

Pb–Sr–Nd Isotope Data from 30 and 300 Ma Collision Zone Carbonatites in Northwest Pakistan

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RECEIVED SEPTEMBER 30, 1997; REVISED TYPESCRIPT ACCEPTED MAY 21, 1998

We present isotope data for the synorogenic Sillai Patti and Loe Shilman (30 Ma) and the pre-orogenic Koga and Jhambil (300 Ma) carbonatites, of the Indus Suture Zone. The younger carbonatites are foliated–banded sheet-like bodies in the metamorphosed belts of the Higher and Lesser Himalaya, within the Indus Suture Zone. They appear to be collision related and have no relationship to the silicate rocks in the complex. The initial isotope ratios are: ϵ_{Nd} -3.1 to -3.8 ; $^{87}Sr/^{86}Sr$ 0.70463 – 0.70486 (ϵ_{Sr} $+2.4$ to $+5.6$); $^{206}Pb/^{204}Pb$ 19.01 – 21.35 ; $^{207}Pb/^{204}Pb$ 15.54 – 15.67 ; $^{208}Pb/^{204}Pb$ 38.29 – 40.63 . The pattern is atypical given that carbonatites generally yield positive ϵ_{Nd} and negative ϵ_{Sr} . The Nd, Sr, and least radiogenic Pb isotope ratios also fit the pattern of carbonatites from the East African Rift, suggesting derivation from similar sources. In that case a lithospheric source for the 30 Ma Pakistan carbonatites was probably transported with the Indian plate during migration from East Africa to the collision with the Asian continent. Intrusion of the carbonatites into rocks of the Himalayan orogenic zone apparently affected only the Pb isotopes in some of the plutons. Nd, Sr and Pb isotope patterns for the 300 Ma Pakistan plutons fit those for most intra-plate carbonatites, with positive ϵ_{Nd} and negative ϵ_{Sr} , showing that neither transport nor collision noticeably disturbed their isotope patterns.

KEY WORDS: carbonatite; isotope tracers; mantle; Pakistan; petrogenesis

INTRODUCTION

Virtually all known carbonatite plutons have been intruded within plates by anorogenic processes, usually in

rift zones, and are thought to provide information on the chemical characteristics of the underlying mantle (e.g. Bell *et al.*, 1982; Bell & Blenkinsop, 1987*a*, 1987*b*; Nelson *et al.*, 1988; Kwon *et al.*, 1989). The carbonatites of Pakistan are especially interesting because they crop out within an orogenic collision zone, and include both synorogenic and pre-orogenic plutons. This opens questions concerning the effect of orogenic activity on the isotope ratios in the bodies. How do the synorogenic carbonatites of Pakistan compare with those of similar age in anorogenic settings? What effect, if any, did transport and the Himalayan orogenic activity have on the pre-orogenic plutons? With those questions in mind we have studied two 30-my-old (Loe Shilman and Sillai Patti) and two 300-my-old (Koga and Jhambil) carbonatites from northwestern Pakistan. These are also the first carbonatite isotope tracer data from the Himalayan belt, and add to the limited amount of data from Asia compared with other continents.

GEOLOGICAL SETTING AND SAMPLE DESCRIPTIONS

The sample locations are given in the geologic sketch-map of Fig. 1. All plutons are situated between the Main Mantle Thrust (Indus Suture Zone) and Main Boundary Thrust. The following descriptions are summarized largely from Le Bas *et al.* (1987).

The synorogenic Loe Shilman carbonatite is exposed in sheets consisting mainly of sövite, with associated

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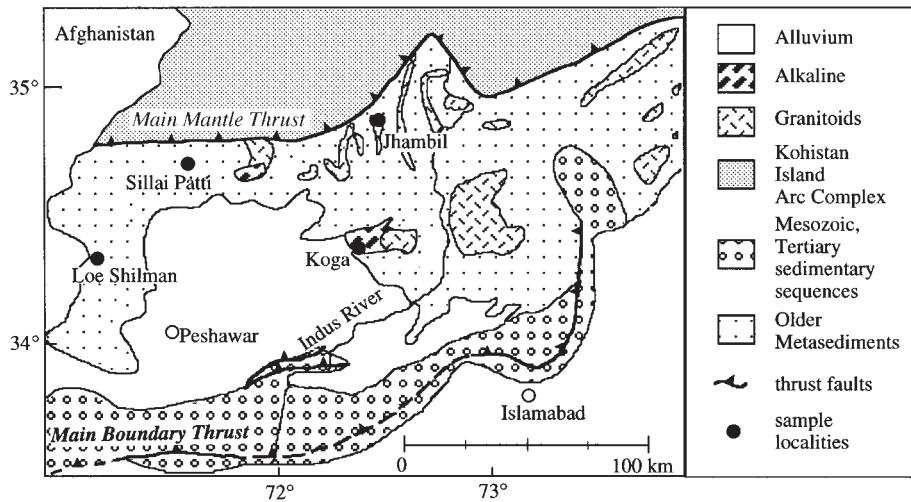


Fig. 1. Geologic sketch-map of sample localities in northern Pakistan. Adapted from Shams *et al.* (1995).

potassic rocks and lamprophyres. The carbonatites are ~150 m wide by 2.5 km long, emplaced along an E–W thrust plane that dips to the north. The country rock consists of Paleozoic(?) metasediments over Precambrian(?) slates and phyllites. The surrounding slates and phyllites are fenitized.

The synorogenic Sillai Patti carbonatite occurs as a sheet along a south-dipping thrust plane. No genetically associated alkaline rocks are noted. The country rock consists of either metasediments of possible Paleozoic age, or granite–gneiss of unspecified age. Le Bas *et al.* (1987) argued that metamorphism did not exceed greenschist facies at Loe Shilman, but reached at least biotite grade at Sillai Patti. Butt *et al.* (1989) have given a more detailed description of the Sillai Patti carbonatite and its country rock.

The pre-orogenic Koga carbonatite occurs as a plug plus veins within a nepheline syenite intrusion that was emplaced into metasediments and gneissic rocks. Ijolite and fenite are also present, as are calcite crystals with diameters up to 1 cm. There is no evidence for post-intrusion metamorphism.

The pre-orogenic Jhambil pluton lacks associated igneous rocks except for a few lamprophyre dikes. Carbonatites occur within the psammitic rocks of the Pacha Formation (Chaudhry *et al.*, 1992). The country rocks have experienced metamorphism up to amphibolite facies. The Jhambil carbonatite also contains large calcite crystals with diameters up to 1 cm.

Le Bas *et al.* (1987) reported age information for three of the carbonatites. Rb–Sr data from euhedral biotites define an age of 31 ± 2 Ma for Loe Shilman and Sillai Patti. Those researchers believed that the biotites date a crystallization rather than a metamorphic event because their texture suggests they crystallized with the calcite.

K–Ar measurements also gave ages of 30–33 Ma for the undeformed biotites, but lower ages of 24 ± 2 Ma for deformed crystals. Le Bas *et al.* ascribed the 24 Ma age to a later phase of movements along the thrusts. More recently, Qureshi *et al.* (1991) have reported fission track ages of 32.1 ± 1.9 Ma for zircon and 22.7 ± 1.1 Ma for apatite from the Sillai Patti carbonatite. Those workers related the 32.1 Ma ages to the emplacement of the carbonatite, and assigned the 22.7 Ma age of the apatite to a later ‘thermal–tectonic episode’. Their results agree reasonably well with the K–Ar data of Le Bas *et al.* (1987). We will use 30 Ma for the age of the carbonatites.

At Koga Le Bas *et al.* (1987) reported an Rb–Sr age of 315 ± 15 Ma for the surrounding granitic rocks and 297 ± 4 Ma for the syenites and ijolites of the complex. Here we assign an age of 300 Ma to the complex, and will assume an age of 300 Ma for the Jhambil carbonatite based upon the similarity to the Koga isotopic data described below.

In the absence of Pb isotope data from igneous rocks and sediments in northwest Pakistan, we measured Pb, Sr and Nd isotope ratios in two Eocene Salt Range limestones (Warwick & Husain, 1990) to provide an approximate idea of the isotopic compositions of Pb, Sr and Nd in crustal sources. The Salt Range is located ~200 km southeast of Peshawar, off the map in Fig. 1.

ANALYTICAL PROCEDURES

All analyses used calcite phases to avoid possible contamination from foreign minerals. The centimeter-size Koga and Jhambil calcite crystals were washed in dilute (0.3 M) hydrochloric acid to clean the surfaces and then were dissolved in the acid. Because our Loe Shilman and

Table 1: Sr–Nd data for calcite from Pakistan carbonatites

Data ¹	Loe Shilman 166	Loe Shilman 608	Sillai Patti 783	Sillai Patti 788	Koga 621	Koga 627	Jhambil 127
ppm Nd	119.6	231.9	176.5	227	198.5	152.4	165.7
ppm Sm	19.12	32.49	30.67	36.42	31.21	23.62	26.72
¹⁴³ Nd/ ¹⁴⁴ Nd ¹	0.512422	0.512457	0.512437	0.512455	0.512627	0.512598	0.512607
¹⁴⁷ Sm/ ¹⁴⁴ Nd	0.0966	0.0847	0.105	0.097	0.0951	0.0937	0.09616
Age (Ma)	30	30	30	30	300	300	300
¹⁴³ Nd/ ¹⁴⁴ Nd ²	0.512403	0.512440	0.512416	0.512455	0.512440	0.512414	0.512418
$\epsilon(\text{Nd})^3$	−3.8	−3.1	−3.6	−3.2	+3.7	+3.2	+3.2
ppm Sr	9450	8250	6850	11700	15000	13300	
⁸⁷ Sr/ ⁸⁶ Sr ⁴	0.704632	0.704668	0.704859	0.704752	0.703549	0.703485	0.703550
$\epsilon(\text{Sr})^5$	+2.4	+2.9	+5.6	+4.0	−8.5	−9.4	−8.5

¹Observed Nd ratios based upon ¹⁴⁶Nd/¹⁴⁴Nd = 0.721903, which yields a ¹⁴³Nd/¹⁴⁴Nd ratio of 0.511895 for Ames Nd, and 0.512638 for BCR-1 inter-laboratory basalt standard. External reproducibility is $\pm 0.004\%$; internal reproducibility is $\pm 0.002\%$.

²Initial ratios.

³Relative to chondritic bulk silicate Earth in units of 10^{-4} . Parameters are: ¹⁴⁷Nd/¹⁴⁴Nd = 0.1967 (Jacobsen & Wasserburg, 1984); age of the Earth = 4.56×10^9 years.

⁴Observed ratios. ⁸⁷Sr/⁸⁶Sr = 0.71020 for the NBS 987 standard.

⁵Parameters for evolution of ⁸⁷Sr/⁸⁶Sr in bulk silicate Earth are: initial ratio at 4.55 Ga = 0.69898; ⁸⁷Sr/⁸⁶Sr in present-day bulk silicate Earth = 0.7045 with ⁸⁷Rb/⁸⁶Sr = 0.08241. All ratios are normalized to ⁸⁶Sr/⁸⁶Sr = 0.119400.

Sillai Patti samples lacked large calcite crystals, residues amounting to 22–30% of the total samples remained after dissolution in dilute HCl. For that reason Pb isotope ratios for those plutons were measured on a second set of samples dissolved in 5 M acetic acid in place of the dilute (0.3 M) HCl; 5 M acetic acid is known to be highly selective compared with other acids for dissolving calcite (e.g. Schultz *et al.*, 1989). Inspection of the Pb isotope data in Table 2 (see below) shows that the dilute HCl dissolution procedures did not appear to release significant Pb contamination from the residues.

All chemical separations employed common ion exchange procedures as described by Tilton & Bell (1994). Concentrations were measured by isotope dilution using a Finnigan MAT 261 multi-collector mass spectrometer. The 2σ uncertainties of the means for Sr and Nd analyses were typically about $\pm 0.002\%$ for single runs, and 0.004% for replicate runs. For lead the 2σ uncertainties are $\sim 0.04\%$ per mass unit.

DISCUSSION

All carbonatite Nd–Sr data are given in Table 1 and plotted in Fig. 2. As an index for isotope patterns that are consistent with mantle sources in this and subsequent figures, we will use the present-day mantle end-member components, DMM, HIMU and EM1, as defined from oceanic volcanic rocks by Hart *et al.* (1992). To compare initial ratios with estimated values for bulk silicate Earth, it is necessary to correct for the age of the samples. For

Nd we use the chondrite uniform reservoir (CHUR) of Jacobsen & Wasserburg (1984) to represent average Earth evolution. Sr isotope evolution is less accurately known. Here we use 0.7045 for ⁸⁷Sr/⁸⁶Sr of bulk silicate Earth today, but others use 0.7047. The true value could be outside those limits. For that reason we will use the observed ratios for all Sr data, and indicate the bulk silicate Earth ratios calculated from our model.

Figure 2 shows the correlation between Nd and Sr for the Pakistan data, and compares them with other young (<130-my-old) carbonatites from around the world. The squares with dots represent four sets of data for intra-plate plutons from three continents, plus two additional plutons from oceanic settings. Because the six isotope patterns are similar in spite of very diverse locations, we will use those samples as a reference group for modern carbonatite isotope patterns.

A striking feature for the 30-my-old Pakistan carbonatites is their very negative ϵ_{Nd} values, coupled with the high ⁸⁷Sr/⁸⁶Sr ratios. For most isotopically analyzed carbonatites the ¹⁴³Nd/¹⁴⁴Nd ratios are greater than the bulk silicate Earth ratio (positive ϵ_{Nd}), as outlined in the summaries by Bell & Blenkinsop (1989) and Harmer & Gittins (1998), and shown by the reference group in Fig. 2. Out of 128 analyses by Harmer & Gittins (1998) for carbonatites from around the world with ages <200 Ma, 100 yield positive ϵ_{Nd} values, generally paired with negative ϵ_{Sr} . The remaining negative ϵ_{Nd} values are predominantly from East Africa, Brazil and Asia (India).

In their original study of East African Rift carbonatites with ages between 0 and 120 Ma, Bell & Blenkinsop

Table 2: Lead isotope data for calcite from Pakistan carbonatites

Data	L. Shilman 166	L. Shilman 608	S. Patti 783	S. Patti 788	Koja 261	Koga 627	Jhambil 127
$^{206}\text{Pb}/^{204}\text{Pb}^1$	19.445	19.025	20.497	21.362	18.872	18.643	18.758
$^{207}\text{Pb}/^{204}\text{Pb}^1$	15.542	15.573	15.634	15.673	15.614	15.601	15.609
$^{208}\text{Pb}/^{204}\text{Pb}^1$	40.629	39.734	39.328	39.513	38.937	38.720	38.910
ppm Pb	6.56	8.75	11.36	14.07	10.98	4.09	6.52
ppm U	0.342	0.494	1.53	0.338	0.0735	0.0132	0.140
ppm Th	0.139	3.12	4.27	0.137	4.559	0.0521	0.508
age (Ma)	30	30	30	30	300	300	300
$^{238}\text{U}/^{204}\text{Pb}$	3.45	3.68	8.91	1.61	0.431	0.206	1.37
$^{232}\text{Th}/^{204}\text{Th}$	1.45	24.0	25.7	0.675	27.6	0.842	5.17
$^{232}\text{Th}/^{238}\text{U}$	0.42	6.53	2.88	0.419	64.1	4.08	3.76
$^{206}\text{Pb}/^{204}\text{Pb}^2$	19.429	19.008	20.455	21.354	18.851	18.633	18.693
$^{207}\text{Pb}/^{204}\text{Pb}^2$	15.541	15.572	15.632	15.672	15.613	15.600	15.605
$^{208}\text{Pb}/^{204}\text{Pb}^2$	40.627	39.698	39.290	39.512	38.524	38.707	38.833
$^{206}\text{Pb}/^{204}\text{Pb}^3$	19.490	19.001	20.465	21.354			18.706
$^{207}\text{Pb}/^{204}\text{Pb}^3$	15.581	15.561	15.618	15.661			15.604
$^{208}\text{Pb}/^{204}\text{Pb}^3$	40.766	39.539	39.239	39.477			39.789

¹Absolute ratios obtained by adjusting observed ratios for replicate runs on the measured ratios normalized to standard values given by Todt *et al.* (1995). The average fractionation corrections were $0.12 \pm 0.01\%$ per mass unit.

²Calculated initial ratios.

³Samples dissolved in acetic acid in place of hydrochloric acid. (See text for explanation.)

(1987*b*, 1989) found that six out of nine carbonatites from Uganda, Kenya, and Tanzania gave negative ϵ_{Nd} values ranging from -0.5 to -3.7 . Figure 2 has eight East African samples with negative ϵ_{Nd} out of 19 samples. Bell & Blenkinsop also found that their samples defined a line in an $\epsilon_{\text{Nd}}-\epsilon_{\text{Sr}}$ diagram that they called the East African Carbonatite Line, which tracks the present EM1–HIMU line in Fig. 2 rather closely. The young Pakistan carbonatite data plot near the EM1 end member along the EM1–HIMU line, although their separation between the HIMU–EM1 and DMM–EM1 lines is minimal. A clearer resolution will be shown in Fig. 3.

The Nd–Sr data from the 30-my-old Pakistan carbonatites differ very substantially from those of the 60-my-old carbonatite complex of Amba Dongar of western India (Simonetti *et al.*, 1995). Although ϵ_{Nd} is negative in both groups, the Amba Dongar carbonatites are distinguished by unusually high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. Simonetti *et al.* (1995) concluded that the Sr isotopes have been altered by low-temperature interaction with crustal materials.

In contrast to the younger carbonatites, the 300-my-old Koga and Jhambil carbonatites have positive values for ϵ_{Nd} and plot within the field of the comparison group carbonatites from both continental and oceanic settings, indicating differing isotopic compositions for Nd and Sr in the carbonatite sources between 300 and 30 Ma. However, it should be noted that, although the Nd and

Sr isotope ratios for bulk silicate Earth can be estimated with reasonable accuracy at 300 Ma, the ratios for DMM, HIMU, and EM1 are less well known because the absence of seafloor volcanics older than ~ 190 Ma precludes obtaining direct information beyond that age. Thus we cannot evaluate the isotope data for the sources in terms of specific mantle end members, although the positive ϵ_{Nd} and negative ϵ_{Sr} values argue for substantial contributions from HIMU- and/or DMM-like components.

The limestone ratios, $^{87}\text{Sr}/^{86}\text{Sr} = 0.7097$ and $\epsilon_{\text{Nd}} = -5.7$ (see Table 3), cannot be shown in the field of Fig. 2. They signify substantial input from aged sialic crustal material.

Figure 3 compares $^{206}\text{Pb}/^{204}\text{Pb}$ data from Table 2 with ϵ_{Nd} for the samples in Fig. 2. This diagram has the distinct advantage that DMM and EM1 are much better separated than in Fig. 2. Accordingly, their contributions can be evaluated more accurately. The Pb and Nd isotopic compositions for the majority of the African Rift carbonatites are described by mixing between the HIMU and EM1 model mantle end members. There is no indication of two-component mixing between DMM and EM1, as might be inferred in Fig. 2. In considering the actual proportions of end members in a magma source, it is necessary to consider the Pb, Sr and Nd elemental abundances as well as the isotope ratios. Although the relative abundances of Pb, Sr and Nd in the HIMU, EM1 and DMM sources seem to be similar (Hart, 1988),

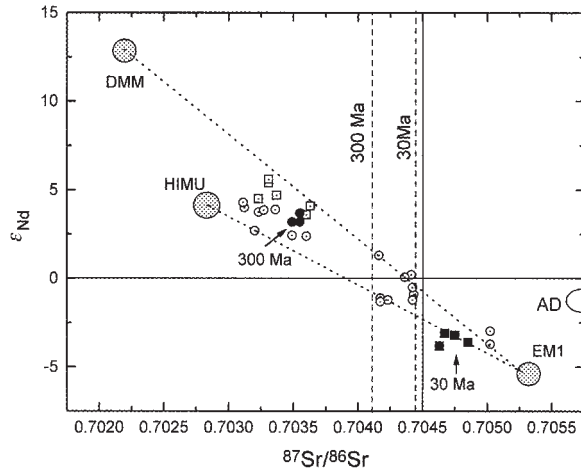


Fig. 2. Nd–Sr correlation diagram for young (<130 Ma) carbonatites. Filled symbols are Pakistan data. Circles with dots are African Rift samples from Bell & Blenkinsop (1987*b*), Nelson *et al.* (1988), Simonetti & Bell (1994*a*, 1994*b*), Bell & Simonetti (1996) and Kalt *et al.* (1997). Squares with dots are Oka (Grünenfelder *et al.*, 1986), Arkansas (Tilton *et al.*, 1987), New Zealand (Barreiro & Cooper, 1987), Kaiserstuhl (Nelson *et al.*, 1988), Canary Island (Hoernle & Tilton, 1991) and Cape Verde (K. A. Hoernle *et al.*, unpublished data, 1995). DMM, EM1 and HIMU are mantle end-member components from Hart *et al.* (1992). $^{143}\text{Nd}/^{144}\text{Nd}$ for modern bulk silicate Earth is 0.512638, for which $\epsilon_{\text{Nd}} = 0$. $\epsilon_{\text{Nd}} = [(R_{\text{MEAS}}/R_{\text{BSE}}) - 1] \times 10^4$, where $R = ^{143}\text{Nd}/^{144}\text{Nd}$ and BSE is bulk silicate Earth at any given time. $^{87}\text{Sr}/^{86}\text{Sr}$ for present bulk silicate Earth is taken as 0.7045. Vertical dashed lines show model bulk silicate Earth ratios at 30 and 300 Ma. AD, Amba Dongar carbonatite complex data from Simonetti *et al.* (1995). The Eocene limestone data ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7096$ and $\epsilon_{\text{Nd}} = -5.6$) plot outside the field of this diagram.

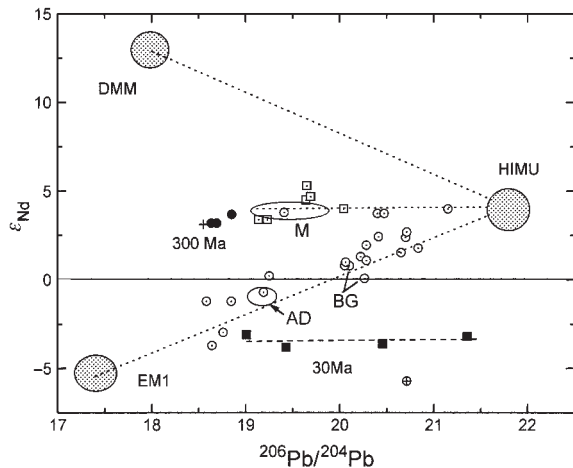


Fig. 3. ϵ_{Nd} vs $^{206}\text{Pb}/^{204}\text{Pb}$ for carbonatite samples in Fig. 2. Pb data are from Nelson *et al.* (1988), Simonetti & Bell (1994*b*), Bell & Simonetti (1996), Kalt *et al.* (1997), and G. R. Tilton & K. Bell (unpublished data, 1995). BG, Bala Gully carbonatite from Kalt *et al.* (1997). (See text for explanation.) M, Malawi data from Simonetti & Bell (1994*b*). Cross is Sokli (Finland) carbonatite, age 350 Ma (Patchett *et al.*, 1981). ϵ_{Nd} for Sokli is derived from the formula $\epsilon_{\text{HF}} = 2\epsilon_{\text{Nd}} + 2$ (Vervoort & Patchett, 1996). Circle with cross is Eocene limestone from Table 3. Other symbols and data sources as in Fig. 2. DMM, HIMU and EM1 as in Fig. 2.

Table 3: Salt Range limestones

Data	1043	1047
$^{206}\text{Pb}/^{204}\text{Pb}^1$	20.230	20.707
$^{207}\text{Pb}/^{204}\text{Pb}^1$	15.816	15.857
$^{208}\text{Pb}/^{204}\text{Pb}^1$	39.528	39.504
ppm Pb	1.95	5.56
ppm U	0.383	1.60
ppm Th	0.535	3.48
$^{238}\text{U}/^{204}\text{Pb}$	13.0	19.1
$^{232}\text{Th}/^{238}\text{U}$	1.44	2.26
$^{87}\text{Sr}/^{86}\text{Sr}^{1,2}$	0.709669 ± 18	0.709632 ± 18
ppm Sr	266	360
$^{146}\text{Nd}/^{144}\text{Nd}^{1,3}$	—	0.512345 ± 15
E_{Nd}	—	−5.7

¹Measured ratios.

²Normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.119400$.

³Normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.721900$.

the absolute abundances are less well known. Assuming approximately equal absolute as well as relative abundances in the mantle end members, the scatter of data about the EM1–HIMU mixing line in Fig. 3 is accounted for by either variations in source coupled Sm/Nd–U/Pb ratios, or contributions of up to a few per cent from DMM sources. The contribution from DMM could be higher because its U, Sr and Nd abundances are lower than those in EM1 and HIMU, but the low concentrations of lithophile elements in mid-ocean ridge basalt (MORB) are generally ascribed to higher degrees of partial melting rather than to lower source abundances.

The young Pakistan carbonatites fit an anomalous Pb–Nd pattern. They are the only samples among the total population in Fig. 3 that plot far outside the mantle end-member field. Significantly, the Loe Shilman sample with the least radiogenic lead also fits the African Rift trend reasonably well. All of the 30 Ma Pakistan data fit the East African Rift trend satisfactorily if one accepts that the other carbonatites experienced varying degrees of Pb contamination, presumably during the collision processes. There is no correlation between U/Pb ratios and Pb isotope ratios in the Pakistan samples, further suggesting inherited radiogenic lead. The limestones contain highly radiogenic Pb from crustal sources, but they do not match the required contamination in the carbonatites.

A possible second group of African Rift carbonatites define a horizontal pattern that emanates from HIMU. This suggests that HIMU might determine an approximate upper limit for ϵ_{Nd} for most carbonatites. The pattern can be explained either by addition of DMM components or some odd type of Pb contamination of

an HIMU source. We return to this question in the discussion for Fig. 6 (see below).

Significantly, the Amba Dongar data, which plotted outside the mantle field in Fig. 2, plot along the EM1–HIMU trend in this diagram. The failure of the Amba Dongar data to fit the mantle model observed in the other carbonatites is thus clearly due to an anomalous Sr isotopic composition, as Simonetti *et al.* (1995) previously noted. We have highlighted the data for the Bala Gully East African Rift carbonatite from Kalt *et al.* (1997) because they, like the Amba Dongar data, do not plot within the mantle field in all subsequent diagrams.

The Koga–Jhambil Nd–Pb data plot in the middle of the mantle field in Fig. 3, close to the reference carbonatites. In this figure it is clear that large proportions of DMM-like components are required to account for the observed ratios in the three-component mantle model. Data for the 300-my-old Pakistan carbonatites can also be compared with those from the 350-my-old Sokli carbonatite of Finland (Vartiainen & Wolley, 1974). The Nd value is estimated from the ϵ_{Hf} value of 8.2 ± 0.7 of Patchett *et al.* (1981), which translates to $\sim +4$ for ϵ_{Nd} compared with $+3.5$ for the 300 Ma Pakistan plutons; the Pb ratio is an unpublished Santa Barbara analysis. The agreement between the two localities is reasonable.

The Pb and Sr data are shown in Fig. 4, which is almost a mirror image of Fig. 3. Therefore most of the remarks made in the discussion above for the Nd diagram apply also to the Sr diagram. The Pb–Sr data from the African Rift exhibit greater scatter along the EM1–HIMU trend than do the Pb–Nd data in Fig. 3. There is some suggestion of a trend between HIMU and the group that describes minimum values of $^{87}\text{Sr}/^{86}\text{Sr}$ as a function of $^{206}\text{Pb}/^{204}\text{Pb}$ for the 30 Ma carbonatites, somewhat similar to the limiting trend observed with Pb and Nd in Fig. 3. The Pb data again plot outside the mantle end-member field, as in Fig. 3. The Koga and Jhambil carbonatite data plot in the middle of the diagram, removed from the field for the Sillai Patti and Loe Shilman samples.

The high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the Amba Dongar carbonatite places it far outside the East Africa–Pakistan field, and even outside the DMM–HIMU–EM1 field. The Bala Gully carbonatite data likewise plot considerably above the HIMU–EM1 mixing line, suggesting Sr contamination similar to that in the Amba Dongar carbonatites, but on a less extreme scale. The limestone Sr ratios plot outside the range of the diagram.

Figure 5 gives $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios from the carbonatites. Lead ratios of the 30-my-old samples define a regression line that yields a slope corresponding to a Pb/Pb age of ~ 35 Ma, but the uncertainty is much too large to provide useful time information. However, Figs 3 and 4 show clearly that the Pb isotope patterns for the Loe Shilman and Sillai Patti carbonatites are

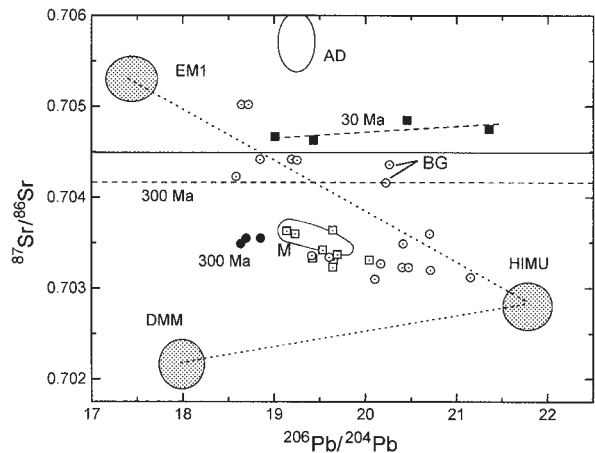


Fig. 4. Pb–Sr correlation diagram for samples in Fig. 3. Symbols and identities as in Fig. 3. Dashed line is estimated bulk silicate $^{87}\text{Sr}/^{86}\text{Sr}$ ratio at 300 Ma. The Eocene limestone data ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7096$) plot far outside the field of the figure.

unusual compared with Pb in the remaining samples, suggesting contamination. With present information, we conclude that the 30-my-old Pakistan carbonatites have experienced varying degrees of Pb contamination from older, crustal(?) rocks as a result of the orogenic processes that accompanied their intrusion. These results are particularly interesting because the East African carbonatites formed in a continental rift setting, whereas the Loe Shilman and Sillai Patti plutons originated in a continental collision setting, yet they fit a similar initial isotope ratio pattern if the Pb contamination model for the Pakistan plutons is correct.

The Amba Donga carbonatites have unusually high $^{207}\text{Pb}/^{204}\text{Pb}$ ratios that differ from those in the young Pakistan samples. The two localities thus differ in Pb as well as in Sr isotopic compositions. The Bala Gully Pb are not noticeably anomalous.

The Eocene limestones contain very radiogenic lead with the highest $^{207}\text{Pb}/^{204}\text{Pb}$ ratios found in this study, indicating aged, probably crustal, sources. The lead does not fit the isotopic composition required for Pb contamination in the above model, which suggests that the Pb contamination in the carbonatite is not due to shallow-level sources.

The Koga–Jhambil Pb isotopic compositions plot on the left side of the field of the younger plutons, which is consistent with their older age. Significantly, the ratios do not exhibit the anomalies found for the 30-my-old Pakistan carbonatites. The ratios also somewhat resemble those for the Sokli (Finland) carbonatite, as noted for the Nd and Pb isotope data in Fig. 3.

Another method of analyzing the results is to view the fit of the Nd–Sr–Pb data to the mantle plane described by the model end members, HIMU, EM1 and DMM,

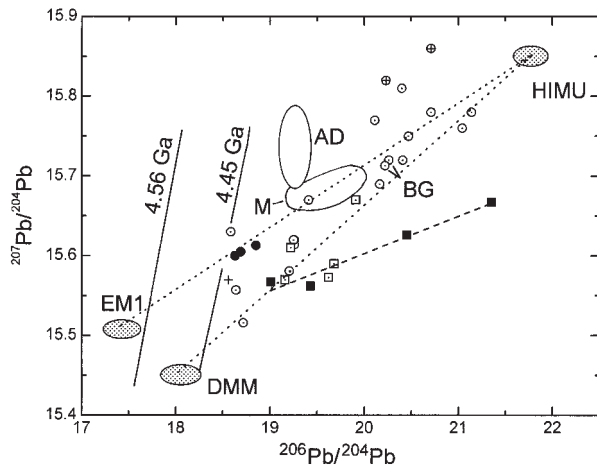


Fig. 5. $^{207}\text{Pb}/^{204}\text{Pb}$ – $^{206}\text{Pb}/^{204}\text{Pb}$ correlation diagram for samples in Fig. 3. The two 0 Ga isochrons are drawn for fractionation of uranium and lead at 4.56 and 4.45 Ga. Allègre *et al.* (1995) have suggested 4.45 Ga for the age of the Earth's U–Pb reservoir, which they interpreted as the time of core separation. Cross is the Sokli, Finland, carbonatite (G. R. Tilton, unpublished data, 1995). Circle with cross is Eocene limestone.

in two-dimensional space. Zindler *et al.* (1982) showed how to depict the plane as a line in two-dimensional space by combining $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ values into a term plotted against $^{87}\text{Sr}/^{86}\text{Sr}$. It should be noted that plotting along the Fig. 6 mantle plane is a necessary, but not sufficient, criterion to establish pure mantle model sources for the magmas. It is certainly possible to combine isotope data from outside the plane so as to produce magmas that plot along the schematic plane; however, this cannot be true in the present case for those ratios that plot within the EM1–DMM–HIMU triangles in Figs 3–5.

Figure 6 illustrates the 30 Ma Pakistan and other young carbonatite data in the simplified diagram. The reference group and African Rift samples, excepting Bala Gully, fit the mantle plane well, especially considering their scattered locations, which include continental and oceanic settings. The ratios from Loe Shilman closely fit the mantle plane, whereas the Sillai Patti data are farther removed, but if we accept the Pb contamination model and adjust the Sillai Patti $^{206}\text{Pb}/^{204}\text{Pb}$ ratios to 19, the fit to the mantle plane is then very good.

The pattern in Fig. 6 answers the question raised in the discussion of data in Fig. 3, namely, whether the horizontal line defined by the reference group and some of the African rift samples resulted from Pb contamination that did not affect the Sr or Nd isotope ratios. Figure 6 shows that the Pb isotope variations appear to involve mantle, rather than random, possibly crustal, sources. The horizontal trend of the data in Fig. 3 remains a puzzle. Perhaps the pattern is accidental, and would disappear if more data were available. Although the

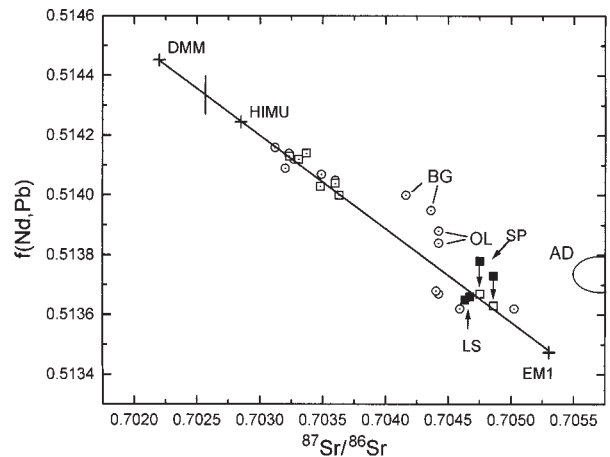


Fig. 6. End-on view of the model mantle plane defined by HIMU, EM1 and DMM as given by Hart *et al.* (1992). The equation for the mantle plane is: $-0.428851(^{87}\text{Sr}/^{86}\text{Sr}) - 1.358437(^{143}\text{Nd}/^{144}\text{Nd}) - 8.75115E - 5(^{206}\text{Pb}/^{204}\text{Pb}) + 1 = 0$. $f(\text{Nd,Pb})$ [based upon Zindler *et al.* (1982)] = $[(^{143}\text{Nd}/^{144}\text{Nd})^2 + (^{206}\text{Pb}/^{204}\text{Pb})^2] \times \{\sin[\arctan(^{143}\text{Nd}/^{144}\text{Nd} + ^{206}\text{Pb}/^{204}\text{Pb}) + 0.000064]\}$. The error bar shows the uncertainty for ± 1.0 in $^{206}\text{Pb}/^{204}\text{Pb}$, or ± 0.00018 in $^{87}\text{Sr}/^{86}\text{Sr}$, or ± 0.000065 in $^{143}\text{Nd}/^{144}\text{Nd}$. Open squares are Sillai Patti carbonatites (SP) corrected for Pb contamination as explained in the text. OL indicates the extremes from 11 analyses of Oldoinyo Lengai carbonatites in Bell & Simonetti (1996). BG, Bala Gully samples from Kalt *et al.* (1997). LS, Loe Shilman. Eocene limestone data, not shown, yield $f(\text{Nd,Pb}) = 0.5136$; $^{87}\text{Sr}/^{86}\text{Sr} = 0.7096$. The 300 Ma Pakistan carbonatites are not included because past values of DMM, EM1 and HIMU are not well enough known at that time.

Amba Dongar and Bala Gully carbonatites do not fit the mantle plane, the reason in those cases involves the Sr, rather than the Pb ratios, as is clearly shown in Fig. 3.

To summarize, a small amount of crustal contamination accompanying the collision process can explain the difference between the Loe Shilman and Sillai Patti carbonatite Pb data. The Amba Dongar field is removed from the plane because of high $^{87}\text{Sr}/^{86}\text{Sr}$ values; the limestone ratios likewise plot far outside the field of the diagram for the same reason.

MODELS

Among the young (<130-my-old) carbonatites of the world, negative values of ϵ_{Nd} are found almost entirely in some of the carbonatites in the African Rift, northern Pakistan, Brazil, and in the 60 Ma Amba Dongar carbonatites of northwestern India. We exclude the Amba Dongar carbonatites because they do not plot on the mantle plane in Fig. 6, and have anomalously high $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios compared with the other carbonatites in Figs 4 and 5. As shown in Fig. 3, the East African Rift carbonatites with negative ϵ_{Nd} are unique in requiring only two mantle end-member components to account for most, perhaps all, of their Nd,

Sr, and Pb isotope compositions. All Pakistan 30 Ma carbonatites share the same unique character when the Loe Shilman $^{206}\text{Pb}/^{204}\text{Pb}$ ratios are corrected to 19. That similarity is especially interesting given that the Indian plate was attached to the present northeast Africa before the break-up of Gondwanaland, and was probably once part of the Neoproterozoic Pan African craton (Bender, 1995). Some evidence for that is the 0.5–0.7 Ga Sm/Nd model ages of 30–50-my-old granites in the Kohistan batholith in northern Pakistan (George *et al.*, 1993). It is thus possible that the lithosphere beneath the Pakistan and Pan African crusts has been rather similar from a geochemical standpoint over at least the last 130 my.

However, the two groups are at present situated in very different tectonic settings: the African plutons in a rift zone and the Pakistan plutons in a collision zone. According to Le Bas *et al.* (1987), the Pakistan plutons are emplaced along thrust faults rather than rifts. It is also significant that the 30 Ma Pakistan carbonatites crop out in close proximity to the 300 Ma Koga and Jhambil carbonatites, whose data resemble those of the reference group three-component, Nd–Sr–Pb isotope patterns with positive ϵ_{Nd} and negative ϵ_{Sr} (compare Figs 3 and 4).

How do we account for those observations? The main question is the source of the negative ϵ_{Nd} values, which are generally associated with aged sialic crustal rocks. However, the close fit of the carbonatite data to the mantle plane in Fig. 6 indicates that the negative values seem to involve mantle rather than crustal sources. If there are crustal contributions to the magmas, those presumably come from rocks that were first subducted into the mantle. The EM1 component dominates the isotope patterns for those plutons with negative ϵ_{Nd} (Figs 2–4), raising the question of its source, which is generally thought to be in the lithosphere [reviewed by Hart (1988)]. Bell & Simonetti (1996) used that model in their detailed study of the petrogenesis of the Oldoinyo Lengai natrocarbonatites. They proposed that the pluton is emplaced along the margin of the Kenya Dome, where HIMU-like plume material penetrated and metasomatized an EM1-like old sub-crustal lithosphere. Kalt *et al.* (1997) emphasized the large variations in isotope ratios between closely situated carbonatites and argued for highly heterogeneous lithospheric sources.

With such models the question remains as to why the East African and Pakistan carbonatites differ so greatly from many other carbonatites around the world. The young Oka, Arkansas, and Kaiserstuhl carbonatites, all emplaced through continental crust, clearly in rift systems at Oka and Kaiserstuhl, have strongly positive values for ϵ_{Nd} coupled with negative values for ϵ_{Sr} [see the summary by Bell & Blenkinsop (1989)] and isotope patterns requiring substantial contributions from all three components, DMM, HIMU and EM1. In addition, older, Precambrian, carbonatites around the world generally

yield similar patterns with positive ϵ_{Nd} and negative ϵ_{Sr} , as first demonstrated by Bell *et al.* (1982) for large areas of Canada. Apparently, formation in, or intrusion through, lithosphere underlying old (Precambrian) crustal rocks is often insufficient in itself to yield negative values for ϵ_{Nd} in carbonatites.

Recent observations from mantle tomography provide another possible reason for the unique African Rift and Pakistan data. As a 'speculative alternative' to other models, Hart (1988) proposed that EM1 and HIMU might be stored in the core–mantle boundary layer. Although Hart envisioned whole-mantle circulation with downwelling at the poles and upwelling at the equator, recent data from tomography indicate that major upwelling from the core–mantle boundary occurs in just two places, one underneath the East African Rift zone and the other underneath the southeast Pacific Ocean (Grand, 1994; Van der Hilst *et al.*, 1997). The upwelling is indicated in both P and S waves, and seems well established. The East African Rift 'plume' is especially prominent.

Is there a connection between the unusual isotope patterns in the East African carbonatites and the unique deep mantle upwelling zone beneath the rift? Combining the tomography with the Hart (1988) model, we can envision the EM1 component along the African Rift being added to the lithospheric mantle from the core–mantle boundary by a deep-seated upwelling mass. This highly speculative model would explain why the negative ϵ_{Nd} and prominent EM1–HIMU mantle-component anomalies occur along the East African Rift. It also raises the issue of whether rising material from the deep mantle must always have a very positive ϵ_{Nd} , as is generally assumed in oceanic hot-spot magmas (e.g. Zindler *et al.*, 1982, and references therein; Hoernle & Tilton, 1991). In any case, with present data it seems necessary to relate the unusual isotope pattern of the African Rift carbonatites in some way to the presence of the rift.

How does that relate to the Pakistan plutons? Although the Indian plate, including Pakistan, was once part of present-day northeast Africa, the 30-my-old plutons are clearly not part of the present rift system. For the mantle upwelling model it is appropriate to link the EM1 anomalies to underlying lithosphere that already contained material from the mantle–core boundary at the time of separation of India and Africa, which was then transported with the plate from Africa to the present location. The model requires that neither the transport of the land mass nor the continental collision process, which started ~50 my ago (Bender, 1995), seriously disturbed the unique isotope ratios in the lithospheric sources of the pluton magmas, except for contamination in some of the lead ratios in the collision. The validity of this model obviously depends critically upon several factors, including better knowledge of the isotope characteristics of

the lithospheric sources underlying Pakistan and north-west India.

Comparison between the 300- and 30-my-old Pakistan carbonatites indicates that the conditions that led to the anomalous features in the 30-my-old carbonatites did not exist 300 my ago in the future Pakistan terrain, and that the Koga and Jhambil plutons were emplaced under tectonic settings that must have differed greatly from present-day conditions. The African plate would have occupied a very different global position, and the Rift probably did not exist at that time. The 300-my-old plutons must have survived the transport and collision processes that affected the Indian plate without losing their isotopic or compositional identity.

CONCLUSIONS

Our results show that Nd, Sr and Pb isotopes in two Pakistan carbonatites that were emplaced along thrust planes in the Himalayan collision zone ~30 my ago are unique compared with most other young (<130-my-old) carbonatites in the world in having negative values for ϵ_{Nd} , and correspondingly higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. That is the opposite of the usual pattern for most intra-plate carbonatites, except for many of the East African Rift plutons with ages ranging from 0 to 130 Ma. Many of the African Rift and the young Pakistan carbonatites are also unique in having Nd, Sr and Pb isotope patterns that fit two-component mantle sources involving only the end members HIMU and EM1, when corrections are made for Pb contamination in some of the Pakistan plutons. However, the low abundances of Nd, Sr and Pb in DMM relative to EM1 and HIMU allow substantial contributions to the magma sources from DMM. Nearly all of the remaining young carbonatites from around the world require three mantle end-member components—EM1, HIMU and DMM—to explain their isotope ratios.

We note that there may be a connection between the rather unusual Nd, Sr and Pb isotope patterns in the African Rift carbonatites and the major upwelling of material from the core–mantle boundary detected by tomography. As the young Pakistan carbonatites crop out in an area that was once part of northeastern Africa and they share some of the anomalies present in the East African plutons, transport from northeast Africa to the Himalayan belt did not erase the unique isotope patterns in their lithospheric magma sources. The collision process does not appear to have affected the isotope ratios in any of the Pakistan carbonatites, except for Pb in the 30-my-old plutons.

The 300-my-old Pakistan carbonatites, like most intra-plate plutons, require substantial proportions of all three mantle end-member components to explain their isotope patterns. They appear to have formed before the rift was

established, and to contain no input from the unique lithospheric source that now underlies it. The transport of the Indian plate from Africa to its present position and collision with Asia did not noticeably alter their Nd, Sr and Pb isotope ratios.

ACKNOWLEDGEMENTS

We thank Michael Roden, Khalil Spencer and Anthony Simonetti for constructive reviews, which have led to a number of improvements in the manuscript. Additional comments from Keith Bell and Bruce Kjarsgaard are much appreciated.

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