Trace element and isotopic studies of Permo-Carboniferous carbonate nodules from Talchir sediments of peninsular India: Environmental and provenance implications

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Syngenetic carbonate nodules constitute an interesting feature of the glaciogene sediments of various Talchir basins in peninsular India. Petrographic, cathodoluminescence and sedimentary results suggest that many of these nodules contain primary carbonate precipitates whose geochemical signatures can be used for determining environment of deposition and provenance of the sediments and drainage source. Several nodules were collected from Gondwana basins of east-central India and analyzed for stable carbon and oxygen isotope ratios, REE and trace element composition, and Sr isotope ratio. The mean δ^{18}O and δ^{13}C values of the calcites in the nodules are $-$ 19.5‰ and–9.7‰ (w.r.t. PDB) respectively suggesting a freshwater environment (probably lacustrine) for formation of these objects. Trace element ratios (Eu/Eu* and La/Yb) of the nodule samples show that the source of the sediments in the Damodar valley basin was the granites, gneisses and intrusives in the Chotanagpur region. The sediments in the Mahanadi valley were derived from granulites, charnockites and granites of the eastern ghat region. The Sr concentration of the carbonate phase of the nodules is low, ranging from 10–60 ng/g . The ^{87}Sr/^{86}Sr ratios of the samples from the west Bokaro basin and Ramgarh basin vary from 0.735 to 0.748 (mean: 0.739) and from 0.726 to 0.733 (mean: 0.730) respectively. These values are consistent with our proposition that water of these basins drained through the granitic rocks of the Chotanagpur region. In contrast, the ^{87}Sr/^{86}Sr ratios of the samples from the Talchir basin (Type area) of Mahanadi valley vary from 0.718 to 0.723 (mean: 0.719). These ^{87}Sr/^{86}Sr ratios are close to those of the granulites in the adjoining eastern ghat belt suggesting that area as the drainage source.

1. Introduction

The late Carboniferous-early Permian Talchir Formation of Indian Gondwana contains sediments comprising conglomerates, sandstones, and siltstones which are occasionally capped by sediments with marine fossils (Ghosh 1954). Considerable information on the sedimentological pattern and the nature of sediments in the Talchir Formation has been acquired in the last few decades (Ghosh and Mitra 1975; Casshyap and Tewari 1988; Bhattacharya *et al* 1989; Pandya 1990; Veevers and Tewari 1995). The lower part of the Formation is now believed to be of glacial origin. However, the environment of deposition of the upper part is still not clear and the observed sedimentary features can be explained equally well by sedimentation in a sea or a big lake.

In the majority of Gondwana exposures, the Talchir strata are overlain by coal-bearing Barakar

Formation with intervening deposits bearing fresh water signature (Casshyap and Kumar 1987). However, in Umaria (figure 1) a marine bed is found sandwiched between the Talchir and the Barakar Formations. This marine bed comprises a three-meter-thick limestone containing fossil shells of *Productus, Spiriferina, Reticularia* etc and a Permo-Carboniferous age (Asselian-Sakmarian) has been assigned to this bed based on this fossil assemblage (Ghosh 1954). Similar marine beds have also been found in the neighbouring Manendragarh and Daltonganj localites (figure 1) (Krishnan 1982) where exposures containing marine fossils occur in several patches attaining a thickness of up to five meters. The marine fauna identified from these two sections includes *Protoretepora, Spirifer, Aviculapecten, Eurydesma, Hyperammina, Glomospira* etc. (Bhatia and Singh 1959). The presence of marine fossils in beds overlying the Talchir Formation was cited as an indicator of an inland sea for Talchir sedimentation even though no marine fossils have been discovered in the Talchir Formation itself (Ghosh 1954). Bose *et al* (1992) interpreted the morphological (Hummocky cross-stratification) features of the (Talchir) sandstone facies of the west Bokaro basin (Dudhinala section) as the product of wave-agitated shelf environment in an open water body (either sea or a vast lake). Recently, Sengupta *et al* (1999) reported the presence of external casts of molluscs from the same place and interpreted this as evidence of marine incursion in west Bokaro. However, the absence of marine body fossils in any of the Talchir exposures and questions regarding the origin of the isolated single external cast raise doubts about the proposed marine environment for the Talchir sediments (Mukhopadhyay and Bhattacharya 1994; Sen 1995; Bhattacharya *et al* 2001). In contrast, the association of plant mega-fossils (Rikba horizon) near the top of the Talchir Formation, the glacial character of the basal conglomerate and the presence of cross bedded sandstone and pebbly conglomerate in the overlying unit indicate a glacio-fluvial, glacio-lacustrine freshwater milieu (Ghosh and Mitra 1975). It is clear that the origin and nature of the Talchir sedimentation cannot be inferred unambiguously on the basis of sedimentary features. For example, hummocky structures are also found associated with sediments from lakes as in lake Ontario, Canada (Eyles and Clark 1986).

Geochemical analysis of the Talchir sediments can play an important role in identifying the environment of sedimentation. The isotopic composition of the authigenic carbonates occurring as spheroidal nodules in the sediments can be used to infer the genesis of the sediments in which they occur. Our recent work (Bhattacharya *et al* 2001) on stable isotopes of oxygen and carbon in Talchir sedimentary carbonates attempts to resolve the controversy pertaining to the deposition of the Talchir sediments (marine vs freshwater regime). In this work, chemical composition of the bulk sediments comprising the nodules of the Talchir deposits has been used in understanding the genesis of the sediments and the environment of their deposition.

2. Materials and methods

Two major Gondwana basins were investigated in this study: The Damodar valley basin and the Mahanadi valley basin, each comprising several exposures classified as sub-basins (figure 1). Each of these basins contains at its base a sequence of conglomerate bed, sandstone and shale for which the name Talchir Formation was proposed after the type area in Mahanadi valley basin. An excellent exposure of the Talchir sediments of the Damodar valley basin was found (Ghosh and Mitra 1975) near the confluence of Dudhi Nala, Dube Nala and Silai Nala about 0.5 km south of the village Jarwa (lat. 85°27′ and long. 23°49′) at the western part of the Bokaro sub-basin (figure 1a). The sediments here rest unconformably on the Archaean basement and comprise conglomerate, sandstone and siltstone. Spheroidal calcareous nodules were found in the bluish grey siltstone bed and rhythmite bed (Ghosh *et al* 2001; Bhattacharya *et al* 2001). Inspite of large variations in the outer appearance of the nodules, they can be broadly classified into three types:

- Tennis ball shaped nodules composed of the same material as the host rock (i.e., arkosic sandstone),
- Discoidal nodules embedded in the siltstone and often displaying multiple growths giving rise to many bizarre shapes, and
- Round to irregularly shaped nodules outcropping on the flat surface of the varve-like siltstone.

It is clear that the nodules were not transported from elsewhere. The *in situ* growth (indicating accumulation of carbonate around a centre) can be clearly identified by the continuity of lamination between the host and the nodular body.

Large numbers of nodules were collected for analysis from the Dudhi Nala section (figure 1a) and from the Nandirjhor Nala section (figure 1b) in the Talchir sub-basin (in Mahanadi valley basin). They occur within the rhythmite bed and the calcareous siltstone bed.
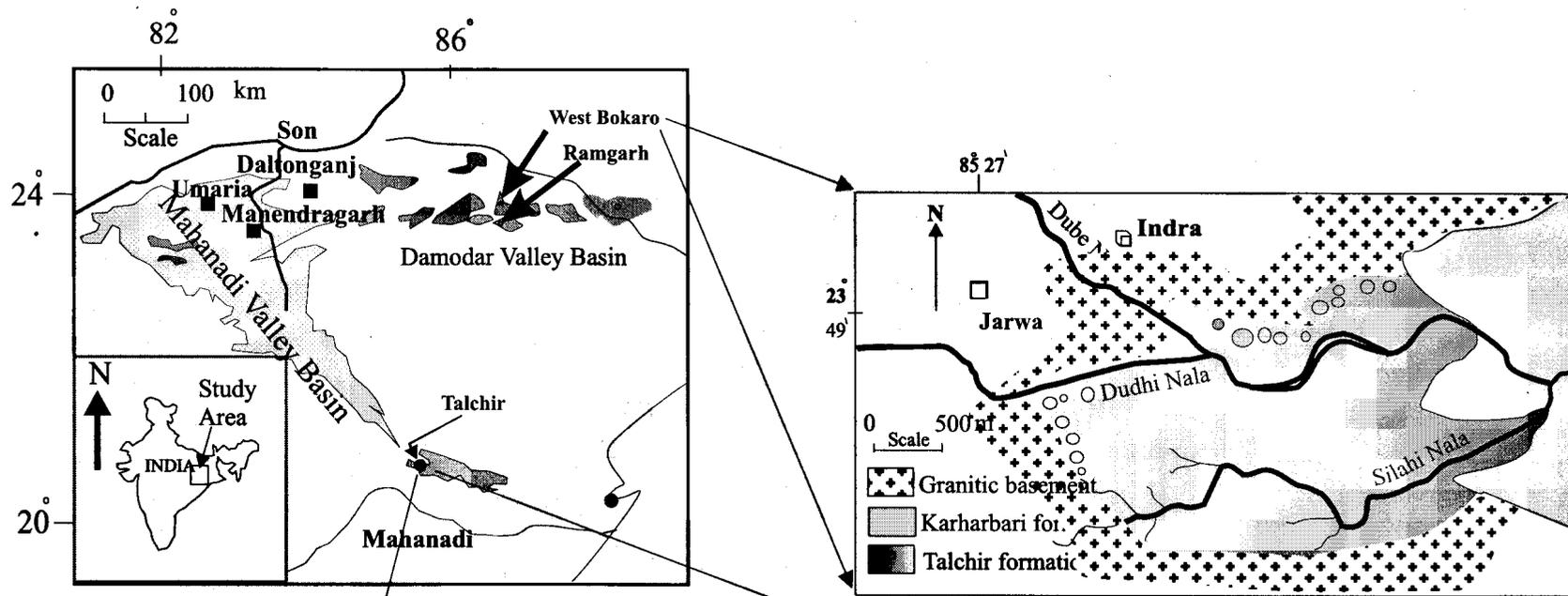
Figure 1(a)
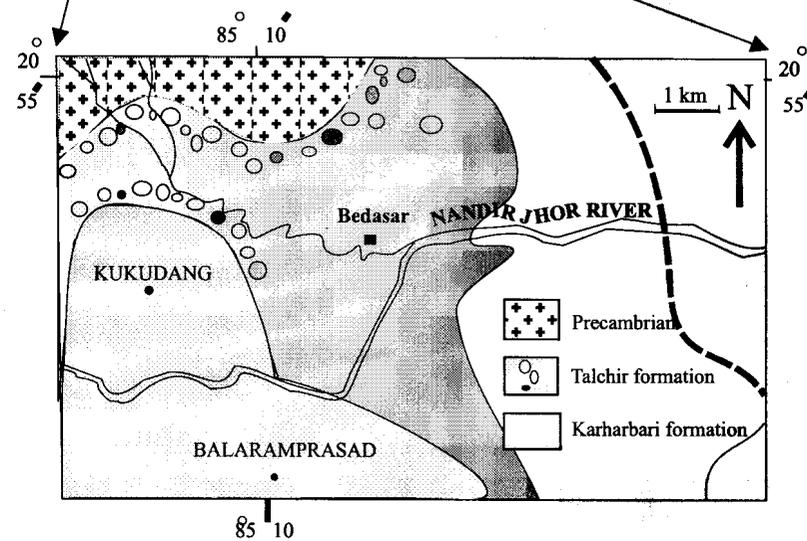
Figure 1(b)

Figure 1. Three major Gondwana basins of peninsular India. Present work is confined to Damodar valley basin (west Bokaro and Ramgarh sub-basins) and Mahanadi valley basin (Talchir sub-basin). (**a**): Geological map of west Bokaro sub-basin. (**b**): Geological map of Talchir basin showing Nandirjhor Nala section.

2.1 *Petrography and X-ray diffraction studies*

X-ray diffraction analysis of powders from the nodule samples shows that calcite is the dominant carbonate mineral in the matrix. Several lines of petrographic evidence indicate that most of the nodular concretions formed very early in the diagenetic history of the sediments and did not alter significantly afterwards (Ghosh 2000). Consequently, the trace elements, REE and isotopic data may be used for deciphering primary signatures of sedimentary environment and sediment provenance.

2.2 *Cathodoluminescence studies*

Cathodoluminescence (CL) is an additional tool for studying the extent of diagenetic alteration in carbonates. Viewed under CL, carbonates show varied luminescence signatures (in orange, brick red, and yellow colors) ranging from non-luminescent to dull luminescent, to moderately luminescent, to bright luminescent. The variation in CL of calcretes, for example, can help in the recognition of different events and stages of carbonate precipitation. Driese and Mora (1993), based on a study of pedogenic calcites, showed that CL components define chemically distinct petrographic fields corresponding to separate environments. Microprobe data show that dull red-brown luminescent micrites typically contain Mn concentrations of 500 to 2000 ppm and Fe concentrations below 2000 ppm. Bright yellow orange luminescence in the late calcite spar cement indicates extremely high Mn concentration (up to 34,000 ppm) and low Fe concentration (below detection limit). Such high concentrations of Mn suggests the availability of Mn in solution during calcite precipitation. This requires a reducing environment which can develop during burial and oxidation of organic matter in the sediments.

The nature of post-depositional cementation was investigated in a few nodule samples with the help of CL images of thin sections. This showed that the majority of the nodules are composed of two generations of calcites characterized by two shades of orange colouration:
- bright orange, indicating precipitation from water having a high concentration of Mn ions (Veizer *et al* 1999; Tobin *et al* 1999) and
- dull red CL (usually in the calcite surrounding detrital quartz grains) indicating low Mn abundance.

Based on these characteristics we can distinguish between calcite precipitated at the sediment water interface (calcite with less Mn) and calcite formed during burial diagenesis (high Mn). Identification of these two major classes of calcite is helpful in the interpretation of isotopic composition of the nodular carbonates. Some of the samples with features of late diagenesis (as revealed by CL), as well as petrological signs of alteration, are discarded during discussion of the results.

2.3 *Analytical procedure*

Measurement of Sr isotopic composition in the carbonate phase was done following the leaching technique of Lord *et al* (1988). About 100 mg of sample powder was taken in a clean teflon vial and soaked with ultrapure water (obtained by double distillation in a quartz system) before leaching the carbonate with 0.2 N HCl for 2 hours at room temperature. The solution was centrifuged and dried on a hot plate at 90°C, redissolved in 2 N HCl and transferred into an elution column containing ion-exchange resin (Dowex 50×8, 200–400 mesh) for separation and purification of Sr. The Sr fraction collected after elution was completely dried and loaded on to a tantalum filament for isotopic measurement. 20 samples along with standards were analyzed in a VG-354 Thermal Ionization Mass Spectrometer (TIMS) at National Geophysical Research Institute (NGRI), Hyderabad. Another batch of 20 samples was analyzed in a home made TIMS at Physical Research Laboratory (PRL), Ahmedabad (9 inch radius, single focussing mass spectrometer fitted with a Faraday cup collector) described in Trivedi *et al* (1995). The Sr isotopic ratio was normalized to $^{86}Sr/^{88}Sr = 0.1194$ (natural abundance ratio). The long term reproducibility of measurement was checked by measuring the $^{87}Sr/^{86}Sr$ of the carbonate standard NBS-987 (in PRL) and SRM (in NGRI) periodically. The mean $^{87}Sr/^{86}Sr$ value of the NBS-987 was found to be 0.71025 ± 0.00007 (at 2σ) in our laboratory while for SRM the mean value was 0.71023 ± 0.00002 (at 1σ) at NGRI. Typical 2 sigma error for a single $^{87}Sr/^{86}Sr$ determination was 0.0005 (when measured in PRL) and 0.00004 (when measured in NGRI). These errors are quite small compared to the magnitude of Sr isotope variation in the samples. As an additional check, some of the samples were analyzed both at NGRI and PRL. The results usually differed only by 0.001 (e.g., for sample number DN/N2/95 the two values are 0.7379 and 0.73699).

For Rare Earth Elements and trace elements neutron activation analysis was used. Several grams of bulk sediment (containing carbonate as well as detrital material) were dried and ground before being irradiated at a neutron flux of $1.5 \times 10^{12} \, cm^{-2} \, sec^{-1}$. Based on activity of short-lived radionuclides the concentration of 32 elements were determined at the Radioactivity Counting Laboratory of Tokyo Metropolitan University using coaxial and planer type pure Ge detector for gamma ray measurements. Elemental abundances were

obtained by comparing γ-ray intensities of the samples with those of reference standard rocks, JB-1 (supplied by Japan Geological Survey) and BCR-1, which were irradiated with the nodule samples. The Allende meteorite reference sample prepared by the Smithsonian Institute, USA also was simultaneously irradiated. Analytical errors due to counting statistics are less than 5% (less than 2% for most elements).

3. Results and discussions

3.1 *Sedimentation in a fresh water lake*

Before discussing the elemental data it is worthwhile to summarize the conclusions based on the stable isotope analysis of the nodules reported in our recent work (Bhattacharya *et al* 2001). An important feature of the stable isotope data is highly depleted oxygen isotopic composition (mean -19.5‰ w.r.t PDB) of a majority of the nodules. This suggests that precipitation of the Talchir carbonates took place in a fresh water environment because marine carbonate cements are characterized by $\delta^{18}O$ values close to zero or slightly negative (Veizer *et al* 1999). This conclusion is also supported by the depleted carbon isotope ratio of the nodules (mean -9.7‰ w.r.t. PDB) indicating

mixing of biogenic carbon with normal sedimentary carbon in making up the bicarbonate reservoir of the basin. The carbon isotope ratio in ancient sedimentary carbonates is least affected during diagenesis and alteration (Schidlowski and Aharon 1992). Therefore, the observed $\delta^{13}C$ values of the nodules strongly indicate that the basin was made up of fresh water (if the precipitation took place in sea water the carbon isotope ratio would have been within ± 2‰).

As mentioned before, petrographic investigations of the nodular carbonates indicate that most of the nodules represent *in situ* primary precipitates. Cathodoluminescence observations suggest that a majority of the calcites were precipitated from water having high Mn/Fe ratio, as expected in glacio-fluvial condition and contrary to that expected if the Talchir carbonates were formed in a marine regime with low Mn/Fe ratio (Tobin *et al* 1999). Moreover, the association of the nodules with glacial diamictites suggests freshwater lakes formed by glacier melt-water as probable milieu for sedimentation and carbonate precipitation (schematically shown in figure 2). The nodules probably formed inside the sediment bed by radial growth around a favourable starting nucleous induced by physico-chemical conditions of the micro-environment. The growth process involves compaction of the detrital minerals by cementation
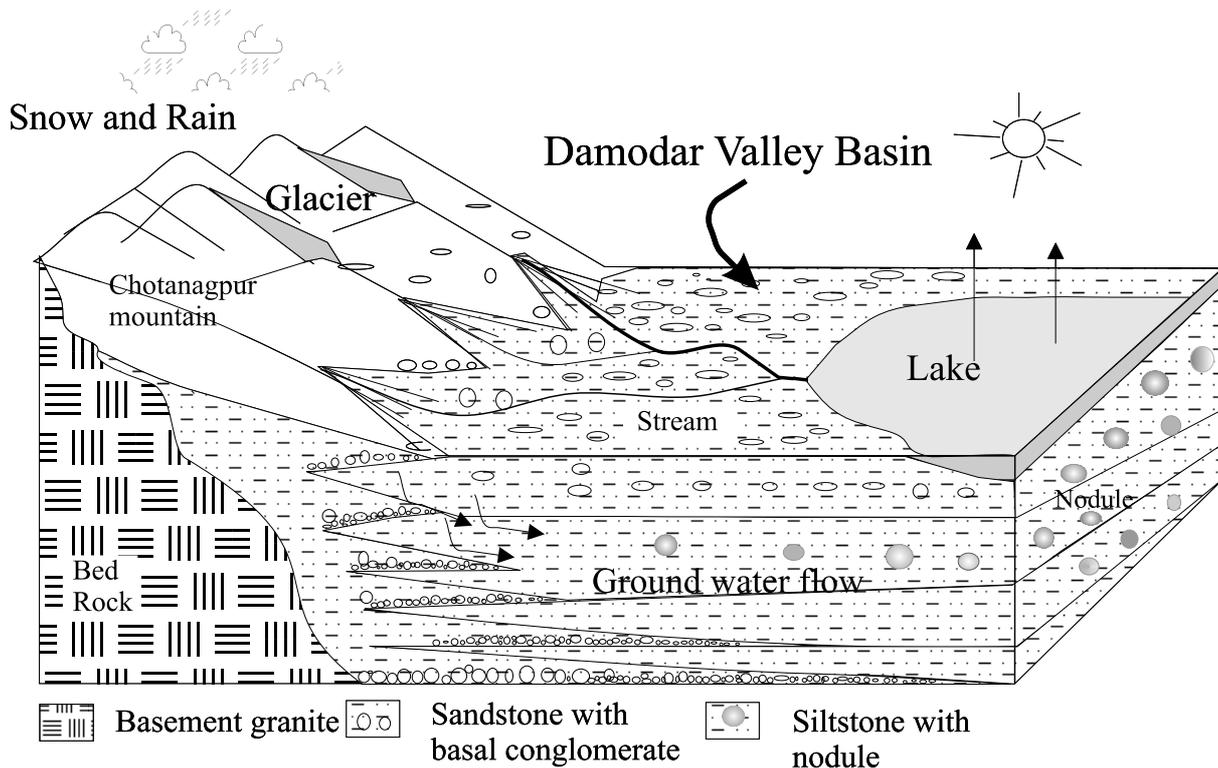
Figure 2. Schematic diagram showing the proposed environment of nodule formation based on the stable isotope analyses of carbonate phase (Bhattacharya *et al* 2001). The Chotanagpur mountain is shown as host for the glaciers draining into Damodar valley basin based on the present geochemical analysis.

and is mediated by supply of ions through pore water by localised chemical or microbial carbonate pumps (Berner 1980; Reading 1981).

3.2 *Provenance of Talchir sediments*

Mineralogical and chemical composition of sediments have been widely used to determine the provenance of detrital matter (Wronkowicz and Condie 1987; Condie *et al* 1991; Hiyashi *et al* 1997). Several factors affect the chemical composition of sedimentary rocks viz., grain size, adsorption of ions by clays, mobilization of elements during diagenesis, degree of chemical weathering of the source rocks, climate and rate of tectonic uplift, bulk composition of the source terrain, tectonic settings etc. (Wronkowicz and Condie 1987; Cullers 1988). Owing to the fine grained nature the mineralogical make-up of siltstone and shale is commonly established through their major element composition. The trace elements like REEs, Th, Sc, LILEs and HFSEs (Cr, Zr, Ni) are commonly used to characterize the source area. Being relatively insoluble in most natural waters these elements are transported with detritals from the source rocks to the site of deposition and are incorporated in the nodules during their formation (McLennan *et al* 1980; Taylor and McLennan 1985; Condie *et al* 1991). The constituent minerals comprising detritals are geochemically unaltered during diagenesis and low-grade metamorphic conditions (Bhatia 1985; Argast and Donnelly 1986; Feng and Kerrich 1990). Therefore, REE and trace element concentrations of the nodules are believed to reflect those of the source material (Ronov 1974; Taylor and McLennan 1985; Cullers *et al* 1987; and Condie *et al* 1991). Most of the sediments in the continental basins are derived from the weathering of exposed platforms. In general, such platforms inside a continent comprise of felsic (granites and gneisses) and mafic (basalt, granulites and amphibolites) rock types. Therefore, samples of these sediments from the continental region represent a mixture of felsic and mafic components and mostly display similar REE distribution pattern (Taylor and McLennan 1985; McLennan *et al* 1980). High-silica source rocks tend to contain higher concentrations of REE, Th, Hf and Ba, lower concentration of Co, Fe, Sc and Cr, and more negative Eu anomalies than the low silica source rocks (Cullers 1994). Therefore, REE concentration and their pattern in the sediments are useful tools for identification of source rock types. In addition, strontium isotope ratio of authigenic carbonate (Quade *et al* 1995) of these nodules can strengthen the provenance interpretation based on REE and trace element data.

3.3 *Rare Earth Elements*

This study is restricted to the nodules from Talchir siltstone beds containing quartz, feldspar, clays and heavy minerals with carbonate as binding cement. The discrete nodules presumably formed by cementation of the siltstone components with calcium carbonate. Mineralogically, the nodules are composed of $CaCO_3$ cement (40–50%), quartz and feldspar (10–20%) and heavy minerals with minor amount of opaques and clays. Hornblende, biotite and zircon could be identified by petrographic observation in thin sections.

The bulk of REE for sedimentary rocks resides in the clay and siltsize fraction whereas, the sand fraction comprising quartz and feldspar and the calcium carbonate cement is poor in REE. However, in some cases the sand fraction may contain heavy minerals enriched in HREE (Gd-Lu) (Cullers 1994). In addition, we note that samples collected from the same strata have different calcium carbonate content (15 to 50%) but possess remarkably similar chondrite normalized REE pattern and concentration suggesting negligible intake of REE in the carbonate phase of the nodules (Ghosh 2000). In view of the above, the REE content of the nodules can be explained in terms of admixture of clay and heavy minerals.

The REE data for 19 samples (bulk) from three sub-basins, namely, west Bokaro, Ramgarh and Talchir are given in table 1. The chondrite-normalized REE distribution patterns of the samples are plotted in figure 3 for west Bokaro, Ramgarh and Talchir basins. All three figures show LREE enrichment and a relatively flat pattern for HREE with moderate negative Eu anomaly (values range between 0.47 and 0.74).

3.4 *Sediment source in Damodar valley basin*

Palaeocurrent direction provides first approximation about the possible provenance of Talchir sediments. Casshyap and Tewari (1988) studied the clastic fabrics and clastic lithology of tillites at several places in Damodar valley basin and showed that the palaeocurrent direction of glacier flows were towards the north from Chotanagpur plateau (located south of the Damodar valley basin). This suggests that the rocks in the plateau (highlands) could be candidate for supplying the sediments of the Damodar valley basin. Chotanagpur plateau mainly comprises of granite, gneisses and small plutonic intrusions (Mazumder 1988; Sarkar 1989). Unfortunately, REE data of granites, gneisses and plutonic rocks in the Chotanagpur region are few and the values vary among samples from different positions within a granitic body (Sarkar 1989). An attempt to relate the composition of sediments

Table 1. *REE concentrations (in ppm) of the bulk fraction of the nodules from three Talchir sub-basins.*

Sample No	La	Ce	Nd	Sm	Eu	Tb	Yb	Lu	Eu/Eu*	(La/Yb)$_N$
West Bokaro sub-basin, Damodar valley basin										
SN/3A	76.1	172.4	83.0	22.5	5.0	2.8	11.2	2.2	0.72	4.6
DN/1A/95	54.8	121.0	57.8	13.4	3.3	2.3	8.1	1.0	0.74	4.6
DN/N4	74.2	138.4	71.4	23.9	5.6	5.1	19.6	3.0	0.66	2.6
DN 3A-1	67.0	128.0	68.0	14.4	2.6	1.9	6.3	0.9	0.58	7.2
DN6-1	77.1	113.2	70.1	16.1	2.7	1.8	6.6	1.0	0.54	7.9
DN/N3/95	90.7	206.1	114.3	37.8	7.1	1.8	7.5	0.9	0.69	8.2
DN/N2/95	58.9	118.1	59.6	12.8	2.6	1.7	6.1	0.8	0.64	6.5
DN-2	42.7	100.9	47.8	10.6	2.0	1.4	5.3	0.7	0.59	5.5
DN/3A/95	73.2	124.3	70.8	21.3	3.3	2.9	12.1	1.5	0.49	4.1
Mean	68.3	135.8	71.4	19.2	3.8	2.4	9.2	1.3	0.63	5.7
Ramgarh sub-basin, Damodar valley basin										
BP/N3a/95	49.6	114.3	49.5	9.9	2.2	1.3	5.0	0.6	0.70	6.8
BP/N3b/95	(195.2)	(302.4)	(121.1)	(20.0)	2.7	1.5	3.0	0.3	0.47	(43.8)
BP/N1/95	63.2	95.7	53.6	9.2	1.5	1.2	5.0	0.7	0.51	8.6
BP/N2/95	86.2	143.3	76.1	16.4	2.7	1.8	6.3	0.8	0.54	9.3
BP/M1/95	73.8	151.9	76.7	15.3	2.9	1.9	6.7	0.9	0.60	7.5
Mean	68.2	126.3	63.9	12.7	2.4	1.6	5.2	0.7	0.56	8.05
Singhbhum Granite type A	49.7	74.7	-	4.2	0.9	0.3	1.0	0.1	0.83	34.6
Singhbhum Granite type B	48.4	89.9	-	4.9	0.8	0.5	1.7	0.3	0.48	20.7
Banresar Margin	81.1	184.5	-	12.5	0.8	2.8	9.8	1.4	0.2	5.9
Banresar Centre	58.0	131.4	-	9.9	1.4	1.9	5.6	0.8	0.43	7.4
Manihari Margin	49.5	141.2	-	9.0	0.4	2.3	10.3	1.6	0.10	3.4
Manihari Centre	83.9	161.9	-	11.7	1.5	2.3	6.6	0.9	0.36	9.1
Mayur.Granite	77.0	137.7	-	10.2	0.6	1.6	6.9	1.0	0.14	8.0
Diorite Enclave	23.9	48.3	-	6.2	2.0	0.8	3.7	0.5	1.10	4.6
Composite Mixture	59.0	121.2	-	8.6	1.0	1.6	5.7	0.8	0.45	7.4
Talchir sub-basin, Mahanadi valley basin										
NDK-A1	161.3	339.6	160.3	32.0	6.3	3.0	6.4	1.2	0.66	16.9
NDK-5	48.6	112.4	64.5	11.6	2.1	1.6	4.5	0.6	0.57	7.3
NDK-1	62.4	144.2	77.1	14.1	2.5	1.9	5.6	0.7	0.57	7.6
NDJ/ST5/95	41.1	66.5	24.9	4.2	0.6	0.4	0.6	0.1	0.47	45.1
NDJ/ST2/95	56.7	103.8	58.9	10.4	1.8	1.1	3.0	0.4	0.57	12.8
Mean	74.0	153.3	77.1	14.5	2.7	1.6	4.0	0.6	0.57	17.9
Granulite (G)	27.2	60.3	26.7	5.8	1.59	1.0	3.5	0.5	0.80	5.0
Granite (gr)	171	368	160	26.2	2.33	0.4	7.2	1.0	0.80	77.1
Charnockite (ch)	49.6	101	44.9	8.2	1.51	1.1	2.3	0.3	0.60	14.1
50G:30gr:20ch	74.8	160.7	70.3	12.4	-	-	4.4	0.6	-	12.2

1. ( ) indicates values not used for calculating the mean.
2. Eu* = (2/3)*(Sm)$_N$ + (1/3)*(Tb)$_N$, where (Sm)$_N$ and (Tb)$_N$ designate chondrite-normalized value.
3. The data for granite, charnockite and granulite are from Sarkar (1989) and Paul *et al* (1990).

with the source lithology requires the definition of a mixture, having contributions from different portions of a granitic body and also from other rocks in the adjoining region. From a perusal of the available rock types occurring in the surrounding area (Sarkar 1989) and their REE data, we define a composite mixture (see table 1) taking average of the concentrations of REE in the granite suites and intrusives using data of Banresar, Manihari, Mayurbhanj, Chotanagpur (an enclave), and Singhbhum granites (phase A and B) (see table 1). The composite mixture shows an LREE enriched fractionated pattern with flat HREE and is characterized by moderate to strong negative Eu-anomaly (figure 3a). The REE pattern of west Bokaro and Ramgarh samples is similar to that of the compos-