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Organic carbon fluxes from the upper Yangtze basin: an example of the Longchuanjiang River, China

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Abstract:

To investigate the effects of anthropogenic activity, namely, land use change and reservoir construction, on particulate organic carbon (POC) transport, we collected monthly water samples during September 2007 to August 2009 from the Longchuanjiang River to understand seasonal variations in the concentrations of organic carbon species and their sources and the yield of organic and inorganic carbon from the catchment in the Upper Yangtze basin. The contents of riverine POC, total organic carbon and total suspended sediment (TSS) changed synchronously with water discharge, whereas the contents of dissolved organic carbon had a small variation. The POC concentration in the suspended sediment decreased non-linearly with increasing TSS concentration. Higher molar C/N ratio of particulate organic matter (average 77) revealed that POC was dominated by terrestrially derived organic matter in the high flows and urban wastewaters in the low flows. The TSS transported by this river was 2.7×10^5 t/yr in 2008. The specific fluxes of total organic carbon and dissolved inorganic carbon (DIC) were 5.6 and 6 t/km^2/yr, respectively, with more than 90% in the high flow period. A high carbon yield in the catchment of the upper Yangtze was due to human-induced land use alterations and urban wastes. Consistent with most rivers in the monsoon climate regions, the dissolved organic carbon–POC ratio of the export flux was low (0.41). Twenty-two percent (0.9 t/km^2/yr) of POC out of 4 t/km^2/yr was from autochthonous production and 78% (3.1 t/km^2/yr) from allochthonous production. The annual sediment load and hence the organic carbon flux have been affected by environmental alterations of physical, chemical and hydrological conditions in the past 50 years, demonstrating the impacts of human disturbances on the global and local carbon cycling. Finally, we addressed that organic carbon flux should be reassessed using adequate samples (i.e. at least two times in low-flow month, four times in high-flow month and one time per day during the flood period), daily water discharge and sediment loads and appropriate estimate method. Copyright © 2011 John Wiley & Sons, Ltd.

KEY WORDS dissolved organic carbon; particulate organic carbon; C/N ratio; water discharge; sediment loads; Longchuanjiang; Yangtze River

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INTRODUCTION

Carbon fluxes from worldwide rivers to the oceans were estimated approximately 1 Gt/yr, with approximately 45% being transported as organic carbon (Schlesinger and Melack, 1981; Ludwig *et al*., 1996). This riverine transportation of organic carbon, composed of dissolved organic carbon (DOC) and particulate organic carbon (POC), from terrestrial ecosystem to marine systems, represents a significant process in the global carbon cycling (Meybeck, 1982; Hedges, 1992; Ludwig *et al*., 1996). The DOC/POC ratio of the carbon flux was approximately 1.2 at a global scale. The total organic carbon (TOC) is closely related to stream water quality or considered as an indicator of organic contamination. Previous studies reported the positive correlations between certain organic pollutants and TOC (Parks and Baker, 1997; Hinga, 2003; Chen *et al*., 2006). Therefore, investigation on the geochemical characteristics and transport process of riverine organic carbon can help us to interpret global carbon budget and assess organic pollution.

Organic carbon is strongly associated with catchment physical characteristics such as soil type, climate, hydrology and land use (Meybeck, 1993b; Hope *et al*., 1994). POC in rivers is the result of terrestrially (i.e. soil erosion, allochthonous source) and aquatically (i.e. phytoplankton, autochthonous source) produced organic matter. POC is mainly terrestrially produced in high turbid rivers such as the Yangtze and the Yellow Rivers (Zhang *et al*., 1992; Cauwet and Mackenzie, 1993; Wu *et al*., 2007), whereas the contribution of aquatic biomass to POC increases evidently in low turbid rivers (Meybeck, 1993a; Kao and Liu, 1996; Hope *et al*., 1997; Gao *et al*., 2007). However, riverine DOC is mostly terrestrial in origin (Raymond and Bauer, 2001; Helie and Hillaire-Marcel, 2006). The C/N ratios have been widely used to decipher autochthonous and allochthonous sources of carbon (Meybeck, 1982). The C/N ratios of terrestrial organic matter have a wide range (~12–400), whereas the C/N ratios of phytoplankton are less variable (~6–8) (Hedges *et al*., 1986, 1997). Anthropogenic activities such as deforestation and dams greatly regulate the transports, concentrations and temporal variations of riverine organic carbon (Syvitski et al., 2005;

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Balakrishna *et al.*, 2006). The fluxes of organic carbon increased in response to deforestation and road construction (Moore and Jackson, 1989; Kao and Liu, 1996) and decreased in response to dam/reservoir construction (Parks and Baker, 1997; Wu *et al.*, 2007; Zhang *et al.*, 2009).

Numerous studies have been conducted on organic carbon fluxes at global and local scales (Meybeck, 1982, 1993a,b; Ludwig *et al.*, 1996; Gao *et al.*, 2002; Balakrishna and Probst, 2005; Balakrishna *et al.*, 2006; Ni *et al.*, 2008; Agren *et al.*, 2008), especially for small rivers (Helie and Hillaire-Marcel, 2006; Agren *et al.*, 2008). However, total amounts and components of riverine organic carbon are closely related to diverse biogeochemical and physical processes within the drainage area, thus the complex nature of riverine systems due to variations in climate, land use, soil composition, hydrology and human disturbances requires reliable data on the concentrations and their controlling factors at various seasons of hydrology. As a consequence, most reports on waterborne matters are still in uncertainty. For instance, organic carbon fluxes were underestimated because of lack of data during rainy events particularly in monsoon-dominated rivers (Meybeck, 1993a,b; Hope *et al.*, 1994). The associations between organic carbon and water discharge as well as daily water flows are required to yield reliable results.

Moreover, because of largely seasonal differences in terrestrial organic carbon outputs caused by monsoon climate, the riverine organic carbon transport study is relatively rare in most monsoon-dominated drainage systems compared with many advances achieved in non-monsoon drainage systems (cf. Gao *et al.*, 2002). Despite some studies on the nature and quantity of carbon transport in the Yangtze River basin, organic carbon fluxes could be underestimated because of the limited sampling (i.e. Zhang *et al.*, 1992; Cauwet and Mackenzie, 1993; Wu *et al.*, 2007).

Previous reports on the Longchuanjiang River located in the upper Yangtze River indicated impacts of anthropogenic activities on sediment, nutrients and water geochemistry (Lu, 2005; Zhu *et al.*, 2007, 2008; Li *et al.*, 2011; Lu *et al.*, 2011). The aims of the present work were (1) to identify seasonal variations in organic carbon concentrations and C/N ratios, (2) to quantify the sources of organic carbon and (3) to determine riverine organic carbon transport change over the past decades. This study could update information on geochemical characteristics and transports of organic carbon in the headwater of the Yangtze River under human activities. Further, there has been an increasing forested land through reforestation for reducing greenhouse gas emissions and therefore climate change mitigation worldwide (Post and Kwon, 2000). How does soil organic matter accumulation, for example, 33 to 34 g/m^2/yr reported by Post and Kwon (2000) through reforestation, influence the riverine organic matter? Thus, the present work also provides a basis for evaluating the riverine organic carbon change under the programme of conversion from farmland to plantation, secondary forests and grasslands in the upper Yangtze River, China.

MATERIALS AND METHODS

Study site

The Longchuanjiang River drains an area of 5560 km^2 (24°45′N–26°15′N and 100°56′E–102°02′E) before joining into the Jinshajiang, a tributary of the upper Yangtze River (Figure 1). The mainstream length and its total elevation fall are 231 km and 2300 m (from 700 to 3000 m a.s.l.), respectively. The upper catchment (upper the Xiaohekou station) has 1788 km^2 with a subtropical monsoon climate, characterised by annual mean temperature of 15.6 °C. The average annual rainfall amount is 825 mm with 86% to 94% of the total rainfall occurring in the rainy season from May to October. The mean annual runoff and the mean annual sediment load at the Xiaohekou Station were 3.2×10^8 m^3 and 4×10^5 t with pronounced seasonal variations (Lu, 2005). The area is dominated by purple soil under the Chinese soil classification (Lu, 2005), namely, inceptisols according to US Department of Agriculture soil Taxonomy (1999), which is very susceptible to water erosion and weathering. Erosion was accelerated by growing populations and economic growth, which have contributed to deforestation (in earlier times), intensified agriculture activity, reservoir building, stone excavation and road construction. There are some small-sized reservoirs situated in the drainage basin, which may result in reduction of sediment and organic matter export at Xiaohekou hydrological station. Also, there are several counties (Nanhua and Chuxiong) along the riverine network, where industrial and domestic wastes discharge directly to the river. Chuxiong County, adjacent to the sampling location, has large impacts on water chemistry.

Sampling and analysis

Daily water discharge and sediment concentration from January 2007 to March 2009 were recorded by the staff of the Xiaohekou gauging station located in the Chuxiong City (Figure 1). Water samples for chemical analysis were collected once a month over a 24-month period (September 2007 to August 2009) using the same mechanised pulley system used by the station workers to determine the discharge rating curve. A total of 24 depth-integrated samples were collected and stored in acid-washed 5-L high-density polyethylene containers, in the outlet of the river basin at Xiaohekou station.

All the samples were filtered by vacuum filtration on the sampling day. The samples for total suspended sediment (TSS) were filtered in pre-cleaned 0.45-μm pore size, 47-mm-diameter pre-weighed Nuclepore filters. The filters were dried at 60°C for 24 h and weighed for quantifying TSS. Samples for POC and C/N ratio were filtered through 0.7-μm pore size Whatman GF/F filters, which were pre-combusted at 450°C for 6 h and pre-weighed. The filter paper was kept in a plastic bag for future POC and C/N ratio analyses after being dried at 50°C for 24 h. Filtrates were acidified to $\mathrm{pH} < 2$ with high purity HNO_3 and preserved in high-density polyethylene bottles (marinate for 24 h in 1:10 HNO_3 acid solution beforehand) in the refrigerator for DOC analysis later in the laboratory.

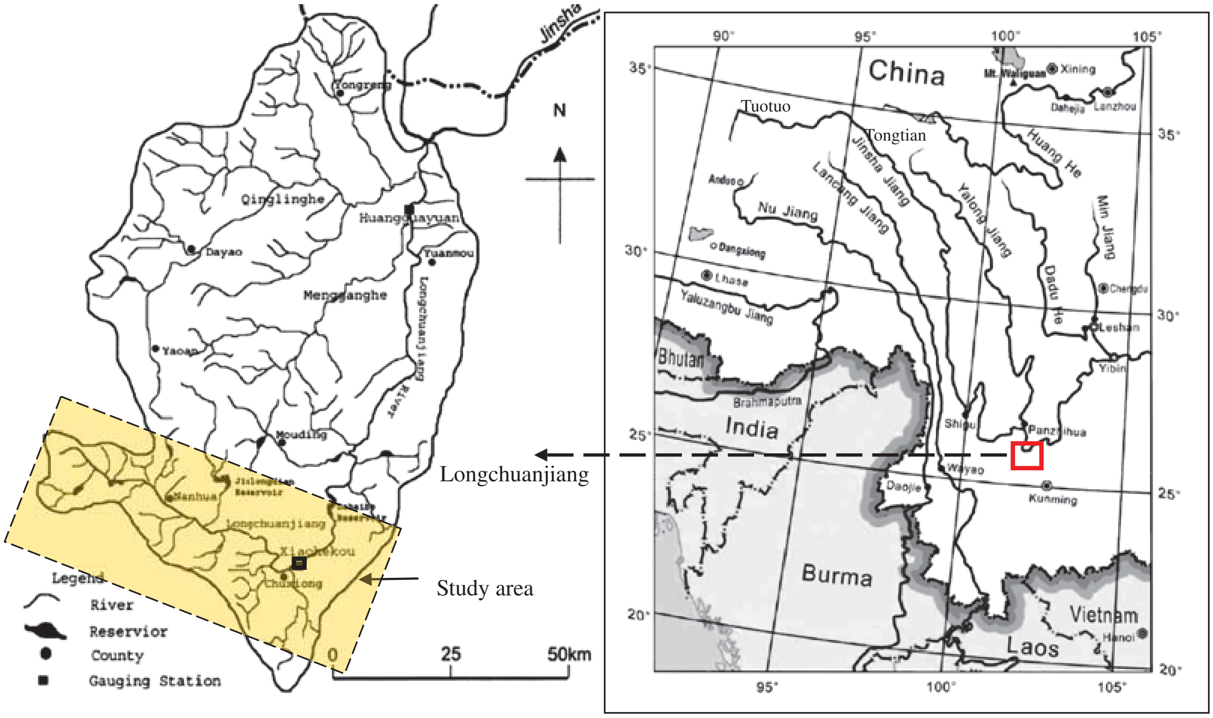

Figure 1. Location map of the Longchuanjiang River with sampling sites and gauge stations and other tributaries in Yangtze River, China.

TSS concentration was determined by the weight difference of filter paper before and after filtering. DOC was measured using TOC analyser (phoenix8000, Tekmar) in the Ailaoshan Forest Ecological Research Station (ASSFERS), Chinese Academy of Sciences. POC and C/N ratio were analysed using a CHN analyser in the Key Laboratory of Tropic Forest Ecology, Chinese Academy of Sciences. TOC is the sum of DOC and POC. Duplicate samples were run, and the precision was within 5% for C and N. In the present work, DIC was recalculated from HCO_3^- concentration because of the river waters with pH ranging from 7.2 to 8.4, measured using acid titration reported by Li *et al.* (2011).

The annual discharge and sediment for 50 years (1958–2008) were also from Xiaohekou gauge station. Kendal Tau tests were used to analyse the trends of annual sediment load and water discharge over the past 50 years. In the beginning of the 1980s, China transferred its socioeconomic policies from a commune system to a so-called land responsibility system. Subsequent rapid economic development during post-1980s resulted in a profound land use alteration, such as deforestation, urban and agricultural expansion and road construction. Severe soil erosion since 1980 together with higher rainfall had increased sediment flux in the river basin (Lu, 2005); thus, the time series were divided into two portions pre- and post-1980 for the tests (Figure 2).

Calculations of organic carbon fluxes and yields

There are generally two flux calculation methods including interpolation and extrapolation on the basis of chemical concentrations and water flow data (Zhang *et al.*, 2009; Lu et al, 2011). Interpolation method assumes that the concentration in water samples is constant in the river for the period between sampling occasions. Extrapolation method develops a rating relationship between concentration and instantaneous water discharge, and this relationship is used to a detailed flow record to calculate annual yield in a basin.

Many studies simply estimated the annual yield of organic carbon using discharge weighted concentration and annual river discharge because of the unavailability of detailed water flow or sediment (i.e. Balakrishna and Probst, 2005). The results obtained were underestimated because of large amounts of organic matter load during the storm period. In the present study, POC and TOC fluxes were calculated using extrapolation, and the rating relationship between organic carbon concentrations and water discharge or sediment concentration could be used for the sampling period. Despite that DOC concentration had no significant relationships with water discharge, DOC yield could be calculated by the difference between TOC and POC yields. It also could be quantified by the relationship between DOC fluxes and instantaneous river discharge and the following empirical equation best fitted to describe the above associations (from Figures 2 and 3):

$$F_{\mathrm{DOC}} = 8.3595\,F_{\mathrm{water}} + 1.7988\ \left(R^2 = 0.96, p < 0.01, n = 24\right) \quad (1)$$

Both depth and lateral integrated sampling have been conducted periodically by the gauging staff, and mean daily water discharge was recorded. Hence, we could obtain reliable annual organic matter loads (Table I). Because of the absence of daily discharge in 2009, we chose organic carbon loads in 2008 representing recent annual yield.

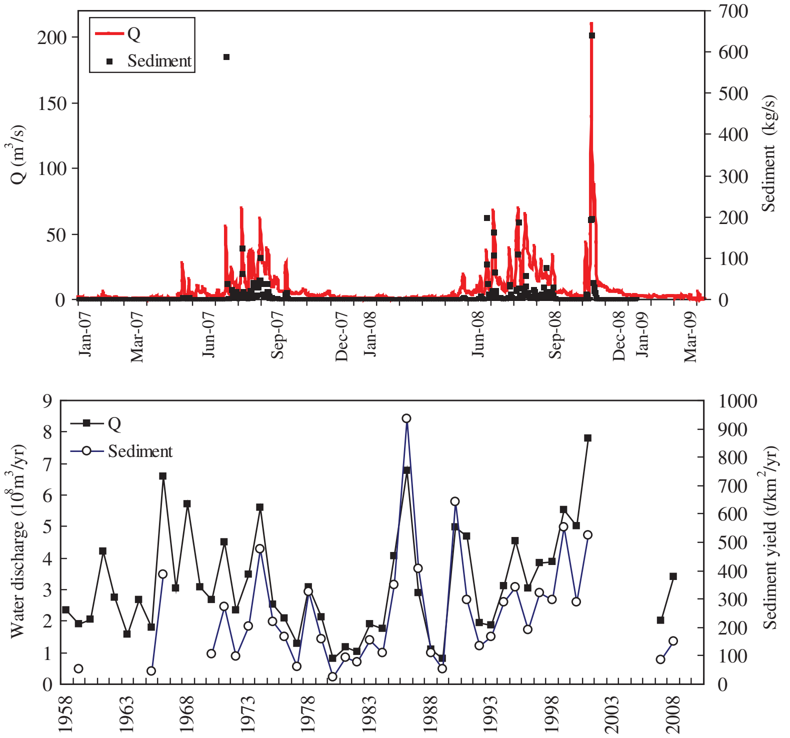

Figure 2. (a) Diurnal mean water discharge (m^3/s) and sediment flux (kg/s) from 1 January 2007 to 30 March 2009 and (b) annual flux of water discharge and sediment in the past 50 years in the Longchuanjiang River (Xiaohekou station), China (*x*-axis in MM/YY) (data are from the Xiaohekou gauging station; a clear trend of increased sediment production from 1980 to 2001 was detectable by Kendal tau test).

The annual yield of organic matter (F_{matter}) in a river was compared using varied calculations in our study:

$$(\text{a})F_{\text{matter}} = C_{\text{w}} \times Q/A \tag{2}$$

where C_{w} is the discharge weighted concentration, Q is the annual discharge of the river (m^3/s) and A is the catchment area;

$$(\text{b})F_{\text{matter}} = \left(\sum_{i=1}^{365} Ci \times Qi\right)/A \tag{3}$$

where C_{i} is obtained using the rating relationship between organic matter and instantaneous water discharge and Q_{i} is the daily river discharge (this study);

$$\begin{aligned}(\text{c})F_{\text{DOC}} &= \text{a}\,F_{\text{water}} + b(\text{see Equation (1)}), F_{\text{POC}} \\ &= 14.612\,F_{\text{water}} \\ &\quad -35.009\ \left(R^2 = 0.90, p < 0.01\right)\end{aligned} \tag{4}$$

(from Figures 2 and 3).

RESULTS AND DISCUSSION

Seasonal variations of water and sediment discharge

Daily water discharge and sediment flux was shown in Figure 2. Daily river discharge was the highest in the beginning of November 2008 (210 m^3/s), and the minimum value was 0.1 m^3/s (April 2008). A flood occurred in the autumn 2008, resulting in a higher annual water discharge in 2008 than that in 2007 ($2 \times 10^8\ m^3/yr$ in 2007 *vs* $3.4 \times 10^8\ m^3/yr$ in 2008, respectively). The total water flow in 2008 was close to the average water discharge of $3.2 \times 10^8\ m^3/yr$ during the 1950s to 2001 (Lu, 2005). High flow period ranged from May to October in 2007 and from May to November in 2008, whereas low flow period ranged from January to April in 2007, November in 2007 to April in 2008, and December in 2008 to March in 2009. Water discharge in high flow period accounted for 88.4% of the total water discharge in 2007 and 93.7% in 2008.

Sediment flux was largely proportional to water flows and varied from 0 kg/s to as high as 640 kg/s (November 2008). The second highest sediment flux (590 kg/s) occurred in July 2007 with a comparatively low water discharge of 55 m^3/s, which was attributed to the extreme storm events possibly occurred in severely eroded areas of the basin. There was no a clear trend for annual sediment production, although a clear increasing trend from 1982 to 2000 was detectable ($p < 0.05$ by Kendal test). The sediment transports in 2007 and 2008 (1.5×10^5 *vs* 2.7×10^5 t/yr, respectively) were markedly smaller than the annual average of 4×10^5 t/yr for the period 1970 to 2001 (Figure 2), which could be due to the recent development of hydropower reservoirs and vegetation recovery by China's Grain for Green Project.

Seasonal variations of organic carbon concentrations

Although instantaneous TSS concentration, varying from 39 to 440 mg/l, peaked before water discharge, it had

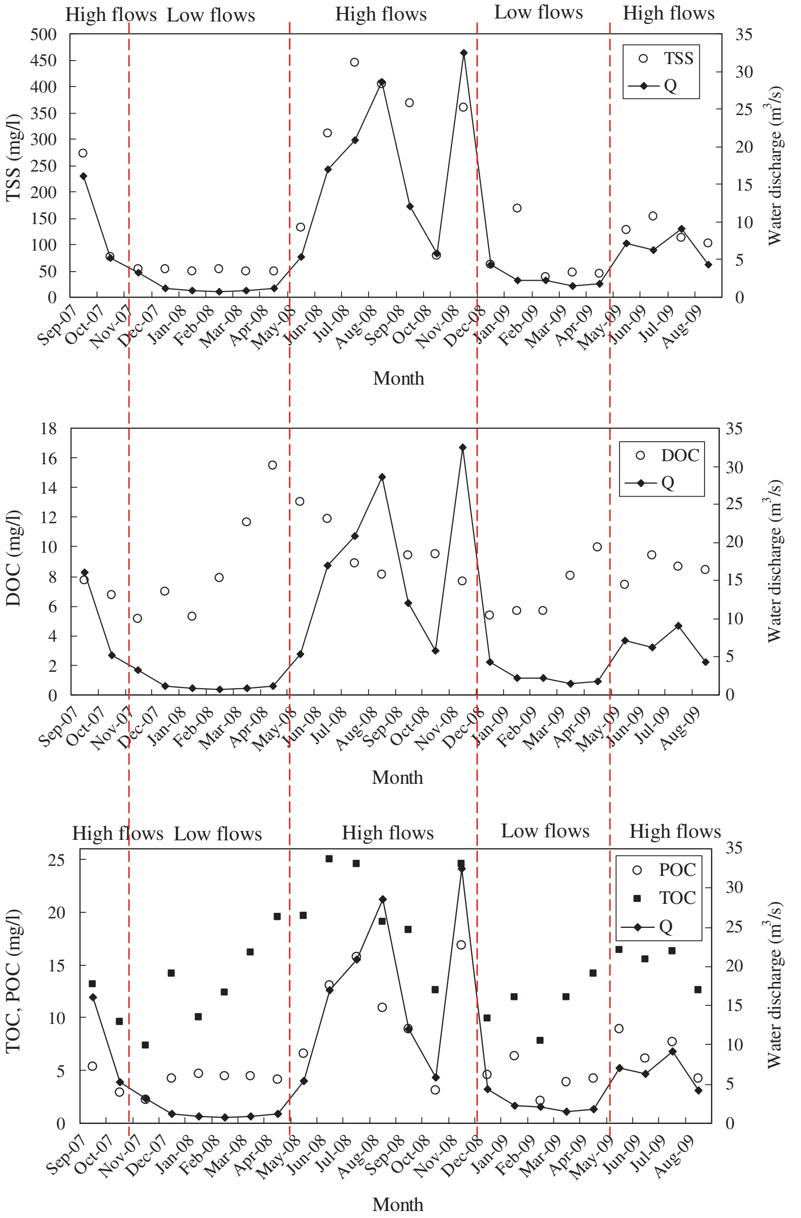

Figure 3. Seasonal variations of (a) TSS, (b) DOC and (c) TOC and POC in the outlet of the river basin (at Xiaohekou station, Chuxiong county) (*x*-axis in MM/YY).

Table I. Organic carbon fluxes of the Longchuanjiang River in 2008 in the outlet of the river basin at Xiaohekou station

	Discharge		POC		DOC		TOC		DOC/POC	DOC[a]	
	Flux (10^6 m^3)	%	Flux (t)	%	Flux (t)	%	Flux (t)	%		Flux (t)	%
Low flows	22	6	100	1.4	210	7	290	3	2.1	200	7
High flows	320	94	7000	98.6	2700	93	9800	97	0.39	2700	93
Total	340	100	7100	100	2900	100	10000	100	0.41	2900	100

[a] Calculated using Equation (1).

similar temporal variations as water discharge (Figure 3), reflecting its close associations and primarily hydrological control ($R^2 = 0.8$, $p < 0.01$; Figure 4). The combined effects of spatial heterogeneity in rainfall and human disturbances may result in an exhaustion of sediment before peak water discharge.

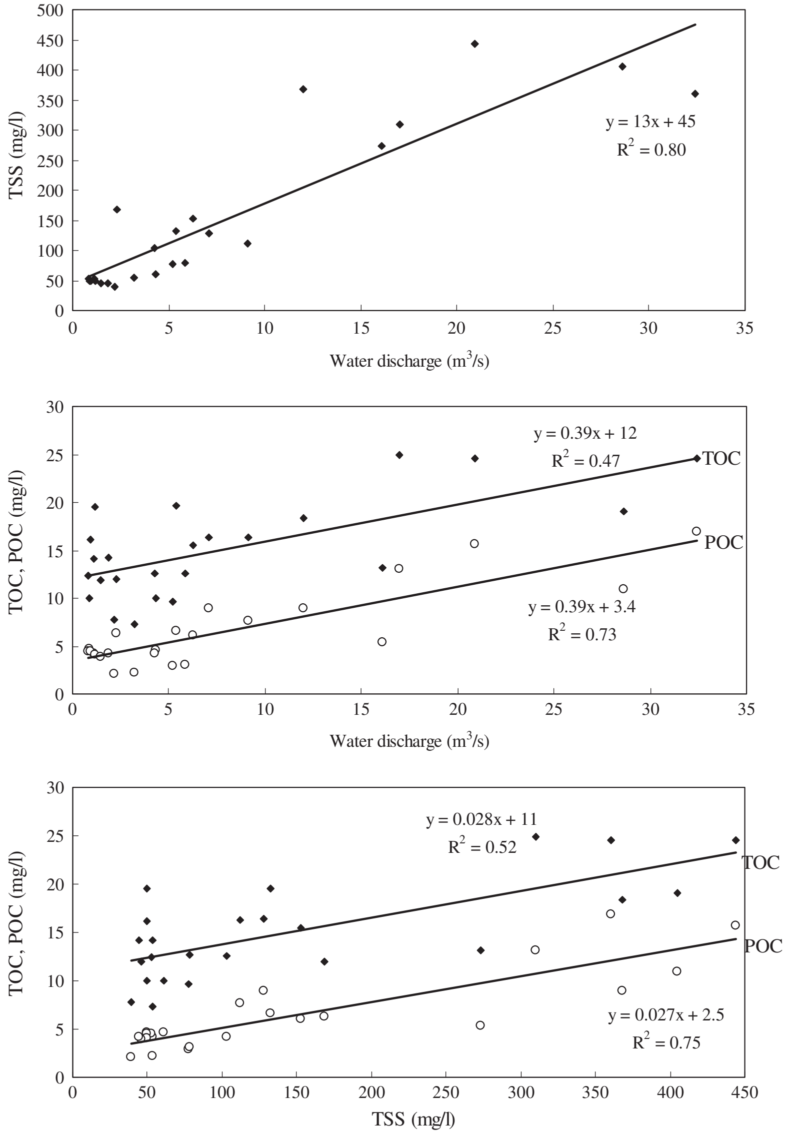

Figure 4. Relationships of (a) TSS and water discharge, (b) organic carbon and water discharge and (c) organic carbon and TSS in the outlet of the river basin (Xiaohekou station).

Compared with seasonal variations in TSS and water discharge, the concentration of DOC varied relatively small, that is, from a low value of 5.2 to a high of 15 mg/l, with an average of 8.5 mg/l, which was approximately 1.6 times of the global average, that is, 5.4 mg/l (Ludwig *et al.*, 1996). Despite the highest concentration (15 mg/l) that occurred in the high flow period (April 2008), the averages in high and low flow periods showed little differences (9 *vs* 8 mg/l, respectively) (Figures 3). A weak correlation between DOC and TSS was found in both high and low flow periods or all data points considered. DOC had a dominant allochthonous sources from terrestrial environment (litters and soils) (Raymond and Bauer, 2001; Helie and Hillaire-Marcel, 2006; Zhang *et al.*, 2009), confirmed by strong linear relationship between DOC flux and water discharge ($R^2 = 0.96$, $p < 0.01$, $n = 24$). The insignificant relationships between DOC concentrations and water discharge/TSS could be due to the interaction of the dilution effects of precipitation and biological processes in the reservoirs (Tipping *et al.*, 1997; Parks and Baker, 1997).

In contrast to the DOC, higher POC concentrations with a range of 2.1 to 17 mg/l were found in the high period, especially in November 2008 (Figure 3). The seasonal variations of POC concentrations and water discharge were almost similar (Figure 3), thus POC had a significant linear relationship with water discharge or TSS ($R^2 = 0.75$, $p < 0.01$; Figure 4). This indicated that large amounts of terrestrial particulate matter were transported to rivers by runoff during high flow period, resulting in high POC concentrations in the high flow period. The resuspension of particles by high water flow could also elevate POC concentrations (Tipping *et al.*, 1997). The organic carbon content of TSS decreased with increasing TSS concentrations (Figure 4), with the percentage content of POC in suspended sediment ranging from 2% to 9.4%. The elevated concentrations of >8% measured at low discharges of <5 m^3/s and the reduced concentrations between 2% and 5% at high discharges of >10 m^3/s (Figure 5). The similar relationship has been observed in other river systems (Meybeck, 1982; Ludwig *et al*., 1996; Balakrishna and Probst, 2005; Gao *et al*., 2002, 2007).

This inverse relationship between POC content (%) and TSS was due to the following processes (Ludwig *et al*., 1996; Ittekkot, 1988; Gao *et al*., 2002; Balakrishna and Probst, 2005). First, with the intensifying mechanic erosion, the riverine POC is diluted by mineral-like material coming from the erosion of terrigenous soils and remobilisation of mineral matter in river bed. Second, surface soil horizon contains higher POC content, whereas soils in the deeper horizon with lower POC are scoured into rivers during the intensive monsoon precipitation period, which also results in decreasing POC content (%) in suspended sediment with increasing TSS concentration. Third, higher TSS concentration leads to reduced availability of light, which restricts the growth of phytoplankton and consequently reduces autochthonous carbon.

TOC concentration varied between 7.4 and 25 mg/l with an average of 15 mg/l (Figure 3). Similar to POC concentration, TOC showed a positive relationship with water discharge and TSS (Figure 4), which could be used for calculation of TOC flux. TOC tended to display similar seasonal variations as POC in the high flow period, but as DOC in the low flow period, reflected by much higher *R*-value between POC and TOC ($R = 0.94$, $p < 0.01$) in the high flow period, and between DOC and TOC in the low flow period ($R = 0.91$). This indicated that TOC was largely regulated by POC in the high flow period but by DOC in the low flow period, which was consistent with higher POC/TOC ratio occurred in the high flow period but higher DOC/TOC ratio in the low flow period (Figure 6).

Sources of organic carbon

The C/N ratio is generally indicative of the source of organic matter, with higher C/N ratios (15–30) in terrestrial organic matter (humus) compared with those in phytoplankton (~5–8; Meybeck, 1982; Hedges *et al*., 1986, 1997). The average value of the C/N ratio of the global riverine POC is 10 to 15 (Meybeck, 1982). Our data showed large variations of the C/N ratios from 12 to 220 with an average of 77 (Figure 7). The C/N ratios were >25 for most of the samples (74%), the critical value for marker of terrestrially derived matter reported by Meyers (1994).

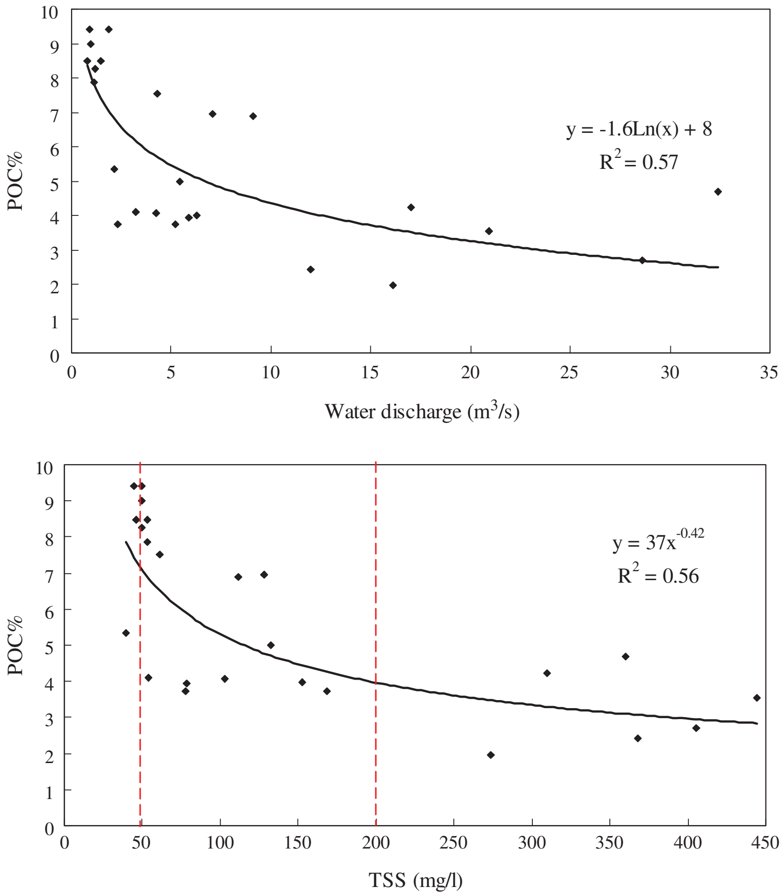

Figure 5. Relationships of POC in percentage of suspended sediment (%) and (a) water discharge and (b) TSS in the outlet of the river basin (Xiaohekou station).

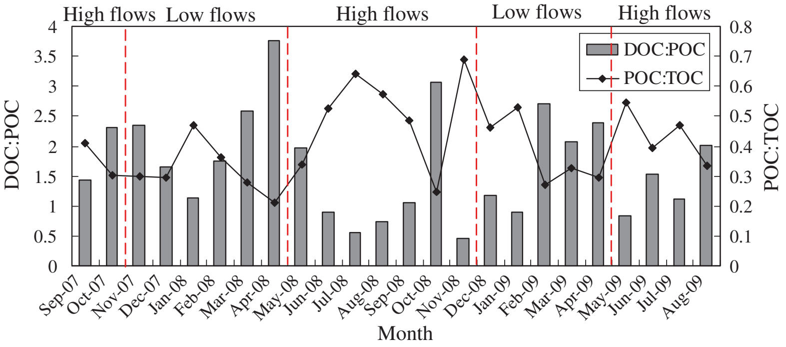

Figure 6. Seasonal variability in relative importance of organic carbon concentrations in the Longchuanjiang River (x-axis in MM/YY).

Thus, the C/N ratios were considerably higher than those for plankton and bacterial biomass, although comparable with those for soil humus and terrestrial plants (Hedges *et al*., 1997), which indicated a predominantly allochthonous source to riverine POC, similar to most of the world's monsoon rivers (Ittekkott and Zhang, 1989).

However, a negative relation between the C/N ratios and the TSS in the Longchuanjiang River was observed, although the R-value was low ($R^2 = 0.5$; Figure 7), which was contrary to many reports (i.e. Balakrishna and Probst, 2005; Zhang *et al*., 2009). Moreover, a weak correlation between POC and the C/N ratios was also found for the high and the low flow periods. This suggested that terrestrially particular nitrogen in the Longchuanjiang catchment increased sharply during the storm season. Also, the C/N ratios were constant when the concentrations of TSS were >200 mg/l, suggesting a relatively stable C/N values (mean = 21, median = 18) for terrestrial material during further intense soil erosion. When a river had low turbidity during low flow period, the contribution from *in situ* phytoplankton would be enhanced because of light availability, thus the nitrogen rich in phytoplankton tissues in comparison with fresh plant material results in lower C/N ratios (5–8) (Balakrishna and Probst, 2005). Thus, the abnormally large variability of the C/N ratios (37–219) in the low flows could be contributable to waste discharges from the Chuxiong County, adjacent to the hydrological gauge station (Figure 7).

POC originates mainly from terrestrially (i.e. soil and litter) and aquatically photosynthesised organic matter in natural fresh waters. The C/N ratios in suspended sediment are much higher in terrestrial plant material and debris (i.e. 20–400; Hedges *et al*., 1997) than those in the global and China's soil organic matter (i.e. 15–30; Meybeck, 1982; Batjes, 1996; Li *et al*., 2007; Wang *et al*., 2011), and previous studies on Chinese rivers reported C/N ratios in

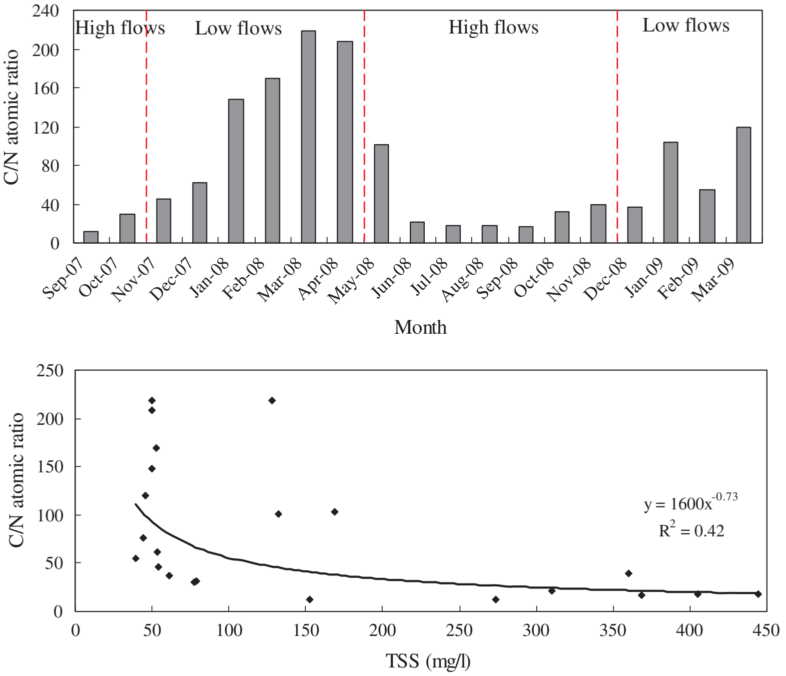

Figure 7. (a) Seasonal variations in atomic C/N ratios and (b) the relationships between C/N ratios and TSS weight in the Longchuanjiang River, China (x-axis in MM/YY).

sediment varying from 6 to 15 in the Yangtze River system (Wu *et al.*, 2007) and from 8 to 12 for Zengjiang (Gao *et al.*, 2007), from 7 to 9 for Zhujiang Delta (Ni *et al.*, 2008) and from 5.9 to 13 for Luodingjiang (Zhang *et al.*, 2009) of the Zhujiang basin, indicating the dominant contribution to riverine POC from soil organic carbon (SOC). Hence, C/N ratios in the Longchuanjiang (12–40 and 12–220 in the high and low flow periods, respectively) demonstrated that terrestrially produced POC was largely controlled by soils in the high flows and by urban wastewaters in the low flows. Moreover, the POC concentrations dropped to 2% to 4.7% as the TSS concentrations rose to more than 150 mg/l (Figure 5); thus, the POC threshold reached (i.e. 2.5%) was comparable with that in the Yangtze River system (0.5%–2.5%) (Wu *et al.*, 2007) and Zhujiang basin (2% or lower) (Gao *et al.*, 2007; Ni *et al.*, 2008; Zhang *et al.*, 2009), where POC was considered erodible soils in origin. This provided further evidence that riverine POC in the Longchuanjiang River primarily originated from soil erosion in the high flow period.

The sources of POC, such as aquatic (autochthonous) and terrestrial (allochthonous) sources, can be further quantified. The contribution from each end member could be calculated using the following model:

$$A_{\text{sample}} = B_{\text{autochthonous}}A_{\text{autochthonous}} + B_{\text{allochthonous}}A_{\text{allochthonous}} \tag{5}$$

$$B_{\text{autochthonous}} + B_{\text{allochthonous}} = 1 \tag{6}$$

where A was the C/N ratio or POC content (%) in TSS and B (%) was the contribution rate. The subscript "autochthonous" and "allochthonous" represented autochthonous and allochthonous sources, respectively. Despite that, $A_{\text{autochthonous}} = 6$ could be designated according to the C/N ratios for aquatic biomass in different water bodies including the Zhujiang basin (Gao *et al.*, 2007), whereas the C/N ratios in soils, wastes and plant litter in the present study area were not measured. Therefore, the quantification for POC sources was not conducted according to the C/N ratios.

We used a rough hypothesis on the differentiation of POC source using the POC content (%) of TSS in different end members. In Figure 5, the curve tended to flatten out at 2.5% of POC and 200 mg/l of TSS, indicating the threshold of terrestrial (allochthonous) organic matter end member. The samples with low TSS weight and a higher percentage of POC corresponded to the phytoplankton and urban wastewaters; thus, a 10% POC was assumed as the autochthonous end member (including urban wastewaters). The instantaneous fluxes of autochthonous and allochthonous POC were determined (Figure 8). Hence, 22% of POC annual flux was from autochthonous production and 78% from allochthonous sources.

Annual organic carbon fluxes

The results of flux calculations for riverine organic carbon species are shown in Table I. The transported load in the year of 2008 was 10,000 t for TOC. Most of the organic carbon load was POC (7100 t/yr), and only 29% was DOC (2900 t/yr). Organic carbon transports displayed great seasonality and nearly 97% of TOC transported during the high flow period from May to November. Compared with DOC transport in the high flow period, POC transport was more sensitive to rising water discharge with approximately 98.6% transported in the high flow period.

The TOC yield in the upper Longchuanjiang River was 5.6 t/km^2/yr, among which 4 t/km^2/yr was POC and 1.6 t/km^2/yr was DOC (Table II). The ratio of dissolved to POC export fluxes was 0.4 for the year 2008, with the high value (2.1) in the low flow period and low value (0.4) in the high flow period (Tables I and II). Compared with the global rivers, the area-specific flux of organic carbon was relatively higher and also higher than the global average level despite that DOC flux was slightly lower than the global average level (Table II; Ludwig *et al.*, 1996). Compared with other Chinese rivers, our results were in the range of the Yangtze River system but higher than that in the low turbidity tributaries in the Zhujiang basin (Table II) (Wei *et al.*, 2003; Gao *et al.*, 2007; Sun *et al.*, 2007). Our results were also lower than those reported by Ludwig *et al.* (1996). Previous studies reported the physical, chemical and economical processes in a drainage basin together controlled the organic geochemical characteristics (Ludwig *et al.*, 1996), and hydrological seasonality and soil erosion were the major contributors to organic matter transports,

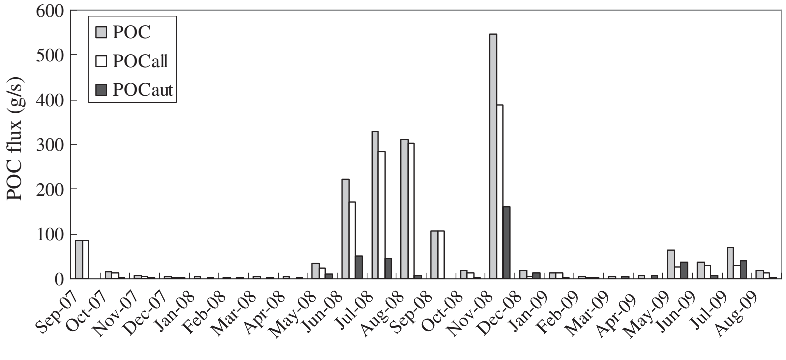

Figure 8. Instantaneous flux of autochthonous and allochthonous POC (g/s) in the Longchuanjiang River, China: POC flux (g/s). POCaut, POC from autochthonous source; POCall, POC from allochthonous source.

Table II. Comparison of organic carbon transported in the Longchuanjiang River and that of other drainage areas

Climate type	River	Area	Discharge	F_{TSS}	F_{POC}	F_{DOC}	F_{TOC}	DOC/POC	Reference
		10^3 km^2	10^9 m^3/yr	t/km^2/yr	t/km^2/y	t/km^2/y	t/km^2/y		
Monsoon									
	Longchuanjiang	1.8	0.34	150	4	1.6	5.6	0.41	This study
	Luodingjiang	3.2	2.7	85	1.1	1.2	2.3	1.2	Zhang *et al*. (2009)
	Zengjiang	2.9	3.8	85	0.8				Gao *et al*. (2007)
	Yangtze River	1800	990	250	1.2	0.5	1.7	0.41	Wu *et al*. (2007)
	Yellow River	750	32	900	8.1	0.3	8.4	0.03	Zhang *et al*. (1992)
	Yellow River	820	34	1100	5.5	0.1	5.6	0.01	Cauwet and Mackenzie (1993)
	Xijiang	350	230	190	8.3	1.9	10	0.23	Gao *et al*. (2002)
	Xijiang	350	230	190	1.2	1	2.2	0.79	Sun *et al*. (2007)
	Xijiang	350	230	190	0.5	1	1.5	2.2	Wei (2003)
	Ganges/Brahmaputra	1600	1000	670	5.2	2.2	7.4	0.42	Ludwig *et al*. (1996)
	Indus	910	100	260	1.9	2.9	4.8	1.6	Ludwig *et al*. (1996)
	Godavari	310	100	550	2.4				Balakrishna and Probst (2005)
	Garonne	53	20	45	1.5				Veyssy *et al*. (1999)
	Irrawaddy	410	430		5.5–10	2.2			Bird *et al*. (2008)
	Salween	270	210		8.8–12	0.8			Bird *et al*. (2008)
Non-monsoon									
	Amazon	5900	5900	190	2.8	4.5	7.3	1.6	Ludwig *et al*. (1996)
	Zaire	3700	1300	11	0.7	2.5	3.2	3.6	Ludwig *et al*. (1996)
	Mississippi	3200	490	120	0.3	1.3	1.6	4.1	Ludwig *et al*. (1996)
	Ob	3100	410	6	0.1	1.2	1.3	10.3	Ludwig *et al*. (1996)
	Parana	2900	470	30	0.3	1.4	1.7	5.1	Ludwig *et al*. (1996)
	Mackenzie	1600	280	23	0.9	0.8	1.7	0.98	Ludwig *et al*. (1996)
	St. Lawrence	1100	480	4	0.3	1.6	1.9	5	Ludwig *et al*. (1996)
	St. Lawrence	1100	330		0.1	1.1	1.2	14	Helie and Hillaire-Marcel (2006)
	Orinoco	1000	1100	150	1.6	4.8	6.4	3	Ludwig *et al*. (1996)
	Yukou	840	190	71	0.3	1	1.3	3.1	Ludwig *et al*. (1996)
	Rhine	160	30	4	0.6	1	1.6	1.8	Ludwig *et al*. (1996)
Average of global rivers		110000	38000	170	1.6	1.9	3.5	1.2	Ludwig *et al*. (1996)

F_{TSS}, flux of total suspended sediment; F_{POC}, flux of particulate organic carbon; F_{DOC}, flux of dissolved organic carbon; F_{TOC}, flux of total organic carbon.

which was responsible for large discrepancies in organic carbon fluxes among rivers.

The 0.4 ratio of DOC to POC flux in the Longchuanjiang River was consistent with most rivers in the Asian monsoon climate regions (except for a much higher ratio of 1.2 for the Luodingjiang) (Table II). This ratio (0.4) was evidently lower than the global average (1.2) and the Zhujiang systems but comparable with those in the Yangtze River and higher than that in the Yellow River (Table II). Because of the decreasing sediment flux in the recent decade due to reforestation and reservoir construction, the dominance of DOC flux over POC flux was expected in the Luodingjiang, a tributary in the Zhujiang (Zhang *et al*., 2009). This phenomenon could also occur in the Longchuanjiang River, with a reduction in sediment loads in the recent years. In contrast to the non-monsoon drainage areas, the flux of particulate species dominated over dissolved species in the monsoon areas (Table II).

Table II also showed the different estimates of organic carbon fluxes for the same river basin, such as the Yangtze River, the Yellow River and the Xijiang River. The sampling scheme and associated processes contributed to the differences. For instance, the Yangtze River had a quite low organic carbon yield (1.7 t/km^2/yr for TOC) reported by Wu *et al*. (2007) because of surface waters sampled in the dry season (April–May). The sampling scheme is similar in the other two studies (Table II; Gan *et al*., 1983; Cauwet and Mackenzie, 1993), that is, surface waters by cruises during the two seasons of high and low water discharge, but their results for organic carbon flux diverged greatly (Table II). The huge discrepancy in the two studies was primarily due to the low sampling frequency, which might not adequately represent the great seasonality of organic carbon flux. Sampling method, that is, either surface samples or depth and lateral integrated samples, is a major control factor for organic carbon. For example, both surface and bottom water samples during the dry (May) and wet (September) seasons in 1987 (Zhang *et al*., 1992) resulted in much higher organic carbon transport compared with that in 1985 (Cauwet and Mackenzie, 1993) using only surface waters by two cruises (May and August) for the Yellow River (Table II). Hence, detailed sampling with integrated waters and diurnal water discharge are essential for reliable estimates of organic carbon transport.

The annual yield of organic carbon, estimated by the three different methods, was compared (Table III).

Similar annual export of DOC was obtained, but a large deviation for POC flux. Thus, reliable and representative data as well as appropriate method were important to estimate an accurate organic matter flux, and organic matter particularly POC fluxes should be reassessed in many studies (Gao *et al.*, 2002; Balakrishna and Probst, 2005).

Flux calculations were also made for dissolved carbon (DIC) in 2008 on the basis of the HCO_3^- data by Li *et al.* (2011), with the objective of estimating the total carbon flux from upper Longchuanjiang River. Similar to the behaviour of organic carbon flux, DIC flux was also sensitive to rising water discharge with approximately 92% transported in the high flows. The DIC concentration varied small with a value of 2 for the ratio of its maximum to minimum contents, despite its negative associations with water discharge. This indicated that DIC as a function of alkalinity had conservative behaviour in the river. The DIC export from the upper Longchuanjiang was estimated at 11,000 t/yr (6 t/km^2/yr), which was 1.5 times higher than the POC flux while close to the TOC flux. Thus, a total flux (DIC + POC + DOC) was approximately 21,000 t/yr (12 t/km^2/yr) from the upper Longchuanjiang River. Out of the total flux, the organic carbon contributed approximately 48% comparable with the flux estimates of worldwide rivers (Ludwig *et al.*, 1996).

Organic carbon fluxes change over the past decades

Studies reported that organic carbon fluxes, particularly POC fluxes, varied with the changes of sediment load under anthropogenic disturbances. This observation was obtained in the Yangtze River systems, that is, decreasing sediment load due to the construction of dams and associated water discharge reduction and therefore decreased POC flux (Wu *et al.*, 2007). Lu (2005) reported that slight increase in vegetation cover and dam building could not counterbalance the increasing sediment flux by climate change and anthropogenic activities such as land use change, road construction, urbanisation in the Longchuanjiang catchment, which could explain increases in annual water discharge and sediment flux in the Longchuanjiang River since the 1980s ($p < 0.05$ by Kendal test for 1980–2001; Figure 2). However, both sediment load and water discharge showed sharp decreases in the 2000s, suggesting the influences of different human disturbances such as reforestation and dam/reservoir construction in the upper Longchuanjiang River, which can consequently result in the large reduction in export of organic matter.

Table III. Comparison of annual organic carbon fluxes estimated using different processes

	DOC (t/km^2/yr)	POC (t/km^2/yr)	Reference
Extrapolation[a]	1.6	4	This study
$C_w \times Q / A$[b]	1.6	1.9	
$F_{OC} \sim F_{water}$[c]	1.6	2.5	This study

[a] Using the rating relationships between organic matter concentrations and real-time water discharge as well as mean daily water discharge (Equation (3)).

[b] Equation (2).

[c] $F_{DOC} = 8.3595F_{water} + 1.7988$; $F_{POC} = 14.612F_{water} - 35.009$ (Equation (4)).

The highest sediment yield (930 t/km^2/yr) occurred in 1986, more than six times than that in 2008 in the catchment. The minimum POC content in suspended sediment 0.5% is close to organic carbon content in shales and represents a lower organic carbon level in riverine suspended sediment (Meybeck, 1982; Ittekkot, 1988). Thus, the minimum POC yield in 1986 with highest sediment yield was 4.7 t/km^2/yr. The POC content was close to 2.5% for the TSS concentration of >350 mg/l in the river basin (Figure 5), so the actual POC flux (i.e. 23 t/km^2/yr) in 1986 was much higher than the value 4.7 t/km^2/yr. Hence, the current annual yield was much lower than the estimated load in 1986 because of the dramatic sediment load decrease. Similar trend for POC flux was observed for Indian rivers such as Godavari, that is, 9.1 t/km^2/yr in 1993 to 2.4 t/km^2/yr during 1998–1999 (Gupta *et al.*, 1997; Balakrishna and Probst, 2005). This reflected the large impact of human activities including reforestation for reducing soil erosion and the construction of dams and reservoirs for sediment trapping in the river basin.

SOC is the dominant source of riverine POC in the Yangtze River (Wu *et al.*, 2007); thus, riverine POC content in suspended sediment can reflect soil fertility. POC content in suspended sediment of the Yangtze River decreased dramatically (from ~2.5% to ~1%) from 1976 to 1986 (Gan *et al.*, 1983; Cauwet and Mackenzie, 1993) and reached to 0.5% in 2003 (Wu *et al.*, 2007), resulting in land degradation in the drainage basin. Thus, SOC sequestration is of great importance for sustainable land use.

Carbon enrichment ratio was higher than clay enrichment ratios during the process of water erosion (Wang *et al.*, 2010). The riverine POC content in sediment (2%–9.4%) of the Longchuanjiang River was higher than SOC (1%–2%) of the soils in Yunnan Province, Southwest China, on the basis of the soil survey in 1992 (Zhang *et al.*, 2008). Further, carbon enrichment ratio was higher in the low flow period than that in the high period (Figure 5). In the wet season, sediments are fully dispersed, more aggregates are disrupted by consecutive rainfall events and fine fraction with high carbon content is removed, thereby resulting in higher carbon enrichment ratio in riverine suspended sediment in the low flow period.

Recent studies indicated a carbon sink from cropland erosion (Van Oost *et al.*, 2007; Harden *et al.*, 2008), which was also supported by Wang *et al.* (2010) who studied small catchment-scale carbon redistribution and carbon mineralisation during erosion and transport. However, the enhancement of mineralisation during sediment transport over long distance could lead to significant carbon degradation, particularly in large rivers (Alewell *et al.*, 2009). Thus, more field data and further study should be conducted, for example, in the Yangtze River basin with severe soil erosion, to understand the fate of carbon transport, carbon redistribution and carbon delivery with increase in river distance.

CONCLUSION

The riverine flux of organic carbon, dominantly by continent erosion substance, plays a key role in the global carbon biogeochemical cycling. This study investigated DOC, POC and atomic C/N of particulate organic matter to understand the source of organic carbon and the net fluxes of organic and inorganic carbon from a catchment in the Upper Yangtze basin. The concentrations of DOC, POC and TOC in this catchment showed great seasonality controlled by water flow and therefore suspended sediment. The atomic C/N ratios of particulate organic matter varied from 12 to 220 with an average of 77, demonstrating the dominance of terrestrial sources in the high flow period and primarily urban wastewaters in the low flow period. Moreover, terrestrial production contributed 78% to POC, and the rest of POC was due to aquatic origin (including urban waste waters). Anthropogenic activities such as agricultural practices and deforestation coupled with heavy rainfall resulted in higher fluxes of organic carbon, that is, 5.6 t/km^2/yr for TOC, composed of 1.6 t/km^2/yr of DOC and 4 t/km^2/yr of POC. The ratio of DOC to POC was 0.4, demonstrating the intense mechanical erosion in the drainage basin. The total carbon export from this river was 21, 000 t/yr (11,000 t/yr for DIC and 10, 000 t/yr for TOC), reflecting the higher transports of carbon in the drainage catchment of the upper Yangtze basin.

Our study indicated a large difference in the POC exports using various calculation methods, thus reliable and representative data should be used for estimating organic matter fluxes in many rivers worldwide. Because of the seasonal heterogeneity of precipitation, particularly for the monsoon-climate rivers, intensified sampling should be conducted in the high flow season, particularly during the flooding period where one sampling is required each day. Meantime, daily water discharge is another important factor for estimating POC exports. Damming, diversion and withdrawal of water from the main channel have greatly reduced sediment fluxes in the Longchuanjiang River and consequently regulated the yield of organic carbon and the seasonal variability in organic carbon concentrations and compositions. It is anticipated that organic matter particularly POC transports will further decrease from the Yangtze to the East China Sea because of the construction of hydropower dams.

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