Sr isotopes in rivers of India and Pakistan: A reconnaissance study

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It is well established that the isotopic composition of Sr in sea water has oscillated on different time scales during the past ~600 Ma. These oscillations result from changes in the relative contributions of dissolved Sr to sea water from its two major sources, continental weathering and hydrothermal interactions, with their characteristic 87Sr/86Sr. The continental input via fluvial weathering is generally more radiogenic (average 87Sr/86Sr 0.7119 ref. 2) compared to the hydrothermal 87Sr/86Sr of 0.7035 (ref. 2). The Sr flux and 87Sr/86Sr of different river systems vary widely2-4 and in some way are related to the lithology of the drainage basin, the intensity of weathering and the efficiency with which they transport their dissolved load. We have measured the dissolved Sr concentrations and 87Sr/86Sr of the major

rivers draining India and Pakistan. These measurements provide data on the flux of Sr isotopes transported by these rivers to the ocean and their impact on the Sr isotope evolution of the oceans. Further, as the river basins sampled comprise a diverse set of lithologic, tectonic and climatic regimes, they provide an opportunity to study the controls of these factors place in determining the Sr isotopic systematics of rivers.

THIS work forms a part of our geochemical reconnaissance study of the Indian rivers. Major ions, Sr and U isotopes were the principal constituents measured in the samples, the results of some of our earlier work are published elsewhere⁵⁻⁹. Figure 1 shows the sampling sites.

The rivers sampled can be conveniently classified into two groups ^{10,11} in terms of their basin geology, tectonics and climate – the peninsular and the extrapeninsular (Himalayan). The peninsular region is a tectonically quiescent shield area that has several mountain systems such as the Aravallis, the western and eastern Ghats and the Nilgiris, most of which are geologically mature. This shield is made of Pre-Cambrian rocks of diverse origin,

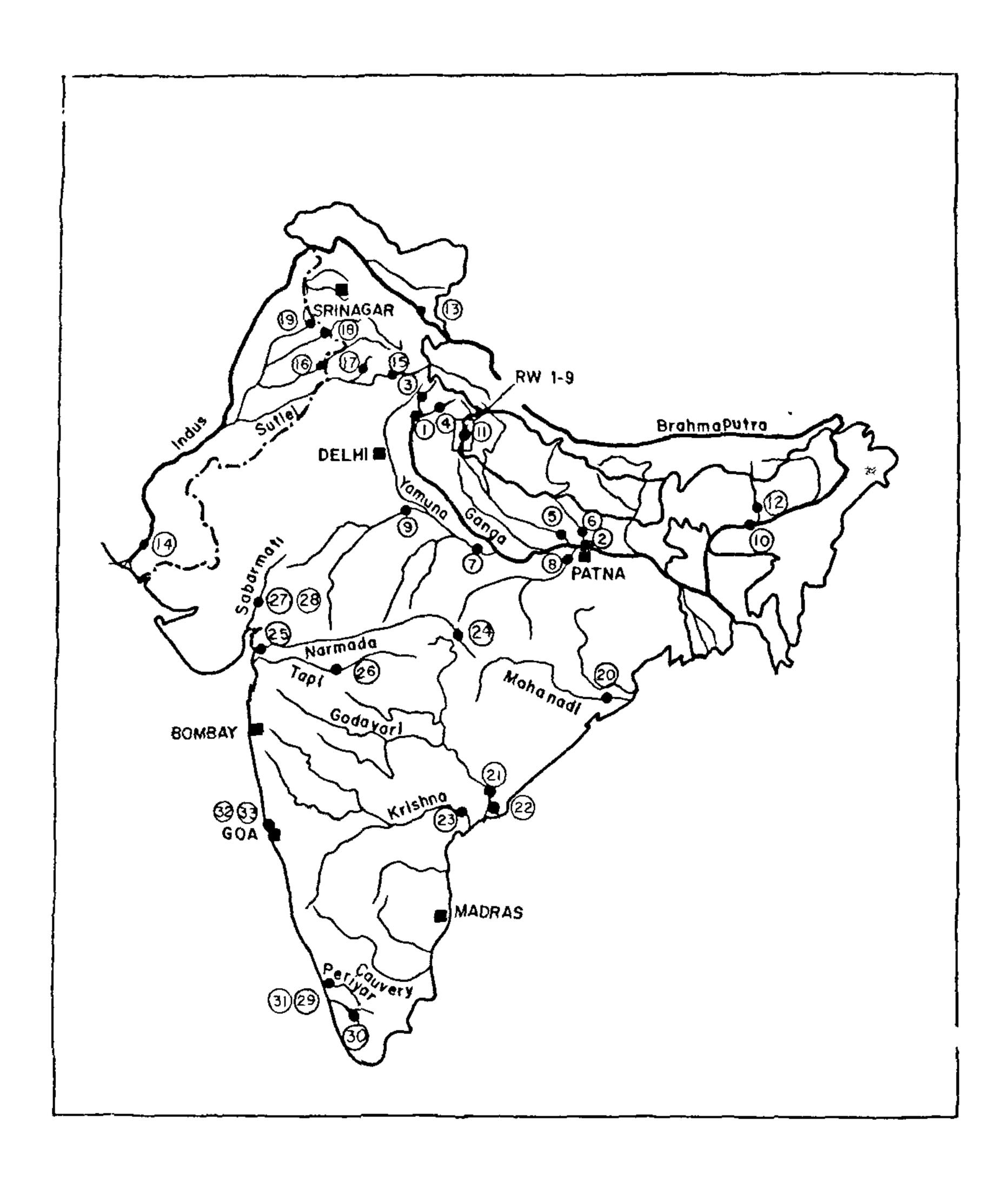


Figure 1. Sampling locations of rivers from India. The sampling location of Indus and its tributaries are given in ref 9. The locations of samples from the Kali-Sarju system (RW 1-9) are marked by rectangle.

most of which have been greatly modified and metamorphosed. These ancient rocks (gneisses, granite, charnokites, greenstones, granulites, etc.) are overlain in many areas by Pre-Cambrian and later sedimentaries (Cuddapah, Vindhyan, Delhis and Gondwanas) and by extensive sheets of lavas of the Cretaceous/Tertiary Deccan traps. Some Mesozoic and Tertiary sediments are also found along the coastal regions 10,11.

There are a number of rivers traversing the peninsular India, the larger ones being Mahanadi, Godavari, Krishna, Pennar and Cauvery, which flow eastward into the Bay of Bengal, and Narmada, Tapi, Sabarmati and Periyar, flowing westward into the Arabian sea. Banas, Luni, Chambal, Sindh, Betwa and Son are the peninsular rivers (lowland rivers⁵) belonging to the Ganga system.

Similar to the peninsular region, the extrapeninsular (Himalayan) region also contains some Pre-Cambrian rocks. These old rocks in the Lesser and Greater Himalayas are comprised predominantly of granitic gneisses and crystallines 12,13. In the Lesser Himalaya there are also extensive exposures of sedimentary sequences (carbonates, sandstones, shales). The peninsular and extrapeninsular regions are separated by the Indo-Gangetic alluvial plain consisting of massive beds of clay, sand and gravel. The clay zones of alluvial plains contain impure calcareous material in the form of irregular concretions called 'Kankar'. The calcareous material at places can be as abundant as 30%. A large part of the soils is impregnated by alkaline and saline salts⁵. In contrast to the peninsular shield area, the Himalayas are

Table 1. K, Ca, Rb and Sr isotopes in rivers from India-Pakistan

# River		Location	K Ca (μmol/l)		Rb Sr (nmol/l)		⁸⁷ Sr/ ⁸⁶ Sr
Himalayan rivers						——————————————————————————————————————	——————————————————————————————————————
_	a-Brahmaputra s	vs <i>ie</i> m					
1.	<u>-</u>	Rishikesh	38	252	20	600	0.7276
2.	Ganga	_	63	353	29	580	0.7365
2. 3	Ganga Phograthi	Patna	51	547 295	i 1	1017	0.7249
د 4.	Bhagirathi Alaknanda	Devprayag	51	385 420	54	529	0.7589
5.		Bagwan	59	429 529	40	644	0 7385
6.	Ghaghara Gandak	Ayodhya		528	23	756	0 7349
7.	Yamuna	Hajipur	84 57	649	58	847	0.7400
8.		Allahabad	57 40	547	7	1509	0.7149
9.	Son	Ara	40 53	434	6	644	0.7200
-	Chambal	Dholpur	53	829	. 5 22	2290	0.7127
10.	Brahmaputra	Goalpara	46	318	32	670	0.7197
11.	Kali	Jauljibi	39	551	70	1320	0.7276
	Indus system						
13.	Indua	Loma	34	522	41	1394	0.7104
14.	Indus	Thatta	109	957	10	3689	0.7111
15.	Sutlej	Salapar	82	622	79	1962	0.7166
16	Ravi	Lahore	71	1070	30	1858	0.7295
17.	Beas	Hanogi Mata	25	148	31	169	0.7448
18.	Chenab	Gujrawala	63	1050	32	2155	0.7218
19.	Jhelum	Lala Musa	40	950	13	2839	0.7125
Penin	sular rivers						
20	Mahanadi	Cuttak	40	303	11	704	0.7193
21.	Godavari	Dhavleswaram	59	350	24	1179	0 7163
22.	Godavari	Ravulapalam	63	400	26	1375	0.7152
23.	Krishna	Vijaywada	77	600	20	3742	0 7142
24.	Narmada	Amarkuntak	18	190	13	316	0 7144
25.	Narmada	Upstream Bharuch	46	742	6	2156	0.7114
26.	Tapi	Near Bhusaval	-	585	14	2083	0 7097
27.	Sabarmati	Kheroj	120	908	15	3358	0 717
28.	Sabarmati	Sijaria	77	1388	10	5199	0.715
29.	Periyar	Alwaye	(920)	90	36	365	0.719
30.	Pambayar	Taliawadı	28	43	35	180	0.7230
31 .	Achankovil	Pathanamthitta	26	63	26	290	0.7149
32.	Mandovi	Near Valpoi	26	242	24	352	0.719
33.	Zuari	Quepem	36	148	32	431	0.723
8.	Son	Ara	40	434	6	644	0.720
9.	Chambai	Dholpur	53	829	5	2290	0.720

Data for Ganga-Brahmaputra from ref 7 and for the Indus system from ref 9 and for the peninsular rivers this study.

Table 2. K, Ca, Rb and Sr isotopes in the Kali-Sarju system (headwaters of Ghaghra)

# Code/river		Location	K Ca (μmol/l)		Rb Sr (nmol/l)		87Sr/ ⁸⁶ Sr
RW-1	Sarju	Sheraghat	33	577	37	263	0 7585
RW-2	Ramganga	Thal	30	379	41	296	0 7757
RW-3	Ghat Gad	Near Patet	34	609	35	316	0.7508
RW-4	Kali	Jauljibi	39	551	70	1320	0.7276
RW-4 (R*)						0 7277	
RW-5	Gori	Baram	41	456	63	768	0 7490
RW-6	Sarju	Rameshwar	34	551	44	311	0 7514
RW-7	Sarju	Near Rameshwar	33	551	36	286	0 7553
RW-8	Panar	Near Rameshwar	52	545	22	1245	0 7408
R W-9	Suyai	Lohali	39	211	17	413	0 7351

^{*}Repeat analysis of RW-4.

[#]Denotes sample location in Figure 1

tectonically active, still rising and being extensively eroded by the fast-flowing rivers. Glaciers abound the higher reaches and also contribute to the accelerated weathering. The lithology of the suture zone in the Trans-Himalaya is dominated by granites and sediments^{9,11}.

Ganga, Brahmaputra and Indus are the major river systems of the extrapeninsular region, the first two flowing eastward into the Bay of Bengal and the last into the Arabian sea. Ganga and its tributaries from the Himalaya, Yamuna, Kali, Gandak, Kosi and Ghaghara, drain the Pre-Cambrian crystallines and sedimentary sequences in the southern slopes of the Himalaya, whereas Brahmaputra flows along the Indus Tsangpo suture zone in Tibet before taking a turn at the syntaxial bend and entering the crystalline terrains of the Himalaya. The main tributaries of the Indus system – Sutlej, Beas, Ravi, Chenab (Chandra and Bhaga) and Jhelum – drain the crystalline terrains of the Himalaya in their upper reaches (similar to Ganga and its tributaries),

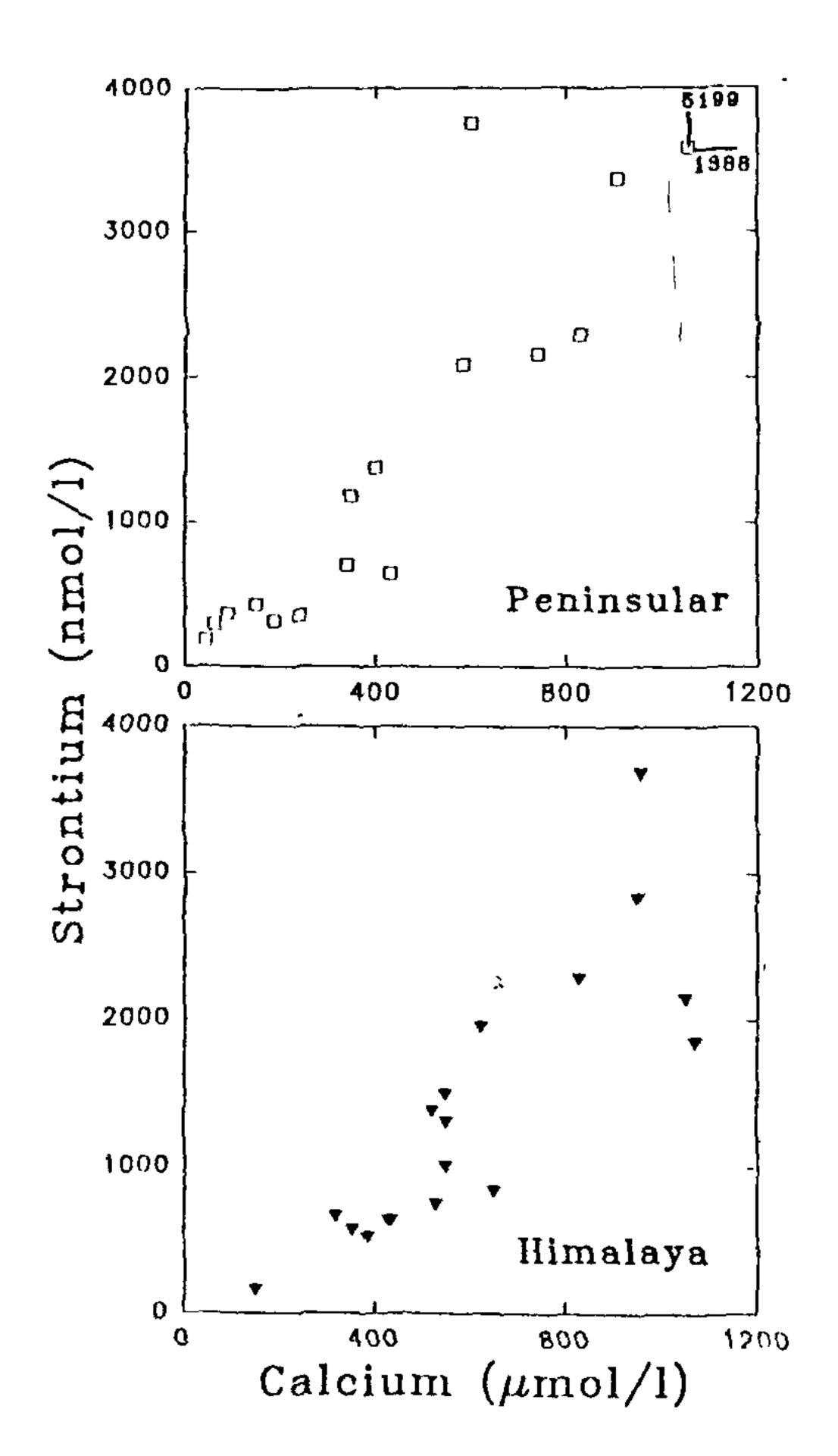


Figure 2. Variation of Sr concentration with calcium in the peninsular and Himalayan rivers. The Sr concentrations generally increase with Ca.

whereas Indus and its northern tributaries, Nubra and Shyok, drain a suture zone lithology in a monsoon shadow region⁹.

The peninsular rivers are seasonal (monsoonal) in contrast to the Himalayan ones, which are fed by glaciers and are perennial, though their water flux increases considerably during monsoon.

Most of the rivers were sampled during the past several years. The sampling of the Kali-Sarju system (headwaters of the Ghaghra) was carried out recently. The sampling procedures are discussed in detail elsewhere. Briefly, the river waters were filtered through 0.4 µm Nuclepore filters within 5-6 h of collection, acidified to pH-2 with HNO₃ and stored in polyethylene bottles till the time of measurement. Rb and Sr isotope measurements were made in about 50 g of water following isotope dilution mass spectrometric procedures¹⁴. The weighed water samples were dried with HF/HNO₃, put in HC1 and spiked with 87Rb and 84Sr tracers, and dried. The dried residue was taken in 2.5 N HCl and the Rb and Sr fractions were separated on a cation exchange resin (Dowex 50×8, 200-400 mesh). The Rb and Sr fractions were separately dried, taken in a drop of orthophosphoric acid and evaporated directly onto an outgassed tantalum filament of the ion source. The samples were run on a 9" radius single-focusing mass spectrometer fitted with a Faraday cup collector and the ion current was recorded digitally.

The Sr isotopic ratios were normalized ⁸⁶Sr/⁸⁸Sr = 0.1194. The long-term stability of the mass spectrometer was checked by measuring the ratio 87Sr/86Sr of the NBS-987 standard periodically. The mean 87Sr/86Sr of the standard based on nine runs during 1991-93 is 0.71025 ± 0.00005 . In addition, to check on the accuracy of 87Sr/86Sr measurements we analysed a sea water sample from the Arabian sea and CaCO3 from a living coral from the Lakshadweep. For these two samples ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ are 0.7097 ± 0.0006 and $0.7092 \pm$ 0.0002, respectively, in good agreement with contemporary sea water values. The total Rb and Sr blanks from reagents and chemical procedures are ~5 and ~50 pmol, respectively, much lower than the typical values of about 1 and 25 nmol for Rb and Sr in 50 g water samples. The overall precision on 87Sr/86Sr (two standard errors of the mean of about 50 ratio measurements) is about ± 0.0005. These errors are considerably smaller than the scale of Sr isotope variations observed in the river waters analysed. Ca and K measurements were made by atomic absorption spectrophotometry.

The dissolved Rb and Sr concentrations and ⁸⁷Sr/⁸⁶Sr of the water samples analysed are given in Table 1 along with their K and Ca abundances. Table 2 lists the same for the Kali-Sarju system (also known as Mahakali-Sarju) headwaters of Ghaghara, a major tributary to the Ganga. More detailed data on Sr isotopes, Rb and major

ions for the rivers belonging to the Ganga-Brahmaputra and Indus systems are published elsewhere^{7,9}.

The Kali-Sarju waters were sampled to test if ⁸⁷Sr/⁸⁶Sr for the headwaters of major tributaries of Ganga, are as radiogenic as of Bhagirathi and the Alaknanda, sampled by us earlier⁷. The results show that the Sr isotope ratios in all these waters are highly radiogenic, and as will be discussed later, such high values probably result from weathering of rich in radiogenic Sr silicates in the drainage basin.

A cursory look at the data in Table 1 reveals that the Sr concentration in the samples ranges between 169 and 5199 nmol/1, similar to that observed for rivers in other parts of the world²⁻⁴.

The Sr concentration shows a general increase with Ca (Figure 2), the correlation being relatively tight for rivers from the peninsular region. The Sr/Ca molar ratios (nmol Sr/µmol Ca) of the peninsular and the Himalayan rivers overlap within the scatter of the data. The mean molar Sr/Ca ratio corresponds to a Sr/Ca weight ratio (g/g) of $\sim 7 \times 10^{-3}$ and $\sim 5 \times 10^{-3}$ for the peninsular and the Himalayan rivers, respectively. The Sr/Ca ratio in rivers is determined by the supply of Sr and Ca to the waters by various rocks and minerals, and their mixing proportions. An important source of Ca to these waters (Table 1) is the weathering of carbonates⁵⁻⁹. The Sr/Ca weight ratio in typical carbonates is $1-2 \times 10^{-3}$ (ref. 15). Congruent weathering of carbonates (for Ca and Sr) would yield Sr/Ca ratio in river waters significantly lower than that we have measured. Such an observation (Figure 2) would require that either Sr is mobilized preferentially over Ca from carbonates such that the Sr/Ca ratio in rivers is higher than that in carbonates or that these rivers receive Sr from other sources such as evaporites and silicates from the upper crust, which have Sr/Ca higher than that in typical carbonates. Basalts, granites and shales have Sr/Ca about 3 to 10 times higher than that in carbonates. As discussed below, 87Sr/86Sr for these river waters also requires that they derive some of the Sr from a more radiogenic source than typical carbonates, such as silicates of the upper crust.

lies in the range 0.7097–0.7589; almost all samples have ⁸⁷Sr/⁸⁶Sr exceeding the global average estimate of 0.7119 (ref. 2). The data also show that ⁸⁷Sr/⁸⁶Sr for the peninsular rivers falls within a narrow range of 0.71–0.72 and is generally lower than that for the Himalayan rivers draining the granite/gneiss crystallines. The contrast is more pronounced when the comparison is made with that for the headwaters of Ganga and Ghaghara (Table 2, Figure 3). All the headwater samples analysed, except Kali, have ⁸⁷Sr/⁸⁶Sr exceeding 0.73, with Sr concentration in the range of 140–1390 nmol/1 (Table 2, ref. 7)

⁸⁷Sr/⁸⁶Sr of dissolved Sr in rivers is governed by the magnitude of Sr contribution from various source rocks

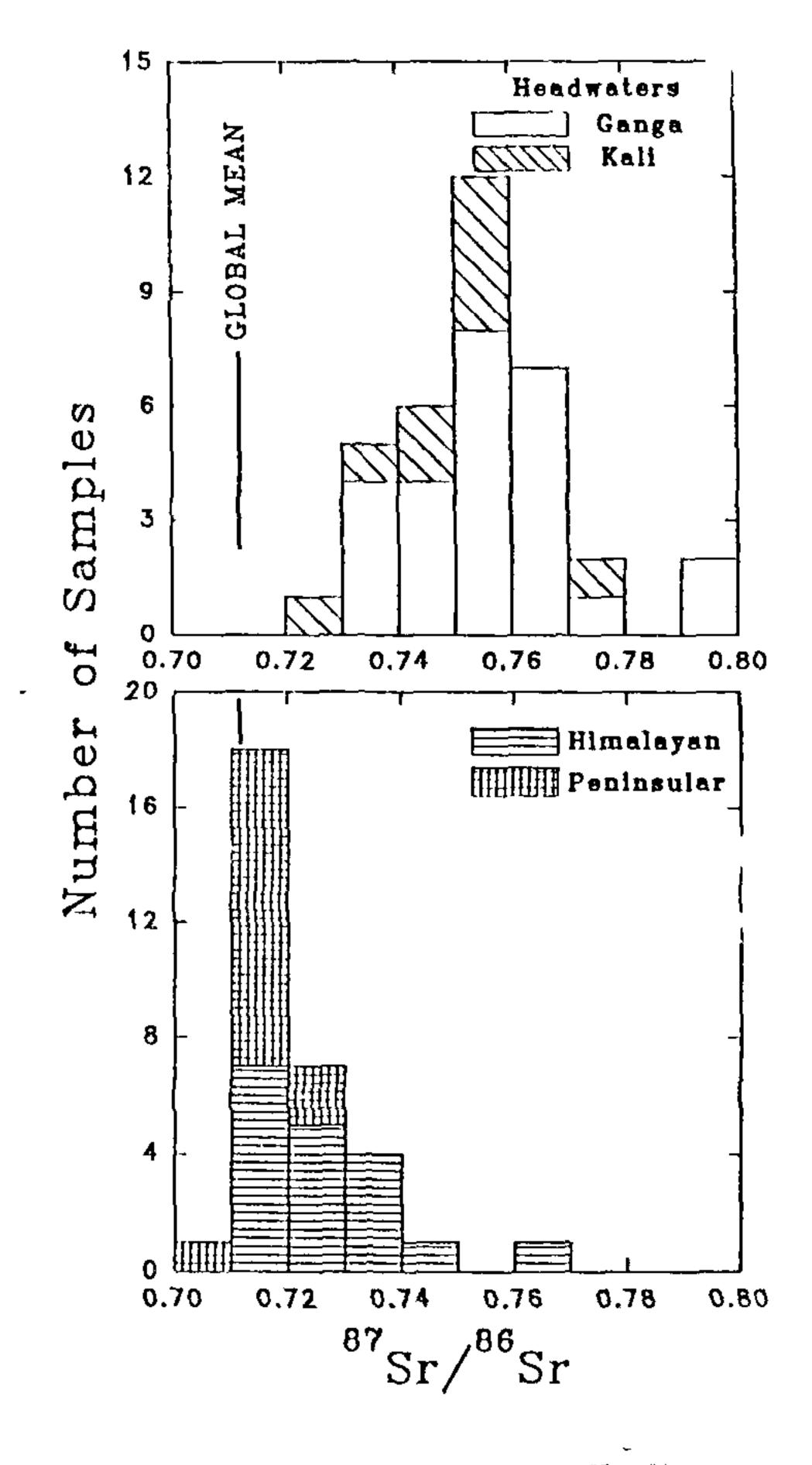


Figure 3. Frequency distribution of dissolved ⁸⁷Sr/⁸⁶Sr in the rivers sampled Data for Ganga headwaters from ref. 7 and Kali River (this study) For comparison the data for Himalayan and peninsular rivers from Table 1 are also shown. The distinctly higher radiogenic ⁸⁷Sr/⁸⁶Sr of the headwaters is evident.

(e.g. carbonates, silicates, evaporites) to the effluent waters and their respective $^{87}\text{Sr}/^{86}\text{Sr}$. For typical carbonates and evaporites this ratio is constrained within a narrow range of 0.706-0.709, whereas that for silicates it can vary widely both within and among various drainage basins. Basaltic terrains set a lower limit on $^{87}\text{Sr}/^{86}\text{Sr}$ for crustal silicates with values of ~ 0.705 . The upper limit on $^{87}\text{Sr}/^{86}\text{Sr}$ in silicates is less constrained, though values exceeding 1.0 are encountered in individual rock suites, such high values are unlikely to be representative of river basins as a whole.

For all the river waters sampled in this study (Tables 1 and 2) 87Sr/80Sr is higher than that for typical marine carbonates, suggesting that at least a part of the Sr in

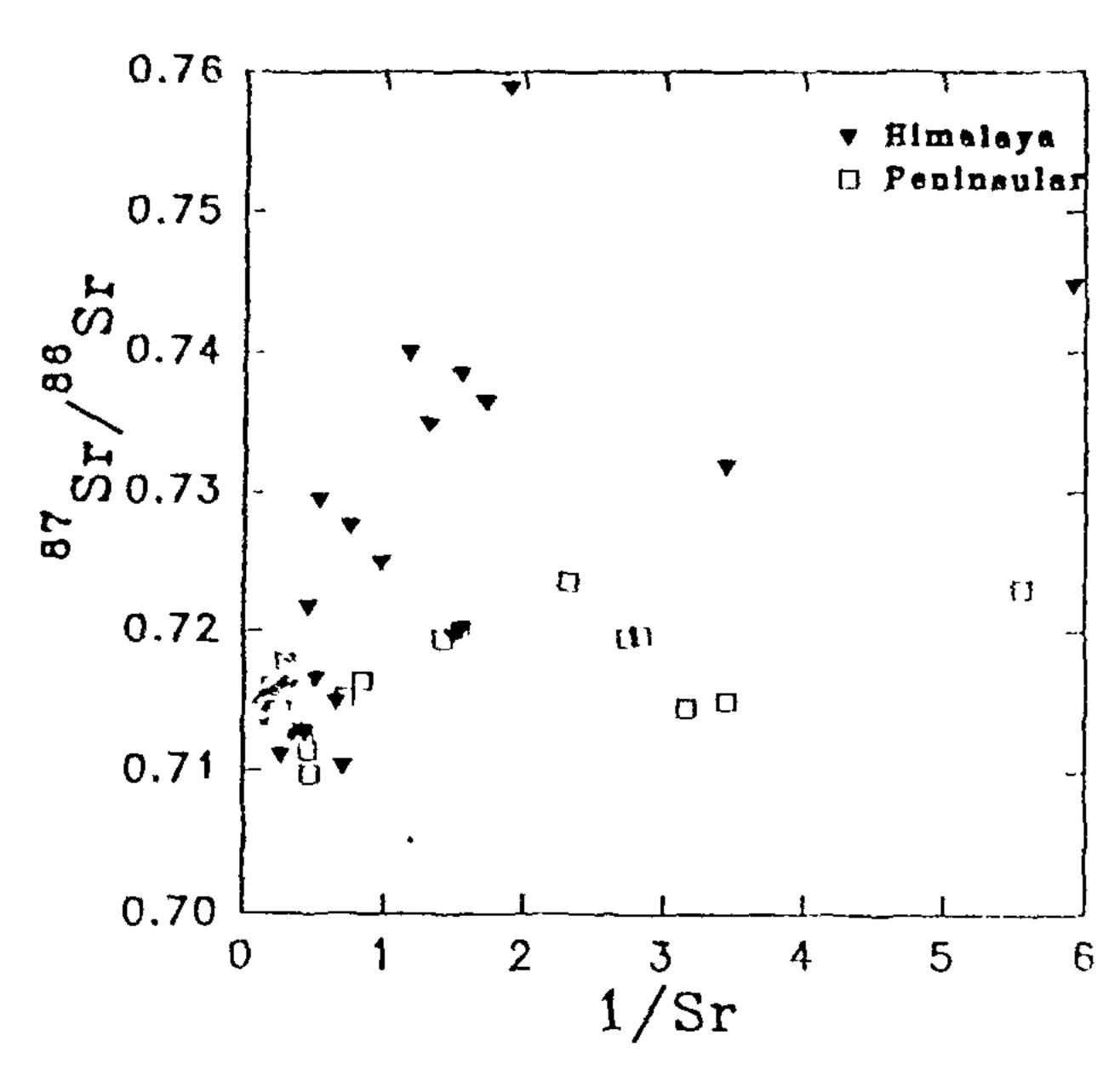


Figure 4. Sr isotope systematics in the rivers sampled. The *7Sr/**Sr increases sharply with decreasing Sr concentration in the Himalayan rivers. The penninsular rivers also show a similar trend, but with a gentler slope

these rivers is derived from a source having more radiogenic 87Sr/86Sr. The plot of 87Sr/86Sr versus 1/Sr (Figure 4) reveals that for the Himalayan rivers there is a steep increase in 87Sr/86Sr as the Sr concentration decreases. The peninsular rivers also seem to show an increase in 87Sr/86Sr as Sr decreases, but with a much gentler slope. The broad trend in Figure 4 can be interpreted in terms of mixing of Sr between a high 87Sr/86Sr end member with a low 87Sr/86Sr component (e.g. typical carbonates/evaporites). The relation between 87Sr/86Sr and Sr concentration for the Himalayan rivers, particularly those belonging to the Ganga system, are distinctly different from other global rivers, in accordance with our earlier observations and other studies^{2,7,9,16,17}. Many of these rivers are characterized by high 87Sr/86Sr and high Sr concentrations (Figure 4). The source of such high 87Sr/86Sr in these rivers is not clear. Weathering of Pre-Cambrian granites and gneisses^{7,9} and rocks and minerals enriched in radiogenic Sr due to regional metamorphism, such as the leucogranites and gneisses 16 and carbonates¹⁷, have been suggested as possible sources of high 87Sr/86Sr in these rivers. Studies 18 of 87Sr/86Sr in sediments from the Bay of Bengal indicate that the dominant source of these sediments for the past ~17 Ma is the higher Himalayan crystallines. Available data from the drainage basin of the Ganga headwaters and its tributaries show that whole-rock 87Sr/86Sr are quite radiogenic, with the bulk of the samples clustering around 0.76 and a significant fraction of the samples

having values in excess of 0.90 (Figure 5). Such high ⁸⁷Sr/⁸⁶Sr in the silicates can account for the high ⁸⁷Sr/⁸⁶Sr in the headwaters of the Ganga system. However, data on Sr isotope composition of carbonates from the drainage basin are needed to confirm this. Whatever be the source of ⁸⁷Sr/⁸⁶Sr to Ganga, its high value has considerably influenced the Sr isotope evolution of the oceans since the Cenozoic⁷.

The data in Figure 4 show an overall increasing trend in ⁸⁷Sr/⁸⁶Sr with decreasing Sr, but there is a significant scatter. The scatter within each trend probably reflects heterogenity in the Sr isotope composition of the silicate end member. Such a heterogenity can arise as the Himalayan and the peninsular regions are large geographical areas having lithologically distinct sub-basins which the different rivers drain. The wide range of ⁸⁷Sr/⁸⁶Sr for the silicates from the drainage basin of the Ganga system headwaters (Figure 5) attests to this hypothesis.

The Sr isotope composition of the peninsular rivers is similar to that reported for rivers from other continental shield areas. The source of high radiogenic Sr to Zuari and Mandovi rivers could be the granites and gneisses in their basin, some of which have 87Sr/86Sr of 0.713-1.13 (ref. 19). Periyar and Pambayar flow mainly through granulites; the Sr isotopic analysis on these rocks are meagre, and those which are available are in the range 0.78-0.89 (refs. 20 and 21). Mahanadi and Godavari traverse through Deccan trap basalts, unmetamorphosed sedimentary sequences and the high-grade metamorphic terrain of the eastern Ghats. The measured 87Sr/86Sr for these river samples integrates the contributions from all these lithologies. Tapi, flowing mainly through the Deccan trap basalts, has the lowest 87Sr/86Sr among all the samples (Table 1).

Based on the Sr concentration and 87Sr/86Sr in the peninsular rivers (Table 1) and those reported earlier by us^{7,9} for Ganga, Brahmaputra and Indus, the dischargeweighted mean Sr concentration in these rivers is calculated to be $\sim 1.5 \,\mu\text{mol/l}$, with a $^{87}\text{Sr/}^{86}\text{Sr}$ of 0.716. The mean Sr concentration in the rivers of India-Pakistan is higher than the global average value of $0.89 \,\mu \text{mol/l}^{-2}$; our sampling of some of the rivers (e.g. Indus) during their lean flow may be a likely cause for the high mean concentration. The discharge-weighted Sr/Ca molar ratio of these rivers is 2.7×10^{-3} , which compares with a value of $\sim 2.2 \times 10^3$ for the world rivers (based on Sr and Ca concentrations of 0.89 and 398 µmol/l, respectively). The annual flux of Sr transported by the rivers from India-Pakistan to the ocean is ~2200 million moles if our measurements can be considered representative of these rivers. These calculations show that the rivers from lndia-Pakistan account for ~6.5% of the global riverine Sr flux to the oceans, with a 87Sr/86Sr marginally more radiogenic than the global average.

The influence of anthropogenic effects in modifying the natural Sr flux and its isotopic composition is diffi-

Table 3.	Rb, Sr concentrations an	d 87 Sr/86 Sr in fertilizers
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Code	Type	Source	Rb (ppm)	Sr (ppm)	⁸⁷ Sr/ ⁸⁶ Sr*
NPK	Potassium ammonium phosphate	RAMBAN Co. (Dehradun)	24	15	0.7087
MOP	Muriate of potash	~-	122	6 4	0 7494
DAP	Ammonium phosphate	SPIC (Dehradun)	2 0	130	0.7080
GUUREA	Urea	KRIBHCO (Ahmedabad)	3 1	0 05	-
NBPHOS	Ammonium phosphate	NARMADA Fertilizers (Ahmedabad)	1 3	554	0 7080
GUJDAP	Ammonium phosphate	IFFCO	3 6	12	0 7091

^{*}Error ($\pm 2\sigma$) is ~0 0005

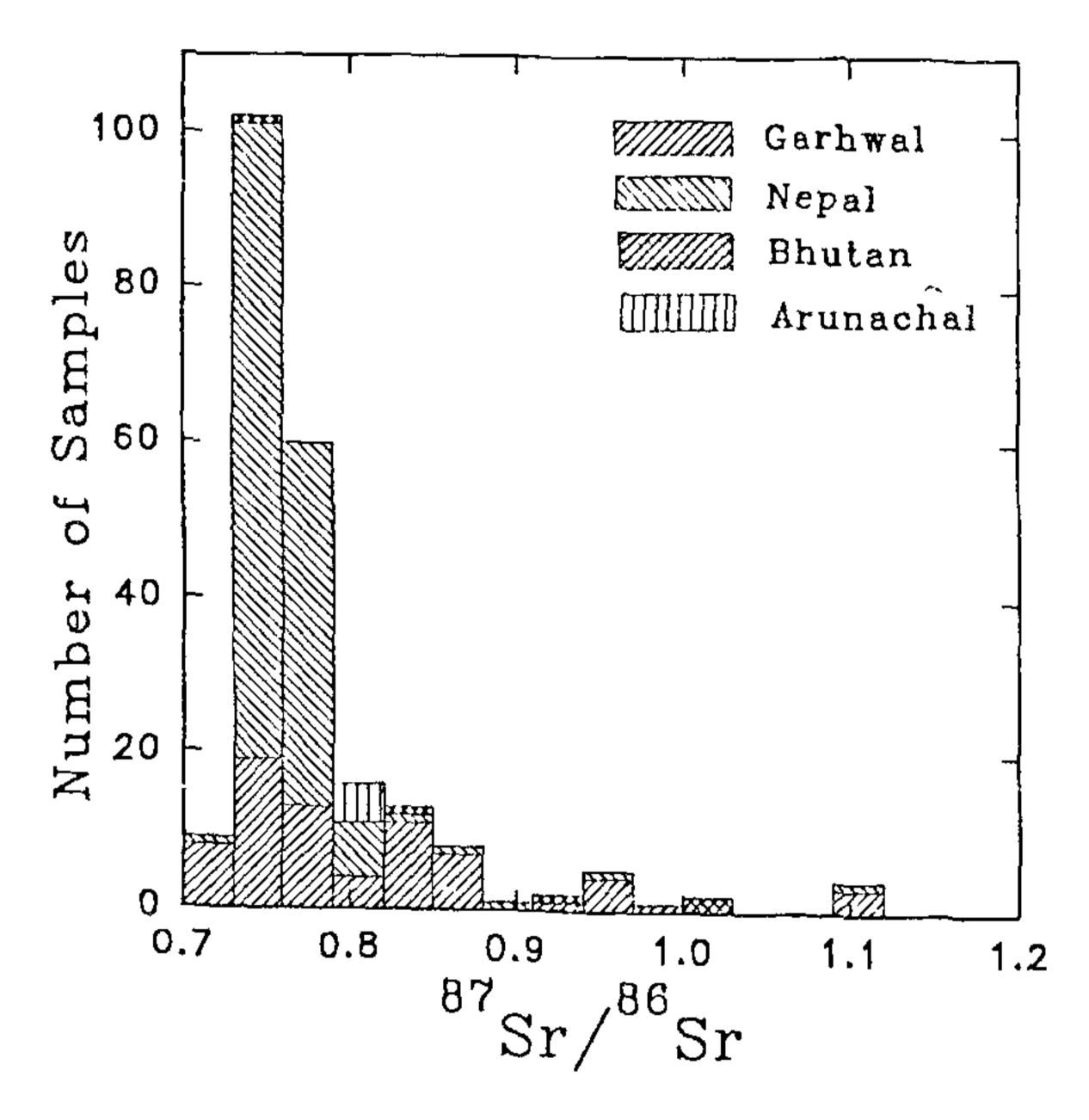


Figure 5. Frequency distribution of *7Sr/*6Sr in whole-rock samples from the drainage basins of the headwaters of Ganga and its tributaries (Garhwal and Nepal) and Brahmaputra (Bhutan and Arunachal) Data compiled from refs 14, 23-32. The rocks from this region have highly radiogenic *7Sr/*6Sr.

cult to assess. Damming and impounding of rivers may reduce their water discharge, whereas the use of fertilizers may alter the natural Sr concentration of rivers and their ⁸⁷Sr/⁸⁶Sr. We measured the Sr concentration and ⁸⁷Sr/⁸⁶Sr in a few fertilizer samples used in northern and western India. The results (Table 3) show that all the samples, except for the muriate of potash (MOP), have ⁸⁷Sr/⁸⁶Sr of ~0 708, consistent with that expected, as bulk of Sr in fertilizers is derived from limestones and phosphates of marine origin. The muriate of potash, which is abundant in potassium and rubidium, has a

highly radiogenic ⁸⁷Sr/⁸⁶Sr of ~0.749 (Table 3). This high value raises concern about the impact such fertilizers can have in enhancing the natural ⁸⁷Sr/⁸⁶Sr of rivers. Our earlier observation⁷ that ⁸⁷Sr/⁸⁶Sr for Ganga and many of its tributaries varies only marginally during different river stages is an indication that contamination, if any, of natural Sr by fertilizer may not be significant. Moreover, the most radiogenic Sr isotope composition is in the headwaters of Ganga and Ghaghara, where agricultural influences are minimal.

Summing up, our results on the Sr isotope systematics of rivers from India and Pakistan show that almost all the rivers sampled have ⁸⁷Sr/⁸⁶Sr in excess of the global average, with the headwaters of Ganga and its tributaries having the most radiogenic Sr isotope composition. Studies on the source of radiogenic Sr to the Ganga system (i.e. silicates or metamorphosed carbonates) would help in understanding better the coupling between Himalayan tectonics, weathering and climate change²².

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