Variations of carbon transport in the Yellow River, China

Jianrong Liu, Xiangfang Song, Zhimin Wang, Lihu Yang, Zhenyu Sun and Wenjia Wang

ABSTRACT

The Yellow River is the second largest river in China. Carbon transport by the Yellow River has significant influence on riverine carbon cycles in Asia. In order to monitor seasonal and spatial variations of carbon concentrations and to estimate carbon exports, water and suspended solids were sampled every 10 days at three representative stations (Qingtongxia, Tongguan, and Luokou) along the mainstream of the Yellow River. Results showed that riverine carbon was mainly in dissolved form, except during flood period and water and sediment regulation (WSR) scheme, when particulate organic carbon (POC) dominated. Concentration of dissolved inorganic carbon was mostly 5 to 10 times higher than that of dissolved organic carbon (DOC). DOC was mainly related to a natural process (leaching effect) in the upstream and anthropogenic activities in the midstream (domestic sewage and fertilizer application) and downstream (industrial wastewater). POC was connected with high suspended solids. Annually carbon delivered to the Bohai Sea was 1.34×10^{12} g/yr, accounting for 0.15% of the global total riverine carbon flux. Mean DOC exported accounted for 0.12% of the Asian rivers' DOC flux. WSR played an important role in the carbon transport, which accounted for 1/5 to 1/3 of the corresponding annual fluxes.

Key words | carbon concentration, carbon export, China, water sediment regulation, Yellow River

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INTRODUCTION

Rivers, in particular, large rivers, play a critical role in the global carbon cycle by linking the atmosphere, the land, and the ocean system (Ludwig *et al.* 1996; Lal 2003; Dagg *et al.* 2004; Alvarez-Cobelas *et al.* 2012). The unidirectional flux from terrestrial to ocean carbon reservoirs influences the biogeochemical processes within the aquatic and coastal environment (Ran *et al.* 2013). During the latter half of the 20th century, perturbations from natural and human activities, including climate and land use changes, population increases, water consumption, fertilizer application, industrial wastewater disposal, and damming of rivers, have significantly altered the global river systems and increased nonconservative behaviors of carbon in waters (Klavins *et al.* 2012; Liu *et al.* 2014). Additionally, the increasing anthropogenic emissions to atmospheric CO_2 leads to difficulties in attempts to balance the biogeochemical cycle of carbon on a global scale (Schlesinger & Melack 1981). Therefore, a full understanding of carbon transport and transformations within rivers, especially under anthropogenic interferences, has crucial implications for the global carbon budget.

It is estimated that about 0.9 Gt of carbon is carried every year by the world's large rivers (Meybeck 1993b), among which Asian rivers make significant contributions as they deliver about 70% of the global total suspended solids (Milliman & Meade 1983). The Yellow River is the second largest river in China and ranks one of the highest in terms of the sediment load among the world's rivers (Wang *et al.* 2007). A few previous studies have been carried out to estimate the amount of carbon exported by the Yellow River. However, these studies were mostly based on a single sampling event or separated cruises (Hu *et al.* 1982; Gan *et al.* 1983; Zhang *et al.* 1992; Cauwet &

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Mackenzie 1993; Zhang *et al.* 1995; Chen & Wang 1999), which could not fully reflect the carbon delivery dynamics, as the hydrological regime was highly variable between different seasons (Ran *et al.* 2013). With 2–3 orders of magnitude in variance, the obtained carbon fluxes were also likely misleading. Additionally, these studies were principally based on carbon concentration and water discharge before 2000. Since 1999, the Yellow River has never dried up. The annual water discharge actually shows an increasing trend (Zhang *et al.* 2009). However, the suspended sediment supply from the Yellow River has decreased by 84% due to both anthropogenic (e.g. dam and reservoir constructions) and natural causes (Dai *et al.* 2009). Furthermore, in order to flush away the sediment deposited in the watercourse and reservoirs and prepare for the coming flooding in the wet season, since 2002, the Sanmenxia and Xiaolangdi reservoirs have been operated to regulate water and sediment during late June to early July to control the speed of deposition in the lower reaches (Yellow River Conservancy Commission 2008; Fu *et al.* 2012; Zhang *et al.* 2013). The water and sediment regulation (WSR) scheme is the only regular activity in the world with extreme human disruption of river-matter transport (Wang *et al.* 2013a). Therefore, the current study aims to: (1) systematically monitor seasonal and spatial variations of carbon concentrations in both organic and inorganic forms; (2) calculate carbon fluxes in the upstream, midstream, and downstream of the river and the annual exports of carbon to the Bohai Sea; and (3) estimate the impacts of the WSR scheme on annual transport of carbon species in the Yellow River, under the natural conditions and human activities which changed greatly after 2000. The obtained results could be of benefit for ongoing assessment of dynamics and biogeochemical processes of Asian and global riverine carbon cycling.

DATA AND METHODS

Description of sampling sites

The Yellow River originates from Qinghai–Tibet Plateau with most of the water contributed from the ice melt and snowfall. It flows about 5464 km and drains 75.2×10^4 km^2 before entering the Bohai Sea. The climate of the river basin is arid to semi-arid. Average annual rainfall is 476 mm, while average annual evaporation is 1100 mm (Chen *et al.* 2003). The midstream of the river flows through the Loess Plateau, which contributes 90% of the sediment load and makes the Yellow River one of the most turbid rivers in the world. Therefore, compared with other large rivers of the world, its low water discharge and high sediment load are distinctive characteristics.

In the current study, three hydrological stations, namely, Qingtongxia (QTX), Tongguan (TG), and Luokou (LK), are selected as the representative observation and sampling sections, which stand for the upstream, midstream, and downstream of the Yellow River, respectively (Figure 1).

Sampling and measurement

Water samples were collected every 10 days to measure the concentrations of dissolved total carbon (DTC, mg/l), dissolved organic carbon (DOC, mg/l), and particulate organic carbon (POC, mg/l). Water samples were filtered through preweighted and precombusted (at 450 °C for 6 h) GF/F glass-fiber papers. Filters were dried at 50 °C for 24 h for POC analysis. Filtrates were separated into two parts and kept in 1-liter high density polyethylene bottles, respectively, which have been pre-washed by acid and neutralized. The part for DTC analysis was untreated, while the other for DOC analysis was acidified with H_3PO_4 at pH 2 to remove dissolved inorganic carbon (DIC). All samples were preserved at 0 to 4 °C before measurement. Concentrations of carbon were determined using a high-temperature catalytic oxidation technique with the Apollo 9,000 TOC Analyzer. Analysis of DOC concentration was carried out during 2005 to 2007 at three stations. Concentrations of DIC were obtained by subtracting DOC from DTC, and concentrations of total carbon (TC) were basically the sum of DTC and POC. Analysis of DTC and POC concentrations was mainly carried out for the year of 2006. However, due to some sampling problems, POC analysis was mainly obtained for LK station and QTX (July–November).

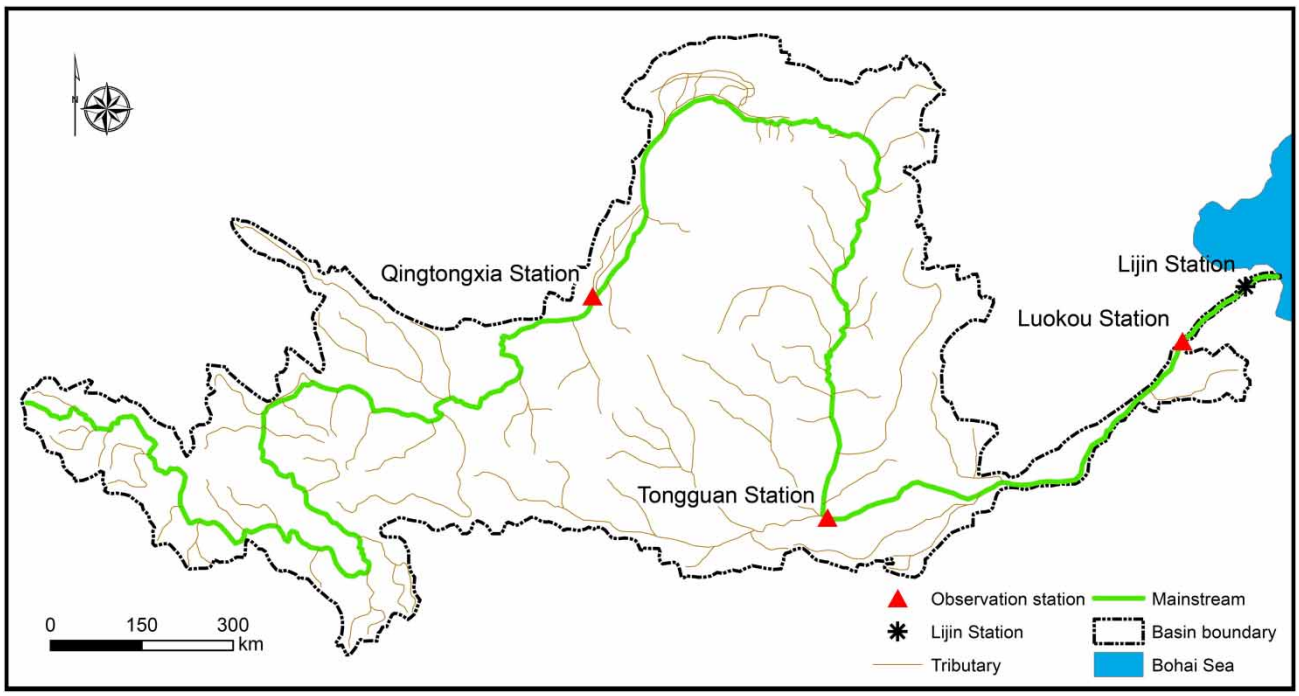

Figure 1 | Location of the observation and sampling sites.

Environmental parameters were also measured during sampling, including water flow (Q, m^3/s), water temperature (T, °C), electrical conductivity (EC, μs/cm), pH value (pH), dissolved oxygen (DO, mg/l), total suspended solids (TSS, g/l), and chemical oxygen demand (COD, mg/l).

Flux calculation

Carbon flux within a time interval was calculated from the total water flow within the time interval multiplied by the concentration at the midpoint of the interval. Total flux was calculated as the sum of loadings for all time intervals (Dolan *et al.* 1981; Parks & Baker 1997). Carbon flux is calculated using the equation:

$$\mathrm{Flux_C} = \sum \mathrm{con} \times Q \quad (1)$$

where $\mathrm{Flux_C}$ refers to daily (g/day) or annual carbon flux (g/yr), *con* is the concentration of carbon in different forms (mg/l), Q refers to the water discharge through a section in unit time (m^3/s).

RESULTS AND DISCUSSION

Carbon concentration

DOC concentration showed relatively complex seasonal variations at three stations along the mainstream of the Yellow River (Figure 2). On average, DOC concentrations varied between 1.12–6.66, 1.29–9.37, and 1.00–10.16 mg/l for the upstream, midstream, and downstream, respectively (Table 1). Relatively high DOC concentrations were found for the TG station, with 5.24 and 5.19 mg/l for 2005 and 2007, respectively, whereas relatively low DOC concentrations were found for the LK station. Variations of DOC were generally more intense in the downstream.

Concentrations of DIC and DTC varied synchronously with time at QTX, TG, and LK in the mainstream of the Yellow River (Figure 3). DIC dominated DTC as ratios of DIC/DTC were more than 75%, and the highest reached more than 90%. For QTX, TG, and LK, DIC/DTC ratios varied in the ranges 75–93%, 78–95%, and 82–98%, respectively. Concentrations of DIC were mostly 5 to 10 times higher than concentrations of DOC. At QTX and TG,

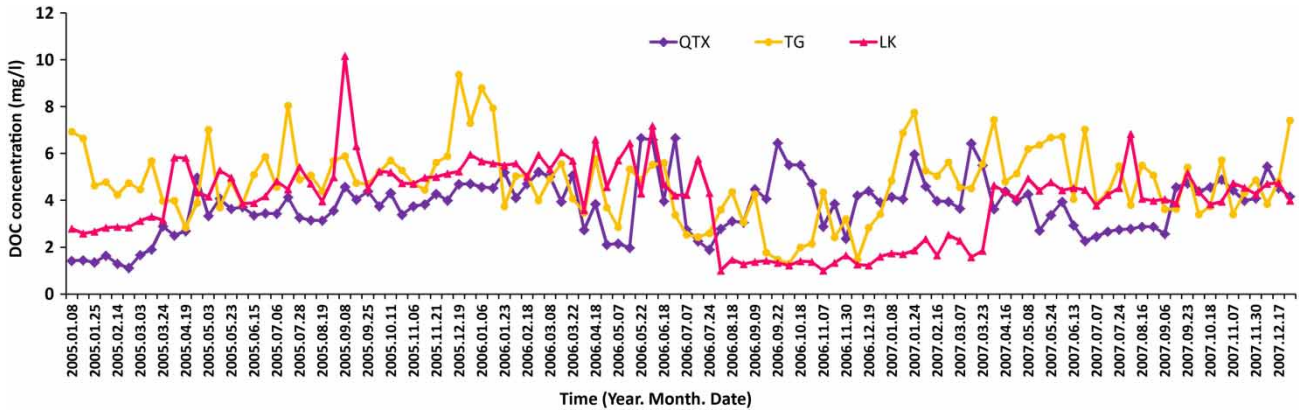

Figure 2 | Seasonal variations of DOC concentrations at QTX (upstream), TG (midstream), and LK (downstream) during 2005 to 2007.

DIC/DOC ratios were concentrated at 3–12.9 and 3.5–19.8, respectively. At LK this ratio was distributed from 4.7 to 46. Therefore, the dissolved carbon in the water of the Yellow River was mainly in the inorganic form, and the DIC gradually increased in the proportion of DTC from upstream to downstream.

Statistical information regarding carbon species is described in Table 2. The mean DIC concentrations were 27.31–38.17 mg/l and increased from upstream to downstream, indicating that the inorganic carbonous substances were continuously added (i.e. by $CaCO_3$ dissolution, bacteria respiration), or the increased substances were more than those removed by physical process (i.e. dilution, sedimentation), biogeochemical activities (i.e. adsorption, $CaCO_3$ precipitation, biological production), or material exchange with surrounding environments during the flow of the river (Abril *et al.* 2003; Cai 2003; Ortega *et al.* 2005, 2008; Hans *et al.* 2011; Guo *et al.* 2012; Huang *et al.* 2012; Liu *et al.* 2014). From upstream to midstream, the minimum DIC at the TG station was less than that at the QTX station, indicating some periods in which the environments helpful in removing DIC probably existed. However, the average DIC at the TG station was slightly higher than that at the QTX station, indicating that the added DIC was still more than that removed in general. The minimum and average DIC at the LK station were much higher than those at the TG station, which showed that the added DIC substances were still more than those removed from midstream to downstream. The variation coefficients of DIC at the QTX, TG, and LK stations were 0.33, 0.42, and 0.18, respectively, reflecting the fluctuations of DIC at upstream and midstream were more intense than that at downstream. As the DIC dominated DTC, variations of DTC were similar to those of DIC in ranges, averages, and variation coefficients at the three stations.

Concentrations of POC were larger than other forms of carbon (Figure 3). At LK, POC concentration was relatively low during fall and winter, but relatively high during spring. POC concentrations reached their maximum values during late June to early July (the WSR period) and September (the flooding period). At QXT, although the POC concentration was high during the flood period, the highest value was lower than that at LK during the same period. Furthermore, in the downstream of the Yellow River, carbon transport was mainly in dissolved form as the POC/DTC ratio was mostly lower than 1 (Figure 4), except during the periods of WSR and flooding, when the particulate carbon dominated. The POC% (POC/TSS × 100%) of LK station ranged between 0.63 and 1.20 (averaged at 0.82%), and generally decreased exponentially with increasing TSS concentration.

Factors controlling carbon concentrations

Principal component analysis (PCA) results

The PCA was applied to extract the most important physicochemical factors controlling carbon species along the mainstream of the Yellow River (Table 3). For the QTX station, four components were extracted. The first PC

associated with the DOC in water. The second PC indicated that particulate carbon input into the upstream was correlated with EC and TSS. The third PC was mainly related to the DIC, and the water discharge showed contrary contributions to DIC and DTC levels. The fourth PC was correlated with COD and pH, indicating the acid-base status and the amount of organic pollutants may not connect with the carbon species in the upstream.

For the TG station, three components were extracted. The first and the third PC represented the factors related to water quality and discharge, respectively. The second PC mainly related to the dissolved carbon in the midstream. For the LK station, three components were extracted, which were generally associated with the dissolved inorganic, particulate organic and dissolved organic forms of carbon to the downstream, respectively. The first PC indicated that the main environmental impacts to DIC were water temperature, DO, pH, and EC. The second PC denoted that water discharge and suspended solids were favorable to contribute the particulate carbon content. The third PC showed the amount of organic pollutants influenced the DOC level in the downstream.

Factors controlling DOC concentration

Natural DOC in rivers ranged from labile forms, such as simple sugars and organic acids, to highly refractory forms, such as humic acids, polysaccharides, polypeptides and some colloidal materials (Schlesinger & Melack 1981; David *et al.* 1999). The main sources of DOC included decomposition of terrestrial organic matter (allochthonous) and *in situ* production by aquatic plants and microbes (autochthonous). In addition, impacts of anthropogenic activities, such as fertilizer application, domestic sewage and industrial wastewater disposal cannot be ignored either (Zhang *et al.* 2013). Within aquatic systems, the quantity and form of DOC could be altered by a variety of processes including microbial breakdown, sorption/desorption to particles, sedimentation and photodegradation (Stanley *et al.* 2011).

The main source of DOC in the upstream of the Yellow River was terrestrial. The strong leaching effect of high organic matter soil in the Tibetan Plateau (Finlay *et al.* 2006) made a great contribution to DOC. The soil organic carbon content in the headwater region was mostly larger

Table 1 | Concentration and export of DOC for three stations in the mainstream of the Yellow River during 2005–2007

Year	Water yield (10^9 m^3)			DOC concentration (mg/l)*			DOC export ($\times 10^9$ g/yr)		
	QTX	TG	LK	QTX	TG	LK	QTX	TG	LK (WSR, Ratio %)
2005	19.51	22.71	22.89	3.24 (1.12–4.97, 0.34)	5.24 (2.84–9.37, 0.25)	4.56 (2.59–10.16, 0.31)	66.0	113.7	107.7 (21.1, 19.6)
2006	19.99	23.13	21.96	4.09 (1.90–6.66, 0.34)	3.86 (1.29–8.79, 0.44)	3.66 (1.00–7.18, 0.58)	81.6	84.5	83.3 (20.7, 24.8)
2007	21.02	24.80	23.43	3.96 (2.27–6.43, 0.25)	5.19 (3.40–7.75, 0.24)	3.83 (1.56–6.83, 0.32)	82.2	121.2	100.5 (16.1, 16.0)
Total	20.17	23.55	22.76	3.76 (1.12–6.66, 0.33)	4.76 (1.29–9.37, 0.33)	4.01 (1.00–10.16, 0.41)	76.6	106.5	97.2 (19.3, 19.9)

*Mean (ranges, variation coefficient) values.

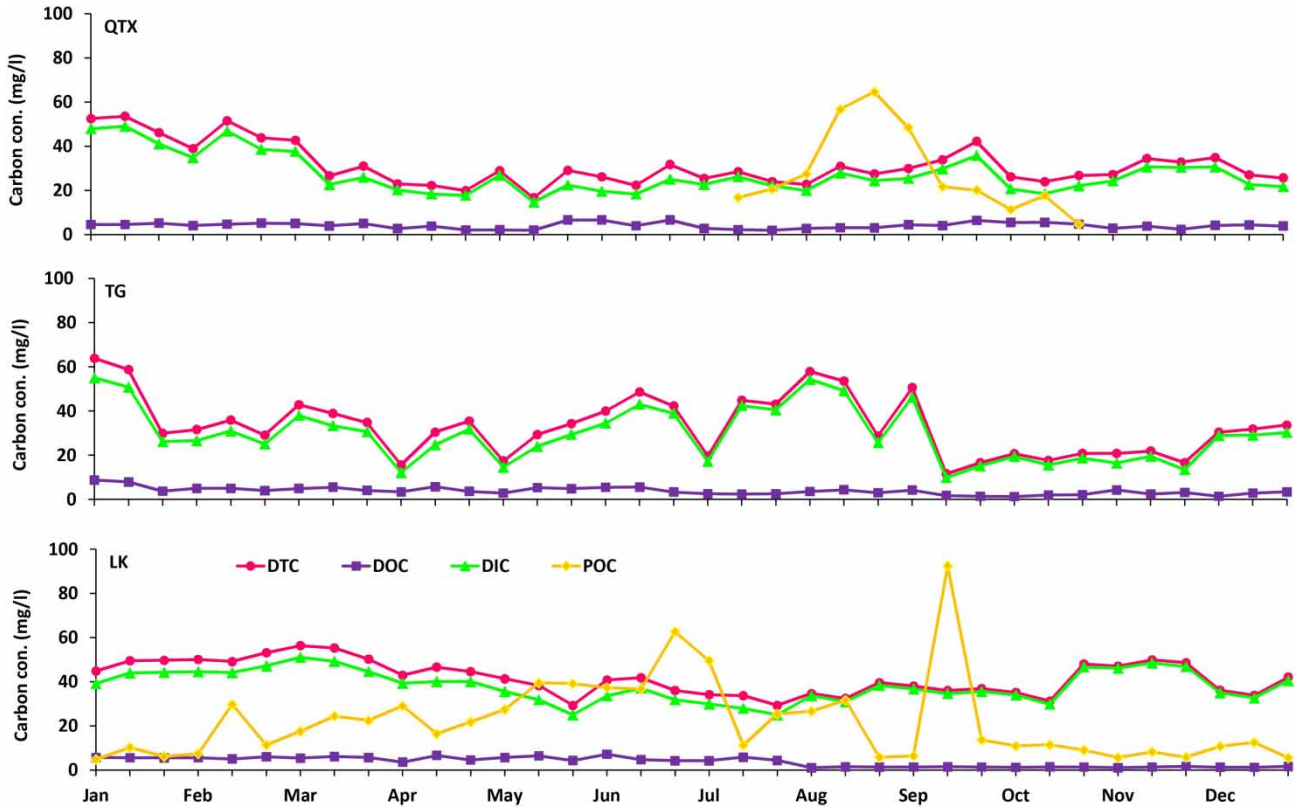

Figure 3 | Seasonal variations of carbon concentrations in different forms at QTX (upstream), TG (midstream), and LK (downstream) during 2006.

than 1.5% and could reach up to 39% in some areas. Comparatively, the soil organic carbon content in the middle reaches (the Loess Plateau) was quite low, normally below 0.8% in most parts and even less than 0.5% in the catchments near the desert (Ran 2013). However, the DOC concentration in the midstream was relatively higher than that in the upstream. About 107 million people live in the Yellow River basin. According to the statistics (National Bureau of Statistics of China 1986–2008), from 2005 to 2007, the population increased by 3.70 times in the areas from QTX to TG, which was much higher than that in the catchment above QTX (0.34 times) and from TG to LK (0.36 times). Accordingly, disposal of domestic sewage in the middle reaches was comparatively higher than those from upper and lower reaches. In addition, irrigation is the key to agriculture in the basin, as nearly half of the farmland (1.2×10^5 km^2) was irrigated by the Yellow River (Chen *et al.* 2003; Yang *et al.* 2004). The nitrogen fertilizer applied in the catchment from TG to LK was increased on average by 5.68 times during 2005 to 2007. The inefficient irrigation methods, mainly as flood irrigation, resulted in a large amount of high-DOC water returning to the Yellow River (Zhang *et al.* 2008). Therefore, the high concentration of DOC in the midstream was mostly connected with agricultural pollution and domestic sewage. This also explained why the PCA result showed that the natural environment controls had less impact on the carbon species in the midstream. Furthermore, fertilizer utilization increased by 3.18 times from 1985 to 2007 in the upstream (Yu *et al.* 2010). Therefore, besides the high leaching effect additional DOC input from agricultural activities was also a significant contribution. Downstream of the Yellow River was the only section that was dramatically influenced by industrial wastewater. From TG to LK, the input of industrial wastewater increased by 2.56 times, whereas there were only slightly increasing trends or even decreasing trends observed for the QTX–LK section and upper QTX, respectively. However, as the leaching effect and contributions from agricultural (nitrogen fertilizer was only increased

by 0.5 times) and sewage were all dramatically decreased, the concentration of DOC was lower than TG and QTX. This also explained why the fourth PC extracted for DOC was mainly related to COD.

DOC concentration also showed different correlations with water discharge at the three stations (Figure 5). At QTX, DOC concentration did not show significant correlation with water discharge. For TG and LK, DOC concentrations decreased exponentially with the elevated water discharge, which indicated a dilution effect. In particular, at LK, the DOC concentration presented different decreasing rates with changes in water discharge. The DOC with concentration <3.5 mg/l was mainly generated by low water discharge (mostly <800 m^3/s), and the DOC decreased more rapidly with the increasing water discharge. When water discharge was higher than 2000 m^3/s (WSR scheme or flooding periods), the corresponding DOC concentration was also relatively high (>3.5 mg/l). There are two main reservoirs (Sanmenxia and Xiaolangding) located along the mainstream from TG to LK. Most of the suspended sediments were captured and deposited due to impoundment. With extension of water residence time and removal of the limitation of light, *in situ* production of DOC was promoted. During the high discharge period, water containing relatively high DOC was released from reservoirs. Furthermore, for all stations no EC was extracted by the DOC components, as the conductivity in water was primarily affected by the presence of inorganic dissolved solids (which usually carry positive or negative charges) but was unaffected by the organic compounds, which did not conduct electrical current very well.

Factors controlling DIC concentration

Aquatic inorganic carbon exists in three species, HCO_3^-, CO_3^{2-}, and CO_{2aq} (including H_2CO_3). DIC is defined as the sum of all three species, with most in the form of HCO_3^- (Meybeck 1993a). The total dissolved solids (TDS) in the Yellow River water are among the highest in China and even in the world. HCO_3^- is the most abundant anion, constituting 30–50% of TDS. The mean HCO_3^- in the mainstream of the Yellow River was 180–224 mg/l, much higher than concentrations of other ions (Chen *et al.* 2003; Li & Zhang 2005). Carbonate and silicate weathering consumed

Table 2 | Concentration and export of carbon species in the mainstream during 2006

	Carbon concentration (mg/l)*					Carbon export ($\times 10^9$ g/yr)				
	DIC	DOC	DTC	POC		DIC	DOC	DTC	POC	TC
QTX	27.31 (14.69–49.03, 0.33)	4.09 (1.90–6.66, 0.34)	31.40 (16.66–53.55, 0.30)	28.17 (4.65–64.49, 0.69)		533.6	81.6	615.2	–	–
TG	29.50 (9.86–55.05, 0.42)	3.86 (1.29–8.79, 0.44)	33.36 (11.63–63.84, 0.40)	–		673.8	84.5	758.3	–	–
LK	38.17 (24.94–51.02, 0.18)	3.66 (1.00–7.18, 0.58)	41.83 (29.24–56.35, 0.18)	20.96 (4.78–92.41, 0.85)		788.7	83.3	872.0	668.1	1540.1
Total	31.66 (9.86–55.05, 0.34)	3.87 (1.00–8.79, 0.45)	35.53 (11.63–63.84, 0.32)	22.76 (4.65–92.41, 0.81)	WSR	159.1	20.7	179.8	251.8	431.6
					Ratio (%)	20.2	24.8	20.6	37.7	28.0

*Mean (ranges, variation coefficient) values.

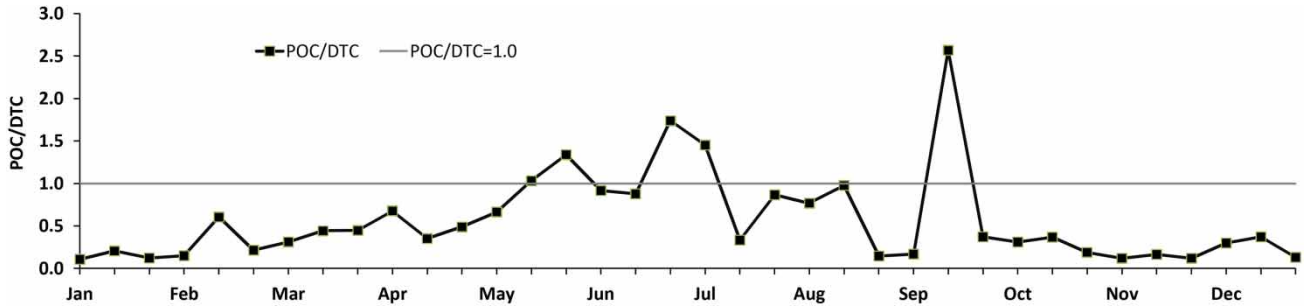

Figure 4 | Ratios of particulate carbon and dissolved carbon at the LK station. Carbon transport was mainly in dissolved form as the POC/DTC ratio was mostly lower than 1 except during the periods of WSR and flooding.

atmospheric CO_2, which was transferred into the river in the form of HCO_3^-, through the following simplified reactions:

$$\text{Carbonates: } CaCO_3 + CO_2 + H_2O \rightarrow Ca^{2+} + 2HCO_3^-$$

$$\text{Silicates: } CaSiO_3 + 2CO_2 + 3H_2O \rightarrow 2HCO_3^- + Ca^{2+} + H_2SiO_4$$

For the Yellow River basin, 55% of HCO_3^- was derived from atmospheric CO_2 by rock weathering, while 45% of HCO_3^- was supported by the carbonates (Li & Zhang 2005). The Loess Plateau, the biggest and deepest plateau in the world, largely overlaps the middle reaches of the Yellow River basin. The mineral composition of the loess is mainly evaporates, silicates, clays, and carbonates (Zhang *et al.* 1995). When the river flowed through the Plateau, the intense weathering of the carbonate-rich loess resulted in quite a high HCO_3^- concentration, and led to increasing DIC concentration from upstream to midstream. In the downstream, reservoirs prolonged the contact between near-bottom and sediment-interstitial waters and carbonate-rich particles, which resulted in very high DIC concentrations. Furthermore, the concentrating effect of high evaporative water loss, resulting from the arid climate and the agriculture use of water through irrigation systems, also elevated the HCO_3^- concentration (Cai *et al.* 2008).

At QTX and LK, DIC concentrations were generally negatively correlated with water discharge. High DIC concentrations were observed during the dry, low-discharge period of winter and early spring, whereas low concentrations were observed during the wet, high-discharge season of late summer and early fall (Figure 3). During high-discharge periods, DO in water was low, and the river was mainly in a reduction environment, which favored denitrification, while during dry seasons, the water discharge was smaller and the river was in an oxidation environment (Wang *et al.* 2013a). Internal processes such as denitrification, ammonium oxidation, net biological production and respiration, and $CaCO_3$ dissolution and precipitation may contribute to nonconservative behavior of DIC (Liu *et al.* 2014). These processes were probably more significant in the downstream due to the reservoir impoundment, as the first PC extracted for LK was related to environmental conditions (e.g. DO, water temperature, pH, and EC). However, there was no clear correlation between DIC concentration and discharge for TG. In the previous section, an environment in favor of removing DIC was detected in the midstream. It could probably be attributed to the organic acid contained in the DOC in the midstream neutralizing the HCO_3^- in the water, leading to decreasing DIC concentration. It may also explain the PCA result for TG where the DIC and DOC showed reverse contributions to the dissolved carbon in water.

Factors controlling POC concentration

In poorly turbid waters (TSS <100 mg/l), riverine POC originates mostly from soil erosion, whereas in highly turbid rivers or during major river floods in semi-arid environments, riverine POC probably originates from rock erosion (Meybeck 1993a). In the Yellow River, a considerable portion of POC originated from the mechanical erosion of sedimentary rocks and a small portion originated from soil horizons (Ran *et al.* 2013). Among all the factors that may control the POC concentration, the sediment solids showed a most significant

Table 3 | PCA results – rotated component matrix

		Component			
		1	2	3	4
QTX	T	– 0.941	0.154	0.026	0.185
	DOC	0.872	0.004	0.224	0.193
	DO	0.778	– 0.192	0.064	– 0.376
	EC	0.312	0.917	0.029	0.088
	POC	– 0.394	0.891	0.079	0.027
	TSS	– 0.454	0.865	0.058	0.009
	DIC	– 0.015	0.166	0.940	0.098
	DTC	0.220	0.152	0.913	0.140
	Flow	0.026	– 0.205	0.557	– 0.329
	COD	– 0.219	0.171	– 0.199	0.873
	pH	0.024	– 0.146	0.433	0.751
TG	T	0.928	– 0.013	0.239	
	pH	0.897	– 0.234	0.072	
	EC	0.894	0.191	– 0.186	
	DO	– 0.885	0.023	– 0.290	
	DTC	0.063	0.983	0.002	
	DIC	0.121	0.961	0.028	
	DOC	– 0.379	0.765	– 0.191	
	Flow	0.147	– 0.051	0.975	
LK	DO	0.906	– 0.245	– 0.126	
	DIC	0.895	– 0.134	0.239	
	T	– 0.869	0.341	0.048	
	DTC	0.850	– 0.141	0.430	
	pH	– 0.730	– 0.343	0.325	
	EC	0.677	– 0.314	0.241	
	TSS	– 0.176	0.931	0.103	
	POC	– 0.122	0.926	– 0.116	
	Flow	– 0.322	0.694	0.277	
	DOC	0.094	– 0.069	0.859	
	COD	0.054	0.330	0.578	

Notes: QTX – 4 components extracted; TG – 3 components extracted; LK – 3 components extracted; T – water temperature, DO – dissolved oxygen, EC – electrical conductivity, TSS – total suspended solids, Flow – water discharge, COD – chemical oxygen demand.

contribution. The second PCs extracted for QTX and LK both denoted the factor affecting soil and water losses. TSS showed a positive contribution to the POC concentrations in the upstream and downstream (Figures 6(a) and 6(b)). In winter, the upstream and midstream of the Yellow River was almost frozen. Consequently, water discharge and TSS stayed at very low levels, leading to low POC concentrations. As the temperature gradually increased, the ice and snow started to melt during late February to mid-April. As a result, water discharge, TSS and POC concentration all increased (Figure 3). The POC concentration reached its highest during flooding periods (August to September). Furthermore, during the WSR scheme, a large amount of sediment accumulated in reservoirs and the watercourse in the lower reach of the river was flushed out. The water discharge and TSS increased sharply within a short period, and resulted in the second highest POC concentration within the year. The WSR reflected an extreme human disruption of river-matter transport (Zhang *et al*. 2013).

The organic carbon transported in the Yellow River was mainly in particulate form, as the DOC/POC ratio at QTX and LK was generally lower than 0.6 (Figures 6(c) and 6(d)). The ratio decreased exponentially with the increasing TSS concentration. The lowest DOC/POC ratio (0.02–0.20) for LK was observed during WSR and flooding periods when the TSS was higher than 5000 mg/l. During these two periods, POC accounted for approximately 80–98% of the total organic carbon transport.

Carbon flux

Fluxes of dissolved and particulate carbon

Based on 10-day DOC concentrations (Figure 2) and 1-day flow using Equation (1), daily DOC fluxes in the mainstream of the Yellow River were calculated (Figure 7). Daily DOC fluxes were significantly influenced by water discharge, especially during the periods of the WSR scheme and flooding. On average, annual DOC flux for the upstream, midstream, and downstream was 76.6, 106.5, and 97.2 thousand tons ($\times 10^9$ g/yr), respectively (Table 1). During the WSR scheme of 2005–2007, the DOC flux at LK accounted for 19.6, 24.8, and 16.0% of the corresponding annual mean DOC flux, respectively.

Daily fluxes of dissolved inorganic and particulate carbon in the mainstream were also calculated (Figure 8). Daily DIC and POC fluxes were also highly influenced by water discharge, especially during high discharge periods and the WSR scheme in the downstream. As DIC dominated DTC, variations of daily DTC flux were similar to those of

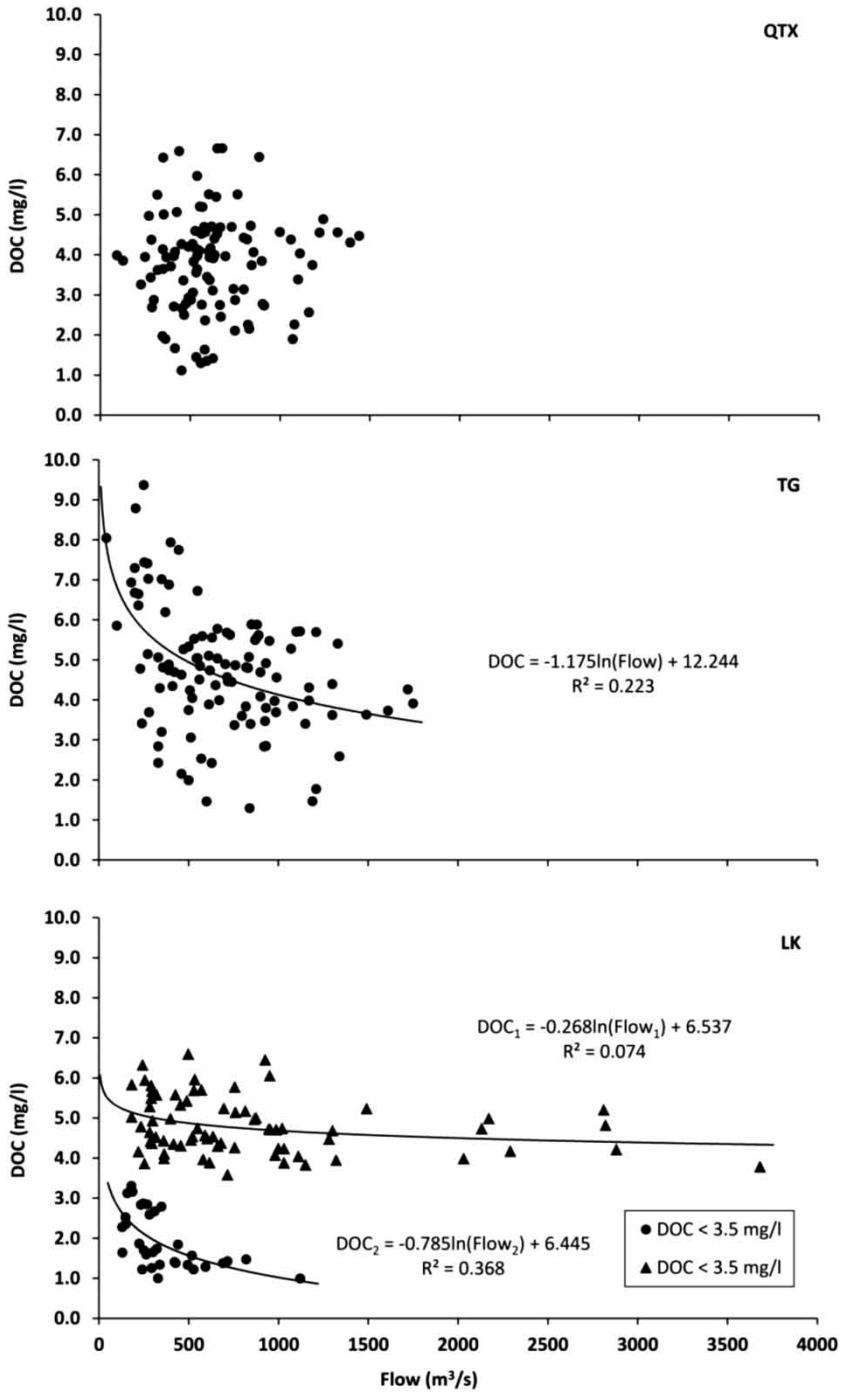

Figure 5 | Correlations between DOC concentrations and water flow. No significant correlation was found for QTX. Exponentially decreasing trends in DOC concentrations were found with the increasing of water discharge for TG and LK. At LK, different decreasing rates were found for DOC higher or lower than 3.5 mg/l.

DIC. From upstream to downstream, annual DIC fluxes were 533.6, 673.8, and 788.8 thousand tons, respectively (Table 2). With the addition of DOC fluxes in 2006, the DTC fluxes were 615.2, 758.3, and 872.0 thousand tons per year, respectively. Both of the DIC and DTC fluxes showed increasing trends from upstream to downstream. The DIC fluxes

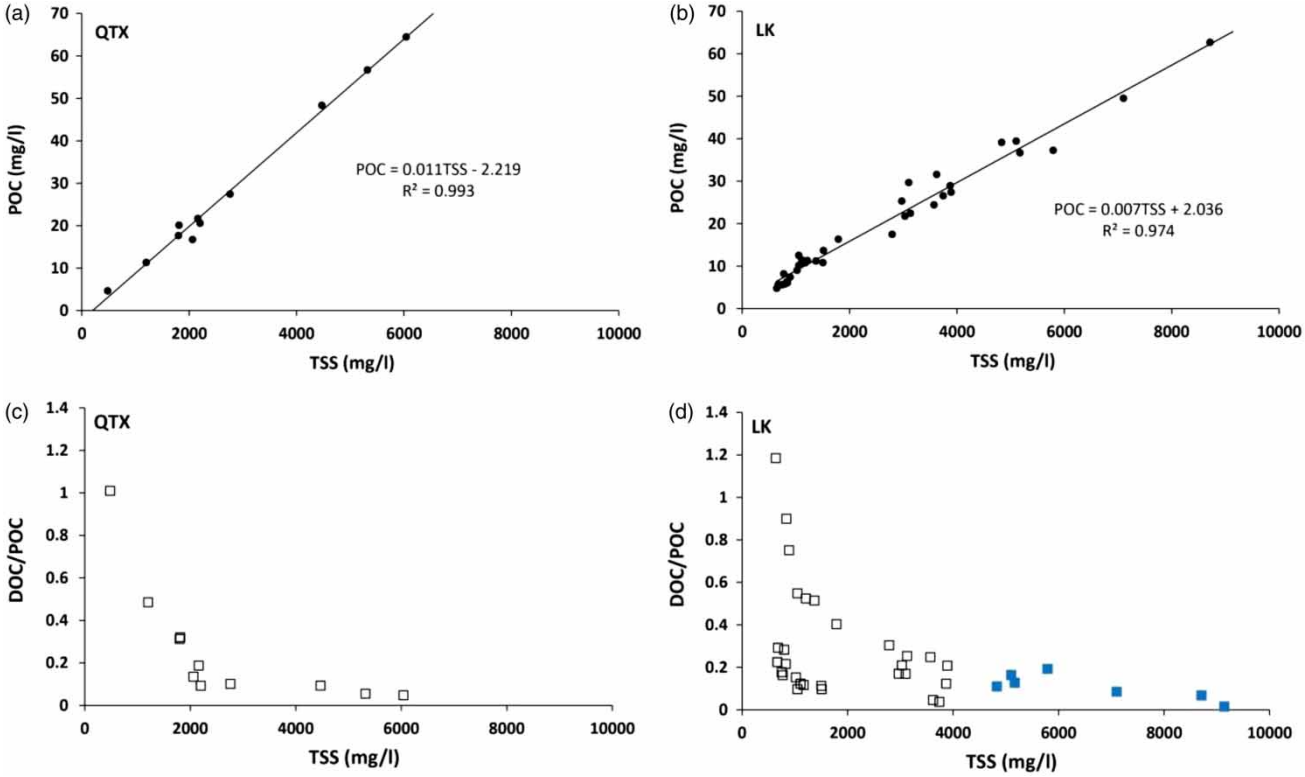

Figure 6 | Correlations between POC concentrations and TSS at QTX (a) and LK (b). TSS showed a positive contribution to POC concentrations. Ratios of DOC/POC decreased exponentially with the increasing of TSS (c) and (d).

accounted for 86.7, 88.6, and 90.4% of DTC fluxes, respectively. DIC fluxes were generally one order of magnitude higher than those of DOC. The POC flux was mainly calculated in the downstream. The annual POC flux was 668.1 thousand tons, thus the dissolved (including organic and inorganic) and particulate carbon accounted for 56.6 and 43.4% of the annual TC transport, respectively. During the WSR scheme, DIC, DTC, POC, and TC fluxes in the downstream accounted for 20.2, 20.6, 37.7, and 28.0% of the annual mean carbon fluxes, respectively. Therefore, the WSR scheme had significant influence on the carbon transport of the Yellow River.

The Lijin hydrological station is located approximately 100 km from the entrance of the Yellow River to the Bohai Sea (Figure 1). The LK station in the current study is 280 km from the Bohai Sea, which is the nearest station other than Lijin. As the river level of the downstream is higher than the surrounding ground level (suspended river), which means no tributaries enter the Yellow River below (Zeng & Zeng 2004), the carbon exported to the Bohai Sea by the Yellow River can be calculated based on the carbon fluxes at the LK station (Wang *et al.* 2013a). The annual mean water discharges at LK and Lijin stations were 22.13 and 19.17 billion (10^9) m^3, respectively (Yellow River Conservancy Commission 2002–2007). Thus, from LK to Lijin about 13% of the annual water discharge was subtracted. If the carbon concentrations were not changed from LK to Lijin, approximately 87% of the carbon fluxes at the LK station were transported to Lijin. Therefore, based on the data in Table 1, from 2005 to 2007, about 93.7×10^9 g/yr, 72.5×10^9 g/yr, and 87.4×10^9 g/yr DOC have been transported from Lijin, respectively. In addition with the about 0.69×10^{12} g/yr and 0.66×10^{12} g/yr of DIC and POC, a total of 1.34×10^{12} g/yr of carbon is exported to Lijin (Table 4).

Implications for global carbon export

Compared with the world's major rivers, the DIC concentration in the Yellow River water was among highest

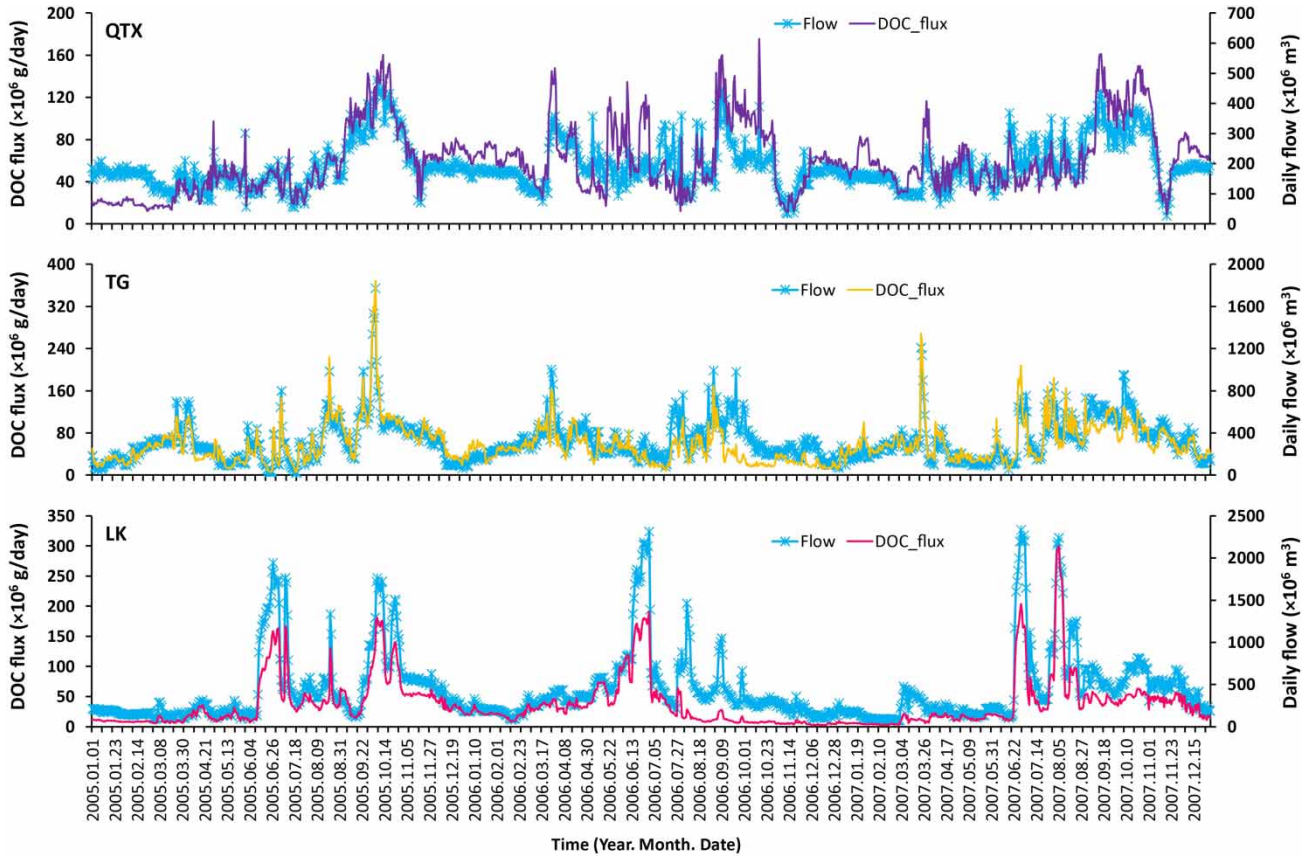

Figure 7 | Daily DOC fluxes in the mainstream of the Yellow River during 2005 to 2007. (Flow data were from Data-sharing Network of China Hydrology: http://www.hydrodata.gov.cn.)

(Table 4). Similarly high DIC concentration was found in the Mississippi River, which was also due to the extremely high weathering rate (Cai 2003). Additionally, relatively high DIC concentrations at the Ganges, Indus, and Nile rivers were probably due to the concentrating effect caused by high water evaporation (Cosa & Tremblay 1983; Kempe 1983). However, due to the low water discharge, the annual DIC exported by the Yellow River was quite small. DOC concentration (4.18 mg/l) of the Yellow River was lower than the global average level of 5.75 mg/l (Meybeck 1982), but could be comparable to that for the Amazon, Mackenzie, and Yukon rivers. Similarly, due to the low water discharge, the DOC flux in the Yellow River was also among the lowest in the world. The average POC concentration of the Yellow River was extremely high among the world's rivers. With respect to the Amazon and Ganges rivers, which also had tremendous TSS yields, the annual POC export of the Yellow River was approximately two orders of magnitude lower. Globally, the DOC/POC ratios range mostly approximately from 1–4, with a quite exceptionally high value of 10.3 for the Ob River and a low value of 0.4 for the Ganges River (Figure 9). Based on the correlation between DOC/POC and TSS (DOC/POC $= 2.83 \times e^{-0.003TSS}$), when the sediment yield exceeds around 1100 t/km^2 yr, the DOC/POC ratio begins to remain roughly stable. In such a situation, the organic carbon transport is mainly in particulate form (90%). Therefore, the extraordinary low DOC/POC ratio further contrasts the Yellow River from other large rivers and exemplifies the unique nature of the OC transport of the Yellow River (Wang *et al.* 2013b).

The Yangtze River is the largest river in China. It was estimated that annually 17.7×10^{12} g/yr carbon was exported to the East China Sea, including 14.6×10^{12} g/yr and 0.9×10^{12} g/yr as DIC and DOC, respectively (Wu *et al.* 2007). Therefore, the TC, DIC, and DOC exported

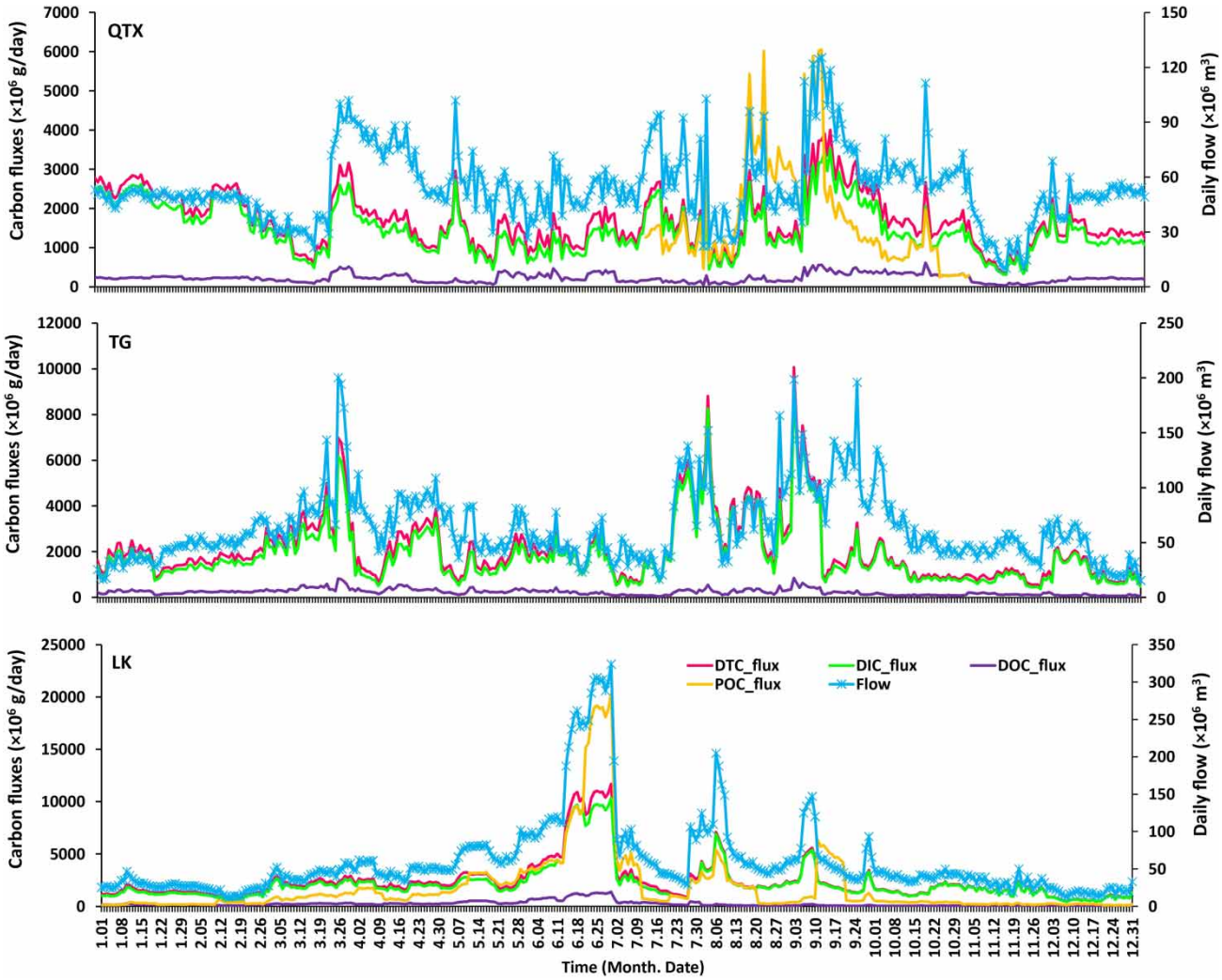

Figure 8 | Daily carbon fluxes in the mainstream of the Yellow River during 2006.

by the Yangtze River were about 13, 21, and 13 times higher than those of the Yellow River. Globally, a total of 0.9 Gt of carbon are carried every year by the world's rivers (Meybeck 1993b), among which approximately 0.34 Gt are as DIC (Wang *et al.* 2013b), 0.21 Gt are as DOC, and 0.17 Gt are as POC (Ludwig *et al.* 1996), respectively. Therefore, carbon transported by the Yellow River accounted for approximately 0.15, 0.04, and 0.34% of the global annual TC, DTC, and POC fluxes, respectively. Furthermore, considering the 69.01×10^{12} g DOC transported by the Asian rivers (Ludwig *et al.* 1996), the mean DOC exported by the Yellow River accounted for 0.12%.

CONCLUSIONS

Three stations along the mainstream of the Yellow River have been selected to monitor the variations of carbon concentrations and fluxes to the Bohai Sea under new natural hydrological and anthropogenic conditions after 2000. The main results found were that the DIC concentration of the Yellow River was among the highest of the world's major rivers, which was mainly due to the extremely high weathering rate of carbonate-rich loess and a concentrating effect of high evaporative water loss. DOC in the upstream mainly originated from natural processes, while it was principally related to the

Table 4 | Basic information, carbon concentration and fluxes for world's and China's major rivers

	Water	Sediment	Basin	Concentration (mg/l)			Flux (10^{12} g/yr)				
River	(10^9 m^3/yr)	(10^6 t/yr)	(10^6 km^2)	DIC	DOC	POC	DIC	DOC	POC	TC	Reference
Amazon	5500	1150	6.15	8–10	4.46	2.83	26	28 [1]	13.2	67.2	[1] Moreira-Turcq *et al.* (2003)
Congo	1300	43	3.70	3.1[2]	10.6[2]	5.1	3.7[2]	8.9	1.1	13.7	[2] Wang *et al.* (2013b)
Orinoco	1200	150	0.99		4.39	1.45		3.2[3]	1.9[3]		[3] Depetris & Paolini (1991)
Ganges	459	1050	1.48	20–21	3.87	9.13	7.9	1.7	18	27.6	
Mississippi	547	210	3.27	30–32	5.87	1.02–1.98	13.5	3.1	0.93	17.53	
St. Lawrence	413	3	1.03		3.75	0.75	7.02	1.55	0.31	8.88	
Ob	400	16	2.99		9.09	0.88		3.05[4]	3.69[5]		[4] Raymond *et al.* (2007)
Mackenzie	310	100	1.81		4.93	5.03	6.24	1.4[4]	1.92[4]	9.56	
Columbia	249	8	0.67		2.12	0.26	3.77[6]	0.52	0.06	4.35	[6] Cai *et al.* (2008)
Indus	240	50	0.97	28	14.4	8.82	2.1	2.67	1.64	6.41	
Yukon	210	60	0.84		4.14	1.8[7]	4	1.7	0.3	6	[7] Zou *et al.* (2006)
Niger	152	40	1.21		3.71	2.59	1.24	0.53	0.66	2.43	
Yangtze	971	480	1.94	17.76[8]	1.44[8]	0.69[8]	14.6	0.9	2.2	17.7	[8] Wu *et al.* 2007
Pearl	330	80	0.45	12–15	1.01–3.78	0.14–6.33	5.8	0.66	2.93	9.39	
Yellow	25.2	1080	0.75	31.66	4.18	22.76	0.69	0.07	0.58	1.34	This study

i. Water, sediment discharge, and drainage basin data are from Milliman & Meade (1983) and Meade (1996).
ii. DIC concentrations are from Xia & Zhang (2011) and the references therein.
iii. DOC and POC concentrations are from Ludwig *et al.* (1996) and the references therein.
iv. DIC, DOC, and POC fluxes data are from Ran *et al.* (2013) and the references therein.
[5] Lobbes *et al.* (2000).

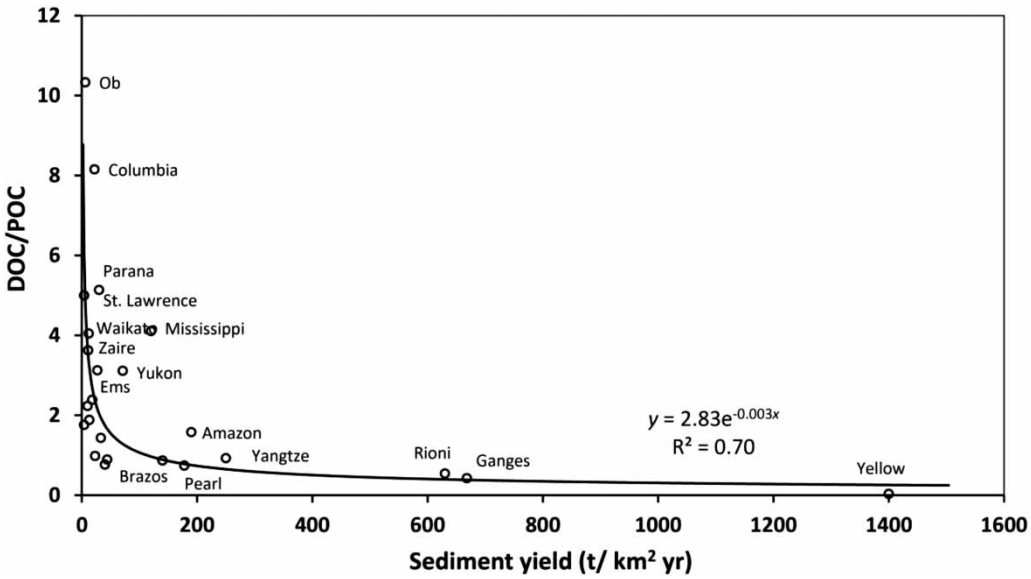

Figure 9 | Correlation between DOC/POC ratio and sediment yield in world's major rivers. In addition to rivers presented in Table 4, data were from Ludwig *et al*. (1996). Yellow River data were from the current study.

anthropogenic inputs from fertilizer application and domestic sewage in the midstream and industrial wastewater disposal in the downstream. POC was mostly connected with high TSS. Carbon exports by the Yellow River were significantly influenced by water discharge. In 2006, carbon exported to the Bohai Sea amounted to 1.34×10^{12} g/yr, among which 56.6 and 43.4% were as dissolved and particulate form, respectively. During WSR, the mean carbon fluxes accounted for approximately one-fifth to one-third of the total annual flux.

The current obtained concentrations and fluxes of carbon in the Yellow River provide essential information for the ongoing assessment of Asian and global riverine carbon cycling and budgets. The distinctive characteristic of low water discharge and high sediment load also has implications for dynamics in transport of carbon species as well as the associated biogeochemical processes in highly turbid rivers. However, due to sampling problems, continuous POC observations were only available in the downstream. Supplementary samples of suspended solids will be collected in future studies to fully determine spatial POC variations along the mainstream of the Yellow River. Furthermore, measures should be taken to control the disposal of domestic and industrial wastewater from the Yellow River to reduce the organic content transported to the Bohai Sea.

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