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Research papers

Damming alters the particulate organic carbon sources, burial, export and estuarine biogeochemistry of rivers

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ABSTRACT

The long-term changes in composition and export of particulate organic carbon (POC) by rivers due to dam construction are poorly known. Based on observations, incubation experiments and modelling, this study analyzed the sources, spatial and temporal distribution and annual export of POC in the Changjiang River during recent decades to explore the POC changes due to dam construction. Changes in POC composition resulted from both increasing POC sequestration and carbon fixation in the river basin. The allochthonous POC (from terrestrial input) export by the Changjiang River to its estuary decreased by ~90% between 1956 and 2002 and 2013–2019. We estimated that 2.3 ± 0.5 Mt C/yr allochthonous POC has been sequestered in the Changjiang watershed since 2013. The autochthonous (entirely from in-stream riverine primary production) contribution increased from 1% to 55% of total POC export during the same period due to improved light transparency caused by decreased sediment discharge. Global POC trapping in nine large rivers strongly impacted by dams is 12% of the global riverine POC export. The reduced POC export and increase in labile autochthonous POC strongly impact the estuarine carbon cycle. Therefore, with continued dam construction in the future, important changes in the riverine and estuarine carbon cycle can be expected.

1. Introduction

Rivers play an important role in the global carbon (C) cycle by transporting terrestrial organic carbon (OC) to the ocean (Battin et al., 2009; Galy et al., 2015). Global river OC export from the land to the ocean is ~300 Tg C yr^{-1}, of which 140 Tg C yr^{-1} is particulate organic carbon (POC) (Seitzinger et al., 2010) and the remainder is dissolved organic carbon (DOC) (Derrien et al., 2019; Szymczycha et al., 2017). During C transport in rivers, transformations of DOC and POC occur as well as burial, adsorption/desorption and aggregation/dissolution (He et al., 2016). The burial of POC in river sediments is a long-term sink of atmospheric CO_2 (Bouchez et al., 2014; Galy et al., 2007) while POC decomposition is a CO_2 source (Fearnside and Pueyo, 2012; Maavara et al., 2019).

Riverine POC has different origins, i.e. allochthonous POC from terrestrial vegetation in riparian and flooded areas or from eroded soil material (Galy et al., 2015; Hilton, 2017), and autochthonous POC derived from in-situ primary production (Ittekkot, 1988; Meybeck, 1982b; Schlesinger and Melack, 1981; van Hoek et al., 2019). Generally, autochthonous POC is more easily decomposable than allochthonous POC (Lin et al., 2019; Mendonça et al., 2012) with approximately 10–50% of total POC (POC_{tot}) being labile, and the complement more resistant to degradation (POC_{res}) (Fabre et al., 2019; Ittekkot and Laane, 1991; Li et al., 2015).

Anthropogenic perturbations, such as dam construction, water diversion, sand mining and land-use change in river basins have substantially changed the POC delivery to inland waters and its export to coastal waters (Butman et al., 2015; Catalán et al., 2016; Dean and

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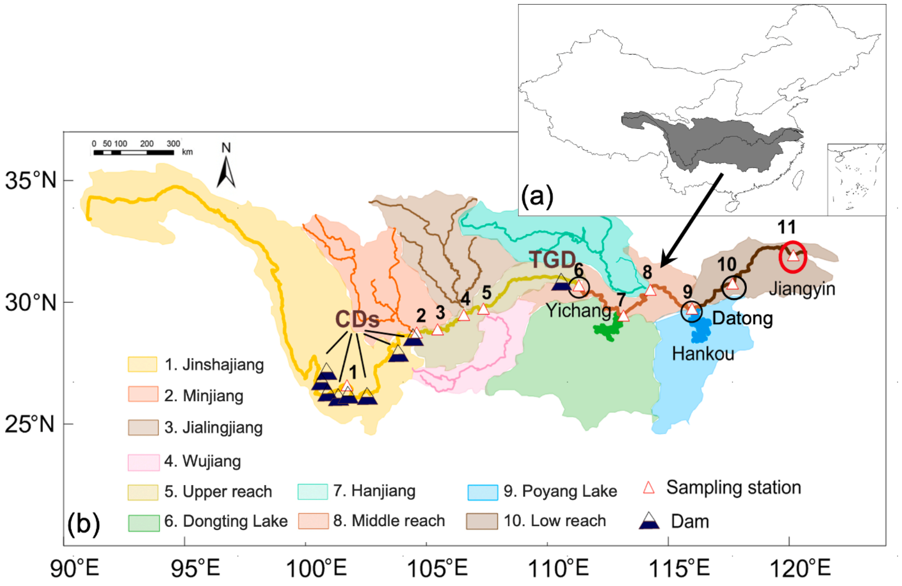

Fig. 1. Location of the Three Gorges Dam (TGD), cascade dams (CDs) and sampling stations in the Changjiang River.

Gorham, 1998; Maavara et al., 2017; Regnier et al., 2013; Walling, 2006). Dams may have an important impact on POC export (Li et al., 2015; Maavara et al., 2017; Mendonça et al., 2012; Mulholland and Elwood, 1982) by increasing the travel time of water, by promoting sedimentation and thus interrupting the transport of POC (Battin et al., 2009; Dean and Gorham, 1998; Stallard, 1998), and by enhancing primary production in reservoirs (Mendonça et al., 2012). Such changes of riverine POC flux and composition have important impacts on the ecology of coastal waters, especially in estuaries (Bauer et al., 2013).

Many global large rivers experienced a marked decrease of suspended sediment discharge (Walling and Fang, 2003; Walling, 2011). Some rivers, like the Yellow River, Indus River and Krishna River, show a progressive decline of sediment and water discharge after the construction of large dams (Rao et al., 2010; Walling, 2011; Wang et al., 2016). This combined reduction in sediment and water discharge may also be related to water diversion and climatic change also play vital roles (Kong et al., 2015; Milliman et al., 1984a; Zhang et al., 2017). Other rivers, including the Changjiang, Red, Chao Phraya, Godavari, Mississippi, Danube and Pearl Rivers, have experienced a decrease of sediment load without significant decrease in annual runoff during the past decades (Das et al., 2021; Meade and Moody, 2010; Vinh et al., 2014; Wu et al., 2019). Previous studies show that the reduction in sediment discharge due to damming in the Red, Chao Phraya, Godavari, Mississippi, Danube and Pearl Rivers fall into the range of 60%–80% (Das et al., 2021; Keown et al., 1986; Le et al., 2007; Walling, 2006; Walling, 2011; Wu et al., 2012), which is less than in the Changjiang River basin (up to 99%) (Yang et al., 2018). Therefore, the Changjiang River could be regarded as a typical example to explore the impact of river damming on POC export from rivers to the sea.

The Changjiang River (Yangtze River) is the third largest river in the world with respect to discharge with a considerable contribution to the global material flux from land to the oceans (Chetelat et al., 2008; Milliman and Meade, 1983; Viers et al., 2008). Rapid dam construction in the Changjiang River basin since the 1950 s has resulted in the current > 50,000 reservoirs with a total storage volume of 140 km^3 which is 16% of the average annual water discharge (Dai and Lu, 2014; Nilsson et al., 2005; Yang et al., 2018). As a consequence, the total suspended sediment (TSS) export from the Changjiang River has decreased since the 1970 s (Chen et al., 2008; Yang et al., 2011), with a sharp decline after the closure of the Three Gorges Dam (TGD) and filling of the Three Gorges Reservoir (TGR) in 2003 (Yang et al., 2007). In 2012, the completion of cascade dams (CDs) in Jinshajiang River, the headwater zone of the Changjiang River basin stretching from Wudongxi to Xiangjiaba Dams, further increased the trapping of sediment (Li et al., 2015; Wang et al., 2012; Wu et al., 2015; Wu et al., 2007; Yang et al., 2018; Yu et al., 2011; Zhang et al., 2014). However, the effect of these different dams on the amount of POC trapping is poorly understood.

In this study, we characterized POC, TSS and chlorophyll-*a* (Chl-*a*) in the mainstream of the Changjiang River to investigate the factors controlling the spatial distribution of POC delivery, decomposition and transport along the river continuum. Incubation experiments were used to estimate decomposition rates of autochthonous POC. Monthly labile and recalcitrant POC fluxes, C stable isotopes and TSS variation close to the river mouth were analyzed to estimate different POC sources and their seasonal variability. To understand the long-term POC alteration due to damming, we modelled the allochthonous supply, autochthonous production and burial of POC in the whole Changjiang River basin during the past 60 years. Finally, comparisons were made with nine global rivers strongly impacted by reservoirs.

2. Methods

2.1. Study area

The Changjiang River basin covers an area of 1.8 million km^2 which is 20% of China's land area (Fig. 1). The mountainous Jinshajiang subbasin has relatively low temperatures and a dry climate, while the middle and lower reaches mainly consist of fluvial plains with wet and warm climates (Chetelat et al., 2008). The Changjiang River downstream of Datong (about 500 km from the river mouth) is influenced by the tidal range. Most of the Changjiang River basin is dominated by the subtropical monsoon climate and 70–80% of rainfall and water and sediment discharge occur in the flood season from May to September. The vegetation cover along the river is diverse and abundant (Yu et al., 2007) reflecting a heterogeneous pattern of various soil types (Guo et al., 2015) with the highest soil organic carbon (SOC) content in the Jinshajiang subbasin (Figure S1). SOC content has been increasing during the past two decades due to improved land-use management and increasing temperatures and precipitation (Guo et al., 2015; Kong et al., 2020).

2.2. Sampling, experiments and analytical methods

The methods employed in this study include water sampling and incubation experiments to analyze decomposition rates and dissolution of C, nitrate, and phosphate, and the analytical techniques to determine C and stable C isotope concentrations.

2.2.1. Sampling

Water samples were collected in April 2017 (low-flow season) and July 2018 (flood season) at a range of stations (1–11) to obtain longitudinal profiles along the Changjiang River (Fig. 1). To study the seasonality, monthly samples were taken at station 11 (Jiangyin) in the lower reach of the Changjiang River in 2013–2014 and 2018–2019. High turbidity can shrink the thickness of the photic zone (Cloern, 1987; Cole and Cloern, 1987). To make sure water samples come from the photic zone and avoid the pollution caused by vehicle emission, all water samples were collected at 0.2-m depth horizontally in the middle of the river (i.e. of the same distance from both river sides), and filtered in situ through pre-ashed (at 450 °C for 12 h) glass fibre filters (Whatman, 0.7-μm pore size, 47 mm diameter). The filters were folded and stored at −20 °C before analyzing TSS, element (C), C-isotope ($\delta^{13}C$) and chlorophyll-*a* (Chl-*a*) in the laboratory.

2.2.2. Decomposition incubation experiment

A laboratory incubation experiment using unfiltered water sampled in Jiangyin station was performed in August 2018: (1) to estimate the removal efficiency (decomposition + dissolution) of POC_{labile} (POC_{tot} – POC_{res}) and (2) to determine the C:N:P ratio of this POC_{labile}. Pre-ashed quartz flasks were filled with 2 L of sampled water and cultured at 25 °C for 15 days in the dark. One quartz flask was taken out from the incubation environment every other day. A 1000 mL well-mixed water sample was filtered through pre-combusted (at 450 °C for 12 h) glass fibre filters (Whatman, 0.7 μM, 47 mm). Filtered water samples, poured in an acid-cleaned polyethene bottle, were stored at −20 °C for dissolved nutrient measurement.

2.2.3. Experiment analyses

Prior to determination of C and $\delta^{13}C$ content, labile OC was removed from POC_{res} by oxidation with 30% of H_2O_2. POC_{tot} and POC_{res} filter samples were then acidified with 1 $mol \cdot l^{-1}$ HCl to remove inorganic carbonate and then oven-dried at 45 °C for 24 h (Li et al., 2015). The dried filters were packed into solvent-cleaned tin boats. Elemental and stable isotopic analyses were performed simultaneously with a Carlo Erba NA 1500 Series elemental analyzer (Fisons Instruments, USA) interfaced with a DELTA PLUS^{xp} continuous flow isotope ratio mass spectrometer (Thermo Finnigan Instruments, USA). The precision of lab standards (IVA99995 for POC content and USGS40 for $\delta^{13}C$) was ±0.02% for POC contents and ±0.1 ‰ for $\delta^{13}C$ ($n = 6$). The results of $\delta^{13}C$ were reported using the conventional delta notation relative to Vienna Pee Dee Belemnite (VPDB).

Nitrate (NO_3^-), ammonium (NH_4^+) and phosphate (PO_4^{3-}) concentrations were determined using a nutrient auto-analyzer (SEAL Analytical Gmbh, QuAAtro, Germany) with a precision >0.3%. Chl-*a* was determined using the acetone extraction method (Lorenzen, 1967). Filters were extracted in 10 mL of 90% acetone overnight. The absorbance of the sample was measured at 750-nm and 650-nm wavelength using Trilogy (Turner Design, 10-AU, USA) before and after acidification by 0.3 $mol \cdot l^{-1}$ HCl.

2.3. Modelling POC delivery and export

2.3.1. Modelling allochthonous and autochthonous POC

The yearly delivery of allochthonous POC from the terrestrial biosphere and production of autochthonous POC in the river channel were simulated at the spatial scale of ten sub-basins and aggregated to the main stream (divided into upstream, midstream and downstream reach) (Fig. 1). Soil erosion is a primary source of allochthonous POC in the Changjiang River (Wang et al., 2012; Yu et al., 2011). Estimation of allochthonous POC was therefore based on soil erosion and associated OC contents in different soil types (Figure S1, Table S1). The average SOC content of each sub-basin was calculated using the weighted average of SOC contents in different soil types and their corresponding areas:

$$SOC_j = \frac{\sum (SOC_i/100 \times A_{ij})}{A_j} \tag{1}$$

Where SOC_j and SOC_i represent the average contents of SOC (%) in sub-basin *j* and the OC content in soil type *i*, respectively; And A_j and A_{ij} are the area (km^2) of sub-basin *j* and occupied area of soil type *i* in sub-basin *j*.

We linked the sediment erosion and deposition fluxes from recent reports (Yang et al., 2014; Yang et al., 2007), the weighted average SOC contents in different soil types and their corresponding areas to estimate POC input from erosion:

$$C\_SERO_j = S\_SERO_j \times (SOC_j + q_e \times 10)/100 \tag{2}$$

Where C_SERO_j and S_SERO_j represent the POC and sediment fluxes ($Mt \cdot yr^{-1}$) from soil erosion in sub-basin *j*; other equations we used to compute the allochthonous POC budget in the whole river basin are listed in Text S1.

We quantified the adsorption of OC using the approach developed by Wu et al. (2020):

$$q_e = k_{ad} \times TSS^{-0.610} \tag{3}$$

Where q_e is the OC adsorbed on suspended sediment at equilibrium ($mg \cdot g^{-1}$); TSS is the concentration of TSS ($mg \cdot l^{-1}$). k_{ad} is the empirical parameter (unitless) with assumed maximum value of 3.64 based on particle size, particle density, surface density and particle morphology, DOC concentration and pH value in the Changjiang River (Bao et al., 2015; Wang et al., 2012; Wu et al., 2020). We thus obtain an uncertainty range of $0 < k_{ad} < 3.64$.

Autochthonous POC production was simulated on the basis of the modelling approach developed by van Hoek et al. (2021) based on environmental variables including temperature, irradiation, water discharge and TSS concentration:

$$C_{npp} = C_{pp} \times (t_{re})^{k_{re}} \tag{4}$$

$$C_{pp} = \left(f_{pp}(T) \times I_{lim} \times DIC_{lim} \times k_{pp} - k_{res} - k_{em}\right) \times t_{re} \times \mathrm{TALG} \times Q_{lower} \tag{5}$$

Where C_{pp} represent the POC flux originating from riverine primary production (Mt C yr^{-1}); C_{npp} represent the remainder of C_{pp} after deducting the rapid removal part by dissolution and decomposition; $f_{pp}(T)$ is the temperature-dependent function for riverine primary production (-) (Billen et al., 1994); I_{lim} is the light limitation (-). DIC_{lim} (-) is the DIC limitation of photosynthesis calculated with DIC concentration ($mol \cdot l^{-1}$) and half-saturation constant of DIC (k_{DIC}). k_{pp}, k_{res} and k_{em} are the maximum photosynthesis, respiration and emission rates (d^{-1}). TALG represents the initial primary biomass in situ ($mg \cdot l^{-1}$). The variablet_{re} is the water travel time (d). Q_{lower} is the annual water discharge ($l \cdot yr^{-1}$) obtained from the hydrological yearbook. Values of these parameters and data sources are listed in Table S2.k_{re} is the rapid removal function of POC_{labile} in the Changjiang River (unitless, see 3.1.4). Details about the calculation of $f_{pp}(T)$, I_{lim}, DIC_{lim} and the autocthonous POC budget in the whole river basin are listed in Text S2.

2.3.2. Modelling POC fluxes to the Changjiang estuary

Using the monitoring data of monthly total POC (POC_{tot}) and recalcitrant POC (POC_{res}) concentrations and daily water discharge for Datong station (http://www.cjw.gov.cn/), we applied the LOAD ESTimator (LOADEST) to calculate the monthly and yearly POC_{tot} and POC_{res} fluxes to the estuary. Based on a time series of streamflow and constituent concentration, LOADEST can estimate constituent loads in rivers

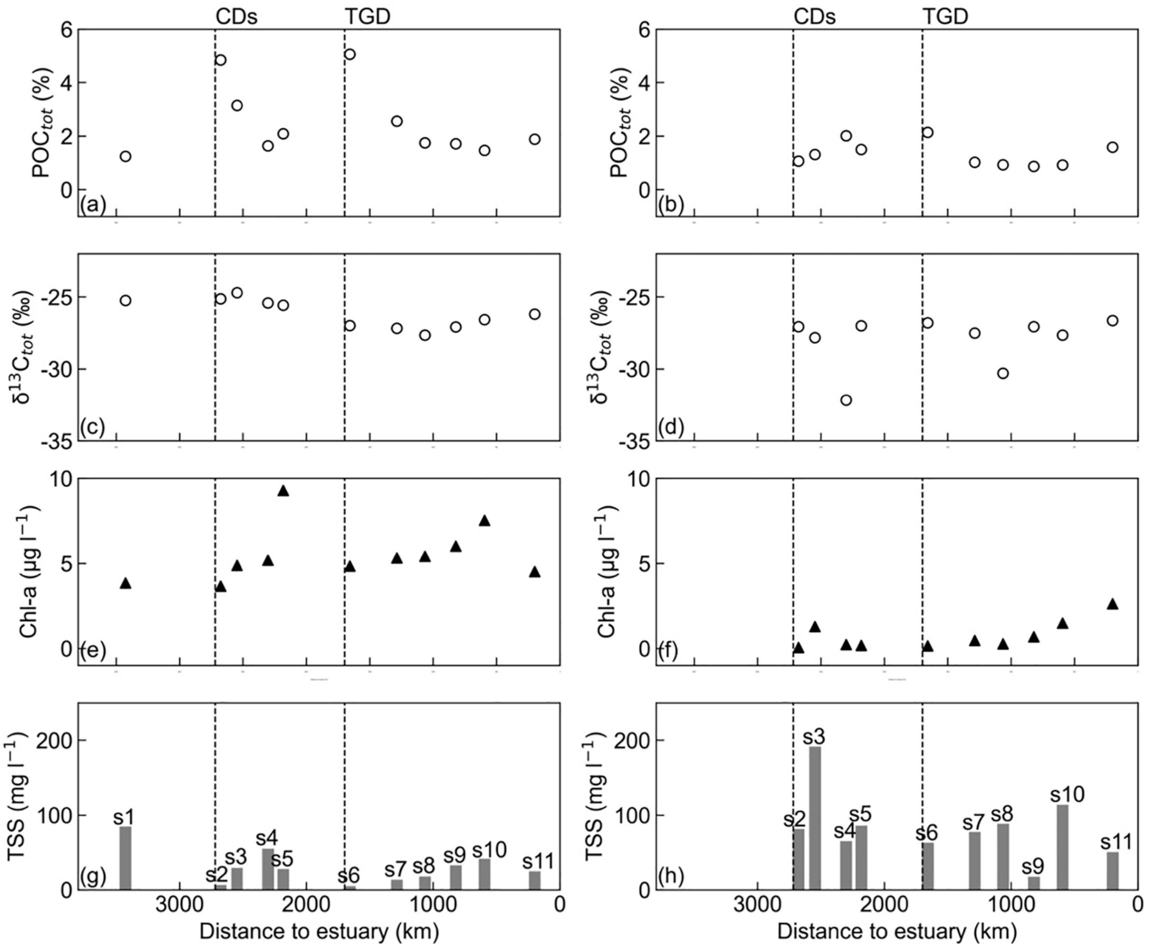

Fig. 2. Spatial distribution of POC content in TSS and concentrations of δ ^{13}C, and Chl-*a* and TSS in the main branch of the Changjiang River.

through developing a regression model (Runkel et al., 2004) by adjusted maximum likelihood estimation (AMLE) including the uncertainty at the 95% confidence level.

3. Results

3.1. Observed POC concentrations and fluxes

3.1.1. Spatial distribution of POC, δ ^{13}C, Chl-a and TSS along the Changjiang River

In April 2017, the TSS concentration in the mainstream of the Changjiang River ranged from 4.3 to 83.9 mg·l^{-1}, with an average value of 30.3 mg·l^{-1} (Fig. 2). The highest value occurred at station 1 located in the Jinshajiang River. Along the river continuum, the first low value of TSS concentration occurred at station 2 downstream of the CDs, and the TSS concentration gradually increased from station 2 to station 4, the head of Three Gorges Reservoir. The TSS concentration reached the lowest value at station 6 and from there it gradually increased downstream along the river. In August 2018, the TSS concentration ranged between 17 and 191 mg·l^{-1} and maintained a high level without clear differences between the stations.

In April 2017, the POC$_{\text{tot}}$ content in the mainstream of the Changjiang River ranged from 1.2% to 5.1% with an average of 2.5%. High POC$_{\text{tot}}$ contents observed at stations 2 and 6 correspond to low TSS concentrations (Fig. 2a).

In July 2018, the content of POC$_{\text{tot}}$ averaged 1.3% and ranged from 0.9% to 2.1%. The POC content was lower in July 2018 than in April 2017 at nearly all stations (Fig. 2b). The difference between the stations in the POC$_{\text{tot}}$ content in July 2018 was small.

In April 2017, the δ ^{13}C$_{\text{tot}}$ values varied from −27.60 ‰ to −24.70 ‰, with high values mainly at the upstream stations (Fig. 2c). However, in July 2018 the range of δ ^{13}C$_{\text{tot}}$ values was –32.15 ‰~–26.63 ‰ (Fig. 2d).

The Chl-*a* concentration in the Changjiang River ranged between 3.7 and 9.3 μg·l^{-1} in April 2017 and was much lower (0.06–2.64 μg·l^{-1}) in August 2018 (Fig. 2e & f). The Chl-*a* concentration was generally higher in the low-flow season than in the flood season. The highest value in the low-flow season was at station 5 in the upstream reach of the TGD. The Chl-*a* concentration gradually increased downstream towards the estuary both in the flood and low-flow season.

3.1.2. Seasonal variation in POC content, δ ^{13}C and TSS in the lower reach of the Changjiang River

According to the monthly investigations during two hydrological periods from May 2013 to May 2014 and from May 2018 to August 2019 (Fig. 3a), the POC$_{\text{tot}}$ content in the downstream reach of the Changjiang River ranged from 1.1% to 4.6% with an average of 2.4% (Fig. 3b). During the whole sampling period, there was an inverse pattern between the monthly POC$_{\text{tot}}$ content and water/sediment discharge. The observed δ ^{13}C$_{\text{tot}}$ value ranged from −26.97 ‰ to −25.54 ‰. The POC$_{\text{res}}$ content varied from 0.3% to 1.5% with an average of 1.0% during the period from May 2018 to August 2019. The variation of the δ ^{13}C$_{\text{res}}$ was small (from −27.03 ‰ to −25.84 ‰) similar to that of the δ ^{13}C$_{\text{tot}}$ (Fig. 3c).

3.1.3. POC export to coastal waters

Based on the monthly TSS fluxes and POC contents in the downstream reach of the Changjiang River, we calculated the monthly POC

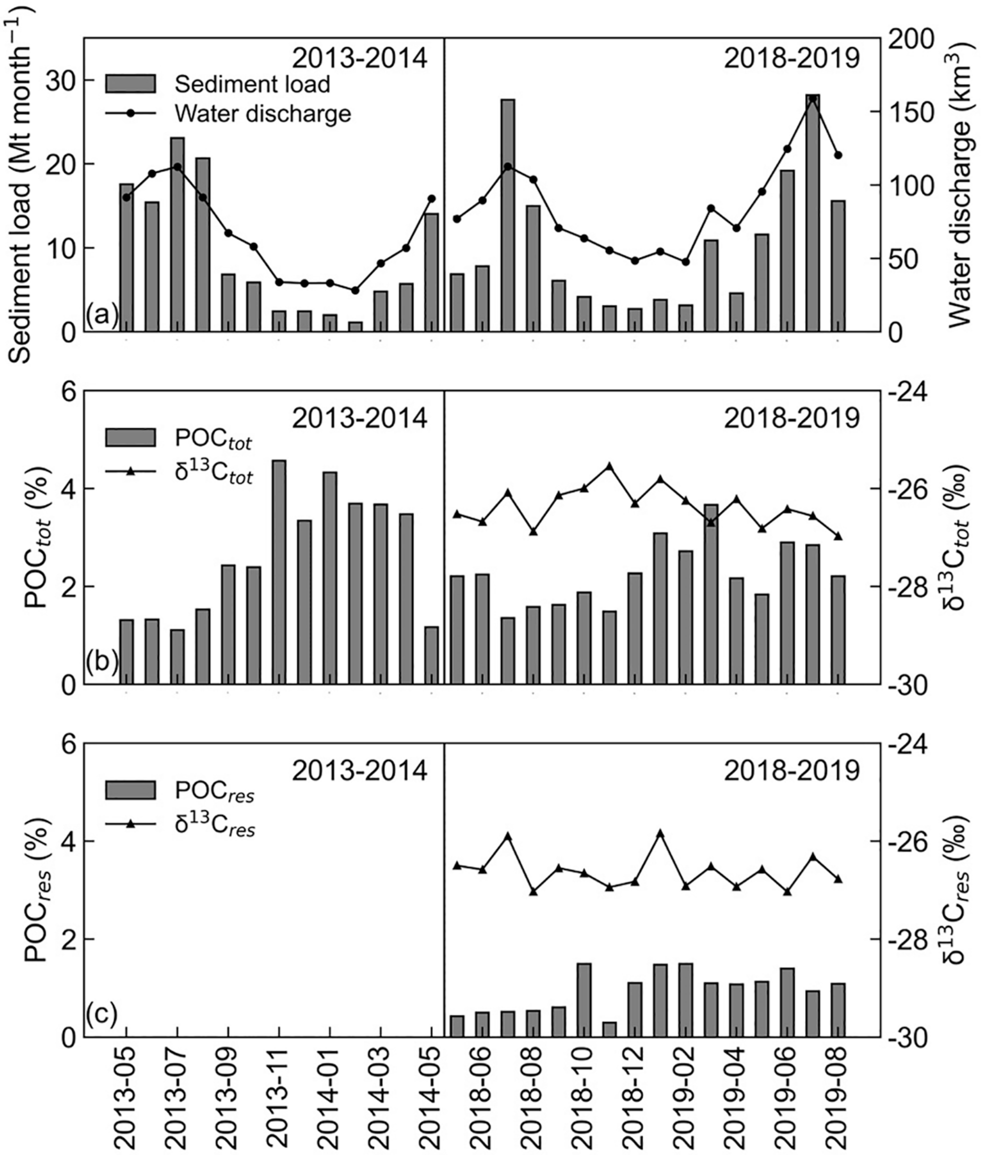

Fig. 3. Monthly water/sediment discharge, POC content of TSS and δ ^{13}C value in the downstream reach of the Changjiang River during May 2013-May 2014 and May 2018-August 2019.

fluxes of the Changjiang River during 2013–2014 and 2018–2019. The monthly fluxes of POC$_{tot}$ in the Changjiang River ranged from 0.04 Mt C month^{-1} to 0.8 Mt C month^{-1} and were higher during the period May-August (Table 1), similar to the monthly variations in the sediment flux. The calculated annual fluxes of POC$_{res}$ during May 2018 to April 2019 and during September 2018 to August 2019 contributed 37% and 50% to the POC$_{tot}$ fluxes of that year, respectively.

3.1.4. POC decomposition

The POC$_{tot}$ concentration decreased with time, with a sharp drop from 96 μmol·l^{-1} to 73 μmol·l^{-1} in the first four days and a slow decrease afterwards (Fig. 4). After the 15th day of incubation, the POC$_{tot}$ concentration was constant at 66.3 μmol·l^{-1}, close to the POC$_{res}$ concentration in Jiangyin station (61.5 μmol·l^{-1}). The remineralization rate of POC$_{labile}$ was 7.6 μmol·l^{-1}·day^{-1} during the period from day 0 to day 4 and 0.67 μmol·l^{-1}·day^{-1} during days 5–15. The high correlation coefficients ($r > 0.8$) indicated that the loss rate of POC$_{labile}$, and the variation in PO_4^{3-}, NO_3^- and NH_4^+ are strongly correlated. On the basis of the declining of NH_4^- and POC and the parallel increase of PO_4^{3-} and NO_3^- in the incubation experiment, we calculated that the ratio of C (POC): N ($NO_3^- + NH_4^+$): P (PO_4^{3-}) for removed organic material ranged from 83:6:1 to 83:13:1. Based on the incubation experiment, we estimated a value of −0.14 for parameter k_{re} for the rapid removal (dissolution + decomposition) function in the Changjiang River (see equation (4); Fig. 4a).

3.2. Simulated and observed POC$_{tot}$ inputs, trapping and export

The simulated average POC$_{tot}$ flux (sum of allochthonous and autochthonous POC) at Datong station is 2.7 ± 0.7 Mt C yr^{-1} during 1956–2002 (Fig. 5a). This number decreased to 0.9 ± 0.1 Mt C yr^{-1} during 2003–2009. After 2010, the POC$_{tot}$ flux at Datong station was stable and increased by ~20% relative to the average value of the period 2003–2009, with an average annual value of 1.1 ± 0.2 Mt C yr^{-1}, due to declining allochthonous and increasing autochthonous POC.

Comparison of the simulated POC$_{tot}$ fluxes at Datong station and observed POC$_{tot}$ fluxes at Datong and Jiangyin stations (Fig. 5a) shows that the model properly describes the observed POC$_{tot}$ fluxes before 2012 (all fall into the 95% confidence interval), and the spatial distribution of simulated Chl-*a* concentration in the mainstream is also

Table 1
Monthly POC_{tot} and POC_{res} fluxes in the Changjiang River.

	Sampling Date	POC_{tot} flux(Mt C month^{-1})	Contribution to annual (%)	POC_{res} flux (Mt C month^{-1})	Contribution to annual (%)
2013–2014	May-13	0.22	10.4	NA	NA
	Jun-13	0.21	9.53	NA	NA
	Jul-13	0.25	11.5	NA	NA
	Aug-13	0.31	14.3	NA	NA
	Sep-13	0.17	7.74	NA	NA
	Oct-13	0.14	6.38	NA	NA
	Nov-13	0.11	5.20	NA	NA
	Dec-13	0.08	3.81	NA	NA
	Jan-14	0.09	4.07	NA	NA
	Feb-14	0.04	1.97	NA	NA
	Mar-14	0.18	8.24	NA	NA
	Apr-14	0.20	9.21	NA	NA
	May-14	0.17	7.65	NA	NA
2018–2019	May-18	0.15	7.84	0.03	1.53
	Jun-18	0.18	9.11	0.04	2.02
	Jul-18	0.38	19.5	0.14	7.44
	Aug-18	0.24	12.3	0.08	4.19
	Sep-18	0.10	5.15	0.04	1.93
	Oct-18	0.08	4.07	0.06	3.24
	Nov-18	0.05	2.36	0.01	0.47
	Dec-18	0.06	3.21	0.03	1.57
	Jan-19	0.12	6.14	0.06	2.94
	Feb-19	0.09	4.45	0.05	2.45
	Mar-19	0.40	20.7	0.12	6.21
	Apr-19	0.10	5.18	0.05	2.58
	May-19	0.21	7.34	0.13	4.51
	Jun-19	0.56	19.1	0.27	9.23
	Jul-19	0.80	27.6	0.27	9.12
	Aug-19	0.35	11.9	0.17	5.85

NA: no data available.

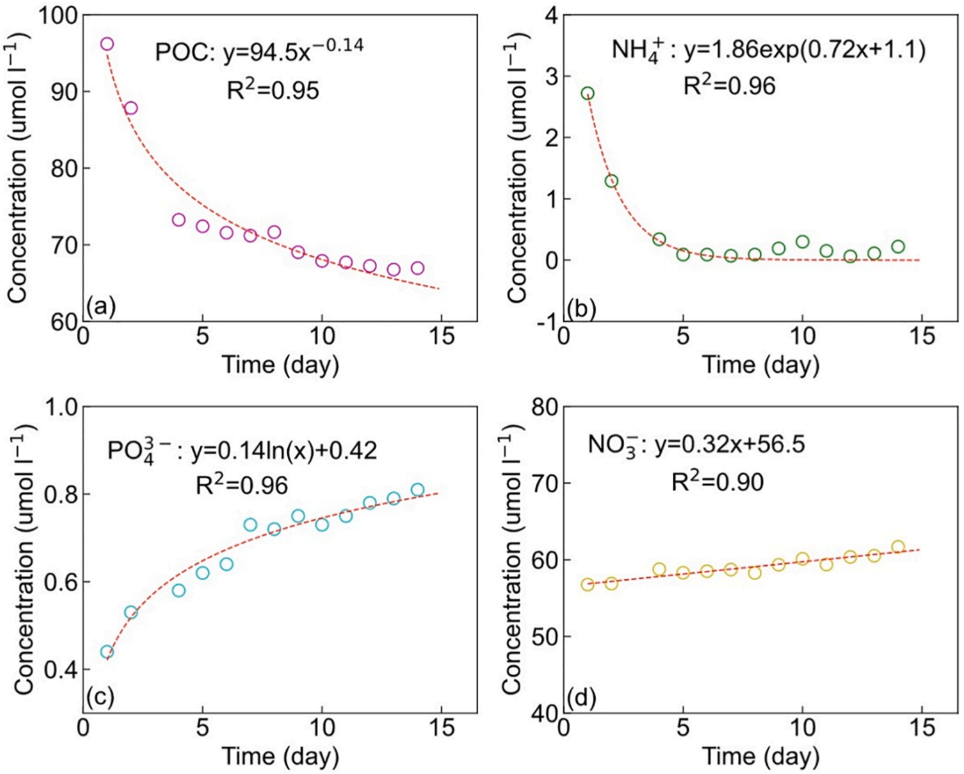

Fig. 4. Change of POC (a) and nutrients (b, NH_4^+; c, PO_4^{3-}; d, NO_3) during the laboratory incubations.

consistent with the observations (Fig. 5b).

The long-term changes in the POC flux and its spatial distribution is related to changes of its composition and delivery patterns in the Changjiang River during different historical periods (Fig. 6). POC_{tot} trapping in the upstream reach increased from 2 to 5% before 2003 to 66–88% during the post-TGD period. Since the autochthonous POC production further increased, the proportion of autochthonous POC also increased. Before 2003, the fraction autochthonous of total POC was

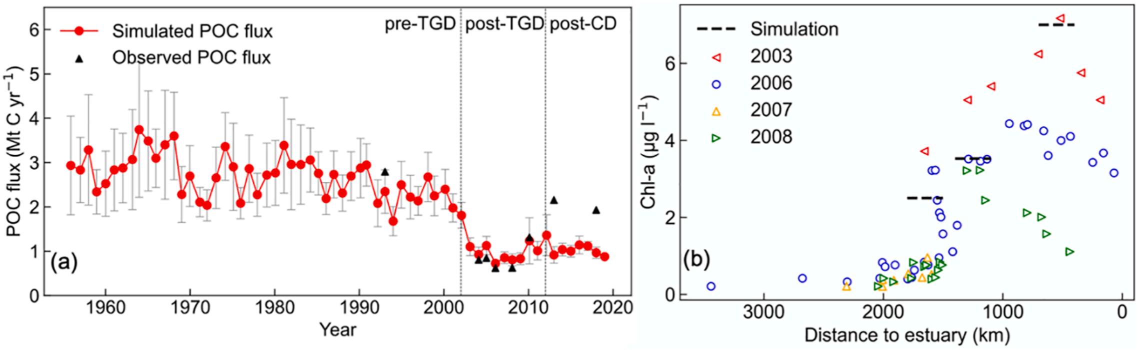

Fig. 5. (a) Long-term changes in POC_{tot} flux at Datong station in the Changjiang River (Error bars indicate uncertainty ranges as described in Section 2.3. Observed POC fluxes at the river mouth (Datong and Jiangyin) were obtained from Cui et al. (2009), Lin (2007), Wang et al. (2011), and Xu et al. (2011)); (b) Simulated and observed Chl-a concentrations distributed along the Changjiang River (Dashed lines are the average of the simulation result during 2000-2009; Chl-a observations for 2003, 2006, 2007 and 2008 are from Ran et al. (2013); Yu et al. (2011); Zhang et al. (2014)).

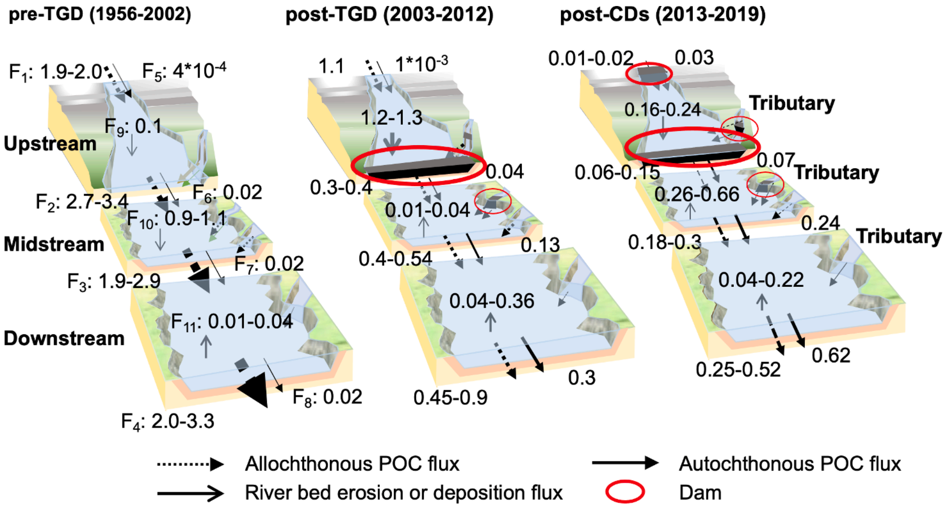

Fig. 6. POC delivery (in Mt C yr^{-1}) in the Changjiang River basin during different periods.

<2% in the whole river basin, while this fraction increased to 10%–13%, 25%–27% and 26%–41% in the upstream, midstream and downstream reaches of the Changjiang during the period 2003–2012, respectively.

POC transport from the Jinshajiang subbasin decreased by 80–88% in the post-CDs period compared to the years between 2003 and 2012. The combined retention in TGD and CDs aggravated the riverbed erosion intensity in the midstream reach. POC_{tot} export from the whole river basin to coastal waters declined by 1.7 ± 0.6 Mt C yr^{-1} from 1956 to 2002 to 2013–2019, but also altered the POC_{tot} composition from allochthonous-dominant to equal proportions of allochthonous and autochthonous POC. Observed ratios of POC_{labile}/POC_{tot} at Jiangyin station during May 2018 to April 2019 (63%) and September 2018 to August 2019 (50%) (Table 1) are close to our simulated range of autochthonous POC/POC_{tot} ratio (46%–71%) for the same period (Fig. 6) and incubation experiments (Fig. 4).

3.3. Simulated allochthonous and autochthonous POC

3.3.1. Simulated allochthonous POC supply and trapping

During the years prior to the TGD closure (i.e. before 2003), the simulated allochthonous POC flux at Yichang (representing the outflow from the upstream reach), Hankou (outflow from midstream to downstream) and Datong (downstream) stations (Fig. 1) was 3.1 ± 0.4, 2.5 ± 0.5 and 2.7 ± 0.7 Mt C yr^{-1}, respectively. About 97% of the allochthonous POC was transported from upstream to the mid- and downstream reaches of the Changjiang. However, the model suggests that after the TGD closure the allochthonous POC flux decreased by 89%, 82% and 75% at Yichang, Hankou and Datong stations, respectively, due to trapping in the TGR (Fig. 7).

According to the model, the Jinshajiang subbasin was the major source of allochthonous POC before 2012 due to its high sediment load and SOC content (Dai and Lu, 2014; Wu et al., 2003). However, after the operation of the CDs in 2012, almost no allochthonous POC was exported from the Xiangjiaba reservoir (Fig. 7d). As a consequence, after 2012 large masses of allochthonous POC that would otherwise be trapped in the TGR were actually deposited upstream of the TGR. The allochthonous POC fluxes at Yichang, Hankou and Datong stations further decreased by 70%, 48% and 43%, respectively, between the periods 2013–2019 and 2003–2012 (Fig. 7a–c). The model results indicate that compared to the allochthonous POC flux during the period 1955–2003, on average approximately 85% (2.3 ± 0.5 Mt C yr^{-1}) of allochthonous POC has been trapped within the Changjiang basin since 2013.

3.3.2. Simulated autochthonous POC supply

After the closure of TGD in 2003, the annual total autochthonous POC load in the upstream, midstream and downstream reaches increased by over 2, 7 and 15 times compared with the average values for the period 1960–2002, respectively (Fig. 8a). Correspondingly, the net autochthonous POC fluxes from the upstream, midstream and downstream reaches increased from 0.02, 0.02 and 0.02 Mt C yr^{-1} during 1960–2002 to 0.04, 0.13 and 0.30 Mt C yr^{-1} after 2003,

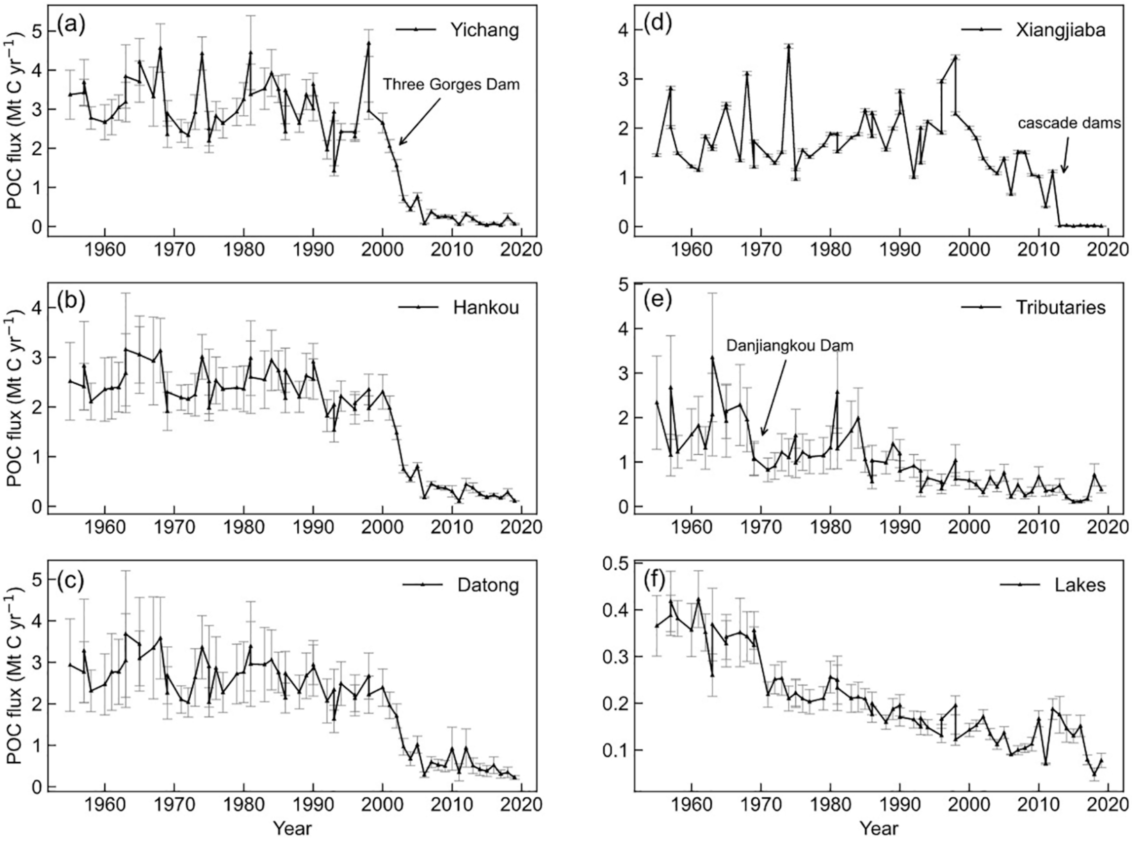

Fig. 7. Simulated allochthonous POC fluxes at different stations in the Changjiang River basin for the period 1956–2019.

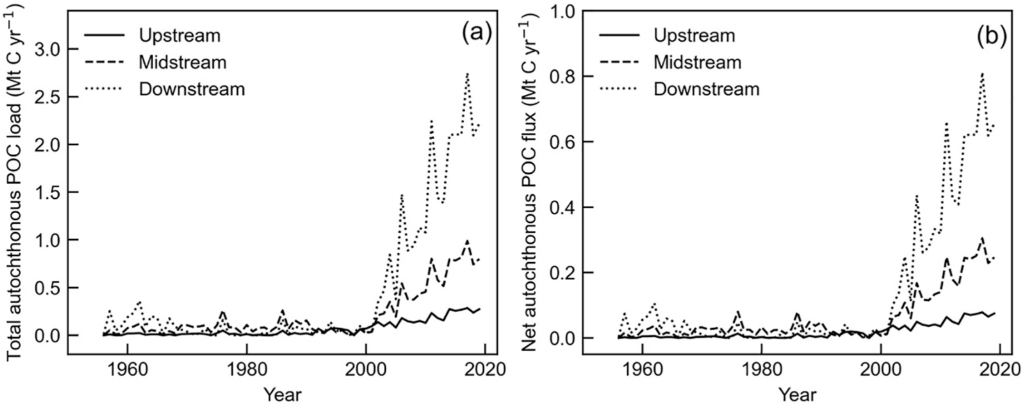

Fig. 8. Simulated autochthonous POC load and net transport fluxes in the upstream, midstream and downstream reaches of the Changjiang River basin for the period 1956–2019.

respectively (Fig. 8b). After 2013, the autochthonous POC production in the downstream reach of the Changjiang River further increased by more than a factor of 20 compared to the average value before 2003 (Fig. 8).

4. Discussion

4.1. *Characterization of POC export from the Changjiang River*

POC and TSS flux are closely related (Fig. 9a and c). A positive correlation between POC and TSS flux is clear during the whole time period, with two significant changes: (1) POC fluxes decreased sharply after 2003; (2) the POC_{tot} export flux from the Changjiang River has increased during the recent decade. As a result, the proportion of POC_{tot} in sediment export gradually increased from < 0.5% to 2% since 2003 (Fig. 9b).

4.1.1. *Decline of POC export*

Our model results show that the longitudinal pattern of the simulated allochthonous POC was in line with that of the observed TSS (Dai and Lu, 2014; Wu et al., 2003) (Fig. 7a–d). The sediment discharge in the upstream and downstream reaches decreased by 96% and 74% in 2006–2017 compared to 1950–1985, respectively (Guo et al., 2019). Nearly all TSS was trapped in the upstream reach after the operation of TGD and CDs (Guo et al., 2018; Yang et al., 2018). Our model suggests that the rapidly decreasing allochthonous POC flux dominates the

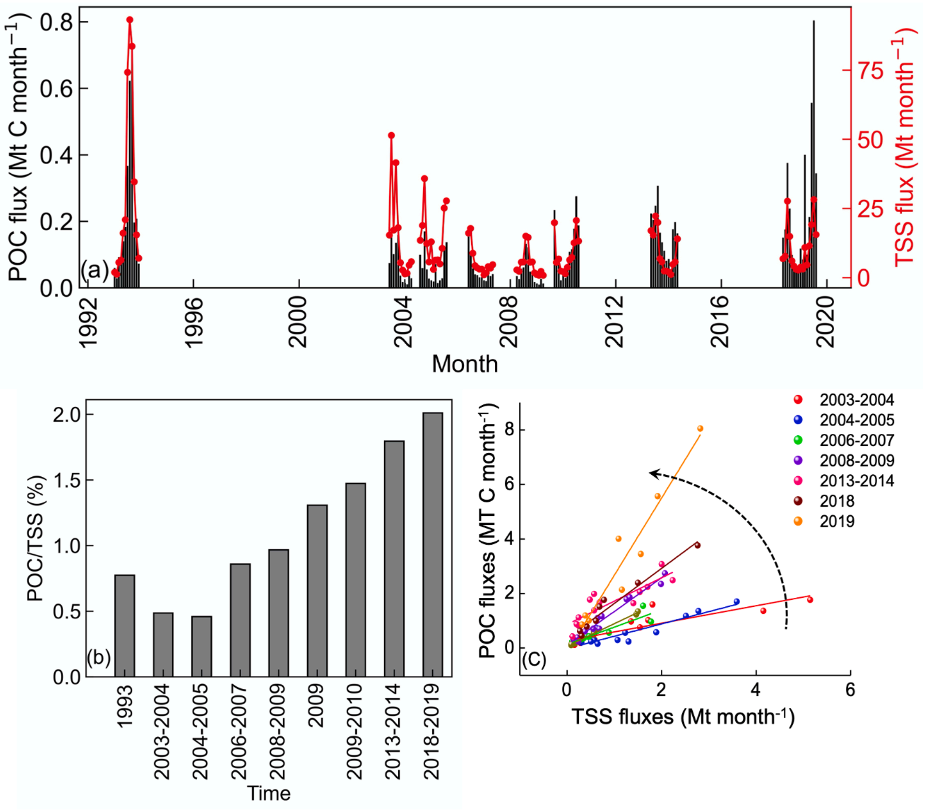

Fig. 9. (a) Long-term monthly variations in POC_{tot} and TSS fluxes at Datong and Jiangyin station of the Changjiang River during 1993–2019, (b) proportions of POC_{tot} in the annual TSS flux during 1993–2019 and (c) relationships between monthly TSS flux and POC_{tot} flux during 2003–2019. Data of monthly POC_{tot} and TSS fluxes in 1993, 2003-2005, 2006-2007 and 2008-2009 were obtained from Cui et al. (2009), Lin (2007), Wang et al. (2011), and Xu et al. (2011) ; POC_{tot} and TSS fluxes in 2013-2014 and 2018-2019 were from this study (Fig. 3).

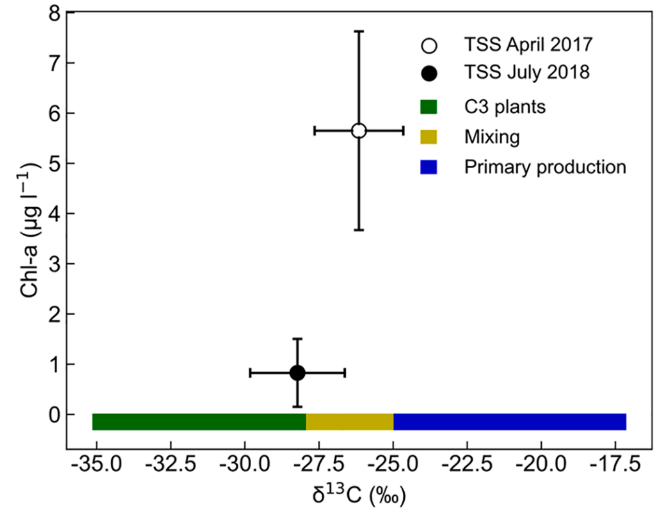

Fig. 10. δ^{13}C values of C3 plants in the Changjiang River basin, riverine primary production, and total suspended sediments and Chl-*a* concentrations in the water column (open and solid dots represent the ^{13}C values in the TSS of the Changjiang River in the low-flow season (April 2017) and flood season (July 2018), respectively). Data of ^{13}C in C3 plants and primary production were obtained from Huang et al. (2003) and Li et al. (2012); data of the ^{13}C values and Chl-a concentrations in the Changjiang River in the low-flow season and flood season are from this study.

changes of POC_{tot} flux before and after 2003. In addition, the high trapping efficiency in the upstream reach caused a decline of transport, which induced a shift from a system with POC retention to one dominated by riverbed POC erosion in the midstream reach. Although riverbed erosion supplied 41% of the TSS export from the midstream reach (Yang et al., 2014), the re-suspended POC flux only added 16–23% of the POC_{tot} export from the midstream to downstream reach due to the low POC content of sediment in the riverbed (Fig. 6).

4.1.2. *Increased POC/TSS ratio and autochthonous POC*

The POC content of TSS generally decreases with increasing TSS concentrations (Meybeck, 1982a; Thurman, 2012) due to dilution in water with high sediment loads, and improved light transparency and production at low sediment loads. This is also shown in the Changjiang River. To understand the change of the POC/TSS ratio, we analyzed the differences in erosion intensity driven by different hydrological patterns. The $\delta^{13}C_{tot}$ range in the flood season (July 2018, Fig. 2d) is more negative than the $\delta^{13}C_{tot}$ range in the low-flow season (April 2017, Fig. 2c), and is within the δ^{13}C range of C3 plants in the Changjiang River basin (Fig. 10). The $\delta^{13}C_{tot}$ range in the low-flow season is consistent with the δ^{13}C range of the overlap of C3 plants and primary production (Mixing), and the higher Chl-a concentration is due to more primary production compared to the flood season (Fig. 10). This indicates that higher soil erosion rates in the flood season lead to more allochthonous POC compared with the low-flow season with a more strongly negative δ^{13}C signal stemming from terrestrial C3 plants. Higher turbidity (Fig. 2) limits algal growth, i.e. causes a decrease in autochthonous POC production. In contrast, a low erosion intensity would effectively enhance the production of autochthonous POC (Sullivan et al., 2001) especially with near-optimal temperature conditions in the low-flow season in April (Garnier et al., 1999; Zhong et al., 2016).

However, the comparison of the POC characteristics before and after reservoir filling indicates that damming has gradually become the dominant factor controlling water turbidity and POC. According to the spatial distribution along the mainstream of Changjiang River in different years, the POC_{tot} contents of TSS began to increase in the TGR area in the post-TGD period after 2003 (Fig. 11a). Despite the sharp decrease from midstream to downstream in 2006, 2008 and 2017, the POC/TSS ratio (>1.7) in the downstream reach was still higher than that in 1993 and 2003 (<1). This indicates that the increasing POC/TSS ratio

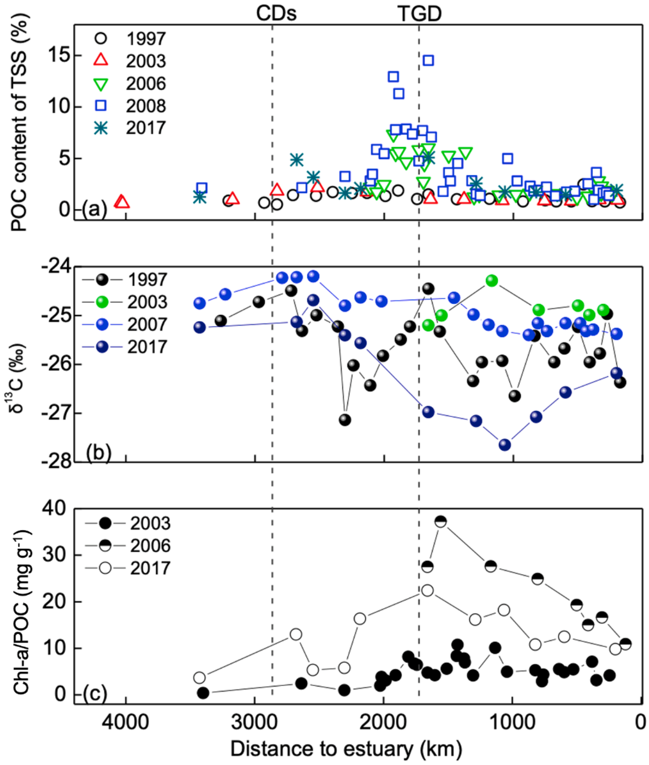

Fig. 11. Spatial distribution of POC_{tot} contents of TSS, $\delta^{13}C_{tot}$ values and Chl-*a*: POC_{tot} ratios along the continuum of the Changjiang River in the moderate-discharge period. POC_{tot} contents for 1997, 2003, 2006 and 2008 were obtained from Wu et al. (2007) and Zhang et al. (2014); $^{13}C_{tot}$ values in 1997, 2003 and 2007 were obtained from Wu et al. (2007), Yu et al. (2011) and Mao et al. (2011); data for Chl-*a*:POC_{tot} ratios in 2003 and 2006 were obtained from Yu et al. (2011) and Zhang et al. (2014); data for 2017 are from this study.

in the downstream reach is related to dam construction in the upstream reach. In 2006 and 2017, the Chl-a:POC_{tot} ratios in the midstream and downstream reaches are higher than those in 2003 (Fig. 11c), which indicates that the proportion of autochthonous POC increased in the midstream and downstream reaches after TGR filling. Moreover, the C:N:P ratio obtained from the incubation experiment (Redfield, 1963; Zhang et al., 2018) (Fig. 4), the approximation of observed ratios of POC_{labile}/POC_{tot} at Jiangyin station during May 2018 to August 2019 (Table 1) and the simulated autochthonous POC/POC_{tot} ratio for the same period (Fig. 6), all confirm that most of the labile POC is autochthonous POC in the Changjiang River. Reservoirs prolong the water residence time, create conditions prone to settling of material which decreases turbidity (He et al., 2017) and thus improve the light transparency, providing suitable conditions for primary production (Maavara et al., 2020; Maneux et al., 2001) as observed in the TGR area after the operation of TGD (Gao et al., 2018; Liu et al., 2012; Ran et al., 2013).

A drop of $\delta^{13}C_{tot}$ occurred downstream the TGD in 2007 and 2017 (Fig. 11b). Deposition of coarse particles, such as carbonate-interlayered black shale which is widespread in the Changjiang River basin (Jiang et al., 2012) with a less negative δ^{13}C (-21.5 ± 7 ‰) values compared to other POC sources in the river basin (Hartmann and Moosdorf, 2012; Li et al., 2015) (Fig. 10), could lead to declining $\delta^{13}C_{tot}$ in the water column. This is confirmed by the decrease in the mean particle size at Yichang station downstream of TGD (Fig. 1) from 8 μm in 2003 to 3 μm in 2008 (Dai and Lu, 2014), which points to intensive particle-size sorting in TGR. Therefore, dam construction, through increasing primary production and sedimentation of coarse particles, leads to an increase in the ratio of POC/TSS ratio in the downstream reaches of the Changjiang.

4.2. Uncertainties of POC fluxes

Li et al. (2015) estimated a trapping flux of POC_{tot} of 9.0 ± 2.8 Mt C yr^{-1} in the Changjiang basin after 2003, which exceeds our estimated POC_{tot} trapping of 2.1 ± 0.5 Mt C yr^{-1} during the same period by more than a factor of 4. Li et al. (2015) used the linear POC_{tot}-TSS relationship to estimate the reduction of POC_{tot} export from the Changjiang River to the estuary, which was regarded as the amount of POC_{tot} trapping in the river basin. However, the POC_{tot} content of TSS before 2000 used by Li et al. (2015) was based on two cruises per year in the Changjiang estuary with salinity > 5 (Cauwet and Mackenzie, 1993; Milliman et al., 1984b). Using higher POC content of TSS (~10%) in the estuary than in Jiangyin and Datong stations (Fig. 9) results in overestimation of POC trapping in the whole river basin. Moreover, combining our estimated POC_{tot} trapping with the silica (Si) retention in the Changjiang River basin during the same period reported by (Liu et al., 2020), we estimate a ratio of C:Si retention ranging between 1.7 and 9, which is within the ranges found for freshwater systems (Conley et al., 1989; Ran et al., 2017).

In our study, uncertainties of POC fluxes calculated from monitoring data in the downstream reach of the Changjiang largely depend on uncertainties of water discharge and POC content estimates associated with differences in the frequency of observations. The estimates in Table 1 are based on monthly POC concentration measurements and daily discharge, assuming that one single POC observation represents the monthly average. This ignores the temporal variability of the POC concentration between two observations across days/months/seasons. However, it is difficult to estimate the uncertainty due to sampling frequency without a statistical comparison of different measurement frequencies. For the model simulations, we calculated the uncertainty of POC flux from the uncertainty of OC adsorption on suspended sediment. However, our uncertainty neglects the effect of particle size change and other factors leading to the decline in TSS fluxes (e.g. afforestation, sand mining and climate change), and does not include uncertainties in the processes of production, decomposition and sedimentation.

4.3. Implications in inland and coastal waters

Dams have dramatically altered POC sources and composition in the Changjiang River, and thus changed the C export to the estuary and coastal waters (Fang et al., 2019; Liu et al., 2019; Xu et al., 2020b; Zhao et al., 2021a). Along with the declining allochthonous POC export by the Changjiang River, the decreasing POC burial has been observed in sediment cores in the Changjiang estuary (Li et al., 2016; Liu et al., 2019). A recent study reported that the terrestrial biomarker content in surficial sediment in the Changjiang estuary decreased by > 80% after the construction of TGD (Wang et al., 2020). Although the increase in autochthonous POC flux partly compensates for the increasing burial of allochthonous POC in reservoirs, most of this labile POC is not preserved due to rapid oxidation in the estuary (Galy et al., 2015; Ittekkot, 1988; Li et al., 2014). As a result, the function of the Changjiang estuary as a site for terrestrial OC burial has substantially decreased. With more C trapping in river basins and less C burial in coastal ecosystems, the spatial distribution of C burial shifted. Meanwhile, the previous estimate of terrestrial OC burial flux in the Changjiang estuary and the East China Sea inner shelf decreased by 1.64 ~ 1.78 Mt/yr from the pre-TGD to the post-TGD period (Deng et al., 2006; Zhao et al., 2021b), which is less than our estimation of terrestrial POC burial flux in the Changjiang River basin. Reduction of POC loss during POC transport in the downstream implies more efficient burial rate of terrestrial POC in the river basin (Battin et al., 2009; Li et al., 2015).

The decreases in POC and TSS export from the Changjiang River may also influence the in-situ C biogeochemistry related to primary production and decomposition in the coastal waters (Li et al., 2016; Zhao et al., 2021a). The reduction in TSS discharge creates better light conditions for photosynthesis, uptake of atmospheric CO_2 and oxygen production, which increases the capability of seasonal in-situ C fixation

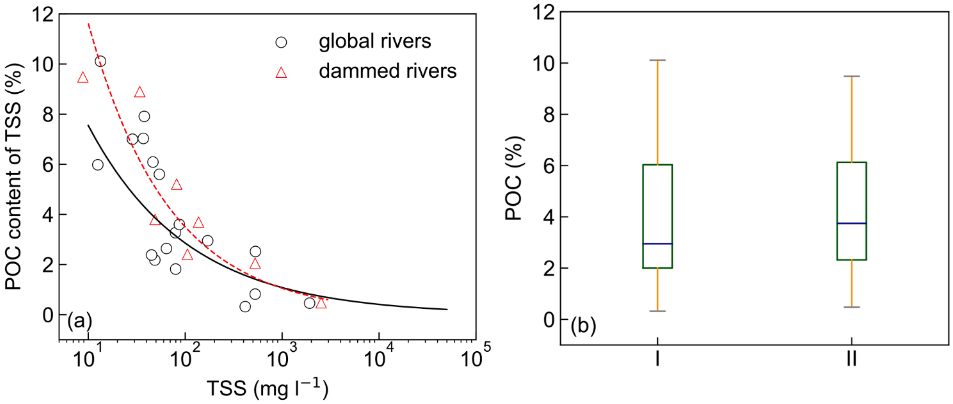

Fig. 12. The plot of POC_{tot} content of TSS versus TSS concentration (a) and boxplot of POC_{tot} content of TSS in global rivers lacking dams (I) and global rivers with dams (II) (b).

in the coastal ecosystem; the decrease in river POC export may reduce the oxygen consumption related to its decomposition (Chen et al., 2017; Dai et al., 2016; Li et al., 2016; Yang et al., 2011). The seasonal hypoxia and acidification in the coastal waters near Changjiang estuary may thus be relieved to some extent.

Many other large rivers in the world, including the Danube, Mississippi, Godavari, Chao Phraya, Krishna, Red, Yellow, Pearl and Indus Rivers (Panda et al., 2011; Walling, 2011; Wang et al., 2011) (Table S3), have experienced reduction in sediment discharge due to dam construction during the past decades (Walling and Fang, 2003). With drastically decreased sediment load in rivers with strong human interference in the hydrology due to dam construction, the POC-TSS relationship seems to have changed (Fig. 12a). The overall range of POC content of TSS does not change much, while the median value of POC content of TSS in global rivers with dams is 16% higher than those in global rivers lacking dams (Fig. 12b). Although the change of POC content is limited, it can have important impact on river POC export to the coastal waters. Based on the empirical relationship shown in Fig. 12, we estimated that total POC export from these nine intensively dammed rivers has declined by about 27.9 Mt C yr^{-1}, equivalent to 12% of the global riverine POC export (Blair and Aller, 2012; Kandasamy and Nagender Nath, 2016; Middelburg, 2019) (Figure S2), which includes the 3.4 Mt C yr^{-1} increase in the export of more labile allochthonous POC.

The increased autochthonous POC in the Yellow River and Krishnia River is negligible (contributed $< 1\%$ to the POC_{tot} flux) (Figure S2). The increase of autochthonous POC in the Indus River contributed 14% to the POC_{tot} flux which is less than that of the Red, Chao Phraya, Godavari, Mississippi, Danube and Pearl Rivers (21–35%; Figure S2). The current autochthonous POC export by the Changjiang River increased much more (50–70% of the POC_{tot} flux) during the past two decades. Hence, the impact of the dams in the Changjiang River is probably larger than in other rivers in the world, which may have profound consequences for biogeochemistry in the Changjiang estuarine ecosystems.

5. Conclusion and perspective

The combination of modelling and observations of POC and TSS fluxes in the Changjiang River basin reveals a dramatic impact of the construction of dams on POC export to the Changjiang estuary. About 90% of allochthonous POC is trapped within the river basin, while the export of more labile autochthonous POC has increased by a factor of 20 and makes up about half of the total POC flux in recent years. The decrease in total POC export and increase in the labile POC fraction in the export has reduced the sink strength for terrestrial OC of the Changjiang estuary. The decline of POC export by nine intensively dammed rivers is 12% of the global riverine POC export, with increasing fractions of labile OC. With more C trapping in river basins and less C burial in coastal ecosystems, the spatial distribution of C burial has been shifted.

Our estimation of POC trapping in reservoirs is uncertain. However, our analysis clearly shows that the changes of river POC transport and their impact on the estuarine C cycle in many large river basins are strongly affected by dams, whereby the influence of the dams on the C cycle in the Changjiang River is probably larger than in other rivers in the world.

With the projected increasing demand for renewable energy, dam construction for hydropower generation will continue worldwide (Grill et al., 2015; Mulligan et al., 2020), with major consequences for riverine and estuarine C cycles. These changes in the C cycle of river basins and estuaries need to be considered when assessing the impact of dams for hydropower generation, water storage and other purposes on the global C cycle.

CRediT authorship contribution statement

Hao Wang: Conceptualization, Formal analysis, Investigation, Methodology, Writing – original draft. **Xiangbin Ran:** Conceptualization, Investigation, Methodology, Writing – review & editing. **Alexander F. Bouwman:** Conceptualization, Methodology, Writing – review & editing. **Junjie Wang:** Conceptualization, Writing – review & editing. **Bochao Xu:** Writing – review & editing. **Zhaoliang Song:** Methodology. **Shaobo Sun:** Writing – review & editing. **Qingzhen Yao:** Writing – review & editing. **Zhigang Yu:** Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data Availability

The data that support this study are available in the Supplementary materials. Some are available from the corresponding author upon reasonable request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jhydrol.2022.127525.

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