁸⁷Sr/⁸⁶Sr variations within each of the three investigated madupite occurrences (based on 15 samples) do not exceed the analytical uncertainty; Sr isotopic variations within wyomingite/orendite exposures (based on 36 samples from 10 locations) may be significant but are then very much smaller than the total observed variations.

Comparison with high-potassium rocks from Italy, Virunga and West Kimberley (Fig. 2) reveals several distinctive features:

(1) Variations in εNd and particularly in ⁸⁷Sr/⁸⁶Sr (if we exclude sample L79-B1) are comparatively small. It is remarkable that the Sr isotopic ratios of the madupites fall completely within the range of the orendites despite distinct εNd ranges.

(2) Not only are Nd isotopic ratios but also Sr isotopic compositions the least radiogenic so far observed for potassic volcanic rocks. This contrasts with generally coupled LREE and Rb/Sr enrichment or depletion in mantle processes as inferred from the inverse Sr–Nd isotopic correlation for oceanic basalts (Fig. 2).

The comparison also reveals similarities:

(1) In common with all high potassium volcanic rocks investigated so far (Italy, Virunga, West Kimberley and Gaussberg) Leucite Hills magmas have negative εNd signatures. By defining εNd as deviation from a chondritic reference Nd isotopic composition (DePaolo and Wasserburg 1976), negative εNd values indicate that these magmas are derived from, or contain a contribution from, an old source enriched in the (chondrite normalized) light REE relative to heavy REE.

(2) As with Nd–Sr isotopic relations in rocks from Italy, Virunga and West Kimberley, ⁸⁷Sr/⁸⁶Sr and εNd in Leucite

Hills rocks appear to be inversely correlated (Fig. 2). This could merely be an apparent effect produced by two populations. Should it be significant however, it could indicate binary mixing either as a source event or accompanying the volcanic activity. Binary mixing as the dominant cause of present-day isotopic variations is well supported by Pb isotope systematics and Pb–Sr–Nd isotopic correlations for the Italian and Virunga rocks (Vollmer 1976, 1977; Hawkesworth and Vollmer 1979; Vollmer and Hawkesworth 1980; Vollmer and Norry 1983a) and tentatively supported by $\epsilon\text{Nd} - 1/\text{Nd}$ covariance for West Kimberley rocks (McCulloch et al. 1983).

Discussion

The first question to be examined is whether, or to what extent, contamination with continental crustal material during magma ascent has affected the isotopic compositions. We will argue that observed isotopic variations are unlikely to have resulted from contamination but reflect source variations. Questions then discussed are: What can the isotope data tell us about the scale of source heterogeneities and the origin and evolution of the madupite and orendite magma sources?

Crustal contamination

Contamination of sub-continental magmas during their ascent through the crust has been convincingly demonstrated in several cases from the spread of isotopic compositions imparted by the contaminant on the original isotopic characteristics of the melts (e.g. Thompson et al. 1982; Carlson et al. 1981; Dickin 1981). Inherent difficulties of this method are uncertainties in defining the isotopic and element abundance and variability of Nd, Sr and Pb in the mantle and crustal end-members. Evidence will usually involve covariance between isotope ratios as well as major and trace elements, but contamination has also been suggested even if trace or major element correlations are lacking (Thompson et al. 1982; Carlson et al. 1981; Dickin 1981). This may be due to elemental abundances – unlike Nd, Sr and Pb isotope ratios – being determined not only by the mass fraction of the contaminant to the magma, but also by other processes such as fractional crystallization (Taylor 1980; DePaolo 1981). It may also be due to selective contamination without extensive assimilation (Patchett 1980).

Could the negative ϵNd isotopic compositions of the Leucite Hills magmas be due to *bulk contamination* of a melt which originally had a positive or near zero ϵNd signature as shown by most oceanic and continental basalts? An ϵNd value of -30 has been measured for a rock from the Archean Wyoming craton (DePaolo and Wasserburg 1979) and these authors consider such a value to be typical for $\sim 2,800$ Myr old continental crust. Although the isotopic compositions of both madupites and wyomingites/orendites are intermediate between a kimberlitic (Group I) melt (Smith 1983) and average Archean crust (De Paolo and Wasserburg 1979), most incompatible trace element concentrations are not. This is readily evident in Fig. 3 which shows that light REE abundances in Leucite Hills lavas are higher than either average crust or kimberlites, rather than intermediate as the model would require. Mixing of crustal material with melts other than kimberlites,

for instance basalts (alkali or tholeiitic), would accentuate the problem since they have lower incompatible element contents than kimberlites. Neither do considerations based on major elements such as the *mg*-value (Table 1) support this model.

Alternatively, could the Nd isotopic difference between madupite and wyomingite/orendite be a result of bulk contamination of a *madupite* magma with crustal material? Assuming for the contaminant $\epsilon\text{Nd} = -30$ and 26 ppm Nd, the average Nd composition ($\epsilon\text{Nd} = -11$) and concentration (290 ppm Nd) of madupite would require up to 85% assimilation to change the Nd isotopic composition of the magma to an orendite type with a $\epsilon\text{Nd} = -17$.

Such a mixture would have only about half the Nd concentration of wyomingite/orendite. The difficulties are similar to the ones discussed above. We conclude that *bulk contamination* can neither account for the negative ϵNd isotopic features of the Leucite Hills magmas nor for the Nd isotopic relation between madupite and orendite magmas.

Patchett (1980) considers *selective contamination* by partial melts and aqueous fluids to be more probable for basic magmas than bulk contamination. He points out that absence of mixing relationships may not provide an argument against this type of contamination as a magma may be contaminated repeatedly during its ascent through the crust. Possibly, such modified crustal contamination model for an ascending kimberlitic melt would remove some of the trace element objections we have noted with bulk contamination, since light REE abundances conceivably could be even higher in a contaminating partial melt of crustal origin than in orendite melts. However, plagioclase feldspar, will probably remain as a residual liquidus-phase and would retain Sr. The partial melt crustal contaminant would be depleted in this element. Since orendite and particularly madupite are strongly enriched in Sr relative either to kimberlite or usual crustal rocks (Fig. 3), such a model would not satisfy all the relative LIL-element relationships shown in Figure 3. The higher Ni content of wyomingite/orendite relative to madupite, but similar *mg*-values, would also be difficult to explain in such a model.

Sample L79-B1 from Boars Tusk is more radiogenic in Sr ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7078$) than any other rock from the Leucite Hills although its Nd isotopic composition is within the error of two other samples from Boars Tusk and Nd and Sr concentrations are not unusual either. It was taken from the outer xenolith-rich zone of Boars Tusk and its high Sr isotope ratio could reflect contamination by assimilation of crustal xenoliths but only if these had several thousand ppm Sr. This simply reflects the difficulty of changing, by assimilation, the Sr isotopic composition of a magma with 3,000 ppm Sr. An alternative interpretation for the high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of this sample will be discussed later.

With the possible exception of sample L79-B1, we consider it unlikely that either bulk or selective contamination had any significant effect on the isotopic compositions of the magmas. Isotopic ratios are regarded to be source features.

Source heterogeneities

Barton and van Bergen (1981) describe salitic pyroxenes as megacrysts and as cores in diopside phenocrysts in a

Leucite Hills lava and conclude that they originate by the disaggregation of xenolithic wall-rock fragments incorporated by the magma. Likewise, salites and other megacrysts in Campanian magmas were found to be in isotopic disequilibrium with each other and with the groundmass (Vollmer et al. 1981); these xenocrysts are suggested to be either source fragments, to be derived from different magma batches or to represent fragments of subvolcanic rocks incorporated by the magma. In all these cases isotopic heterogeneities within a rock will reflect small-scale source variations rather than crustal contamination effects. However, a search for Sr isotopic heterogeneities on a hand-specimen scale (samples L79-D3, L79-MT2, L79-P1: Tables 2, 3) showed no signs of isotopic disequilibria on this scale.

Sr isotopic variations between lavas from the same locality (butte, mesa) are usually small and Nd isotopic variations exceeding the analytical uncertainty have only been observed once, at Zirkel Mesa. Isotopic source heterogeneities are most pronounced *between* volcanic centres and are therefore estimated to be on a km to 10 km scale.

Madupite – orendite relations

The two discrete ranges in ϵNd for madupites (-10.5 to -12.30) and wyomingites/orendites (-13.7 to -17.0) demonstrate that both magma series come from two separate source regions, both of which had a history of light REE enrichment. The Nd isotope variations rule out derivation of these two magma types by different degrees of partial melting from a single source as suggested by Kay and Gast (1973), liquid immiscibility (e.g. Eby 1981), or derivation from a common magma by fractional crystallization as already rejected by Carmichael (1967), Ogden (1979) and Kuehner et al. (1981). However, despite the fact that the two source regions have distinct Sm/Nd histories the chemical affinity between madupite and wyomingite/orendite (Table 1 and Fig. 3) strongly suggests that not only their volcanic history but also their source evolution must have somehow been related.

In general, two alternative sets of models may explain a source relationship (Vollmer 1976): Observed isotopic variations may be dominated by, and primarily a result of, radiogenic growth within an ancient reservoir that has variable parent/daughter element ratios (model 1) or alternatively, (model 2) isotopic variations may now be predominately a result of mixing of two (or possibly more) reservoirs whose previous geochemical relationship may have been lost or is no longer apparent. Of course, any combination of these two basic sets of models may also be possible. The available data for the Leucite Hills do not justify discussion of any but the simplest possible model for each set of models.

Model 1: This model may in its simplest form be regarded as a two-stage model. We assume that the sources of the Leucite Hills magmas differentiated from a primitive mantle reservoir with chondritic Sm/Nd ratio and Rb/Sr=0.03, which is believed to be the best estimate for the bulk Earth ratio (DePaolo and Wasserburg 1976; O'Nions et al. 1977). Differentiation is certainly an old event, which can be broadly constrained by the Sm/Nd ratios in the rocks and the least radiogenic Nd to have occurred before 1.2 Gyr.

As the most likely time we will assume that this mantle enrichment event coincides with the stabilization of the Archean lithosphere in the Wyoming Province about 2.7 Gyr ago. Calculations show that a small enrichment in Nd/Sm (to $[\text{Nd}/\text{Sm}]_{\text{E.F.}}=1.24-1.36$) and Rb/Sr (to 0.036–0.042) is sufficient to generate the observed Nd and Sr isotopic variations. This corresponds to only 20–30% enrichment relative to bulk earth estimates for Nd/Sm and Rb/Sr. Total source heterogeneities would be 9% in Nd/Sm and 16% in Rb/Sr, but only half these values if madupite and orendite sources are considered separately. Larger and possibly more realistic source heterogeneities and higher Nd/Sm ratios would result were the enrichment event more recent. Sample L79-B1 could possibly represent a source domain with an exceptionally high Rb/Sr ratio, although this is not reflected in the measured Rb/Sr ratio. An upper mantle slightly enriched in LIL-elements during the Archean is a possible source for the Leucite Hills magmas.

Although this model implies distinct source Sm/Nd ratios (madupite source: 0.259; orendite source: 0.236 for $t=2.7$ Gyr) there is no discernible difference in Sm/Nd between madupites ($\text{Sm}/\text{Nd}=0.137\pm 0.014$; 1σ) and wyomingites/orendites ($\text{Sm}/\text{Nd}=0.134\pm 0.014$). This could provide an argument against the model although the effect of 10% source variations may be lost in the $\pm 20\%$ total variations in the measured ratio.

The degree of partial melting calculated for the two 2.7 Gyr old sources would be in the range of 1% or less, thus not quite as small as suggested by Kay and Gast (1973) who assumed a single source with chondritic REE abundances. A younger age for the enrichment event would require and allow higher degrees of melting since the two sources would still have to satisfy the Sr and Nd isotopic constraints of this two-stage evolution model, but then the former problem, that of distinct source Sm/Nd ratios for madupite and orendite magmas, would become more severe.

This model suggests a possible consanguinity between kimberlites and lamproites but leaves a major question unanswered: If both are incipient partial melts of an undepleted or LIL-element enriched mantle why are then their trace- and major-element chemistries significantly distinct (Fig. 3)? In particular, why is potassium enriched by an order of magnitude in lamproites relative to kimberlites and why is Nb depleted relative to other *hyg*-elements (Fig. 3)?

Model 2: Mixing models for the source evolution of potassic alkaline rocks have been discussed by Vollmer (1976, 1977), Hawkesworth and Vollmer (1979) and Vollmer and Hawkesworth (1980) for the Italian volcanism, by Bell and Powell (1969) and Vollmer and Norry (1983a) for Virunga, and by McCulloch et al. (1983) for West Kimberley. Dominance of radiogenic isotope variations by a mixing process can be suspected whenever it is suggested that metasomatising fluids enriched the magma sources in LIL-elements and therefore in the parent and/or daughter elements for the isotope systems considered here (Navajo minettes: Roden 1981; Rogers et al. 1982; Central Sierra Nevada ultrapotassic rocks: Van Kooten 1980, 1981). A metasomatic mixing process enriching the Leucite Hills magma source region in LIL-elements some time prior to partial melting is suggested by their extreme abundances in the magmas.

A presupposition of such a model is that at least the

observed Nd isotopic extremes¹ are pre-metasomatic features. The *difference* in ϵNd between the end-members as well as the strongly negative ϵNd signature of one end-member is in this model *not* a result of metasomatic light REE source enrichment but predates this event. The two magma series and their distinct sources – if not an effect of limited sampling associated with the partial melting event – could reflect an essential two-layered premetasomatic solid or specific and distinct mixing proportions for the madupite and orendite sources. The poor Sr–Nd isotopic correlations (Fig. 2) could, according to this model, reflect either pre-metasomatic isotopic heterogeneities in the solid (mixing between more than two components) or radiogenic growth during a time interval (if any) between the metasomatic event and the 1 Myr old partial melting event.

The data are insufficient to permit identification of the fluid end-member except, perhaps, the lesser scatter observed for madupites in the Nd–Sr isotope diagram (Fig. 2) may suggest the fluid to be the high ϵNd endmember, this is assuming the scatter reflects premetasomatic heterogeneities in the solid and not subsequent radiogenic growth.

The timing of a metasomatic event is difficult to estimate. Observed Sm/Nd variations are probably within the analytical error. This not only suggests that this ratio could have been little dispersed by variable degrees of partial melting but also that metasomatism caused strong enrichment of the light REE and resulted in uniform Sm/Nd ratios. These inferences reinforce the argument that observed Nd isotopic variations must predate metasomatism; it also follows that the Sm–Nd system is unsuitable for placing restrictions on the timing of the metasomatic event. A broad positive correlation between Rb/Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ for both the madupites and wyomingites/orendites is observed. If detailed source modelling should prove able to assess the degree of Rb/Sr fractionation during partial melting, then it may perhaps be possible to date a metasomatic event (see also discussion by Vollmer and Norry 1983a).

One could argue that model 2 is unlikely because it requires a pre-metasomatic reservoir (or source component) already possessing a strongly negative ϵNd signature. This implies two independent LIL-element enrichments of the same mantle region with a long time interval. However, analogy with the Italian and Virunga high-K magmas suggests exactly this. Strongly negative ϵNd source components in conjunction with clear Nd, Sr and Pb isotopic correlations are observed for these two occurrences; evidence for two enrichment events is therefore much stronger there than for the Leucite Hills. It has been proposed (e.g. Vollmer 1983; Vollmer and Norry 1983a) that for the Italian and Virunga high-K magma source, the early event was not a metasomatic event but the crust formation event and that the $-\epsilon\text{Nd}$ source component is continental crustal material. However, the present data for the Leucite Hills are insufficient to distinguish which of the two basic sets of models describes better the observed isotopic characteristics of the Leucite Hills magmas and it is therefore uncertain whether a general genetic model common for all three occurrences of high-K magmas can be found.

¹ The same may not necessarily be the case for Sr and Pb isotopic extremes because of the much shorter half-life of the radioactive parent and because metasomatism may in these isotope systems possibly cause large parent/daughter element variations. For an example and further discussion see Vollmer and Norry (1983a, b)

Conclusions

1. Crustal contamination by either bulk assimilation or selective contamination is unlikely to have been significant in determining the Nd and Sr isotopic characteristics of the Leucite Hills lamproites. Any contamination effect may be masked by the 5 to 10 times higher Nd and Sr abundances in these magmas compared to common crustal rocks. The maximum possible effect is likely to be less than the isotopic heterogeneities between lavas from a single volcanic centre (butte), that is less than the analytical uncertainty for ϵNd and less than 0.0003 for $^{87}\text{Sr}/^{86}\text{Sr}$ with the possible exception of one sample (L79-B1). We regard the Nd and Sr isotopic compositions of the lavas generally as being representative of the magma sources.

2. In common with highly potassic volcanic rocks from Italy, Virunga, West Kimberley and Gaussberg the Leucite Hills magmas contain a contribution from an old light rare earth element enriched source.

3. The lack of any systematic Nd and Sr isotope difference between wyomingites and orendites supports geochemical and petrological evidence for their derivation from a common magma series under differing cooling conditions.

4. The distinct Nd isotopic characteristics of madupite and wyomingite/orendite magmas rules out the possibility of a comagmatic origin by either fractional crystallization from a common primary magma, by liquid immiscibility or by variable partial melting conditions from a common source. Two distinct but heterogeneous sources are required for these two magma types. Isotopic and probably also trace element source heterogeneities are most pronounced on an inter-volcanic-centre scale.

5. The trace element abundance patterns leave no doubt that madupite and wyomingite/orendite magmas are not only related by their volcanic history but also through their source evolutions. The simplest relationship between the two sources is either by their differentiation from a common reservoir (a two-stage source evolution) or by mixing of two reservoirs which themselves may be unrelated or whose relationship may no longer be apparent. The first case could correspond to an upper mantle source for the Leucite Hills magmas which has been slightly and heterogeneously enriched in LIL-elements during the Archean. The second possibility could correspond to metasomatism of a $-\epsilon\text{Nd}$ solid by a fluid with an isotopic signature around $\epsilon\text{Nd}=0$ (or vice versa). Both models are possible although we favour the latter because it would be analogous to metasomatic source enrichment events involving an old LREE enriched component inferred for the Italian and Virunga potassic volcanism.

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