Silicate and carbonate weathering in the drainage basins of the Ganga-Ghaghara-Indus head waters: Contributions to major ion and Sr isotope geochemistry

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The role of silicate and carbonate weathering in contributing to the major cation and Sr isotope geochemistry of the headwaters of the Ganga-Ghaghara-Indus system is investigated from the available data. The contributions from silicate weathering are determined from the composition of granites/gneisses, soil profiles developed from them and from the chemistry of rivers flowing predominantly through silicate terrains. The chemistry of Precambrian carbonate outcrops of the Lesser Himalaya provided the data base to assess the supply from carbonate weathering. Mass balance calculations indicate that on an average ~ 77% (Na + K) and ~ 17% (Ca + Mg) in these rivers is of silicate origin. The silicate Sr component in these waters average ~40% and in most cases it exceeds the carbonate Sr. The observations that (i) the $^{87}Sr/^{86}Sr$ and Sr/Ca in the granites/gneisses bracket the values measured in the head waters; (ii) there is a strong positive correlation between $^{87}Sr/^{86}Sr$ of the rivers and the silicate derived cations in them, suggest that silicate weathering is a major source for the highly radiogenic Sr isotope composition of these source waters. The generally low $^{87}Sr/^{86}Sr$ (≤ 0.720) and Sr/Ca (~ 0.2 nM/μM) in the Precambrian carbonate outcrops rules them out as a major source of Sr and $^{87}Sr/^{86}Sr$ in the headwaters on a basin-wide scale, however, the high $^{87}Sr/^{86}Sr$ (~ 0.85) in a few of these carbonates suggests that they can be important for particular streams. The analysis of $^{87}Sr/^{86}Sr$ and Ca/Sr data of the source waters show that they diverge from a low $^{87}Sr/^{86}Sr$ and low Ca/Sr end member. The high Ca/Sr of the Precambrian carbonates precludes them from being this end member, other possible candidates being Tethyan carbonates and Sr rich evaporite phases such as gypsum and celestite. The results of this study should find application in estimating the present-day silicate and carbonate weathering rates in the Himalaya and associated CO_2 consumption rates and their global significance.

1. Introduction

It is well recognised that the high $^{87}Sr/^{86}Sr$ and Sr concentration of rivers draining the Himalaya, particularly the Ganga-Brahmaputra (G-B) have significantly influenced the Sr isotope evolution of the oceans since the Cenozoic (Palmer and Elderfield 1985; Palmer and Edmond 1989; Richter *et al* 1992; Krishnaswami *et al* 1992). The source(s) contributing to the high $^{87}Sr/^{86}Sr$ of these rivers remains controversial, though many recent studies have addressed this topic as it has relevance to atmospheric CO_2 budget and global change (Edmond 1992; Krishnaswami *et al* 1992; Palmer and Edmond 1992; Derry and France-Lanord 1996; France-Lanord and Derry 1997; McCauley and DePaolo 1997; Quade *et al* 1997; Harris *et al* 1998; Blum *et al* 1998). The high $^{87}Sr/^{86}Sr$ of silicates (granites, gneisses and metasediments) in the drainage basins of these rivers have led to the suggestion (Edmond 1992; Krishnaswami *et al* 1992; Harris 1995) that they exert dominant control over the $^{87}Sr/^{86}Sr$ of rivers flowing through them. Palmer and Edmond (1992) based on the observation that the major ion chemistry of the G-B rivers is mainly regulated by

Keywords. Sr isotopes; major ions; silicate weathering; Himalayan rivers.

Proc. Indian Acad. Sci. (Earth Planet. Sci.), **107**, No. 4, December 1998, pp. 283–291

carbonate weathering (Sarin *et al* 1989; 1992) proposed that carbonates enriched in radiogenic strontium during metamorphism is responsible for their high ^{87}Sr/^{86}Sr. Recently Quade *et al* (1997) from Sr isotope measurements in carbonates from paleosoles and river particulate matter have attested to the suggestion that carbonates are an important contributor to the high ^{87}Sr/^{86}Sr of these rivers. Besides this, data on Sr isotope composition of carbonates from the Himalaya are sparse. Sarkar *et al* (1996) have reported ^{87}Sr/^{86}Sr in a few samples of carbonates from the Krol-Tal formations, Derry and France Lanord (1996) have quoted values of ≈ 0.710–0.714 for marbles from the Higher Himalayan Crystallines and Tethyan Sediments and ≈ 0.8 (with low Sr) for samples from the Lesser Himalaya.

Precambrian carbonates of the Lesser Himalaya are one of the most extensive carbonate deposits of the Himalaya through which many of the source waters of the Ganga-Ghaghara-Indus flow. Studies on the chemical and Sr isotope composition of these carbonates (Singh *et al* 1998) have provided insight into their role in contributing to the Sr isotopic budget of these rivers. These results and available data on the chemistry and ^{87}Sr/^{86}Sr of silicates from the drainage basins of the Ganga-Ghaghara source waters have been synthesised in this manuscript to determine their controls on the major ion chemistry and Sr isotope composition of these waters.

2. The data set

2.1 *Source waters and lithology of their drainage basins*

The source waters of the Ganga (Bhagirathi, Alaknanda and Bhilangna), the Ghaghara (Kali, Sarju, Ramganga) all originate in the Higher Himalaya and Tibet. Granites, gneisses and metasediments (Gansser 1964) are the dominant lithology of their drainage basins in this region. Presence of marbles along with

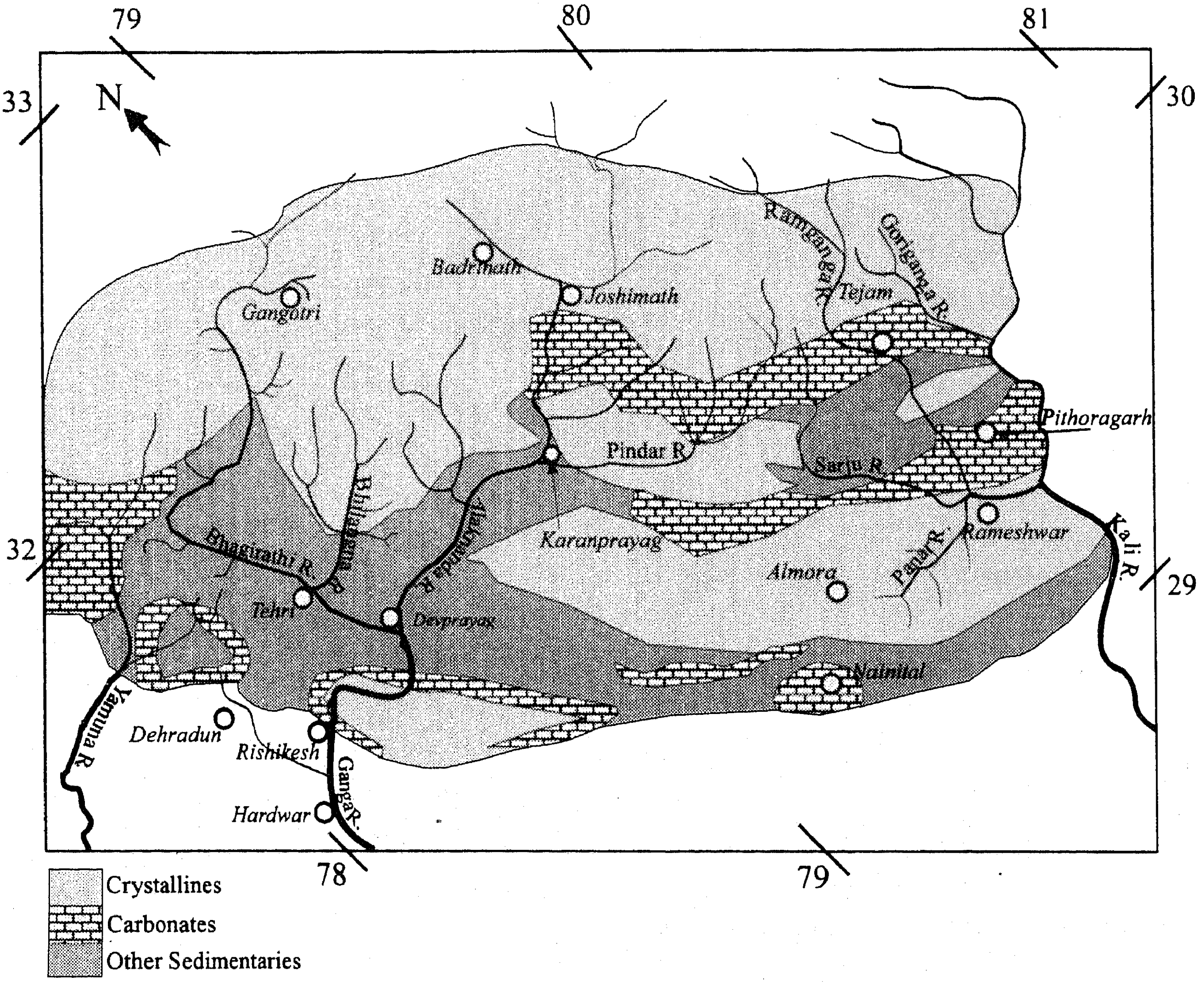

Figure 1. The source waters of the Ganga-Ghaghara and a broad lithological map of their drainage basins.

crystallines and calc-silicates have been reported in the drainage basins of the Alaknanda, Kali and Goriganga (Gansser 1964). Tethyan sediments containing carbonates are exposed in the Higher Himalaya and Tibet, in the drainage basins of Kali and Sutlej. A large fraction of the drainage basins of the source waters of the Ganga-Ghaghara and the Indus lie in the Lesser Himalaya, where in addition to the crystallines, Precambrian carbonates and sedimentaries constitute a major component of their lithology (Valdiya 1980). The chemical weathering in the Lesser Himalaya is likely to be more intense than in the Higher Himalaya because of relatively higher temperature, rainfall and the availability of more biogenic soil CO_2. Figure 1 shows a broad lithological map of the drainage basins of the Ganga-Ghaghara source waters.

2.2 *Contributions of silicate weathering to major ion chemistry*

The major ion chemistry of the source waters and their Sr isotopic composition are presented in some of our earlier publications (Krishnaswami *et al* 1992; Sarin *et al* 1992; Pande *et al* 1994; Trivedi *et al* 1995). The primary sources of major ions to these source waters are (i) atmospheric deposition, (ii) dissolution of evaporites and (iii) weathering of silicates and carbonates. To determine the silicate component of major cations and Sr in river waters it is necessary to apportion them into various sources. It is estimated from the available data on the chemical composition of rain, snow and glacier ice that atmospheric deposition on an average contributes about 15% of Na and K each to the source waters of the Ganga (Sarin *et al* 1992; Krishnaswami *et al* 1998); the supply to individual streams/rivers, however, could be different from the average estimated above. The dissolution of evaporites (halites and gypsum) can be an important source for Na and Ca in rivers. One approach to calculate the contribution of Na to rivers from atmospheric deposition and halite dissolution is to assume that it equals the chloride concentration in them (Sarin *et al* 1989, Krishnaswami *et al* 1998). Based on the above approach the silicate component of sodium and potassium in the source waters of the Ganga, Ghaghara and Indus (Sutlej, Beas, Chandra, Bhaga and Dharcha) can be calculated as:

$$(\mathrm{Na}_s + \mathrm{K}_s) = (\mathrm{Na}_r - \mathrm{Cl}_r) + 0.85\,\mathrm{K}_r \qquad (1)$$

where the subscript s and r refer to silicate and river. These calculations show that on an average ~ 77% of Na and K (range 53 to 90%) in these waters is of silicate origin (figure 2). The validity of these calculations requires that there are no alkaline/saline deposits containing carbonates/bicarbonates and sulphates of sodium along the drainage basins of these source waters, a condition very likely to be met for the rivers considered in this study. Estimation of silicate components of Ca and Mg (Ca_s and Mg_s) in these waters is a more difficult task and rely on a few simplifying assumptions which can be contentious. Calcium is supplied to rivers from the weathering of silicates and carbonates and dissolution of evaporites whereas the sources of Mg are silicates and carbonates. The silicate component of Ca and Mg in rivers can be derived if the ratio of (Ca/Na) and (Mg/Na) released to solution during silicate weathering can be ascertained. It is possible to place some constraints on these ratios (i) by assuming that they are the same as their abundance ratio in granites/gneisses of the Lesser Himalaya. This requires that Ca(Mg) and Na weather congruently from granites/gneisses, a condition which may be difficult to satisfy as silicate rocks generally weather incongruently; (ii) from the Ca(Mg) and Na abundances in granites/gneisses and in soils developed from them (Krishnaswami *et al* 1998). This approach would yield a time averaged value for (Ca/Na) and (Mg/Na) ratio released to solution; and (iii) by assuming that they are the same as those measured in rivers flowing predominantly through silicate terrains. In this case, minor abundances of carbonates and gypsum in the drainage basin can be important as they weather more easily relative to silicate rocks and thus can contribute to Ca and Mg budget of the rivers. In such cases, the (Ca/Na) and (Mg/Na) measured in the rivers is likely to be higher than those released from silicates.

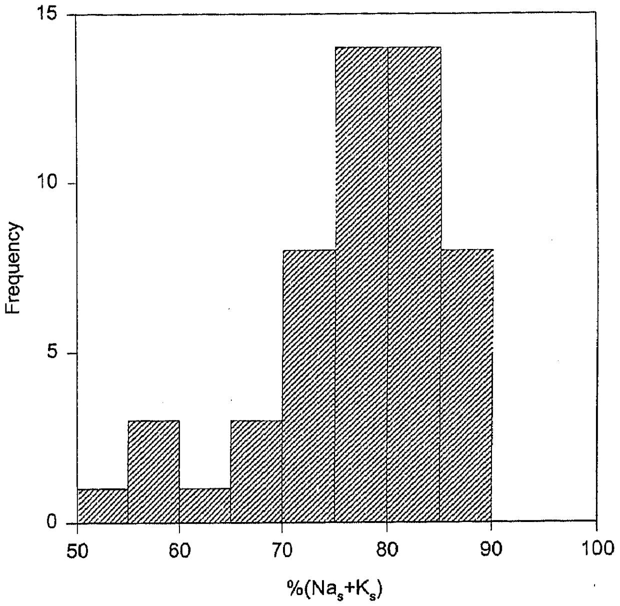

Figure 2. The fraction of silicate (Na + K) in the head waters of the G-G-I system. On an average silicate (Na + K) accounts for ~ 77% of their abundance in the rivers.

The mean Ca/Na abundance ratio (μmole/μmole) in granites/gneisses and metasediments of the Higher and Lesser Himalaya are in the range of 0.32 ± 0.29 to 0.46 ± 0.28 (Krishnaswami *et al* 1998). The (Ca/Na) ratio released to rivers during the weathering of these silicates would be 0.45 ± 0.3 if they weather congruently. The (Ca/Na) solution ratio, calculated (Krishnaswami *et al* 1998) from the soil profile data (Gardner and Walsh 1996) is 0.47 ± 0.08 nearly identical to those in the silicates. The measured (Ca/Na) in Jola gad, a tributary of Bhilangana draining predominantly silicate terrain is 1.15 (Sarin *et al* 1992, Singh *et al* 1998). Thus, the three approaches discussed above yield values in the range of 0.45–1.15 for Ca/Na ratio released to rivers from silicates during their weathering. Using a value of 0.7 ± 0.3 for Ca/Na solution ratio, the silicate Ca in the G-G-I source waters is estimated to range between 2% and 61% with a mean of 16% (Krishnaswami *et al* 1998). This compares with the reported value of ~ 17% for the G-B river system calculated based on Sr isotopic mass balance and Sr/Ca ratios in granites and carbonates (McCauley and DePaolo 1997).

The Mg/Na solution ratio determined from their abundances in granites/gneisses and from the Jola gad waters are similar and centre around ~ 0.5, much higher than that calculated from the soil profile data, 0.05–0.17 (Krishnaswami *et al* 1998). Considering these, the silicate Mg in the source waters have been calculated using a Mg/Na solution ratio of 0.3 ± 0.2. Thus, the sum of silicate Ca and Mg in these source waters, $(\mathrm{Ca}_s + \mathrm{Mg}_s)$, equals that of silicate Na, $[(\mathrm{Ca}_s + \mathrm{Mg}_s)/\mathrm{Na}_s = (1 \pm 0.35)]$.

The foregoing discussions have brought out some of the difficulties associated with the precise determination of Ca_s and Mg_s to the source waters of the G-G-I river systems. The approaches described above, however, provide reasonable estimates of Ca_s and Mg_s in these rivers. The fraction of silicate derived cations in these rivers, $(\Sigma\mathrm{Cat})_s$ is given by (Krishnaswami *et al* 1998)

$$(\Sigma\mathrm{Cat})_s = \left(\frac{\mathrm{Na}_s + \mathrm{K}_s + \mathrm{Ca}_s + \mathrm{Mg}_s}{\mathrm{Na}_r + \mathrm{K}_r + \mathrm{Ca}_r + \mathrm{Mg}_r}\right) = \left[\frac{(\mathrm{Na}_r - \mathrm{Cl}_r) \times 2 + 0.85\mathrm{K}_r}{\mathrm{Na}_r + \mathrm{K}_r + \mathrm{Ca}_r + \mathrm{Mg}_r}\right] \qquad (2)$$

The term, $(\mathrm{Na}_r - \mathrm{Cl}_r) \times 2$ equals the sum of silicate Na, Ca and Mg. Figure 3 presents the $(\Sigma\mathrm{Cat})_s$ in the G-G-I source waters in the form of histograms taking into consideration $\pm 1\sigma$ uncertainty in the $(\mathrm{Ca}_s + \mathrm{Mg}_s)$ estimation. These calculations though have been carried out for all the source waters analysed by our group (Sarin *et al* 1992; Pande *et al* 1994; Trivedi *et al* 1995; Krishnaswami *et al* 1998), they are likely to be more reliable for the larger streams/rivers which integrate contributions from different silicate lithologies. These estimates show that on an average the $(\Sigma\mathrm{Cat})_s$ in the G-G-I source waters is ~ 30%, a significant part of which is contributed by Na and K. The silicate (Ca + Mg) in these waters average only about 15%–20% of their measured concentrations, much unlike (Na+K) which is predominantly of silicate origin. These calculations suggest that the bulk of (Ca + Mg) in these waters have a non-silicate source, carbonates and evaporites for Ca and carbonates for Mg.

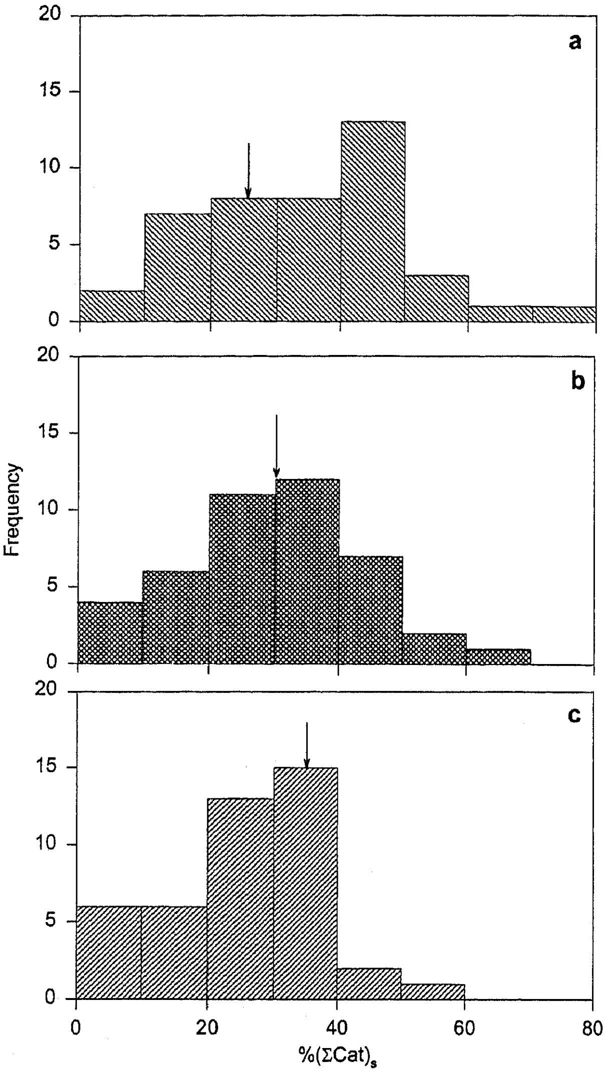

Figure 3. $(\Sigma\mathrm{Cat})_s$ in the head waters of G-G-I system. Three histograms are shown. (**a**) $(\mathrm{Ca}_s + \mathrm{Mg}_s) = 0.65\mathrm{Na}_s$; (**b**) $(\mathrm{Ca}_s + \mathrm{Mg}_s) = \mathrm{Na}_s$; and (**c**) $\mathrm{Ca}_s + \mathrm{Mg}_s = 1.35\mathrm{Na}_s$. Figures (a) and (c) are calculated by considering $\pm 1\sigma$ uncertainty in the estimates of $(\mathrm{Ca}_s + \mathrm{Mg}_s)$. On an average, $(\Sigma\mathrm{Cat})_s$ accounts for ~30% of the total cations in the source waters (figure b). The arrow represents the mean of the distribution.

2.3 *Ca and Mg from carbonate weathering*

The weathering of carbonates (limestone/dolomite) releases both Ca and Mg to rivers. Precambrian carbonate outcrops are abundant in the Lesser Himalaya along the drainage basins of the source waters of the G-G-I river systems (figure 1). The sources of Mg to rivers are silicates and carbonates, therefore the difference between the Mg concentration in rivers and the silicate Mg, calculated by the method described earlier, provides an estimate of carbonate magnesium, Mg_c, in these rivers. A similar approach to derive carbonate calcium, Ca_c, renders the results ambiguous as Ca can also be supplied by the dissolution of gypsum. Use of SO_4 concentration in rivers as an index of gypsum derived Ca is also fraught with problems as SO_4 in rivers can be derived from two sources, dissolution of gypsum and oxidation of pyrites. Thus, from the available data it is not possible to quantitatively apportion the measured Ca in rivers among its three primary sources. An upper limit of carbonate Ca can, however, be estimated by assuming that there is no supply of Ca from gypsum i.e., Ca budget in these source waters is governed by only two sources, silicates and carbonates, analogous to that of Mg. The upper limit of (Ca_c+Mg_c) in these rivers, thus estimated range between 43% and 98% of their measured concentration (Krishnaswami *et al* 1998). These results also show that the ($Ca_c + Mg_c$) component in the Alaknanda and the source waters of the Ghaghara and the Indus is generally higher relative to that in the Bhagirathi and the Bhilangna, broadly consistent with the lithologies of their drainage basins (figure 1).

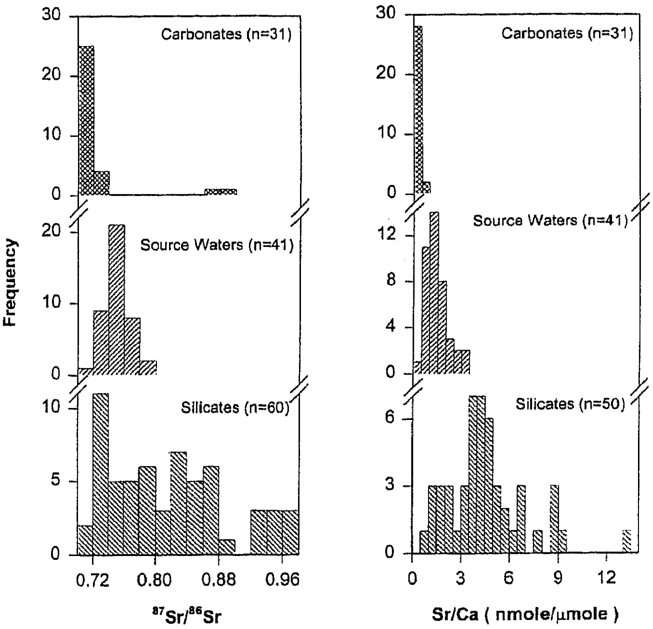

Figure 4. Comparison of $^{87}Sr/^{86}Sr$ and Ca/Sr in G-G-I source waters, silicates (granites/gneisses) and Precambrian carbonate outcrops of the Lesser Himalaya. The data show that the bulk of the carbonates have $^{87}Sr/^{86}Sr$ and Ca/Sr much less than those measured in the head waters (Singh *et al* 1998). Granites/gneisses having $^{87}Sr/^{86}Sr > 1.0$ though reported, they are excluded from the histogram.

2.4 *Sr and $^{87}Sr/^{86}Sr$ budget of G-G-I source waters*

The relevance of silicate weathering in the Himalaya and the associated CO_2 consumption to long term global change (Raymo and Ruddiman 1992) has prompted the search for suitable "proxies" to characterise silicate weathering. In this context identifying the major source(s) for the high $^{87}Sr/^{86}Sr$ in the G-G-I source waters has gained significance. It has been suggested that the weathering of Precambrian granites/gneisses (Krishnaswami *et al* 1992), leucogranites (Edmond 1992) and metasediments (Harris 1995) can all serve as sources to the highly radiogenic Sr isotopic composition of these waters. In addition, the dominant control of carbonate weathering on the major ion chemistry of these waters has led Palmer and Edmond (1992) to hypothesise that metamorphosed carbonates enriched in radiogenic Sr could also be an important contributor to the high $^{87}Sr/^{86}Sr$ of these rivers. Considering that most of the G-G-I source waters flowing along the Himalaya (east to west) have high $^{87}Sr/^{86}Sr$ with moderate Sr concentration (Krishnaswami *et al* 1992; Pande *et al* 1994; Trivedi *et al* 1995) for silicates and/or carbonates to be an important source it is required that (**i**) they have $^{87}Sr/^{86}Sr$ sufficiently high to account for the measured values in the rivers; (**ii**) they contribute significantly to the Sr budget of the rivers; and (**iii**) they occur widespread all along the Himalaya. The silicates, granite/gneisses and metasediments by and large satisfy the above requirements. They are ubiquitous all along the Himalaya. The $^{87}Sr/^{86}Sr$ of granites/gneisses from the Lesser Himalaya is quite radiogenic with typical values in the range of 0.71 to 1.0 (figure 4). The mean whole rock $^{87}Sr/^{86}Sr$ for this range, ≈ 0.78, overlaps with the highest $^{87}Sr/^{86}Sr$ measured in the source waters, 0.7986 ± 0.0008 for the Jola gad (Krishnaswami *et al* 1992). The whole rock Sr/Ca of the LH granites/gneisses are in the range of 0.85 to 25.5 (figure 4) which bracket the values measured in source waters (Singh *et al* 1998). Thus, the whole rock $^{87}Sr/^{86}Sr$ and Sr/Ca of silicates seem adequate to dominate the Sr concentration and its isotopic compositions in source waters, however, better understanding of the effects of their incongruent weathering is needed to assess more quantitatively their role on the riverine Sr isotope budget. The control of silicate weathering on the Sr isotope composition of rivers is evident from the plot (figure 5) of $^{87}Sr/^{86}Sr$ vs $(\Sigma Cat)_s$, the fraction of silicate derived cations. The data in figure 5 show a strong positive correlation ($r^2 = 0.56$) reflecting the control of silicate weathering on the $^{87}Sr/^{86}Sr$ of the source waters.

The silicate component of Sr in these source waters can be estimated following the procedure adopted for the major ions. The Sr/Na abundance ratio (mole) in the HH and LH granites and gneisses are 1.68 ± 0.72 and 1.22 ± 0.65 respectively (Krishnaswami *et al* 1998). The soil profile data yield values in the range of 2.5 to 2.9 for Sr/Na solution ratio, which compares with the measured Sr/Na molar ratio of 1.96 in the Jola gad waters. Following the assumptions employed for major ions, the Sr/Na ratio (molar) released to rivers during silicate weathering would be in the range of 1.2–2.9; for calculations a value of 2.0 ± 0.8 have been used (Krishnaswami *et al* 1998). The fraction of silicate Sr in the various source waters range between

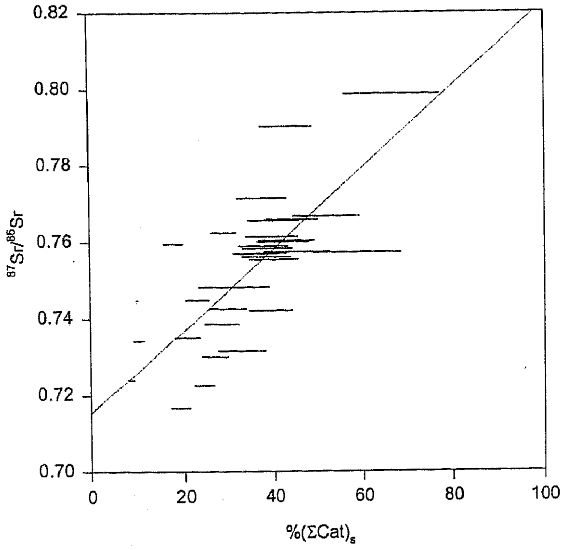

Figure 5. Variation of the measured $^{87}Sr/^{86}Sr$ in G-G-I source waters with their calculated $(\Sigma Cat)_s$. There is an overall positive correlation between the two ($r^2 = 0.56$) suggesting that silicate weathering is an important source for the highly radiogenic $^{87}Sr/^{86}Sr$ of the rivers. The bars represent the range in $(\Sigma Cat)_s$ calculated by considering $\pm 1\sigma$ uncertainty.

2% and 100% with the Bhilangna, Bhagirathi and their tributaries having higher Sr_s component, 30%–100% relative to the Kali, Sutlej, Chandra and Bhaga, which have $\leq 10\%$. The average Sr_s component in G-G-I source waters is 40%. McCauley and Depaolo (1997) have estimated the silicate Sr contribution to the G-B rivers to be 39% based on Sr isotopic mass balance assuming a two component system, silicates and carbonates.

In addition to silicates, another source which could control the Sr concentration and its isotopic composition in rivers is carbonates. The detailed studies of chemical and isotopic composition of Precambrian carbonate outcrops from the Lesser Himalaya (Singh *et al* 1998) provide necessary data to test their role on the Sr budget and $^{87}Sr/^{86}Sr$ of the source waters. The important findings are (i) these carbonates show definite evidence of alteration such as low Sr concentration with enriched $^{87}Sr/^{86}Sr$; (**ii**) the Sr/Ca ratio (mole) in them are only 0.04 to 0.64, with a mean of 0.2 ± 0.15 significantly lower than those in source waters (figure 4); and (**iii**) their $^{87}Sr/^{86}Sr$ are generally < 0.72, though there are a few samples with much higher values, ~ 0.85 (figure 4). These findings have led to the conclusion (Singh *et al* 1998) that these extensive exposures of Precambrian carbonate outcrops are unlikely to be a major contributor to the high $^{87}Sr/^{86}Sr$ of the source waters on a basin-wide scale, though they could be important for particular streams. This conclusion is not unexpected considering that for these carbonates to be important on a large scale their $^{87}Sr/^{86}Sr$ has to be considerably enriched all along the Himalayan range drained by the G-G-I source waters.

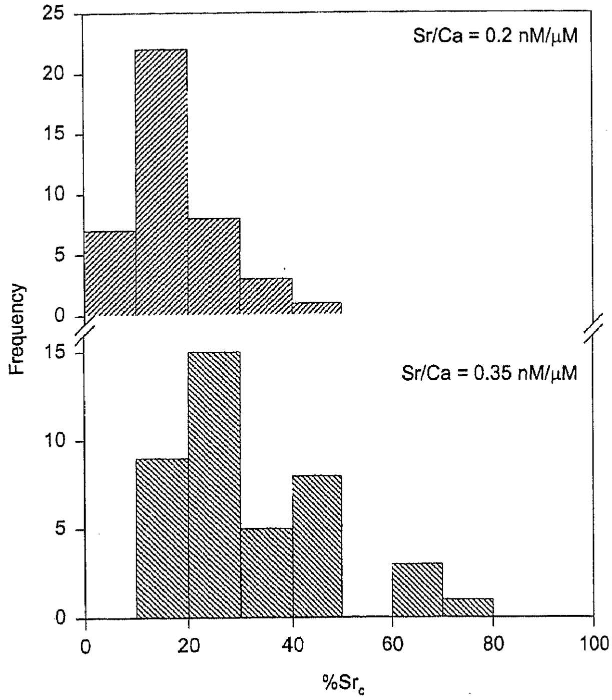

Figure 6. Carbonate Sr component in the G-G-I source waters. The values are upper limits and are calculated by assuming that all the Ca in the headwaters is of carbonate origin and Sr/Ca in the carbonates as 0.2, their mean abundance ratio in Precambrian carbonate outcrops (**a**) and 0.35, 1σ over the mean abundance ratio in carbonates (**b**).

How important are these carbonates in regulating the Sr budget of the source waters? This question can be addressed by estimating the carbonate Sr in them. The primary sources of Sr to rivers are silicates, carbonates and evaporites. As discussed earlier, it is not possible to estimate the evaporite contribution to major ions (and Sr) from the available data; however an *upper limit* on the carbonate Sr can be derived by assuming that (**i**) *all* Ca in the rivers is of carbonate origin; and (**ii**) the Sr/Ca ratio in the carbonates is $0.20 \pm 0.15\,nM/\mu M$, the mean value measured in the Precambrian carbonate outcrops (Singh *et al* 1998). The results (figure 6) show that the carbonate Sr can account for a maximum of 6 to 44% (mean 18%) of measured riverine Sr. (The limit would increase to 11 to 77%, (mean 31%) if the Sr/Ca ratio in the carbonates is taken as 0.35). A more realistic estimate of carbonate Sr can be calculated based on the carbonate Ca component of rivers (Krishnaswami *et al* 1998) which equals the difference between the measured Ca in them and their calculated silicate Ca. The results of these calculations yield a value of 5% to 42% (mean 15%) for the carbonate Sr contribution. The contribution of silicate and carbonate Sr in the source waters are compared in figure 7. The data, inspite of substantial uncertainty, show that in most of the source waters the silicate Sr component exceeds that of the carbonate Sr, exceptions being some of the tributaries of the Ghaghara and the Indus, where

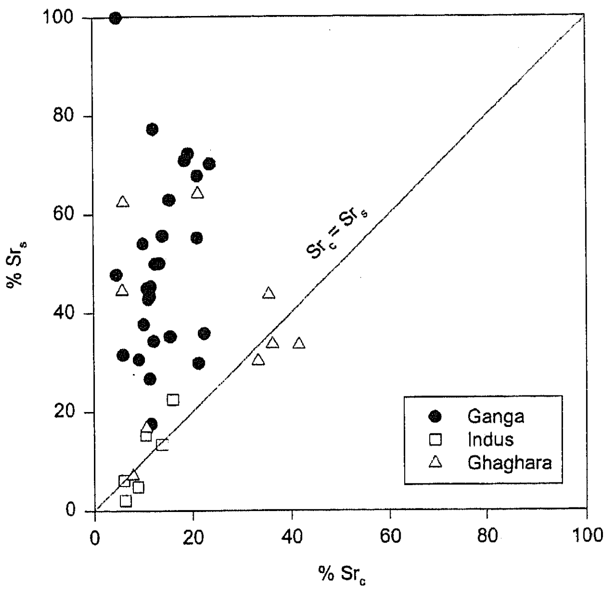

Figure 7. Scatter plot of Sr_s vs Sr_c (Sr_c calculated using Sr/Ca = 0.2). The silicate Sr exceeds the carbonate Sr in a majority of the headwaters. The data for the Ganga, Ghaghara and the Indus headwaters are shown by separate symbols. In Kali, Gori (Ghaghara system), Sutlej, Beas, Chandra, Bhaga, Dharcha (Indus system) $Sr_s \approx Sr_c$ (see text for calculations).

$Sr_s \approx Sr_c$. Thus the extensive outcrops of the Precambrian carbonate deposits of the Lesser Himalaya do not seem to be a major source for both Sr concentration and $^{87}Sr/^{86}Sr$ in the source waters on a large scale, they could, however, be still important for particular streams.

Can the silicates and the Precambrian carbonates account for Sr balance in the source waters? The fraction of riverine Sr that is supplied from the weathering of silicates and Precambrian carbonates is presented in figure 8 which show that only in a few of the source waters the Sr budget can be balanced if input is solely from these sources. It is possible to balance Sr budget in a few more rivers in which the sum of silicate and carbonate Sr contribution exceeds 70% by considering 1σ uncertainties associated with the estimates (figure 8). It is seen from this exercise that in a number of source waters, particularly Kali, Gori (Ghaghara system), Sutlej, Beas, Chandra, Bhaga and Dharcha (Indus system) it is difficult to account for their measured Sr if the two end members chosen (silicates and Precambrian carbonates) are its only sources and if the approaches used for estimating their contributions are valid. These calculations point to the need for another source for balancing the Sr budget in some of these source waters. The analysis of Sr isotope data of these rivers also seems to suggest this inference. The plot of $^{87}Sr/^{86}Sr$ vs Ca/Sr in the source waters (figure 9; note that unlike in earlier discussions where the Sr/Ca abundance ratios were compared, in figure 9 Sr isotope ratios are plotted as a function of Ca/Sr, following the conventional mixing diagrams) shows that the data fan out from a low Ca/Sr and low $^{87}Sr/^{86}Sr$ end member, similar to the trend in the $^{87}Sr/^{86}Sr$ vs 1/Sr plot (Singh *et al* 1998) suggesting the need for more than two end

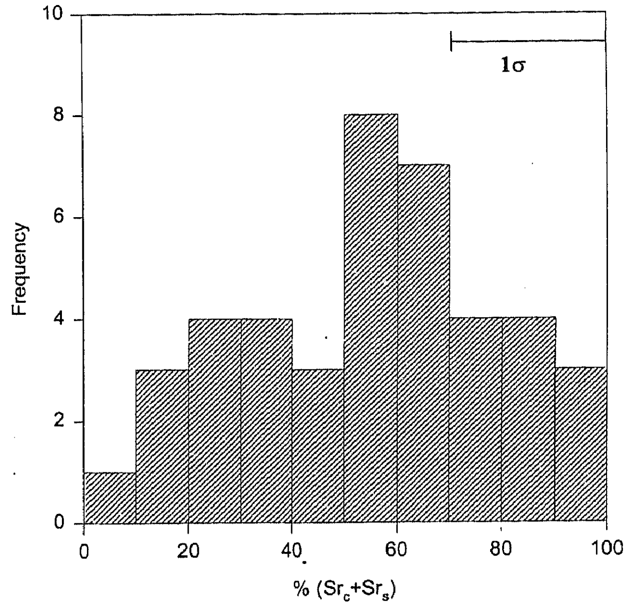

Figure 8. The sum of silicate and carbonate Sr contributions to the Sr budget of the G-G-I source waters. The data show that only in a few of the head waters, the Sr budget can be balanced based on the approaches discussed in the text. Samples with ($Sr_s + Sr_c$) contribution > 70% can be made to balance the budget by considering 1σ uncertainty.

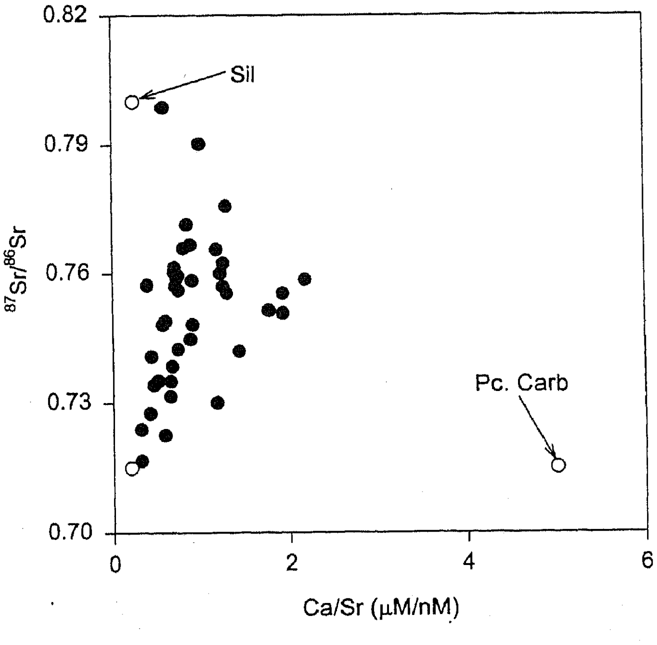

Figure 9. Plot of measured $^{87}Sr/^{86}Sr$ vs Ca/Sr in the headwaters. The end member values for silicates (Sil: $^{87}Sr/^{86}Sr = 0.8$; Ca/Sr = 0.2μM/nM) and Precambrian carbonates (Pc. Carb: $^{87}Sr/^{86}Sr = 0.715$; Ca/Sr = 5) are also given. The data fan out from a low $^{87}Sr/^{86}Sr$ and low Ca/Sr end member. Possible candidates for this end member are Tethyan carbonates and Sr rich evaporite phases.

members to explain the data. The low $^{87}Sr/^{86}Sr$ and Ca/Sr (i.e. high Sr) end member is commonly taken to be carbonates. The results of Singh *et al* (1998) and those presented in this study (figure 9) suggest that the extensive outcrops of Precambrian carbonates of the Lesser Himalaya because of their relatively lower Sr abundance (i.e. high Ca/Sr) are not the low $^{87}Sr/^{86}Sr$ and low Ca/Sr end member. Possible candidates for this end member are the Tethyan carbonates (present in the drainage basins of some of the Ghaghara and Indus source waters) and/or evaporites rich in Sr (gypsum, celestite etc.). Very recently, Blum *et al* (1998) based on the chemical and Sr isotope study of Raikhot watershed from the Higher Himalayan crystallines have suggested that weathering of vein calcites contributes to their high $^{87}Sr/^{86}Sr$. These vein calcites are inferred to have high $^{87}Sr/^{86}Sr$ (~ 0.8) and Ca/Sr ~ 5 (Blum *et al* 1998) and thus preclude them from being the end member with low $^{87}Sr/^{86}Sr$ and low Ca/Sr (figure 9).

3. Summary

Studies on the silicate weathering rates in the Himalaya and associated CO_2 consumption have gained considerable importance recently because of their possible influence on long term global change. Such studies naturally require "ideal proxies" to derive silicate weathering rates and its temporal variations. Sr isotope though have proved to be informative in this context, proper interpretation of $^{87}Sr/^{86}Sr$ in rivers needs better understanding of the various sources contributing to the Sr budget and their magnitudes. The major emphasis of this manuscript has been to estimate the contributions of silicates and carbonates to the major ion and Sr budget of the G-G-I source waters. The results show that on an average about a third of the major cations in these rivers is of silicate origin and that silicates exert a dominant control on their high $^{87}Sr/^{86}Sr$. Precambrian carbonate outcrops, a major component of the lithology of the head waters in the Lesser Himalaya, though may contribute to the high $^{87}Sr/^{86}Sr$ in particular streams, they are unlikely to be important for the Sr budget and the high $^{87}Sr/^{86}Sr$ of the headwaters on large, basin-wide scale. The Sr budget of many of the headwaters is difficult to balance if its supply is only from the two sources, granites/gneisses and Precambrian carbonates from the Lesser Himalaya, the data, therefore, indicate the need for an additional Sr rich source with low $^{87}Sr/^{86}Sr$.

Acknowledgements

This work is a synthesis of our continuing investigations on the major ion and Sr isotope geochemistry of the rivers draining the Himalaya. Discussions with Drs. M M Sarin and J R Trivedi who have contributed significantly to these investigations over the years have been very helpful and productive. We thank Mr. Tarun K Dalai for his help with the synthesis and Prof. J Edmond and an anonymous reviewer for comments on an earlier version of the manuscript.

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