Biogeosciences Discuss., 9, 14365–14405, 2012
www.biogeosciences-discuss.net/9/14365/2012/
doi:10.5194/bgd-9-14365-2012

Biogeosciences Discussions

Organic carbon transport and impacts of human activities in the Yellow River

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Received: 2 October 2012 – Accepted: 3 October 2012 – Published: 17 October 2012

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Based on four field investigations during 2003 and 2009 along the Yellow River mainstream, we examined the distributions, seasonal variations and transport features of organic carbon, with a focus on the impacts of human activities (reservoir construction and regulation scheme). The results showed that organic carbon transport processes in the Yellow River were different from other major rivers. Particulate organic carbon (POC) dominated in the Yellow River and it mainly originated from the Loess Plateau. POC levels in suspended sediment (POC %) ranged between 0.25 % and 2.20 % and more than 80 % of POC concentrated in the particles with grain size smaller than 16 µm. On time scale, dissolved organic carbon (DOC) correlated negatively with discharges, indicating the influence of dilution effect. Along the river on spatial scales, DOC in the Qinghai-Tibet Plateau was closely related with temperature while DOC in the Loess Plateau showed the concentration effect, due to the abundant human input and the high ratio of evaporation to precipitation. Organic carbon in the Yellow River was very refractory and about 90 % of POC and 70 % of DOC cannot be degraded. Due to the high turbidity, the Yellow River suffers more impacts from the reservoirs in the transport of total suspended solids (TSS) and organic carbon. Ratios of DOC/POC ranged between 2.0 and 12 in the reservoirs and organic carbon was mainly in the dissolved from. POC deposited in the reservoirs of the Yellow River achieved 0.0033 Gt a^{-1}, about 8 times its annual POC flux discharged to the ocean. During the 2008 Water and Sediment Regulation (WSR) period, DOC and POC fluxes was as high as 1.1×10^{10} g and 2.2×10^{11} g respectively, accounting for 35 % of annual DOC flux and 56 % of the annual POC flux to the ocean. Discharges and material fluxes to the ocean decline sharply due to the reservoir construction while large amounts of water and sediment are transported to the ocean in such a short WSR period. These two human disturbances totally altered the processes of substance transport in the Yellow River, and may change the water chemical characteristics in the coastal zones.

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1 Introduction

Rivers are the main conduits for the transport of terrestrial materials to oceans. They provide a detailed, integrated record of natural and anthropogenic activities within the drainage basin and control the cycling of elements within rivers, as well as within estuaries and coastal zones (Meybeck, 1982; Balakrishna and Probst, 2005). World rivers transport about 0.38×10^{15} g or Gt organic carbon per year to the oceans, 0.21 Gt in dissolved form and 0.17 Gt in particulate form (Ludwig et al.,1996), accounting for 17 % of the net carbon accumulaqtion on continents or in the oceans (Aufdenkampe et al., 2011). Consequently, it is of great significance to understand the mechanisms and characteristics of riverine organic carbon transport, to advance global carbon cycle research.

There have been many researches on the organic carbon transport and transformation in large rivers of the world, mainly focusing on the rivers in the mid or low latitude, humid climate regions, such as Amazon River (Hedges et al., 1986, 1994; Moreira-Turcq et al., 2003; Townsend-Small et al., 2008; Neu et al., 2011; Cerri et al., 2012), Mississippi River (Wang et al., 2004; Dubois et al., 2010; Bianchi et al., 2004, 2007, 2011), Congo River (Coynel et al., 2005a; Spencer et al., 2012), Tana River (Bouillon et al., 2007, 2009; Tamooh et al., 2012) and Yangtze River (Wu et al., 2007; Wang et al., 2012). On the contrary, few studies have been carried out in the arid and semi-arid areas, although rivers there suffer more human disturbances (dams and water consumptions). Meanwhile, global warming will aggravate the drought degree in these areas. Therefore, studies of riverine organic carbon in these areas are helpful to reveal the influences of human activities on the riverine carbon transport, to expand the understanding of riverine organic carbon transport regularities and to balance the global carbon budget more precisely.

The Yellow River is located in the mid latitude area with arid and semi-arid climate. It has the fourth largest total suspended solids (TSS) discharge to the sea over millennial-scale historical time (Wang et al., 2006). As a result of agricultural irrigation,

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reservoir construction and the Water and Sediment Regulation (WSR) scheme, the Yellow River has become one of the most human disturbed rivers in the world and possesses very special organic carbon transport features. However, previous studies were mainly from the perspective of land-ocean interactions and focused more on estuarine areas. Zhang et al. (1992) studied the differences of organic carbon between dry and wet seasons in the lower reach and estuarine waters, and found that 17 % of particulate organic carbon (POC) and 3 % of dissolved organic carbon (DOC) were labile and easily degradable. In addition, POC in the Yellow River estuary was predominantly of terrestrial origin during the winter while it got supplement from aquatic production in the summer (Cauwet and Mackenzie, 1993). Moreover, Zhang et al. (2007) found that the phytoplankton contribution to organic carbon was significant only when TSS was smaller than 200 mg l^{-1}. While the above estuarine studies were useful, they only provided limited insight regarding the internal flux and cycle of organic carbon in the Yellow River drainage basin. To further understand the distributions, seasonal variations and transport features of the Yellow River organic carbon, four field campaigns were carried out in 2003, 2006, 2007 and 2009, respectively. Furthermore, observations were also conducted during a whole hydrological year at the Huayuankou station from November 2005 to November 2006 and an overall WSR period at the Lijin station in 2008. Reservoir construction and the WSR scheme are emphasized especially, in order to discuss the impacts from human activities on organic carbon transport.

2 Data sets and methods

2.1 Study Area

The Yellow River originates in the Yueguzonglie Basin on the Qinghai-Tibet Plateau at an elevation of 4500 m. It flows about 5464 km in distance in the main course and accumulates 75.2×10^4 km^2 of drainage area before debouching into the Bohai Sea (Fig. 1). The area upstream the Huayuankou station (Qinghai-Tibet Plateau and Loess

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Plateau) accounts for about 97 % of the whole Yellow River basin while that downstream the Huayuankou station, characterized by highly raised riverbed over the surrounding grounds, only occupies 3 % (Wang et al., 2006, 2007). Moreover, the Yellow River has different sources for water and sediment. Water comes mainly from the Qinghai-Tibet Plateau while sediment principally originates from the Loess Plateau (Zhang et al., 1990). In addition, most of the Yellow River basin is located in the arid and semi-arid areas, where annual evaporation (1712 mm) greatly exceeds precipitation (440 mm), especially in the Loess Plateau region (Yang et al., 2004). Furthermore, the organic material content in the soil is much higher in the Qinghai-Tibet Plateau (∼ 6.6 %) (Zeng et al., 2004) than that in the Loess Plateau (0.6 %) (Wen, 1989).

Affected by factors such as climate change, reservoir construction and irrigation, the water and sediment fluxes from the Yellow River to the Bohai Sea have declined significantly in recent years (Wang et al., 2006, 2007). In order to guarantee the freshwater flux discharged to the estuarine area and flush the sediment deposited in the watercourse and reservoirs away, the Yellow River Conservancy Commission (YRCC) has carried out the WSR trials during late June and early July since 2002, and these were put into operation formally in 2005. During the WSR period, more than 1/3 of the annual water and 1/2 of the annual sediment discharges were transported in less than one month (Yellow River Water Resource Bulletin, 2002–2005), changing the material transport pattern downstream completely.

The Yellow River mainstream is divided into three sections in our research, according to its geographic features. The upper stream Qinghai-Tibet Plateau reach (I) is located between the river origin and the Lanzhou station while the Loess Plateau reach (II) situates between the Lanzhou station and the Huayuankou station. The reach below the Huayuankou station, where the riverbed is generally above the surrounding areas, is considered as the Suspended River reach (III).

2.2 Data sets

Four investigations along the Yellow River mainstream were carried out in October 2003, November 2006, July 2007 and July 2009 (Fig. 1). In addition, field observations were also conducted once a week at the Huayuankou station from November 2005 to November 2006. In order to investigate the influences of the WSR scheme on the organic carbon transport of the Yellow River, the whole 2008 WSR process was observed at the Lijin station. Moreover, the Lijin station is about 100 km upstream from the estuary, and it is the last hydrological station in the lower river. In order to obtain an integrated view of the Yellow River mainstream, data in the low salinity estuarine area (LSA) ($0 < S < 2$) downward Lijin station was also taken into consideration. This included data from 2003 to 2009 collected by our team and data quoted from Zhang et al. (1992) and Cauwet and Mackenzie. (1993).

Samples for studying TSS classification were taken from the Lijin station (June, 2005), the Yellow River estuary (September, 2005), and the Lanzhou, Tongguan and Huayuankou station (November, 2006). Furthermore, samples for determine the labile organic carbon were collected during the 2006 investigation.

Natural discharge (Q_n) is characterized as the water source transferred directly from regional precipitation without human disturbance, and it is defined as actual discharge (Q_m) plus water consumption and reservoir variation (Wang et al., 2006). In this paper, natural discharges and sediment fluxes (S_{flux}) are quoted from the Yellow River Water Resources Bulletin (1998–2010) and the Yellow River Sediment Bulletin. (1998–2010). However, we could not get every Q_n and S_{flux} data for each sampling station due to limited or mismatch of hydrologic stations with the sampling stations. In that case, we applied average values of the neighboring stations instead (Fig. 1, Fig. 2).

2.3 Sampling and experiments

All the parameters were analyzed within a week after sampling. Methods for determining the parameters are described below.

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TSS: Samples were processed first by filtering through 0.45 µm pre-weighed cellulose acetate membrane filters. Then, the filters were dried at 45–50 °C and weighed on an electronic balance (AL 104, Mettler Toledo, Switzerland). The precision of the measurement was 0.0001 g.

POC, DOC: Pre-weighed and pre-combusted GF/F glass-fiber filters (0.7 µm, Whatman, Maidstone, UK) were used to separate POC and DOC in the field. Filtrates were collected in the pre-combusted bottles and poisoned with 8–10 µl saturated $HgCl_2$ for later analysis. Returning to the laboratory, filters were acidified with 2 $mol\,l^{-1}$ Hcl for 24 h to remove the inorganic carbon and then dried at 45 °C. After that, high temperature oxidation method was used to determine POC with an element analyzer (Vario ELIII CHONS, Germany). Furthermore, DOC was determined using high-temperature catalytic oxidation techniques using the Shimadzu TOC analyzer (see the TOC-$V_{CPH/CPN}$ User Manual for more details).

Chlorophyll-*a* (Chl-*a*): Chl-*a* was determined using a flurospectrophotometer (F4500, Hitachi Co. Japan) after the membrane samples had been extracted with 90 % acetone. The standard curve was constructed with SIGMA C-5753 (Sigma Co. Japan).

COD_{Mn}: The original water samples and the filtrates, after filtration using GF/F glass-fiber filters (0.7 µm, Whatman, Maidstone, UK), were processed by permanganate titration to determine Chemical Oxygen Demand (COD_{Mn}). f-COD stands for the permanganate index of the filtrates while m-COD_{Mn} represents the deviation between the permanganate index of the original samples and filtrates.

Labile organic carbon: Aerobic pollutants in the surface water are mainly the organic materials and a small amount of inorganic materials (Wang, 2010). If reductive inorganic materials can be neglected, COD_{Mn} is defined as the amount of oxygen

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consumed by labile organic materials (L_{TOC}). The processes are described as follows:

$$\mathrm{CmHnOxNaPbSc} + \left(m + \frac{n}{4} + \frac{a}{2} + \frac{5b}{4} + c - \frac{x}{2}\right)O_2 = mCO_2 + \frac{n}{2}H_2O + aNO + \frac{b}{2}P_2O_5 + cSO_2 \quad (1)$$

$$COD_{Mn} = \left(m + \frac{n}{4} + \frac{a}{2} + \frac{5b}{4} + c - \frac{x}{2}\right)W_{O_2} \quad (2)$$

$$L_{TOC} = mW_C \quad (3)$$

$$COD_{Mn} = \frac{W_{O_2}}{W_C} \times L_{TOC} + \left(\frac{n}{4} + \frac{a}{2} + \frac{5b}{4} + c - \frac{x}{2}\right) \times W_{O_2} \quad (4)$$

Where W_{O_2} and W_C denote the molar mass of O_2 and C, respectively; and m, n, x, a, b and c denote the numbers of C, H, O, N, P and S atoms in the organic carbon, respectively; $(\frac{n}{4} + \frac{a}{2} + \frac{5b}{4} + c - \frac{x}{2}) \times W_{O_2}$ indicates the difference between the oxygen consumed by the organic materials and oxygen in the organic materials themselves.

For the reason that N, P and S contents in the organic materials are much smaller than that of C (Zhang et al., 1999), and we just want to discuss the lability of organic material from the point of carbon. Therefore, $(\frac{n}{4} + \frac{a}{2} + \frac{5b}{4} + c - \frac{x}{2}) \times W_{O_2}$ can be neglected and formula (4) can be rewritten as

$$COD_{Mn} = 2.67 L_{TOC} \quad (5)$$

Labile organic carbon content can be defined as

$$\text{Percentage of Labile Organic Carbon} = \frac{L_{TOC}}{TOC} \times 100\,\% = \frac{COD_{Mn}/2.67}{TOC} \times 100\,\% \quad (6)$$

where TOC denotes total organic carbon in the samples.

TSS classification: the water elutriation system, based on Stokes' Principle (Walling and Woodward, 1993; Zhang et al., 2009) and Mastersize-2000 (Malvern Co. U.K),

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was used to classify the particles into five categories: < 8 µm; 8–16 µm; 16–32 µm; 32–63 µm; > 63 µm. The measuring range of the instrument is 0.02–2000 µm and the precision is better than 3 % on replicate analyses.

3 Results

3.1 Spatial-temporal distribution of organic carbon in the Yellow River mainstream

Water diverted from the Yellow River, mostly for agricultural irrigation use, is higher than the discharge to the ocean (Wang et al., 2006, 2007; Yellow River Water Resources Bulletin, 1998–2010). Thus, the relationship between actual discharge and material concentrations is not quite meaningful and practical. For this reason, we use natural discharges to illustrate and discuss the behaviors and mechanisms of substance transport in the Yellow River mainstream (Fig. 2).

TSS along the Yellow River mainstream (reservoirs excluded) ranged from 91 to 8400 $mg\,l^{-1}$ during our four investigations and showed the similar changing trend that TSS was very low in the Qinghai-Tibet Plateau reach (I), peaked in the Loess Plateau reach (II) and then decreased in the Suspended River reach (III). Correspondingly, POC showed the similar variation trend as TSS and ranged between 0.65 and 27.4 $mg\,l^{-1}$. In addition, POC % of the Yellow River ranged between 0.25 % and 2.2 % (averaged at 0.7 %) and the high value appeared in the reach (I). Regarding to DOC, it increased first and then decreased from the headwater to the river mouth, varying from 1.6 to 3.8 $mg\,l^{-1}$. What's more, it is worth noting that DOC in the reach (I) during the 2009 investigation was much higher. Chl-*a* concentrations changed dramatically along the watercourse, with a range between 0.46 and 122.4 $\mu g\,l^{-1}$. Ratios of DOC/POC of the Yellow River (reservoirs excluded), ranging between 0.11 and 2.4 (averaged at 0.84), were bigger than 1 in the reach (I) whereas smaller than 1 in the other two reaches (II, III).

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The Liujiaxia Reservoir (storage capacity, 5.7 km^3) in the reach (I), the Sanmenxia Reservoir (storage capacity, 9.7 km^3) and Xiaolangdi Reservoir (storage capacity, 12.7 km^3) in the reach (II) were selected to analyze what impacts the reservoirs have on the transport of organic carbon. Compared with the mainstream, TSS (3.8–175.3 $mg\,l^{-1}$) and POC (0.27–1.7 $mg\,l^{-1}$) were lower whereas DOC (2.7–5.1 $mg\,l^{-1}$) and POC % (0.94 %–24.9 %) were higher. Chl-*a* concentrations ranged between 0.66–5.9 $\mu g\,l^{-1}$ and ratios of DOC/POC were among 2 and 12.

3.2 Variations of organic carbon during a one-year observation

Water consumption and reservoir variations cannot be acquired at each sampling time due to the data limitations. However, water consumption at the Huayuankou station only accounts for 5 % of the whole basin (Yellow River Water Resources Bulletin, 1998–2010), and reservoir inventory variations almost exclusively occurred during the WSR period. Hence, it is appropriate to apply actual discharges (Q_m) to the discussion of organic carbon variations.

In winter, the Yellow River in the section I and II were almost frozen while the average temperature at the Huayuankou station was about 4 °C. Consequently, discharges at the Huayuankou station stayed at a very low level (456 $m^3\,s^{-1}$), resulting in the decrease of exogenous input, thus the decline of TSS, POC and DOC (Fig. 3). In contrast, POC % increased significantly. Concentrations of Chl-*a* ranged between 1.4 and 7.4 $\mu g\,l^{-1}$ during the winter period, much higher than those in the rest of the year. Temperature increased as ice melt at the end of winter, leading to the increase in discharge, TSS, POC and DOC. Discharge remained at the high level (1059 $m^3\,s^{-1}$) in the spring, therefore each parameter decreased because of dilution, especially DOC. The Water and Sediment Regulation (WSR) scheme of 2006 was carried out during 15 June and 3 July in the summer. As a result of outputs from the reservoirs in the middle and upper reaches, TSS and POC increased sharply at the Huayuankou station with the maximum values reaching 2576 $mg\,l^{-1}$ and 4.8 $mg\,l^{-1}$, respectively. After the WSR period, levels of TSS and POC decreased together with discharges except for occasions such as flood events (Fig. 3) and periods during which reservoirs started to release water (Fig. 3). DOC stayed at relatively high levels during the autumn season. It is clear that the natural seasonal change was relatively small and the WSR action dominated the large changes in the time series recorded in Fig. 3. However, we have to point out that while the sharp increase in discharge and TSS and the associated POC and DOC that occurred between 15 June and 3 July was caused by the WSR, this is also the wet season in the Yellow River basin, during which precipitation accounts for nearly 70 % of the annual total (Yang et al., 2004). Thus, variations of organic carbon on the time scale were dominated by both natural and human factors.

3.3 Variations of organic carbon during the Water and Sediment Regulation (WSR) period

Since 2002, the Yellow River Conservancy Commission (YRCC) has carried out the Water and Sediment Regulation (WSR) scheme, including the drainage period (Period I) and desilting period (Period II). More than 1/3 and 1/2 of the annual water and sediment load were transported in less than one month (Yellow River Water Resource Bulletin, 2002–2005), changing the material transport pattern downstream completely. In order to find out how the WSR scheme influenced the organic carbon transport in the Yellow River mainstream, the overall WSR period (19 June–3 July) of 2008 was observed at the Lijin station. Owing to the fact that water consumption at the Lijin station was small and reservoir variations almost entirely happened during the WSR period, we used the actual discharge (Q_m) instead of natural discharge in the discussion.

Before the 2008 regulation period, average values of Q_m, TSS, POC, POC % and DOC at the Lijin station were 319 $m^3 s^{-1}$, 378 $mg l^{-1}$, 4.1 $mg l^{-1}$, 1.1 % and 2.7 $mg l^{-1}$, respectively (Fig. 4). Afterwards, discharges increased sharply on 23 June and achieved the highest value (4110 $m^3 s^{-1}$) on 30 June due to large amounts of water released from the Xiaolangdi Reservoir (Period I). During this period, TSS and POC increased sharply and achieved 3194 $mg l^{-1}$ and 14.9 $mg l^{-1}$, respectively, while

DOC showed a declined trend. POC % ranged between 0.35 % and 0.65 % (averaged at 0.49 %). Large quantities of small grain size sediment was discharged from the the Xiaolangdi Reservoir as a density flow on 30 June and they reached the Lijin station on 4 July (Period II). As a result, TSS and POC increased sharply and reached 27.1×10^3 mg l^{-1} and 188.3 mg l^{-1} on 6 June, respectively. During Period II, DOC showed the same increasing trend with POC and achieved 2.94 mg l^{-1}, while POC % ranged between 0.48–0.70 % and averaged at 0.62 %.

As a whole, although TSS and POC changed significantly during the WSR period, they correlated very well with each other ($R^2 = 0.99$). TSS and POC averaged at 8.5×10^3 and 52.7 mg l^{-1}, respectively, much larger that those before the scheme. In addition, average DOC was 2.7 mg l^{-1} during the WSR period, close to the value before the scheme started.

4 Discussion

4.1 POC transport features of the Yellow River

The Qinghai-Tibet Plateau (I) supplies about 60 % of river discharge but only 10 % of the sediment load, whereas the Loess Plateau (II) contributes 40 % of the river discharge and 90 % of the river sediment load (Zhang et al., 1990). Therefore, concentrations of POC are very low for the dilution effect in the reach (I) whereas concentrations of POC are extraordinary high due to the contribution from large amounts of TSS in the reach (II) (Fig. 2). In addition, discharges and sediment loads are mainly dominated by reservoir regulation and rainfall in the Suspended River section (III), where the riverbed is generally above the surrounding areas.

Natural discharges in 2003 in the reach (II) were lower than the other three years (Fig. 2), but due to some floods happened to occur during our investigation (November 2003), discharges at the Lanzhou station were huge and the monthly discharge (November 2003) accounted for more than 20 % of the annual discharge (Yellow River

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Sediment Bulletin, 2003). Therefore, the intense scouring effect caused very high TSS and POC concentrations downstream the Lanzhou station during the 2003 investigation. Furthermore, we also observed very high TSS at the Yinchuan station, which is located in the middle of the Ningxia irrigation district in the upper Loess Plateau (Fig. 1), in both July of 2007 and 2009, but annual sediment load in this station was not that high compared to the lower Tongguan station (Fig. 2). The Ningxia irrigation area is only 2.6 % of that in the Yellow River basin (Yang et al., 2004), but its agricultural water diversion accounts for nearly 22 % of the whole basin (Yellow River Sediment Bulletin, 1998–2009). It is important to notice that about half of the diverted water returns to the river after irrigational use (Luo et al., 2010). In addition, the irrigation use of water reached a peak between May and June (Yan et al., 2007), which was just before our investigation. Therefore, temporary and unexpected high TSS and POC in this area are possible.

As the carrier of POC, TSS is the controlling factor for its temporal and spatial distribution. During our four investigations, TSS and POC were highly correlated, even during the WSR period and in the reservoirs (Fig. 5a, Eq. 7).

$$C_{\mathrm{POC}} = 0.0064 \times C_{\mathrm{TSS}} - 0.83 \quad (R^2 = 0.97, n = 181) \tag{7}$$

More specifically, when concentrations of TSS were less than 500 $\mathrm{mg\,l^{-1}}$, the relationship between TSS and POC was similar to that of other major rivers (Ludwig and Probst, 1996). In contrast, when concentrations of TSS were much greater than 500 $\mathrm{mg\,l^{-1}}$, especially during the WSR period, POC content was very low and the same as that of the loess (0.6 %) (Wen, 1989), indicating the contribution of Loess Plateau. In addition, POC/Chl-*a* can be used to represent the autochthonous contributions, the smaller this ratio, the more autochthonous contributions (Kim et al., 2000; Park et al., 2009; Tamooh et al., 2012). POC/Chl-*a* of the Yellow River mainstream averaged at 3988 (50.1–22518), similar to that of the Tana River (Bouillon et al., 2009; Tamooh et al., 2012), but much higher than that of the lower Mississippi River (256), the Pearl

River (472) (Duan and Bianchi, 2006) and the Yangtze River (48–136) (Yu et al., 2011), indicating that POC in the Yellow River mainly derived from terrestrial input. On the other hand, TSS and POC in the reservoirs decreased dramatically due to the precipitation while Chl-*a* increased for the removal of light limitation. Therefore, POC/Chl-*a* ratio declined to 964, but still much higher than that in the reservoirs on other less turbid rivers (Kim et al., 2000; Bouillon et al., 2009; Park et al., 2009).

POC % of the Yellow River ranged between 0.25 % and 1.5 %, lower than the values of the Tana River (1.1 %–4.6 %) (Bouillon et al., 2009), the Mississippi River (1.4 %–7.7 %) (Trefry et al., 1994) and most other rivers in the world (0.5 %–5.0 %) (Ludwig and Probst, 1996). Furthermore, POC % decreased logarithmically with increasing TSS in the Yellow River (Fig. 5b). Although this relationship was similar to that observed in other major rivers (Ludwig and Probst, 1996; Ittekkott, 1988), the curve of the Yellow River was below that of other major rivers in the world, indicating that the POC content of the Yellow River was lower at the same TSS level. Three reasons may be responsible for this phenomenon: (1) severe mechanical erosion in the Loess Plateau caused large amounts of mineral materials exist in the suspended solids, causing the decline of POC % (Ludwig et al.,1996); (2) the higher the TSS concentration, the bigger the median diameter, the smaller POC content (Coppola et al., 2005); (3) in the high TSS circumstances, light limitation influenced the photosynthesis of the phytoplankton, thus the autochthonous contribution decreased (Balakrishna and Probst, 2005). The curve tended to flatten out at TSS bigger than 500 $mg\,l^{-1}$ when POC % decreased to about 0.6 %, especially during the WSR period. However, in the Mississippi River and Nivelle River, values of POC % stayed stable at 1.8 % (Trefry et al., 1994) and 3 % (Coynel et al., 2005b) when TSS increased. Comparisons between these rivers suggested that the Yellow River possessed very low POC content and small organic carbon background values. On the contrary, POC % was very high in the reservoirs, even exceeded 20 %, probably due to the removal of light limitation and more autochthonous contributions. In addition, the dominance of small grain size particles of TSS in the reservoirs also contributes to the higher POC content as they are rich in organic carbon (Coppola

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et al., 2005). More specifically, POC % in the reach (I) were always very high, due to the high organic material content (Zeng et al., 2004) and low degradation efficiency in the cold and high altitude areas (Coûteaux et al., 2002; Finlay and Kendall, 2007).

The total effective POC bearing area of TSS is dependent upon its grain size (Mayer, 1994; Keil et al., 1997), so the acknowledgement of the relationship between the POC % and TSS grain size is the basis for studying the POC transport features. In our research, TSS samples were classified into five catagories (Table 1), using a water elutriation system based on Stokes' Principle (Walling and Woodward, 1993; Zhang et al., 2009).

Although the Yellow River mainstream is more than 5000 km long and different reaches have appearantly different geological backgrounds, overall POC has a consistent transport pattern. More than 80 % of POC is concentrated in particles with grain size smaller than 16 µm while over 95 % of POC was found in particles smaller than 32 µm. Thus, TSS grain size was the dominant factor controlling POC transportation in the Yellow River.

There is a negative exponential relationship between POC % and median diameter (Fig. 6). It appears that two reasons are responsible for this phenomenon: (1) the smaller the median diameter, the greater the specific surface area, and the more organic carbon loading that occurred (Coppola et al., 2005), and (2) quartz and feldspar content in the TSS increased when the median diameter increased, diluting the organic carbon content (Ludwig et al., 1996). From Fig. 6, we can determine the highest POC content along the Yellow River was 0.56 %, quite similar to the organic material content of the loess (0.60 %) (Wen, 1988).

4.2 DOC transport features of the Yellow River

DOC along the Yellow River mainstream (reservoirs excluded) averaged at 2.9 $mg\,l^{-1}$ (1.6–3.8 $mg\,l^{-1}$), much lower than that of the global rivers (> 5 $mg\,l^{-1}$) (Ludwig and Probst, 1996; Dai et al., 2012). Several possible reasons are responsible for this phenomenon: (1) extremly low precipitation (Yang et al., 2004), (2) a weak leaching effect,

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(3) low vegetation coverage (Wang et al., 2001), and (4) low content of organic materials in the loess, covering a large part of the Yellow River basin (Wen, 1989). Spatial distribution of DOC along the Yellow River corresponded very well with the geological features and climate conditions (Fig. 2). The reach (I) is the waterhead area of the Yellow River, therefore DOC concentrations in this area were at the lowest level, probably due to the dilution effect. On the contrary, DOC increased clearly in the reach (II), probably due to severe human disturbances and high ratio of evaporation to precipitation (Yang et al., 2004; Wang et al., 2007). In the reach (III), DOC changed with the same pattern as discharge did. In addition, DOC in the reservoirs ($3.3\,mg\,l^{-1}$) was much higher than that of the mainstream, probably due to more contributions from phytoplankton (Parks and Baker, 1997).

It is worth noting that DOC of the 2009 investigation in the reach (I) was much higher that that of other investigations, even reaching as high as $3.1\,mg\,l^{-1}$ (Fig. 2). Temperature might have cuased these differences. From Fig. 7, we can get that temperature increased gradually in recent years and had a oscillating period about 4 to 5 yr. Besides, the average temperature of July in 2009 (11.3 °C) was almost 2 degrees higher than that in 2007 (Fig. 7), leading to faster snow melting and thus a stronger leaching effect on the soils with high organic material content.

As it integrated the upper stream signals and thus eliminated or reduced some geographic differences, it was appropriate to use the one-year observed DOC data at the Huayuankou station to reflect the DOC seasonal variation features of the Yellow River.

Most DOC data during the Huayuankou hydrological year was negatively correlated with discharges ($R^2 = 0.3$) (Fig. 8a), different from that of other major rivers where positive correlations were observed (Finlay et al., 2006; Raymond et al., 2007). Therefore, dilution was supposed to be the dominant controlling factor for DOC on timescales. In contrast, the correlation between DOC and POC was the same as that of other world rivers (Coynel et al., 2005). However, there were some exceptional cases. First, at the end of winter and the beginning of spring, materials accumulated during winter time were flushed into the river, causing increases of DOC (circles in Fig. 8). Second,

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during the WSR and flood periods, DOC clearly increased (triangles in Fig. 8). Last, DOC increased when the reservoirs above started to release water (stars in Fig. 8).

4.3 Transport form of organic carbon

Transport form of riverine organic carbon has already attracted attention. Degens et al. (1991) pointed out that ratios of DOC to POC (DOC/POC) in global rivers ranged between 0.1 and 5 while this value reached 1.5, 3.6, 2.3, and 2.7 for the Amazon River, the Zaire River, the Orinoco River and the Yangtze River, respectively (Dagg et al., 2004). In other word, organic carbon was primarily transported in the dissolved form (Ludwig et al., 1996; Coynel et al., 2005a; Alvarez-Cobelas et al., 2012; Dai et al., 2012). However, few researches had demonstrated clearly the relationship between TSS and organic carbon transport form. To our knowledge, only two researches were carried out but both were in the estuarine areas (Abril et al., 2002; Zhang et al., 2007). As the Yellow River is one of the most turbid and human-disturbed rivers in the world, its DOC/POC ratio and the relationship between this ratio and TSS are essential to the further organic carbon research in this basin.

With the data came from various sources, including four field investigations of the mainstream, water and sediment regulation (WSR) period of 2008 and one-year observation in the Huayuankou station (HYK), we found that DOC/POC ratio correlated consistently and negatively with TSS (Fig. 9). Specifically, TSS is 366 $mg\,l^{-1}$ when the DOC/POC ratio equals 1, suggesting that when TSS exceeds 366 $mg\,l^{-1}$, POC is the main form of transported organic carbon; otherwise DOC becomes the dominant form in the Yellow River.

TSS of the Yellow River mainstream (reservoirs excluded) was extremely high (averaged at 1527 $mg\,l^{-1}$), indicating organic carbon was mainly transported in the particulate form. In contrast, TSS in the reservoirs was much lower (only 40 $mg\,l^{-1}$), so DOC became the dominant form. In addition, a one-year observation (WSR period excluded) at the Huayuankou station showed that TSS averaged at 334 $mg\,l^{-1}$ and DOC was transported mainly in the dissolved from. The Huayuankou station was only 150 km

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downstream the Xiaolangdi Reservoir. As a result, lots of TSS was deposited in the reservoir and then POC decreased sharply while DOC increased due to more contributions from phytoplankton (Parks and Baker, 1997). In addition, during the 2008 WSR period, TSS averaged at 8.5×10^3 mg l^{-1} due to river channel erosion and reservoir desilting. Undoubtedly, orangic carbon was transported in the particulate form. To sum up, POC dominates the organic carbon transport from in the Yellow River mainstream.

4.4 Lability of the Yellow River organic carbon

It is very difficult to clearly distinguish natural labile organic carbon from refractory ones. Ittekkot (1988) chose sugars and amino acids to represent the labile part in the organic carbon and concluded that percentage of labile particulate organic carbon of major rivers was 35 %. Zhang et al. (1992) also applied this method to the Yellow River estuary and concluded that percentages of labile particulate organic carbon and dissolved organic carbon were 18 % and 3 %, respectively. Although it is said that the organic carbon of the Yellow River originates from loess and possesses refractory features, so far no report has been published to explicitly support this viewpoint. For the reason that the permanganate index (COD_{Mn}) can be considered as the amount of oxygen consumed by labile organic materials, we determined the permanganate index of the original samples and filtrates during the 2006 investigation, together with the POC and DOC data, to discuss the lability of the Yellow River organic carbon.

The permanganate index of the filtrates (f-COD) correlated positively with DOC (Fig. 10a) while the deviation between the permanganate index of the original samples and filtrates (m-COD_{Mn}) correlates positively with POC (Fig. 10b). In addition, with the method described in Sect. 2.3, we concluded the percentages of labile POC and DOC were 8.3 % and 31.6 %, respectively, indicating that 90 % of POC and 70 % of DOC in the Yellow River cannot be degraded and transported to the sea. However, these two values in the reservoirs were much higher (18.9 % and 41.7 %, respectively), due to more autochthonous contributions.

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Aerobic pollutants in the surface water are mainly the organic materials and a small amount of inorganic materials (Wang, 2010). However, organic carbon of the Yellow River mainly derived from the loess, which is highly degraded and almost harmless to the environment (Diao and Wen, 1988; Chen et al., 2004). Although this method still cannot accurately quantify the labile part of the organic carbon, it is meaningful to point out the refractory features of organic carbon in the Yellow River and assess the impact of the organic materials to the environment.

4.5 Impacts of human activities on the organic carbon transport in the Yellow River

4.5.1 Reservoirs

Reservoir construction is considered to be one of the most disruptive human activities for rivers. Water and sediment impounded in reservoirs lead to the declined water and sediment fluxes to the ocean (Wang et al., 2006, 2007) and form a very special environment (Kim et al., 2000; Bouillon et al., 2009; Williamson et al., 2009).

More than 3000 reservoirs scattered over the Yellow River and the total storage capacities exceeded $57.4 \times 10^9\,m^3$ (Zhang et al., 2001; Wang et al., 2006), about four times the annual water flux ($14.6\,km^3\,a^{-1}$) transported by the Yellow River to the Bohai Sea (Yellow River Sediment Bulletin, 2008). In addition, the Yellow River has extremely high turbidity, and therefore suffers more impacts from the reservoirs in the transport of TSS and organic carbon. Taking the Xiaolangdi Reservoir (storage capacity: $12.7\,km^3$) as an example, its average level of trapped TSS was $0.24\,Gt\,a^{-1}$ between 1997 and 2003, about 1.9 times that of the Three Gorges Dam (storage capacity: $39.3\,km^3$) during 2003 and 2005 (Dai et al., 2009).

The Loess Plateau contributes about 90 % of sediment load to the Yellow River (Zhang et al., 1990), so the material sedimentation in the reservoirs mainly exists in this reach. Storage abilities of the reservoirs in the Loess Plateau amounted to $29.3\,km^3$ (Wang et al., 2006) and TSS trapped in this reach achieved $0.55\,Gt\,a^{-1}$, concluded

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from the case of the Xiaolangdi Reservoir (Dai et al., 2009), further higher than the TSS annual flux to the ocean (0.056 Gt a^{-1}) (Wang et al., 2012; Yellow River Sediment Bulletin, 2009). Based on the fact that the POC content of the loess was 0.6 % (Wen, 1989), we concluded the deposited POC in the reservoirs in the Loess Plateau achieved 0.0033 Gt a^{-1}, about 8 times the annual POC flux of the Yellow River to the ocean (Wang et al., 2012). On the contrary, reservoirs on the Verde River only deposited about 78 % of TSS (Baker et al., 1994) and 72 % of POC (Parks and Baker, 1997). This comparision further suggests that turbid rivers suffers more impacts from the reservoirs in the transport of materials.

4.5.2 Water and Sediment Regulation (WSR)

The imbalance transport of water and sediment in the Yellow River leads to serious siltation downstream and the same situation exists in the reservoirs, especially in the reach (II). Meanwhile, suffered from natural and human impacts, the water and sediment fluxes from the Yellow River to the Bohai Sea have declined significantly in recent years (Wang et al., 2006, 2007), severely affecting the eco-systems in the estuarine area. In order to guarantee the freshwater flux to the sea and flush the sediment deposited in the watercourse and reservoirs away, the Yellow River Conservancy Commission (YRCC) has carried out the Water and Sediment Regulation (WSR) scheme during late June and early July every year since 2002.

TSS and POC increased sharply at the Lijin station during period I (Fig. 4), owing to large amounts of water released from the Xiaolangdi Reservoir. However, the increased discharge did not lead to the decline of DOC. Two reasons may be accounting for this phenomenon, (1) the removal of light limitation and then more autochthonous contributions in the reservoirs (Parks and Baker, 1997); (2) DOC released from the re-suspended materials (Gao et al., 2002). At the end of period I, although actual discharge stayed at a nearly constant level around 4000 $m^3 s^{-1}$ (Fig. 11a), concentrations of TSS (POC) went through a sharp decline, indicating that the sediment in the watercourse had already been flushed away. However, concentrations of DOC showed an

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increasing trend (Fig. 4), probably due to the fact that DOC in the watercourse almost originated from the reservoirs. Furthermore, actual discharge and TSS appeared as a clockwise hysteresis during period I, suggesting that TSS mainly originated from the large grain size sediment in the watercourse (Fig. 11a), and this was also the reason that values of POC % during this period were smaller than those before the scheme (Ludwig et al., 1996; Coppola et al., 2005).

Then the regulation scheme went into period II, the desilting period. During this period, the upstream Sanmengxia and Wanjiazhai Reservoirs released large amounts of clear water to flush out the sediment in the Xiaolangdi Reservoir, for the purpose of increasing the reservoir storage capacity. As a result, concentrations of TSS and POC increased sharply and average POC % reached 0.62 %, larger than that in the period I (0.49 %), due to large amounts of small grain size particles flushed out as a density flow from the Xiaolangdi Reservoir. Concentrations of DOC in period II were larger than those in period I and correlated very well with POC ($R^2 = 0.69$), indicating that a transformation from POC to DOC happened during period II (Gao et al., 2002). During this period, actual discharge and TSS at the Lijin station appeared as a counter-clockwise hysteresis (Fig. 11b), suggesting that TSS mainly originated from the Xiaolangdi Reservoir rather than from the watercourse. The research on the Rhine River showed similar results that counter-clockwise hysteresis occurred during a low discharge period with most of the sediment originating from upstream tributaries, while a clockwise hysteresis seemed to be related to erosion or early sediment supply just upstream of the measurement location (Asselman, 1999).

Ratios of DOC/POC during the WSR period ranged between 0.02–0.49 and averaged at 0.16, indicating that organic carbon transported mainly in the particulate form. According to the data of DOC and POC and discharges, we concluded that DOC and POC fluxes during the 2008 WSR period achieved 1.1×10^{10} g and 2.2×10^{11} g, respectively, accounting for 35 % of annual DOC flux (3.2×10^{10} g a^{-1}) and 56 % of the annual POC fluxes (3.9×10^{11} g a^{-1}) (Wang et al., 2012). Such a large amount of materials transported in this short time leaded to the deposition and burial of organic carbon

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in the estuarine area, changing the water chemical characteristics and influencing the eco-system balance in the coastal zones.

Discharges and material fluxes to the ocean decline sharply due to the reservoir construction while large amounts of water and sediment are transported to the ocean in such a short WSR period. These two human disturbances totally change the laws of substance transport in the Yellow River. What is more, sediment export flux of the Xiaolangdi Reservoir during the whole 2008 WSR period achieved 4.8×10^{13} g (Wu, 2010), only 20 % of the annual sedimentation of the Xiaolangdi Reservoir (Dai et al., 2009), so how to take the deposition in the reservoir away and increase its storage capacity is the urgent problem to solve.

Most of the Yellow River is located in the arid and semi-arid regions in the middle latitudes, where agricultural activities are intense and human disturbances are severe. Hence, riverine organic carbon transport is mainly dominated by the human activities. Milliman et al., (2008) found that lots of rivers in the world suffered declined discharges due to damming, irrigation and interbasin water transfers and most of these rivers drain arid and semi-arid regions in Africa, Asia and Australia, 18 of which (including the Indus and the Yellow River) experienced more than 50 % decline. Therefore, researches in this region are essential in revealing the human impacts on the riverine organic carbon transport, enriching the knowledge of riverine organic carbon transport laws and accurately balancing the global carbon budget.

5 Conclusions

1. Organic carbon transported in the Yellow River was mainly in the particulate form while dissolved organic carbon dominated in other major rivers. Particulate organic carbon in the Yellow River mainly originated from the Loess Plateau. Values of POC % ranged between 0.25 % and 2.2 % and more than 80 % POC concentrated in the particles with grain size smaller than 16 µm.

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2. DOC in the Yellow River was much lower than that of the world rivers. On timescales, a weak and nagetive relationship exiested between DOC and discharges, indicating the influence of dilution effect. At spatial scales, DOC in the Qinghai-Tibet Plateau was closely related with temperature while DOC in the Loess Plateau showed the concentrations effect, due to the abundant human input and the high ratio of evaporation to precipitation. Ratios of DOC/POC were bigger than 1 in the Qinghai-Tibet Plateau (I) while smaller than 1 in the Loess Plateau (section II) and the Suspended River (III). About 90 % of POC and 70 % of DOC in the Yellow River cannot be degraded and transported to the sea. These two values in the reservoirs were much higher due to more autochthonous contributions.

3. The Yellow River has extremely high turbidity, and therefore suffers more impacts from the reservoirs in the transport of TSS and organic carbon. POC in the reservoirs was much lower than that of the mainstream while DOC showed an opposite side. Ratios of DOC/POC ranged between 2.0 and 12 in the reservoirs and DOC is the dominant form of organic carbon. POC deposited in the reservoirs in the Loess Plateau achieved 0.0033 Gt a^{-1}, about 8 times the annual POC flux of the Yellow River to the ocean.

4. The Water and Sediment Regulation (WSR) scheme is considered as the most human-disturbed action on the Yellow River material transportation. During the 2008 regulation period, DOC changed little but POC increased dramatically. Therefore, ratios of DOC/POC only ranged between 0.02 and 0.49, organic carbon was transported dominantly in the particulate from. DOC and POC fluxes during the 2008 WSR period achieved 1.1×10^{10} g and 2.2×10^{11} g, respectively, accounting for 35 % of annual DOC flux and 56 % of the annual POC flux to the ocean.

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Acknowledgements. This work was financially supported by the Foundation for Innovative Research Groups of National Science Foundation of China (NSFC): Marine Organic Biogeochemistry (Grant No. 41221004) and Public Benefit Special Fond of State Oceanic Administration (Grant No. 201005009). We are thankful to Ming Xue, Zhiyuan Liu, Baosen Wang, Yan Cui and Chunchao Xiao for meaningful suggestion.

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Table 1. The median diameters ($d_{0.5}$) and POC % of each grain size and the original samples.

		$\Phi_{<8\,\mu m}$	$\Phi_{8\text{–}16\,\mu m}$	$\Phi_{16\text{–}32\,\mu m}$	$\Phi_{32\text{–}63\,\mu m}$	$\Phi_{>63\,\mu m}$	original samples
Lanzhou	$d_{0.5}$(µm)	3.9	8.3	21.9	42.3	71.1	7.2
	POC %	0.51 %	0.43 %	0.28 %	0.14 %	0.088 %	0.43 %
Tongguan	$d_{0.5}$(µm)	3.9	10	26.8	51.7	89.2	17.4
	POC %	0.48 %	0.41 %	0.23 %	0.14 %	0.090 %	0.38 %
Huayuankou	$d_{0.5}$(µm)	3.6	9.2	23.7	48.8	78.7	10.6
	POC %	0.59 %	0.52 %	0.3 %	0.13 %	0.12 %	0.44 %
Lijin	$d_{0.5}$(µm)	3.6	9.2	23.7	50.7	71.5	18.6
	POC %	0.48 %	0.40 %	0.18 %	0.066 %	0.037 %	0.33 %
the Yellow River delta*	$d_{0.5}$(µm)	4.3	9.0	14.3			6.9
	POC %	0.63 %	0.56 %	0.48 %			0.57 %

* Only three categories can be achieved in the Yellow River estuary due to small grain size.

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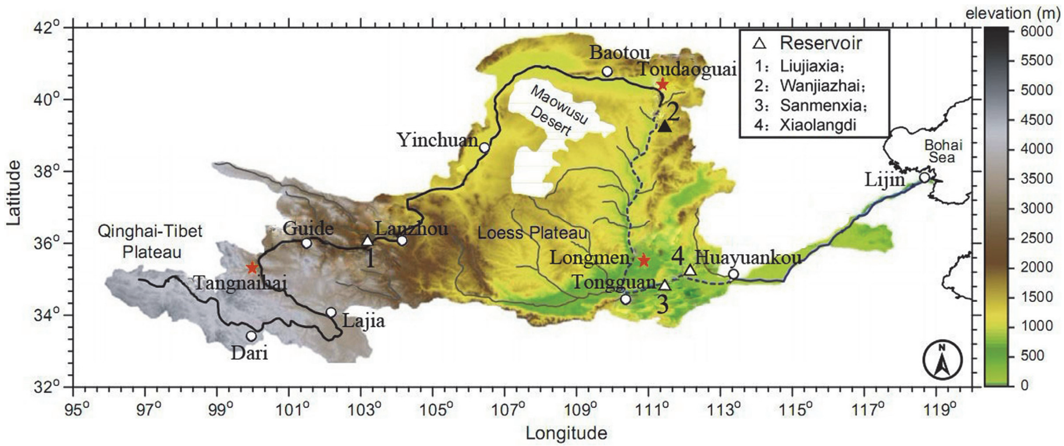

Fig. 1. The Yellow River basin and sample locations (open symbols).

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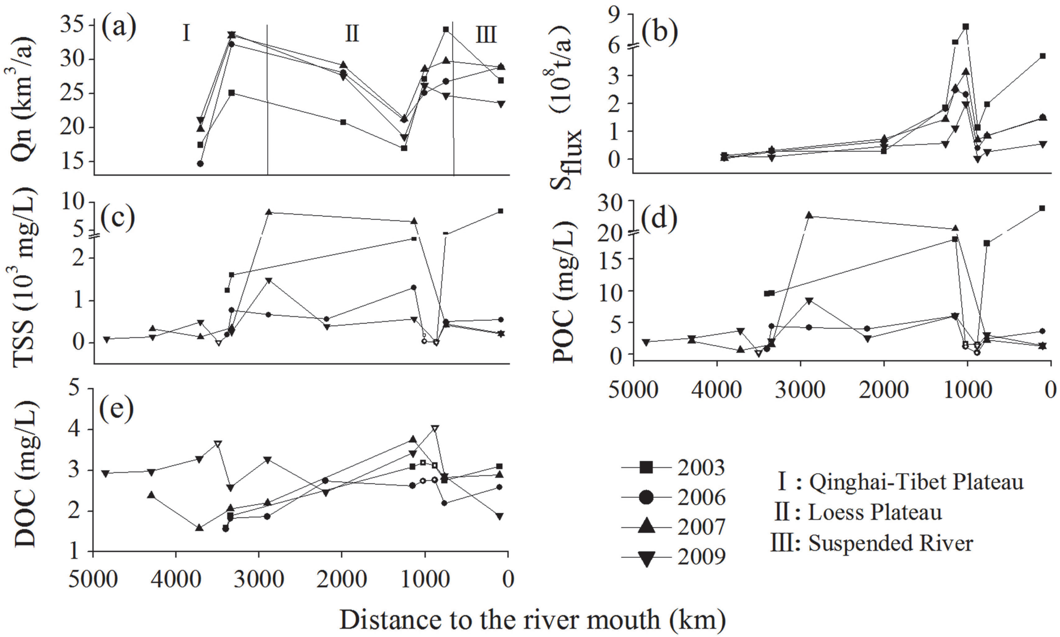

Fig. 2. Spatial and temporal variations of natural discharge (Q_n) **(a)**, sediment flux (S_{flux}) **(b)**, TSS **(c)**, POC **(d)** and DOC **(e)** along the mainstream of the Yellow River (open circles represent the reservoirs of our investigation).

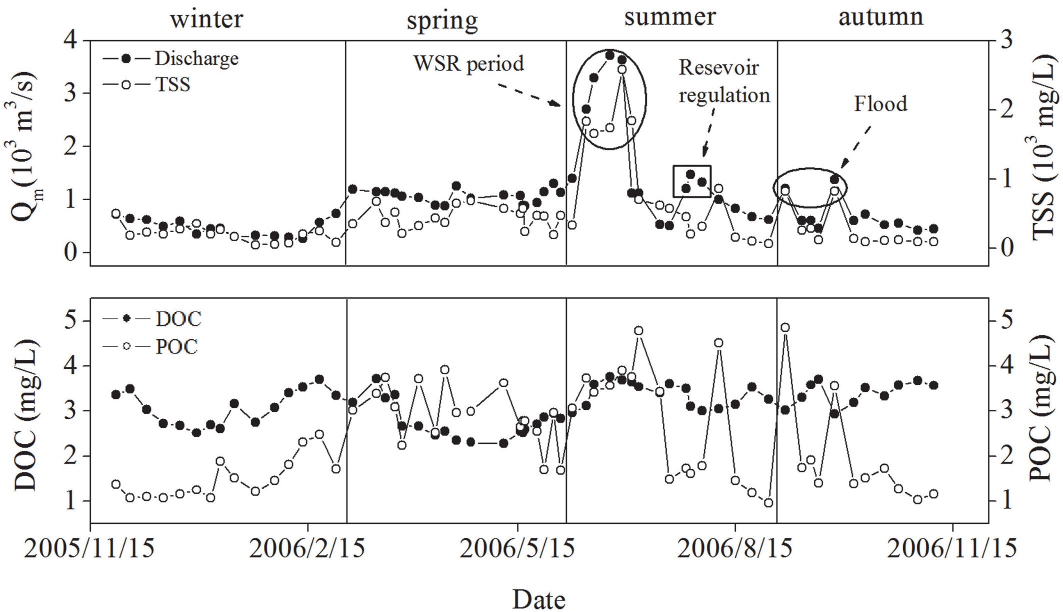

Fig. 3. Distributions of discharge (Q_m), TSS, POC and DOC at the Huayuankou station between November 2005 and November 2006.

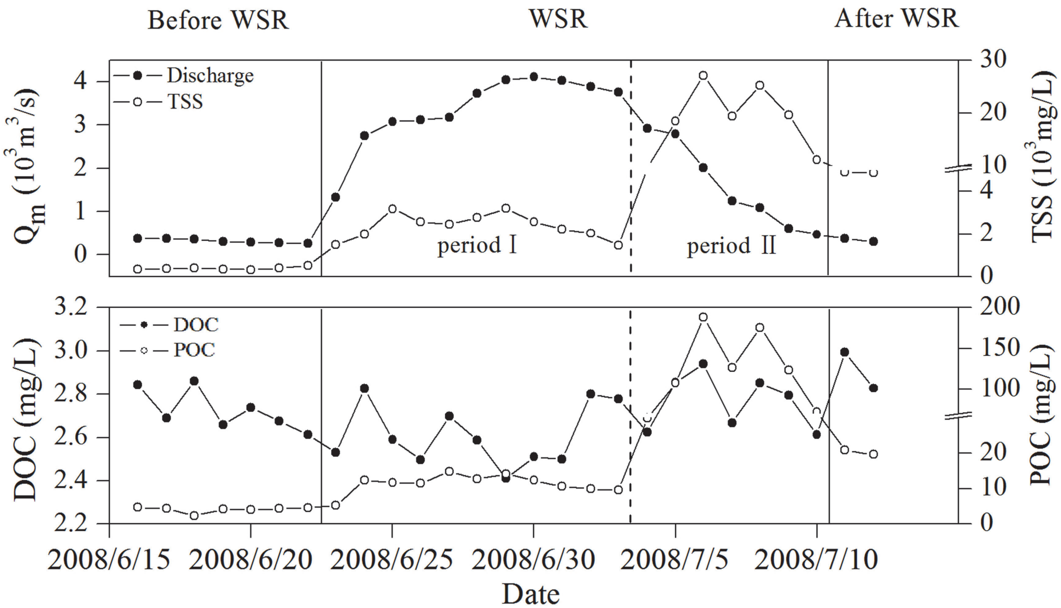

Fig. 4. Distributions of actual discharge, TSS, POC and DOC during the 2008 water and sediment regulation (WSR) period at the Lijin station.

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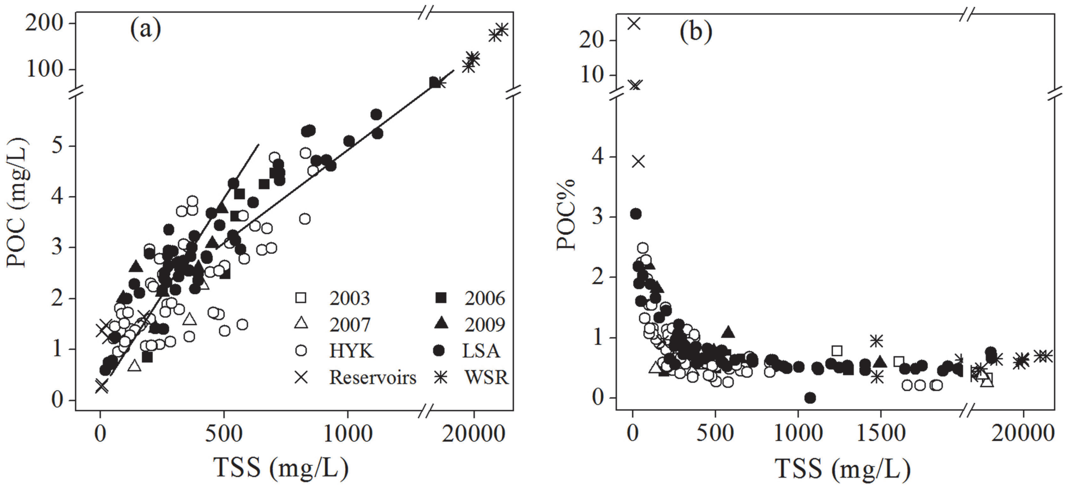

Fig. 5. Relationship between POC and TSS **(a)**, POC % and TSS **(b)** of samples in the Yellow River.

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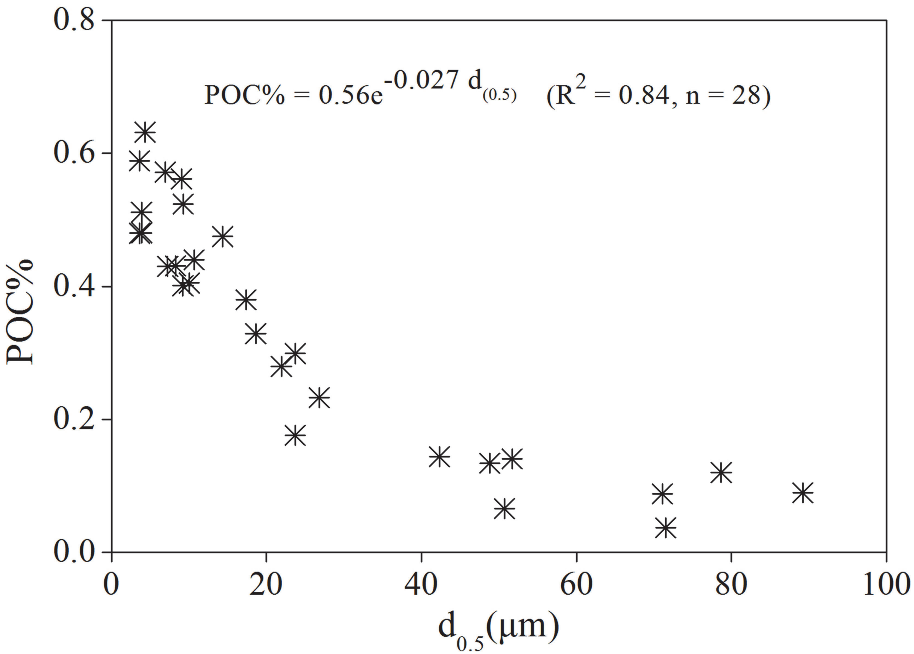

Fig. 6. Relationship between POC % and median particle diameter ($d_{0.5}$).

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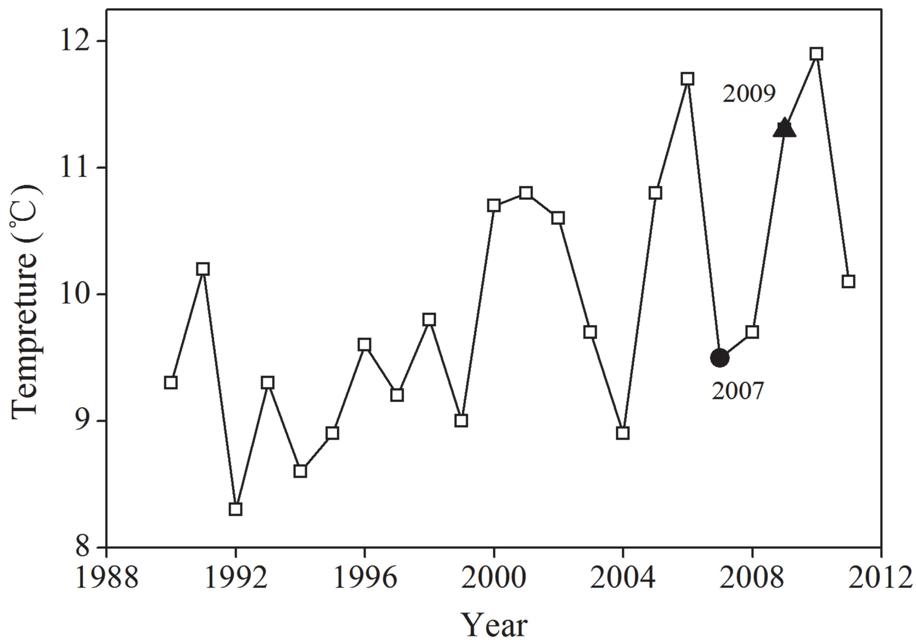

Fig. 7. Temperature Variations of July at the Dari station in the Qinghai-Tibet Plateau (data quoted from http://cdc.cma.gov.cn).

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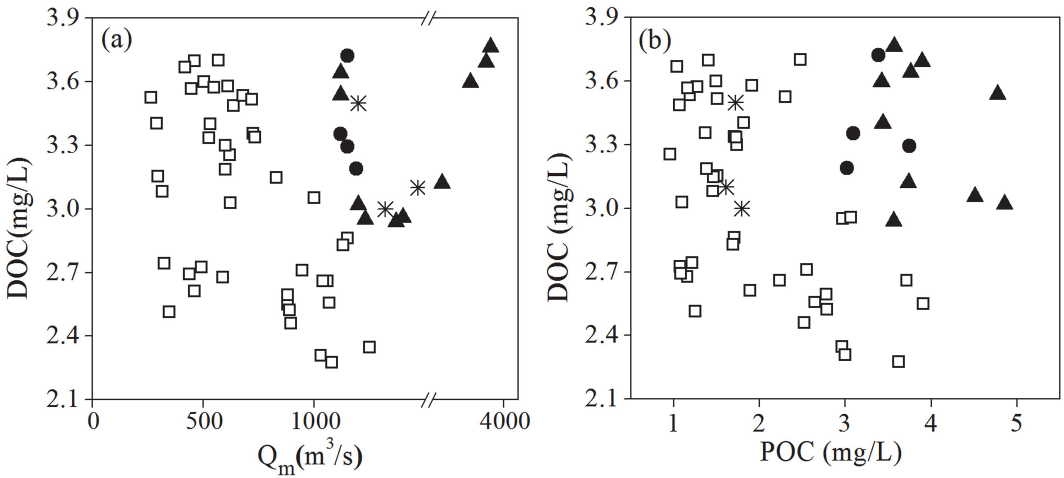

Fig. 8. Relationship between DOC and actual discharge (Q_m) **(a)**, DOC and POC **(b)** during the whole hydrological year at the Huayuankou station (circles represent the period of the end of winter and the beginning of spring, triangles represent the water and sediment regulation (WSR) and flood periods, stars represent periods during which reservoirs start to release water).

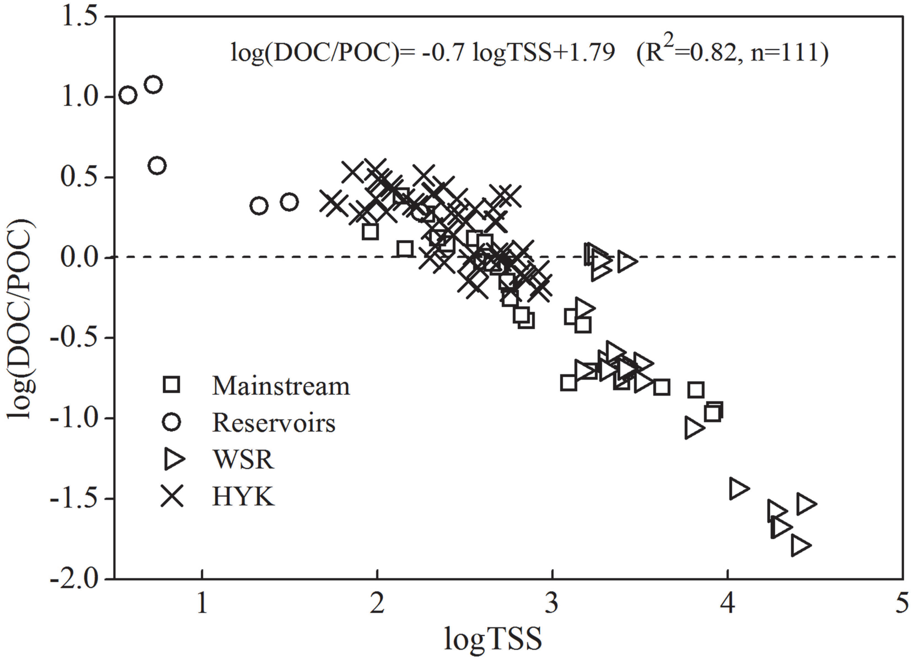

Fig. 9. Relationship between log(DOC/POC) and logTSS in the Yellow River mainstream.

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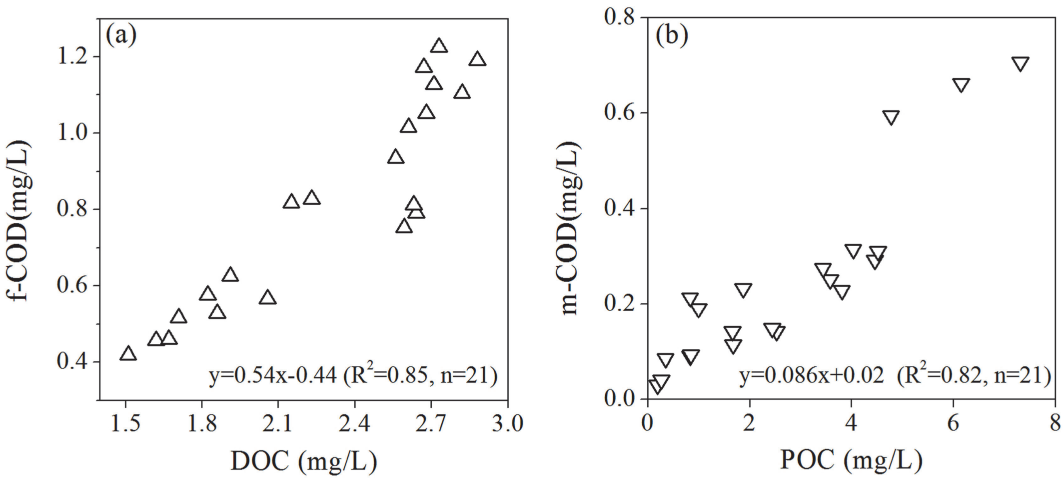

Fig. 10. Relationship between f-COD_{Mn} and DOC **(a)**, m-COD_{Mn} and POC **(b)** of the samples during the 2006 investigation along the Yellow River mainstream.

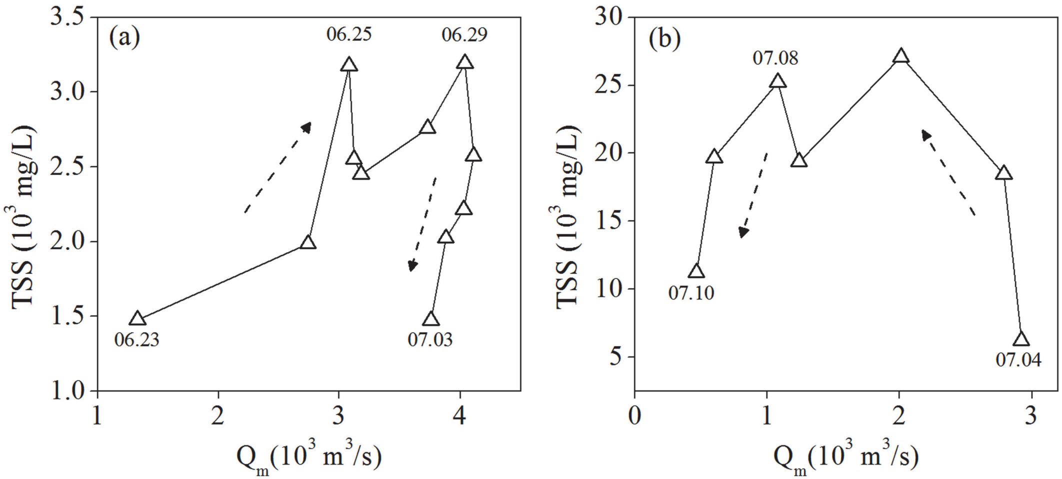

Fig. 11. Relationship between actual discharge (Q_m) and TSS in period I **(a)** and period II **(b)** during the water and sediment regulation (WSR) period in 2008, Lijin station.

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