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Mechanisms controlling dissolved CO_2 over-saturation in the Three Gorges Reservoir area

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ABSTRACT

The emission of CO_2 to the atmosphere from inland waters is an important part of the global carbon cycle. In this study, we made spatial and temporal measurements of CO_2 partial pressure (pCO_2) along the Three Gorges Dam system. The pCO_2 ranged from 619 to 2383 µatm and was supersaturated relative to atmospheric CO_2. Further, pCO_2 showed obvious spatial and temporal variations: pCO_2 at the high-flow season was much lower than that at the low-flow season near the upstream part of the reservoir, whereas pCO_2 in the reservoir water and after the dam showed an opposite seasonal trend. Organic matter mineralization produced more CO_2 in the surface water of the reservoir area at the high-flow season and should be responsible for the $\delta^{13}C$-depleted dissolved inorganic carbon ($\delta^{13}C_{DIC}$). In addition, organic carbon mineralization is sensitive to temperature variability, which is expected to be an important driver of the dissolved CO_2 over-saturation in the reservoir. This study suggested that the construction of Three Gorges Reservoir increased the water transit time and accelerated the organic carbon mineralization in the Changjiang River. The results indicate that carbon cycling changes markedly in large impounded rivers.

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KEYWORDS
organic carbon mineralization; pCO_2; $\delta^{13}C_{DIC}$; temperature sensitivity; Three Gorges Reservoir

Introduction

Inland waters link terrestrial and oceanic ecosystems by transporting materials from land to ocean (Barth et al. 2003, Wang et al. 2014a) and also exchange materials with the atmosphere (Kosten et al. 2010, Raymond et al. 2013). Although the fluvial carbon export by inland waters only occupies a small portion (10^{15} g yr^{-1}) of the global carbon cycle (Meybeck 1982, Aucour et al. 1999), it plays an important role in regional carbon cycling (Wang et al. 2014b). In the last few decades, the natural fluvial processes in many rivers have been disturbed by anthropogenic activities (Humborg et al. 1997, Raymond et al. 2008, Regnier, 2013, Guo et al. 2015, Liu et al. 2017), and the consequences of dam construction have been intensively studied (Humborg et al. 1997, Barros et al. 2011, Wang et al. 2011, 2013, 2014b, Bao et al. 2014). Impoundment converts a river into an "artificial lake" and consequently modifies the ecological function and biogeochemical processes of the inland waters. River regulation by dam construction has become an important environmental problem affecting greenhouse gas release from rivers, although hydropower is regarded as a "green energy" (Humborg et al. 1997, Chen et al. 2011, Wang et al. 2011). Enhanced dam construction in rivers has greatly changed the transport of sediments, dissolved silicon, and terrestrial organic carbon (Humborg et al. 1997, Yu et al. 2011, Bao et al. 2014, Yang et al. 2015). The construction of dams moderates the organic matter flux and composition downstream, and sediment trapping within a reservoir results in intense respiration, thus increasing the proportion of aquatic carbon as well as CO_2 emission from inland waters (Bao et al. 2014).

The Three Gorges Dam is the largest hydropower dam in the world. To assess its environmental effects, the ecological environment and biogeochemical processes in the Three Gorges Reservoir (TGR) have been widely studied (e.g., Bao et al. 2014, Zhang et al. 2014). Previous studies in TGR have estimated the changes in hydrology and sediment dynamics (Xu and Milliman 2009, Dai and Liu 2013, Yang et al. 2015, Deng et al. 2016, Li et al. 2016), biogeochemistry (Bao et al. 2014, Mao et al. 2017), and greenhouse gases emissions (Chen et al. 2011, Zhao et al. 2013). Few studies, however, have focused on the sources of the dissolved CO_2 and the relative biogeochemical processes in inland waters. Artificial reservoirs are known to be potential CO_2 contributors to the atmosphere

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(Raymond et al. 2013, Wang et al. 2015). Dissolved CO_2 over-saturation with respect to the atmosphere is the main driver of CO_2 emissions.

Multiple controlling factors have been proposed for CO_2 over-saturation in inland waters. Maberly et al. (2013) found that catchment productivity controls CO_2 emissions from lakes. Marcé et al. (2015) showed that carbonate weathering is a driver of CO_2 over-saturation in lakes. Inorganic carbon loading was regarded as a primary driver of dissolved CO_2 concentrations in lakes and reservoirs of the contiguous United States (McDonald et al. 2013). Ward et al. (2013) found that degradation of terrestrial macromolecules contributes significantly to CO_2 out-gassing from inland waters.

In this study, we investigated the temporal and spatial patterns of dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), particulate organic carbon (POC), the partial pressure of CO_2 ($p$$CO_2$), and stable carbon isotope of DIC ($\delta^{13}C_{DIC}$) in surface water of the TGR area. The objectives of the study were to (1) investigate the carbon dynamics in TGR, (2) trace the main sources of the dissolved CO_2 in TGR, and (3) understand the controlling mechanism of the dissolved CO_2 over-saturation in TGR.

Study site

TGR, the largest hydropower project in the world, is located between the upper and middle reaches of the Changjiang River, upstream of Yichang city in Hubei province (Fig. 1; Zhao et al. 2013, Deng et al. 2016). TGR is a narrow V-shaped valley-type reservoir with steep slopes along the river channel. Mountainous areas occupy up to 96% of the TGR area, with 4.3% plains only in the river valley (Zhao et al. 2013). TGR experiences a humid subtropical monsoon with an annual mean temperature of 18 °C (Mao et al. 2017). The local annual rainfall is ~1250 mm and occurs mainly from May to September (Mao et al. 2017).

TGR has been fully operated since the end of 2008 (Zhao et al. 2013). The water level ranges from 145 m at high-flow to control floods and 175 m at low-flow to retain water, with corresponding storage capacities of 17.2 and 39.3 km^3, respectively (Yang et al. 2014). High-flow season is defined as occurring May to October and low-flow season from November to April of the subsequent year, based on water regulation at TGR, according to the water level.

Methods

Six sampling sites (Qingxi [QX], Wanzhou [WZ], Zhongzhou [ZZ], Taipingzi [TPX], Huanglingmiao [HL], and Yichang [YC]) were chosen in the TGR area (Fig. 1), of which 4 (QX, TPX, HL, and YC) were chosen for long-term observation. QX is located near the inflow to the reservoir, WZ, ZZ, and TPX are located sequentially down-stream within the reservoir, and HL and YC are located downstream of the TGR discharge and Gezhou dams, respectively. We collected samples at QX monthly for 2 hydrological years and added extra sampling occasions during high-flow. TPX, HL, and YC were sampled monthly for a hydrological year, and additional samples were added during high-flow.

Water temperature (T), pH, and electric conductivity (EC) were measured directly at the time of surface water sampling using a portable EC/pH meter (WTW, pH 3210/Cond 3210, Germany). Water samples were collected in sealed high-density polyethylene bottles, and the alkalinity was measured by Gran titration with 0.02 M HCl within 24 h of sampling. The concentration of DOC was analyzed using an OI Analytical Aurora 1030 TOC analyzer. Total suspended solids (TSS) were trapped on a glass-fibre filter paper (0.7 µm, Whatman, GF/F) and then freeze dried and weighed. POC was measured with an elemental analyzer (PE2400 [II], Perkin Elmer) after acidification. The $\delta^{13}C_{DIC}$ was determined by the method of Li et al. (2010): 20 mL aliquots of water were purified with a precision of 0.2‰ on a vacuum line with 2 mL 85% phosphoric acid and magnetic stir bar. Daily water discharge and water level data were obtained online from the Ministry of Water Resources (http://www.hydroinfo.gov.cn/). The $p$$CO_2$ was calculated based on mass balance relationships and the relative equilibrium constants.

Results

Hydrological characteristics

Although the Changjiang River carries a tremendous volume of water, the Three Gorges Dam can moderate the downstream delivery of water. The water level ranged from 145 to 175 m a.s.l. during the study period (Fig. 1). The Changjiang River water was retained in TGR in the low-flow season, and the water level was maintained at a relatively high level to meet water navigation and hydropower requirements (Fig. 1). The water level was decreased to a low level from April to June to provide capacity for flood control (Fig. 1). From September to October, the water level was increased to impound water, and a high level was maintained during the dry season (Fig. 1). At QX, in the upper reaches of the reservoir, discharge varied from 4293 to 36 484 $m^3\ s^{-1}$ (average 10 242 $m^3\ s^{-1}$) from February 2015 to February 2016. Because of water regulation, discharge at HL and YC was

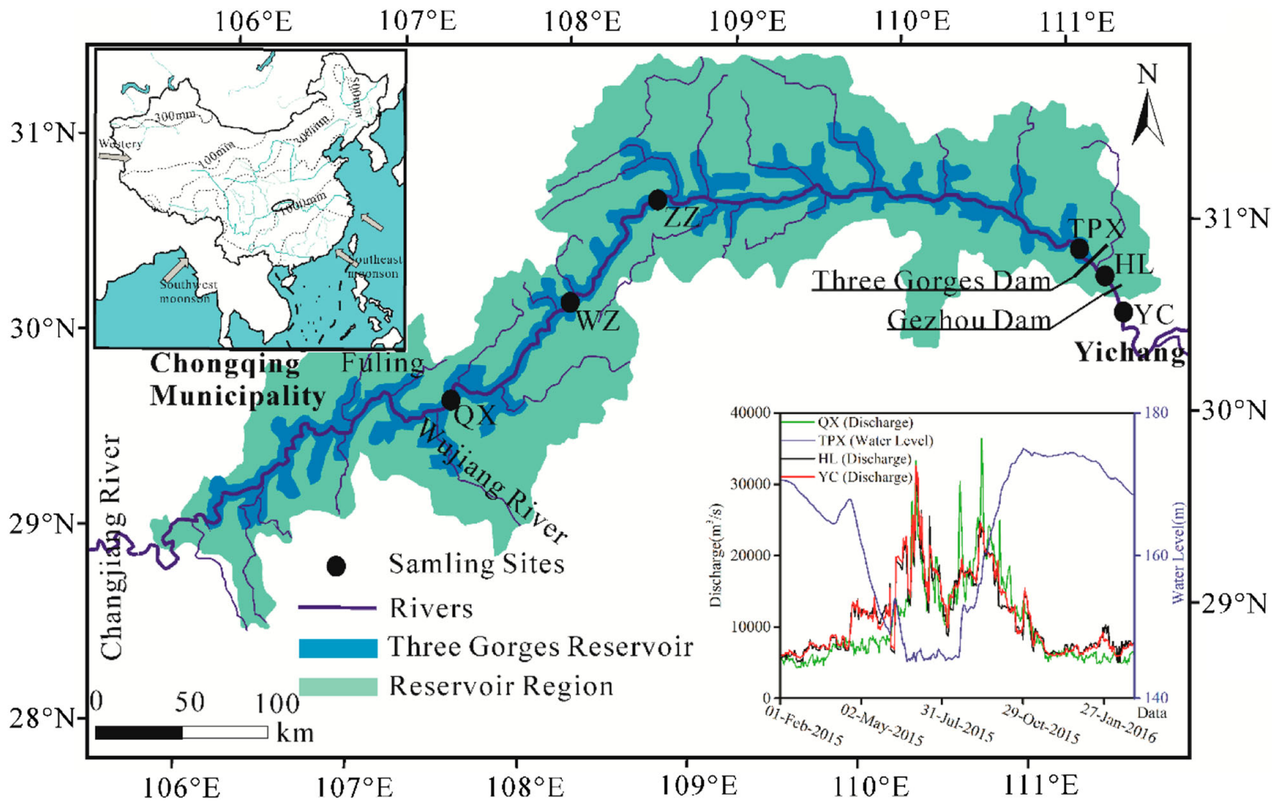

Figure 1. Sampling sites in the Three Gorges Reservoir area: Qingxi (QX), Wanzhou (WZ), Zhongzhou (ZZ), Taipingxi (TPX), Huanglingmiao (HL), and Yichang (YC). Upper inset: location of the region within China; lower inset: discharge at QX, HL, YC and water level at TPX from Feb 2015 to Feb 2016.

less variable, from 5050 to 31 800 $m^3 s^{-1}$ and from 5620 to 32 600 $m^3 s^{-1}$, respectively. The average discharge was not significantly different at the 3 hydrological stations, however, indicating that the contribution of other inflowing rivers was minor for the TGR area.

Variations of carbon species and $\delta^{13}C_{DIC}$ in TGR

Temporal variation in water temperature was much larger than spatial variation. Temperature varied temporally from 11.2 to 28.9 °C, with little variation in the surface water of the reservoir (TPX) and the discharged water at HL. This finding was markedly different from other reservoirs (Wang et al. 2014a), possibly because of the weak stratification in TGR (Wu et al. 2012). The pH values varied from 7.75 to 8.31 for all water samples, with little spatial variation. Conductivity varied from 295 to 410 µS cm^{-1}, with both the maximum and minimum values observed in QX. The more stable status of the other sites could be ascribed to the regulating effect of TGR. DOC ranged from 0.86 to 2.05 mg L^{-1}, again with lower spatial than temporal variation. Alkalinity ranged from 1.97 to 2.60 mequiv L^{-1}, and the alkalinity at QX was higher than at other stations. The pCO_2 ranged from 619 to 2383 µatm (Fig. 2a), and therefore all samples were supersaturated relative to atmospheric CO_2 and hence sources to the atmosphere. The pCO_2 values of the water samples in WZ and ZZ were between those of QX and TPX in January 2016, decreasing from QX to YC in the low-flow season and increasing in the high-flow season (Fig. 2a). The pCO_2 values were higher in the low-flow season (1150 ± 343 µatm) than in the high-flow season (987 ± 309 µatm) at QX, but the pCO_2 values were lower in the low-flow season at the other sites (Fig. 2a). The pCO_2 in the reservoir area was always lower than in the in-flowing water and the out-flowing water in reservoirs of Southwest China; however, similar pCO_2 values between reservoir area and the out-flowing water were found at TGR (Fig. 2a). The $\delta^{13}C_{DIC}$ varied from −13.2‰ to −6.6‰ for all the water samples, and the $\delta^{13}C_{DIC}$ of QX was much heavier than that at other stations, especially at high-flow (Fig. 2b). The $\delta^{13}C_{DIC}$ was lower in the high-flow season at all sites.

Discussion

Response of pCO_2 and $\delta^{13}C_{DIC}$ to hydrological change

Emissions of CO_2 from waters to the atmosphere have been explored in relation to CO_2 sources and processes

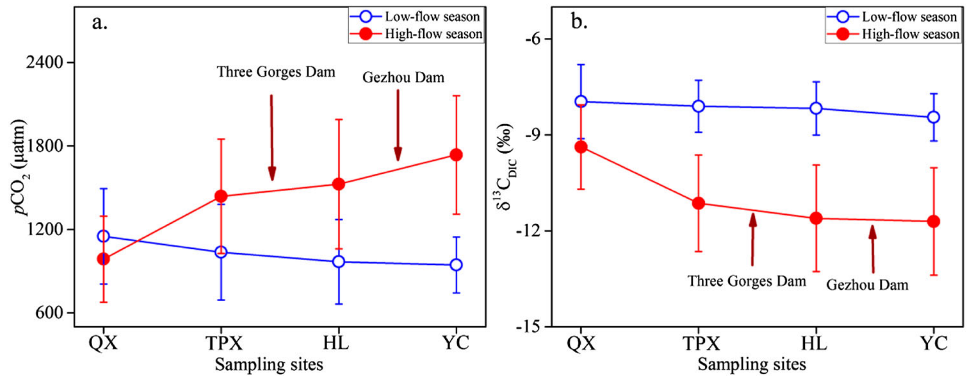

Figure 2. Changes in pCO_2 and $\delta^{13}C$ at 4 sites along the Three Gorges Reservoir system at low and high flow. (a) Variation in pCO_2; (b) variation in $\delta^{13}C_{DIC}$. Average of all samples in each season is shown along with standard deviation.

(Whitfield et al. 2010), including soil CO_2 influx, *in situ* degradation of organic carbon, catchment productivity, carbonate weathering, inorganic carbon loading, and photosynthesis (Johnson et al. 2008, Li et al. 2010, Larsen et al. 2011, Maberly et al. 2013, McDonald et al. 2013, Marcé et al. 2015). A negative correlation existed between pCO_2 and discharge near the upper reaches of the reservoir at QX (Fig. 3a), indicating a dilution effect by overland flow at high-flow. Water pCO_2 values were lower in high-flow than in low-flow, but pCO_2 values were variable in both seasons (Fig. 3a), indicating that hydrological variation was not the main controller of pCO_2 at QX. The negative correlation between pCO_2 and discharge showed that pCO_2 exhibited strong biogeochemical stationarity, which meant relatively stable behavior in response to changing discharge. Soil CO_2 influx or degradation of organic matters may be responsible for this biogeochemical stationarity with high discharge, which was similar to a previous study in the Wujiang River (Zhong et al. 2017).

A large dynamic range of $\delta^{13}C_{DIC}$ values were measured at QX, with the most negative value in high-flow conditions and the most positive value in low-flow conditions (Fig. 3b). The $\delta^{13}C_{DIC}$ values were negatively correlated to the discharge for QX (Fig. 3b). Negative $\delta^{13}C_{DIC}$ values were related to higher discharge, which should not be ascribed to simple dilution. High concentrations of CO_2 derived from terrestrially fixed carbon broken down in the soil can enter the water directly (Maberly et al. 2013). Large amounts of soil CO_2 were discharged into the river during high discharge, producing more negative $\delta^{13}C_{DIC}$ values in the water (Li et al. 2010, Zhong et al. 2017). Mineralization of macromolecules in the channel can also produce lighter $\delta^{13}C_{DIC}$ values in the water (Ward et al. 2013). Soil CO_2 recharge was likely to be the main driver of CO_2 dynamics responding to hydrological variation in QX (Zhong et al. 2017).

A negative relation between pCO_2 and discharge occurred at QX (Fig. 3a), but positive relationships were found between pCO_2 and discharge at HL and YC (Fig. 4a), both of which are located downstream of the Three Gorges Dam. Compared to the negative relationship at QX (Fig. 3a), the positive relationships at HL and YC may be ascribed to the absence of dilution effect (Fig. 4a). Higher values of pCO_2 were recorded with high discharge, contrary to findings at QX, owing to multiple biogeochemical processes in the reservoir.

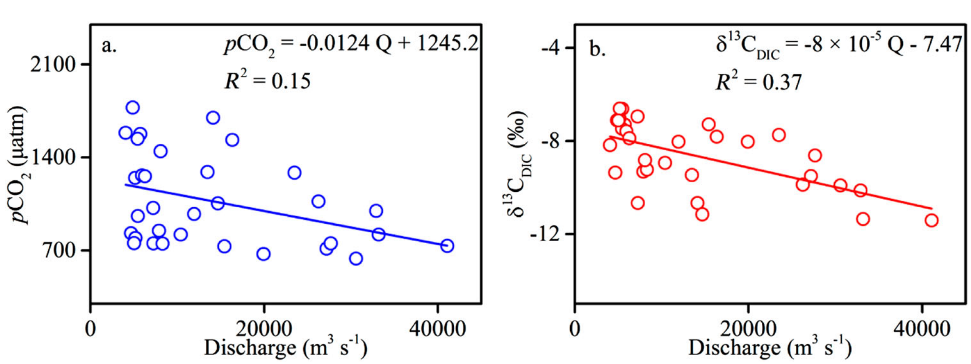

Figure 3. The relationship between pCO_2 or $\delta^{13}C_{DIC}$ and discharge at the upstream site on the Three Gorges reservoir Qingxi (QX). (a) pCO_2 vs. discharge; (b) $\delta^{13}C_{DIC}$ vs. discharge.

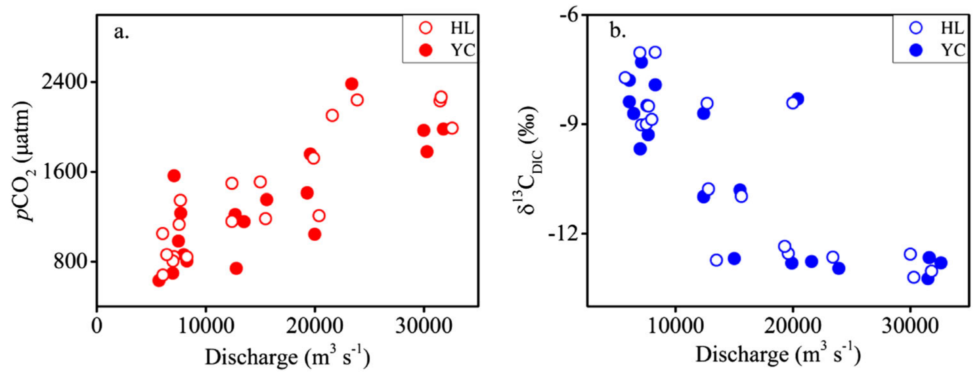

Figure. 4. The relationship between pCO_2 or $\delta^{13}C_{DIC}$ and discharge at Huanglingmiao (HL) and Yichang (YC). (a) pCO_2 vs. discharge; (b) $\delta^{13}C_{DIC}$ vs. discharge.

Biogeochemical processes in TGR may be responsible for the pCO_2 over-saturation at TPX, HL, and YC (Algesten et al. 2005, Brothers et al. 2012, McDonald et al. 2013).Thus, $\delta^{13}C_{DIC}$ values were significantly and negatively correlated to increasing discharge (Fig. 4b).

Relationships between pCO_2 and organic carbon concentration

The transformation between inorganic and organic forms of carbon would alter the pCO_2 in inland waters. When terrestrial or autochthonous organic carbon was mineralized, CO_2 was produced in reservoirs (Kosten et al. 2010), whereas phytoplankton productivity would remove CO_2 from waters (Wang et al. 2015). Although no significant linear relationship occurred between pCO_2 and discharge at QX, pCO_2 values at HL and YC were positively related to DOC concentration (Fig. 5a–b), similar to the results of Larsen et al. (2011) and Sobek et al. (2005). CO_2 over-saturation at HL and YC may be derived from the degradation of DOC in high-flow. Large amounts of DOC entered the inland waters, largely in the form of allothogenic C (Hope et al. 1996, Striegl et al. 2001, Whitfield et al. 2010). Despite the low contents, allothogenic DOC mineralization may be an important source of CO_2 over-saturation. Although no markedly spatial variation of DOC was measured in the TGR areas, allothogenic DOC inputs should counteract the effect of DOC degradation. Intense photosynthesis and submerged respiration would induce both high DOC and pCO_2 concentrations in the high-flow season.

POC concentration was high in the TGR system and was strongly related to the concentration of total suspended matter (TSM). TSM in surface waters of the Changjiang main stream ranged from 0.9 to 123.6 mg L^{-1}. The relationship between POC% (POC/TSM × 100%) and TSM followed that observed previously (Zhang et al. 2014) showing the power-law function (POC% = 16.59 × $TSM^{(-0.57)}$) for samples collected on both upstream and downstream of the Changjiang River. The same pattern in both upstream and downstream sites indicated that TGR did not have a major effect on the relationship between POC% and TSM (Fig. 6a).

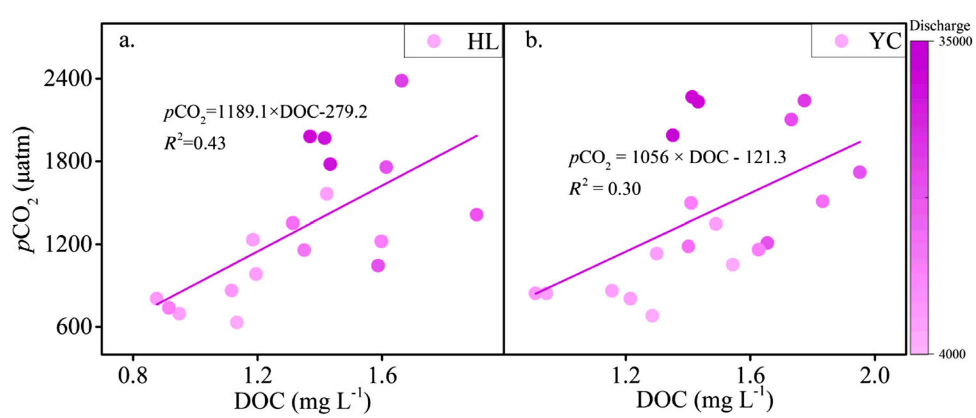

Figure. 5 Correlation between pCO_2 and concentration of DOC at (a) Huanglingmiao (HL) and (b) Yichang (YC). Discharge (m^3 s^{-1}) is indicated by the color density of the symbol.

Significant positive correlations between pCO_2 concentration and POC were found at HL and YC (Fig. 6b), indicating that organic matter mineralization may be a potential source of pCO_2 over-saturation in TGR. Large amounts of POC presented at high discharge in the high-flow season. Concurrently, POC mineralization increased in the high-flow season, contributing to the pCO_2 over-saturation. Dai and Liu (2013), Xu and Milliman (2009), and Yang et al. (2014) found that TGR noticeably traps sediment. Respiration in sediment contributes significantly to summer CO_2 emission for boreal and subarctic lakes (Algesten et al. 2005), so qualifying the contribution ratio of POC decomposition to water pCO_2 over-saturation was difficult. These results, however, were consistent with other studies that showed mineralization of organic carbon was a main diver of pCO_2 over-saturation (Hope et al. 1996, Algesten et al. 2005, Sobek et al. 2005, Whitfield et al. 2010, Weyhenmeyer et al. 2012, Ward et al. 2013).

In recent years, isotope proxy application has been widely used in studying the riverine carbon cycle (Li et al. 2010, Tamooh et al. 2013). $\delta^{13}C_{DIC}$ signatures have been used to trace the transport and transformation of DIC in inland waters based on the distinct isotopic values of various carbon sources (Barth et al. 2003, Li et al. 2010, Tamooh et al. 2013, Goodwin et al. 2016, Zhong et al. 2017). Riverine $\delta^{13}C_{DIC}$ dynamics were primarily controlled by both chemical weathering in the catchment and biogeochemical processes in inland waters. In general, rock weathering and biological CO_2 dissolution were 2 primary DIC sources, and photosynthesis, calcite precipitation, and CO_2 degassing were primary mechanisms of DIC transformation and loss. At QX, soil CO_2 influx and organic carbon decomposition should be responsible for the DIC temporal dynamics for the soil CO_2 contribution from various tributaries (Zhong et al. 2017). Soil CO_2 influx was related to the reactive interface between water and soil and would therefore play a minor role in DIC dynamics in the reservoir area because of the limited soil–water interface. Both pCO_2 and $\delta^{13}C_{DIC}$ were negatively correlated with increasing discharge (Fig. 2a–b). Although large amounts of soil CO_2 discharged into the river, the dilution effect on pCO_2 can conceal the soil CO_2 influx at QX.

Significant negative relationships between $\delta^{13}C_{DIC}$ and pCO_2 presented in HL and YC (Fig. 7). Relatively higher pCO_2 concentrations with lighter $\delta^{13}C_{DIC}$ values occurred in high-flow conditions. Although the stratification was not significant in the reservoir area, the average residence time of water was 6–30 d (Zhao et al. 2013), sufficient time for organic carbon degradation. The over-saturation of pCO_2 in TGR would result in degassing of dissolved CO_2 to the atmosphere, but minor spatial variations occurred in water pCO_2. Inorganic carbon loading and organic carbon decomposition may be the primary driver of pCO_2 in TGR. Inorganic carbon loading would shift to $\delta^{13}C$-enriched DIC values, but the $\delta^{13}C_{DIC}$ values became more negative at TPX, HL, and YC than that at QX. Therefore, inorganic carbon loading should not be regarded as the primary driver of pCO_2 in TGR.

In general, the upper Changjiang catchment has C3 plant coverage, suggesting that the organic carbon would deplete in ^{13}C in terms of water DIC for TGR. Impoundment of TGR would increase the riverine water transit time. Although the Three Gorges Dam released water for flood control in the high-flow season, organic carbon increased with increasing discharge, and mineralization of organic carbon likely contributed to the elevated dissolved CO_2 (Whitfield et al. 2010). The biological CO_2 dissolution would shift the $\delta^{13}C_{DIC}$ to more negative values.

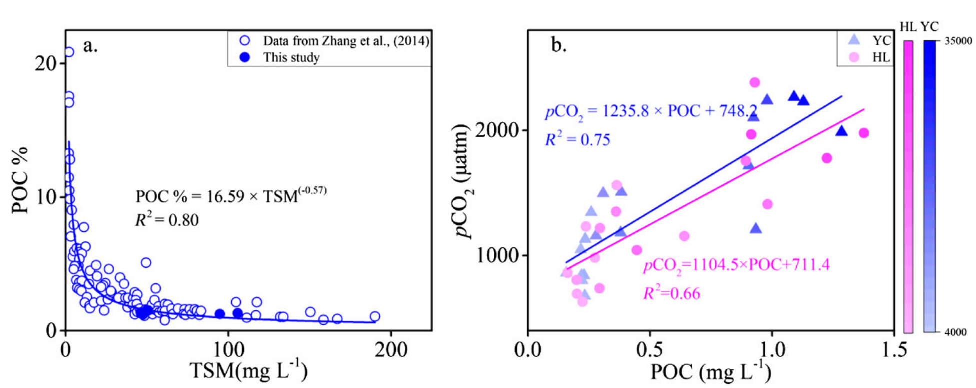

Figure 6. Relationships between POC and TSM or pCO_2 in the main stem of the Changjiang River. (a) Relationship between POC% and TSM at this study compared to data from Zhang et al. (2014); (b) relationship between pCO_2 and POC at Huanglingmiao (HL) and Yichang (YC). Discharge (m^3 s^{-1}) is indicated by the color density of the symbol.

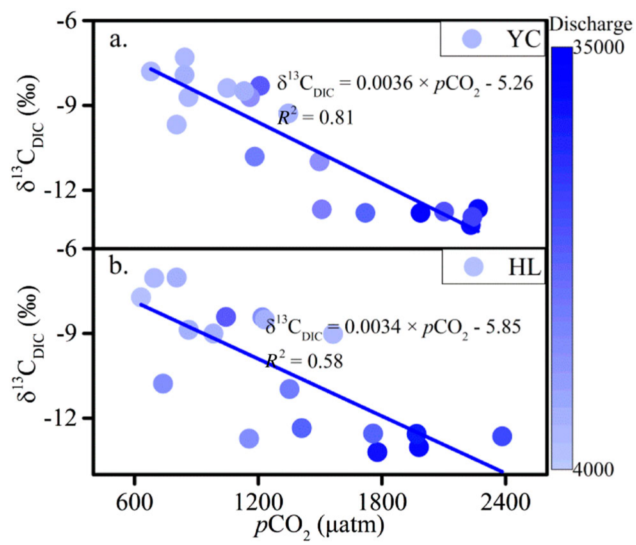

Figure 7. Correlation between $\delta^{13}C_{DIC}$ and pCO_2 at (a) Yichang (YC) and (b.) Huanglingmiao (HL). Discharge ($m^3\ s^{-1}$) is indicated by the color density of the symbol.

The relations of $\delta^{13}C_{DIC}$ versus pCO_2 were consistent with our hypothesis that organic carbon decomposition, depleted in ^{13}C, was responsible for the water pCO_2 over-saturation in the reservoir (Fig. 7). Therefore, the pCO_2 over-saturation in the TGR area can be explained by not only the attributing soil CO_2 influx with the inflowing water, but also the mineralization of organic carbon in the reservoir area as the primary driver of CO_2 over-saturation.

Sensitivity of pCO_2 to temperature variability in TGR

Mineralization of organic carbon and primary productivity were sensitive to temperature (Sobek et al. 2005, Acuna et al. 2008, Maberly et al. 2013,), which regulates the water pCO_2. A negative relationship between pCO_2 and T was found in QX, but the explained variance in pCO_2 by T was low ($R^2 = 0.135$; Fig. 8a). Because of the turbid and fast-flow water to the QX, especially in high-flow, the pCO_2 dynamics could not be elucidated by primary production. As discussed earlier, dilution effects of pCO_2 and soil CO_2 recharge with inflow of tributaries should control the pCO_2 dynamics; therefore, temperature was not the primary driver of pCO_2 over-saturation at this site. The pCO_2 concentration increased with increasing T for TPX, HL and YC (Fig. 8b–d), supporting the hypothesis that a

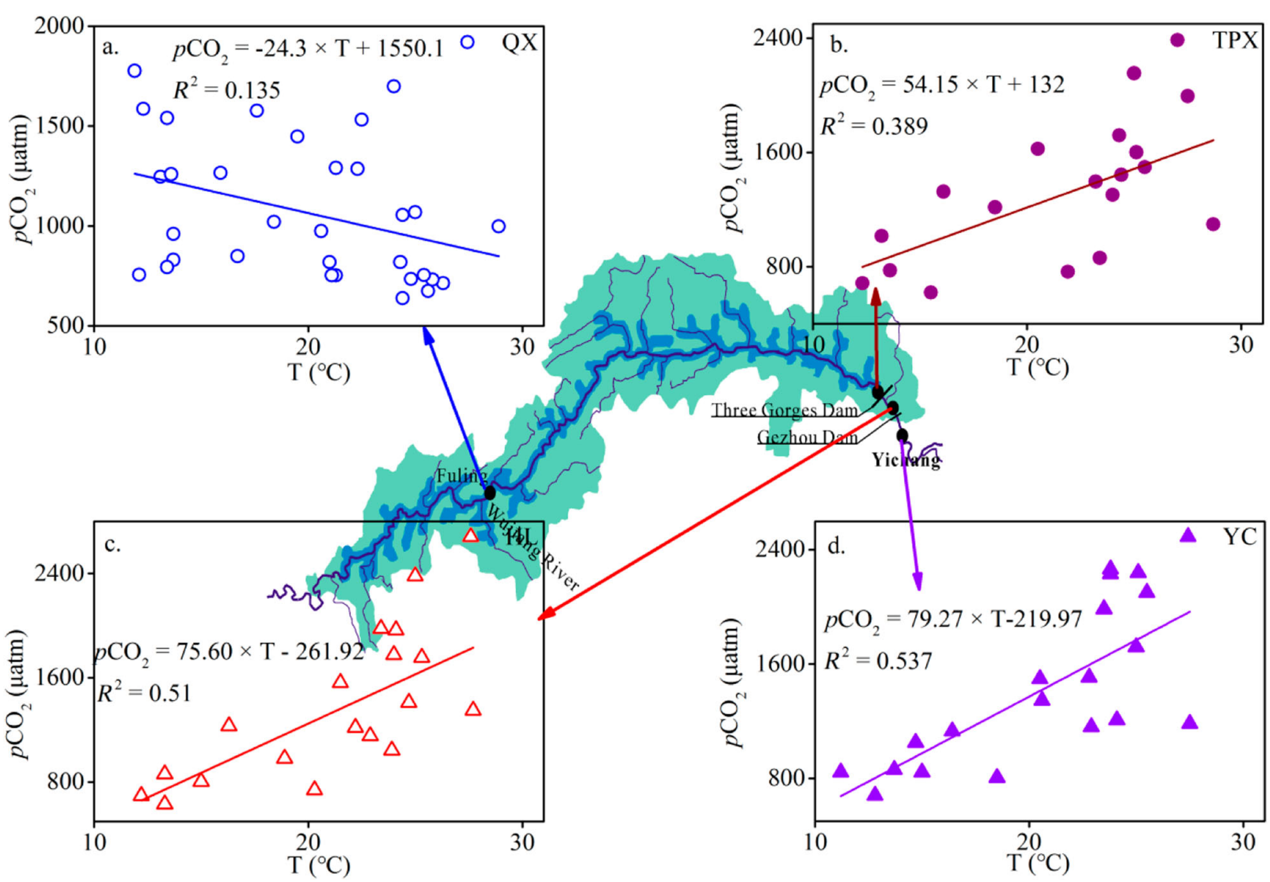

Figure 8. Scatter plot of pCO_2 vs. water temperature at (a) Qingxi (QX), (b) Taipingxi (TPX), (c) Huanglingmiao (HL), and (d). Yichang (YC).

lower rate of primary production than organic carbon mineralization occurred in the reservoir. High temperature stimulated high organic carbon mineralization rate, thus increasing the water pCO_2. Organic carbon mineralization should therefore be responsible for the water pCO_2 over-saturation, with high temperature as the primary driver.

High pCO_2 in the water was the main driver of CO_2 emission for inland waters. Mineralization of organic carbon was the main source to replenish the dissolved CO_2 lost to the atmosphere or taken up by phytoplankton. The pCO_2 decreased along the main stream for the TGR area in the low-flow season (Fig. 2a), likely ascribed to the aquatic CO_2 emission and low organic carbon mineralization rate at low temperature. The pCO_2 increased along the main stream for the TGR area in the high-flow season (Fig. 2a), indicating that organic carbon mineralization produced more CO_2 than that emitted from reservoir waters to the atmosphere. Organic carbon mineralization was therefore the primary driver of TGR CO_2 over-saturation with respect to the atmosphere.

Acknowledgements

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