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Fluxes and sources of particulate organic carbon in the Ganga-Brahmaputra river system

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[1] Sources and fluxes of particulate organic carbon (OC) in the Ganga-Brahmaputra river system were estimated from $^{13}\text{C}/^{12}\text{C}$ ratios of bedrocks, soils, bank and suspended river sediments from a Himalayan watershed (Narayani, Nepal), and the Bangladesh floodplain. In the watershed of the Narayani, OC $\delta^{13}\text{C}$ values for bank and suspended sediments have similar values and narrow ranges, with a mean of about -24.2‰ at the base of the Himalayas. On the Bangladesh floodplain, bank and suspended sediments are enriched in ^{13}C ($\delta^{13}\text{C}$ about -22.5‰) relative to those collected at the base of the Himalayas. The OC exported at the base of the Himalayan range is estimated to include about 15% from the ^{13}C enriched C_4 biomass while the C_4 contribution is about 25% of the OC exported by the Ganga and Brahmaputra. A calculated total OC flux of $0.65 \times 10^{12} \text{ molC yr}^{-1}$ is exported to the ocean or trapped in the plain. In sediments of the Ganga and Brahmaputra, the clay-size fraction is depleted in ^{13}C ($<2.6\text{‰}$) relative to the bulk sediment. Possible explanations are that either the organic matter associated with clays is largely inherited from the Himalayan watersheds, or, and considered more likely, the clays and coarser sediments sample different OC pools, possibly with different ages (the growth of C_4 crops has dramatically increased since the mid twentieth century), on the floodplain. The OC budget of the actual Ganga-Brahmaputra system is broadly comparable to that derived from the Quaternary sediments of the Bengal fan that represents about 10% of the global OC contribution to the continental margins.

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1. Introduction

[2] Among world rivers the Ganga (or Ganges) and Brahmaputra rank first for sediment transport [Sarin *et al.*, 1989], with high erosion rates in the Himalayan range driving the high suspended sediment flux [Galy and France-Lanord, 2001]. The annual sediment discharge is $400\text{--}600 \times 10^6 \text{ t yr}^{-1}$ for the Brahmaputra and $330\text{--}550 \times 10^6 \text{ t yr}^{-1}$ for the Ganga [River Survey Project, 1996]. This generates an organic carbon (OC) burial flux that, associated with sediment trapping in the Bengal fan, has been estimated to be around $1.1 \times 10^{12} \text{ molC yr}^{-1}$ for the Neogene [France-Lanord and Derry, 1994, 1997]. It represents about 10% of the global burial flux of OC in continental margins [e.g., Keil *et al.*, 1997] and therefore contributes signifi-

cantly to the modulation of the ocean-atmosphere budget of CO_2 .

[3] High erosion rates and sediment discharge, organic matter composition [Hedges *et al.*, 1986; Leithold and Blair, 2001] and interaction of organic matter with mineral particles [Keil *et al.*, 1997] can be implicated as key factors for the preservation of riverine organic matter in the fan setting. This underscores the importance of elucidating the sources of the riverine organic matter in the Himalayan watersheds at the locus of physical erosion and of following organic matter loss and addition during the transit of particles in the floodplain. While Himalayan watersheds dominate the sediment production, one central question is whether the floodplain has any significant impact on the organic matter delivered to the fan. Floodplain processes that may affect the organic matter include photosynthesis, oxidation [Hedges *et al.*, 1986], sedimentation-resuspension of particles [Meade *et al.*, 1985; Aucour *et al.*, 2003a] and desorption-sorption of organic compounds on mineral surfaces [Keil *et al.*, 1997; Aufdenkampe *et al.*, 2001; Aucour *et al.*, 2003a]. It can be predicted that the fine and coarse fractions of organic matter would present strong differences in their behavior with regard to these processes [Hedges *et al.*, 1986]. The finer, clay-size, particles appear to play a

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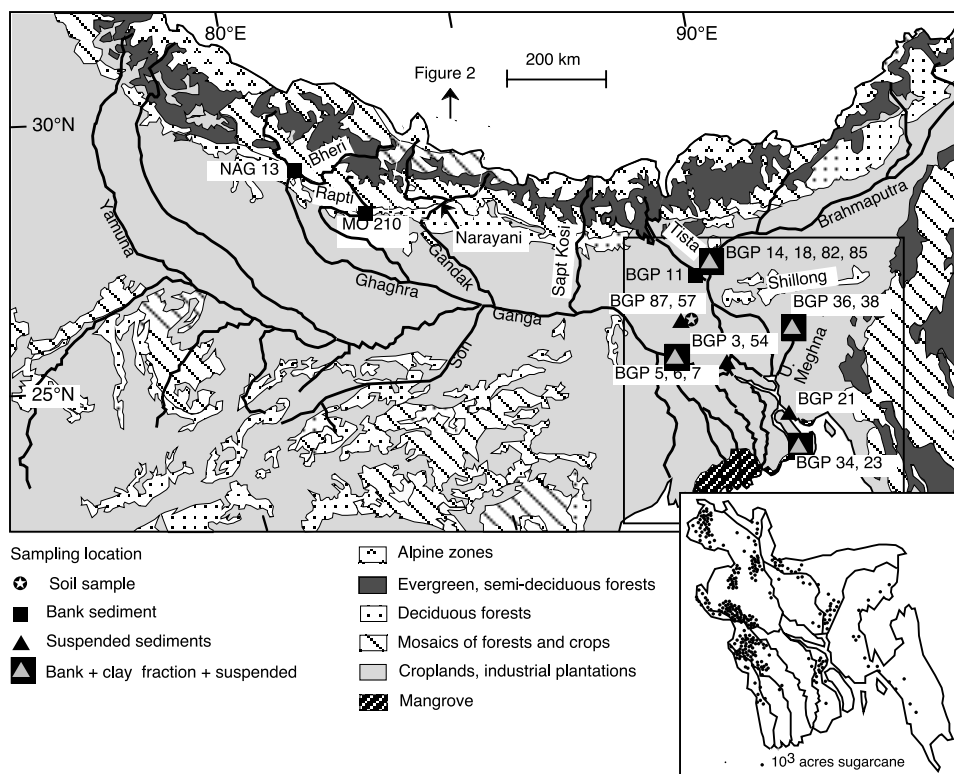


Figure 1. Map of the Ganga-Brahmaputra basin. The location of samples collected in Bangladesh (BGP) and in Western Nepal in the Bheri (NAG 13) and Rapti (MO 210). Vegetation types are drawn from *Blasco et al.* [1996]. The inset shows the distribution of sugar cane acreage (1 acre \approx 0.4 hm^2) in Bangladesh [*Rashid*, 1991]. The location of the Narayani watershed that is represented at a larger scale on Figure 2 is also shown.

major role in the riverine export of OC, by both their concentration in OC and refractory nature of their OC [*Hedges et al.*, 1986; *Keil et al.*, 1997].

[4] Previous studies of OC in the Ganga and Brahmaputra have documented the concentration and composition of organic matter in the dissolved and suspended load in the Indo-Gangetic plain [*Chowdhury et al.*, 1982; *Ittekkot et al.*, 1985; *Subramanian et al.*, 1985; *Safiullah et al.*, 1985]. They have provided insights into the sources and state of degradation of the organic matter near the mouth [*Ittekkot et al.*, 1985] and first estimates of particulate OC fluxes [*Subramanian et al.*, 1985]. Our main objectives here are to evaluate the sources of organic matter in the Himalayan hinterland and to follow the addition of the organic matter during the transit of particles on the floodplain. A further objective is to reestimate the whole flux of exported particulate OC and assess the contributions of the main sources to this flux.

[5] For the purpose of tracing the sources of organic matter in the Himalayan watersheds and the organic matter addition in the floodplain, we used $^{13}\text{C}/^{12}\text{C}$ ratios and OC concentrations in bedrocks, soils, suspended sediments and their clay fraction. Variations of the $^{13}\text{C}/^{12}\text{C}$ ratio of exported OC are linked to (1) the variable proportions of C_3 and C_4 terrestrial biomass on the watershed [*Hedges et al.*, 1986; *Bird et al.*, 1994; *Martinelli et al.*, 1999], (2) altitude effects on the isotopic composition of the C_3

terrestrial biomass [*Hedges et al.*, 1986; *Körner et al.*, 1988, 1991], (3) the contribution of OC inherited from bedrock relative to that inherited from soil and biomass [*Leithold and Blair*, 2001], and (4) the input of phytoplanktonic carbon [*Barth et al.*, 1998]. The stable carbon isotope composition of particulate organic carbon may vary with season in response to a change-over of the dominant source of organic matter [*Bird et al.*, 1998]. Within the river sections, it is expected to vary with depth and turbidity, reflecting algal blooms and/or size sorting of the particles. Size sorting effects are accounted for here by the study of the whole sediment and of its clay-size fraction. We then propose estimates for the total flux of exported particulate OC and of the contributions of the main sources to this flux using OC and ^{13}C mass balance considerations and previous budgets of solid matter in the system [*River Survey Project*, 1996; *Galy and France-Lanord*, 2001].

2. Vegetation and Geology

[6] The Ganga-Brahmaputra basin covers an area of about $1.6 \times 10^6 \text{ km}^2$ (Figure 1), including the Shillong plateau, the Indo-Gangetic plain and parts of the Himalayan range of the Tibetan plateau and Indian Shield. The basin is exposed to a monsoon climate characterized by heavy rainfall from May to November, with a peak in July and August. The main types of vegetation that occur in the basin are shown

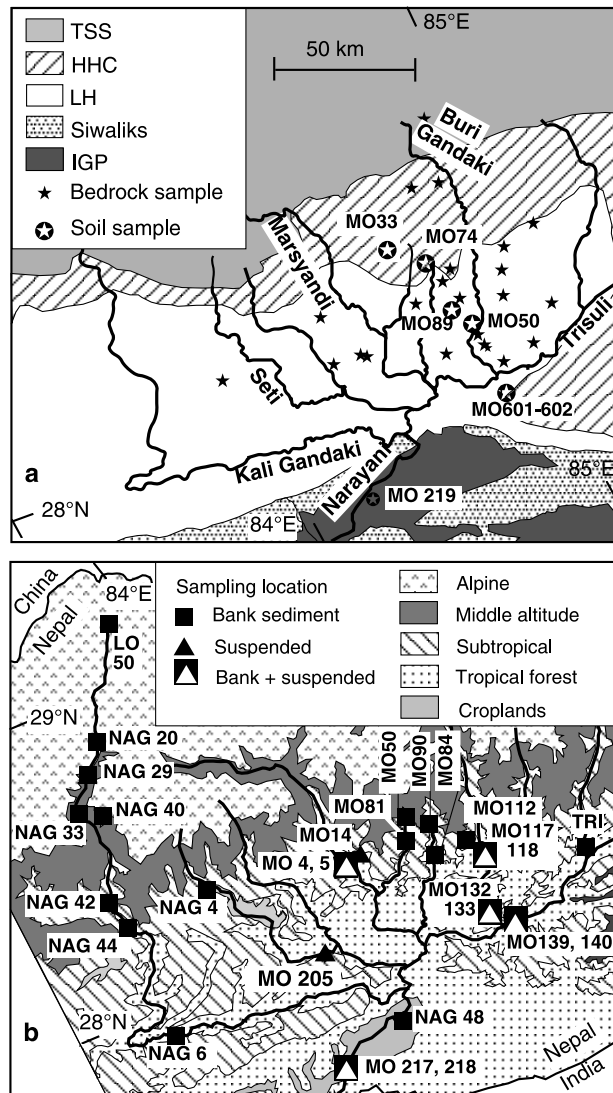


Figure 2. Location of the samples in the Narayani watershed (central Nepal). (a) Geological map showing the location of the bedrock and soil samples. (b) Ecological map showing the location of the river sediment samples. Vegetation levels are drawn from *Dobremez et al.* [1978]. TSS, Tethyan Sedimentary Series; HHC, High Himalaya Crystalline Series; LH, Lesser Himalaya; IGP, Indo-Gangetic Plain.

in Figure 1. The Indo-Gangetic plain is densely populated and agricultural activity is intense. Prior to clearance, tropical deciduous forests composed the vegetation cover. In the Himalayan range, the distribution of vegetation is mainly dictated by altitude (Figure 2b) [Blasco *et al.*, 1996]. In Nepal, the subtropical levels, between 1000 and 2000 m altitude, is intensely populated and deforested [Dobremez *et al.*, 1978]. Surviving subtropical forests of *Pinus roxburghii* and *Shima wallichii* are sparse. Mixed temperate coniferous forests are found in the middle altitudes of the Himalayas, between 2000 and 3000 m altitude. These have come under enormous pressure due to large-scale deforestation during

recent decades. Subalpine fir forests dominate altitudes between 2900 and 3500 m while alpine scrub grasslands extend between 3000 m and 5500 m.

[7] Areas that could deliver organic matter derived from C_4 biomass include patches of tropical grasslands [Sankhla *et al.*, 1975; Raghavendra and Das, 1978] and the areas cultivated with C_4 crops. The *Phragmites-Saccharum-Imperata* grassland is found in humid regions of the Gangetic plain and Brahmaputra valley. The *Dicanthium-Cenchrus-Lasiurus* and *Sehima-Dichanthium* grasslands occur in the western part of the basin. Subtropical grasslands of *Themeda-Arundinella* type are found in the Himalayas up to 2000 m altitude. C_4 crops now cover substantial areas of the basin (Table 1). The C_4 biomass, due to its concentration on periodically flooded areas, may provide relatively large amounts of exported carbon compared to its areas. The distribution of sugar cane (C_4) in Bangladesh (Figure 1) shows that this crop is concentrated in the Brahmaputra valley and along the Ganga levees. Sugar cane fields may also contribute more carbon to the river sediment as a result of their higher standing biomass per unit area and productivity than other crops (Table 1).

[8] The three main geological units that are encountered in the High Himalayan range all outcrop in the Narayani watershed (Figure 2a). The Tethyan Sedimentary Series (TSS) consists of clastic sediments and carbonates, and variably metamorphosed clastic platform sediments. The High Himalaya Crystalline series (HHC) is composed of highly metamorphosed paragneisses and some marbles, while the Lesser Himalaya (LH) is composed of metamorphosed metasediments, including quartzo-pelitic schists, minor black shales and dolomitic limestones.

3. Methods and Sampling

[9] The studied Himalayan river courses are the Narayani in Nepal that becomes the Gandak in India, the Bheri and Rapti that join the Ghaghra (Figure 1). The areas of the Himalayan watershed of the Ghaghra and Gandak cover, respectively, 32 and 18% of the Himalayan watershed of the Ganga and their flux of suspended sediments represents about one third and one fourth of that exported at the base of the range [e.g., Galy, 1999]. Samples include (1) bank sediments of the Bheri and Rapti, that were collected near the base of the range (Figure 1), and (2) bedrocks, soils, bank and suspended sediments that were sampled in the watershed of the Narayani (Figure 2). Samples of bedrock (see auxiliary material¹) were unaltered, except for four black shales from the LH. Soil profiles (Table 2), bank and suspended sediments (Table 3) were collected on the HHC and LH units, as well as on alluvial sediments. The sediments were taken at various stages of the hydrograph (Figure 3). Suspended sediments were collected by filtering 0.6 to 1.5 L of river waters on precombusted glass-microfiber filters (GF/F Whatman).

[10] Samples collected in the Bangladesh floodplain (Figure 1) include soil (Table 2), bank and suspended sediments of the Ganga, Brahmaputra, Tista, Upper Meghna

¹Auxiliary material is available at <ftp://ftp.agu.org/apend/gb/2004gb002324>.

Table 1. Area, Production, and Biomass of C₃ and C₄ Major Cultivated Plants of the Ganga-Brahmaputra River Basin^a

State	Area, 10 ⁶ ha	Sugar cane Area, 10 ⁶ ha	P, ^d 10 ⁶ T	Maize Area, 10 ⁶ ha	P, ^d 10 ⁶ T	Millet Area, 10 ⁶ ha	P, ^d 10 ⁶ T	Rice Area, 10 ⁶ ha	P, ^d 10 ⁶ T	Wheat Area, 10 ⁶ ha	P, ^d 10 ⁶ T	C ₄ ^b Area, %	C ₄ /C ₃ + C ₄ ^c	
													Area	B ^d
Assam ^e	203	0.03	1.29	-	-	-	-	2.49	3.38	-	-	0.001	0.012	0.30
Bihar ^e	174	0.11	5.04	0.69	1.17	-	-	4.98	6.77	2.12	4.40	0.046	0.101	0.40
Uttar Pradesh ^e	284	1.96	120	1.07	1.66	1.19	1.23	5.66	12.20	9.32	23.47	1.149	0.220	0.79
West Bengal ^e	87	0.03	1.83	0.04	0.13	-	-	5.9	13.20	0.37	0.78	0.008	0.011	0.14
Him. Pradesh ^e	56	-	-	0.31	0.62	-	-	-	-	0.38	0.64	-	0.449	0.60
Nepal ^f	141	-	0.81	-	0.90	-	0.15	-	3.28	-	0.74	0.014	-	-
Bangladesh ^g	144	0.17	6.95	-	-	0.11	-	10.00	15.40	0.59	1.05	0.002	0.026	0.32

^aDash, not given.^bC₄, area percentage by state of C₄ crops.^cC₄/C₃ + C₄, ratio between C₄ crops (sugar cane, maize, millet) and main crops (wheat, rice, sugar cane, millet).^dP, production; B, aboveground biomass, calculated with a B/yield ratio of 1.6 for sugar cane, 2.3 for maize, 1.4 for rice, and 1.5 for wheat.^eFor 1997–1998, from www.Indian Commodity.com. All-India areas under maize and sugar cane were, respectively, 3.26 × 10⁶ ha and 1.47 × 10⁶ ha in 1949–1950. They were 6.32 × 10⁶ ha and 3.97 × 10⁶ ha in 1997–1998 (from www.Indian Commodity.com).^fFor 1987–1988, from *National Planning Commission* [1990].^gFor 1987–1988, from *Rashid* [1991].

and Lower Meghna (Table 4). The Tista (Figure 1), which drains the Sikkim Himalaya, has an annual sediment discharge of 150 × 10⁶ t yr⁻¹ [Goswami, 1985]. The Upper Meghna has an annual sediment discharge around 13 × 10⁶ t yr⁻¹ [Coleman, 1969]. The Lower Meghna forms after the confluence of the Ganga and Brahmaputra. The river sediments were mainly collected in August 1996 during the monsoon season (Figure 3). Soils and some river sediments were collected in March 1997, in the dry season. The suspended sediments were collected by sampling 2 L of water from the middle of the river, within 50 cm of the

surface, and allowing them to settle for 24 hours in a decantation bottle.

[11] To assess the effect of particle size on the behavior of organic matter, bank sediments from the Bangladesh floodplain were suspended in a small amount of distilled water, sonicated for 5 min, then washed through a 50-μm stainless steel sieve. The <50-μm fractions were mixed in distilled water with Na-amberlite for 16 hours in order to destroy cationic bridges [Bartoli *et al.*, 1991]. The clay-size fraction (<2 μm) was then isolated by standard settling techniques and weighed (Tables 4 and 5).

Table 2. Organic Carbon (OC) Content and δ¹³C Value of Soil Samples From the Narayani Watershed (Central Nepal) and From the Bangladesh Floodplain

Location Number	Altitude, m a.s.l.	River Basin	Bedrock and Vegetation	Sample Depth, cm	OC, %	δ ¹³ C, ‰
<i>Central Nepal</i>						
MO 74	2970	Chepe	HHC paragneiss rhododendron, montane level	10–25	3.54	-26.5
				40–50	0.87	-25.5
				120–130	0.84	-25.1
MO 33	2700	Dordi	HHC paragneiss forest, montane level	0–10	8.51	-28.0
				20–30	4.43	-27.5
				60–70	2.18	-26.3
				100–110	0.39	-25.9
				180–190	0.17	-27.1
MO 89	1520	Darondi	LH schist subtropical, deforested level	0–20	1.35	-23.0
				50–60	0.27	-25.8
				110–130	0.27	-20.6
MO 122	380	Bhuri Gandaki	LH schist, jungle	0–20	2.80	-28.1
MO 601	325	Trisuli	LH black shale	0–10	2.68	-27.4
				10–20	1.38	-25.9
				20–50	0.83	-24.5
				40–60	1.33	-26.7
MO 602	325	Trisuli	forest, LH black shale tropical level	0–20	1.04	-24.0
				20–40	1.86	-26.3
				60–80	0.40	-24.9
				80–90	0.25	-25.4
MO 219	220	Narayani	Quaternary river sediments	0–10	1.48	-27.0
				20–30	1.73	-26.5
				50–60	0.33	-26.0
				80–90	0.25	-25.4
<i>Bangladesh</i>						
BGP 87 ^a	50	Ganga-Brahmaputra	Paddy field	0–20	0.40	-26.0
BGP 89 ^a	50	Ganga-Brahmaputra	banana/jute	0–20	0.60	-25.0
BGP 91 ^a	50	Ganga-Brahmaputra	sugar cane/sweet potato	0–20	0.30	-22.7
BGP 60 ^a	50	Ganga-Brahmaputra	alluvial clays, sugar cane/sweet potato	0–20	0.90	-13.7

^aAll samples were collected in the same area, in the floodplain near Mahasthan.

Table 3. Chemical (SiO₂, Al₂O₃, OC) and Isotopic Composition (OC δ¹³C Value) of River Sediments of the Narayani Watershed (Central Nepal)^a

Sample No	Type of Sediments	River	Bedrock	Sampling Period	Altitude, m a.s.l.	TSS, ^b mg/L	SiO ₂ , %	Al ₂ O ₃ , %	OC, %	δ ¹³ C, ‰
LO 50	bank	Kali Gandaki	TSS	May 1993	3550		72.68	11.37	0.07	-23.0
NAG 20	bank	Kali Gandaki	TSS	Dec 1995	2790		52.67	6.59	0.16	-25.5
NAG 29	bank	Kali Gandaki	TSS	Dec 1995	2700		49.24	6.48	0.15	-23.1
NAG 33	bank	Kali Gandaki	TSS	Dec 1995	2700		51.15	8.12	0.21	-22.1
NAG 40	bank	Miristi ^c		Dec 1995	1490		47.17	8.49	0.10	-25.4
NAG 42	bank	Kali Gandaki	TSS + HHC + LH	Dec 1995	1180		53.09	7.93	0.12	-22.4
NAG 44	bank	Kali Gandaki	TSS + HHC + LH	Dec 1995	740		64.26	8.56	0.10	-25.1
NAG 6	bank	Kali Gandaki	TSS + HHC + LH	Nov 1995	460		62.22	7.60	0.16	-24.0
MO 5	bank	Marsyandi	TSS + HHC + LH	May 1997	600		62.41	10.47	0.10	-21.0
MO 4 ^d	suspended	Marsyandi	TSS + HHC + LH	May 1997	600	103	-	-	0.61	-24.0
MO 14	suspended	Dordi ^c	HHC + LH	May 1997	1000	236	-	-	5.78	-22.0
MO 50	bank	Chepe ^e	HHC	May 1997	3600		72.56	11.79	0.15	-24.2
MO 81	bank	Chepe	HHC	May 1997	1700		72.00	11.94	0.08	-23.8
MO 90	bank	Darondi ^c	HHC + LH	May 1997	1000		-	-	0.07	-24.1
MO 94	bank	Darondi	HHC + LH	May 1997	600		85.57	6.42	0.04	-26.3
MO 93 ^d	suspended	Darondi	HHC + LH	May 1997	600	14	-	-	1.96	-23.7
MO 112	bank	Isul k. ^f	LH	May 1997	600		79.89	6.36	0.08	-24.8
MO 117	bank	Bhuri Gandaki	TSS + HHC + LH	May 1997	600		-	-	0.10	-23.7
MO 118 ^d	suspended	Bhuri Gandaki	TSS + HHC + LH	May 1997	600	83	-	-	0.85	-24.5
MO 132	bank	Bhuri Gandaki	TSS + HHC + LH	May 1997	300		80.77	7.84	0.06	-23.8
MO 133 ^d	suspended	Bhuri Gandaki	TSS + HHC + LH	May 1997	300	87	-	-	1.10	-23.4
NAG 4	bank	Seti	LH	Nov 1995	870		83.44	7.32	0.09	-25.2
MO 204	suspended	Seti	TSS + HHC + LH	May 1997	300	49	-	-	0.85	-24.0
TRI	bank	Trisuli	TSS + HHC + LH	Aug 1993	750		77.06	10.76	0.01	-23.6
TRI	bank	Trisuli	TSS + HHC + LH	Dec 1993	750		78.46	10.35	0.08	-25.3
MO 139	bank	Trisuli	TSS + HHC + LH	May 1997	300		73.42	11.84	0.06	-24.1
MO 140 ^d	suspended	Trisuli	TSS + HHC + LH	May 1997	300	45	-	-	1.62	-24.2
NAG 48	bank	Narayani	TSS + HHC + LH	Dec 1995	225		67.22	8.83	0.10	-24.0
MO 217	bank	Narayani	TSS + HHC + LH	May 1997	220		68.48	8.44	0.07	-24.7
MO 218 ^d	suspended	Narayani	TSS + HHC + LH	May 1997	220	25	-	-	1.30	-24.2
NAG 13	bank	Bheri	TSS + HHC + LH	Nov 1995	370		72.09	6.10	0.07	-24.6
MO 210	bank	Rapti	LH + Siwaliks	May 1997	385				0.10	-25.3

^aDash denotes "not determined."^bTSS, total suspended solids.^cTributary of the Kali Gandaki.^dSame location as the sample just above.^eTributary of the Marsyandi.^fTributary of the Bhuri Gandaki.

[12] Bulk samples (bedrocks, soils, river sediments) and their isolated clay-size fractions were all ground in an agate mortar. The samples contain up to 35% calcite and up to 5% dolomite. Those containing carbonate were treated with 6N HCl, for 2 hours, using frequent agitation to remove calcite and dolomite. The decarbonated powder was combusted with copper oxide in a sealed preevacuated quartz tube at 850°C. The released CO₂ was manometrically measured, yielding the total OC content in weight%. The precision for organic carbon is typically better than 10% of the measured value in the range of OC values of 0.1–1%. The CO₂ was then analyzed on either a VG Prism (ENS Lyon) or a VG602D (CRPG Nancy) mass spectrometer. The δ¹³C values are given versus V-PDB with an uncertainty of 0.1‰.

4. Results

4.1. Narayani Watershed

[13] Mean OC contents in the TSS and HHC bedrock samples (Figure 4a) are, respectively, 0.25% and 0.04%. In the LH, OC content is high in unaltered black shale (7 to 11%) but otherwise varies between 0.01 and 0.44% in

sandstones and schists. Assuming that black shales, a minor lithology, represent one per cent or less of LH outcrops, the mean OC content for unaltered LH is ≤ 0.18%. The δ¹³C values of the bedrock samples present a wide range from -31 to -18‰ (Figure 4a). Mean δ¹³C values in the TSS and HHC are, respectively, -25.8‰ (*n* = 3) and -26.4‰ (*n* = 3). Excluding the black shales, the LH is very heterogeneous with values ranging between -28.6 to -18.7‰ (*n* = 17). Unaltered LH black shales have relatively low δ¹³C values between -31 and -25.2‰ (*n* = 5). Taking the percentage of unaltered black shales as 1%, the average δ¹³C value for the LH bedrocks is -25.8‰.

[14] The OC content decreases with depth, as expected, in a given soil profile (Table 2), so that for a given profile increasing OC content on Figure 4b indicates decreasing depth. The term "soil-surface layer" will be used here for the sample collected within 20 cm of the surface. The OC content of soil surface layers tends to increase with altitude. However, the δ¹³C value of organic matter in the soil-surface layer of central Nepal does not present any simple altitude trend between 220 m and 2700 m asl (Figure 5b). The highest δ¹³C value is found in the deforested subtrop-

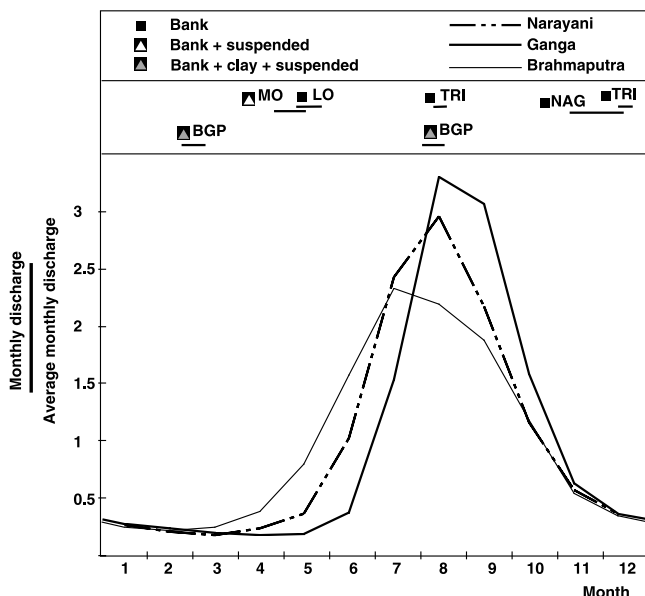


Figure 3. Hydrographs of the Narayani, Ganga and Brahmaputra. Also shown are the sampling periods (horizontal black bars) in Nepal, for samples LO, TRI, NAG, and MO (Figures 1 and 2), and in Bangladesh, for samples BGP (Figure 1). Hydrographic data for the Ganga and Brahmaputra are taken from *Hossain et al.* [1987]. Hydrographic data for the Narayani are unpublished data from the Hydrological Survey of Nepal.

ical level (MO 89). In the deeper soil layers, organic matter presents $\delta^{13}\text{C}$ values around -26‰ , with the exception of the soil profile sampled in the subtropical zone (MO 89).

[15] Bank sediments collected in the Narayani watershed (Figure 4c) are poor in OC with an average content of 0.10%. Suspended sediments have a mean of 1.7%. At the station located furthest downstream on the Narayani, the OC content of bank and suspended sediments are, respectively, 0.08 and 1.3%. Enrichment in OC of the suspended sediment versus the bulk bank sediment collected at the same location (Table 3) ranges from sixfold to fiftyfold. The $\delta^{13}\text{C}$ values of the bank and suspended sediments yield means of, respectively, -24.0‰ and -23.7‰ . Those collected at the outflow of the range, in the Narayani and its major tributaries, the Trisuli, Marsyandi and Kali Gandaki, and in the Rapti, yield a narrow range of $\delta^{13}\text{C}$ values around -24.2‰ .

4.2. Bangladesh Floodplain

[16] The soil-surface layers, which were sampled under cultivated areas in the Bangladesh floodplain, present $\delta^{13}\text{C}$ values between -26 and -14‰ , with the highest values being found under areas where sugar cane cultivation alternates with C_3 crops (Table 2). Bank sediments present a mean OC contents of 0.16% (Table 4). Suspended sediments that were collected in August 1996 during the peak discharge present a mean OC content of 0.6% whereas the few samples that were collected in March 1997 during the dry season have higher OC contents (1.2 to 7.6%; $n = 3$). The bank sediments have relatively high $\delta^{13}\text{C}$ values of -21.1 to -24.5‰ , mean -22.8‰ ($n = 6$), except that of the Upper Meghna (-27.3‰). The suspended sediments collected during the peak discharge have $\delta^{13}\text{C}$ values ranging from -23 to -21.7‰ , mean -22.5‰ ($n = 5$), the Upper

Table 4. Chemical (SiO_2 , Al_2O_3 , OC) and Isotopic (OC $\delta^{13}\text{C}$ Value) of River Sediments From Bangladesh^a

Sample Number	Type of Sediments	River	Sampling Period	TSS, ^b mg/L	SiO_2 %	Al_2O_3 , %	OC, %	$\delta^{13}\text{C}$, ‰
BGP 5	bank, bulk	Ganga	Aug 1996		66.04	11.91	0.17	-22.3
	bank, $<2\ \mu\text{m}$				46.83	20.86	1.23	-23.5
BGP 6 ^c	bank, bulk	Ganga	Aug 1996		78.45	7.86	0.04	-24.5
BGP 7 ^c	suspended	Ganga	Aug 1996	1128	54.66	17.25	0.63	-21.7
BGP 11	bank, bulk	Tista	Aug 1996		73.72	12.22	0.08	-23.7
	bank, $<2\ \mu\text{m}$				40.08	23.50	2.06	-23.8
BGP 14	bank, bulk	Brahmaputra	Aug 1996		73.02	11.88	0.18	-22.9
	bank, $<2\ \mu\text{m}$				40.24	22.41	3.61	-24.9
BGP 82 ^c	bank, bulk	Brahmaputra	March 1997		76.51	10.30	0.04	-22.2
	bank, $<2\ \mu\text{m}$				43.13	22.74	1.36	-24.8
BGP 18 ^c	suspended	Brahmaputra	Aug 1996	439	60.74	16.86	0.50	-23.0
BGP 85 ^c	suspended	Brahmaputra	March 1997	6	-	-	1.20	-22.6
BGP 3	suspended	Brahmaputra	Aug 1996	289	58.24	18.37	0.60	-23.0
BGP 54 ^c	suspended	Brahmaputra	March 1997	34			7.60	-32.1
BGP 36	bank, bulk	Upper Meghna	Aug 1996		74.65	10.34	0.38	-27.3
	bank, $<2\ \mu\text{m}$				41.03	20.41	1.63	-25.3
BGP 38 ^c	suspended	Upper Meghna	Aug 1996	6,3	-	-	1.00	-26.8
BGP 21	suspended	Lower Meghna	Aug 1996	377	52.83	18.10	0.54	-22.7
BGP 34	bank, bulk	Lower Meghna	Aug 1996		63.23	15.47	0.25	-21.1
	bank, $<2\ \mu\text{m}$	Lower Meghna	Aug 1996		44.42	22.05	1.59	-23.6
BGP 23 ^c	suspended	Lower Meghna	Aug 1996	271	55.84	19.66	0.66	-21.9
BGP 57	suspended	Residual Lake	March 1997	34	-	-	7.30	-26.1

^aDash denotes "not determined."

^bTSS, total suspended solids.

^cSame location as the sample just above.

Table 5. Mineralogical and Isotopic (ϵ_{Nd}) Composition of the Clay-Size Fraction of Bank Sediments in Bangladesh^a

Sample Number	Composition of the Clay-Size Fraction					ϵ_{Nd} , ‰
	Mineralogy					
	Clay, %	Illite, %	Chlorite, %	Smectites, %	Mixed Layer, %	
BGP5	6.7	11	0	57	32	-14.6
BGP 11	1.1	65	35	0	0	-
BGP 14	3.0	70	30	0	0	-15.7
BGP 82	1.1	60	40	0	0	-14.5
BGP 36	1.5	25	25	50	0	-
BGP 34	14.1	40	40	20	0	-

^aDash denotes "not determined."

Meghna ($-26.8‰$) being an exception. All suspended samples have $\delta^{13}C$ values within a few tenths of a per mil of their nearby bank sediments. Samples collected in the Brahmaputra during the low water stage present $\delta^{13}C$ values either within that for the peak discharge (BGP 85 at $-22.6‰$) or much more negative (BGP 54 at $-32.1‰$).

[17] The OC content of the clay-size fractions, that were extracted from bank sediments, ranges from 1.2% (BGP 5) to 3.6% (BGP 14) and is from 6 to 34 times higher (average 16) than its bulk sample (Table 4). Compared to bulk sediment, five of six clay-size fractions have more negative $\delta^{13}C$ values by 0.1 to 2.6‰ (average 1.7‰). One exception is the Upper Meghna for which the clay extract presents less negative $\delta^{13}C$ values than the bulk sediment (by 2‰).

5. Discussion

5.1. Sources of OC in the Himalayan Watersheds

[18] Comparison between the OC contents of the Himalayan continental crust and of the Himalayan river sediments may provide constraints on the recycling of organic matter inherited from bedrocks. The OC content is expected to vary within the river according to grain size and mineralogical characteristics of the sediments. Bank sediments present low OC concentrations of about 0.1%, which are similar to the average Himalayan continental crust [France-Lanord and Derry, 1997]. On the other hand, samples of suspended sediments have significantly (sixfold to fiftyfold) higher concentrations. They likely overestimate the OC content of the average suspended sediment exported, because they were collected during the low-discharge period and correspond to low suspended sediment loads of 25 to 236 mg L⁻¹ (Table 3). From the global inverse trend that is observed for suspended sediments between their concentration in river waters and their OC content [Ittekkot, 1988], it can be expected that our suspended sediments would present higher OC contents than those exported during the peak sediment discharge. Note also that the comparison of the OC contents of the river sediments with those of the Himalayan continental crust rely on data for unaltered bedrocks. The soil samples developed on black shales (MO 601, 602; Table 2) indicate that a large fraction of the organic matter has been lost, at least for these organic-rich bedrock samples, during exposure to oxidizing surface waters.

[19] Comparison of $^{13}C/^{12}C$ ratios of bedrocks from the three units (TSS, HHC and LH) and river sediments may provide additional constraints on the extent of OC recycling from bedrocks in the river sediments. At the Narayani's outflow from the Himalaya, the mineralogical composition and ϵ_{Nd} values of the silicate fraction were measured in bank sediments by Galy [1999]. They indicate that the

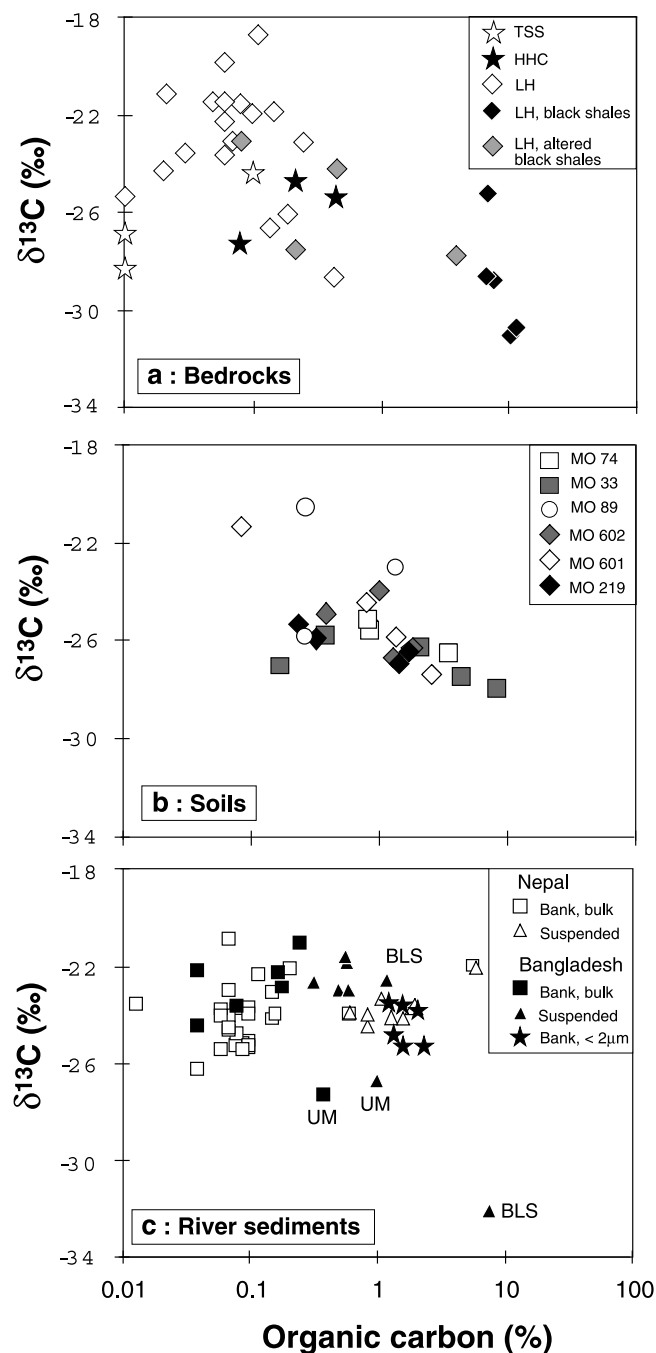


Figure 4. The $\delta^{13}C$ value versus organic carbon content for (a) bedrocks, (b) soil profiles, and (c) river sediments (UM, Upper Meghna; BLS, Brahmaputra, Low Stage). See Figure 2 for abbreviations.

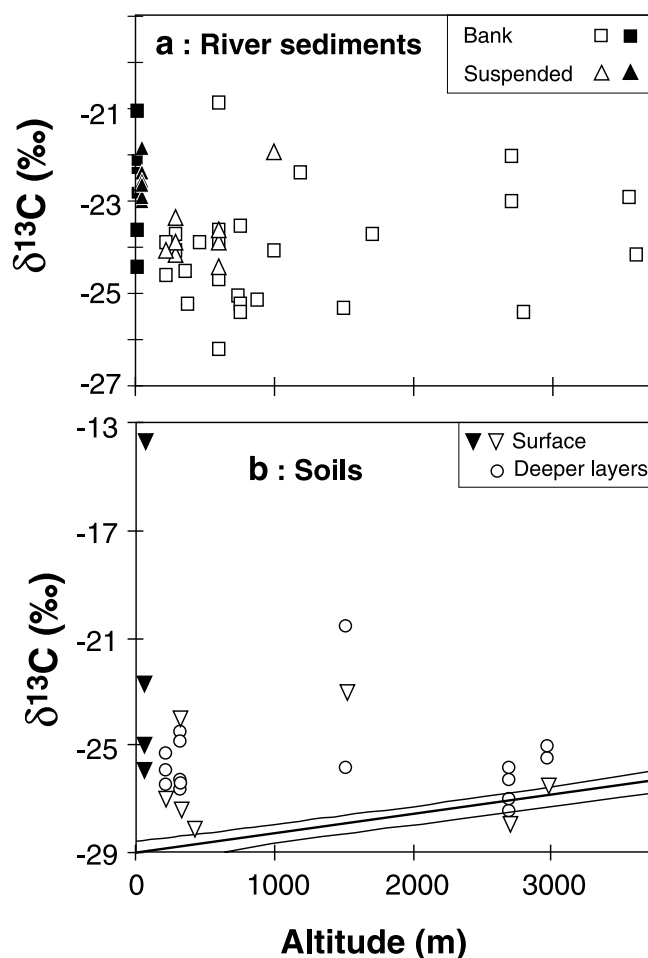


Figure 5. The $\delta^{13}\text{C}$ value of organic carbon versus altitude of sampling site for (a) river sediments and (b) soils. The curves denote the relationship between plant $\delta^{13}\text{C}$ value and altitude (central heavy line) with the 95% confidence interval (outer curved lines) established from a global sampling of plant leaves [Körner *et al.*, 1988]. Solid symbols represent samples collected in Bangladesh. Open symbols represent samples collected in Nepal on Himalayan watersheds.

proportion of silicate that is supplied by each of the three units is about one third from each one. Using these proportions and average $^{13}\text{C}/^{12}\text{C}$ ratio and OC content in each of the three units, the calculated $\delta^{13}\text{C}$ value for the OC inherited from the bedrock is -25.8‰ . It is depleted ($\sim 1.4\text{‰}$) in ^{13}C relative to the value measured at the Narayani's outflow (NAG 48, MO 217). While the OC $\delta^{13}\text{C}$ data base for TSS and HHC bedrocks is limited, that for LH bedrocks is more extensive, giving a wide variation in $\delta^{13}\text{C}$ values (-28.6 to -18.7‰). Therefore the estimated difference between bedrocks and river sediments does not provide a clear constraint on the recycling by river sediments of OC inherited from the bedrock.

[20] The isotopic composition of soil organic matter, which is another potential source of riverine OC, was

investigated in the Narayani's watershed at different altitudes between 220 and 2900 m asl. Himalayan soils appear to be essentially dominated by organic matter derived from C_3 plants, with a possible minor input of C_4 grasses and crops in the lower parts of the Himalayan watersheds. The $\delta^{13}\text{C}$ values of most soils appear to be slightly lower than those of river sediments on Figure 5. However, our sampling tends to underestimate the importance of cultivated areas, which comprise significant areas of C_4 crops (maize and the finger millet *Eleusine coracana*) particularly in the Lesser Himalaya [Dobremez and Jest, 1976]. Comparison between the present data base for river sediments and soils suggests a significant contribution from the deforested Lesser Himalaya to the OC exported by the Himalayan rivers and indicates that this zone deserves further soil sampling.

5.2. Phytoplanktonic Carbon Input

[21] Phytoplanktonic organic matter was detected by the presence of short-chain *n*-alkanes in the suspended sediments of the Ganga, in July [Ittekkot *et al.*, 1985]. It was thought to come from the flushing of the floodplain into the main channel with the rising waters. Dissolved inorganic carbon has $\delta^{13}\text{C}$ values of -8‰ in rivers sampled at the base of the Himalayas and of -12 to -8‰ in the Ganga and Brahmaputra [Galy and France-Lanord, 1999]. Phytoplanktonic matter is estimated to have relatively negative $\delta^{13}\text{C}$ values of about -35 to -30‰ [Mook and Tan, 1991; Aucour *et al.*, 2003b]. The $\delta^{13}\text{C}$ value (-32.1‰) and high OC content (7.6%) of sample BGP 54, that was collected during the low-flow stage, at a time of low water turbidity, is consistent with a dominant contribution from freshwater phytoplanktonic matter. Otherwise, the OC's $^{13}\text{C}/^{12}\text{C}$ ratios and contents do not record a clear contribution of phytoplanktonic organic matter. This observation can be linked to the high turbidity and low light prevailing in the river waters except under low-flow conditions (BGP 54).

5.3. Carbon Isotope Gradient Between Foothills and Floodplain

[22] On the Bangladesh floodplain, sediments of the Ganga and Brahmaputra, both before and after their confluence, have an average $\delta^{13}\text{C}$ value of $-22.5 \pm 0.9\text{‰}$ ($n = 11$). This is significantly higher than the average of $-24.2 \pm 0.5\text{‰}$ ($n = 11$) observed for the Himalayan rivers at their foothills. Samples from the Upper Meghna (BGP 36, 38) were not included in the average because it is a minor tributary and not representative of the floodplain. Similarly, sample BGP 54 was excluded as it was collected at a time of low sediment discharge and strongly influenced by phytoplanktonic production, as discussed above. The observed ^{13}C -enrichment of OC in river sediments could be due to the lag in sampling period rather than to a real gradient in ^{13}C content. Note, however, that bank and suspended sediments, which integrate the $\delta^{13}\text{C}$ on different timescales, yield a similar difference in ^{13}C content between the rivers sampled at the base of the Himalayas and the Bangladesh floodplain. This argues for a real gradient in $\delta^{13}\text{C}$ between the carbon

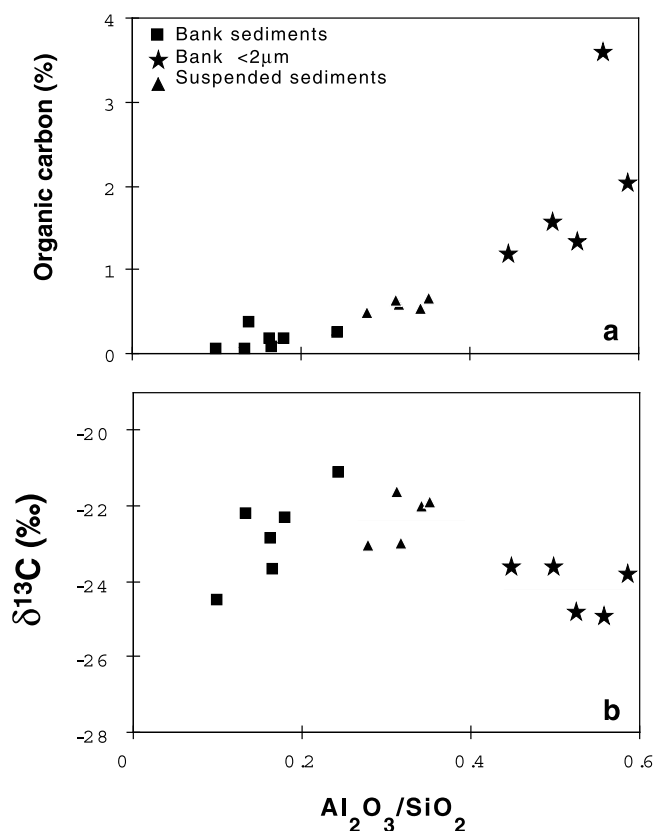


Figure 6. (a) Organic carbon content versus Al_2O_3/SiO_2 and (b) $\delta^{13}C$ of organic carbon versus Al_2O_3/SiO_2 in samples of river sediment collected in the Bangladesh floodplain.

exported from the foothills and that passing through the floodplain.

[23] This difference indicates an input of ^{13}C -enriched organic matter that should be derived from the mixed C_3 – C_4 terrestrial biomass. It may come (1) from major tributaries of the Ganga that were not sampled here and/or (2) directly from the floodplain. All the sampled Himalayan rivers present the typical topographic profile of an Himalayan river [Galy and France-Lanord, 1999]. They represent an important fraction of the suspended sediments exported by the Ganga [e.g., Galy, 1999]. If the Tista sample can be included because it is relatively close to the foothills, then the foothills were sampled over nearly 1000 km, strongly supporting that the average value of -24.2‰ is representative of the base of the range. Further sampling of the Sapt Kosi, the Ghaghra and upper reaches of the Ganga watersheds would be useful to check this point. The ϵ_{Nd} values of the silicate fraction of the sediments of the Ganga indicate that they are predominantly derived from the Himalayan watersheds and that the sediment input from the southern tributaries of the Ganga is minor [Galy and France-Lanord, 2001]. Suspended sediments, that were collected in Himalayan and southern tributaries during the low-discharge period, exhibit similar OC content of 1.1 to 1.3% [Subramanian et al., 1985]. Assuming that average OC

contents of the suspended and bedload are similar in both types of tributaries, most of the river's OC should be carried by Himalayan tributaries. All available data thus indicate that ^{13}C -enriched organic matter is probably added in the floodplain with a possible contribution from the Indo-Gangetic plain.

5.4. Organic Matter and Al_2O_3/SiO_2 Ratios of Floodplain Sediments

[24] Sediments collected in the floodplain include suspended sediments, that were collected in the middle of the river, within 50 cm of the surface, bank sediments and their clay fractions. The OC contents of these three types of sediments indicate that they are controlled by grain size and mineralogy. The OC versus Al_2O_3/SiO_2 plot (Figure 6a) shows that there is a positive relationship between the OC content and Al_2O_3/SiO_2 ratio. Low Al_2O_3/SiO_2 ratios reflect an enrichment in coarse minerals and quartz whereas high ratios indicate an enrichment in clay minerals [Galy, 1999]. The observed trend on Figure 6a is consistent with previous observations that the OC concentration of the suspended sediments increases with decreasing grain size and increasing mineral surface area [Keil et al., 1997]. Using the percentage of clays in the bulk sediments (Table 5), the OC associated with the clay-size fraction should represent 35–50% of the total OC of the bank sediment. The process that is thought to determine the distribution of OC in the fine fractions is the adsorption of organic matter on mineral surfaces [Keil et al., 1997; Aufdenkampe et al., 2001], with the amount of adsorbed organic matter increasing with mineral surface area. One Brahmaputra clay-size fraction has a very much higher OC content (3.6 wt.%) but comparable Al_2O_3/SiO_2 ratio to all other samples. However, the mineralogical compositions and sources of the minerals do not seem to be a determining factor, and their minerals originate from a similar source, as indicated by the ϵ_{Nd} values (Table 5).

[25] The stable carbon isotope composition of the organic matter in the Ganga and Brahmaputra varies with the Al_2O_3/SiO_2 ratio and between the size classes (Figure 6b). The clay-size fractions are more depleted in ^{13}C than the suspended and many bank sediments. Their $\delta^{13}C$ values are similar to those of sediments collected at the outflow of the Himalayan range. A possible explanation is that the organic matter associated with the clay-size fraction essentially originates from the Himalayan watersheds whereas that of coarser fraction is derived from the floodplain. This explanation, however, is not supported by the mineralogical composition of clays for the Ganga. Smectites, which are newly formed on the plain, are the dominant phase in the clay fraction of the Ganga sediments [Konta, 1985; Galy, 1999; Heroy et al., 2003]. An alternative explanation for the difference in isotopic composition between bulk sediments and their clay extracts is that they sample different OC pools on the plain. The area under C_4 crops has increased greatly in the basin since the mid-twentieth century (Table 1). Older organic matter would be preferentially associated with the finer fraction [Hedges et al., 1986] and therefore the finer fraction would be less influenced by inputs from C_4 crops. Also, clay fractions could selectively favor the adsorption of

^{13}C -depleted, phytoplanktonic, organic matter. Advancing on this problem would require an investigation of the organic matter on a molecular level.

5.5. Particulate OC Flux

[26] Peak discharge occurs between June and September (Figure 3) and represents about 85% of the annual sediment discharge [*River Survey Project*, 1996]. Suspended sediments that were previously collected during the peak discharge have yielded OC contents of 0.6 to 1.0% in the Brahmaputra and 0.9% in the Ganga [*Safiullah et al.*, 1985]. Our data yield similar to lower OC contents, of about 0.6%, for the suspended sediments collected during the peak discharge. Since our suspended sediments were sampled near the water's surface, this implies that they have higher $\text{Al}_2\text{O}_3/\text{SiO}_2$ than deeper samples, as observed by *Singh and France-Lanord* [2002] in the Brahmaputra. Our surface suspended sediment analyses should yield, according to the trend observed in Figure 6a, an upper limit for depth-weighted OC concentration in the river suspended load. Taking the OC contents measured in our study and fluxes of suspended sediments of $500 \times 10^6 \text{ t yr}^{-1}$ and of $440 \times 10^6 \text{ t yr}^{-1}$ for the Brahmaputra and Ganga, respectively, we obtain a total flux of $0.5 \times 10^{12} \text{ molC yr}^{-1}$ for particulate OC transported as suspended load. This flux does not account for OC transported as bedload or sequestered in the plain. Assuming that the flux of sediments transported as bedload or sequestered in the floodplain is approximately equal to that of suspended load [*Galy and France-Lanord*, 2001] and using a sediment flux of $940 \times 10^6 \text{ t yr}^{-1}$, with an OC content of 0.16% for the bedload and sequestration in the plain, we calculate a total flux of $0.65 \times 10^{12} \text{ molC yr}^{-1}$ for the OC that is buried in the floodplain or exported to the ocean. This value is lower than a previous estimate of $3 \times 10^{12} \text{ molC yr}^{-1}$ for the particulate OC flux at the mouth [*Subramanian et al.*, 1985] based on an OC content of 1–2% for the suspended load. Our data show that the OC content of river sediments largely varies with its $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio and that physical sorting of the particles must be accounted for when estimating total particulate OC flux from the discrete sampling of the suspended load through the river section and through time.

5.6. Particulate OC Mass Balance

[27] First estimates of the contribution of the main sources of organic matter to the whole flux exported may be derived from the OC content and ^{13}C mass balance. The OC and ^{13}C data point toward three potential sources of OC in the system: terrestrial C_4 plants, terrestrial C_3 plants and bedrocks. From the present database, the average organic matter that is inherited from bedrock and that derived from terrestrial C_3 biomass are indistinguishable at a $\delta^{13}\text{C}$ value of -26‰ . The $\delta^{13}\text{C}$ of organic matter that is derived from the C_4 terrestrial biomass can be set at -13‰ [*Deines*, 1980]. The river sediments of the Narayani system (Figure 2), Bheri and Rapti (Figure 1) of the Himalaya are quite homogeneous with $\delta^{13}\text{C}$ of $-24.0 \pm 1.1\text{‰}$ ($n = 32$). They are therefore generally less negative than the above -26‰ value. The OC and ^{13}C mass balance calculations for these sediments indicate that the

organic carbon derived from the C_4 terrestrial biomass represents about 15% of the OC exported by the range's outflow. In the Bangladesh floodplain, a similar calculation to above indicates that, close to the mouth, about 25% of the OC in the Ganga and Brahmaputra is derived from the terrestrial C_4 biomass. As discussed above, the input of particulate C_4 organic carbon in the Ganga and Brahmaputra appears to occur preferentially in the fraction coarser than $2 \mu\text{m}$.

[28] Comparison between the average OC content of the Himalayan continental crust (0.1%) and that of the sediments of the Ganga and Brahmaputra may provide constraints on organic matter inherited from bedrocks. We assumed that the flux of sediments transported as bedload or sequestered in the plain is approximately equal to the flux of suspended sediments [*Galy and France-Lanord*, 2001] and we took an OC content of 0.6% for the suspended sediments and of 0.16% for the bedload. On the basis of OC contents in both types of sediments, we estimate that the average sediment exported to the ocean or sequestered in the plain should have an OC content of around 0.4%. Comparison with the OC content of the Himalayan continental crust (0.1%) [*France-Lanord and Derry*, 1997] indicates that bedrock OC may represent one fourth of the particulate OC exported from the Himalayan range. This is an upper estimate, assuming that no oxidation of source rocks occurs during alteration.

[29] The OC budgets in the modern Ganga-Brahmaputra system can be compared with available data from the Bengal fan. Around 0.9 Ma, fan sedimentation becomes coarser, illite-rich and presents a negative shift of the OC $\delta^{13}\text{C}$ values. The Quaternary fan sediments exhibit OC $\delta^{13}\text{C}$ values from -25 to -21‰ , and OC contents ranging between 0.1 and 1.7% [*France-Lanord and Derry*, 1994; *Bouquillon et al.*, 1990; *Kudrass*, 1996]. The OC $\delta^{13}\text{C}$ values of modern river sediments of the Ganga and Brahmaputra during their peak discharge therefore fall in the range observed for Quaternary sediments, whereas their OC contents match the minimum OC values observed in Quaternary sediments of the fan. The reloading of river particles with marine organic carbon, which is observed in other delta systems [*Keil et al.*, 1997], should be minor because the organic matter trapped in the Bengal fan is predominantly derived from terrigenous sources [*Poynter and Eglinton*, 1990; *Meyers and Dickens*, 1992; *Freeman and Colarusso*, 2001]. Changes in $\delta^{13}\text{C}$ values of fan sediments essentially reflect variations of the contribution of terrestrial OC derived from C_4 plants [*France-Lanord and Derry*, 1994; *Freeman and Colarusso*, 2001]. A similar calculation as above yields a proportion of 5–40% for the OC derived from the C_4 terrestrial biomass in the Quaternary sediments. Otherwise, high OC contents in studied fan sediments ($>0.6\%$) could be due to (1) grain-size sorting and/or (2) input of terrestrial carbon that is higher for particles brought in during the Quaternary (and Pliocene) than for modern river particles. In order to choose between these two hypotheses, it will be useful to apply the coupled $\text{Al}_2\text{O}_3/\text{SiO}_2$ versus OC approach to the fan sediments. This would also improve previous estimates [*France-Lanord and Derry*, 1997] of average OC content and of consumption of

atmospheric CO₂ by OC burial in the fan (that equals uncorrected OC burial minus bedrock OC recycling).

6. Conclusions

[30] Compared to the average Himalayan continental crust, the suspended sediments, that are carried at the mouth of the Ganga-Brahmaputra in the monsoon season, present a significant enrichment in OC, from 0.1 to 0.6%. Their OC contents are comparable to the minimum values observed in the Quaternary sediments of the Bengal fan. They confirm the importance of the flux of organic matter exported with the Himalayan sediments as a major component to the global flux of OC delivered to the sea.

[31] Variations in the ¹³C/¹²C ratio of OC in modern soils and river sediments of the Ganga-Brahmaputra basin essentially reflect the differential input of C₃ and C₄ derived organic matter. Significant, but minor, inputs of organic matter are derived from C₄ plants. The analyses confirm the dominance of C₃ vegetation in the modern system that is also consistent with the record of the Bengal fan.

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