Carbonate versus silicate weathering in the Raikhot watershed within the High Himalayan Crystalline Series

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ABSTRACT

The major element and Sr isotope geochemistry of surface waters, bedrock, and river sands was investigated in the Raikhot watershed within the High Himalayan Crystalline Series (HHCS) in northern Pakistan. Mass-balance calculations of mineral-weathering contributions to the dissolved flux of ions from the watershed indicate that 82% of the HCO$_3^-$ flux is derived from the weathering of carbonate minerals and only 18% is derived from silicate weathering, even though the bedrock is predominantly quartzofeldspathic gneiss and granite with only ~1% carbonate in the watershed. This study demonstrates the importance of trace amounts of bedrock carbonate in controlling the water chemistry of glacial watersheds. We suggest that the flux of Sr with a high $^{87}$Sr/$^{86}$Sr ratio in the major Himalayan rivers may be derived in large part from weathering of trace amounts of calcite within the largely silicate HHCS. Models that use the flux of radiogenic Sr from the Himalayas as a proxy for silicate weathering rates may, therefore, overestimate the amount of CO$_2$ consumption due to silicate weathering in the Himalaya.

INTRODUCTION

The Himalayan uplift has been implicated as a major factor in the global climatic cooling of the past 40 m.y. because of enhanced silicate weathering and consequent atmospheric CO$_2$ drawdown (Raymo and Ruddiman, 1992). Supporting evidence for this mechanism is the dramatic increase in marine $^{87}$Sr/$^{86}$Sr ratios at 40 Ma, shortly after the beginning of the Asia-India continental collision, which appears to be related to Sr inputs from major rivers draining the Himalaya (Palmer and Edmond, 1992; Edmond, 1992; Richter et al., 1992). Several workers have studied regional patterns of $^{87}$Sr/$^{86}$Sr ratios in tributaries of the Ganges-Brahmaputra and Indus River systems as well as bedrock $^{87}$Sr/$^{86}$Sr ratios in the high-radiogenic Sr in Himalayan rivers is weathering of silicate minerals in the HHCS (Krishnaswami et al., 1992; Pandit et al., 1994; Harris, 1995). Here we report on a geochemical investigation of a single watershed within the HHCS, with the aim of elucidating the systematics of Sr release from individual minerals in the HHCS bedrock. We chose the ~200 km$^2$ Raikhot watershed in the western Himalaya on the north side of the Nanga Parbat massif in northern Pakistan, because it is one of the most thoroughly studied areas in the HHCS with respect to bedrock geology and geochemistry.

STUDY SITE DESCRIPTION

The Raikhot watershed has its headwaters in the glaciated cirques of the 8125 m Nanga Parbat massif and drains ~28 km to the north terminating at 1200 m in the Indus River gorge. Currently, ~20% of the watershed is covered by glacial ice, and most of the watershed area is believed to have been covered by ice during the last glacial maximum. Much of the watershed is covered by steep cliffs and talus slopes, and only ~10% of the watershed is forested. The bedrock of the Nanga Parbat massif is predominantly high-grade quartzofeldspathic biotite gneiss and schist with dikes and small plutons of anatetic biotite granite. The gneisses and schists had metasedimentary protoliths that were predominantly pelitic sedimentary rocks. Minor amounts of calcareous sedimentary rocks and basaltic dikes give rise to thin layers and lenses of marble as well as calc-silicate and amphibolite schist, which together make up only a small (~1%) proportion of the outcrop exposure area. Bedrock mapping has been carried out in the ~50% of the watershed that is both ice free and accessible (e.g., Zeitler et al., 1993; Chamberlain et al., 1995), and observations of rock types in glacial moraines confirm that the unmapped parts of the watershed have bedrock similar to that of the well-mapped areas.

Although the protolith ages of these rocks are ca. 1.8 Ga, they have been metamorphosed and rapidly uplifted within the past 10 m.y. (e.g., Zeitler et al., 1993). As a result of this recent metamorphism, biotite and feldspars have young and nearly concordant Ar-Ar and RbSr ages (e.g., Zeitler et al., 1993; Gazis et al., 1995). The bedrock protolith and geologic history of this area are believed to be generally representative of the entire HHCS, which extends over 2000 km across the length of the Himalayan Mountain Range and dominates the geology of the steep southern slopes that are subjected to rapid physical erosion during the monsoon season (Harris, 1995). The ages of rocks currently at the surface in the Nanga Parbat region are, however, younger than those in much of the HHCS, which underwent peak metamorphism and granitic intrusion at 10 to 40 Ma (e.g., LeFort et al., 1987).

SAMPLING AND ANALYTICAL METHODS

Samples for analysis were taken of rainwater, snow, clear streams, Raikhot River water (which has abundant suspended glacial flour), Raikhot riverbed sand, bedrock from outcrops, and bedrock boulders collected on the surface of the Raikhot Glacier (Tables 1 and 2). All water samples were collected in acid-washed polyethylene bottles. Stream and river samples were filtered in the field through acid-washed 0.45 µm polypropylene filters, and at each sampling site an unfiltered sample was also collected and filtered 6 to 12 months later. Gran titrations were carried out in the field to determine carbonate alkalinity, and SO$_4^{2-}$, NO$_3^-$, and Cl$^-$ concentrations were determined in the laboratory by ion chromatography. NO$_3^-$ and Cl$^-$ concentrations are negligible, and the ratio of HCO$_3^-$/SO$_4^{2-}$ varies from ~10 in the Raikhot River to ~4 in the clear streams.

Rock samples containing calcite were leached for 1 h in 4N acetic acid to dissolve calcite for analysis. Silicate rock samples were digested with a LiBO$_2$ flux for elemental analysis and with hydrofluoric and perchloric acids for Sr isotope analysis. A sample of Raikhot riverbed sand (believed to be representative of unweathered bedrock from the watershed) was collected below the glacier terminus. It was sieved to <2 mm and split into two representative subsamples, one of which was left uncrushe and the other of which was crushed to a fine powder. Each of these two subsamples was first leached in 4N acetic acid, which was assumed to remove for measurement the average carbonate fraction present in the watershed, and then the remaining silicate material was digested in hydrofluoric and perchloric acids to allow analysis of the average silicate fraction.

Concentrations of major elements and Sr were measured to an accuracy of ±10% by inductively coupled plasma optical emission spectrometry, and Ca and Sr were measured on rain and snow by inductively coupled plasma mass spectrometry. Sr was separated with Eichrom Sr-specific resin; total procedural blanks were <70 pg of Sr. $^{87}$Sr/$^{86}$Sr ratios were measured on a Finnigan MAT 262 thermal-ionization mass spectrometer to a preci-
sion of at least ±0.000 020 (2σ). The 86Sr/88Sr ratio was normalized to 0.1194 and analyses of the NBS-987 standard yielded a mean 87Sr/86Sr ratio of 0.710 243 ± 0.000 022 (2σ, n = 44).

RESULTS AND DISCUSSION

Major Elements

The most abundant minerals in the bedrock of the watershed are quartz, plagioclase, K-feldspar, and biotite. Calcite is present in only minor abundance but is highly reactive in glacial environments (Anderson et al., 1997). In recently glaciated temperate environments, plagioclase and K-feldspar are assumed to weather to kaolinite, and biotite to vermiculite (or hydrobiotite) (e.g., Blum et al., 1994; Blum and Erel, 1997). The dissolved weathering products of calcite, plagioclase, K-feldspar, and biotite are plotted in a ternary diagram with apices (Na + K), (Si), and (Ca + Mg) (Fig. 1).

**TABLE 1. ANALYTICAL RESULTS FOR WATER SAMPLES**

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Filtered</th>
<th>Notes</th>
<th>Ca (µmol/L)</th>
<th>K (µmol/L)</th>
<th>Mg (µmol/L)</th>
<th>Na (µmol/L)</th>
<th>Si (µmol/L)</th>
<th>Sr (µmol/L)</th>
<th>87Sr/86Sr</th>
<th>% HCO₃⁻ from carb*</th>
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</thead>
<tbody>
<tr>
<td>Raikhot River</td>
<td>yes</td>
<td>gneiss</td>
<td>257</td>
<td>53</td>
<td>43</td>
<td>38</td>
<td>83</td>
<td>103</td>
<td>0.770 649</td>
<td>84.3</td>
</tr>
<tr>
<td>no</td>
<td></td>
<td></td>
<td>730</td>
<td>77</td>
<td>57</td>
<td>46</td>
<td>112</td>
<td>379</td>
<td>0.743 101</td>
<td>91.5</td>
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<tr>
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<td>yes</td>
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<td>740</td>
<td>70</td>
<td>52</td>
<td>28</td>
<td>89</td>
<td>375</td>
<td>0.734 718</td>
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<td>358</td>
<td>64</td>
<td>57</td>
<td>49</td>
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<td>171</td>
<td>0.766 901</td>
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<tr>
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<td>672</td>
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<td>425</td>
<td>0.750 599</td>
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<td></td>
<td>217</td>
<td>50</td>
<td>56</td>
<td>34</td>
<td>53</td>
<td>101</td>
<td>0.774 488</td>
<td>84.3</td>
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<tr>
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<td>445</td>
<td>51</td>
<td>42</td>
<td>36</td>
<td>67</td>
<td>296</td>
<td>0.749 649</td>
<td>90.6</td>
</tr>
</tbody>
</table>

* Calculated percent of HCO₃⁻ in water from carbonate dissolution; see text for explanation.

**TABLE 2. ANALYTICAL RESULTS FOR ROCK LEACHATES AND DIGESTS**

<table>
<thead>
<tr>
<th>Number</th>
<th>Notes</th>
<th>Ca (µmol/g)</th>
<th>K (µmol/g)</th>
<th>Mg (µmol/g)</th>
<th>Na (µmol/g)</th>
<th>Si (µmol/g)</th>
<th>Sr (µmol/g)</th>
<th>87Sr/86Sr</th>
<th>88Sr/86Sr</th>
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</thead>
<tbody>
<tr>
<td>P1590</td>
<td>gneiss</td>
<td>4419</td>
<td>1.73</td>
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<td>6.03</td>
<td>2021</td>
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<tr>
<td>P198a92</td>
<td>gneiss</td>
<td>4001</td>
<td>2.33</td>
<td>3.48</td>
<td>3.41</td>
<td>8.00</td>
<td>2466</td>
<td>0.722 258</td>
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<td>gneiss</td>
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<td>6.96</td>
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<td>0.719 859</td>
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<td>gneiss</td>
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<td>6.77</td>
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<td>9.27</td>
<td>2399</td>
<td>0.725 597</td>
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<tr>
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<td>gneiss</td>
<td>584</td>
<td>3.51</td>
<td>2.44</td>
<td>5.01</td>
<td>6.64</td>
<td>216</td>
<td>0.823 624</td>
<td></td>
</tr>
<tr>
<td>96R6b</td>
<td>gneiss</td>
<td>443</td>
<td>3.95</td>
<td>7.42</td>
<td>6.58</td>
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<td>0.741 695</td>
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<td>gneiss</td>
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<td>3.64</td>
<td>3.69</td>
<td>17.74</td>
<td>774</td>
<td>0.726 148</td>
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<tr>
<td>96R6e</td>
<td>gneiss</td>
<td>169</td>
<td>3.40</td>
<td>2.54</td>
<td>4.58</td>
<td>8.19</td>
<td>81</td>
<td>0.755 181</td>
<td></td>
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<tr>
<td>96S15</td>
<td>gneiss</td>
<td>125</td>
<td>16.51</td>
<td>20.30</td>
<td>9.85</td>
<td>6.08</td>
<td>97</td>
<td>0.743 220</td>
<td></td>
</tr>
<tr>
<td>96S15</td>
<td>sand*</td>
<td>92</td>
<td>3.37</td>
<td>11.58</td>
<td>1.94</td>
<td>2.54</td>
<td>66</td>
<td>0.723 124</td>
<td></td>
</tr>
</tbody>
</table>

Silicate digestes - hydrofluoric and perchloric acid

96S15  | river sand* | 323 | 801 | 193 | 841 | -------- | 1530 | 0.866 939 |
96S15  | river sand* | 365 | 885 | 185 | 888 | 12544 | 1597 | 0.861 837 |
96S15  | river sand* | 235 | 1136 | 20 | 1136 | 12381 | 719 | 0.887 760 |
96S15  | river sand* | 399 | 1384 | 298 | 732 | 11250 | 1792 | 0.823 580 |
95R4   | gneiss | 414 | 1499 | 624 | 678 | 10934 | 1849 | 0.818 230 |
95R5   | gneiss | 52  | 2278 | 754 | 161 | 9636   | 713  | 0.883 870 |

*Crushed

† Uncrushed

§ Total digest of samples after 4N acetic acid leach.

A mass-balance calculation was performed to quantify the proportions of each of the important bedrock minerals that weathered to yield each water composition. This six-step calculation—based on the stoichiometry of the assumed weathering reactions—also allowed the estimation of the proportion of HCO₃⁻ that was derived from silicate versus carbonate weathering (Table 1). (1) Assessment of the importance of atmospheric input to stream- and river-water compositions. Two rainwater and five snow-pack samples were collected and analyzed from the region. Concentrations were very low (<2.5 µmol/L for Ca, K, Mg, Na, and Si; <5 nmol/L for Sr); thus atmospheric correction is unnecessary, and omitting it does not affect any of the results of this investigation.

(2) Attributing of all Na in stream and river waters to the weathering of albite to kaolinite. According to this reaction when balanced, 2 mol of Si and 1 mol of HCO₃⁻ are also subtracted for each mole of Na. (3) Attributing of Ca in proportion to the Ca/Na ratio of plagioclase to the weathering of anorthite to kaolinite. We use the Ca/Na ratio of 0.38 measured in the silicate fraction of the riverbed sand as an estimate because plagioclase dominates the Ca and Na budgets of the silicate fraction. According to this reaction, 2 mol of HCO₃⁻ are subtracted for each mole of Ca.

(4) Attributing any remaining Si to the weathering of orthoclase to kaolinite. According to this reaction, 0.5 mol of K and 0.5 mol of HCO₃⁻ are subtracted for each mole of this Si. (5) Attributing the remaining K to the weathering of biotite to vermiculite. According to this reaction 1 mol of HCO₃⁻ is subtracted for each mole of K. (6) Attributing excess Ca and Mg to carbonate dissolution.

For the field-filtered Raikhot River samples, the above mass-balance calculation yields an estimate of the relative amounts of weathering of bedrock minerals as follows: 14% plagioclase, 2% orthoclase, 11% biotite, and 73% carbonate. This estimate corresponds to a riverine HCO₃⁻ flux that is 18% derived from silicate and 82% derived from carbonate weathering reactions. Separate analyses of the carbonate and silicate fractions of the riverbed sand indicate that only 1.0 wt% of the sediment load is carbonate (Table 2). The calculated percentages of weathering of bedrock minerals for the average quartzofeldspathic gneiss and granite bedrock stream waters were: 24% plagioclase, <1% orthoclase, 12% biotite, and 64% carbonate. High proportions of HCO₃⁻ are also de-

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derived from carbonate (68%–78%) in the stream waters known to drain exclusively quartzofeldspathic gneiss and granite bedrock.

**Ca/Sr Ratios and Sr Isotope Systematics**

The Ca/(1000Sr) ratios of silicate rocks (gneiss and granite) range from 0.07 to 0.4, whereas marble layers range from 1 to 2. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the silicate rocks (and all their constituent minerals; Gazis et al., 1995) range between 0.82 and 0.89, whereas marble layers range from 0.71 to 0.73. The riverbed sand silicate and carbonate samples are taken to be reliable average values for silicate and carbonate in the watershed. Surprisingly, in plots of both Ca/(1000Sr) versus $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 2) and Ca/(1000Sr) versus the percent of HCO$_3^-$ derived from dissolution of carbonate (Fig. 3), river and stream-water samples from the watershed do not plot on a mixing line between the average silicates and marbles. In both Figures 2 and 3, water samples plot within a triangle defined by silicate, marble, and an additional weathering end member, which must have a Ca/(1000Sr) ratio over 4.5 and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio over 0.82 to satisfy mixing relationships in Figure 2, and also must be calcite to satisfy mixing relationships in Figure 3.

Leachates of marble bands and the carbonate fraction of calc-silicate gneiss samples containing calcite range from Ca/(1000Sr) and $^{87}\text{Sr}/^{86}\text{Sr}$ values similar to the average defined by the riverbed sand up to values approaching those observed for silicate carbonate bands in high-grade metamorphic rocks (Bickle et al., 1995). Traces of finely disseminated calcite veins and blebs are also found commonly in thin sections of quartzofeldspathic gneiss and granite and associated with quartz veins in the watershed (Craw et al., 1994; this study). Although we have not measured it directly, we infer that this vein calcite end member has a Ca/(1000Sr) of >4.5 (Fig. 2), which is higher than that observed for most

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**Figure 1.** Ternary plot showing proportions of major elements released by weathering reactions. Quartz, calcite, and biotite dissolved weathering products plot at the three apices, and K-feldspar weathering products plot on edge of triangle. Position of plagioclase weathering products depends on Ca/Na ratio of plagioclase; average for watershed is shown as open circle with centered dot. Average water compositions of Sierra Nevada springs and Loch Vale, Colorado, watershed (Mast et al., 1990) are also shown as examples of water compositions that result from “normal” granitic weathering, and weathering of gneiss containing trace calcite, respectively. Raikhot watershed water analyses are shown as open symbols that are identified on plot.

**Figure 2.** Ca/(1000Sr) ratio plotted versus $^{87}\text{Sr}/^{86}\text{Sr}$ for water samples, rocks, and rock leachates. Arrows connect field-filtered and laboratory-filtered Raikhot River samples. Carbonate rock analyses are designated “m” for calcite from massive marble bands and “cs” for calcite from finely laminated calc-silicate-rich rocks. Vein-calcite end member is inferred on the basis of mixing relationships.

**Figure 3.** Ca/(1000Sr) ratio plotted versus calculated percent of HCO$_3^-$ in waters derived from carbonate dissolution, as described in text. Average marble and silicate rock values (solid squares) are estimated from Raikhot riverbed sand analyses. Vein-calcite end member is inferred on basis of mixing relationships.
conclude that the pervasive young metamorphism and
dissolution of carbonate minerals, which has no
Himalayan uplift was predominantly derived from
highly radiogenic Sr to the oceans related to the
single watershed can be extrapolated to the entire
pathic gneisses and granites. If the results from this
finely disseminated as veins within quartzofelds
that had reequilibrated with silicates having high
processes, and the strontium isotope record in ma-
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CONCLUSIONS AND IMPLICATIONS

We conclude that even in the predominantly
silicate Raikhot watershed of the HHCS, the cation
and HCO₃⁻ flux is predominantly derived from the
dissolution of a small amount of calcite (~1%) found in thin layers of metasedimentary rocks and
finely disseminated as veins within quartzofeldsp-
atic gneisses and granites. If the results from this
single watershed can be extrapolated to the entire
HHCS (and we think that they can, although the
extrapolation remains to be demonstrated), then
the implication of our finding is that the flux of
highly radiogenic Sr to the oceans related to the
Himalayan uplift was predominantly derived from
dissolution of carbonate minerals, which has no
net effect on rates of atmospheric CO₂ consump-
tion (e.g., Berner, 1994). Thus, models using the
flux of radiogenic Sr from the Himalayas that
assume this flux is due to silicate weathering may
overestimate the amount of CO₂ consumption due
to silicate weathering in the Himalayas.

Our conclusions are generally consistent with
large-scale regional studies of the Ganges-
Brahmaputra and Indus Rivers which have sug-
gested that about two-thirds of the dissolved
ations are derived from carbonate weathering and
about one-third from silicate weathering (Krishnaswami et al., 1992), although we add the
observation that carbonate weathering dominates
even in the predominantly silicate HHCS. Our
work supports the speculation of Palmer and
Edmond (1992) that the high Sr flux and 87Sr/86Sr
ratio of major Himalayan rivers are likely con-
trolled by carbonates with high 87Sr/86Sr ratios
that had reequilibrated with silicates having high
87Sr/86Sr ratios during metamorphism. We find
that the precipitation of disseminated calcite by
hydrothermal fluids plays a much more import-
ant role than previously recognized.

Quade et al. (1997) recently argued that meta-
morphosed carbonate rock has played a role in the
high 87Sr/86Sr ratios of the Himalayan rivers by
studying paleosols from the Himalayan foreland
basin. We concur with this conclusion and add the
finding that weathering of vein calcite in the high-
land catchments contributes an additional source of carbonate-derived Sr with even higher 87Sr/86Sr
ratios. We also support the conclusion that the
geologic record of 87Sr/86Sr variation in Himala-
ayan rivers through time (Derry and France-
Landor, 1996; Quade et al., 1997) cannot be used
to directly calculate the amount of CO₂ consump-
tion by silicate weathering in the Himalaya
(Quade et al., 1997). Instead, we find that weath-
ering reactions releasing the high 87Sr/86Sr flux of
dissolved Sr from the HHCS derive only ~18% of
their HCO₃⁻ from silicate weathering.

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