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Strontium isotopes and major ion chemistry in the Chambal River system, India: Implications to silicate erosion rates of the Ganga

R. Rengarajan *, Sunil K. Singh, M.M. Sarin, S. Krishnaswami

Planetary & Geosciences Division, Physical Research Laboratory, Navrangpura, Ahmedabad 380 009, India

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The dissolved concentrations of major ions and Sr isotopes ($^{87}Sr/^{86}Sr$) were measured in the headwaters of the Chambal river and its tributaries draining the Deccan Trap basalts and the Vindhyan sediments of peninsular India. The total dissolved solids (TDS) ranged from 181 to 547 mg L^{-1}; much higher than the global "mean" river water. A significant fraction of solute abundance in the Chambal river is derived from sodium salts, unlike the Himalayan rivers which exhibit dominance of (Ca+Mg) salts. It is estimated that the Chambal river supplies about one-third of sodium via the Yamuna to the Ganga at Rajshahi (Bangladesh), with only ~6.5% of water discharge. The presence of Na salts not associated with chloride in the Chambal headwaters constraints the application of Na* (Na corrected for Cl) as an index of silicate derived component. This finding brings out the need to revisit the estimates of silicate erosion rate (SER) and associated CO_2 consumption in the Ganga basin, downstream Allahabad, based on Na* as an index.

The Sr concentration in the Chambal tributaries varied from 1.9 µM to 5.9 µM and $^{87}Sr/^{86}Sr$ ratio from 0.70923 to 0.71219. Unlike the Himalayan Rivers, Sr isotope composition in the Chambal river is far less radiogenic as the major sources of Sr to the Chambal are the Deccan Trap basalts and the Vindhyan sediments, which are low in $^{87}Sr/^{86}Sr$. The Sr isotope budget of the Ganga, based on available data of the Chambal, Betwa, Ken, Yamuna and the Ganga shows that, weathering of the Deccan Trap basalts and the Vindhyan sediments (the drainage basin of the Chambal, Betwa and the Ken) contribute ~70% of the dissolved Sr to the Ganga at Varanasi. This study highlights the key role of peninsular rivers draining the Deccan and the Vindhyan regions in the major ion and Sr budget of the Ganga.

1. Introduction

The chemical composition of rivers and fluxes of elements they transport to oceans depend on a number of factors that include lithology of the basins, runoff, relief, vegetation and temperature (Berner and Berner, 1996; Meybeck, 2005). These parameters differ significantly among various river basins, which make it necessary to study rivers from widely different regions, characterized by diverse physical and environmental factors, to obtain global mean river water composition and representative values of various fluxes. Among the major global rivers studied during the past few decades, the Ganga and the Brahmaputra draining the southern Himalaya are two of the river systems that have been investigated in detail for their chemical and isotopic composition (particularly $^{87}Sr/^{86}Sr$) to quantify chemical erosion rates of the region (Palmer and Edmond, 1992; Krishnaswami et al., 1992, 1999; Galy and France-Lanord, 1999, 2001; Dalai et al., 2002; Bickle et al., 2005; Singh et al., 2005, 2006; Tipper et al., 2006; Hren et al., 2007; Rai and Singh, 2007). These studies have been prompted by two main considerations (i) these rivers are two of the large river systems of the world draining the Himalaya. They transport significant fraction of global river water and sediments to oceans (Milliman and Syvitski, 1992; Hay, 1998) which makes their studies important from the perspective of marine elemental and isotopic budgets and (ii) to assess the role of silicate weathering and associated CO_2 draw-down in these basins as a potential driver of long-term global change (Walker et al., 1981; Raymo and Ruddiman, 1992; Ruddiman, 1997).

The Ganga is one of the large rivers draining the Himalaya and the alluvial plains of India. It has many tributaries, most of which originate in the Himalaya and merge with it in the plains. Among the various tributaries, the Yamuna is the largest in terms of drainage area (Rao, 1975; Jain et al., 2007) with most of its drainage in peninsular India and the Ganga plain. The Yamuna has a number of tributaries joining it both in the Himalaya and in the plains (Fig. 1a). Among these, the tributaries merging in the plain, the Chambal, Betwa and the Ken (Fig. 1a) are relatively larger both in terms of water discharge and basin area. Studies of these tributaries can yield data to evaluate the role of chemical erosion in the peninsular regions and the Ganga plain in contributing to the overall chemistry of the Ganga and to the elemental fluxes transported by it. Further, as these tributaries predominantly drain the Deccan Traps and the Vindhyan system in peninsular India

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* Corresponding author. Tel.: +91 79 26314314; fax: +91 79 26301502.
E-mail address: rajan@prl.res.in (R. Rengarajan).

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(Krishnan, 1982), their chemistry would be dictated by weathering of these lithologies unlike their Himalayan counterparts whose chemistry is known to be dominated by weathering of granites/gneisses and carbonates of the Tethyan Sedimentary Series and the Lesser Himalaya (Valdiya, 1980; Sarin et al., 1989; Krishnaswami et al., 1992; Galy and France-Lanord, 1999; Krishnaswami et al., 1999; Dalai et al., 2002; Bickle et al., 2005). Thus, a comparative study of these two groups of rivers can provide insight into the role of lithology in determining their water chemistry.

In this work, study of major ion composition and ^{87}Sr/^{86}Sr of the Chambal river and its tributaries has been carried out. The Chambal is the largest tributary of the Yamuna, with its entire drainage in peninsular India. The focus of this study is (i) to characterize the water chemistry of the Chambal in terms of supply from various sources and to compare it with the chemistry of the Yamuna headwaters draining the Himalaya (Dalai et al., 2002) and (ii) to evaluate the contribution of dissolved major elements, Sr and ^{87}Sr/^{86}Sr from the Chambal to the Yamuna and finally to the Ganga. These studies, as mentioned earlier can provide a measure of the impact of chemical weathering in peninsular India not only in contributing to elemental and isotope chemistry of the Ganga, but also to the global riverine flux to the oceans.

2. Geologic setting of the Chambal river system

The Chambal (Fig. 1a) rises in the Vindhyan range, near Mhow, at an elevation of 354 m (Rengarajan, 2004). The Banas, Kali Sindh, Shipra, Newaj and the Parbati are its major tributaries (Fig. 1b). All these rivers are fed entirely by rain during the SW monsoon (June–September, http://www.grdc.sr.unh.edu) with the result that many of these rivers shrink to rivulets during summer. A broad lithological map of the Chambal basin along with sampling locations is shown in Fig. 1b. The Chambal basin lies between 22° 27' to 27° 20' N and 74° 30' to 79° 20' E and has a drainage area of ~1.4×10^5 km^2 (http://www.grdc.sr.unh.edu). The headwaters of the Chambal, Kali Sindh, Shipra and the Newaj all flow through theoliitic lava flows of the Deccan Traps, which cover significant area of the basin (Fig. 1b). Elemental composition of basalts from the Mhow area (Table A1; Peng et al., 1998), the source region of the Chambal (Fig. 1b), overlap within errors with that of average Deccan Trap basalts. The ^{87}Sr/^{86}Sr of the Mhow region basalts ranges from 0.70585 to 0.71610, suggesting significant levels of crustal contamination (Peng et al., 1998; references there in). At several locations the Chambal system rivers incise the Deccan Trap basalts to expose the Vindhyan and the Aravalli systems in the valleys. The Vindhyan system is dominated by shallow marine deposits of neoproterozoic to paleoproterozoic age (Ray et al., 2002, 2003). Outcrops of granites, sandstones, limestone and dolomites belonging to the Aravalli and the Vindhyan screen the Deccan Traps in the Chambal basin (Valdiya et al., 1982). The northern part of the Chambal mainstream and some of its tributaries, the Parbati and the Kali Sindh drain the Vindhyan system which forms a prominent plateau-like range of sandstones and carbonates (Krishnan, 1982). These carbonates composed both of calcites and dolomites (Ghosh, 1976; Ray et al., 2003) are widely exposed in the Chambal drainage, particularly in the basin of the Banas, and the lower reaches of the Chambal, Kali Sindh, Prabati and their tributaries (Fig. 1b). In addition to the Deccan Traps and the Vindhyans, Quaternary deposits comprising mainly of alluvium occur in the river valleys (Fig. 1b). Black soils and laterites formed by chemical weathering of basalts under semi-arid climate occur in the Deccan plateau part of the drainage (Landey et al., 1982; Bhargava and Bhattacharjee, 1982). Further, a significant part of the Chambal basin is impregnated with alkaline and saline salts (Fig. 1c) analogous to that in the Ganga plain (Bhumbla, 1975; Sarin et al., 1989). The spatial coverage of these alkaline/saline soils in the Chambal basin (CSSRI, 2007; NRSA, 2008) show that they are dispersed all along the course of various rivers sampled (Fig. 1c). Investigations on alkaline/saline soils mainly from the Ganga plain (Gupta, 1968; Bhargava and Bhattacharjee, 1982; Pal et al., 2003) suggest that the source of their alkalinity and salinity is dissolved ions supplied by rivers and shallow

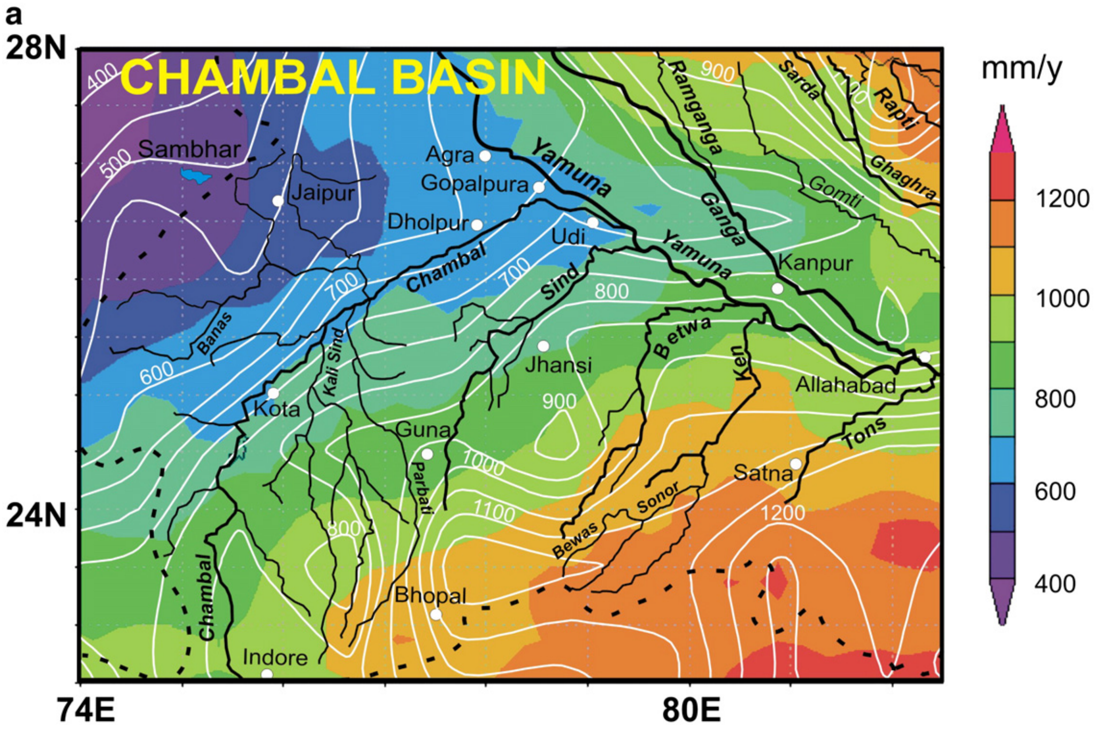

Fig. 1. (a) Map of the Chambal basin along with rainfall contours. The Ganga, Yamuna, Betwa and the Ken are also shown. Rainfall data from (http://disc2.nascom.nasa.gov/Giovanni/tovas/TRMM_V6.3B43.shtml) and IITM. (b) Simplified lithological map of the Chambal river catchment with sampling locations. The sampling locations are indicated as numbers enclosed in circles. (c) Distribution of saline/alkaline soils in the Chambal basin (Source: CSSRI, 2007; NRSA, 2008). The ellipses in the figure are based on CSSRI (2007) and the irregular shapes are based on NRSA (2008).

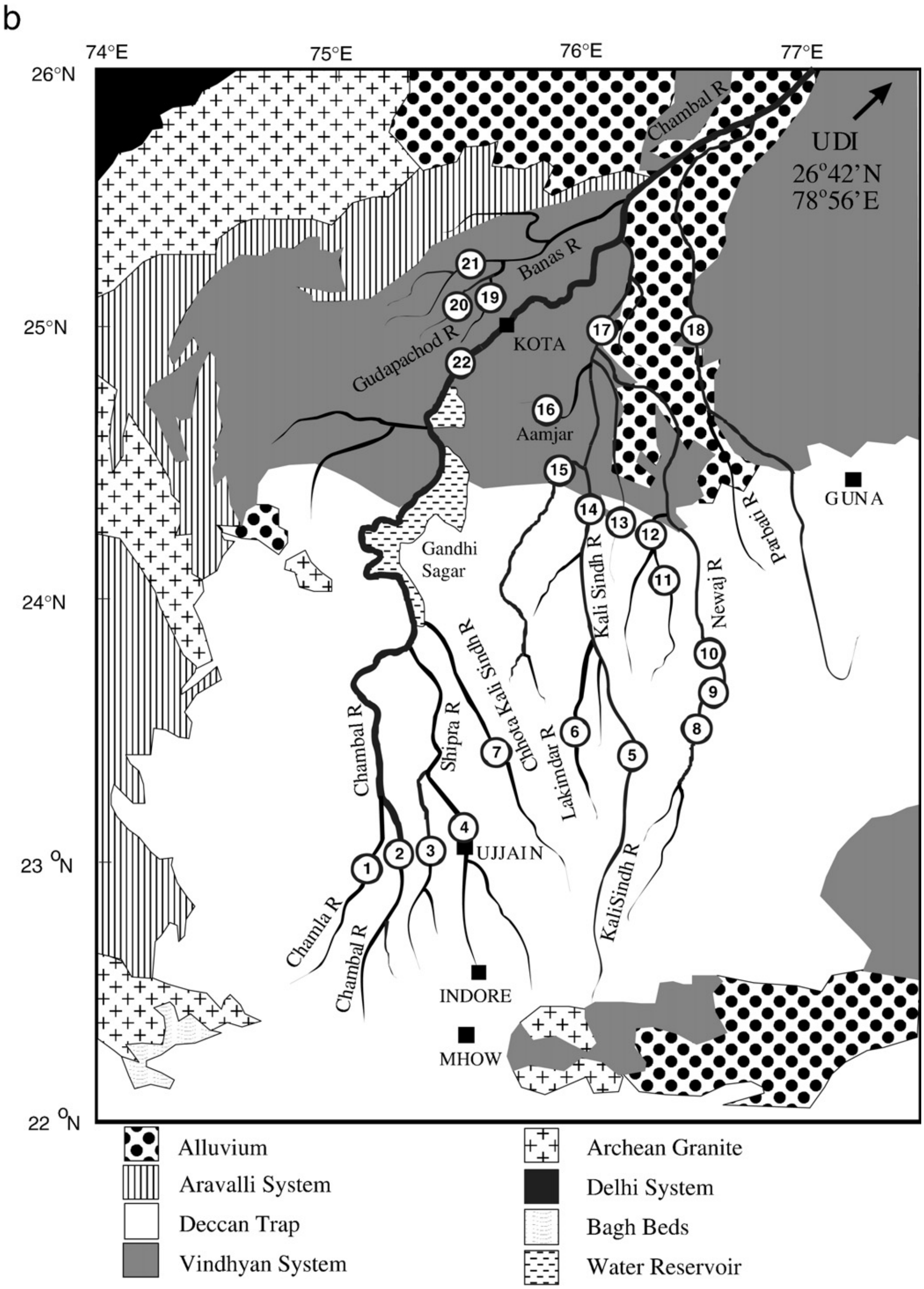

Fig. 1 (*continued*).

ground waters. These dissolved weathering products get trapped in the poorly drained endoreic areas of the drainage basins and evaporate during summer causing the deposition of various salts, some of which (NaCl, Na_2SO_4, $NaHCO_3$ and Na_2CO_3) contribute to the alkalinity and salinity of the soils (Bhargava and Bhattacharjee, 1982; Pal et al., 1994; Datta et al., 2002). This inference is also attested by studies on the aqueous extracts of saline/alkaline soils from the Ganga plain, which yield high concentrations of leachable Na, Cl, SO_4 and alkalinity (Table A2; Gupta, 1968; Bhargava and Bhattacharjee, 1982; Pal et al., 1994). Thus, repeated cycles of wetting and drying in the basin results in salinisation and alkalisation of the soils in the region. The excessive use of surface and ground waters for irrigation during the recent past has further enhanced the salinisation of the basin. In addition, these wetting and drying cycles also promote the formation and deposition of calcium carbonate in the soils. These carbonates locally known as "kankar" are generally made of calcium carbonate though magnesium carbonate is also reported in some of them (Aggarwal et al., 1992; Singh et al., 2004).

The climate of the Chambal basin is warm, humid and semi-arid, with mean annual minimum and maximum temperatures of 18.9 °C and 32.2 °C respectively (IMD, 1999). Maximum temperature displays a bimodal distribution, with peaks in summer (May) and fall (October). The average annual rainfall in the Chambal basin based on the Tropical Rainfall Measuring Mission (TRMM) Gridded Rainfall Data (http://disc2.nascom.nasa.gov/Giovanni/tovas/TRMM_V6.3B43.shtml; Fig. 1a), is ~80 cm. The TRMM based annual rainfall is consistent with the average

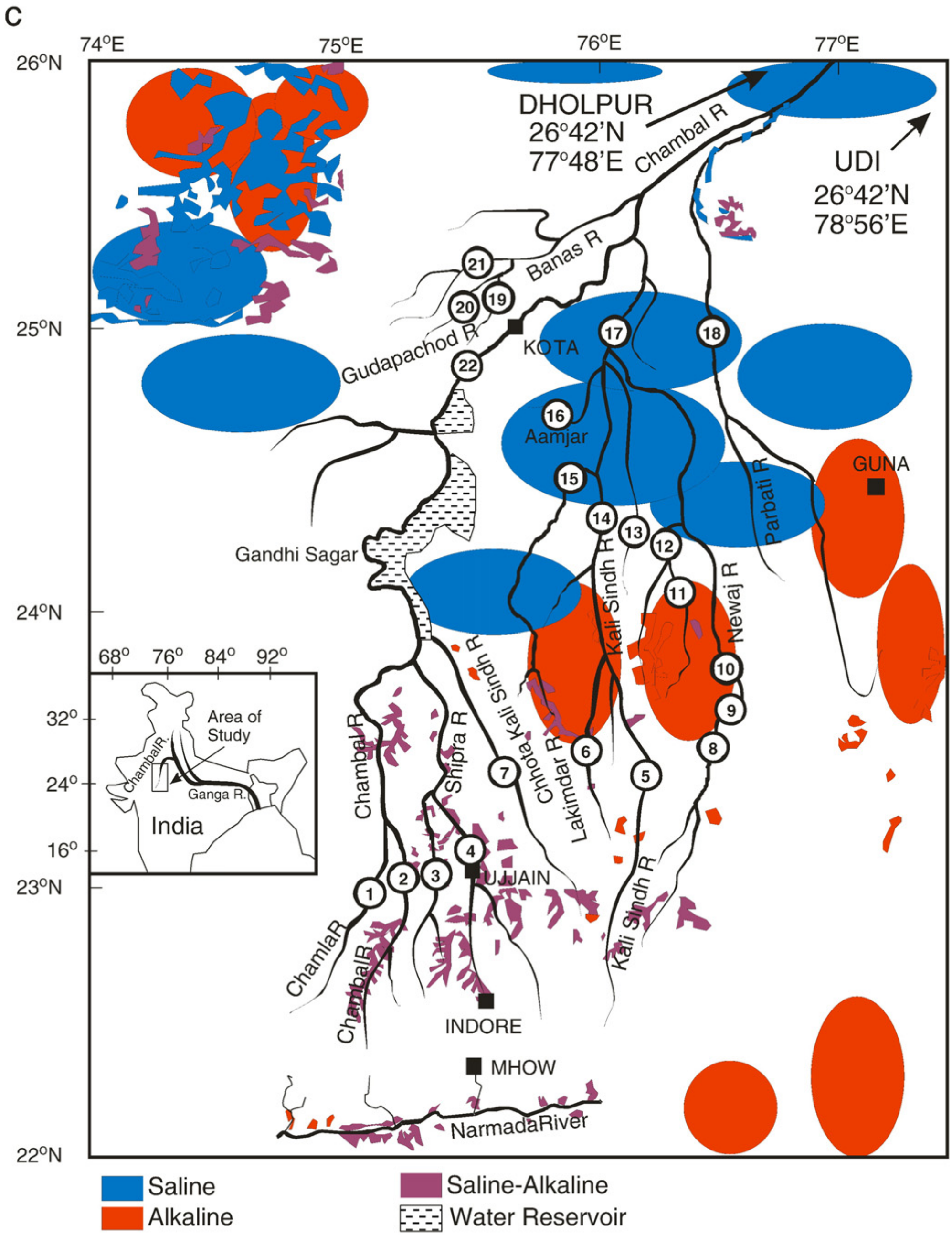

Fig. 1 (*continued*).

of ~81 cm for the years 1995–2004 compiled by the Indian Institute of Tropical Meteorology (IITM), Pune (Dr. B. N. Goswami, personal communication). Both TRMM and IITM data in the Chambal basin show significant spatial variability in precipitation, gradually decreasing towards the north–northwest (Fig. 1a). The rainfall is more (~100 cm y^{-1}) in the headwaters of the Chambal, the Kali Sindh and the Parbati. In contrast, the Banas catchment receives only ~50 cm rain annually. There have been concerns on the use of TRMM data for rainfall amount estimation (Bookhagen and Burbank, 2006; Kamal-Heikman et al., 2007). This is unlikely to be an issue in this study as its interest is on the spatial distribution of rainfall which is nearly identical as derived from both TRMM and IITM data. Further, the average annual rainfall over the Chambal basin estimated from both TRMM and IITM data are also quite similar. The overall low rainfall in the Chambal drainage promotes the development of alkaline and saline soils in its basin (FAO, 2005). The mean annual water discharge of the Chambal at Udi (26°42'N, 78°56'E) prior to its confluence with the Yamuna is ~30×10^{12} L (Rao, 1975; CBPCWP, 1982) with maximum discharge, ~90%, during monsoon.

3. Sample collection and analytical methods

The details of the sampling and analysis are given in Rengarajan (2004). Briefly, water and sediment samples from the headwaters of the Chambal and its tributaries were collected during September 1998 from 22 locations (Fig. 1b, c). Sampling was done from the mid-stream of rivers whereas sediments were collected from river beds/banks. The sampling was carried out during monsoon, when the water stages are highest in the rivers, to minimize the influence of ground water discharge and anthropogenic inputs on their major ion chemistry. Measurements of pH (precision ± 0.1 unit) and alkalinity (by titration with HCl using Merck® mixed indicator) were performed at site. For measurement of dissolved major ions, water samples were filtered through Gelman® 0.4 µm filters at site and stored in clean polyethylene bottles (Rengarajan, 2004). Soon after filtration, an aliquot of the filtered water was acidified to pH ~2 using Merck® ultra pure HNO_3 and stored separately for measuring trace elements and Sr isotopes.

In the filtered unacidified water samples, Na and K were measured by AAS, and Si, Ca and Mg by ICPAES and Cl, NO_3 and SO_4 by Ion Chromatography. In addition, a few samples of acidified water samples were also measured for Ca and Mg. The results showed no measurable difference in their concentrations with respect to unacidified samples, implying that during the interval between sampling and analysis, Ca and Mg have behaved conservatively. Measurement precision for various cations and anions, based on repeat analysis of samples and standards, are generally better than 5% (Rengarajan, 2004). Dissolved Sr was measured in the filtered, acidified samples by ICPAES using ultrasonic nebulizer (Dalai et al., 2002). Along with the samples, USGS reference materials were also analyzed to monitor the accuracy of measurements. This was better than ±5%.

For measurement of Sr isotope composition, strontium was separated from a known weight of filtered and acidified water (typically ~100 g) by ion-exchange techniques (Singh et al., 2008). Isotope composition of purified Sr fractions was measured using ISOPROBE-T thermal ionization mass spectrometer in static multi-collection mode. Measured $^{87}Sr/^{86}Sr$ ratios were corrected for instrumental mass fractionation using $^{86}Sr/^{88}Sr$ as 0.1194. Internal precision of analysis was better than ±10 ppm (1σ). The total procedural blanks for Sr varied between 1.2 to 3 ng compared to sample strontium loads of several µg. Along with samples, NBS 987 standard was also measured which yielded $^{87}Sr/^{86}Sr$ of 0.710234±0.000024 (2σ, $n=3$) during the measurement period.

4. Results

Concentrations of dissolved major cations and anions in the Chambal river system along with their strontium isotope ratios are given in Table 1. The water in the Chambal mainstream and its tributaries is alkaline in nature with pH ranging from 8.2 to 9.2 (Table 1). Total dissolved cation (TZ^+) and anion (TZ^-) charges in the Chambal waters varied from 2416 to 7656 µEq and 2166 to 7062 µEq respectively. The normalized inorganic charge balance, NICB [= $(TZ^+ - TZ^-)/TZ^+$], show a marginal excess of TZ^+, averaging 8.8±3.4% (Table 1). The higher deficit of negative charge in some of the samples (10–15%) needs further study; it could be due to organic anions such as acetates, oxalates and humic components, the importance of which in the charge balance of tropical rivers is known (Edmond et al., 1995; Viers et al., 2000). Among cations, Na is often the most abundant (in molar units) and constitutes ~25–58% of total cations. The cation abundance in these streams decreases as Na≈Ca>Mg>K. This trend differs from that of the Himalayan rivers, (e.g. the Yamuna headwaters) where Ca≈Mg>Na>K. This difference, as discussed later, can result from contribution of Na to rivers of the Chambal system from alkaline and saline soils.

The anions in Chambal vary as Alk>Cl>SO_4>NO_3, with Alk constituting ~60–90% (molar). Chloride accounts for ~8 to 29%; the variations in its concentration parallel that of Na. The concentrations of NO_3 and SO_4 are quite high in many samples and range from 14 to 281 µM and 42 to 301 µM, respectively (Table 1). The high concentrations of Cl, NO_3 and SO_4 in the Chambal has been persistent at least over four decades, the measurements reported for samples from Kota, made during 1968/1969 show that over a year monthly values range from 2 to 105 µM and 50 to 350 µM for NO_3 and SO_4 (CWPC, 1973). The source of NO_3 in rivers can be natural (from soils where oxidation of organic matter is prevalent) and/or anthropogenic (fertilizers and industrial wastes, Meybeck, 1982; Probst, 1985; Berner and Berner, 1996; Subramanian 2008). Samples with relatively high concentrations of NO_3 and SO_4 are also chloride rich (Table 1). All these anions also correlate significantly with Na ($r^2=0.74$, 0.59 and 0.82 for Cl, NO_3 and SO_4 respectively, $p<0.001$) hinting at the possibility of a common source for them such as saline/alkaline soils and/or anthropogenic input (see later discussion). Some of these ions can be from fertilizers as part of the Chambal basin is intensely farmed agricultural land where a variety of fertilizers are in use (CBPCWP, 1982; FAO, 2005).

The dissolved Si concentration in the Chambal streams range from 157 to 607 µM (Table 1) and are similar to those reported for the Narmada, Tapi and the Krishna rivers flowing through the Deccan Trap basalts (Dessert et al., 2001; Das et al., 2005b). The (Alk/Si) molar ratios

Table 1
Major ion and Sr isotope composition of the Chambal river system

Sample Code	River	Location	pH	Na	K	Mg	Ca	Alk	Cl	NO_3	SO_4	Si	NICB %	TDS mg L^{-1}	Sr µM	$^{87}Sr/^{86}Sr$
				µM												
Western Rivers																
CH-1	Chamla	Barnagar	8.2	1965	188	518	876	3221	935	138	108	279	9	366	3.17	0.71024
CH-2	Chambal	Barnagar–Ujjain Road	9.2	1247	176	424	604	2279	600	–	122	225	10	256	2.18	0.71021
CH-3	Ghambir	Barnagar – Ujjain Road	8.2	925	140	366	716	2527	316	14	67	189	7	248	2.20	0.71038
CH-4	Shipra	Near Ujjain	8.4	2455	247	938	1539	4212	1967	281	301	386	8	547	3.78	0.71025
CH-7	Chhota Kali Sindh	Bat village	8.4	554	88	313	574	1784	239	21	61	157	10	181	2.01	0.71025
CH-16	Aamjar	Before Kota	8.4	2398	161	580	973	4063	746	101	259	214	4	433	5.85	0.71219
CH-19	Talera	Talera village	8.2	719	59	473	878	2478	269	63	80	171	15	248	2.46	0.71135
CH-20	Gudapachod	Between Kota & Bundi	8.7	1007	58	453	913	2924	387	31	92	168	7	286	2.72	0.71113
CH-21	Mangli	Between Kota & Bundi	8.6	1749	71	605	826	3072	802	53	187	171	8	338	3.99	0.71088
CH-22	Chambal	Jawahar Sagar Dam	8.4	785	73	399	611	2230	378	–	117	168	1	226	1.94	0.71030
Eastern Rivers																
CH-5	Kali Sindh	Sarangpur	8.7	892	110	864	1135	3716	436	86	98	450	11	375	2.65	0.70927
CH-6	Lakimdar	Choma village	8.5	1172	95	1033	1068	4162	453	83	114	550	10	418	3.26	0.70923
CH-8	Newaj	Pachor	8.2	1032	130	757	1008	3766	491	69	99	343	4	369	2.98	0.71023
CH-9	Dhudhi	Biaora	8.3	1164	109	782	749	3171	571	51	88	479	8	334	2.46	0.71003
CH-10	Newaj	Kisanghat	8.4	999	103	749	863	3270	480	47	95	418	8	333	2.63	0.70999
CH-11	Gherganga	Before Aklera	8.5	587	47	601	1090	3171	271	42	54	607	11	321	1.91	0.71091
CH-12	Chappi	Arnia	8.4	678	57	650	1043	3122	294	37	57	521	13	315	2.13	0.70944
CH-13	Ujar	Asnawar	8.6	480	76	490	896	2725	234	–	42	393	9	264	1.86	0.70989
CH-14	Kalisindh	Jalawar	8.6	859	91	774	951	3072	401	71	96	454	15	323	2.47	0.70926
CH-15	Aav	Suket	8.7	974	68	741	828	3171	332	39	80	432	11	318	2.54	0.70936
CH-17	Kalisindh	Kota-Kisanganj Road	8.7	735	75	630	906	3072	312	56	82	411	7	306	2.28	0.70966
CH-18	Parbati	Kisanganj	8.7	727	73	576	828	2973	267	50	61	354	5	288	2.17	0.71007

– below detection limit.

in the Chambal waters range from 5 to 19, much higher than that expected and observed for weathering of the Deccan Trap basalts in the Krishna headwaters (~2, Das et al., 2005a,b). These high ratios bring out the importance of additional sources of carbonate to the Chambal waters such as weathering carbonates and saline/alkaline soils.

Fig. 2a and b are ternary cation [Mg, Ca, (Na+K)] and anion [Alk, Cl+SO_4, Si] diagrams of the samples analysed. It is seen in Fig. 2a that the water data cluster around the centre suggesting that Ca, Mg and (Na+K) all contribute roughly equally to the cation budget. This inference also draws support from the inter-relation of (Na+K) and (Ca+Mg) with ΣCations. In contrast, the anions plot close to the alkalinity apex along the mixing line of alkalinity and Cl+SO_4 (Fig. 2b). This reiterates the dominant role of alkalinity and its mixing with (Cl+SO_4) in determining the anion budget of these rivers. The plot also shows that silica has only a minor role on the anion budget.

The total dissolved solids (TDS) in the samples ranged from 181 to 547 mg L^{-1} (Table 1, Fig. 3). Among the tributaries, the Chhota Kali Sindh has the lowest TDS, ~181 mg L^{-1}, while the Shipra has the highest, ~547 mg L^{-1}. The TDS of all the rivers sampled in the Chambal system significantly exceed that of the global mean river water (Meybeck and Helmer, 1989; Berner and Berner, 1996). The TDS of the Chambal near its source (CH-2) and at the outflow of the Jawahar Sagar Dam (CH-22) are 256 and 226 mg L^{-1} respectively (Table 2), very similar to that reported for the Chambal at Dholpur (247 mg L^{-1}; Sarin et al., 1989) ~275 km downstream of CH-22 (Fig. 1a) sampled during September 1982 (Table 2). These data, though limited, indicate that TDS of the Chambal does not vary significantly along its course, from the source to near its confluence with the Yamuna. The TDS of the Chambal is roughly similar to that of the Yamuna at the foothills of the Himalaya (Table 2).

The frequency distribution of TDS in rivers of the Chambal system is compared with that of the Yamuna headwaters (both collected during Sept–Oct 1998) in Fig. 3. The range of TDS of the Yamuna brackets that of the Chambal river system; however, the average TDS of the Yamuna headwaters is 181 mg L^{-1} compared to 322 mg L^{-1} of the Chambal. It is seen that nearly two thirds of the Yamuna headwaters have TDS < 175 mg L^{-1}, whereas almost all the Chambal samples have TDS in excess of 175 mg L^{-1} (Fig. 3), reinforcing their highly saline nature.

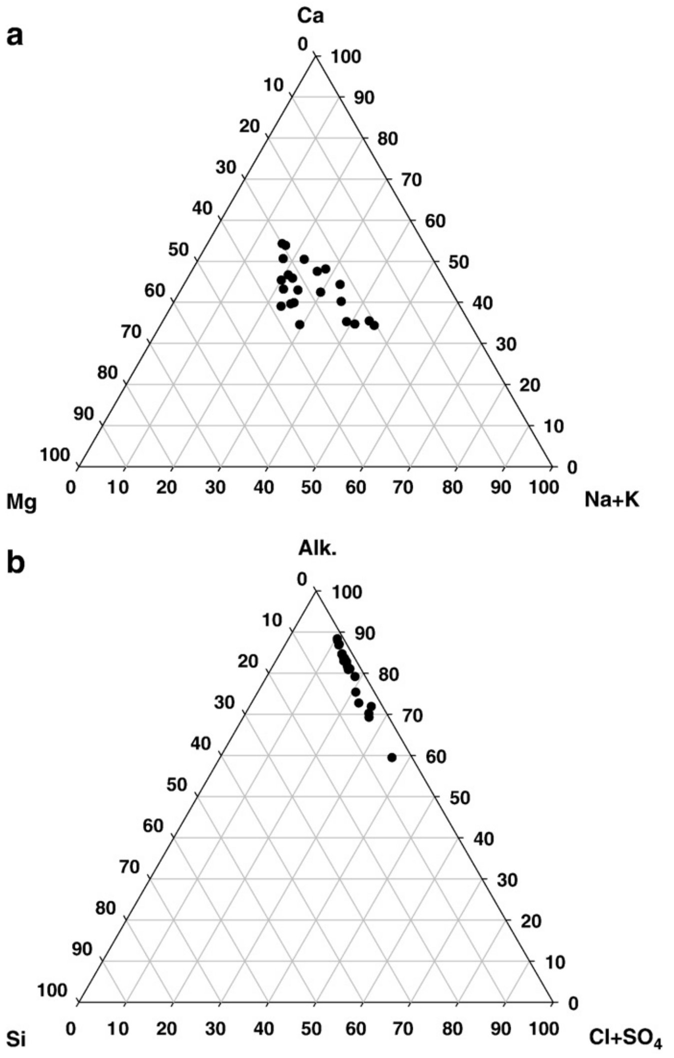

Fig. 2. Ternary plots of major cations (a) and anions (b) in rivers of the Chambal river catchment (μEq). In most samples the cations plot near the centre suggesting that all these ions contribute nearly equally to the cation budget. (b) Anions plot along the mixing line of Alkalinity and (Cl+SO_4), with most samples close to the alkalinity apex. The data bring out the important contribution of Alk from weathering of basalts and carbonates by CO_2 and solution of alkaline/saline soils.

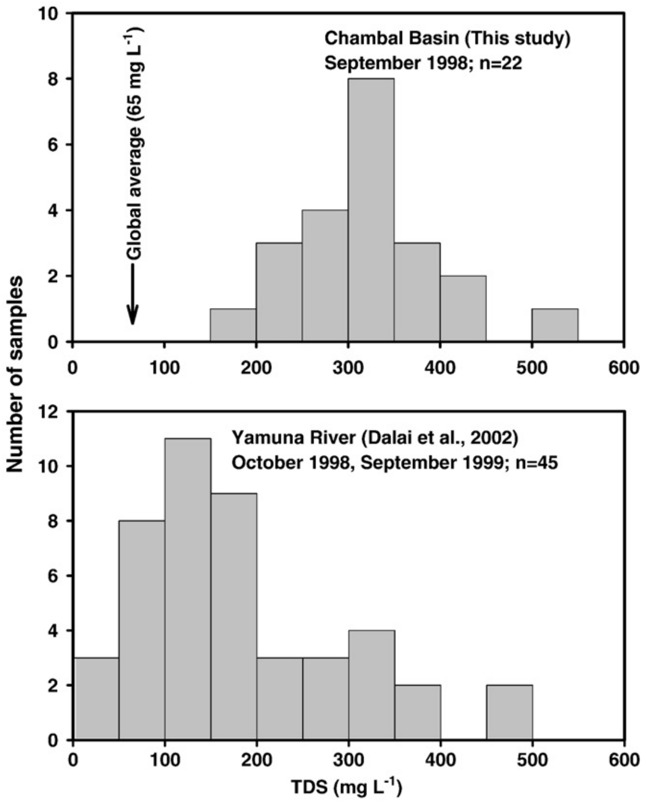

Fig. 3. Distribution of total dissolved solids (TDS) among the rivers of the Chambal system. The TDS ranges from 181 to 547 mg L^{-1}. Data for the Yamuna at Batamandi and Saharanpur (September, 1999; Dalai et al., 2002) are also given for comparison. The discharge-weighted average of TDS in world rivers (Meybeck and Helmer, 1989) is also shown.

Table 2

Temporal variation of TDS in the Chambal and the Yamuna rivers

Sampling date	Location	TDS mg L^{-1}	References
Chambal			
September 1998	Barnagar–Ujjain Road	256	This study
September 1998	Jawahar Sagar Dam	226	This study
March 1982	Dholpur	333	Sarin et al. (1989)
September 1982	Dholpur	247	Sarin et al. (1989)
Yamuna			
October 1998	Batamandi	229	Dalai et al. (2002)
September 1999	Batamandi	274	Dalai et al. (2002)
October 1998	Saharanpur	236	Dalai et al. (2002)
September 1999	Saharanpur	151	Dalai et al. (2002)

The dissolved Sr in the Chambal sample near its source (CH-2) is 2.18 µM with $^{87}Sr/^{86}Sr$ of 0.71021; the corresponding values for the downstream sample at Jawahar Sagar Dam (CH-22) are 1.94 µM and 0.71030 respectively. The Sr concentration in the Chambal tributaries varies from 1.86 µM to 5.85 µM and $^{87}Sr/^{86}Sr$ from 0.70923 to 0.71219 (Table 1). Some of the western rivers, of the Chambal system (headwaters of the Chambal, Shipra, Aamjar and the Banas, Fig. 1b) sampled from locations in the Vindhyan system have marginally more radiogenic Sr. The Sr concentration of the Chambal measured in this study is similar to the reported value of 2.3 µM (Krishnaswami et al., 1992) for a sample collected from Dholpur, in September 1982, which, however, had a marginally more radiogenic $^{87}Sr/^{86}Sr$, 0.7127 (Krishnaswami et al., 1992) due to contribution from the Vindhyan silicates. More importantly, the $^{87}Sr/^{86}Sr$ of the Chambal river and its tributaries measured in this study (and the limited data reported earlier, Krishnaswami et al., 1992; Palmer and Edmond, 1992) show that they are far less radiogenic than the $^{87}Sr/^{86}Sr$ of the headwaters of the Ganga and the Yamuna (e.g. Krishnaswami et al., 1992; Galy et al., 1999; Bickle et al., 2003; Dalai et al., 2003) but is within or close to the range of values for the Narmada, Tapi and the headwaters of the Krishna system, all of which predominantly drain the Deccan Trap basalts (Dessert et al., 2001; Das et al., 2006).

5. Discussion

5.1. Ca, Mg distribution

In majority of the streams, (Ca+Mg) is nearly balanced (within ~±10%) by alkalinity (Fig. 4). Such an observation is often interpreted in terms of a common source for Ca, Mg and alkalinity such as chemical weathering of Ca, Mg dominated silicates and/or carbonates (dolomites) by carbonic acid. Both these options are possible in the Chambal headwater basin considering that Ca and Mg silicates are major components of the Deccan Trap basalts (Subbarao et al., 2000) and limestone and dolomite are abundant in the drainage basin. Samples which deviate from this trend (Fig. 4) are CH-1 (Chamla), CH-3 (Ghambir), CH-4 (Shipra) and CH-16 (Aamjar). Some of these samples also have high concentrations of Na, Cl, SO_4 and NO_3 (Table 1) indicating that their chemistry is influenced significantly by contributions from other sources. In the Shipra sample alkalinity is ~15%, deficient relative to (Ca+Mg) whereas in the Chamla and Aamjar samples, alkalinity is ~15 to 30% excess. The deficiency in alkalinity can result if part of (Ca+Mg) is balanced by SO_4, whereas its excess can be due to supply of alkalinity from alkaline soils some of which are known to contain $NaHCO_3$ and Na_2CO_3 (Table A2; Sehgal et al., 1975; Bhargava et al., 1981; Pal et al., 1994; Datta et al., 2002).

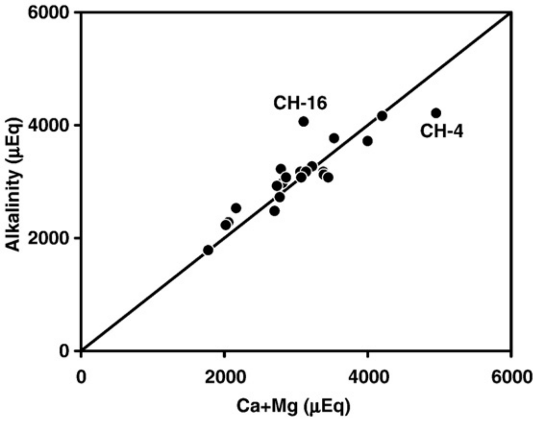

Fig. 4. Scatter plot of (Mg+Ca) with Alk. Most of the points fall close to the equiline. The four data points plotting away are CH-1, CH-3, CH-4 and CH-16. The overall equivalence of Alk and (Ca+Mg) is suggestive of CO_2 mediated weathering of (Ca+Mg) dominated silicates and carbonates.

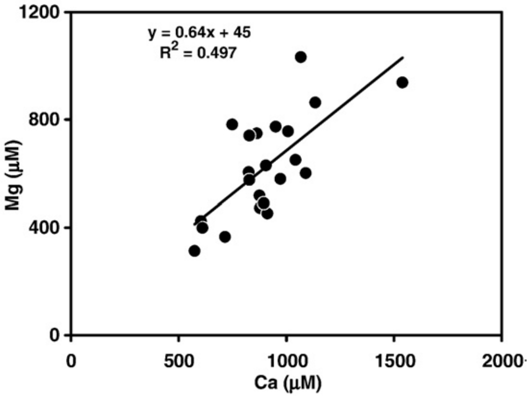

Fig. 5. The variation of dissolved Mg concentration with Ca. There is an overall positive correlation between Ca and Mg (r^2=0.497, n=22).

Mg shows an overall correlation (r^2=0.497, n=22) with Ca albeit significant scatter (Fig. 5). The Deccan Trap basalts, the Vindhyan and *kankar* carbonates are the primary sources of Ca and Mg to these waters. The variability in their relative contributions from these sources can be a cause for the scatter in Fig. 5. Incongruent weathering of basalts for Ca, Mg is unlikely to be the source of scatter in Fig. 5, as earlier studies on the Krishna system waters and sediments (Das et al., 2005b; Das and Krishnaswami, 2007) concluded that the Deccan Trap basalts weather nearly congruently for these elements (Section 5.2). Another factor that can contribute to the scatter is the behaviour of Ca in waters. Calculation of CSI (calcite saturation index) following Drever (1988) shows that all the samples analyzed are supersaturated in calcite (at 25 °C) with CSI values in the range of +0.33 to +1.26. Such a high degree of calcite supersaturation is suggestive of its precipitation. Support for this suggestion comes from the presence of *kankar,* impure calcium carbonate, in the Chambal, Yamuna and the Ganga basins (Aggarwal et al., 1992; Pal et al., 1994). The bed sediments of the Chambal system rivers collected from the same locations as the water samples contain 5.1 to 44.8 (wt.%) of $CaCO_3$ (Dalai et al., 2004) part of which can be *kankar* carbonates. The precipitation of calcite, however will have only a minor effect on the dissolved Mg (and Sr) abundances of rivers as its partition coefficient in calcite is <<1 (Jacobson et al., 2002 and references there in).

On a ratio plot, Mg/Na exhibits a linear trend with Ca/Na (Fig. 6). The data fall along a two end-member mixing line between a low Ca/Na,

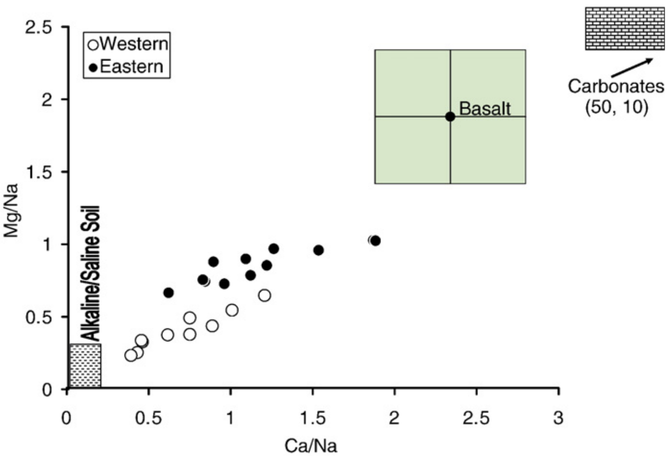

Fig. 6. Property plot of Ca/Na vs Mg/Na; data for various end members contributing Ca, Mg and Na to the Chambal system are also shown. The river water data, as expected, lie along the mixing line among alkaline/saline soils, basalts and carbonates. The results show two trends, one for the western and the other for the eastern rivers. The composition data of end members are given in Tables A1, A2 and A3 of the Appendix. The Ca/Na and Mg/Na ratios in carbonates are from Gaillardet et al. (1999).

Mg/Na end-member (alkaline/saline soil; Table A2) and a high Ca/Na, Mg/Na end-member (the Deccan Trap basalts and/or the Vindhyan carbonates). The composition and elemental ratios of these different end-members are given in the Appendix. Data on the average composition of the Deccan Trap basalts and those from the Mhow region given in Table A1 are from Peng et al. (1998) and Das et al. (2005b). Plotting the elemental ratios of basalts in Fig. 6 implies that Ca, Mg and Na are released to rivers from basalts in the same ratio as their abundances. The validity of this assumption is discussed in the subsequent section. The Ca/Na and Mg/Na ratios of alkaline/saline soil end-member for the Chambal basin are unavailable. Therefore the available data on the composition of aqueous extracts of saline/alkaline soils from the Ganga plain (Bhargava et al., 1981; Pal et al., 2003) are used to characterize this end-member (Table A2). The composition of carbonates is presented in Table A3 (Singh et al., 1998; Ray et al., 2003). The data in Fig. 6 seem to follow two trends, one set by the western rivers (headwaters of the Chambal, Shipra and the Banas) and the other by the eastern rivers (headwaters and tributaries of the Kali Sindh, Newaj and the Parbati). These trends seem to be diverging from the high ratio end member, probably a result of variations in the Mg/Na ratios of the saline /alkaline end member. Calcite precipitation is another process that can alter the Ca/Na (and to a much less degree Mg/Na) ratios of samples compared to that expected from the end-member ratios. The effect of precipitation would be to decrease the Ca/Na in rivers relative to the end-member and thus shift the data points to the left in Fig. 6, i.e. towards Mg/Na axis.

5.2. Contribution of major ions from different sources to the Chambal river system

Major ions are supplied to rivers via atmospheric deposition, chemical weathering of various lithologies in the basin and anthropogenic sources. The dissolved concentrations of major ions and their ratios in the Chambal river are used to constrain contributions from these sources.

5.2.1. Atmospheric deposition

The contribution of atmospheric deposition to the dissolved load can be determined from the chemical composition of rain in the region. Available data on the composition of rainwater from the plains of the Yamuna and the Chambal basins are given in Appendix (Table A4). The average Cl concentration in rainwater from these sites is 31 µM compared to the average concentration of 508 µM in the Chambal system rivers (Table 1). A rough estimate of average rainwater contribution of Cl to the rivers of the Chambal basin is calculated to be ~8%. The atmospheric contribution to individual rivers, however, can vary from the mean depending on local rain and river water Cl and the extent of evapotranspiration. Das et al. (2005b) used a value of ~40% for evapotranspiration in the Krishna basin. If this evapotranspiration factor is applied to the Chambal system, the average atmospheric contribution of Cl would increase to ~13%. The Na contribution from atmospheric deposition on average is ~3% of its river concentration and it would increase to ~5% if evapotranspiration is considered. Mean Ca and Mg in rainwater are 41 and 25 µM respectively. These values are <8% of the lowest concentration measured in Chambal streams (CH-7, Table 1) or <4% of the average river water concentration. It is evident from the above estimates that ≥90% of the major ions in the Chambal, including Cl, has to be derived from sources other than precipitation. The other sources of major ions, as mentioned earlier are (i) chemical weathering of various lithologies of the drainage basin (ii) groundwater and (iii) anthropogenic inputs.

5.2.2. Chemical weathering of the drainage basin

The Deccan Trap basalts and the Vindhyan carbonates are the dominant lithologies of the Chambal headwater basin (Fig. 1b). In addition, *kankar* carbonates and easily weatherable saline/alkaline soils are also dispersed throughout the basin.

5.2.2.1. Solution of alkaline/saline soils. The alkaline/saline soils containing NaCl, Na_2SO_4, Na_2CO_3 and $NaHCO_3$ dispersed in the Chambal basin (Fig. 1c) can be important sources of Cl, SO_4, alkalinity and Na to the rivers. The Cl concentration of rivers in excess of rainwater contribution range from 182 to 1912 µM. If all this Cl is associated with Na as NaCl, then it can be estimated that on average NaCl contribution from these soils can account for ~40% of dissolved Na, an exception being the Shipra (CH-4) sample for which the contribution can be much higher, ~80%. The source(s) for the balance Na can be weathering of silicates and supply of sulphate/carbonate/bicarbonate of Na from alkaline and saline soils, groundwater and anthropogenic inputs. Anthropogenic input, in terms of domestic and industrial wastes (http://cgwb.gov.in/NCR/geochemical.htm) and fertilizers, can be important locally as parts of the Chambal basin are thickly populated and intensely farmed (CBPCWP 1982; FAO, 2005). The impact of this source, however, on the chemistry of samples of this study is expected to be minimal as they were collected during monsoon (September), when the river stage is the highest. In light of this and the widespread occurrence of saline/alkaline soils in the Chambal headwater basin (Fig. 1c) strongly favours them as an important source of Na salts to balance the Na budget of these rivers. The role of silicate weathering and saline/alakaline soils in contributing to major ions is discussed in the subsequent section.

5.2.2.2. Silicate weathering. Apportioning the contribution of major ions to river waters from chemical weathering of silicate rocks is important as it impacts on the long term budget of atmospheric CO_2 and hence global climate (Walker et al., 1981; Raymo and Ruddiman, 1992; Ruddiman, 1997). The contribution from silicate weathering to river water chemistry is derived either by forward modelling using suitable proxies (Galy and France-Lanord, 1999; Krishnaswami et al., 1999; Das et al., 2005b) or by inverse modelling (Negrel et al., 1993; Gaillardet et al., 1999; Millot et al., 2003; Moon et al., 2007). One of the proxies used in the forward modelling approach is Na* generally taken to be the concentration of Na in rivers derived from silicate weathering in their basins [$Na^* = (Na_r - Cl_r)$ where the subscript r refers to concentration in rivers]. Forward modelling assumes that dissolved Na in rivers is derived from two sources, (atmospheric + halite) and silicate weathering. Further, the (atmospheric + halite) contribution of Na is generally taken to be the concentration of Cl in rivers, making the silicate derived Na equal to Na*. Knowing Na*, the contribution of other major cations (Ca, Mg, K) to rivers from silicate weathering is calculated by assuming that they are released to the rivers from silicates of the basin in a fixed proportion relative to Na. The dominant source of silicate cations to the Chambal rivers sampled is the Deccan Trap basalts as they cover a large fraction of these river basins (Fig. 1b). This is also consistent with the observation that the Sr isotope composition and Fe/Al ratio of the silicate fraction of bed/bank sediments of the headwaters of the Chambal system (unpublished data) are within the range reported for the Deccan Trap basalts from the Mhow region (Peng et al., 1998). The abundances of silicate derived cations estimated based on Na* could lead to overestimation if part of Na* is supplied from sources other than silicates, such as sulphates/carbonates of Na, as inferred for the Chambal rivers. Indeed, Das et al. (2005b), in their studies of the Krishna headwaters, which drain the Deccan Trap basalts almost exclusively, used concentration of dissolved Mg of silicate origin as a proxy to estimate silicate derived cations in rivers as there were concerns that part of Na* in these rivers could be of non-silicate origin. Das et al. (2005b) in the process of estimating the silicate-derived cation concentrations, observed that dissolved Mg/Na and Ca/Mg ratios (corrected for rain input) in a number of small streams of the Krishna basin and the western ghats where water chemistry is determined only through contributions from atmospheric deposition and basalt weathering, overlaps within errors with the corresponding ratios in the Deccan Trap basalts. For example, the Ca/Mg and Na/Mg molar ratios in the Krishna headwaters are 1.27 ± 0.08 and 0.49 ± 0.08 compared to values of 1.19 ± 0.25 and 0.52 ± 0.12 in the Deccan Trap basalts (Das et al., 2005b). This led Das et al. (2005b) to conclude that the Deccan Trap basalts, in general, weather nearly congruently for Na, Ca and Mg. These results on the ratios with which Ca, Mg and Na are released from "whole rocks" of the basin to rivers provide a reliable approach to calculate silicate-derived cation abundances in rivers and silicate erosion rates of the basin. The conclusion of Das et al. (2005b) further indicates that the cumulative impact of differences in the abundances of minerals hosting Na, K, Mg and Ca in the Deccan Trap basalts and their weathering rates on the release of these elements from basalts to rivers is within the uncertainties of their abundance ratios in water and basalts.

The Na*/Mg ratio in the Chambal rivers range from 0.50 to 2.85 with almost all the western rivers having ratios in excess of 1.0 (considering that Mg concentrations used in these calculations are the measured values (Table 1) which have both silicate and carbonate contributions, the Na*/Mg_{sil} would be even higher than the calculated ratios). These high Na*/Mg ratios, far in excess of the ratio in basalt (0.52 ± 0.12), therefore, suggest either additional sources for Na* to the Chambal headwaters and/or incongruent weathering of Na/Mg in the Chambal headwater basin, with Na being weathered preferentially over Mg. The results of Das et al. (2005b) on the near congruent release of Na, Mg and Ca from the Deccan Trap basalts to rivers, however, argue against the latter hypothesis. Therefore, the high Na*/Mg ratios in the Chambal rivers is more likely to be due to supply of Na from additional sources such as non-chloride salts of saline/alkaline soils (e.g. NaHCO_3, Na_2CO_3, Na_2SO_4). The impact of anthropogenic inputs on the Chambal headwaters analysed should be minimal considering that sampling was carried out during peak water discharge. This inference, that there are non-silicate source(s) for Na*, suggests that the silicate Na calculated based only on Cl correction would be upper limits.

The role of additional sources of Na is also borne out from Si/(Na* + K) ratio. The Si/(Na* + K) molar ratio in the Chambal range between 0.12 to 1.67 with a majority of samples having values < 0.70. Among the samples, the western rivers generally have lower Si/(Na* + K) due to both low Si and high (Na* + K) concentrations. The Si/(Na* + K) ratio has been used to infer the nature of silicate weathering provided these elements are derived only from silicates (Stallard and Edmond, 1983). The Si/(Na* + K) molar ratio of > 4 is expected for weathering of the Deccan Trap basalts to kaolinite. The observation that most of the measured Si/(Na* + K) ratios in the Chambal headwaters are significantly lower than the expected value is an indication of additional supply of Na and K to the waters. This argument, however, would be negated if Si behaves non-conservatively in the Chambal headwaters. Si, being a biogeochemically active element, can be subject to removal from dissolved phase of rivers through uptake by diatoms (Humborg et al., 2000; Meybeck 2005), phytoliths (Webb and Longstaffe, 2000) and sequestration in secondary phases formed during weathering. The occurrence of lakes/reservoirs along the course of the rivers can promote the uptake of Si by diatoms (Meybeck, 2005). In the Chambal system studied, the Gandhi Sagar dam is a major reservoir (Fig. 1a) and can be a potential site for the uptake of Si by diatoms. However, considering that the sampling was done during monsoon, the peak discharge period of rivers and when the waters are generally turbid due high particulate matter concentration, diatom and other plant growth in rivers and associated uptake of Si is expected to be minimal. The low Si/(Na* + K) in the samples, therefore is more likely due to additional sources of (Na* + K).

The use of Mg as a proxy to derive silicate contribution of cations in the Chambal headwaters is also limited. This approach was possible in the Krishna head-waters as Mg in them is almost entirely of basalt origin. In contrast, in the Chambal headwaters, Mg is sourced from both silicates (the Deccan Trap basalts) and carbonates (the Vindhyan and *kankar*).

It is evident from the above discussion that, in the Chambal basin, estimation of silicate erosion and associated CO_2 consumption rate has to depend on surrogates other than Na and Mg. In this context, dissolved Si in rivers has often been used to derive CO_2 consumption by silicate weathering in river basins (Stallard and Edmond, 1983, 1987; Edmond and Huh, 1997). Extension of this application to obtain concentrations of silicate derived cations in rivers and silicate erosion rates is feasible if (i) the ratios of release of Na, Mg and Ca relative to Si from silicates to rivers are known and (ii) Si behaves conservatively in rivers. Das et al. (2005a,b) based on their studies of δ^{13}C in dissolved inorganic carbon (DIC) and major ion chemistry of Krishna headwaters and western ghat rivers observed that samples most-depleted in δ^{13}C (−18 ± 2‰) have (Si/HCO_3) molar ratio of 0.5 ± 0.05. These highly depleted δ^{13}C values was attributed to DIC derived almost entirely from basalt weathering with CO_2 from C_3 plants, making the measured (Si/HCO_3) representative of the Deccan Trap basalt weathering. This measured ratio is in good agreement with the value ~0.52, calculated for weathering of the Deccan Trap basalts based on their mineralogical and chemical composition (Das et al., 2005a). Further, the HCO_3/Mg molar ratio calculated for weathering of the Deccan Trap basalts 6.4±0.6, was also found to be in good agreement with the measured value of ~5.8 ± 0.5. Based on these HCO_3/Mg and Si/HCO_3 ratios and assuming congruent release of Na, Mg and Ca from basalts to rivers (Das et al., 2005b), the Na/SiO_2, Mg/SiO_2 and Ca/SiO_2 molar ratios are calculated to be 0.16 ± 0.04, 0.30 ± 0.04 and 0.36 ± 0.09 respectively. The errors are based on uncertainties in average elemental abundances and HCO_3/SiO_2 ratios. Using these release ratios and the dissolved Si concentration in the Chambal water samples, the cation contribution from the Deccan Trap basalts are derived as

$$(X_i)_{\text{sil}} = \left[\left(\frac{X_i}{\text{Si}}\right)_{\text{sol}}\right] \times \text{Si} \tag{1}$$

$$f\text{cat}_{\text{sil}} = \frac{\sum(X_i)_{\text{sil}}}{\sum(X_i)_r} = \frac{\text{Na}_{\text{sil}} + \text{K}_{\text{sil}} + \text{Ca}_{\text{sil}} + \text{Mg}_{\text{sil}}}{\text{Na}_r + \text{K}_r + \text{Mg}_r + \text{Ca}_r} \tag{2}$$

where $\left(\frac{X_i}{\text{Si}}\right)_{\text{sol}}$ is the ratio of (Ca, Mg, Na)/Si released to rivers from basalts and subscripts *r* and sil refer to rivers and silicate-derived. The calculated silicate derived cations are given in Table 3. In Eq. (2), K_{sil} is calculated by subtracting the rain contribution of K from its measured concentration in rivers. K in the Deccan Trap basalts resides mainly in plagioclase and ground mass (Sethna and Sethna, 1988; Subbarao, 1988). Implicit in the above calculation is the assumption that dissolved Si behaves conservatively in the Chambal system. Therefore, if dissolved Si behaves non-conservatively in these rivers (i.e. gets removed by biogeochemical processes), the calculated silicate derived cations and $f\text{cat}_{\text{sil}}$ would be lower limits.

The estimated fraction of cation contribution from the Deccan Trap basalts, $f\text{cat}_{\text{sil}}$, range from 6 ± 0.6 to 23 ± 3% with a mean of 14 ± 4%. The $f\text{cat}_{\text{sil}}$ for the eastern rivers (the Kali Sindh, Newaj and the Parbati) seem marginally higher 17 ± 2% than that of the western rivers (the

Table 3

Silicate derived cations in the Chambal sub-basins

River	Sample code	SiO_2	Ca_{sil}	Mg_{sil}	Na_{sil}	K_{sil}	ΣSil
		μM					mg L^{-1}
Chambal	CH-22	168	60.5 ± 15	50.4 ± 10	26.9 ± 6.7	55 ± 18	16.5 ± 0.7
Kali Sindh	CH-17	411	148 ± 37	123 ± 25	65.7 ± 16	57 ± 18	37.3 ± 1.6
Parbati	CH-18	354	127 ± 32	106 ± 21	56.6 ± 14	55 ± 18	32.4 ± 1.4
Banas[a]		169	60.8 ± 15	50.7 ± 10	27.0 ± 6.7	40 ± 18	16.0 ± 0.7
Average		276 ± 127					25.5 ± 5.5
Chambal Dholpur[b]		296	106 ± 27	88.8 ± 18	47.4 ± 11.8	35 ± 18	26.6 ± 1.4

ΣSil = [$(\text{Ca} + \text{Mg} + \text{Na} + \text{K})_{\text{sil}} + \text{SiO}_2$] mg L^{-1}.

[a] Average of samples, CH-19, CH-20 and CH-21.

[b] Based on data of Sarin et al. (1989).

Table 4
Silicate erosion rates and CO_2 consumption in the Chambal river basin and other river basins in the Deccan Traps and the Himalaya

River	Location	Water discharge 10^{12} L y^{-1}	Area 10^3 km^2	Runoff mm y^{-1}	SER[a]		CO_2 drawdown[b]	References
					(i)	(ii)		
Chambal	Udi	30	139	215	5.5	2.1	1.1	This work
Chambal	Dholpur	30	139	215	5.7	2.2	1.0	Sarin et al. (1989); This work
Yamuna	Batamandi	10.8	9.6	1125	25–28	10	4–7	Dalai et al. (2002)
Krishna	Alamati			463	14.0	5.4	–	Das et al. (2005b)
Ganga	Rishikesh	22.4	19.6	1140	12.9	4.9	2–3	Krishnaswami et al. (1999)
Ganga–Brahmaputra		1002	1555	640	13.6	5.3	3.3	

[a] Calculated using the mean density 2.6 g cm^3 for silicates. (i): 10^3 kg km^{-2} y^{-1}; (ii): mm ky^{-1}.
[b] Via silicate weathering in 10^5 mol km^{-2} y^{-1}.

Chambal, Shipra and the Chhota Kali Sindh; 10±3%). The river Gherganga (CH-11) has the highest $fcat_{sil}$, ~23±3%. In contrast, the Deccan Trap basalt contribution is lowest to the river Mangli (CH-21) which drains almost entirely through the Vindhyan system. All tributaries sampled from sites within the Vindhyan system (CH-16, 19, 20, 21) have lower silicate cation abundances.

The major source of uncertainty in the estimated concentrations of cations derived from the Deccan Trap basalts to the Chambal rivers is associated with the various (element/Si) release ratios. The uncertainties in Ca/Si, Mg/Si and Na/Si ratios range between 14 to 27%, these get propagated in the estimates of $(X_i)_{sil}$ and $fcat_{sil}$. In addition, if part of dissolved Si is removed by biogeochemical processes, then the estimates of the concentrations of silicate derived cations have to be increased proportionately. The magnitude of this correction is expected to be minimal considering that the sampling was carried out during the peak river stage and representative (Si/HCO_3) ratios have been used for calculations.

5.3. Erosion rates and CO_2 consumption rates

The silicate erosion rate (SER) for the basin is calculated based on Ca_{sil}, Mg_{sil}, Na_{sil}, K_{sil} derived above and measured dissolved Si in rivers, $(SER)=[(Ca+Mg+K+Na)_{sil}+SiO_2\ (mg\ L^{-1})]\times Q$. where Q is the runoff. The range in $[(Ca+Mg+K+Na)_{sil}+SiO_2]$ among the four sub-basins of the Chambal, viz. Kali Sindh, Parbati, Chambal and the Banas is between 16.0±0.7(Banas river system) to 37.3±1.6 mg L^{-1} (Kali Sindh) with a mean of 25.5±5.5 mg L^{-1} (Table 3). The SER of the entire Chambal basin based on the above mean value and runoff at Udi yields a value of $(5.5\pm1.2)\times10^3$ kg km^{-2} y^{-1} (Table 4). This value is nearly identical to that estimated from major ion chemistry of the Chambal sample collected at Dholpur during September 1982 (Sarin et al., 1989). The calculated SER is expected to be typical of the annual average as the sampling was during monsoon, when ~90% of river discharge occurs. The estimated SER is ~5 times lower than that of the Yamuna river at Batamandi (Table 4; Dalai et al., 2002). This is intriguing considering that the headwaters of the Chambal drain the Deccan Trap basalts, which are expected to weather more easily compared to granites/gneisses of the Yamuna headwaters (Dessert et al., 2001; Das et al., 2005b). This difference therefore has to be attributed to other factors such as the higher runoff and relief of the Yamuna head-water basin, factors which are known to enhance chemical erosion. The runoff in the Yamuna basin is 1125 mm y^{-1} compared to 215 mm y^{-1} of the Chambal basin (Table 4). Similarly, the relief of the Yamuna basin is higher by a factor of 20 relative to Chambal basin (Global Mapper V8.0, www.globalmapper.com). The SER of the Chambal system is also only ~40% of that reported for the Krishna head-waters (Das et al., 2005b), though both of them weather the Deccan Trap basalts. The lower SER in the Chambal system has, therefore to be explained in terms of be cumulative effect of lower run off (463 mm y^{-1} in the Krishna basin vs. 215 mm y^{-1} in the Chambal river system) and the presence of other lithologies in the Chambal basin such as the Vindhyan carbonates, which reduces the effective area of exposure of basalts per unit drainage area. The areal coverage of the Deccan Trap basalts is in the Chambal river system is ~60,000 km^2 (Global Mapper V8.0, www.globalmapper.com), which is ~43% of the total drainage area. An alternative, but less likely explanation, is that the calculated SER of the Chambal basin is a lower limit due to non-conservative behaviour of Si. It is however, pertinent to mention here that even if the highest Si concentration among the four sub-basins, Kali Sindh, is used for calculation instead of the mean value, it will not alter the above observations and conclusions, though the SER of the Chambal basin would increase by ~50% to $(8.3\pm1.8)\times10^3$ km^{-2} y^{-1}.

The flux of CO_2 consumed during silicate weathering (ΦCO_2, moles km^{-2} y^{-1}) in the Chambal basin at Udi is calculated from (HCO_3) released during this process, which in turn is derived from Si/HCO_3 ratio (0.52±0.05) and the mean Si concentration of (276±

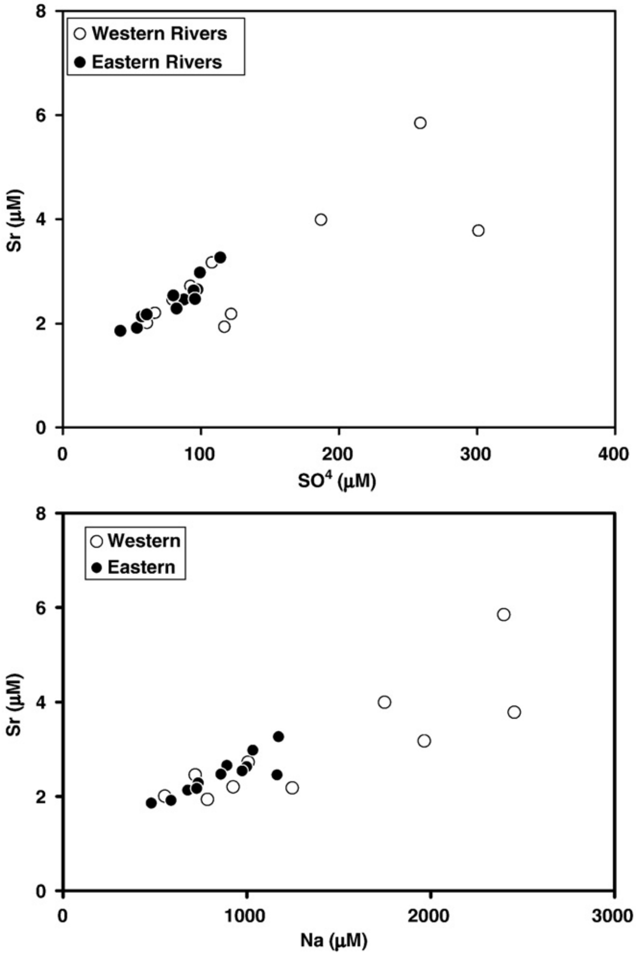

Fig. 7. Scatter plots of Sr vs. SO_4 and Sr vs Na. Good correlations between Sr–SO_4 and Sr–Na are observed, indicative of a common source for them, such as alkaline/saline/ evaporitic salts.

127) μM. Every mole of CO_2 consumption releases one mole of $(HCO_3)_{sil}$ to solution. The mean $(HCO_3)_{sil}$ at Udi is (531 ± 250) μM. This multiplied by runoff 215 mm y^{-1}, yields CO_2 consumption rate of $(1.1 \pm 0.6) \times 10^5$ mol km^{-2} y^{-1}. This estimate is lower by factors of 4 to 7 compared to that reported for the Yamuna river at Batamandi (Dalai et al., 2002, Table 4) and factors of 3 to 8 relative to those for the Deccan Trap basalts (Dessert et al., 2001, 2003; Das et al., 2005b). The low runoff and relief of the Chambal and low effective area of exposure of basalt in its basin, all contribute to its low SER and ΦCO_2.

5.3.1. Sr and $^{87}Sr/^{86}Sr$ isotope ratio in the Chambal system

In the Chambal system, dissolved Sr exhibits an overall positive correlation with SO_4 ($r^2 = 0.69$, $p < 0.001$, Fig. 7) and Na ($r^2 = 0.74$, $p < 0.001$). The Sr–SO_4 relation is similar to that observed for the Yamuna River system (Dalai et al., 2003). These correlations could arise if Sr, Na and SO_4 all have a common source such as alkaline/saline soils. Fig. 8 is a $^{87}Sr/^{86}Sr$ vs. Mg/Sr mixing plot of water samples from the Chambal river system. The end-members which can contribute Sr to the Chambal rivers are: the Deccan Trap basalts ($^{87}Sr/^{86}Sr$ 0.704 to 0.716; Peng et al., 1998), Vindhyan carbonates ($^{87}Sr/^{86}Sr$ 0.707 to 0.710; Ray et al., 2003), saline/alkaline soils and *kankar* carbonates. The Deccan Trap basalts of the Mhow region through which the headwaters of the Chambal system flow is known to be contaminated significantly by crustal materials. This has contributed to making these basalts more radiogenic in $^{87}Sr/^{86}Sr$ with values as high as 0.71610 in some locations (Peng et al., 1998). There are no data on the $^{87}Sr/^{86}Sr$ of the saline/alkaline soils and *kankar* carbonates, however considering that these are formed from rivers of the Chambal system, the range in their $^{87}Sr/^{86}Sr$ is expected to be same as that in river waters, 0.709 to 0.712 (Table 1). The Mg/Sr ratios of these end members are also unavailable. The Mg/Sr ratio released from basalts to rivers is taken to be the same as that in basalts. This is based on the studies of Das et al. (2006) who have shown that Sr and Mg weather nearly congruently from the Deccan Trap basalts. The data in Fig. 8 seem to show that they lie along a mixing line between the Deccan Trap basalts/carbonates and a low Mg/Sr end-member. This low Mg/Sr end-member remains to be identified, a potential candidate, however, is saline/alkaline soils. The strong correlation of Sr with Na and SO_4 (Fig. 7) is supportive of this suggestion.

The Sr_{sil} concentration in the Chambal rivers is also calculated following the approach used for major ions. Das et al. (2006) observed that the average Sr/Ca and Sr/Mg in the Krishna headwaters were same within errors as their ratios in the Deccan Trap basalts indicating congruent release of Mg, Ca and Sr from basalts to rivers. Based on this observation and available data on Mg/Sr in basalts and Mg/SiO_2 release ratio, the Sr/Si molar ratios released from basalts to rivers is calculated to be ~0.00051 ± 0.00013. This ratio, when multiplied with the dissolved Si concentration of the four sub-basins yields Sr contribution in the range of (0.17 ± 0.07) μM from the Deccan Trap basalts to the Chambal rivers. This contribution is <10% of the measured average dissolved Sr concentration. These calculations also assume conservative behaviour of Si in rivers.

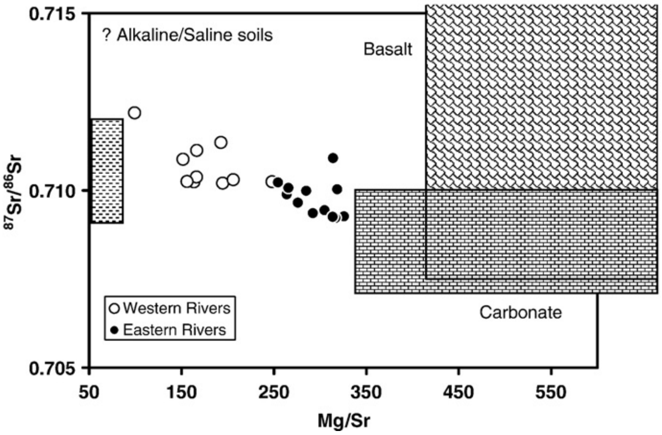

Fig. 8. Plot of $^{87}Sr/^{86}Sr$ vs. Mg/Sr in the water samples from the Chambal river system along with ratios of various end members. The range in $^{87}Sr/^{86}Sr$ of saline/alkaline soils is assumed to be same as that of the river waters, as Sr in these soils are derived from them. The basalt end member is based on the composition of the basalts of Mhow area of the Deccan Traps. These basalts are contaminated with crustal material and have $^{87}Sr/^{86}Sr$ in the range of 0.70585 to 0.71610 (Peng et al., 1998). Carbonate end-member values are taken to be the same as those reported for the Vindhyan carbonates (Ray et al., 2003) as well as from Gaillardet et al. (1999).

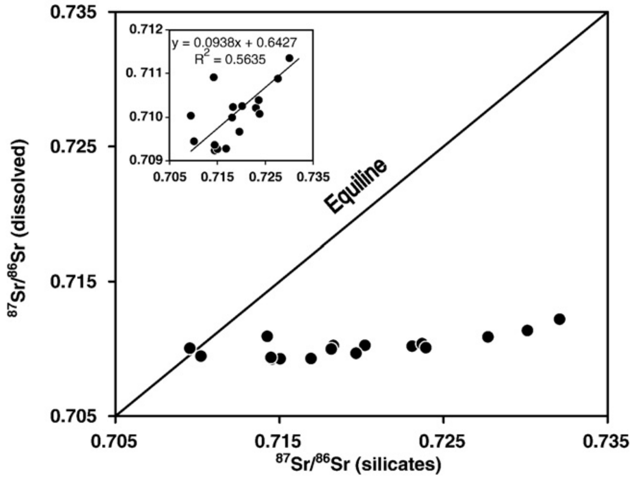

Fig. 9. Plot of dissolved $^{87}Sr/^{86}Sr$ in the Chambal head waters vs. $^{87}Sr/^{86}Sr$ of silicate phase of bed/bank sediments from the same locations. The inset shows the same data plotted with the *Y*-axis scale considerably expanded. It is seen from the inset that though $^{87}Sr/^{86}Sr$ of the Chambal waters vary with $^{87}Sr/^{86}Sr$ of sediment silicates, the dissolved $^{87}Sr/^{86}Sr$ are far less radiogenic in Sr than that of the sediment silicates. This suggests that the contribution of silicates to the dissolved Sr budget is only minor.

Fig. 9 presents the relation between dissolved $^{87}Sr/^{86}Sr$ in the Chambal rivers and the $^{87}Sr/^{86}Sr$ in the silicate fraction of the bed bank sediments collected from the same location. A cursory look at the plot shows that the dissolved $^{87}Sr/^{86}Sr$ is nearly independent of silicate $^{87}Sr/^{86}Sr$. A closer look at the data, however, indicates a marginal increase in dissolved $^{87}Sr/^{86}Sr$ with increasing silicate $^{87}Sr/^{86}Sr$ (inset Fig. 9). The trend in Fig. 9 brings out the important role of Sr input to these rivers from sources less radiogenic in $^{87}Sr/^{86}Sr$ relative to bed sediment silicates. The high $^{87}Sr/^{86}Sr$ in sediments of the Banas system rivers and the Aamjar is attributed to contribution from the Vindhyan silicates. Sr isotope composition of the waters is determined by Sr contribution from silicates, carbonates and saline/alkaline soils. The $^{87}Sr/^{86}Sr$ of these end members though fall in the same range, efforts have been made to place constraints on the silicate Sr contribution to the Sr isotope budget based on their ratios and conventional mass balance equation. The $^{87}Sr/^{86}Sr$ of the combined carbonate and saline/alkaline soils end member is 0.709 (range: 0.707–0.712). The $^{87}Sr/^{86}Sr$ of the silicate end-member is taken to be the same as that of the silicate fraction of sediments collected from the same location as water. This estimate indicate that on average ~12 ± 10% of Sr in the Chambal waters is derived from silicates, consistent with the earlier estimate based on Sr budget.

5.3.2. Impact of the Chambal on dissolved Sr flux and $^{87}Sr/^{86}Sr$ isotope ratio of the Ganga

The $^{87}Sr/^{86}Sr$ of the Ganga at the foothills of the Himalaya at Rishikesh is ~0.7425 (Krishnaswami et al., 1992). This decreases to ~0.725 at its outflow in Bangladesh due to dilution in the Ganga plain with less radiogenic Sr from the peninsular rivers. This study has provided an opportunity to estimate the flux of Sr from the peninsular rivers and its impact on the $^{87}Sr/^{86}Sr$ of the Ganga. This has been done

Table 5
$^{87}Sr/^{86}Sr$ of the peninsular rivers, the Yamuna and the Ganga

Rivers	Location	Date	$^{87}Sr/^{86}Sr$	References
Chambal	Jawahar Sagar Dam	09/98	0.7103	This work
Chambal	Dholpur	09/82	0.7127	Krishnaswami et al. (1992)
Chambal	Dholpur	03/82	0.7111	Palmer and Edmond (1992)
Chambal	Dholpur	11/83	0.7115	Palmer and Edmond (1992)
Betwa	Hamirpur	03/82	0.7125	Palmer and Edmond (1992)
Betwa	Hamirpur	11/83	0.7130	Palmer and Edmond (1992)
Ken	Before Confluence with Yamuna	11/83	0.7130	Palmer and Edmond (1992)
Peninsular average			0.7123	
Yamuna	Saharanpur	10/98	0.72657	Dalai et al. (2003)
Yamuna	Saharanpur	06/99	0.72657	Dalai et al. (2003)
Yamuna	Saharanpur	09/99	0.72624	Dalai et al. (2003)
Yamuna	Saharanpur	03/82	0.7270	Palmer and Edmond (1992)
Yamuna	Allahabad (after confluence with peninsular rivers)	09/82	0.7149	Krishnaswami et al. (1992)
Yamuna	Allahabad (after confluence with peninsular rivers)	03/82	0.7135	Palmer and Edmond (1992)
Ganga	Allahabad (before Yamuna Confluence	09/82	0.7282	Krishnaswami et al. (1992)
Ganga	Allahabad (before Yamuna Confluence	03/82	0.7298	Palmer and Edmond (1992)
Ganga	Varanasi (after Yamuna confluence)	09/82	0.7168	Krishnaswami et al. (1992)
Ganga	Varanasi (after Yamuna confluence)	03/82	0.7164	Palmer and Edmond (1992)
Ganga	Varanasi (after Yamuna confluence)	11/83	0.7182	Palmer and Edmond (1992)

based on the dissolved Sr isotope composition in the Chambal along with available data for the Yamuna and the Ganga at Allahabad and Varanasi respectively (Krishnaswami et al., 1992; Palmer and Edmond, 1992). The major tributaries of the Yamuna draining the peninsular India are the Chambal, Betwa and the Ken. Earlier studies (Krishnaswami et al., 1992; Palmer and Edmond, 1992) show that the $^{87}Sr/^{86}Sr$ of these tributaries (Table 5) range between 0.7111 to 0.7130 over a period of a year or so, with a mean of 0.7123 and a maximum difference of 0.0019 among them. (The $^{87}Sr/^{86}Sr$ of the Chambal headwater at the outflow of Jawahar Sagar Dam, CH-22, measured in this study is 0.7103, marginally lower than the earlier reported values at Dholpur. This is because the Dholpur sample represents the Chambal after mixing with its tributaries). The average $^{87}Sr/^{86}Sr$ of 0.7123, closely matches the value of 0.7127 of the Chambal during monsoon (Table 5). In light of this $^{87}Sr/^{86}Sr$ of the Chambal river during monsoon (0.7127) within an uncertainty of ±0.0019 has been used as representative annual value for all these three rivers. The contribution of Sr from the peninsular drainage to the Yamuna river at Allahabad and to the Ganga downstream Allahabad has been estimated using material balance equation:

$$\left(\frac{^{87}Sr}{^{86}Sr}\right)_{YA} = \left(\frac{^{87}Sr}{^{86}Sr}\right)_{P}(f_P) + \left(\frac{^{87}Sr}{^{86}Sr}\right)_{Yus}(1 - f_P) \qquad (3)$$

where, $\left(\frac{^{87}Sr}{^{86}Sr}\right)_{YA}$ and $\left(\frac{^{87}Sr}{^{86}Sr}\right)_{Yus}$ are Sr isotope composition of the Yamuna river at Allahabad (0.7149±0.0014) and Saharanpur (upstream of the confluence with peninsular rivers, 0.7266±0.0008) respectively, $\left(\frac{^{87}Sr}{^{86}Sr}\right)_{P}$ is Sr isotope composition of the peninsular rivers (0.7127±0.0019), f_p is the fraction of Sr supplied by the peninsular rivers to the Yamuna. In all the above, Sr isotope data of monsoon samples (September) have been used and the errors represent the maximum difference in a sample set. Similarly, the contribution of Sr from the peninsular rivers to the Ganga via the Yamuna is estimated using the end-member $^{87}Sr/^{86}Sr$ values of (0.7282±0.0016) and (0.7168±0.0018) for the Ganga at Allahabad and at Varanasi respectively. The uncertainties in the end-member values will decrease if standard deviations based on the available $^{87}Sr/^{86}Sr$ at a given site are considered instead of using the maximum difference.

Mass balance calculation shows that, at Allahabad, the rivers from peninsular drainage contribute 84±17% of the dissolved Sr to the Yamuna. Similar calculations for the Ganga at Varanasi demonstrate that its dissolved Sr is a result of 14±17, 14±15 and 72±24% contributions each from the Ganga upstream Allahabad, the headwaters of the Yamuna and the peninsular rivers. Even at Rajshahi (Bangladesh), the Chambal contributes about 28% of Sr to the Ganga though it accounts only for ~6.5% of water discharge. This estimate is based on Sr concentration of 0.56 μM for the Ganga (Galy et al., 1999, measured during monsoon at Rajshahi) and 2.36±0.49 μM for the Chambal (average of Chambal (CH-22), Kali Sindh (CH-17) Parbati (CH-18) and the Banas rivers, Table 1). The value of 2.36 μM for the Chambal closely matches its earlier reported value of 2.29 μM at Dholpur (Krishnaswami et al., 1992). The concentration of Sr in samples collected from the Chambal, Betwa and the Ken during November (Palmer and Edmond, 1992) show that they are quite similar, ~3.8±0.3 μM. In light of this, extension of the above calculation to all these three rivers using the Sr concentration of the Chambal (2.36 μM) as the annual average, show that these rivers together account for ~47% of Sr to the Ganga at Rajshahi. These results suggest that the peninsular rivers of the Yamuna system draining the Deccan Trap and the Vindhyan regions contribute significantly to the Sr budget of the Ganga, disproportionately higher than their contribution to water discharge. The peninsular Sr, being nearly un-radiogenic, dilutes the Sr isotopic composition of the Ganga in the plain.

6. Flux of Na from the Peninsular Rivers to the Ganga: implication to SER of the Ganga

Na concentration of the Chambal river is 785 μM at the outflow of the Jawahar Sagar Dam (CH-22). This compares with the value of 851±206 μM for the average concentration of Na in the Chambal, Kali Sindh, Parbati and the Banas system rivers measured in this study. This value yields a flux for $(2.6 \pm 0.62) \times 10^{10}$ mol y^{-1} for Na from the Chambal to the Ganga via the Yamuna. The Na flux of the Ganga, based on Na concentration measured at Rajshahi in August 1996 (Galy and France-Lanord, 1999) and water discharge at this location, is $\sim 8 \times 10^{10}$ mol y^{-1}. It can be inferred from these fluxes that about a third of Na exiting the Ganga at Rajshahi is derived from the Chambal. The major uncertainty in the Na flux of these rivers arises from seasonal variations in their Na concentration and the inter-annual variations in their water discharge. As the Na content of these rivers is measured during monsoon, they can be considered as their annual average as most of the water flow in these rivers is during monsoon. The inter-annual variations in the water discharge of the Ganga at its outflow based on available water discharge data of the Ganga at Farakka between 1949 to1973 (UNESCO, 1971, 1993) is ±20%. This uncertainty in water discharge is similar to the 18% variation in the interannual rainfall in the Chambal basin between 1995 and 2004. These uncertainties when propagated yield values of ~ (2.6 ± 0.77) and $(8 \pm 1.6) \times 10^{10}$ mol y^{-1} for the fluxes of Na from the Chambal and the Ganga at Rajshahi respectively. These fluxes show that the Chambal contributes ~ (32±12) % of Na to the Ganga at Rajshahi, about five times its share of water discharge. Extending this calculation to the Betwa and the Ken, assuming their Na concentration to be the same as the Chambal, would suggest that these three rivers together supply ~ (55 ± 18) % of Na flux from the Ganga.

In the Chambal system, a significant fraction of Na as discussed earlier, is derived from saline/alkaline soils and anthropogenic inputs. Hence, attempts to estimate silicate weathering rate at the outflow of the Ganga using Na* as an index of silicate weathering can lead to the over-estimation of silicate erosion rates and associated CO_2 consumption. In some of the earlier studies of the Ganga system (Galy and France-Lanord, 1999; Krishnaswami et al., 1999; Dalai et al., 2002), Na* has been used as a proxy of silicate weathering. This assumption would be valid for the headwaters of the Ganga as Na* in these waters is by and large from silicate weathering, whereas for the Ganga in the plain and at its outflow, it may be suspect as part of Na* can be of non-silicate origin, e.g. from alkaline/saline soils. Further studies, based on better spatial and temporal coverage in sampling, particularly during peak discharge are needed to confirm this conclusion and evaluate its implications to estimates of SER and associated CO_2 consumption more quantitatively.

In contrast to Na, the contributions to Ca and Mg fluxes from these peninsular tributaries to the Ganga at Rajshahi are significantly lower ~ 11 ± 3 and ~ 17 ± 5% respectively similar to their contribution to water discharge within errors. Precipitation of Ca as calcite in the basin can be a cause for its lower contribution.

7. Conclusions

Dissolved concentrations of Sr isotopes and major ions in the headwaters of the Chambal river, a major tributary of the Yamuna, draining peninsular India have been measured (i) to determine the sources of major ions to the waters and silicate erosion rates and (ii) to evaluate contribution of major ions and dissolved Sr from the rivers draining peninsular India to the Yamuna and finally to the Ganga. Based on a synoptic study of the streams within the Chambal river system, it is observed that the TDS ranged from 181 to 547 mg L^{-1} with Na being the most abundant (molar) constituting 25–58% of total cations. The order of cation abundance in these stream waters is $Na \approx Ca > Mg > K$ and differs from the trend of the Himalayan rivers viz. $Ca \approx Mg > Na > K$.

The (Ca + Mg)/Alk equivalent ratio in general centres around 1, indicating CO_2 mediated weathering of (Ca + Mg) rich silicates of the Deccan Trap basalts and Vindhyan carbonates. A key observation is the large excess of Na over Cl, attributable, in addition to silicate weathering, to supply of non-chloride Na salts from alkaline/saline soils present in the catchment and from anthropogenic sources. The significant input of Na from alkaline/saline soils constrains the use of Na* (Na corrected for Cl) as an index of silicate weathering. The silicate erosion rates (SER) calculated based on reported (cation/Si) ratios released from the Deccan Trap basalts to rivers during chemical weathering and Si abundances in the Chambal rivers yield a value of $\sim 5.5 \times 10^3$ kg km^{-2} y^{-1} for the Chambal basin. The flux of CO_2 consumed during silicate weathering, ΦCO_2, is 1.1×10^5 mol km^{-2} y^{-1}. These values are significantly lower than those reported for the Yamuna and the Krishna headwaters. Lower runoff, relief and aerial exposure of the Deccan Trap basalts in the Chambal basin together contribute to its low SER and CO_2 consumption.

Mass balance calculations show that the rivers draining peninsular India contribute ~85% of the dissolved Sr to the Yamuna at Allahabad. The dissolved Sr of the Ganga, at varanasi, is made up of about 15, 15 and 70% contributions each from the Ganga upstream Allahabad, the headwaters of the Yamuna and the peninsular rivers respectively. Similarly, budget calculation for Na show that the Chambal accounts for about a third of its flux from the Ganga at Rajshahi, five times its share of water flux. This study highlights the key role played by the peninsular rivers in the budget of some of the dissolved major ions and Sr in the Ganga and the potential uncertainties associated with the use of Na* as an index of silicate weathering.

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Appendix A

Table A1
Composition of the Deccan basalts

End member	Na	K	Ca	Mg	Sr	Ca/Na	Mg/Na	$^{87}Sr/^{86}Sr$
	wt.%				μg g^{-1}	Molar ratio		
Mhow	1.80	0.50	7.31	3.33	217	2.35	1.76	0.7058–0.7161
σ	0.11	0.24	0.56	0.39	54	0.30	0.27	
Deccan average	1.85	0.39	7.3	3.74	228	2.27	1.91	0.704–0.719
σ	0.30	0.26	0.8	0.66	32	0.44	0.46	

Data from Peng et al. (1998) and Das et al. (2005b).

Table A2
Composition of saturation extracts of saline/alkaline soils from the Ganga plain*

Sample code	Location	Na	K	Mg	Ca	Cl	HCO_3	CO_3	SO_4	Reference
Pedon 25	Etah	167	0.6	1.1	0.7	48	14	33	74	Pal et al. (2003)
Pedon 24	Etah	104	0.4	0.5	1.3	18	41	42	5.5	Pal et al. (2003)
UP VIII	Kanpur	137	0.3	–	–	13	44	78	9.1	Bhargava et al. (1981)

*Concentration in meq./l averaged over top ion soil.

Table A3
Ca, Mg, Sr and $^{87}Sr/^{86}Sr$ in carbonates

	Ca	Mg	Sr	Ca/Mg	Sr/Mg	$^{87}Sr/^{86}Sr$
	wt.%		μg/g	Molar ratio		
Vindhyan Bhander Fm[a]				147 ± 53	0.05 ± 0.02	0.708 ± 0.001
Vindhyan Dolomite[a]				2.4 ± 1.2	$(4.5 \pm 6.4) \times 10^{-4}$	0.7096 ± 0.0036
Himalayan (Pc-C)[b]	24.3 ± 7.7	8.3 ± 4.7	109 ± 98	1.8 ± 1.2	$(3.6 \pm 3.9) \times 10^{-4}$	0.725 ± 0.043

[a] Ray et al. (2003) ; [b] Singh et al. (1998).

Table A4
Concentration of major ions (μM) in rainwater

Components	Agra[a]	Gopalpura[b]	Mallikadevi[c]	Average
pH	7.01	6.7	6.4	6.7 ± 0.3
Cl	31.8	30.6	31.0	31 ± 0.6
SO_4	18.2	7.7	16.4	14 ± 6
Na	18.4	19.4	41.3	26 ± 13
K	7.6	2.5	23.0	11 ± 11
Ca	28.1	67.2	27.4	41 ± 23
Mg	22.8	39.2	12.3	25 ± 14

[a] Kumar et al. (2002); [b] Satsangi et al. (1998); [c] Mahadevan et al. (1989).

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