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Efficient organic carbon burial in the Bengal fan sustained by the Himalayan erosional system

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Continental erosion controls atmospheric carbon dioxide levels on geological timescales through silicate weathering, riverine transport and subsequent burial of organic carbon in oceanic sediments[1]. The efficiency of organic carbon deposition in sedimentary basins is however limited by the organic carbon load capacity of the sediments and organic carbon oxidation in continental margins. At the global scale, previous studies have suggested that about 70 per cent of riverine organic carbon is returned to the atmosphere, such as in the Amazon basin[2–4]. Here we present a comprehensive organic carbon budget for the Himalayan erosional system, including source rocks, river sediments and marine sediments buried in the Bengal fan. We show that organic carbon export is controlled by sediment properties, and that oxidative loss is negligible during transport and deposition to the ocean. Our results indicate that 70 to 85 per cent of the organic carbon is recent organic matter captured during transport, which serves as a net sink for atmospheric carbon dioxide. The amount of organic carbon deposited in the Bengal basin represents about 10 to 20 per cent of the total terrestrial organic carbon buried in oceanic sediments. High erosion rates in the Himalayas generate high sedimentation rates and low oxygen availability in the Bay of Bengal that sustain the observed extreme organic carbon burial efficiency. Active orogenic systems generate enhanced physical erosion and the resulting organic carbon burial buffers atmospheric carbon dioxide levels, thereby exerting a negative feedback on climate over geological timescales.

Photosynthesis of organic matter is a major pathway for consumption of atmospheric CO_2. Although most photosynthetic organic carbon (C_{org}) is re-oxidized and returns to the atmosphere, a small fraction is buried in sedimentary basins and stored over geological timescales[5]. This burial represents the second largest atmospheric CO_2 sink (after silicate weathering coupled to carbonate precipitation) and contributes to long-term climate regulation[6]. Continental erosion exerts a primary control on C_{org} burial through sediment transport and detrital deposition in sedimentary basins. However, assessing the role of continental erosion in this part of the C cycle is complex, as several processes control its efficiency. First, C_{org} transported by rivers is composed of both recent organic matter and fossil refractory C_{org} derived from erosion of carbonaceous rocks. Erosion-burial of the latter has no effect on the long term C cycle and it is therefore necessary to determine its proportion. Second, it is generally believed that ~70 % of C_{org} exported by global rivers (for example, the Amazon) is oxidized in the continental margins before burial and thus returns to the atmosphere[2–4]. Last, assessing the riverine C_{org} flux is not straightforward, because the C_{org} content of sediment is highly variable owing to transport segregation processes. Understanding the impact of continental erosion on the C_{org} cycle requires identification of the controls on the flux of riverine C_{org}, the proportion of rock-derived fossil C_{org} and the burial efficiency.

Himalayan erosion generates the largest flux of sediments to the oceans. Today, this represents between 1 and 2 billion tons of sediments exported each year from the Himalayas through the Ganges-Brahmaputra (G-B) system and buried in the Bengal fan sedimentary unit[7–9]. The total C_{org} concentration (TOC) in G-B fluvial sediments was hitherto estimated using only surface suspended sediments and without correction for the fossil C_{org} contribution[10,11]. Over the past 15 Myr, the Bengal fan has buried an average of about 0.6×10^{12} mol C_{org} yr^{-1}, that is, 15% of the global burial flux[12]. The Himalayas are thus a key locality for isolating the role of major orogens on the C cycle.

A major sample set covering the whole basin from the Himalayas to the distal part of the Bengal fan has been analysed for C_{org}, ^{14}C and chemistry (Supplementary Tables 1 and 2). The two major rivers (Ganges and Brahmaputra) and their confluence (Lower Meghna) were sampled during three monsoon seasons when ~95% of the sediment flux is exported[13] (Fig. 1, Methods). Bed loads and depth profiles of suspended sediments were collected to integrate the total sediment variability. Simultaneous Acoustic Doppler Current Profiler measurements were performed in order to characterize the hydrodynamic conditions. The Bengal fan sedimentary units are documented by subsurface sediments cored in the shelf, in the mid-fan active channel-levee system and in the distal part of the fan during RV *Sonne* cruise SO93 in 1994[14] (Fig. 1). Detailed study of the modern rivers and their complements in the sedimentary record allows us to quantify: (1) the Himalayan C_{org} flux, (2) the proportion of rock-derived fossil C_{org} and (3) the terrestrial C_{org} burial efficiency.

In the G-B system, sediments have a wide range of composition owing to differentiation during transport under hydrodynamic forces. Sediment concentrations increase, while flow velocities decrease, with depth (Supplementary Fig. 1). Suspended sediments are fine-grained and clay-rich in surface water, but become coarser and quartz-rich at depth; bed sediments are sandy and enriched in quartz. TOC varies from 0.03% to 0.82% and is remarkably well correlated with chemical composition; all G-B sediments define a positive correlation between TOC and Al/Si ratio (Fig. 2). This trend mostly expresses co-variation of TOC with grain size and mineralogy, as Al/Si increases with the proportion of aluminous and fine-grained minerals[8]. Individually, the Ganges, the Brahmaputra and the Lower Meghna define comparable trends, indicating identical C_{org} loading (Fig. 2). In the G-B system, mineral particles and C_{org} appear to have a similar response to hydrodynamic sorting, indicating a tight physical association between these components, either at the grain scale (C_{org} sorption on minerals surfaces[15]) or at larger scale (formation of C_{org}-mineral aggregates[16]). A consequence of the gradient in TOC is

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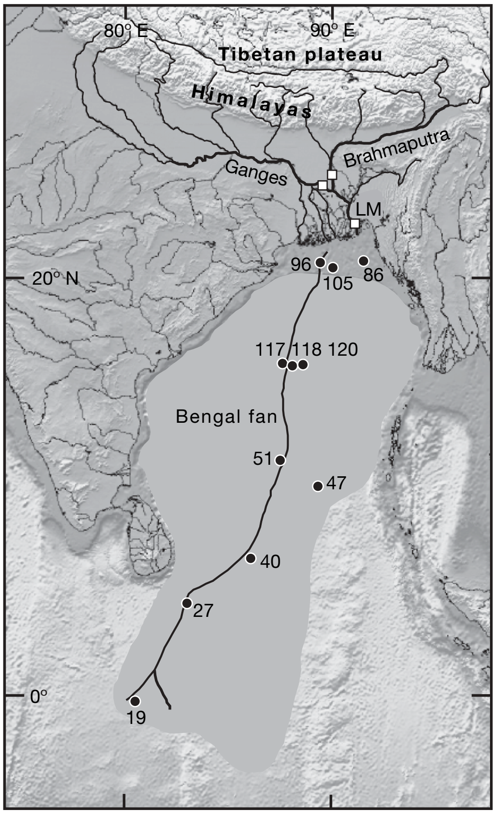

Figure 1 | Map of the Himalayan system. Ganges, Brahmaputra and Lower Meghna (LM) were sampled in Bangladesh (open squares). The different parts of the Bengal fan were investigated using cores from cruise SO 93 (filled black circles and corresponding core numbers). The black line represents the modern active channel.

that earlier estimates of C_{org} fluxes based on surface suspended sediment data were overestimated[10,11]. Thus, assessing the C_{org} flux requires the mean sediment composition transported by the river to be calculated. With our data, the integrated suspended sediment TOC can be estimated by integrating TOC gradient, sediment concentration and flow velocity over the whole river depth. For the Lower Meghna before its discharge into the Bay of Bengal (Fig. 1), we calculate an integrated TOC of 0.41 ± 0.04%. In comparison, bed sediments have low TOC with an average value of 0.05%, consistent with their high proportion of quartz.

C_{org} transported by rivers is a mix of inherited fossil C_{org} (FC_{org}) derived from erosion of carbonaceous rocks, and different sources of recent C_{org} (RC_{org}) that include pre-aged soil C_{org} and fresh plant debris. As FC_{org} burial has no effect on the long-term atmospheric CO_2 content, its proportion must be determined. We measured ^{14}C concentrations of acid-insoluble C_{org} composed of FC_{org} that has no ^{14}C ($\Delta^{14}C = -1{,}000‰$) and RC_{org} that is enriched in ^{14}C (ref. 17). $\Delta^{14}C$ ranges from −814‰ to −215‰ (13.4 to 2.0 kyr) and co-varies with TOC; Ganges, Brahmaputra and Lower Meghna sediments define independent but similar trends. This reflects mixing between bed sediments dominated by FC_{org} and surface suspended sediments dominated by RC_{org} (Fig. 3). The RC_{org} $\Delta^{14}C$ is unknown, as it is derived from sources of variable ages. Surface suspended sediments constrain minimum $\Delta^{14}C$ values for the RC_{org} of each river. $\Delta^{14}C$ of RC_{org} must be higher, as FC_{org} has been detected in these surface sediments by optical and Raman spectroscopy methods (Supplementary Figs 3 and 4, Supplementary Discussion). Therefore, the highest measured $\Delta^{14}C$ sets the minimum FC_{org} to 14% in average suspended sediment and 42% in bed sediment. On the basis of Himalayan river gravel and bedrock[10,12,18] data (Supplementary Table 1, Supplementary Fig. 2), average C_{org} content in the source rocks is between 0.05% and 0.08%. Using the TOC versus Al/Si relationship (Fig. 2) and assuming there is no fossil C_{org} oxidation during erosion and transport, we can limit the maximum proportion of FC_{org} to 20% in average suspended sediment and to 100% in bed sediment. Combining the C_{org} content in source rocks with $\Delta^{14}C$ constraints, we conservatively estimate that FC_{org} represents 10–20% of TOC in suspended sediments and 40–100% of TOC in bed sediments.

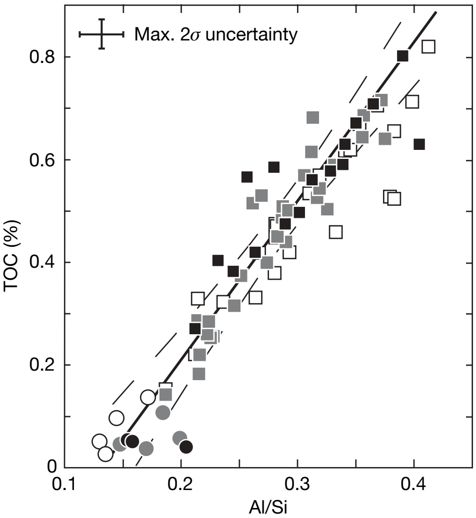

Figure 2 | TOC of riverine sediments as a function of their Al/Si ratio. Filled grey symbols, Brahmaputra; open symbols, Ganges; filled black symbols, Lower Meghna. TOC is very low in the sandy and quartz-enriched bed sediments (circles). In the suspended sediments (squares), TOC linearly increases with the relative proportion of aluminous fine-grained minerals. Sediments from the three rivers define comparable positive trends, indicating they have similar C_{org} loading relative to their composition. Best fit (solid line) and 95% confidence interval (dashed lines) are shown for Lower Meghna sediments.

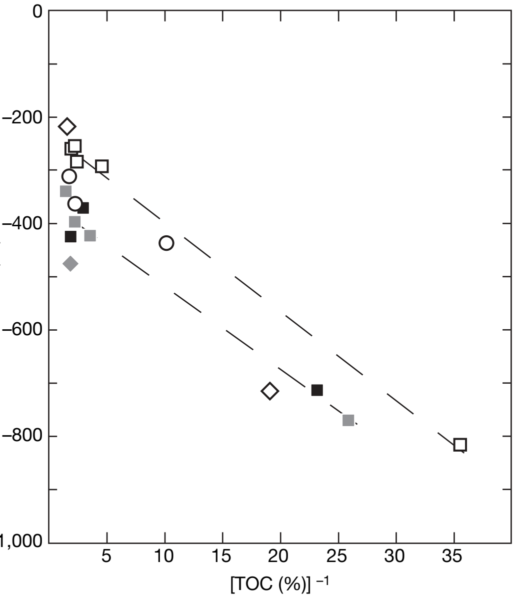

Figure 3 | Radiocarbon composition of acid-insoluble C_{org} extracted from riverine depth profile sediments as a function of 1/TOC. Filled grey symbols, Brahmaputra; open symbols, Ganges; and filled black symbols, Lower Meghna. Circles, monsoon 2002; squares, monsoon 2004; diamonds, monsoon 2005. Sediments from each river define a mixing trend between bed sediment dominated by aged C_{org} and suspended sediments with higher TOC and dominated by modern C_{org}. Maximum 2σ uncertainties on $\Delta^{14}C$ and TOC are respectively 5‰ and 0.03%.

Suspended sediment fluxes have been measured for the Ganges and the Brahmaputra, and the average total flux is around $1 \times 10^9\,\mathrm{t\,yr^{-1}}$ (ref. 13). The flux of bed sediment is not directly measured, but geochemical mass balance implies that bed sediment flux plus floodplain sequestration are almost equal to the suspended sediment flux[8]. Taking into account these sediment fluxes, we estimate that the G-B system delivers $(3.1 \pm 0.3) \times 10^{11}\,\mathrm{mol\,yr^{-1}}$ of RC_{org} to the Bay of Bengal.

Quantifying the corresponding CO_2 uptake requires determination of the proportion of this RC_{org} that is preserved and effectively buried in the Bengal fan. Sediment delivery to the Bengal delta shelf and active channel-levee system[14], which directly reflects the input of the G-B and their sources, has been stable over the past 20 kyr, as shown by constant Sr and Nd isotopic ratios[19]. In these sediments, TOC and Al/Si define a positive linear correlation statistically identical to that defined by G-B sediments (Fig. 4). It indicates similar C_{org} loads for river and marine sediments. The relative abundances of terrestrial and marine biomarkers as well as their stable isotopic compositions show that C_{org} is largely dominated by terrestrial matter (Supplementary Table 3, Supplementary Fig. 5, Supplementary Discussion), as already observed for ancient Bengal fan sediments[20–22]. Therefore, the terrestrial C_{org} loading of Bengal fan sediments roughly equals that of G-B river sediments. The Bengal fan appears to be able to preserve and bury terrestrial C_{org} that is exported by the G-B system. This strongly contrasts with other active margins where the burial efficiency is much lower and limited to ~30% (refs 2–4).

This extreme burial efficiency probably derives from the transport dynamic, with rapid sediment accumulation on the delta shelf (up to $30\,\mathrm{cm\,yr^{-1}}$; ref. 23). In shallow water, where C_{org} oxidation is theoretically most efficient, C_{org} is protected from oxidation by quick burial under a thick layer of fresh sediments. Typical O_2 penetration depth in such sediments is only a few centimetres[24], that is, the same order of magnitude as the Bengal shelf annual accumulation[23]. Therefore, C_{org} exposure time to O_2 is only a few years or so, insufficient for effective C_{org} oxidation. Sediments are finally transferred to the deep-water zone by turbiditic currents and deposited in channel-levee systems[25,26]. Identical C_{org} loading in the shelf, active channel-levee and mid-deep fan sediments (Supplementary Fig. 6) indicates that C_{org} oxidation during sediment transfer from shelf to deep ocean is negligible. In addition, river discharge and precipitation are responsible for a negative salinity anomaly, and maintain stratified waters in the Bay of Bengal[27,28]. Combined with high productivity sustained by high nutrient flux delivered by the G-B, this generates an extended O_2 minimum zone and generally low O_2 concentrations in the Bay of Bengal[27,28]. C_{org} oxidation is thus limited by both short exposure to, and low availability of, O_2.

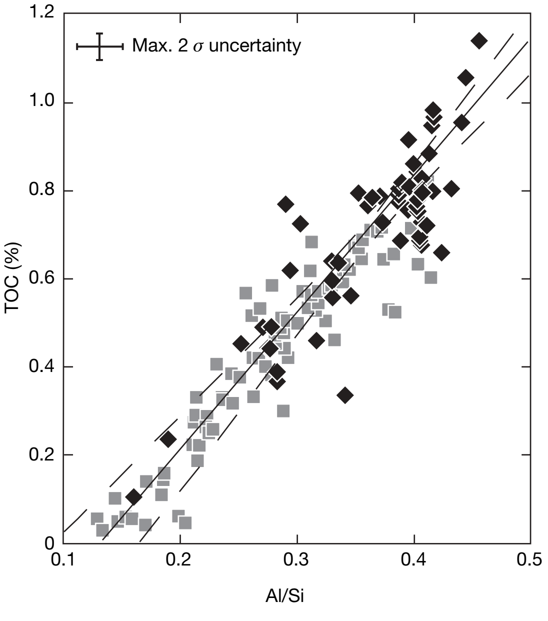

Figure 4 | TOC of riverine and fan sediments as a function of their Al/Si ratio. Filled black diamonds, TOC of the Bengal fan sediments, with best fit (solid line) and 95% confidence interval (dashed lines). Marine sediments define a positive trend highly comparable to that defined by river sediments (filled grey squares), suggesting they have similar C_{org} loads.

Climatic and tectonic factors produce high physical erosion rates and a major influx of particles and nutrients to the ocean that favour long-term C_{org} burial. This is characteristic of the Himalayan basin and is also true for other comparable basins, such as Oceania, that generate comparable sediment influx with high burial efficiency[9,29]. Uplift and erosion result in rapid sediment accumulation, which favours C_{org} preservation with globally significant fluxes. Active orogenesis affects the export and burial of C_{org} for periods of 10^6–10^7 years and tends to buffer long-term atmospheric CO_2 and O_2 levels.

METHODS SUMMARY

Sampling. Detailed sampling of the Ganges, Brahmaputra and Lower Meghna was performed during the 2002, 2004 and 2005 monsoons in Bangladesh. Suspended sediments were taken at different depths in the river channel and bedload was dredged from the bottom of the river. Marine sediments were taken from short cores drilled in the Bengal shelf, channel-levee system and deep-sea fan by the RV *Sonne* in 1994 (cruise SO 93).

Organic carbon analyses. Before organic carbon content and isotopic measurements, sediments were leached with HCl (4 wt%) at 80 °C in order to remove any trace of carbonate. TOC and $\delta^{13}C$ of bulk C_{org} were measured by elemental analyser-isotope ratio mass spectrometer (EA-IRMS) on a modified EuroVector EuroEA3028-HT coupled with a GV Isoprime. The overall 2σ uncertainties associated with the TOC and $\delta^{13}C$ of bulk C_{org} determination are 0.03% and 0.25‰, respectively. ^{14}C measurements were performed on the LMC14 (Gif sur Yvette) AMS national facility. After soluble C_{org} extraction with dichloromethane, n-alkane relative abundances were determined on a HP 5890 gas chromatograph-mass spectrometer (GC-MS) and compound specific $\delta^{13}C$ were measured on a GV-Isoprime gas chromatograph-isotope ratio mass spectrometer (GC-IRMS).

Fossil organic carbon detection. Fossil organic carbon was identified and characterized by Raman microspectroscopy and high-resolution transmission electron microscopy (HRTEM). Raman spectra were obtained on untreated raw sediments with a Renishaw INVIA Reflex Raman micro-spectrometer at the Laboratoire de Géologie of the Ecole Normale Supérieure, Paris, France. HRTEM was performed on a JEOL 2010 microscope with an acceleration voltage of 200 kV, after concentration of carbonaceous material by HCl/HF acid treatment at low temperature (below 70 °C).

Full Methods and any associated references are available in the online version of the paper at www.nature.com/nature.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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Author Contributions V.G. and C.F.L. are the main authors. O.B. conducted the fossil C_{org} characterization. P.F. and F.P. were involved in the organic geochemistry part of the study. H.K. provided the Bengal fan sediments and the framework of sedimentological interpretation.

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METHODS

Sampling. Detailed sampling of the Ganges, Brahmaputra and Lower Meghna was performed during the 2002, 2004 and 2005 monsoons in Bangladesh. On the basis of daily river survey data from the Bangladesh Water Development Board (BWDB), sediment flux during the monsoon is three orders of magnitude higher than during the dry season[13]. As a consequence, ~95% of the sediment flux is transported during the monsoon, and suspended sediments sampled during the monsoon are therefore representative of the total sediment flux. Suspended sediments were taken at different depths in the river channel and bedload was dredged from the bottom of the river. The sampling was systematically performed in the main channel where the velocities are the highest. In 2005, we also performed a depth sampling in a low velocity zone close to the bank to improve the description of the whole river section. Sediments were sampled using a 5-l Nalgene bottle, ballasted and equipped with a depth probe. At a given depth the bottle was opened, then lifted on board after filling. During filling, the depth slightly increases by about one metre; the average depth was recorded as the reference depth. The total contents of the bottle were carefully transferred into opaque plastic containers and sealed free of air. Samples were then filtered at 0.2 μm through a 90 mm nitrile membrane. Special care was taken to recover particles adsorbed at the surface of the container. Suspended sediment concentrations were weighed for each sample. Major and trace element concentrations were measured by ICP-AES and ICP-MS at the Service d'Analyse des Roches et des Minéraux (CRPG-Nancy) on bulk sediment after lithium metaborate fusion.

Marine sediments were taken from short cores drilled in the Bengal fan by the RV *Sonne* in 1994 (cruise SO 93)[14,30]. For cores from the channel-levee system, deposition ages have been estimated on the basis of foraminifer ^{14}C ages and stable isotopes; sediments used in this study were deposited during the last glacial–interglacial cycle. Sediments from the Bengal shelf were dated using gamma-spectrometric and radiocarbon methods[23]. We used a sample set that covers the past 500 yr of sedimentation, including modern samples.

Organic carbon analyses. G-B river sediments as well as Bengal fan sediments contain significant amounts of detrital carbonates including dolomite[31–33]. C_{org} concentration and isotopic measurements must therefore be performed on decarbonated sediments. Efficient dolomite dissolution was achieved through 1 h leaching with 4 wt% HCl at 80 °C. After 50 °C oven drying, the C_{org} content was determined either by elemental analysis on a EuroVector EuroEA3028-HT or by conventional CuO combustion[20]. The problem with aqueous acid leaching is that an unknown proportion of C_{org} is solubilized by acid hydrolysis. For a representative set of river and marine sediments, the amount of C_{org} solubilized during the decarbonatation was measured in order to obtain the real TOC. C_{org} content measured after decarbonation and TOC define a very good linear correlation (Supplementary Fig. 6). We used this relationship to calculate TOC for each sample from C_{org} content measured after decarbonation. The overall 2σ uncertainty associated with the TOC determination is 0.03%.

Δ^{14}C were also determined on decarbonated fractions. Acid-insoluble C_{org} was converted into pure CO_2 at CRPG (Nancy) following the conventional CuO combustion method[20]. ^{14}C measurements were performed on the LMC14 (Gif sur Yvette) AMS national facility. Maximum overall uncertainty on Δ^{14}C calculations was estimated to be 5‰.

Soluble C_{org} was extracted from bulk sediments with dichloromethane using an Accelerated Solvent Extractor (ASE 200). n-alkane relative abundances were determined on a HP 5890 GC-MS and compound specific δ^{13}C was measured on a GV-Isoprime GC-IRMS.

Raman spectroscopy and HRTEM. Raman spectra were obtained with a Renishaw INVIA Reflex Raman micro-spectrometer at the Laboratoire de Géologie of the Ecole Normale Supérieure, Paris, France. Spectra were excited at room temperature with the 514.5 nm line of a 20 mW Ar Spectra Physics laser through a LEICA 50X long-working distance objective (NA 0.50). The laser spot at the sample surface had a diameter of approximately 3 μm and a power of 1 mW, which should be low enough to avoid any spectral change or sample destruction due to light absorption and local temperature increase[34]. Light was dispersed by a holographic grating with 1,800 grooves mm^{-1}. The spectral resolution of about 1.4 cm^{-1} was determined by measuring a neon lamp emission. The spectrometer was calibrated for every session by measuring the position of the neon lamp emission and/or a silicon wafer. The dispersed light was collected by a RENCAM CCD detector. The synchroscan mode from 100 to 2,000 cm^{-1} was selected in order to retrieve Raman modes from both carbonaceous matter (CM; 1,300–1,600 cm^{-1} region) and silicates/carbonates (100–1,100 cm^{-1}). Raman investigations were directly performed on the raw samples, without any preparation. The first-order Raman spectrum of disordered CM exhibits a graphite G band at 1,580 cm^{-1}, corresponding to in-plane vibration of aromatic carbons, and several defect bands (including the most important at 1,350 cm^{-1}), corresponding to physico-chemical defects[34].

HRTEM was performed on a JEOL 2010 microscope with an acceleration voltage of 200 kV. Low- and high-magnification observations as well as electron diffraction were conducted on river and oceanic sediment samples. In particular, the high-magnification 002 lattice fringes mode was used to image directly the aromatic skeleton of CM together with selected area electron diffraction to detect the presence of triperiodic graphite. For HRTEM investigations, CM was concentrated from river and marine sediments using HCl/HF acid treatment at low temperature (always below 70 °C). More details regarding the sample preparation and transmission electron microscopy analysis of natural solid CM may be found in ref. 35 and references therein.

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