Uranium isotopes and radium in the Bhagirathi–Alaknanda river system: Evidence for high uranium mobilization in the Himalaya

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Extensive measurements of dissolved ^{238}U and ^{226}Ra concentrations and $[^{234}U/^{238}U]$ activity ratio have been made on samples collected from the Bhagirathi, Alaknanda and their tributaries—the source waters of the Ganga. The objectives of this study are to determine (i) the sources of U and Ra to the Ganga river; (ii) the weathering rate of uranium in the Himalaya, and (iii) the role of Himalayan–Tibetan rivers on the marine budget of uranium. The dissolved ^{238}U and ^{226}Ra concentrations in the Ganga source waters are typically $\sim 2\ \mu g l^{-1}$ and $\sim 0.2\ dpm l^{-1}$ respectively. The low ^{226}Ra concentrations relative to ^{238}U in these waters indicate that Ra is far less mobile. The Bhagirathi and Alaknanda weather uranium from their drainage basins at a rate of $\sim 2\ kg km^{-2} yr^{-1}$, comparable to that of the other Himalayan rivers like the Yamuna, Gandak and Ghaghara; but orders of magnitude higher than that derived for some of the world's major rivers (Amazon and Congo). These results suggest that large-scale mobilization of uranium in the Himalaya by rivers is ubiquitous. In the global context, the rivers draining the Himalayan–Tibetan region could be a major source of uranium to the oceans and that its supply via these rivers may have considerably influenced the marine budget of uranium.

ONE of the conspicuous characteristics of the Ganga river system is its high dissolved uranium concentration, $\sim 2\ \mu g l^{-1}$, compared to the global average concentration of $0.3\ \mu g l^{-1}$ in river waters[1,2]. The Ganga and the Brahmaputra, together, transport about 1000 tons of dissolved uranium to the estuaries of the Bay of Bengal annually[2]. Where does all this uranium come from? What is the relative contribution of uranium to the Ganga from the streams draining the high altitude Himalaya? What is the role of Himalayan rivers in the marine budget of uranium? To answer these questions, we have sampled the Ganga source waters—the Bhagirathi, the Alaknanda and their tributaries (Figure 1, *a*,*b*). The main lithological units drained by these rivers are the sedimentary and crystalline nappes[3,4]. The sedimentary units are made of carbonates, shales, slates and quartzites; whereas granites and gneisses are the primary constituents of the crystallines (Figure 1,*a*). For detailed lithological features of the drainage basin reference is made to Sarin *et al.*[5]. The Bhagirathi was sampled extensively from its source (at Gangotri) to its confluence with the Alaknanda at Devprayag (Figure 1,*b*). The two rivers merge with each other at Devprayag to form the Ganga.

Details of the sampling procedures have been described in Sarin *et al.*[2]. Briefly, uranium and radium isotopes were preconcentrated at site from ~20 litres of water within 24 h of collection. For uranium, water samples were filtered through 3-μm Gelman cartridge filters. The filtered water was acidified with nitric acid to pH ~1, spiked with ^{232}U and allowed to stand for ~6–8 h for equilibration. Uranium isotopes were extracted from water by coprecipitation with ferric

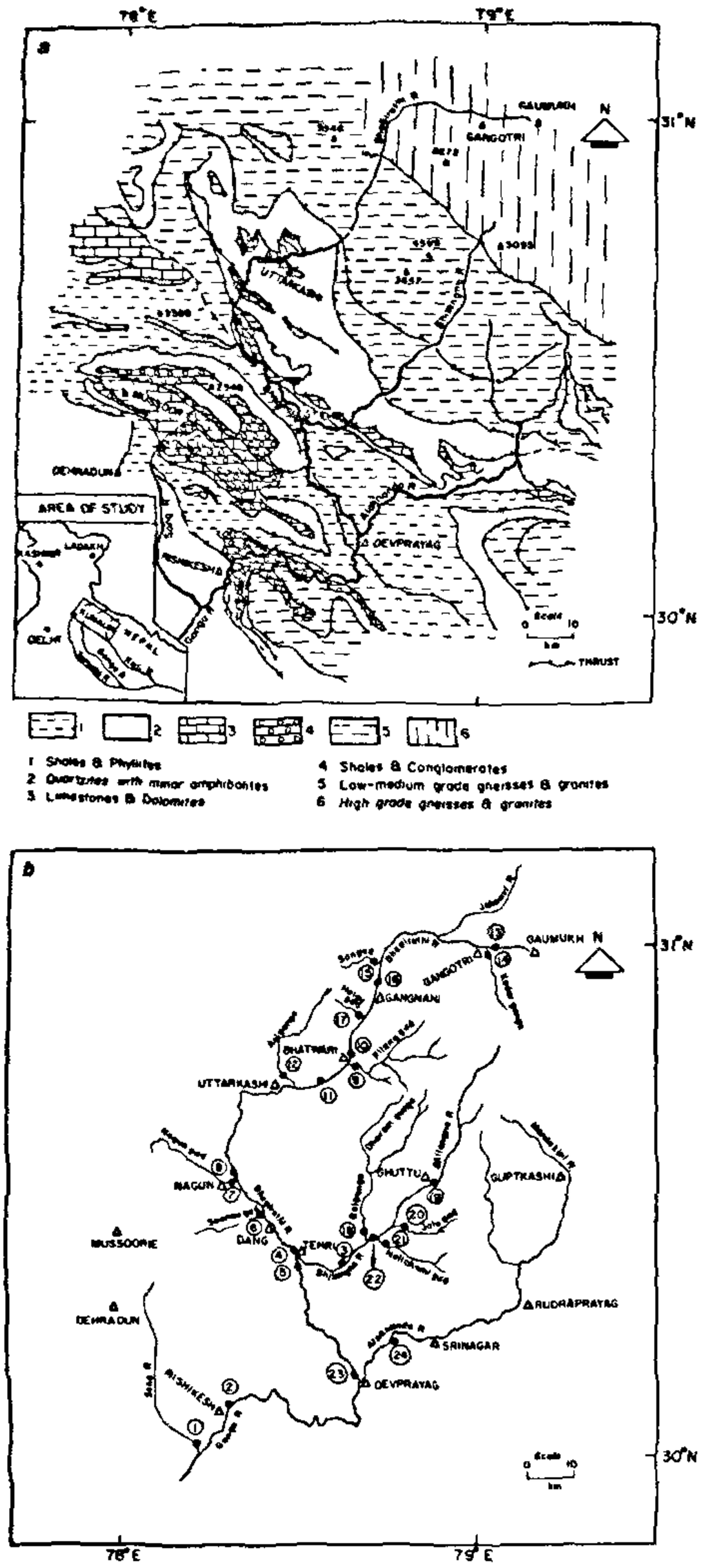

Figure 1. *a*, Lithologic map of the drainage basins of the Bhagirathi and Alaknanda rivers in the Kumaun Himalaya (from Valdiya[4].) *b*, Location map of the samples collected during April 1989, from the Bhagirathi, Alaknanda and their tributaries, see Table 1.

hydroxide; purified later on an anion exchange column and assayed by α-spectrometry[2]. Radium was preconcentrated from the filtered water by absorbing it onto MnO_2-coated acrylic fibres[6]. In the laboratory, the fibres were treated with 3 M hydrochloric acid and radium was measured in acid solution by the radon-emanation technique[7].

The dissolved uranium and radium concentrations and $[^{234}U/^{238}U]$ activity ratios are given in Table 1. The ratio given in square brackets represents activity ratio. The ^{238}U concentration in the Bhagirathi and its tributaries ranges between 0.15 and 3.96 $\mu g l^{-1}$; however, in most samples it centres around 2 $\mu g l^{-1}$, typical of the Ganga in its lower reaches[1,2]. It is noteworthy that the uranium concentration in the Bhagirathi, near its source (at Gangotri), is quite high (2.47 $\mu g l^{-1}$). Another important observation is that in all samples, excluding those from the Nagun gad, Seansu gad and Song, the ^{234}U activity is in radioactive equilibrium with ^{238}U (Table 1). The $[^{234}U/^{238}U]$ in rivers depends on factors such as the lithology of the drainage basin, the kinetics of weathering and $[^{234}U/^{238}U]$ in the source rocks. The rocks of the drainage basin undergoing intense chemical weathering and congruent dissolution would yield $[^{234}U/^{238}U]$, in the solution phase, quite similar to that in the source rocks. In most cases, $[^{234}U/^{238}U]$ in the host rocks is ~1.0. In the Bhagirathi and the Alaknanda, these factors contribute to the near-equilibrium value of $[^{234}U/^{238}U]$. The abundance and chemistry of major ions suggest that these rivers are characterized by intense chemical weathering and that the weathering of carbonate rocks could account for about 70–80% of the cations[5].

The samples from the Nagun gad, Seansu gad and Song (draining mainly carbonates) have high total dissolved solids (TDS) but low uranium concentration with distinct excess of ^{234}U (Table 1). This is not consistent with the interpretation given in the above paragraph; where one would expect high TDS content to be associated with high uranium concentration and $[^{234}U/^{238}U]$ of ~1.0. A likely explanation is that, in these streams the dissolved salts and uranium may have been derived from different source rocks: the dissolved salts from weathering of carbonates and other sedimentary rocks, whereas uranium from selective weathering of silicates. This can also account for the low uranium concentration and associated ^{234}U excess in these samples, the preferential leaching of ^{234}U relative to ^{238}U is well documented in rivers draining the silicate terrains[2,8,9].

Figure 2 is a scatter diagram of ^{238}U vs Σ cations (the sum of Na + K + Mg + Ca concentrations measured in these waters is expressed as Σ cations, data from ref. 5). The results fall into two groups: (i) a set of five samples which have low ^{238}U concentration ($\leqslant 0.3\ \mu g l^{-1}$) and do not show any trend with Σ cations, and (ii) the rest seventeen samples which show an overall positive correlation between ^{238}U and Σ cations, a relationship similar to that observed by us for the samples from the Ganga–Brahmaputra system[2] and the Narmada, Tapti

Table 1. Dissolved salts, uranium isotopes and radium in the Bhagirathi–Alaknanda river system

Sample	Stream	Σ cations* (mg l^{-1})	TDS* (mg l^{-1})	^{238}U ($\mu g\ l^{-1}$)	$[^{234}U/^{238}U]$	^{226}Ra (dpm l^{-1})
13	Bhagirathi	20	79	2.47	1.00	0.47
16	Bhagirathi	25	102	3.96	1.01	NM
10	Bhagirathi	23	96	2.74	1.02	0.21
11	Bhagirathi	24	99	2.11	1.01	0.61
8	Bhagirathi	24	98	2.85	1.03	0.29
4	Bhagirathi	25	107	2.97	1.02	0.16
5	Bhagirathi	24	102	2.22	1.03	0.24
23	Bhagirathi	24	104	2.45	1.02	0.23
14	Kedarganga	23	91	2.50	0.99	1.10
15	Son gad	19	78	0.33	1.04	0.09
9	Pilang gad	11	52	1.38	1.02	0.24
12	Asiganga	14	59	2.00	1.01	0.15
7	Nagun gad	34	151	0.27	1.78	0.0
6	Seansu gad	27	123	0.22	1.21	NM
19	Bhilangna	17	69	1.21	1.01	0.23
22	Bhilangna	18	76	1.87	1.03	0.17
3	Bhilangna	19	84	1.41	1.00	0.28
20	Jola gad	14	67	0.20	1.04	NM
18	Balganga	19	88	1.18	1.00	NM
24	Alaknanda	26	115	1.86	1.03	0.14
2†	Ganga	31	136	1.95	1.00	0.15
1	Song	52	245	0.15	1.44	0.07

*Data from Sarin *et al.*[5]. The errors ($\pm 1\sigma$) associated with radionuclide data are $\leqslant 3\%$ for ^{238}U and $[^{234}U/^{238}U]$; $\sim 5\%$ for ^{226}Ra.

†Ganga at Rishikesh, Figure 1*b*. NM, not measured.

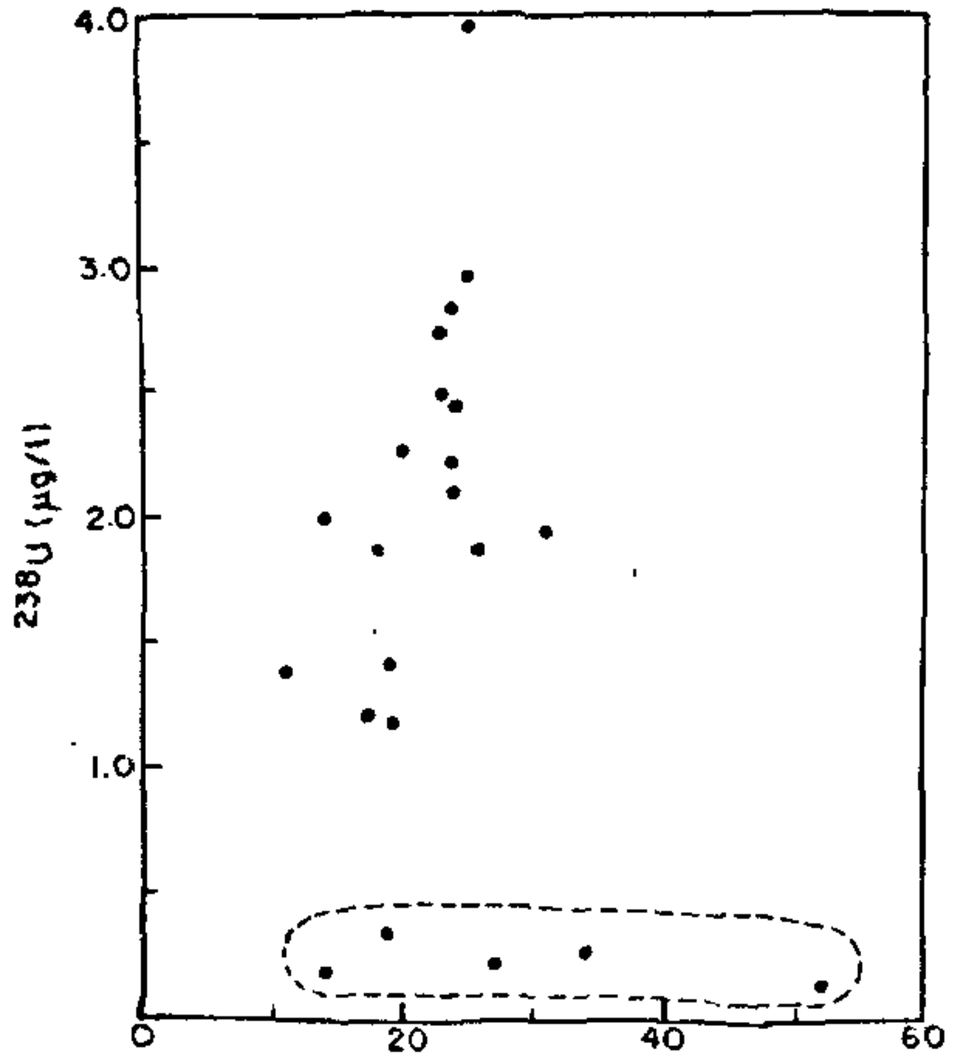

Figure 2. Plot of dissolved ^{238}U concentrations vs Σ cations. Majority of the samples show a positive correlation. Σ Cations data from Sarin *et al.*[5]

rivers[10]. This positive correlation is an indication that the concentration of uranium in these high-altitude streams (Table 1) is in some way dictated by the intensity of chemical weathering of their drainage basins. The scatter in the data (Figure 2) reflects on the variations in $^{238}U/\Sigma$cation ratio in the source rocks.

The ^{226}Ra concentrations of the samples exhibit a wide range, 0.02 to 1.10 dpm l^{-1}, typical of surface water values[2,9,11]. In all these samples, ^{226}Ra content is less than that of ^{238}U, on an average the $[^{226}Ra/^{238}U]$ is ~ 0.1. It is expected that the rocks and soils of the drainage basin would have ^{226}Ra in radioactive equilibrium with ^{238}U, i.e. $[^{226}Ra/^{238}U]$ of ~ 1. Therefore, the low $[^{226}Ra/^{238}U]$ observed in the samples has to be attributed to lesser mobility of ^{226}Ra relative to ^{238}U. This is consistent with the particle reactive behaviour of radium observed in freshwater systems[2,12].

Table 2 lists the flux of uranium transported out of the Himalaya by the Bhagirathi, Alaknanda and some of the major streams of the Ganga river system. For comparison, data on a few other major rivers of the world are also given in Table 2. For calculating the uranium flux (Table 2), we assume that the water discharge of the Bhagirathi at Tehri is the same as that at Devprayag. The average annual discharge at Tehri, for the period 1973–1981, is about 8.4×10^{12} litres[13]. The water discharge of the Alaknanda at Devprayag (Table 2) is estimated to be about 14×10^{12} litres per year from the data of Pal[14]. The most striking feature of the data (Table 2) is that the uranium weathering rate (flux of dissolved uranium transported by rivers per unit drainage area) for the Himalayan streams is about two orders of magnitude higher than that for rivers

Table 2. Dissolved uranium flux of selected Himalayan streams compared with some major rivers

River	Discharge (10^{12} l yr^{-1})	Area (10^3 km^2)	^{238}U (μg l^{-1})	^{238}U flux (ton yr^{-1})	^{238}U flux (kg km^{-2} yr^{-1})
Himalayan streams					
Bhagirathi*	8.3‡	7 8	2 45	20.5	2 63
Alaknanda*	14‡	11.8	1 86	26.2	2.22
Yamuna†	93	140	1.72	160	1 14
Ghaghara†	94	128	1.71	161	1 26
Gandak†	52	46	2.33	121	2 62
World rivers†					
Ganga	393	975	1.81	710	0 73
Brahmaputra	609	580	0 63	380	0.66
Mississippi	530	3270	0.93	493	0.15
Congo	1230	4104	0.08	98	0 02
Amazon	5500	6300	0.04	220	0.03

*This study
†Sarin *et al.*[2]
‡Water discharge at Devprayag[13,14].

such as the Amazon and the Congo. Such high uranium mobilization most likely results from the intense chemical weathering of the Himalaya. The weathering of biotite-rich granites may contribute significantly to the high uranium content in the upper reaches of the Bhagirathi[5,15] (Table 1, Figure 1). It is also likely that some of the uranium is contributed by uraniferous mineral zones in the drainage basin. In the Kumaun Lesser Himalaya, uranium mineralization has been reported at several locations along the Alaknanda and Bhilangna Valley[16,17] and in the Mussoorie phosphorites and black shales[18].

We have also estimated the ^{238}U flux of the major rivers draining the Himalayan–Tibetan region (Table 3). Data on the Ganga and Brahmaputra are based on the measured concentration of uranium[2]. For Indus, Chiang and Mekong, ^{238}U flux is estimated assuming that the ^{238}U/TDS ratio in these rivers is the same as that in the Brahmaputra. (The lithology of their drainage basins is more akin to that of the Brahmaputra.) Our estimates of the ^{238}U flux (Table 3) suggest that (i) the Himalayan–Tibetan rivers supply about 3000 tons of uranium to the oceans annually with a $[^{234}U/^{238}U] \sim 1.0$; (ii) the flux of uranium via the Himalayan–Tibetan rivers is an order of magnitude more than the combined supply of uranium from the Amazon and the Congo, although their water discharge is about thrice that of the Himalayan rivers, and (iii) the global flux of dissolved uranium via rivers to the oceans would be higher than the earlier estimates[2]; the Himalayan–Tibetan rivers being the dominant source representing about a fourth of the dissolved supply. Thus it appears that the intense chemical weathering in the Himalayan–Tibetan region is contributing significantly to the oceanic budget of uranium and that the Himalayan Orogeny has considerably influenced its marine geochemistry, similar to that of Rb and Sr isotopes[19]. The uranium concentration of the oceans during the Miocene, as inferred from the U/Ca ratio of corals[20], was about 30% lower than that of today. It is likely that the supply of dissolved uranium via the Himalayan–Tibetan rivers may have contributed to some of this increase.

Table 3. Dissolved uranium concentration and flux via the Himalayan–Tibetan rivers

River*	Discharge 10^{12} l yr^{-1}	TDS (mg l^{-1})	^{238}U (μg l^{-1})	^{238}U flux (ton yr^{-1})
Ganga	393	178	1.8	710
Brahmaputra	609	100	0.63	380
G–B	1002	131†	1.1†	1100
Indus	238	171	1.1‡	260
Chiang	1063	213	1.3‡	1400
Mekong	*577*	99	0.63‡	*360*

*Water discharge and TDS data from Sarin *et al.*[21], uranium data on the Ganga and Brahmaputra from Sarin *et al.*[2].
†Discharge-weighted average.
‡Estimated assuming that ^{238}U/TDS ratio in these rivers is the same as that in the Brahmaputra.

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