

# Depleted and enriched mantle sources for Paleo- and Neoproterozoic carbonatites of southern India: Sr, Nd, C–O isotopic and geochemical constraints

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## Abstract

Paleoproterozoic (Hogenakal) and Neoproterozoic (Samalpatti, Sevattur, Mulakkadu–Pakkanadu) carbonatites of Tamil Nadu, southern India, have been investigated for whole-rock geochemistry and Nd, Sr and C–O isotopes. These temporally distinct carbonatite complexes are located close to a tectonically active zone that marks the transition between cratonic non-chaonkitic (low- to medium-grade) terrain to the north and the chaonkitic mobile belt (granulite facies) to the south. The carbonatites are variably enriched in LREE; the Hogenakal carbonatites being extremely enriched, with the highest  $\Sigma$ REE among the data while the younger carbonatites show variable enrichment levels and broadly comparable REE patterns. The Hogenakal carbonatites have coherent and typically mantle C- and O-isotopic ratios ( $\delta^{13}\text{C}_{\text{V-PDB}} \sim -6\text{‰}$  and  $\delta^{18}\text{O}_{\text{V-SMOW}} \sim 8\text{‰}$ ). The Neoproterozoic carbonatites are relatively enriched in  $^{13}\text{C}$  ( $\delta^{13}\text{C}_{\text{V-PDB}} = -5.3\text{‰}$  to  $-3.3\text{‰}$ ) and also show a range of O-isotopic composition ( $\delta^{18}\text{O}_{\text{V-SMOW}} = 7.3\text{‰}$  to  $15.4\text{‰}$ ; two samples  $\sim 25\text{‰}$ ). Higher  $\delta^{18}\text{O}$  values indicate variable degree of post-magmatic low-temperature alteration. The older carbonatites have marginally positive  $\epsilon_{\text{Nd}}$  (+0.54 and +1) and significantly low initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.70161 and 0.70174) while younger carbonatites have rather low  $\epsilon_{\text{Nd}}$  (–16.5 to –6.23) and higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.70486–0.70658). The Sr–Nd isotopic data are compatible with a depleted mantle source for the Hogenakal carbonatites and an EM-I-type enriched mantle component for the younger group. This is the first report of the existence of depleted mantle beneath the southern Indian continental crust. The stable isotopic ratios are interpreted as suggesting a depletion event (crustal extraction) in the south Indian subcontinental mantle  $\sim 2.6$  Ga ago. The depleted mantle was subsequently enriched by metasomatic fluids under the influence of the subducting Dharwar plate (sediments and modified oceanic crust).

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## 1. Introduction

A mantle origin for carbonatites is now unequivocal, although the debate still continues whether the

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carbonatite melts are generated within the lithospheric or asthenospheric mantle. A broad consensus, postulating the genesis of carbonatites through the interaction of asthenospheric and lithospheric mantle components, seems to be emerging (Bell et al., 1998 and references therein). Carbonatites have been proposed to be derived from primary magma (Bailey, 1993), by differentiation from a silicate parent (Kjarsgaard, 1997) or separation from a silicate parent by liquid immiscibility (Brooker, 1998). Nevertheless, the isotopic data provide conclusive evidence for their mantle origin. Notwithstanding the ongoing controversy on the source region within the mantle, the carbonatites are particularly useful in deciphering the nature of the upper mantle as their low viscosity facilitates them to sample a potentially large volume of the mantle and their high Sr and Nd abundances (far exceeding the crustal levels) further buffer them against any possible effects of crustal contamination (Bell et al., 1982; Bell and Blenkinsop, 1987a; Tilton and Bell, 1994). Although carbonatites are typically associated with alkaline silicate rocks and preferentially emplaced through continental rifting, the carbonatitic melts are presumed to remain unaffected by crustal contamination because of their low temperatures (600–770 °C, see Secher and Larson, 1980) thus making them suitable candidates to investigate the subcontinental upper mantle. Temporally widespread carbonatite magmatism within a tight geographic terrane would further provide valuable information on mantle evolution (including recycling of the crustal material) and the mantle heterogeneity through time. Secular Nd, Sr and Pb isotopic variations in the 2.7- to 0.11-Ga carbonatites of Canadian Shield have been utilized to model the evolution of a long-lived depleted mantle (Bell et al., 1982; Bell and Blenkinsop, 1987a) and to monitor the terrestrial differentiation of the carbonatite melts (Tilton and Bell, 1994).

The subcontinental upper mantle in the south India has been described as one enriched in alkali metals and LREE, based on the Sr–Nd isotopic data on carbonatites and associated alkaline silicate rocks (Kumar et al., 1998; Schleicher et al., 1998; Miyazaki et al., 2000). Kumar et al. (1998) argue for a long-lived ‘enriched’ lithospheric mantle that remained a ‘closed system’ from 2.6–2.5 Ga to 800–750 Ma and survived convective disruption. Based on the differ-

ences in the isotopic signatures between coexisting Neoproterozoic carbonatites and syenites, Miyazaki et al. (2000, 2001a,b) have envisaged isotopically different ‘enriched’ mantle sources for them, supposedly indicating mantle heterogeneity. Schleicher et al. (1998) and Schleicher (2001) also corroborate an ‘enriched’ mantle component for Neoproterozoic carbonatites; however, they have postulated mixing between an upper mantle reservoir and either an EM-II-like mantle or a crustal component. This communication presents the results of parallel studies carried out on these Paleoproterozoic and Neoproterozoic carbonatites, located within a close tectonic/geographic terrane in southern India. Our isotopic data substantiate the existence of an ‘enriched’ source for Neoproterozoic carbonatites, in agreement with the existing models. However, our identification of an older ‘depleted’ mantle (not reported so far from this region) significantly differs from published results (Kumar et al., 1998). We have attempted to evaluate the evolution of the mantle beneath the southern Indian block in terms of long-lived mantle heterogeneity during the Archaean–Proterozoic or possible metasomatic enrichment, subsequent to the extraction of the Archaean crust.

## 2. Background geology

The carbonatite complexes of southern India are emplaced within the Precambrian granulite terrain along the NE-trending fault system (Subramanian et al., 1978; Subramanian, 1983; Viladkar and Subramanian, 1995). These carbonatite–alkaline complexes are believed to be associated with the Precambrian magmatism within the Eastern Ghat Mobile Belt (EGMB) of eastern and southern India (Schleicher et al., 1998). The studied carbonatite complexes (Hogenakal, Samalaptti, Sevattur and Pakkanadu–Mulakkadu) are located along the SW margin of the EGMB (Fig. 1) within a tectonic zone of intense faulting and thrusting that marks the transition between cratonic non-charnockitic (amphibolite facies) terrain in the north and the charnockitic mobile belt (granulite facies) in the south (Condie et al., 1982; Condie and Allen, 1984; Swaminath, 1974). The Hogenakal carbonatites are Early Proterozoic in age while all other carbonatites were emplaced during the Neoprotero-

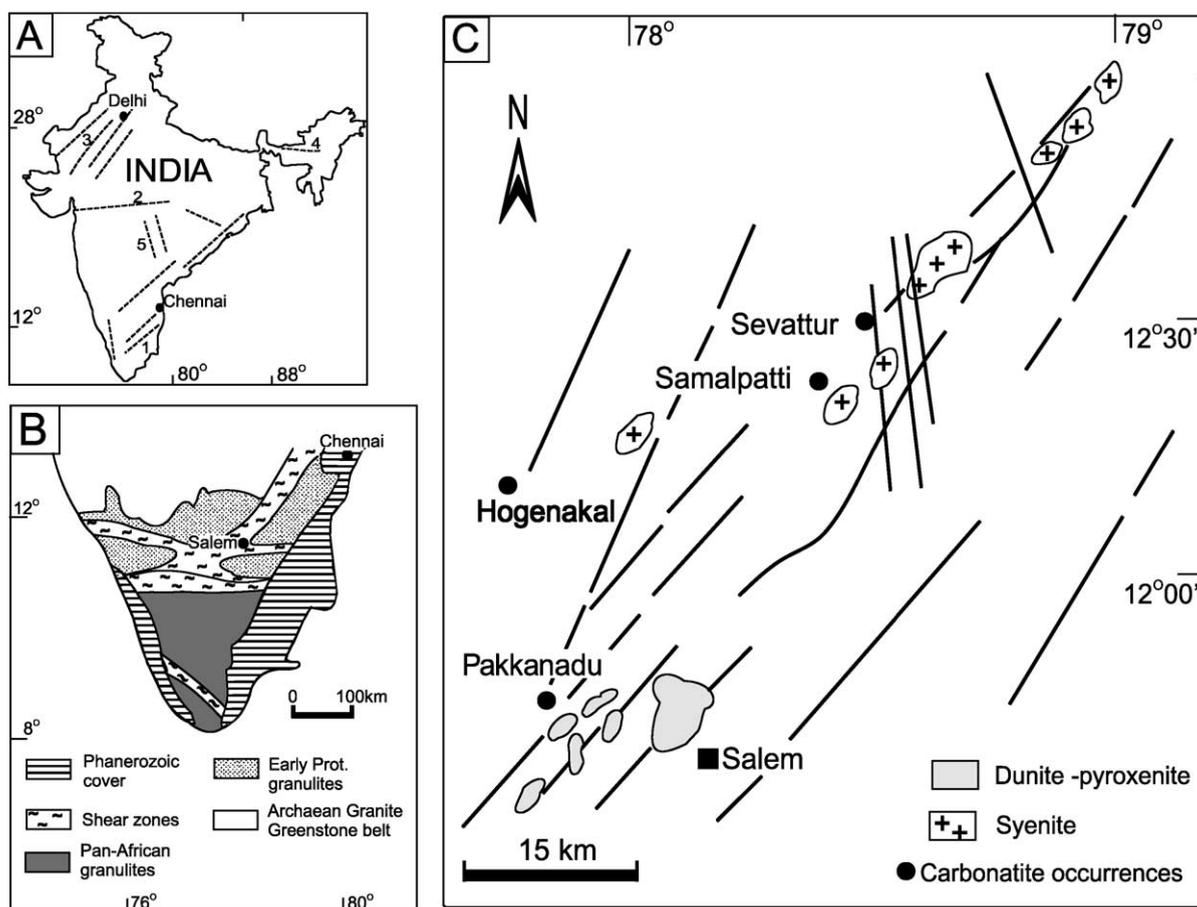


Fig. 1. (A) Carbonatite-related tectonic features in India. 1. Eastern Ghat Belt, 2. Narmada–Son Rift, 3. Aravalli–Delhi Belt, 4. Assam–Meghalaya Belt and 5. Cuddapah–Godavari Rift Zone (modified after Krishnamurthy, 1988). (B) Granulite blocks and shear zones in southern India (compiled from Wickham et al., 1994; Harris et al., 1994). (C) Geological sketch map of Koratti Fault Zone in south India showing carbonatite occurrences.

zoic. The carbonatite emplacement has been guided and controlled by a set of NNE to NE-trending fault system (Grady, 1971).

The Hogenakal carbonatites occur as a series of discontinuous bodies within two sub-parallel NNE-trending pyroxenite dykes that intrude the gneissic charnockite (with xenoliths of amphibolite and quartzite) country rocks. The western dyke coalesces with the eastern one in the south, the latter traceable over a distance of 14 km with further possibilities of southward extension (Ramakrishnan, 1973). Each carbonatite body consists of discrete veins and lenses and shows strong fenitization of the pyroxenite host. Pyroxenite and syenite xenoliths are also present

within the carbonatite. Details on the geological setting of Hogenakal carbonatite are provided by Natarajan et al. (1994) who also reported the whole rock-mineral Rb–Sr isochron age of 1.99 Ga for the Hogenakal carbonatites. However, the five-point Sm–Nd isochron for whole-rock carbonatite and nearby Pikkli Hill syenite gave an age of  $2401 \pm 25$  Ma and the seven-point (two carbonatites and five syenites) whole-rock Rb–Sr isochron corresponds to a precise age of  $2415 \pm 10$  Ma (Kumar et al., 1998). On the basis of a close agreement between two independently estimated age data, we believe the 2.41-Ga age for the Hogenakal carbonatite to be more realistic. The 2.00-Ga age reported by Natarajan et al. (1994) prob-

ably represents a thermal disturbance that has also been noticed by Kumar et al. (1998). As there are no evidence for any other significant metamorphic or igneous event in the Nilgiri–Madras granulite belt (Yoshida et al., 1996) between 2.4 Ga and the time of emplacement of Neoproterozoic carbonatite complexes, the 2.0-Ga thermal disturbance can be regarded of local consequence only. The charnockitic country rocks in the Hogenakal area have yielded U–Pb zircon age of 2.55–2.53 Ga (Peucat et al., 1993), believed to record the accretion and cooling of the juvenile crust. The granulite facies metamorphism in southern India appears to be concurrent with the crust formation and continued up to 2.52 Ga. The Hogenakal carbonatites were thus emplaced within a young crust that was not older than 150 Ma (Kumar et al., 1998).

The Sevattur–Samalpatti carbonatite complexes are located ENE of Hogenakal (Fig. 1). The Samalpatti complex includes carbonatite occurrences in nearby localities (Pallasulakkarai, Onnarakai, Garigapalli). The Sevattur carbonatite occurs as an inwardly dipping arcuate body sandwiched between a large syenitic stock and pyroxenite near village Sevattur (for details on geological setting, see Udas and Krishnamurthy, 1970; Krishnamurthy, 1988), as ring dykes and veins amidst syenites, pegmatites and aplites, as cavity fillings, breccia and diatremes within some contact metasomatic carbonaceous rocks and pyroxenites near village Onnarakai (Ramasamy, 1993). The carbonatites mainly occur as elongated bodies within pyroxenite and to a lesser extent, within syenites. Apophyses and satellite diatremes emanating from the main carbonatite body are also seen. The contact between carbonatite and syenite is marked by shears and fractures. Carbonatite injections have also resulted in brecciation of the gneissic country rocks. Deformed feldspar grains (derived from the country rocks) enclosed within carbonatite further suggest a forceful injection. The carbonatites also incorporate a number of xenoliths of basement gneisses, syenites and pyroxenites (Borodin et al., 1971; Krishnamurthy, 1977; Schleicher et al., 1998). The Sevattur–Samalpatti carbonatites intrude the 2.55- to 2.53-Ga-old (Peucat et al., 1993) granite gneiss terrain. The Samalpatti carbonatites are discordantly emplaced into the hornblende gneisses (Moralev et al., 1975; Subramanian et al., 1978). The alkaline rocks in this region form a central plutonic mass, surrounded by

discontinuous rings of pyroxenite and dunite. The carbonatites are poorly exposed and seen as isolated mounds and boulders as well as linear dykes, veins and lenses within pyroxenite and syenite. The geological relationships in Sevattur indicate that sövite was emplaced first, later veined by dolomitic carbonatite while the ankeritic phase appears to be the youngest (Udas and Krishnamurthy, 1970). The Pakkanadu–Mulakkadu carbonatites occur as small dimension, concordant fracture-fills and diatremes within the pyroxenites and as discordant lenses and dykes within syenite. Their emplacement appears to have been guided and controlled by the subsidiary of the NE-trending Attur Fault (Grady, 1971). Ribbon-like bodies of xenolithic feldspathic breccia are present within the carbonatites. These feldspathic breccias possibly represent the vent rocks that were later disrupted, blown and flushed out by carbonatitic melt, similar to those reported by Bailey (1961) from Zimbabwe. These were described as ‘late-phase hydrothermal carbonatites’ by Borodin et al. (1971) on the basis of the presence of some pegmatitic phase minerals. The studied carbonatites show evidence of variable deformation. The Pakkanadu carbonatites are significantly metamorphosed, as indicated by development of secondary monazite and macro-scale deformation (see Möller et al., 2001) while the Sevattur and Samalpatti carbonatites show the effects of tectonic stress as reflected in development of boudinaged structures, elongated and sheared xenoliths and deformed calcite twin lamellae. The Hogenakal carbonatites appear to be undeformed with well-preserved pristine magmatic texture and exhibit only stress banding in calcite twin lamellae.

Various mineral phases from the Sevattur–Samalpatti carbonatites and associated alkaline rocks have been dated by K–Ar method (biotite, phlogopite), Pb–Pb (pyrochlore), fission track (apatite, zircon) by Moralev et al. (1975), Nagpaul and Mehta (1975), and Parthasarthy and Sankar Das (1976). The reported ages range from 845 to 600 Ma and probably represent the cooling ages. The whole rock–mineral Rb–Sr age of 771 Ma, reported by Kumar and Gopalan (1991), appears to closely approximate the emplacement age. This is also in close agreement with the 800-Ma whole-rock Pb–Pb age reported by Schleicher et al. (1997) and also corresponds with the coeval events of magmatism in other parts of East Gondwana entity,

such as Madagascar, Seychelles and Malani Igneous Suite in NW India (Tucker et al., 1999; Torsvik et al., 2001a,b). The Sevattur and Yelagiri syenites have yielded an identical whole-rock Rb–Sr age of 757–756 Ma (Miyazaki et al., 2000). Möller et al. (2001) have dated the Pakkanadu monazite and obtained ages of  $750 \pm 2$  and  $759 \pm 3$  Ma, interpreted as the intrusion age. The geochronologic data are, thus in general agreement with a Neoproterozoic age of intrusion and correspond with the globally recognised coeval rifting events and alkaline magmatism within other continental blocks related to the break-up of Rodinia.

### 3. Petrography and mineralogy

The carbonatites are extremely variable in their textural attributes and also exhibit a wide range of mineral assemblages. The Hogenakal carbonatites are mainly sövitic and can further be classified into calcite–sövite, biotite–diopside–apatite sövite and agglomeratic sövite, on the basis of silicate minerals present. In the silicate–sövite varieties, biotite, diopside, augite and apatite are set in a mosaic of calcite crystals which mutually define a granular texture. Apatite grains are usually rounded with polished surfaces and in some cases constitute up to 10–15% of the mode. Biotite shows alteration into phlogopite which is observed as replacement rims along the margins of biotite laths. Anhydrous magnetite, present as clusters as well as occupying interstitial spaces

within calcite crystals and cleavage fractures within biotite and pyroxene, constitutes up to 10% of the rock. Aegirine and allanite are the accessory minerals present.

Mineralogical variants identified in the Sevattur carbonatites (from west to east) are, (a) biotite–oligoclase sövite, (b) phlogopite–sövite, (c) aegirine–sövite, (d) para-ankeritic sövite, (e) pyrochlore bearing beforosite and (f) apatite–magnetite sövite.

The Samalpatti carbonatites are mainly sövite and silicocarbonatite. Both varieties are holocrystalline, equigranular and medium- to coarse-grained. Calcite is the most prominent mineral with minor abundance of dolomite. The presence of grossularite, chlorite, scapolite, diopside and barkevikite in the silicocarbonatites indicates significant metamorphism the rocks have undergone. Main mineralogical variants in Onnakarai are riebeckite sövite, riebeckite ferroan sövite and ferroan carbonatitic breccia with abundant opaque minerals and ilmenite–rutile. Two main mineralogical variants have been identified in the Pakkanadu–Mulakkadu carbonatites:

- Highly contorted fine- to medium-grained biotite–phlogopite sövite with accessory magnetite and apatite.
- Magnetite and apatite-bearing ferroan carbonatite with abundant limonitic material.

The sövite shows medium- to coarse-grained granular texture with well-developed rhombic cleavages in

Table 1a  
Major element chemistry of south Indian carbonatites

Sample	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	CO <sub>2</sub>	H <sub>2</sub> O <sup>+</sup>	LOI	Total
H/46	0.20	0.03	0.35	Tr	0.30	Tr	1.20	54.28	0.14	0.11	0.11	42.57	0.50	–	99.79
H/56	7.91	0.07	1.92	0.51	1.20	Tr	2.51	47.10	0.30	0.47	3.17	32.31	0.45	0.50	98.42
SVT/4	1.41	0.01	1.49	1.90	–	0.20	2.95	50.73	0.31	0.21	2.50	36.20	0.30	1.89	100.10
SVT/5	2.00	1.00	4.95	10.30	4.20	0.25	2.38	41.50	0.62	0.33	5.29	24.95	0.28	1.95	100.00
Palla/4	25.93	1.13	5.20	9.56	3.00	2.37	7.58	17.99	3.45	3.09	1.20	18.50	–	1.20	100.20
BG/17	22.90	1.60	3.82	5.12	0.90	Tr	4.20	35.21	1.40	1.92	1.00	22.00	–	0.21	100.28
OK/8	23.50	1.50	4.60	8.91	2.50	1.55	8.95	20.97	2.92	2.20	1.20	21.00	–	0.66	100.44
OK/27	25.97	1.30	4.60	9.71	2.97	1.97	7.12	18.00	3.20	3.10	1.10	18.40	–	1.10	98.54
PK/3	7.22	0.60	1.95	3.20	6.28	0.80	6.96	35.37	0.50	0.95	0.85	33.35	1.00	0.75	99.78
PK/8	6.72	0.55	1.88	2.90	6.20	0.60	6.33	35.12	0.52	1.00	0.90	34.56	0.77	1.50	99.55
MK/28	1.50	0.05	1.10	2.30	2.50	0.25	2.68	51.25	0.17	0.12	4.90	32.34	0.30	0.97	100.43

Tr=traces.

Prefix to the sample number indicates the locality: H=Hogenakal; SVT=Sevattur; Palla=Pallasulakkarai (7 km SW of Samalpatti); BG=Garigapalli (5 km SW of Samalpatti); OK=Onnakarai (6 km SW of Samalpatti); PK=Pakkanadu; MK=Mulakkadu.

the calcite. Most of the pyroxenes are altered to the extent that their optical characteristics have been completely obscured. Some of the less altered grains have been identified as augite to ferro-augite. Apatite, titanite and magnetite are the common accessory minerals. The ferroan carbonatites mainly comprise of calcite, magnetite, apatite, sphene and monazite and accessory abundances of ankerite, biotite, phlogopite, and allanite.

#### 4. Major and trace element chemistry

Major element analysis was carried out by wet-chemical method and spectrophotometrically, follow-

ing the schemes of Groves (1951) and Shapiro and Brannock (1962) with suitable modifications. TriPLICATE analyses of each sample were carried out and the average of the three has been used. The variations in the analytical results were generally of the order of 5–7%. FeO was determined gravimetrically by titrating the solution against ferrous ammonium sulphate using *N*-phenylanthranilic acid as the indicator. Alkalies were determined on an EEL Flame Photometer.  $\text{H}_2\text{O}^-$  was determined by heating the sample at 105 °C for 8–10 h and  $\text{H}_2\text{O}^+$  and  $\text{CO}_2$  were determined on the pre-heated (105 °C) samples. Trace and rare earth elements were determined mass spectrometrically at National Geophysical Research Lab., Hyderabad, India, on ICP-MS (Plasma Quad PQ-1),

Table 1b  
Trace and rare earth elements analyses (ppm) of south Indian carbonatites

	H/46	MK/28	BG/17	PK/8	SVT/4	Palla/4	OK/8	OK/27	PK/15
Sc	11.71	21.77	2.31	7.41	23.77	15.47	4.53	62.95	69.40
V	42.79	35.48	47.58	22.8	72.83	39.99	45.33	285.39	181.16
Cr	19.22	12.35	38.97	11.32	78.12	21.44	14.01	97.89	48.69
Co	5.85	4.51	13.06	0.37	18.09	13.78	5.84	65.31	71.03
Ni	20.19	21.22	21.30	3.08	22.05	14.75	4.20	76.21	74.60
Cu	37.25	38.74	33.16	99.21	83.09	92.75	104.4	676.19	57.20
Zn	43.23	223.64	62.07	4066	87.63	399.5	215.1	520.81	50.2
Ga	70.63	30.3	40.13	16.83	21.75	11.08	6.71	7.15	11.82
Rb	13.49	0.33	2.56	2.72	8.98	1.65	111.4	26.75	3.86
Sr	12253	15678	6176	5545	7454	6316	257.2	1337	2829
Y	202.27	125.22	187.16	34.03	88.13	54.57	8.78	60.8	37.18
Zr	16.06	0.65	10.3	0.47	19.23	2.65	56.49	32.29	8.92
Nb	3.44	1.54	5.57	5.24	50.03	33.19	47.90	92.49	0.50
Cs	0.17	0.00	0.09	0.12	0.69	0.06	0.61	0.09	0.03
Ba	517	4354	766.61	13559	1901	8816	7272	5761	109.62
Hf	0.92	0.17	0.71	0.10	0.64	0.29	2.76	0.88	0.50
Ta	0.21	0.15	0.13	0.35	4.54	1.82	0.34	0.10	0.01
Pb	2.88	16.15	Tr	32.47	Tr	Tr	4.39	Tr	Tr
Th	42.44	2.05	12.94	33.79	9.75	2.67	5.91	12.47	4.00
U	2.08	0.15	2.88	0.38	8.62	6.03	0.66	0.48	0.37
La	1871	875	345.9	1024	545	57.57	32.1	30.62	164.3
Ce	3375	1324	1316	1226	942	178.0	69.5	89.56	290.4
Pr	415	114	234.4	119.5	109	29.33	9.2	14.75	37.26
Nd	1802	570	1241	438.3	446.9	160.3	44.1	83.06	173.4
Sm	290	85.46	268.8	55.36	73.84	40.7	8.55	22.2	30.43
Eu	60.47	19.49	59.62	47.25	16.92	13.85	4.23	7.77	7.08
Gd	188.3	65.27	155.7	39.16	53.99	28.46	9.71	16.71	21.16
Tb	16.97	6.01	15.97	2.73	5.19	3.46	0.61	2.39	2.04
Dy	72.27	29.43	71.85	11.00	26.22	16.82	3.68	13.78	10.59
Ho	8.24	4.07	9.19	1.21	3.48	2.32	0.54	2.07	1.5
Er	34.14	21.86	41.63	6.98	15.97	10.54	2.60	10.61	6.53
Tm	1.68	1.44	2.34	0.43	1.00	0.65	0.21	0.71	0.41
Yb	9.00	9.33	11.74	2.91	5.87	3.97	1.18	1.03	2.25
Lu	1.25	1.41	1.42	0.49	0.76	0.62	0.14	0.56	0.35

following the procedure outlined in Balaram et al. (1996). The instrument was calibrated against international standards such as CCRMP, IGGE, AC-E, JB-2 and GSR-6. Reported analytical precision for trace and REE is >95%. The whole-rock major and trace element compositions are presented in Table 1a and b.

The carbonatites are quite variable in their geochemical characteristics which reflects the wide range of silicate minerals present. Silica content ranges from 0.20% to 25.97% with an average of 12.87%. Sövitic carbonatites have  $\text{CaO} \geq 50\%$  while other carbonatites have MgO and  $\text{FeO}^{\text{I}}$  contents up to 9% and 14%, respectively. All the geochemical variations are within the prescribed ranges for carbonatite (Gold, 1966). In the  $\text{CaO-MgO-FeO+Fe}_2\text{O}_3+\text{MnO}$  classification scheme (Woolley and Kempe, 1989), the carbonatites plot along an array straddling across the calcio-carbonatite and ferrocarbonatite fields (Fig. 2). However, using the molar proportions instead of absolute abundance, and excluding  $\text{Fe}_2\text{O}_3$  (for not being accommodated in the carbonatite phase), as recommended by Gittins and Harmer (1997), the earlier classified ferrocarbonatites plot in the magnesian carbonatite field while the status of calcio-carbonatites remains unchanged.

Trace elements provide useful information to decipher the magma source and to model the evolutionary process of silicate rocks. In carbonatite systems, these are of little help where traditionally ‘incompatible’ elements become frequently ‘compatible’ and field relations show clear evidence of accumulation, veining and layering (Dunworth and Bell, 2001). Very high abundances of Ba and Sr and  $\text{Sr/Ba} > 1$  are characteristic of carbonatites (Keller and Spettel, 1995). The Sr and Ba enrichment levels of the carbonatites in this study are the highest among all other known carbonatite complexes of India (Krishnamurthy, 1988) and the Sr/Ba ratio ranges from  $< 1$  (ferrocarbonatites) to  $> 1$  (calcio-carbonatites). Low to moderate abundances of compatible elements like Ni, Cr, Cs and V indicate some degree of fractionation of the melts before crystallization. Nb and Ta behave as a conjugate geochemical pair in most silicate igneous rocks; however, a decoupling between the two in carbonatites has been considered a result of immiscibility where Nb shows a preference for the silicate melt (Veksler et al., 1998). The mantle normalized  $(\text{Nb/Ta})_{\text{MN}}$  ratios in the present case are high (0.73–16.5), mostly  $> 1$  except two samples  $\sim 0.75$ . A strong enrichment in Nb argues against the origin of carbonatites by liquid immiscibility (Veksler et al., 1998).

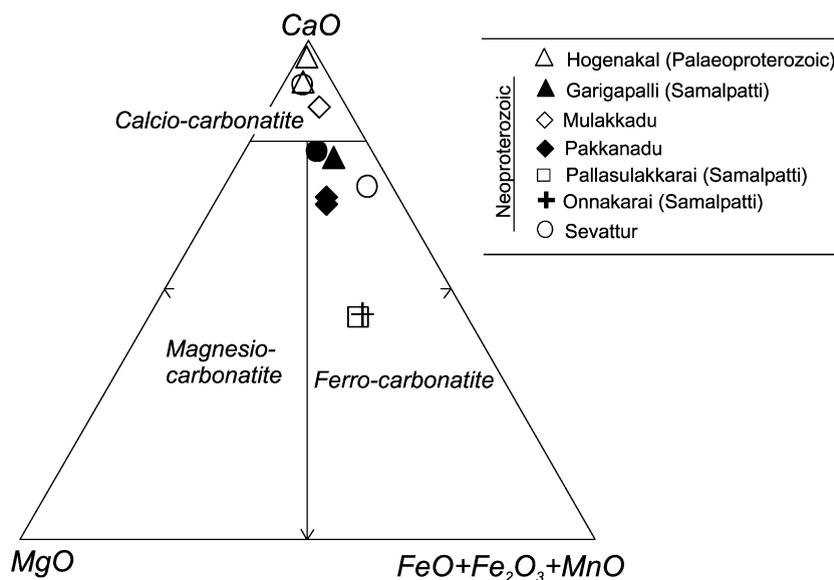


Fig. 2.  $\text{CaO-MgO-FeO+Fe}_2\text{O}_3+\text{MnO}$  diagram showing discrimination of south Indian carbonatites into calcio- and ferro-carbonatite fields (Woolley and Kempe, 1989).

The Zr enrichment (relative to Hf) indicates carbonate metasomatism in the mantle source (Rudnick et al., 1993). In the present case, the  $(Zr/Hf)_{MN}$  ratios are variable (0.12–5.67) and hence inconclusive. The trace elements substitute major elements (Mg, Ti etc.) in crystal lattices, hence their concentration is controlled, to a large extent, by the abundance level of conjugate major elements. The incompatible trace element patterns for Hogenakal carbonatite in the mantle-normalized spidergraph (Fig. 3) show several depletions and enrichments, coherent with normal carbonatite trends (Woolley and Kempe, 1989). Distinct negative anomalies are shown by Rb, Nb, Zr and Yb. A minor negative anomaly for Sr, despite

extremely high abundance is only the reflection of the much higher enrichment in its neighbouring elements (Ce and Nd). Enrichment in Th relative to Rb is a common feature of carbonatites as they normally have much higher Th abundance as compared to other igneous rocks. A high Th/U ratio is normally indicative of lithospheric mantle that resulted from time-integrated Th enrichment by metasomatism (Sun and McDonough, 1989; Williams et al., 1992; Carlson and Irwing, 1994). However, in carbonatites, the ratio may not be truly reflective of the mantle and may result from differences in the degree of compatibility between the two elements. The trace element behaviour for the Neoproterozoic carbonatites shows extreme depletion in K and Ti and negative anomalies for Rb and Zr (Fig. 4). The behaviour of Nb is quite variable as it shows moderate enrichment as well as depletion. This can be attributed to the incoherent abundance of pyrochlore. The behaviour of trace elements (relative enrichment and depletion) can be attributed to significant differences in the type and modal proportion of different accessory mineral species like pyrochlore, monazite and apatite. Except for the differences in the enrichment levels, the trace element patterns are by and large consistent and comparable with those of Hogenakal carbonatites.

Carbonatites are characterized by extremely high  $\Sigma$ REE and also have the highest LREE/HREE ratio among all the igneous rocks (Hornig-Kjarsgaard, 1998). The  $\Sigma$ REE and trends are mainly controlled by the presence and relative abundance of accessory minerals, capable of concentrating the REEs (especially LREEs). The Hogenakal carbonatites have the highest  $\Sigma$ REE with extreme enrichment in La (>5000 times chondrite) and define a straight-line log-linear pattern (Fig. 3). The chondrite-normalized patterns (Fig. 5) for the younger carbonatites define two distinct patterns. Most of the data define nearly straight log-linear patterns with strong and steady increase in enrichment from Lu to La and very high  $(La/Yb)_N$  ratio (Mulakkadu, Sevattur and Pakkanadu carbonatites). The Samalpatti carbonatites (Palla/4, OK/8, OK/27 and BG/17) are convex in the LREE region ( $La_N < Ce_N < Pr_N \leq Nd_N > Sm_N$ ) and show a steady decrease from Sm to Lu, moderate to high  $(La/Yb)_N$  and  $\Sigma$ REE. However, the LREE enrichment levels for Pallasulakkarai and Onnakarai ferro-

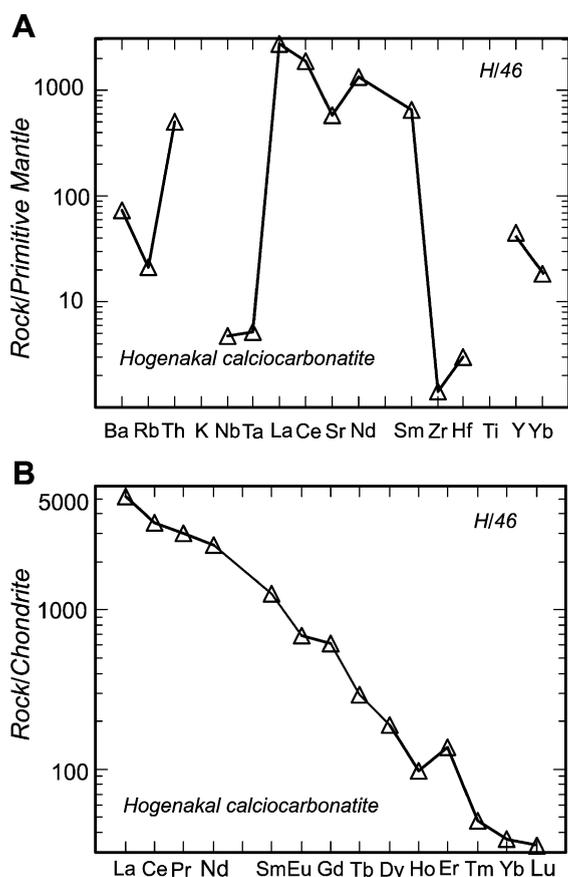


Fig. 3. (A) Primitive mantle (Sun and McDonough, 1989) normalized trace element spidergraph for Hogenakal carbonatite. (B) Chondrite normalized (Sun and McDonough, 1989) REE pattern for Hogenakal carbonatite showing straight line log-linear trend and extreme LREE enrichment.

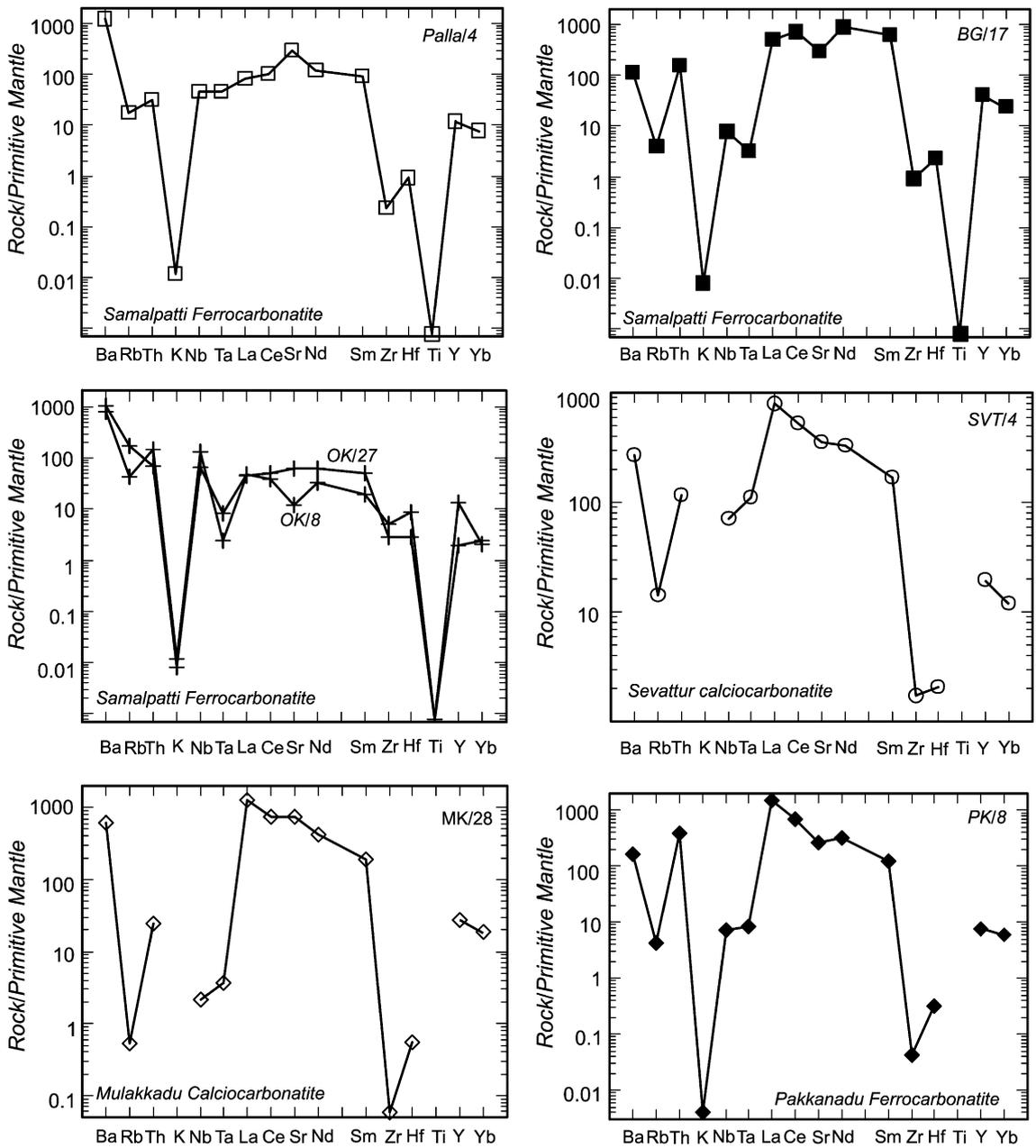


Fig. 4. Primitive mantle (Sun and McDonough, 1989) normalized trace element spidergraphs for south Indian Neoproterozoic carbonatites. Note significant depletions in Rb, K, Ti and Zr.

carbonatites are rather low relative to other samples. This can either be due to the absence of REE-rich accessory phases or LREE mobilization during metamorphic alteration. The Onnakarai samples also show anomalous C- and O-isotopic ratios (discussed

in next section). In the present case, the REE behaviour appears to have mainly been controlled by apatite. Pyrochlore and perovskite (REE-rich accessories) do not seem to have played any role as they can influence the bulk rock REE patterns

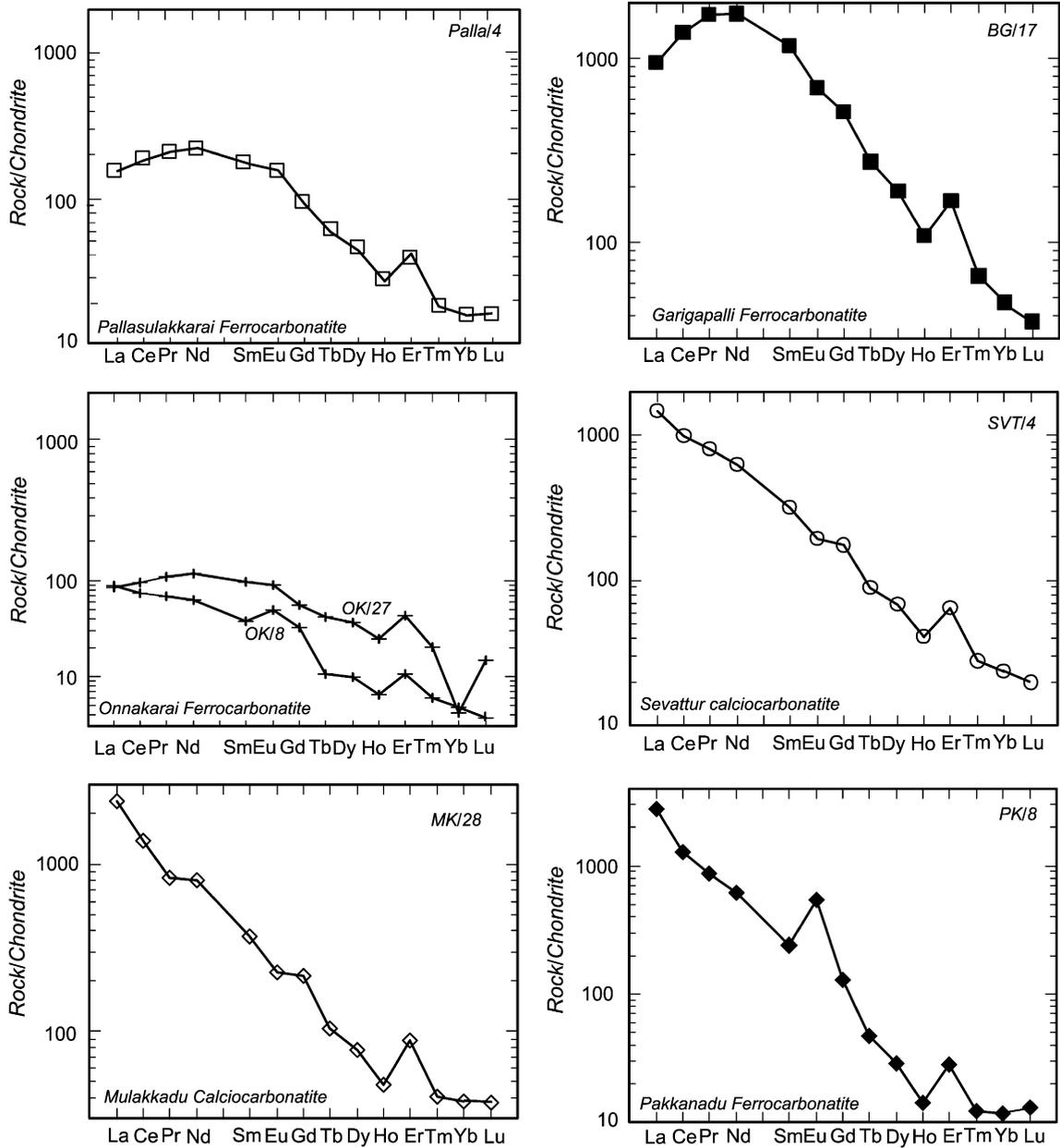


Fig. 5. Chondrite normalized (Sun and McDonough, 1989) REE patterns for Neoproterozoic carbonatite of southern India showing straight line and LREE convex patterns, and variable enrichment levels.

only if present in significant amount (Hornig-Kjarsgaard, 1998). All the samples exhibit significant positive Er anomaly. It appears that this is an analytical problem and does not have any petrogenetic implications. Differences in the REE patterns

(log-linear and convex) can be attributed to the compositional variation in the apatite. Broadly similar REE patterns for carbonatites with variable modal proportions of mineral phases can be associated with common parent magma.

## 5. C–O isotopes

Carbon and oxygen isotopic compositions were determined at NEG-LABISE, Federal University of Pernambuco, Brazil, using the conventional digestion method (McCrea, 1950). Powdered samples were reacted with  $\text{H}_3\text{PO}_4$  at 25 °C to release the  $\text{CO}_2$ . An extended reaction period was preferred for the ferrocarbonatites instead of increasing the reaction temperature. The  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values were measured on cryogenically cleaned  $\text{CO}_2$  (Craig, 1957) in a triple collector SIRA II mass spectrometer. The C and O isotopic data for the carbonatites are presented (as ‰ deviation with reference to V-PDB and V-SMOW, respectively) in Table 2. Borborema Skarn Calcite (BSC), calibrated against international standards, was used as the reference gas and reproducibility of the measurements was better than  $\pm 0.1$  ‰, in general. The values obtained for the standard NBS-20 in a separate run against BSC yielded  $\delta^{13}\text{C}_{\text{V-PDB}} = -1.05$  ‰, and  $\delta^{18}\text{O}_{\text{V-PDB}} = -4.22$  ‰. These results are in close agreement with the values reported by the US National Bureau of Standards ( $-1.06$  ‰ and  $-4.14$  ‰, respectively).

The  $\delta^{13}\text{C}_{\text{V-PDB}}$  in primary igneous carbonatites ranges between  $-4$  ‰ and  $-8$  ‰, and  $\delta^{18}\text{O}_{\text{V-SMOW}}$  between  $+6$  ‰ and  $+10$  ‰ (Taylor et al., 1967). Based on the data from fresh and unaltered natro-carbona-

tites from Oldoinyo Lengai, Keller and Hoefs (1995) proposed a much narrower range of C and O isotopic compositions ( $-7$  ‰ to  $-5$  ‰ and  $+5.5$  ‰ to  $+7$  ‰, respectively), which can be considered to represent undegassed and uncontaminated (by recycled carbon) mantle compositions. However, a number of carbonatites are known to be more enriched ( $\delta^{13}\text{C} > -4$  ‰, see Deines, 1989; Plyusnin, 1980; Dunworth and Bell, 2001). The isotopic differences within the ‘mantle compositions’ can be ascribed to mantle heterogeneity (Deines, 1989). ‘Primary’, as defined by Taylor et al (1967) denotes the carbonatites unmodified by secondary (crustal) processes and does not imply an unmodified partial mantle melt. The  $\delta^{13}\text{C}$  values in south Indian carbonatites range between  $-6.4$  ‰ and  $-3.3$  ‰ (Table 2, Fig. 6). The Hogenakal carbonatites with uniform C isotopic compositions ( $\delta^{13}\text{C} = -6.2$  ‰ to  $-6.0$  ‰, average =  $-6.1$  ‰), in perfect agreement with the calcite separate compositions ( $-6.3$  ‰ to  $-6$  ‰, average =  $-6.1$  ‰) reported by Kumar et al. (1998), define a more coherent and homogeneous group among the total data. The  $\delta^{18}\text{O}$  values for Hogenakal carbonatites are also uniform ( $+8.1$  ‰ to  $+8.4$  ‰, average =  $+8.2$  ‰) and in agreement with the calcite separate compositions ( $+7$  to  $+8.4$ , average =  $+7.8$  ‰), reported by Kumar et al. (1998). Carbonatite parent magmas are believed to be  $\sim 2$  ‰ enriched in  $^{18}\text{O}$  compared to their mantle source (Deines, 1989). A marginal enrichment in  $^{18}\text{O}$ , therefore, is more likely an inherited mantle source characteristic and not a post-magmatic alteration phenomenon. The Neoproterozoic carbonatites are relatively enriched in  $^{13}\text{C}$  and range in  $\delta^{13}\text{C}$  from  $-5.3$  ‰ to  $-3.3$  ‰, except one sample ( $-6.4$  ‰). The isotopic compositions for a single locality are, however, closely similar. Our C-isotopic results for Neoproterozoic carbonatites are also comparable with those of Kumar et al. (1998) for Sevattur calcite separates ( $-6.2$  ‰ to  $-4.8$  ‰; average =  $-5.3$  ‰) and Schleicher et al. (1998) for whole-rock carbonatites ( $-7.6$  ‰ to  $-4.9$  ‰; average =  $-5.6$  ‰). However, our Mulakkadu and Onnakarai samples are significantly enriched in  $^{13}\text{C}$  (average  $\delta^{13}\text{C} = -3.7$  ‰, range  $-3.9$  ‰ to  $-3.3$  ‰) and statistically distinguished from other Neoproterozoic carbonatites (Sevattur, Pakkanadu, Pallasulakkarai, Garigapalli), which are relatively depleted in  $^{13}\text{C}$  (average  $\delta^{13}\text{C} = -5.0$  ‰, range  $-4$  ‰ to  $-6.4$  ‰). Most of the Neoproterozoic

Table 2

C and O isotopic analysis (‰ relative to V-PDB and V-SMOW, respectively) of Paleoproterozoic (1–4) and Neoproterozoic (5–16) carbonatites of south India

Sample		$\delta^{13}\text{C}$	$\delta^{18}\text{O}$
1	H/46	-6.2	8.4
2	H/56	-6.2	8.2
3	H/47	-6.0	8.2
4	H/60	-6.1	8.1
5	SVT/5	-4.9	8.0
6	SVT/4	-6.4	13.2
7	Palla/4	-4.4	15.4
8	Palla/5	-5.1	10.7
9	PK/4	-5.3	8.61
10	BG/17	-4.0	11.1
11	OK/25	-3.3	25.4
12	OK/27	-3.6	24.0
13	MK/20	-3.9	7.3
14	MK/33	-3.8	7.4
15	MK/22	-3.7	7.3
16	MK/23	-3.8	7.4

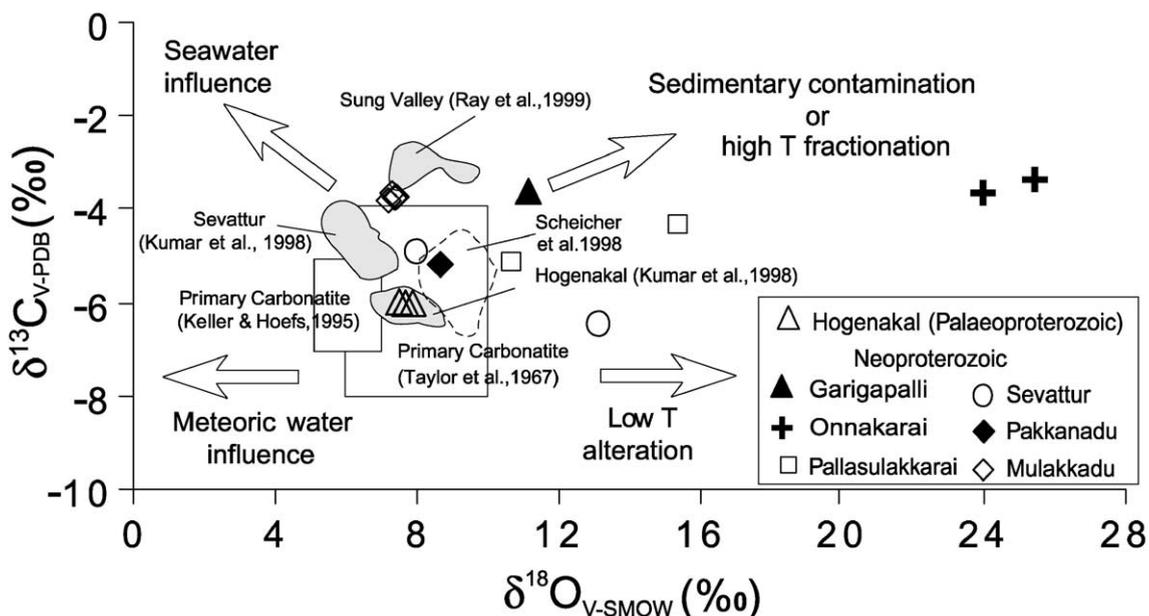


Fig. 6. Stable C- and O-isotopic compositions of south Indian carbonatites (relative to V-PDB and V-SMOW, respectively) plotted in the conventional  $\delta^{18}\text{O}$ – $\delta^{13}\text{C}$  diagram. The rectangular boxes represent the fields for unaltered primary carbonatites of Taylor et al. (1967) and Keller and Hoefs (1995). The arrows indicate possible effects of main processes on isotopic composition (details in Deines, 1989). The Hogenakal samples plot as a close cluster within the field of primary carbonatites while the Neoproterozoic carbonatites show a spread, mainly due to wide variation in O-isotopic composition. The fields of Seivattur carbonatite and Hogenakal calcite (Kumar et al., 1998) and Neoproterozoic carbonatites (Schleicher et al., 1998) have also been shown for comparison. Note uniform isotopic compositions of the Hogenakal whole rock (present study) and calcite separates (Kumar et al., 1998), and close similarity between Mulakkadu and Sung Valley carbonatites (Ray et al., 1999).

carbonatites have  $\delta^{18}\text{O}$  values consistent with mantle composition (+7.3‰ to +8.6‰); however, some of them show a non-correlative enrichment in  $^{18}\text{O}$  ( $\delta^{18}\text{O}$ =+10.1‰ to +15.4‰); two samples of Onnakarai ankeritic carbonatite being extremely enriched ( $\delta^{18}\text{O}$ =24.0‰ and 25.4‰). The Amba Dongar carbonatites of western India (Simonetti et al., 1995), southern African carbonatites (Horstmann and Verwoerd, 1997) and Brazilian carbonatites (Santos and Clayton, 1995) have also been reported to be variably enriched in  $^{18}\text{O}$ . The observed variations in C-isotopic composition in Neoproterozoic carbonatites cannot be related to any preferential escape of  $^{13}\text{C}$ -rich  $\text{CO}_2$  as this would not only produce regionally homogenous  $\delta^{13}\text{C}$  values within individual locality but would also affect the O-isotopic composition too (Demény et al., 1998).

On the conventional  $\delta^{13}\text{C}$ – $\delta^{18}\text{O}$  diagram (Fig. 6), the Hogenakal samples plot as a tight cluster within the primary mantle field of Taylor et al. (1967) and along

the boundary of the field proposed by Keller and Hoefs (1995). Our Hogenakal whole-rock data also overlap with the field of calcite separates of Kumar et al. (1998). We attribute the observed homogeneity in C- and O-isotopic compositions to batch crystallization of carbonatite under plutonic conditions, without any significant isotopic fractionation. The Neoproterozoic carbonatites, in contrast to Hogenakal ones, show a wide scatter, mainly on account of a range of O-isotopic compositions. Our Neoproterozoic carbonatite samples are relatively enriched in  $^{18}\text{O}$  as compared to the Seivattur carbonatite data of Kumar et al. (1998) and Schleicher et al. (1998), the C-isotopic compositions being comparable. The two extremely  $^{18}\text{O}$ -enriched Onnakarai samples plot far away from rest of the population. The significantly metamorphosed Pakkanadu carbonatites plot well within the primary field. In the absence of any discernible covariance between C- and O-isotopic compositions, the data do not support any isotopic fractionation and the non-

correlative enrichment in  $^{18}\text{O}$  can be attributed to post-magmatic modifications. The Hogenakal samples define a group isotopically distinct from the relatively  $^{13}\text{C}$ -enriched Neoproterozoic carbonatites. The latter can further be discriminated into two groups on the basis of coupled C- and O-isotopic compositions; one that overlaps with the mantle compositions and second with very high (and non-correlative)  $\delta^{18}\text{O}$  values. Such a distinction between Hogenakal ( $^{13}\text{C}$ -depleted) and Sevattur ( $^{13}\text{C}$ -enriched) carbonatites is also shown by the calcite separate data published by Kumar et al. (1998); however, they did not recognise the difference in C-isotopic compositions and preferred to interpret all the carbonatites to be derived from a single long-lived enriched mantle source. The Mulakkadu and Onnakarai carbonatites constitute a more enriched group ( $\delta^{13}\text{C}_{\text{V-PDB}} > -4\text{‰}$ ) within the Neoproterozoic carbonatites.

In contrast to the wide variability in C-isotopic composition of meteorites and mantle material such as diamonds, basalts and high-temperature volcanic gases, the range shown by carbonatites is rather narrow, representing the average composition of a large volume of the mantle source. The carbonatite melts are capable of concentrating volatiles about 1000 times their volume (Deines, 1989). The carbonatites are, however, widely variable in  $\delta^{18}\text{O}$  because the O-isotopic compositions are likely to be affected by post-magmatic modifications during various crustal processes. Unlike oxygen, the carbon in a parent carbonatite magma is believed to have completely been extracted from its mantle source (Deines, 1989) and therefore, the C-isotopic variations can be reliably used to model the mantle source. Moreover, the C-isotopic compositions, generally unaffected by a number of crustal processes, can be utilized to model the mantle source even in low-grade metamorphosed and hydrothermally altered samples (Deines, 1989). However, the carbon budget of the deep mantle reservoir itself can be significantly affected by the  $^{13}\text{C}$ -enriched  $\text{CO}_2$  from recycled carbonates from a subducting oceanic crust. Carbonatites originating from such magmas are likely to preserve the  $^{13}\text{C}$ -enriched character. Experimental studies have shown that carbonates from a subducting oceanic crust can penetrate to great depths of mantle (Biellman et al., 1993) even in the presence of silicate melts (Yaxley and Green, 1994).

Before attempting to model the mantle source, it would be necessary to evaluate the data for being representative as a number of processes can potentially alter the isotopic compositions. The likely effects of such processes (Deines, 1989) have been superimposed onto the  $\delta^{13}\text{C}$ – $\delta^{18}\text{O}$  plot (Fig. 6). The low-temperature isotopic exchange between carbonatite and water-rich magmatic fluids will lead to a positive shift in  $\delta^{18}\text{O}$  values. Such post-emplacement positive shift is quite common in carbonatites on account of presence of hydrous mineral phases such as micas and amphiboles (Deines and Gold, 1973; Santos and Clayton, 1995). High-temperature O-isotopic exchange with meteoric waters will result in  $^{18}\text{O}$  depletion. Mixing with a sedimentary source would result in an increase in both  $\delta^{18}\text{C}$  and  $\delta^{18}\text{O}$ . A correlative increase in  $\delta^{18}\text{C}$  and  $\delta^{18}\text{O}$  can also result from a high-temperature fractionation in the silicate–carbonate fluid system (Pineau et al., 1973; Deines, 1989); however, the  $\delta^{18}\text{O}$  enrichment beyond 15‰ cannot be explained through such a fractionation mechanism. The  $^{18}\text{O}$  enrichment in Garigapalli, Onnakarai and Pallasukkarai carbonatites appears to have resulted from alteration and therefore we have excluded these samples from further discussion of primary compositions. Although the  $\delta^{13}\text{C}$  values are not significantly affected during fluid–rock interaction, the most reliable C-isotopic compositions are found in low  $\delta^{18}\text{O}$  carbonatites ( $\delta^{18}\text{O} = 5.5\text{‰}$  to  $8.5\text{‰}$ ). Because the low  $\delta^{18}\text{O}$  carbonatites are unlikely to have experienced any low-temperature alteration, they represent isotopically primary carbonatites. We interpret the Hogenakal compositions to be isotopically primary and representative of the mantle source. Our C-isotope data point toward a  $^{13}\text{C}$ -depleted mantle source for Hogenakal, quite different from the mantle reservoir for Neoproterozoic carbonatites, the latter being significantly enriched in  $^{13}\text{C}$ . The granulite facies rocks (charnockites) of southern India and Sri Lanka have been reported to be rich in  $\text{CO}_2$  as a result of pervasive infiltration of mantle  $\text{CO}_2$  (Hoernes et al., 1991; Wickham et al., 1994). Equilibration of such mantle  $\text{CO}_2$  with charnockites in southern India yields oxygen isotopic compositions in the  $\delta^{18}\text{O}$  range of 7–13‰. This makes any deep crustal contamination indistinguishable from mixing with a different reservoir (Schleicher et al., 1998). Alternatively, the  $^{13}\text{C}$  enrichment in Neoproterozoic

carbonatites can be attributed to the incorporation of a subducted oceanic crust-derived recycled carbon. Similar high  $\delta^{13}\text{C}$  values ( $>-3.4\text{‰}$ ) in the Amba Dongar (western India) and Sung Valley (NE India) carbonatites have also been attributed to a subduction process (Ray and Ramesh, 1999; Ray et al., 1999). The observed C-isotopic variations in the Neoproterozoic carbonatites can be attributed to varying influence of migratory fluids from subducted oceanic crust (Nelson et al., 1988; Barker, 1996). The most likely process that can cause such an enrichment is the mixing of primordial carbon and recycled crustal inorganic carbon ( $\delta^{13}\text{C}=0\text{‰}$ ) in the mantle source region. Demény et al. (1998) have correlated the variations in  $\delta^{13}\text{C}$  in the Canary Island carbonatites to assimilation of sediments by the melt before its emplacement. Such a possibility seems unlikely in the present case due to the absence of any supportive evidence for crustal contamination. The C–O isotopic differences (within the mantle values) can also indicate mantle heterogeneity (Deines and Gold, 1973; Zaistev and Bell, 1995).

## 6. Sr–Nd isotopes

Rb–Sr and Sm–Nd isotopic analyses were carried out at the Geochronology Laboratory, University of Brasília. Sr was separated from whole-rock powders using a conventional cation exchange procedure. The Sr samples were loaded onto a double Re filament and the isotopic measurements were carried out on a multi-collector Finnigan MAT-262 mass spectrometer. Mass fractionation corrections were made using

$^{88}\text{Sr}/^{86}\text{Sr}$  ratio of 8.3752. The  $1\sigma$  uncertainty on the measured  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{87}\text{Rb}/^{86}\text{Sr}$  ratios was better than 0.2% and 2%, respectively. Sm and Nd extraction from whole-rock powders followed the technique of Richard et al. (1976), in which the separation of the REE as a group using cation-exchange columns precedes reversed-phase chromatography for the separation of Sm and Nd using columns loaded with HDEHP (di-2-ethyl-hexyl phosphoric acid) supported on Teflon powder. We have also used, more recently, the RE-Spec and Ln-Spec resins for REE and Sm–Nd separation. A mixed  $^{149}\text{Sm}$ – $^{150}\text{Nd}$  spike was used. Sm and Nd samples were loaded onto Re evaporation filaments of a double filament assembly. Uncertainties on Sm/Nd and  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios are considered to be better than  $\pm 0.05\%$  ( $1\sigma$ ) and  $\pm 0.00001$  ( $1\sigma$ ), respectively, based on repeated analyses of international rock standards BCR-1 and BHVO-1. The observed values for various standards were BHVO (rock standard)— $^{143}\text{Nd}/^{144}\text{Nd}=0.512971\pm 8$  ( $1\sigma$ ); La Jolla— $^{143}\text{Nd}/^{144}\text{Nd}=0.511853\pm 6$  ( $1\sigma$ ); NBS 987 (Sr standard)= $0.71025\pm 2$  ( $1\sigma$ ). The  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios were normalized to an  $^{146}\text{Nd}/^{144}\text{Nd}$  ratio of 0.7219. Nd procedure blanks were smaller than 100 pg.

The Sr and Nd data are presented in Table 3 and plotted in the conventional Nd–Sr anti-correlation plot (Fig. 7). The Sm and Nd concentrations in both Hogenakal and Neoproterozoic carbonatites are well within the limits generally shown by carbonatites (Bell and Blenkinsop, 1989), although much higher than most sialic rocks. The Paleoproterozoic Hogenakal carbonatites have a model age of  $>2.6$  Ga while the Neoproterozoic carbonatites range from 2.12 to 1.76 Ga. The  $^{147}\text{Sm}/^{144}\text{Nd}$  ratios do not allow any

Table 3  
Sr and Nd isotope results for Paleo- and Neoproterozoic carbonatites of south India

Sample	Sm (ppm)	Nd (ppm)	$^{147}\text{Sm}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$	TDM (Ga)	$\varepsilon_{\text{Nd}}(t)$	$^{87}\text{Sr}/^{86}\text{Sr}$ (measured)	$^{87}\text{Sr}/^{86}\text{Sr}$ (initial)	Sr (ppm)	Rb (ppm)
H/46	228	1336	0.103	0.511136	2.62	+1.00	0.70186	0.70174	12252	13.49
H/56	233	1284	0.110	0.511238	2.65	+0.54	0.70172	0.70161	12156	12.65
PK/7	206	1530	0.081	0.511186	2.12	−16.5	0.70536	0.70534	5545	2.72
OK/27	19.1	64.6	0.178	0.512022	—	−10.1	0.70566	0.70500	1337	26.75
OK/25	137	675	0.123	0.511814	2.05	−8.55	0.70550	0.70486	1323	25.67
SVT/5	54	288	0.113	0.511867	1.76	−6.48	0.70532	0.70528	7515	8.78
SVT/4	61	348	0.105	0.511838	1.68	−6.23	0.70536	0.70532	7454	8.98
MK/23	78	474	0.099	0.511476	2.07	−12.7	0.70658	0.70658	15678	0.33
BG/17	271	1314	0.125	0.511835	2.06	−8.34	0.70545	0.70544	6176	2.56

Parameters for calculating  $\varepsilon_{\text{Nd}}$ :  $^{143}\text{Nd}/^{144}\text{Nd}$  CHUR=0.512638 and  $^{147}\text{Sm}/^{144}\text{Nd}$ =0.1967.  $^{143}\text{Nd}/^{144}\text{Nd}$  are the measured ratios.

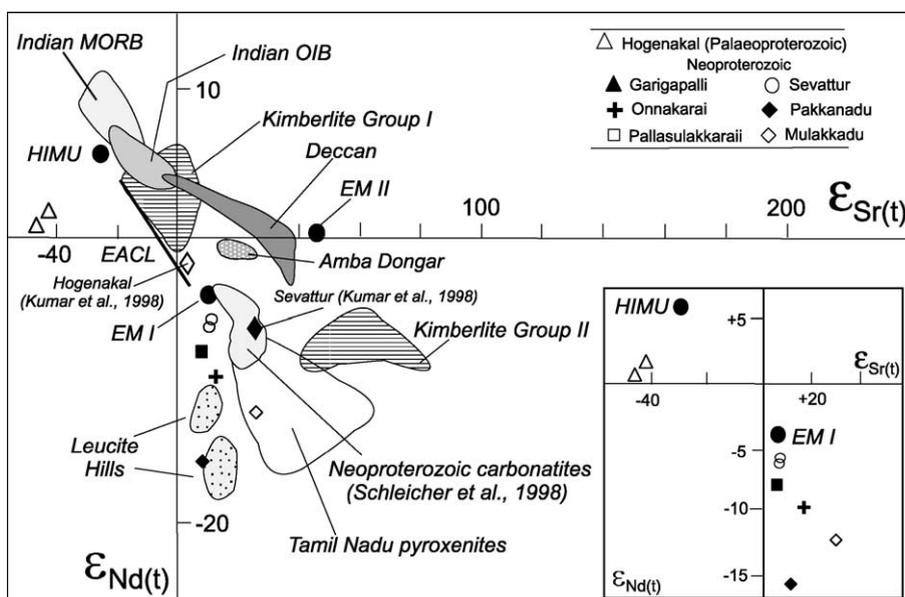


Fig. 7.  $\epsilon_{\text{Nd}}-\epsilon_{\text{Sr}}$  diagram for carbonatites of southern India. The fields of other known occurrences and carbonatites are superimposed for comparison. Compiled from Vollmer et al. (1984), Nelson et al. (1988), Schleicher et al. (1990), Peng and Mahoney (1995), Simonetti et al. (1995), Schleicher et al. (1998) and Miyazaki et al. (2000). Note a close correspondence of our Neoproterozoic carbonatites data with Schleicher et al. (1998). The Hogenakal and Setattur calcite separate data from Kumar et al. (1998) have also been shown. (EACL—East African Carbonatite Line; Bell and Blenkinsop, 1989). The inset figure shows the discrimination of south Indian Paleo- and Neoproterozoic carbonatites into ‘depleted’ and ‘enriched’ quadrants and affinity of the younger carbonatites with the EM I-like mantle reservoir. EM I is the age corrected (800 Ma) composition while HIMU and EM II are present day compositions.

discrimination between older and younger carbonatites and range from 0.099 to 0.178. The carbonatites show rather low present day  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios, varying between 0.511136 and 0.511867 with the exception of one sample (OK/27), which shows a higher ratio (0.512022). The south Indian carbonatites have been reported to have generally low  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios as compared to most other carbonatite occurrences (Kumar et al., 1998; Schleicher et al., 1998; Schleicher, 2001). Our Nd-isotopic results for Hogenakal carbonatites differ from Kumar et al. (1998), who reported  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios of 0.511239 and 0.511317 and corresponding  $\epsilon_{\text{Nd}}$  values of  $-1.1$  and  $-0.9$ . However, they are in general agreement with many Late Archean igneous rocks (see Shirey and Hanson, 1986, for a summary), and Late Archean to Early Proterozoic Canadian carbonatites, such as 2.68 Ga Superior Province ( $\epsilon_{\text{Nd}}=-0.5$  to  $+0.8$ ; Tilton and Bell, 1994), 2.6 Ga Slave Province ( $\epsilon_{\text{Nd}}=+1.8$ ; Villeneuve and Relf, 1998) and  $\sim 1.9$  Ga Borden and Cargill carbonatites ( $\epsilon_{\text{Nd}}=+1.8$ ; Bell and Blenkinsop,

1987a). Similar Nd-isotopic compositions have been reported for the associated alkaline rocks ( $+1.4$  for apatite in Poohbah Lake syenite, Bell and Blenkinsop, 1987a;  $+2.3$  for Otto stock syenite, Tilton and Kwon, 1990). The south Indian Neoproterozoic carbonatites have variable Nd-isotopic ratios ( $\epsilon_{\text{Nd}}=-12.7$  to  $-6.23$ , one sample with a rather low value of  $-16.5$ ), significantly different from the Hogenakal data. Our Nd-isotopic data for the Neoproterozoic carbonatites show an overlap with the results of Schleicher et al. (1998); however, they report a much narrower  $\epsilon_{\text{Nd}}$  variation ( $\epsilon_{\text{Nd}}=-8.35$  to  $-5.45$ ). Kumar et al. (1998) have reported higher  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios for Sevtatur carbonatites (0.511815–0.511931) and corresponding  $\epsilon_{\text{Nd}}$  values ( $-5.9$  to  $-5.1$ ).

Kumar et al. (1998) have suggested the Hogenakal mantle source to have evolved to yield Sevtatur carbonatites. Our Nd-isotopic data, in contradiction, point to isotopically different mantle sources for Hogenakal and Neoproterozoic carbonatites. Jayananda et al. (1995) have published isotopic results

on the mantle-derived quartz monzonite from the 2.5-Ga Closepet batholith, located to the west of the carbonatites discussed in this paper. Their results yield marginally negative  $\epsilon_{\text{Nd}}$  that approach the CHUR and indicate crustal interaction. Relatively younger and uncontaminated, 2 Ga Agali Coimbatore Dykes (tholeiitic) yield significantly negative  $\epsilon_{\text{Nd}}$  values, indicating an enriched source (Radhakrishna et al., 1995).

Initial Sr ratios are equivalent to the measured values (within analytical uncertainty) due to extremely small Rb/Sr ratios (Table 3). The Hogenakal carbonatites yield significantly lower  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.70161–0.70174). Kumar et al. (1998) have reported  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.70185 and 0.70236) for two Hogenakal carbonatite samples, which are different from our results; however, their pyroxenite (host for Hogenakal carbonatite) data (0.70172 and 0.70177) show a close similarity to our Hogenakal carbonatites data. Our Sr-isotopic results are in general agreement with Late Archean and Early Proterozoic carbonatites of Canadian Shield, such as 2.68 Ga Superior Province (0.70125–0.70135, Tilton and Bell, 1994), 2.6 Ga Slave Province (0.701184, Villeneuve and Relf, 1998) and ~1.9 Ga Borden and Cargill carbonatites (0.70181 and 0.70198, respectively; Bell and Blenkinsop, 1987a). The south Indian Neoproterozoic carbonatites are significantly enriched in the radiogenic Sr ( $^{87}\text{Sr}/^{86}\text{Sr}$ =0.70486 to 0.70544, sample MK/23 having a very high ratio of 0.70658). The Sr-isotope data are in agreement with the results of Schleicher et al. (1998) and Kumar et al. (1998) who reported  $^{87}\text{Sr}/^{86}\text{Sr}$  ranges between 0.70455 and 0.70537, and 0.70508 and 0.70523, respectively, for the same carbonatites. Mass balance calculations carried out by Schleicher et al. (1998) preclude any contamination in the Neoproterozoic carbonatites, as extremely high contents of contaminants would be required to effect the changes in the melt that would also seemingly alter the melt characteristics.

In relation to bulk Earth, the depleted source would require high Sm/Nd, and low Rb/Sr ratios. However, the measured Sm/Nd ratios for Hogenakal (0.171 and 0.182) and Neoproterozoic carbonatites (0.135–0.206) are rather low relative to bulk Earth and to the depleted mantle reservoir (Sm/Nd=0.358). This implies a light REE enrichment relative to chondrites (Bell and Blenkinsop, 1987a). In contrast, the Neoproterozoic carbonatites have higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios

and lower  $\epsilon_{\text{Nd}}$  values, thus implying their genesis from an LILE-enriched source relative to CHUR.

In the Sr–Nd diagram, the south Indian carbonatites plot in two different quadrants that correspond to depleted and enriched mantle components (Fig. 7). The Hogenakal carbonatites plot in the ‘depleted’ quadrant, being much lower in  $^{87}\text{Sr}/^{86}\text{Sr}$  as compared to the bulk Earth and marginally higher in  $\epsilon_{\text{Nd}}$  relative to CHUR. The mantle reservoir for Hogenakal carbonatites, however, appears to significantly differ from the present day HIMU or MORB compositions, being more depleted in  $^{87}\text{Sr}/^{86}\text{Sr}$  and slightly enriched in Nd. Absence of HIMU component during Early Precambrian is also indicated by the 2.7-Ga-old carbonatites of Canadian Shield (Tilton and Bell, 1994). The existing models for Archean mantle differentiation yield depleted reservoirs (Chase and Patchett, 1988; Galer and Goldstein, 1991; McCulloch, 1993). Vidal (1992) and McCulloch (1993) have postulated near complete melting (accompanied by dehydration) of subducted oceanic lithospheric slab at shallow depth during the Early Precambrian to yield LILE-depleted outer mantle. Partial melting of basalt derived from such a source resulted in abundant tonalites. The isotopic signatures pointing to a depleted component can be attributed to large-scale differentiation and extraction of the continental crust to produce a depleted source. Later on, as the earth cooled, the slabs could be subducted deeper into the mantle and dehydration reactions resulted in creating an enriched reservoir. Such a model explains a ‘depleted’ source for the 2.4-Ga Hogenakal carbonatites and ‘enriched’ one for the Neoproterozoic carbonatites.

Data on younger African carbonatites suggest a mixing of HIMU and EM-I-type mantle components (Bell and Blenkinsop, 1987b). The depleted source has been postulated to be lithospheric and the other, possibly asthenospheric (Bell and Blenkinsop, 1989). Our Sr–Nd isotopic results identify isotopically different mantle sources for Paleo- and Neoproterozoic carbonatites of south Indian and also preclude any mixing of depleted and enriched mantle components in carbonatite melt generation. The isotopic data from the Canadian Shield (Tilton and Bell, 1994; Bell and Blenkinsop, 1989; Tilton and Kwon, 1990) identify a depleted mantle source for 2.7–0.11 Ga carbonatites, postulating a major depletion event at ~2.9 Ga. As

the depleted mantle component has been fairly widespread and the source for most of the Precambrian carbonatites, its absence in the Indian subcontinental region, thus appears to be rather incongruous. Recognition of the ‘depleted’ mantle component in the south Indian region indicates the Archean ‘depleted’ mantle to be more widespread.

## 7. Discussion and conclusion

Miyazaki et al. (2000) have attributed significant differences in isotopic composition between south Indian Neoproterozoic carbonatites and associated silicate rocks to isotopically different mantle sources. High to very high REE, Sr and Ba contents and variable abundances of HFS elements postulate a rather fractionated stage for the south Indian carbonatitic melt. Typically mantle and seemingly unaltered (low  $\delta^{13}\text{C}$ ) C isotopic ratios for Hogenakal and significantly  $^{13}\text{C}$ -enriched ones for Neoproterozoic carbonatites argue in favour of isotopically discrete mantle sources for them. Observed  $\delta^{13}\text{C}$  variations in Neoproterozoic carbonatites can be linked to variable enrichment of the mantle source under the influence of metasomatizing fluids. The south Indian subcontinental mantle has previously been described as a ‘long-lived enriched’ one that preserved the ‘enriched’ character through the Proterozoic (Kumar et al., 1998; Schleicher et al., 1998; Miyazaki et al., 2000). Our recognition of an Early Proterozoic ‘depleted’ mantle, based on new isotopic data on 2.4 Ga Hogenakal carbonatites, thus assumes a wider significance in constraining the mantle evolution in the region. Such an interpretation is documented by complementary isotopic evidence such as mantle  $\delta^{13}\text{C}$  values, positive  $\epsilon_{\text{Nd}}$  and extremely low  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. Trace element abundances and isotopic signatures also preclude any crustal contamination. The coherence between Nd and Sr isotopic data, in concert with the C-isotopic signatures, provides conclusive evidence for a depleted source for Hogenakal carbonatites, analogous to the mantle region in the Canadian Shield (Tilton and Bell, 1994; Bell and Blenkinsop, 1987a). Similar Sr and Nd isotopic compositions of the pyroxenite (hosting Hogenakal carbonatite) also support a Late Archaean ‘depleted’ mantle in the south Indian subcontinental region. However, in contradiction to

the Canadian Shield region where such a depleted mantle source remained preserved from 2900 to 110 Ma (Bell and Blenkinsop, 1987a), the isotopic data on Neoproterozoic carbonatites of south India (present study; Schleicher et al., 1998; Kumar et al., 1998) indicate an enriched mantle reservoir too.

Schleicher et al. (1998), based on the Pb-isotopes, have recognized some crustal component and interpreted the mantle source for Neoproterozoic carbonatites to have resulted from two-component mixing (one end-member being possibly crustal). The Pb isotopes are very sensitive to contamination on account of low Pb abundance in carbonatites as compared to very high Sr and Nd contents that are buffered against any such effect. The C–O isotope systematics cannot be used to evaluate the crustal contamination because any such contaminant (mantle  $\text{CO}_2$  equilibrated high-grade rocks) may not be isotopically discriminated from an enriched mantle component (Schleicher et al., 1998). The  $^{13}\text{C}$  enrichment in Neoproterozoic carbonatites (relative to Hogenakal ones) can be explained through mantle metasomatism under the influence of subducting Dharwar crust, subsequent to the differentiation of Hogenakal carbonatite melt. This would also explain the relatively higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios and negative  $\epsilon_{\text{Nd}}$  values in Neoproterozoic carbonatites. Ray and Ramesh (1999) and Ray et al. (1999) have attributed similar  $^{13}\text{C}$ -enrichment in Amba Dongar and Sung Valley carbonatites (average  $\delta^{13}\text{C} = -3.4\text{‰}$  and  $-3.2\text{‰}$ , respectively) to a mantle source enriched by incorporation of crustal inorganic carbon. With the recognition of an Archean depleted mantle and further evidence provided for a Proterozoic enriched one, the isotopic evolution of the mantle in south Indian subcontinental region can be interpreted in two different ways:

a. The mantle remained heterogeneous, with co-existing depleted and enriched regions, from the Late Archean through to the Proterozoic. The Hogenakal and Neoproterozoic carbonatites were produced from such reservoirs at different times. This suggests a possible long-lived mantle heterogeneity. The mantle heterogeneity has already been established as Zindler and Hart (1986) have identified at least six components in the mantle, postulating the mantle to be more heterogeneous than the simple model envisaging three large-scale reservoirs (Allègre, 1982). Published

results on Neoproterozoic carbonatites and coeval mantle-derived silicate rocks of south India point toward their derivation from an ‘enriched’ source. Absence of ‘depleted’ mantle signatures in the Neoproterozoic rocks does not support the heterogeneity in the south Indian subcontinental mantle.

b. The Paleoproterozoic Hogenakal carbonatites were derived from an LILE-depleted mantle, following the extraction of the crust during Late Archaean. Later subduction of part of the Dharwar craton around 2.5 Ga subsequently metasomatized the upper mantle and the younger carbonatites were produced from this (metasomatically enriched) reservoir. Differences in C isotopic composition in the Neoproterozoic carbonatites can be related to variable enrichment by such migratory fluids. This model explains the observed isotopic trends and appears to be in agreement with the existing models of Precambrian subcontinental mantle evolution (Vidal, 1992; McCulloch, 1993). Similar model has also been proposed for the evolution of the subcontinental mantle in Canadian Shield (Bell and Blenkinsop, 1987a; Tilton and Bell, 1994). Recycling of crustal carbon into the mantle source for some younger carbonatites of western and eastern India has also been recognized by Ray and Ramesh (1999) and Ray et al. (1999).

High  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios (0.70486–0.70544) and low  $\epsilon_{\text{Nd}}$  (–12.7 to –6.23), and absence of any crustal contamination necessitate an ‘enriched’ source for south Indian Neoproterozoic carbonatites. The enrichment can result from interaction of two mantle components, one of them enriched. Alternatively, metasomatism through a subducting oceanic crust can also lead to such enrichment. The Pb–Pb array in Sevattur carbonatites has been interpreted by Schleicher et al. (1998) as a mixing between two lead reservoirs; one of them a mantle reservoir with lower  $\mu$  value while the other a U-enriched reservoir, most likely a crustal one. However, they did not spell out the details and mechanism of such a mixing. On the strength of the arguments presented in preceding sections, we interpret this ‘crustal’ end-member to be the subducted oceanic crust. The subduction process is also indicated by diagnostic trace-element signatures of the Neoproterozoic syenites (Miyazaki et al., 2000). Major tectonic activities, such as crustal generation, magmatism and metamorphism in southern India have been dated at around 2.5 Ga (Friend

and Nutman, 1991; Peucat et al., 1993). These occurred at the southern edge of the Dharwar craton representing a convergent margin. Subduction of sediments and modified oceanic crust yielded ample  $\text{CO}_2$  necessary for the concurrent charnockite forming processes (Kumar et al., 1998). Equilibration of  $\text{CO}_2$  derived from the oceanic crust with the existing carbon reservoir resulted in relative enrichment of  $^{13}\text{C}$  in the source region that subsequently yielded the Neoproterozoic carbonatites.

The Nd and Sr isotopic data do not provide constraints on the depth of carbonatite melt generation within the mantle, but along with the C- and O-isotopic data, do provide critical evidence of distinct mantle sources for Paleoproterozoic (Hogenakal) and Neoproterozoic carbonatites. Whether the mantle has remained heterogeneous throughout the Late Archaean to Proterozoic or whether it became enriched during the Early Proterozoic by metasomatism during subduction needs to be substantiated.

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