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$\delta^{13}\text{C}$  of organic carbon in the Bengal fan: source evolution and transport of C3 and C4 plant carbon to marine sediments

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**Abstract** - Carbon isotopic measurements on organic carbon in sediment cores from the Bengal Fan (ODP Leg 116) show a dramatic 10 ‰ increase beginning ca. 7 Ma ago, and a rapid decrease after 0.9 Ma. These shifts reflect changes in the mixing ratio of terrigenous carbon derived from C3 and C4 plants. The rapid increase in  $\delta^{13}\text{C}$  of Bengal Fan OC at 7 Ma show that C4 expansion already documented in the Siwaliks was widespread overall the Himalayan foreland. After 7 Ma, relations of  $\delta^{13}\text{C}$  with sedimentological parameters show that C4 plants dominates in the foreland whereas C3 plants remain in the mountain range. Variations in the source of the sediments and of the OC appear to be sensitive to climate-hydrologic conditions in the basin. Major changes in the isotopic composition of the carbon flux in one of the largest river system modify the isotopic budgets of both marine dissolved carbon and the sedimentary carbon mass.

### **Introduction**

The Bengal Fan is one of the largest sedimentary units in the world, containing ca.  $3 \times 10^{19}$  kg of sediment, primarily derived from the erosion of the Himalayan orogen (Curry, 1991; Curry and Moore, 1971; Rea, 1992). The rate of sediment delivery appears to have increased markedly in early Miocene time (ca. 21 Ma (Cochran, 1990; Lindsay, et al., 1991), and again at 10-11 Ma (Rea, 1992), implying an average sediment delivery rate of ca.  $1.5 \times 10^{12}$  kg/yr during the late Neogene. This mean rate is similar to estimates of the modern rate (Milliman and Syvitski, 1992). Measurements obtained from drill core of early Miocene to Quaternary age in the distal portion of the fan (ODP Leg 116) have an average total organic carbon (TOC) content of 0.85 wt % (n=155) (Cochran et al., 1989) and this study). A smaller number of analyses (n=12) from Quaternary sediments in the submarine portion of the proximal Bengal delta are similar (=0.90wt %, (Bouquillon, 1987)). If these results are representative of the fan as a whole, the mean burial rate of OC in the Bengal Fan since early the Miocene has been,  $\approx 1.1 \times 10^{12}$  mol/yr. Recent published estimates for global OC burial are from 4 to  $10 \times 10^{12}$  mol/yr (Lasaga, et al., 1985; Smith and Hollibaugh, 1993), thus the Bengal Fan may account for 10 to 25% of the total global OC burial flux during the Neogene. If the Indus Fan, Indo-Gangetic Plain and Ganga-Yamuna delta are included, the estimate of OC burial in Neogene sediments derived from the Himalaya increases by a factor of roughly 1.5. It is evident that the global OC burial budget may have been significantly perturbed by rapid erosion of the Himalayan region.

The fraction of biologically labile particulate organic carbon (POC) in the Ganga river is low, which suggests (Ittekkot, 1988) that deep sea sediments in the Indian Ocean are an important sink for OC derived from terrestrial plants. Data from ODP Legs 116 and 117 support this hypothesis. Gas chromatograph and pyrolysis data show that OC in the Bengal Fan is dominated by organic matter derived from terrigenous sources (Bertrand, et al., 1991; Meyer and Dickens, 1992; Party, 1989; Poynter and Eglinton, 1990). Carbon isotopic analyses ( $\delta^{13}\text{C}$ ) of paleosol carbonates, organic matter and fossil teeth (Cerling, et al., 1993; Morgan, et al., 1994; Quade and Cerling, in press; Quade, et al., 1989) show evidence of a major shift in terrestrial vegetation patterns in the

Himalayan foreland in late Miocene time. A terrestrial plant community dominated by C4 plants appears to replace a C3-dominated community between 7 and 6 Ma. The stable isotopic results (Cerling, et al., 1993; Morgan, et al., 1994; Quade and Cerling, in press; Quade, et al., 1989) suggest several questions. Was the expansion of C4 plants confined to a relatively small area in the foreland, or was it of general importance in the northern Indian subcontinent? What caused the shift in the dominant photosynthetic pathway? What effects did the expansion of C4 biomass have on the budget of OC exported to the ocean and buried in marine sediments? Since the bulk of the OC preserved in the Bengal Fan is derived from the Ganga and Brahmaputra river basins,  $\delta^{13}\text{C}$  analyses of sedimentary OC provide insight into this question.

### **The Bengal Fan**

Sedimentation of the fan can be illustrated by variations in rate of deposition, clay mineralogy, grain size and TOC (Fig 1). Bengal Fan sediments older than 7 Ma or younger than 0.9 Ma are dominantly silts with an illite-rich clay fraction, TOC content usually  $< 0.5$  wt %, show relatively little chemical weathering, and were deposited at high sedimentation rates. In contrast, between 7 and 0.9 Ma, sedimentation rates of mostly smectite-rich muds and fine silts were lower, the degree of chemical weathering was much more intense, and TOC  $> 1$  wt %. However Nd, Sr, O and H isotopic data show that these variations were not caused by changing sediment sources, but rather by changing continental alteration histories (Bouquillon, et al., 1990; France-Lanord, et al., 1993). Comparable variations in sedimentation rate and weathering intensity are also observed in the Siwalik Himalayan foreland sequence (Burbank, et al., 1993; Quade and Cerling, in press). These variations have been interpreted to reflect a change in the rate of sediment supply and the residence time of sediment in the foreland basin (Burbank, et al., 1993; France-Lanord, et al., 1993). According to this hypothesis, between 7 and 0.9 Ma sediments were stored longer and weathered more intensely in the Indo-Gangetic Plain. The beginning of this interval follows oceanographic evidence for the intensification of the Asian monsoon ca. 7-8 Ma (Kroon, et al., 1991; Prell, et al., 1992)

### **Methods and Results**

Total Organic Carbon has been analyzed on whole rock sediments. Samples were first acidified with 2N HCl at 100°C to eliminate carbonate, rinsed, collected on a silica filter and dried at 80°C. The sample on its filter was then oxidized under vacuum at 950°C with CuO/Cu<sub>2</sub>O. The CO<sub>2</sub> released was purified at -140°C, manometrically measured for TOC content, and analysed on a VG602D mass spectrometer. Reproducibility is better than 0.2% and  $\delta^{13}\text{C}$  of NBS-22 was measured at  $-29.7 \pm 0.1$  relative to PDB.

$\delta^{13}\text{C}$  on total organic carbon (TOC) was analyzed for twenty eight samples from ODP Holes 717C and 718C (Table 1, Fig 2). Together, these two holes provide a continuous 18 Ma record of turbidite sedimentation (Stow, et al., 1990). Prior to ca. 7 Ma,  $\delta^{13}\text{C}$  values in TOC are -27 to -25‰, typical of C3 plants. The  $\delta^{13}\text{C}$  data show a dramatic positive shift of 10‰ beginning near 7 Ma.  $\delta^{13}\text{C}$  values in Pliocene sediments of the

Bengal Fan reach -15 to -17%. In late Quaternary sediments  $\delta^{13}\text{C}$  values decrease rapidly to -24%. Limited  $\delta^{13}\text{C}$  data from OC at DSDP Hole 218 show comparable values with the exception of one sample (Erdman, et al., 1974). Mixing of carbon from terrestrial C3 plants with OC derived from marine primary production ( $\delta^{13}\text{C} \approx -20\%$ ) is unlikely to account for the observed shifts because its relative abundance in Bengal Fan sediments is low (Bertrand, et al., 1991; Meyer and Dickens, 1992; Party, 1989; Poynter and Eglinton, 1990). Rather, we interpret these variations as primarily resulting from changing mixing ratios of terrigenous C3 and C4-derived OC. Between 7 Ma and ca. 0.9 Ma, OC burial in the fan appears to have been dominated by material derived from C4 biomass. In agreement with paleosol studies in the Himalayan foreland (Quade and Cerling, in press; Quade, et al., 1989), we find no evidence for a significant C4 component in Bengal Fan OC prior to 7 Ma. The timing and magnitude of the 7 Ma  $\delta^{13}\text{C}$  shift in both carbonates and OC in the Siwalik paleosols is very similar to that observed in the Bengal Fan OC. This remarkable agreement implies that the expansion of C4 plants was widespread and rapid across the G-B basin.

## **Discussion**

### **C3/C4 plants distribution**

$\delta^{13}\text{C}$  of TOC recorded in the Bengal Fan reflects the C3/C4 abundance in the Himalaya-Ganga-Brahmaputra system. However, in order to interpret the observed variations correctly, we must first consider the processes of carbon export and transport. Net organic carbon export from a variety of ecosystems is roughly proportional to net primary productivity (Kaplan and Newbold, 1993), and productivity of C4 plants is known to be significantly higher than that of C3 plants (Jones, 1992). Therefore, the C4 /C3 ratio recorded in Bengal Fan OC can be amplified with respect to the ratio of C4 to C3 plants in the whole catchment area.  $\delta^{13}\text{C}$  of Bengal OC after 7 Ma is variable and is closely related to the other sedimentological variables such as TOC and water content of the sediments (Fig. 3). The increase of TOC in sediments younger than 7 Ma (Fig. 1) likely results from the decrease of the mean grain size (and increase of specific surface area) of the particles delivered to the fan, as has recently been shown for other marine clastic sediments (Keil, et al., 1994). Whole rock water contents ( $\text{H}_2\text{O}^+$ ) reflect the relative proportion of hydrous supergene minerals (mainly smectite and vermiculite) derived via weathering in the Indo-Gangetic Plain versus less-hydrated minerals produced by physical erosion in the Himalaya (quartz, feldspars, micas, illite) (France-Lanord, et al., 1993). The relation between  $\delta^{13}\text{C}$  and  $\text{H}_2\text{O}^+$  implies that the secondary mineral assemblage is associated with OC derived from a C4-dominated ecosystem, while the "primary" mineral assemblage carries carbon from a C3-dominant ecosystem. Thus the relationship for post-7 Ma samples shown in Fig. 3 can be interpreted as a mixing line between coarse, relatively unaltered sediments associated with C3 carbon, and fine-grained, weathered sediments carrying mostly C4 carbon. The  $\delta^{13}\text{C}$  variability of Bengal Fan OC contrasts with the relatively constant  $\delta^{13}\text{C}$  of Siwalik paleosol carbonate and OC through this interval (Cerling, et al., 1993; Quade and Cerling, in press; Quade, et al., 1989). This suggests that, after 7 Ma, OC  $\delta^{13}\text{C}$  values were

primarily controlled by changes in the source of particulate organic matter rather than by C3/C4 change over the whole source region. The coarser-grained, less weathered sediments, which we interpret to have been derived more or less directly from the Himalayan hinterland during intervals of rapid transport, may have derived more of their OC from C3-dominated forests upstream. Sediments which became incorporated into soils in the C4-dominated foreland and were weathered during intervals of slower transport carry C4 carbon. We propose that, once C4 plants became established in the G-B basin at 7 Ma,  $\delta^{13}\text{C}$  variations in Bengal OC result from changes in the dynamics of the river transport system leading to changes in the source region of river POC. Essentially, under different hydrologic conditions the rivers appear to "sample" different parts of the drainage basin. For example, two mid-Pliocene samples show a brief return to low  $\delta^{13}\text{C}$  values indicative of increased C3 plant carbon. These samples are illite-rich silts, associated with an interval of increased sedimentation rate and grain size. The post-0.9 Ma shift to lower  $\delta^{13}\text{C}$  values is also associated with a return to rapidly deposited, coarse, illite-rich sedimentation.

The increase in the ratio of chemical weathering to physical erosion appears to be synchronous with the expansion of C4 biomass recorded both in the Bengal Fan and the Siwalik foreland. Oceanographic evidence indicates a more pronounced monsoonal climate in the region beginning 7-8 Ma (Kroon, et al., 1991; Prell, et al., 1992). Increased seasonality should favour C4 plants, and the expansion of C4 vegetation in the Siwaliks could be a consequence of the development of the monsoon (Quade, et al., 1989). More recent work has raised the possibility that the late Miocene expansion of C4 plants was a global event, perhaps driven by falling atmospheric  $\text{CO}_2$  levels (Cerling and Quade, 1993; Ehleringer, et al., 1991), but African evidence (Morgan, et al., 1994) suggests a diachronous history. Similarly, changes in the weathering regime could reflect either climatic or tectonic control (Burbank, et al., 1993). We argue that the close coupling of the shifts in both vegetation and weathering in the Himalayan foreland can only be reasonably explained by climate change. The marked change in sediment supply beginning near 0.9 Ma correlates with a significant cooling of global climate and ice volume expansion, and the beginning of the strong 100 kyr periodicity in the marine  $\delta^{18}\text{O}$  record (Ruddiman, et al., 1986). The corresponding decrease in  $\delta^{13}\text{C}$  in Bengal Fan OC could indicate lower abundance or productivity for C4 plants in the G-B basin precipitated by a cooler and/or wetter climate. However, the relationship between sediment source and  $\delta^{13}\text{C}$  for post-0.9 Ma samples (Fig. 3) suggests that the light carbon was primarily derived from higher in the G-B drainage system. This relationship further suggests that the hydrology and transport dynamics of the G-B river system exert first-order control over the type of OC exported to the oceans after 7 Ma.

#### **Implications for the marine and sedimentary carbon isotope budget**

The present day Ganga, Brahmaputra and Indus rivers carry 3 to 5% of the river dissolved inorganic carbon (DIC) flux (Meybeck, 1979), and > 10% of the river TOC flux (Subramanian and Ittekkot, 1991). Although the

Bengal Fan records only refractory POC from the G-B river system, it is likely that DOC and labile POC were isotopically similar (In the Amazon, for example,  $\delta^{13}\text{C}_{\text{DOC}} - \delta^{13}\text{C}_{\text{POC}}$  usually  $<2\%$  (Quay, et al., 1992)). Because the G-B river system is such an important source of OC, it is thus very likely that the mean global  $\delta^{13}\text{C}$  of the riverine TOC input to the ocean increased significantly ca. 7 Ma. The mean  $\delta^{13}\text{C}$  of the inorganic carbon input to the ocean was probably similarly affected. A large fraction of DIC in rivers is derived from respiration of organic matter, which results in river DIC with low  $\delta^{13}\text{C}$  values relative to the atmosphere (Hitchon and Krouse, 1972; Longinelli and Edmond, 1983). Since isotopically heavy carbon derived from C4 plants began to contribute significantly to the carbon load of the G-B rivers at ca. 7 Ma, the mean  $\delta^{13}\text{C}$  of marine DIC should also have increased. A 5% increase in the  $\delta^{13}\text{C}$  of 10% of the global river TOC flux would be sufficient to change the mean  $\delta^{13}\text{C}$  of marine bicarbonate by  $+0.5\%$ . The  $\delta^{13}\text{C}$  record of marine carbonates is complex during this interval, and not all records agree (Broecker and Woodruff, 1992). We note that  $\delta^{13}\text{C}$  of bulk marine carbonate, which has been proposed to be the best estimate of the global ocean carbon budget, increases by ca.  $0.5\%$  near 7 Ma (Shackleton and Hall, 1984), as do several foraminiferal records from both the Atlantic and Pacific e.g. (Wright, et al., 1991).

The expansion of C4 photosynthetic plants in the terrestrial environment has implications for interpreting the Late Cenozoic carbon isotope record as well. As noted above, the Neogene OC burial flux in the Bengal and Indus Fan, associated deltas and foreland basins probably represents  $\sim 15\%$  of the global total. From ca. 7 to 1 Ma, the OC buried in the Bengal Fan was 7-10% less depleted in  $^{13}\text{C}$  than it had been prior to 7 Ma. Consequently, the global  $^{13}\text{C}$  depletion of sedimentary organic carbon relative to carbonate,  $\delta^{13}\text{C}_{\text{carb}} - \delta^{13}\text{C}_{\text{org}} = \Delta B$  (Hayes, et al., 1989), must have decreased by 1-2%. If C4 plant-derived carbon was an important component of OC burial in other regions (Cerling, et al., 1993) this effect would have been correspondingly greater. Carbon isotope depletion produced by marine algae appears to have decreased by 5-7%, mostly by early Neogene time (Popp, et al., 1989). The value of  $\Delta B$  at any given time depends on the relative proportions of marine and terrestrial organic carbon burial as well as the depletion associated with each. Because of the large changes in carbon isotope depletion associated with photosynthesis in both the marine and terrestrial habitats models of the Cenozoic sedimentary carbon isotope mass balance must take these effects into account (Derry, 1993; Kump, 1989).

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Table 1 - Total Organic Carbon contents and  $\delta^{13}\text{C}$  data from Holes 717C and 718B

Sample	#depth (m)	Age* (Ma)	TOC (wt %)	$\delta^{13}\text{C}$ (‰)
<b>Hole 717C</b>				
2 H 2 - (60-61)	15.6	0.1	0.22	-24.0
4X2 - (9-10)	28.6	0.2	0.47	-22.3
5X1 - (51-52)	37.0	0.2	1.75	-20.8
6X3 - (22-23)	49.2	0.3	0.19	-24.4
7X2 - (20-21)	57.2	0.3	0.13	-23.1
8X1 - (50-51)	65.5	0.4	0.21	-21.3
15XCC - (6-7)	114.8	0.7	0.18	-24.8
21X3 - (145-146)	164.5	1.1	2.26	-18.3
22X5 - (25-26)	175.8	1.3	1.58	-16.6
30X2 - (8-9)	247.1	2.4	1.72	-15.5
31X2 - (10-11)	256.6	2.5	1.68	-15.5
36X5 - (77-78)	309.3	3.4	1.66	-19.4
37X1 - (79-80)	312.8	3.4	0.31	-23.1
40X4 - (105-106)	346.1	3.9	0.53	-21.9
41X1 - (36-37)	350.4	4.0	1.81	-17.1
51X5 - (13-14)	451.1	5.6	1.82	-18.3
53XCC - (32-33)	473.5	5.9	1.45	-19.7
55X2 - (95-96)	485.5	6.1	1.92	-19.0
61X2 - (50-51)	542.0	6.9	0.46	-26.0
66X4 - (7-8)	592.1	7.4	1.19	-25.1
71X3 - (99-100)	639.0	7.9	0.38	-25.4
81X1 - (49-50)	730.5	8.5	0.48	-24.9
91X1 - (70-71)	819.4	8.9	0.40	-24.8
<b>Hole 718C</b>				
60X1 - (100-101)	570.8	13.0	0.15	-24.8
68X1 - (67-68)	646.5	14.2	0.40	-25.8
78X5 - (34-35)	747.1	15.9	0.21	-25.5
86X1 - (89-90)	817.7	17.1	0.59	-25.8
95X1 - (59-60)	897.6	18.4	0.35	-25.0

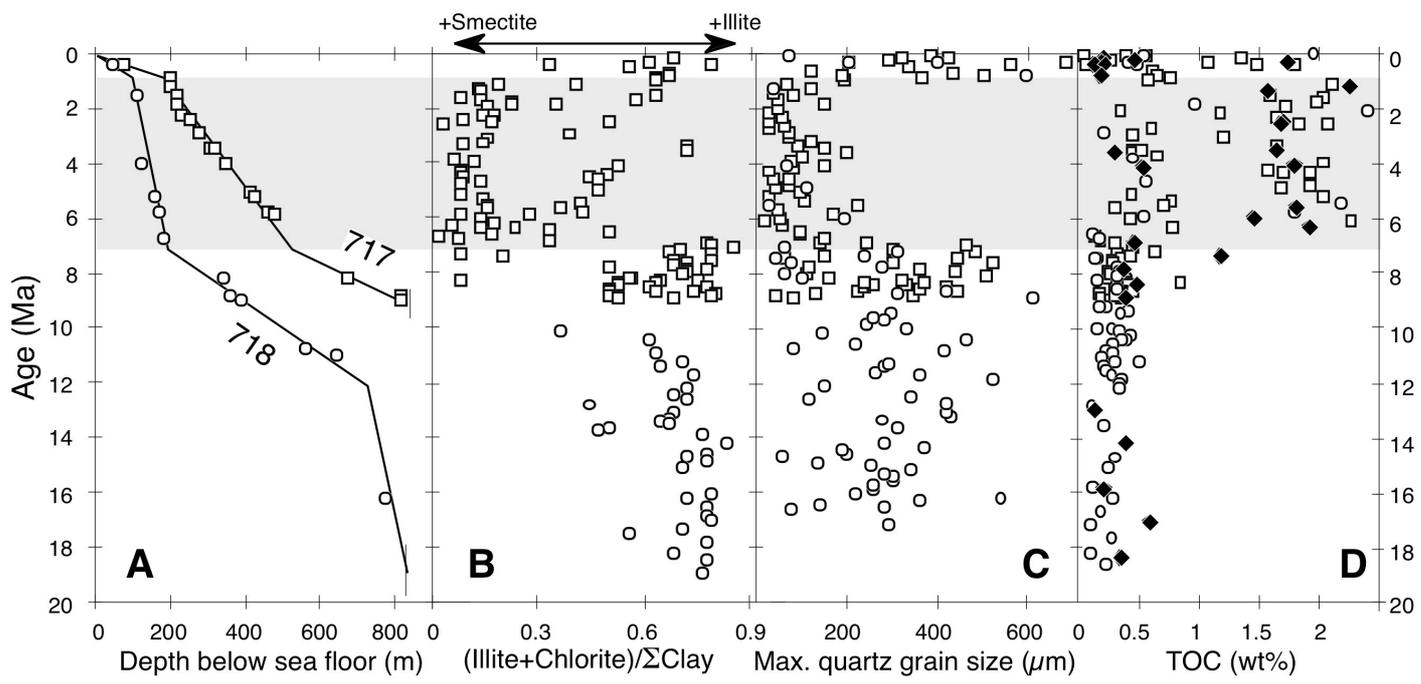
\*Ages are estimated from biostratigraphic data (Gartner, 1990).

## Figure captions

**Fig. 1.** Age dependence with (A) sediment thickness (Gartner, 1990), (B) clay mineralogy (Bouquillon, et al., 1990), (C) maximum quartz grain size and (D) TOC content (Cochran et al., 1989) at ODP Sites 717 ( $\square$ ) and 718 (o) and data in this study( $\blacklozenge$ ). A : lower slopes correspond to higher sedimentation rates. B : (Illite+chlorite)/ $\Sigma$ clay ratios are measured on  $<2\mu\text{m}$  fractions. They mostly reflect the relative abundance of illite versus smectite. In the Bengal Fan, illite and chlorite are derived from metamorphic precursors by physical erosion whereas smectite is primarily derived from weathering of the same material (France-Lanord, et al., 1993). %TOC appears to reflect grain size/specific surface area of sediments (Keil et al. 1994).

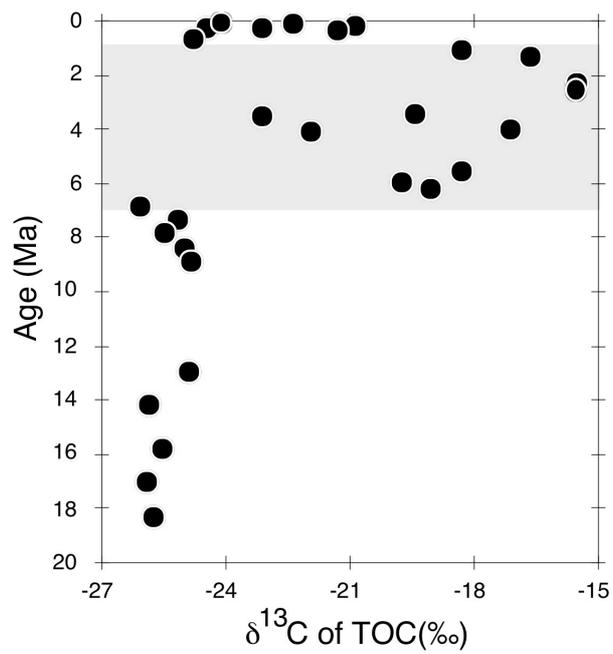
**Fig. 2.**  $\delta^{13}\text{C}$  of TOC in Bengal Fan sediments plotted against age in Holes 717C and 718B. The increase of  $\delta^{13}\text{C}$  values at ca. 7 Ma reflects the increase of the C4/C3 plant ratio in the source of the organic matter. The  $\delta^{13}\text{C}$  increase near 7 Ma closely parallels that observed in paleosol studies in the Himalayan foreland (e.g., Cerling, et al., 1993), but the decrease after 0.9 Ma is not observed in the paleosol data. Variations in  $\delta^{13}\text{C}$  are correlated with variations in sedimentation rate, grain size and clay mineralogy (Fig. 1).

**Fig. 3.** Variations of  $\delta^{13}\text{C}$  of TOC with (A) TOC content, and (B)  $\text{H}_2\text{O}^+$  content for sediments of Holes 717C and 718B.  $\text{H}_2\text{O}^+$  is calculated from loss on ignition of whole rock analysis corrected for carbonate and TOC contents, after drying at  $110^\circ\text{C}$  for 24 hours. Symbols:  $\square$  samples  $> 7$  Ma;  $\bullet$  = 7 to 0.9 Ma;  $\Delta$   $< 0.9$  Ma. In samples  $< 7$  Ma, the relationships between  $\delta^{13}\text{C}$ , TOC and  $\text{H}_2\text{O}^+$  suggest that Bengal Fan OC contains a mixture of C3-derived material associated with coarser, unweathered sediments and C4-derived carbon associated with weathered, fine-grained sediments.



France-Lanord & Derry  
Fig 1

France-Lanord & Derry  
Fig 2



France-Lanord & Derry  
Fig 3

