

RESEARCH ARTICLE

Organic carbon transport in the Songhua River, NE China: Influence of land use

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Abstract

Carbon transported by rivers is an important component of the global carbon cycle. Here, we report on organic carbon transport along the third largest river in China, the Songhua River, and its major tributaries. Water samples were collected seasonally or more frequently to determine dissolved organic carbon (DOC) and particulate organic carbon (POC) concentrations and C/N and stable carbon isotopic ratios. Principal component analysis and multiple regression analysis of these data, in combination with hydrological records for the past 50 years, were used to determine the major factors influencing the riverine carbon fluxes. Results indicate that the organic carbon in the Songhua River basin is derived mainly from terrestrial sources. In the 2008–2009 hydrological year, the mean concentrations of DOC and POC were 5.87 and 2.36 mg/L, and the estimated fluxes of the DOC and POC were 0.30 and 0.14 t·km⁻²·year⁻¹, respectively. The riverine POC and DOC concentrations were higher in subcatchments with more cropland, but the area-specific fluxes were lower, owing to decreased discharge. We found that hydrological characteristics and land-use type (whether forest or cropland) were the most important factors influencing carbon transport in this system. Agricultural activity, particularly irrigation, is the principal cause of changes in water discharge and carbon export. Over the last 50 years, the conversion of forest to cropland has reduced riverine carbon exports mainly through an associated decrease in discharge following increased extraction of water for irrigation.

KEYWORDS

agricultural activity, land use, riverine organic carbon, stable carbon isotope, the Songhua River

1 | INTRODUCTION

Riverine carbon is an important component in the global carbon cycle that combines natural processes and anthropogenic activities. Annually, a large amount of terrestrial carbon is transferred into inland water systems (5.7 Pg-C/year; Raymond et al., 2013), which is much greater than the terrestrial carbon sink associated with anthropogenic emissions (2.8 Pg-C/year; Tranvik et al., 2009). However, the potential effects of human activities on surface water systems and the fate of the eroded carbon remain poorly understood (Wehrli, 2013).

Organic carbon transport in a river is driven by complex interactions involving multiple factors. Discharge has been suggested as the most dominating factor related to the riverine carbon flux (Lauerwald, Hartmann, Ludwig, & Moosdorf, 2012; Ludwig, Probst, & Kempe, 1996; Meybeck, 1982). Among other relevant factors, such as the watershed geomorphology, land use, soil chemistry, and temperature

(D'Amore, Fellman, Edwards, & Hood, 2010; Evans, Chapman, Clark, Monteith, & Cresser, 2006; Evans, Monteith, & Cooper, 2005; Hossler & Bauer, 2013; Köhler, Buffam, Seibert, Bishop, & Laudon, 2009; Worrall, Burt, & Adamson, 2004), land use has profound effects on riverine carbon transport (Lauerwald et al., 2012; Longworth, Petsch, Raymond, & Bauer, 2007) because it can affect soil properties and runoff, as well as the concentrations, fluxes, and geochemistry of the riverine carbon (Chantigny, 2003; Guo & Gifford, 2002; Longworth et al., 2007; Sickman, DiGiorgio, Davisson, Lucero, & Bergamaschi, 2010).

In a natural forest ecosystem, the soil erosion intensity is generally low, but the ground surface and soil with enriched organic matter from the decomposition of wood debris provide plentiful dissolved organic carbon (DOC) to the aquatic system (D'Amore et al., 2010; Michalzik, Kalbitz, Park, Solinger, & Matzner, 2001). Deforestation may drastically enhance the soil erosion rate and lead to the loss of soil organic carbon

(SOC), thus increasing the transport of sediment and particulate organic carbon (POC) to rivers (Eswaran, Van Den, & Reich, 1993; Montgomery, 2007; Van Oost, Cerdan, & Quine, 2009; Wang, Tian, Liu, & Pan, 2003). Additionally, severe DOC leaching out of the soil has been observed in farmlands (Kindler et al., 2011; Liu et al., 2010). However, deforestation and agricultural activities may reduce DOC export (Cronan, Piampiano, & Patterson, 1999; Guo & Gifford, 2002; Lauerwald et al., 2012). Studies have also shown that the riverine carbon concentration and flux may increase in the short term but decrease in the long term after the conversion of forest to cropland (Chantigny, 2003; Moore, 1989; Schelker, Eklöf, Bishop, & Laudon, 2012). Due to the lack of long-term monitoring data, it remains difficult to draw a concrete conclusion regarding the influence of land-use change on riverine carbon, even at the basin scale.

The Songhua River (SR) is located in Northeast China and drains the most fertile and organic-rich black soil region in the country. The Songhua River basin (SRB) has undergone significant land cover change due to massive migration and intense reclamation in the last century (Ye & Chen, 1992). Much of the area has been cultivated from its pristine state over the past 50 years (SRLRC, 2004). Fortunately, some mid- and small-sized subbasins largely remain in wild status with natural forests. These areas provide an opportunity to compare

cultivated and uncultivated subbasins and analyse the effects of land use on riverine organic carbon.

Here, for the first time, we perform a comprehensive survey on the concentrations, fluxes, and stable isotopic compositions of the riverine organic carbon (DOC and POC) in the SR. The objectives of this study were as follows: (a) to characterize the spatial and temporal patterns of riverine organic carbon in the SRB, (b) to evaluate the carbon sources, fluxes, and their relationships with basin properties, and (c) to explore the regional cropland- or land-use effect on riverine organic carbon transport and the implications for the carbon cycle.

2 | STUDY AREA

The SR is geographically distributed within 119°12'–132°33' E and 41°42'–51°40' N (Figure 1) and drains an area of $5.67 \times 10^5 \text{ km}^2$. The annual discharge of the river is $75.9 \times 10^9 \text{ m}^3/\text{year}$ (the third largest river in China). The SR consists of the Nen River to the north and the Ersong River to the south in the upper reaches, and it merges into the Heilong River (Amur) in the lower reaches, totaling 2,309 km in length. The climate varies from temperate to cool temperate. The annual precipitation ranges from 450 to 650 mm, of which 70–80% occurs during the summer season from June to September. The

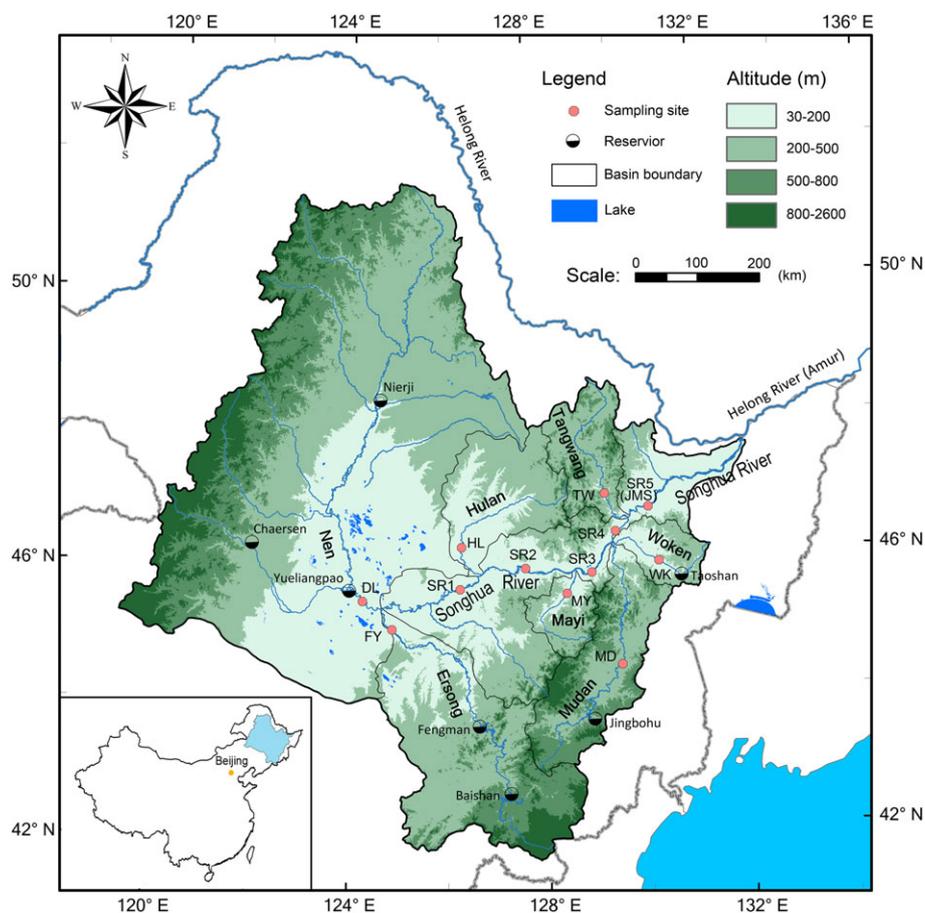


FIGURE 1 Map showing the Songhua River system and sampling sites. Some sampling sites were named after abbreviations of the hydrometric stations: FY = Fuyu station at the outlet of the Ersong River, DL = Dalai station at the outlet of the Nen River, and JMS = Jiamusi station at the outlet of the Songhua River. The other sites were named from abbreviation of the river's name it represented

multiyear mean temperature (1956–2002) is 4 ± 1 °C (SRLRC, 2004). The highest monthly mean temperature ranges from 20 to 25 °C in July, and the lowest is -20 °C in January. Rivers in this region are ice covered from November to March of the following year.

The SRB is a Late Mesozoic and Cenozoic volcanic region (Liu, 2007) dominated by silicates and less than 1% of carbonates. Quaternary fluvial sediments are abundant in the alluvial plains and river valleys, and they account for approximately 41% of the total basin area. The topography of the SRB is characterized by long and gentle slopes, including 61% montane area, 15% foothills, and 24% plains (Liu, Liu, Wan, & Yang, 2008). The vegetation consists mainly of conifer and broadleaf mixed forests, as well as temperate grasses.

The SRB is widely covered with dark-colored soils, including black soil, chernozem, dark brown forest soil, and brown soil. The parent materials of the black soil are Quaternary lacustrine and fluvial deposits or loess (Zhang, Wu, Liu, Zheng, & Yin, 2007). According to the soil taxonomy, the black soil group belongs to udicisohumisol of the isohumisol order (Wen & Liang, 2001). The black soil has a high organic matter content (20–60 g/kg). Much of the area remained intact until massive reclamation transformed the forest or wetland into farmland during the past 100 years (Xu, Xu, Chen, Xu, & Zhang, 2010). The natural forest cover has declined from more than 90% to approximately 30% (Ye & Chen, 1992). Consequently, soils have been seriously degraded since the 1950s. The organic matter content of the black soil declined from 40 to 60 g/kg in its natural state to 20–30 g/kg after cultivation (Liu & Yan, 2009).

Along the SR, there are 34 large-sized (capacity ≥ 100 million m^3) and 180 mid-sized (capacity of 10–100 million m^3) reservoirs, with a total volume of 49.5 billion m^3 . Four reservoirs, Nierji, Baishan, Fengman, and Chaersen, in the upper reaches (Figure 1) account for 26.8 billion m^3 or 54% of the total reservoir volume (Dang, 2014). Due to extensive reclamation for cropland development, the total area of the lakes and wetlands in the SRB has diminished considerably, and

now, only approximately 2.5 thousand km^2 remains, mainly in the lower reaches of the Nen subbasin (Figure 1). Detailed SRB information is presented in Tables 1 and 2.

3 | METHODS

3.1 | Sampling and lab analysis

Seasonal samplings were performed in the SR main stem and its seven major tributaries in May (spring), August (summer), and October (autumn) 2008, and January (winter) 2009 (Figures 1 and 2). The sampling site of each tributary was chosen at or close to the lowest hydrometric station. In the SR main stem, five sampling sites were selected right below the confluences of the major tributaries. To conduct a robust estimation of the riverine organic carbon flux, high-frequency sampling was conducted at the Jiamusi (JMS) hydrological station, which is located at the outlet of the SR (Figure 1). Fifty-three samples were collected at 3- to 14-day intervals over a 1-year cycle according to the hydrologic conditions. Samples were taken at a depth of 0.5–1.0 m below the water surface on the central line of the river, except for the winter samples, which were taken from 20- to 30-cm depths beneath the ice cover.

On the sampling day, the collected water samples were filtered by vacuum filtration through 0.7 μm Whatman GF/F filter paper (47 mm in diameter) that was precombusted at 450 °C for 6 hr and preweighed in the laboratory. Two parallel sediment samples were obtained during each sampling. One was dried at 103 °C for 24 hr and was then weighed to calculate the net total suspended sediment (TSS) by deduction of the filter paper weight. The other was dried at 50 °C for 24 hr and then kept in a plastic bag for POC, C/N ratio, and stable carbon isotope analyses. Before the POC, particulate nitrogen (PN) and stable carbon isotope analyses, the samples were acidified with diluted

TABLE 1 Terrain features and reservoir distributions in the subbasins of the Songhua River

Basin	Terrain features	Reservoirs
Ersong	Upstream: mountains covered with dense forest, narrow river valley Middle and lower reaches: hills, open river valley, agricultural area	Baishan reservoir (5.91 billion m^3) Fengman reservoir (10.99 billion m^3)
Hulan	Upstream: the Xiaoxinganling range (mountain virgin forest) Middle reaches: hills, broad floodplains, concentrated distribution of cropland Downstream: wide plain, major agricultural area for the Songhua basin	A number of mid-sized reservoirs (79.73 million m^3 in total)
Mayi	A mountainous region (mountains account for 70% of the whole basin) Upstream: highlands with forest cover Middle and lower reaches: hills and alluvial plain, most croplands distributed in the river valley	A number of mid-sized reservoirs (82.54 million m^3 in total)
Mudan	Upstream: undulating mountainous area, scattered distribution of cropland Middle reaches: expansive river valley, concentrated distribution of cropland Downstream: mountainous area with deep narrow valley	Lianhua reservoir (4.2 billion m^3) Jingbohu reservoir (1.63 billion m^3)
Nen	Upstream: the Daxinganling range (mountain virgin forest) Middle reaches: transition zone between mountains and plains Downstream: vast plain with gentle hills, major agricultural area for the Songhua basin	Nierji reservoir (8.61 billion m^3) Yueliangpao reservoir (1.20 billion m^3) Chaersen reservoir (1.25 billion m^3)
Tangw	A hilly and mountainous basin covered with dense forest, little cropland, a national nature reserve	No large- and mid-sized reservoirs
Woken	Upstream: hilly area featured with massive relief, cropland distributed in the narrow river valley Middle and lower reaches: hilly area featured with broken relief, wider river valley dominated by croplands	Taoshan reservoir (0.26 billion m^3)

TABLE 2 Environmental factors, riverine organic carbon concentrations, fluxes, and geochemistry in the Songhua River basin

	Ersong	Hulan	Mayi	Mudan	Nen	Tangw	Woken	Songhua
Area (10 ⁴ km ²) ^a	7.2 (7.4)	2.8 (3.1)	1.0 (1.1)	2.2 (3.7)	22.2 (29.3)	1.9 (2.1)	0.4 (1.1)	52.8 (55.1)
Flow (10 ⁸ m ³ /year) ^b	123	4.8	3.7	35.9	48.3	26	0.8	248.1
Rainfall (mm/year)	700	560	510	615	484	512	548	547
Altitude (m)	441	243	318	542	413	440	235	385
Slope (‰) ^c	1.62	0.88	0.7	1.23	0.93	0.89	0.71	0.1
Relief	277	113	178	237	267	151	102	256
Cropland (%)	39.1	49.7	29	26.1	42.3	1.9	48.1	38.3
Forest (%)	47.1	42.3	60.8	66.7	44.5	94.9	34.6	48.7
EroM (t·km ² ·year ⁻¹) ^d	561	515	657	646	936	554	653	776
Pop (person/km ²) ^d	224	153	86	76	57	44	111	104
[DOC] (mg/L)	6	8.98	4.56	6.29	6.07	10.09	11.57	5.64 (5.87 ^e)
[POC] (mg/L)	2.6	2.25	0.89	1.06	1.25	0.41	4.34	1.86 (2.36 ^e)
[POC] (%)	3.31	7.75	4.05	1.84	3	4.53	6.25	3.19 (2.44 ^e)
POC/PN	7.26	6.34	9.42	10.88	8.16	9.97	7.49	7.2
δ ¹³ C _{POC} (‰)	-24.3	-25.3	-27.1	-24.5	-26.1	-25.6	-24.8	-23.6
F _{DOC} (t·km ² ·year ⁻¹)	0.81	0.16	0.17	0.87	0.12	1.57	0.15	0.28(0.30 ^e)
F _{POC} (t·km ² ·year ⁻¹)	0.43	0.04	0.03	0.18	0.03	0.06	0.09	0.11(0.14 ^e)

Note. The concentrations, fluxes, C/N ratios, and stable carbon isotopes are averaged from the results of the seasonal samples as listed in Table S1.

^aIn this row, the number outside brackets denotes the draining area of the hydrometric station or sampling site, whereas these in the brackets are the total area of the basin.

^bIt was monitored at the hydrological station where the sampling was conducted.

^cData from SRLRC, 2004.

^dEroM = erosion modular; Pop = population.

^eResults are based on the high-frequency sampling conducted at the JMS station.

HCl (20 vol.%) to remove carbonate. For the DOC analysis, filtrates were acidified with concentrated HNO₃ until pH values were less than 2 and kept in precleaned 100-ml brown glass bottles. All DOC and POC samples were kept frozen until analysis within 2 weeks. DOC measurement was performed using a TOC analyzer (Shimadzu TOC-Vwp) with an analytical error of less than 2%. POC and PN were analysed using a Perkin Elmer-2400 II (Elemental Analyzer CHNS/O) with an analytical error of less than 0.3%. For quality control, an analytical standard of acetanilide with 71.09% C and 10.36% N was inserted into every five sediment samples. Although inorganic nitrogen may exist in PN, it is a rather small fraction (Meybeck, 1982); thus, the POC/PN ratio is used for the POC/PON (particulate organic nitrogen) ratio in this study. To measure the stable carbon isotopes of POC, each sediment sample was oxidised in a sealed quartz chamber at temperatures of 850–900 °C for 5 hr, and the CO₂ yield was collected and purified. Then, the CO₂ was analysed using a Mat-252 mass spectrometer with dual inlets to determine the stable carbon isotopes at the Institute of Geology and Geophysics, Chinese Academy of Sciences (CAS). The results were reported using the conventional delta notation (δ¹³C) relative to the VPDB standard in permil. Replicate measurements yielded an analytical error (1σ) of ±0.05‰.

3.2 | Data sets of environmental characteristics

Rainfall, the soil erosion modulus and land use were selected to examine their potential influences on riverine carbon. Daily rainfall data collected in 2008 at 32 meteorological stations that best cover

the study area were used to represent the annual rainfall and the associated spatial pattern in the SRB. To calculate the soil erosion modulus of each subbasin, we applied a GIS technique to obtain the area of each land-use type and then calculated the soil erosion modulus of each subbasin by multiplying the area weights of various land-use types by their soil erosion modulus, as recommended in previous studies (Ministry of Water Resources of China, 2010; Wang, Jian, Jiao, & Su, 2013). To examine the effects of land use on hydrology, the runoff coefficient of each subbasin was calculated using area-specific discharge and precipitation.

Land-use data were obtained from the GlobCover 2009 database in a 300 m × 300 m grid produced from satellite images taken by the MERIS sensor in 2009. There are 22 land cover classes according to the United Nations Land Cover Classification System (LCCS). In this study, these original land cover classes were integrated into five categories: forest, cropland, grassland, water body, and residential. Undefined or incorrectly defined areas in the data set were verified or adjusted by field investigation or by comparison with the GLC2000 database or Google satellite maps. For example, some grassland in the original data set was converted to cropland. As forest and cropland dominate the SRB (>85%), both land types were addressed in this study (Table 2).

3.3 | Calculation of the riverine carbon flux

Because the organic carbon concentrations were measured at different frequencies but daily discharge and TSS were monitored continuously,

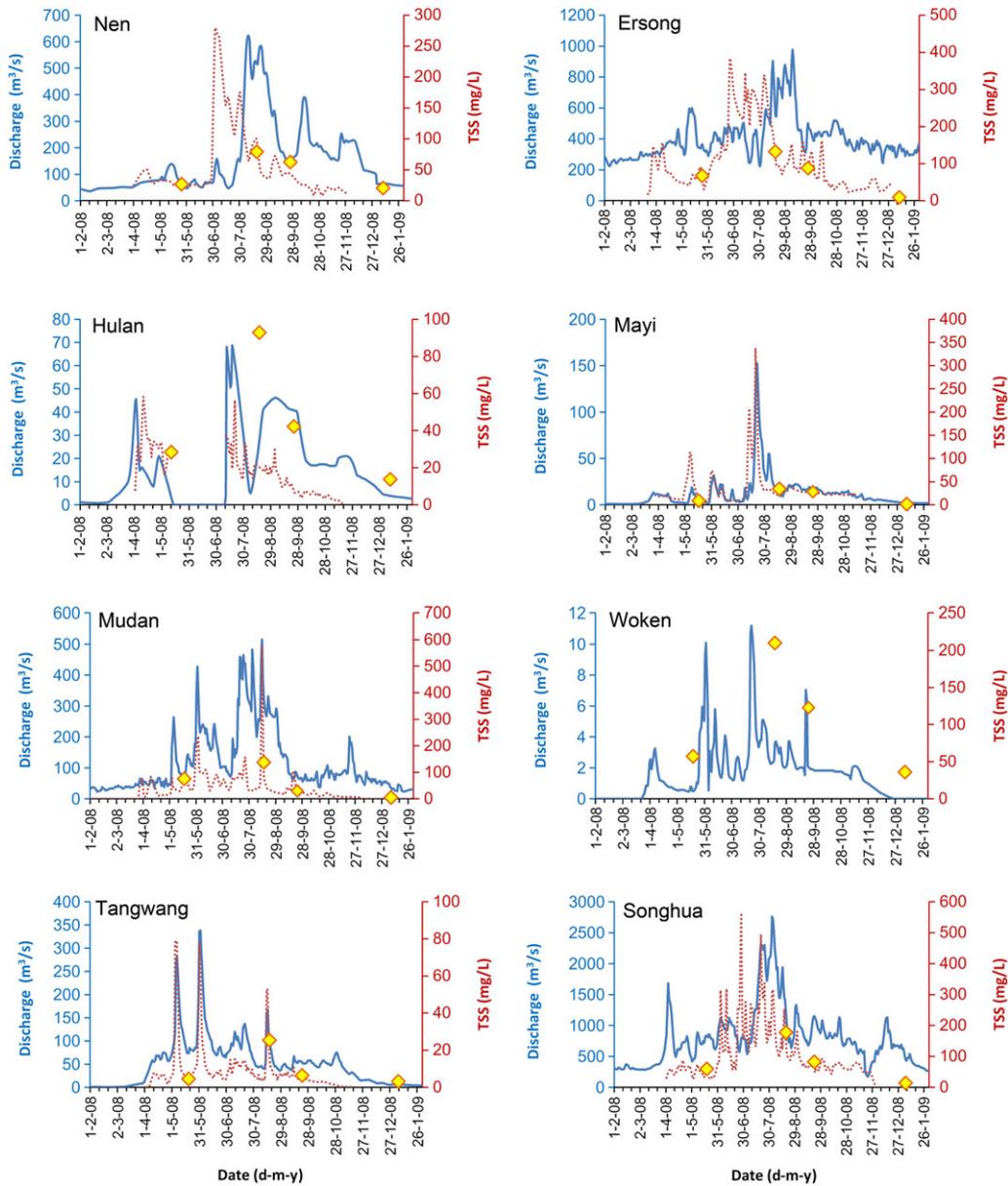


FIGURE 2 Daily discharge (solid line) and TSS (dashed line) of the Songhua River and its tributaries. Diamond indicates sampling date and TSS concentrations. TSS = total suspended sediment

we applied two methods to calculate the carbon flux. For sites with synoptic seasonal sampling, an interpolation method was used to calculate the flux, in which the concentration of each riverine carbon species in a water sample was taken as a mean value of the river water for the period of the sampling interval. The equation used is expressed below:

$$F = \sum(C_i * U_i), \quad (1)$$

where F is the estimated flux, C_i represents the measured concentration of the riverine carbon species (DOC and POC% in TSS) in sampling season i (i = spring, summer, autumn, and winter), and U_i represents either the total water discharge volume (for the DOC flux calculation) or the TSS load (for the POC flux calculation) in season i .

To calculate the carbon flux at the JMS station based on high-frequency sampling, we used Beale's stratified ratio estimator (Parks & Baker, 1997). This equation can be expressed as follows:

$$F = \mu_x \frac{m_y}{m_x} \left(\frac{1 + \frac{1}{n} \frac{S_{xy}}{m_x m_y}}{1 + \frac{1}{n} \frac{S_x^2}{m_x^2}} \right), \quad (2)$$

where F is the estimated flux, μ_x is the mean daily water discharge over a year, m_y is the mean daily DOC or POC flux on the days that the DOC or POC concentrations were measured, m_x is the mean daily water discharge on the days that the concentrations were measured, and n is the number of the days when the concentrations were measured. S_{xy} and S_x^2 are determined by the following equations:

$$S_{xy} = \frac{1}{(n-1)} \sum_{i=1}^n x_i y_i - n m_x m_y, \quad (3)$$

$$S_x^2 = \frac{1}{(n-1)} \sum_{i=1}^n x_i^2 - n m_x^2, \quad (4)$$

where x_i is the individual measured discharge and y_i is the daily DOC or POC load on each day that the concentration was measured. In Equation 2, m_y/m_x is defined as the ratio of the mean flux to the mean water discharge on the days that fluxes were calculated, and this ratio is used with the overall mean water discharge (μ_x) to estimate the annual carbon flux.

3.4 | Principal component analysis (PCA)

Riverine carbon export is affected by many intercorrelated factors in the basin. PCA is a technique that can reduce the dimensionality of a multivariate data set and extract the important influencing factors. It converts a set of observations of possibly correlated variables into a smaller number of linearly uncorrelated variables called principal components, which are linear combinations of the original variables. Here, we performed PCA to determine the principal components of the potential controlling factors of riverine carbon export in the SRB, including rainfall, the percentages of cropland and forest, the river slope, altitude, land relief, the soil erosion modulus, and the population density.

3.5 | Statistical analyses

A number of statistical analyses were performed using Microsoft Excel and Matlab software. Pearson correlation analysis was used to examine the relationships between the riverine organic carbon concentrations or fluxes and environmental factors. Additionally, simple regression analysis was used to determine the trends in the measured data or historical records, and multiple regression analysis was performed to explore the relationships between riverine carbon fluxes and environmental factors. *T* test and *F* test methods were used to evaluate the statistical significance.

4 | RESULTS

4.1 | Discharge and TSS

Water discharge values in the SR main stem and its major tributaries, as presented in Figure 2, were highly variable throughout the year; for example, they ranged from 170 to 2,760 m³/s at the JMS station in the hydrological year 2008–2009. In general, discharge sharply

increased in April and June, and the peak flow occurred in July or August. In the frozen period, the flow decreased to the lowest level and even entirely stopped in some smaller tributaries, such as the Woken and Tangwang rivers. The fluctuations in discharge were generally consistent with precipitation in the SRB, although discharge can also be affected by the agricultural consumption of water and reservoir regulation (SRLRC, 2004; Yang, 1994). The annual discharge of the SRB in 2008 was 248.1×10^8 m³, which was notably lower than the mean value of 653×10^8 m³ on the basis of the historical record from 1956 to 2002.

The temporal variation in the TSS concentration was positively related to discharge (Figure 2), with a maximal value in summer (i.e., the flood season) and a minimal value in winter (the dry season). The peak TSS concentration in the Ersong River, Nen River, and SR main stem occurred before the maximum discharge. On the basis of the daily data collected at the hydrological stations in 2008, the lowest annual mean concentration of TSS was observed in the Tangwang River (5.8 mg/L), and the highest was observed in the Ersong River (79.0 mg/L). There was no daily TSS monitoring data available for the Woken River in 2008, but historical records and our samples showed that the annual mean concentration of TSS in this river exceeded 100 mg/L, which was the highest in the studied tributaries.

4.2 | Concentrations and fluxes of riverine organic carbon

The DOC concentrations measured in the SRB are presented in Figure 3a (see also Table S1). The mean value of each tributary ranged between 4.56 and 11.57 mg/L (Table 2). The SR main stem and the large tributaries, including the Nen, Ersong, and Mudan Rivers, exhibited similar seasonal variations. For example, the minimum concentration occurred in summer (August), and the maximum occurred in spring (May) or autumn (September; Figure 3a). At the outlet of the SR (the JMS station), the DOC concentrations mainly fell in the range of 3–8 mg/L, with a mean of 5.71 mg/L. The highest DOC concentration was observed in the cropland-dominated Woken River (average of 11.57 mg/L and maximum of 24.76 mg/L), followed by the Tangwang (average of 10.09 mg/L, forest dominated), Hulan (8.98 mg/L, cropland dominated), and Mayi (4.56 mg/L) Rivers.

The riverine POC concentrations exhibited large variations seasonally and differed considerably between tributaries (Figure 3b).

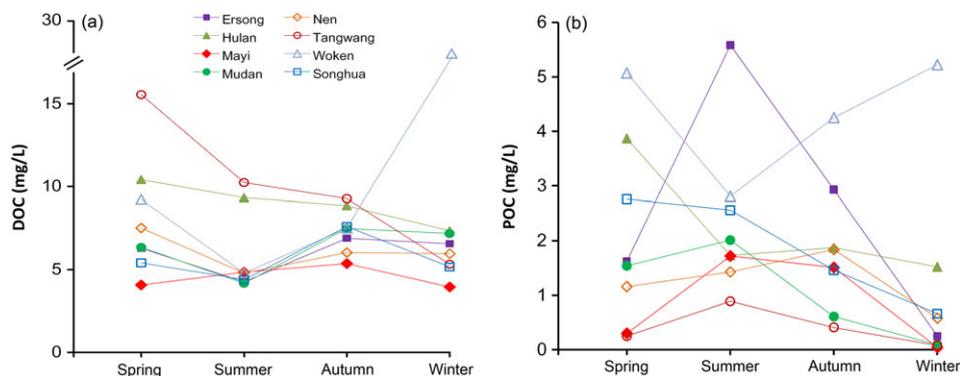


FIGURE 3 Seasonal variations of (a) DOC and (b) POC concentrations in the Songhua River system. DOC = dissolved organic carbon; POC = particulate organic carbon

The mean value in each tributary (Table 2) varied between 0.41 and 4.34 mg/L, with an overall average of 1.83 mg/L. The mean content of POC in the TSS (POC%) was 4.2% (ranging from 1.8% to 7.8%), which was substantially higher than the global mean of 0.95% (Ludwig et al., 1996). The highest mean value of 4.34 mg/L was found in the Woken basin, and the lowest of 0.41 mg/L was observed in the Tangwang basin. At the JMS station, the POC% generally ranged from 2% to 4%, with a few cases lower than 2% during flood events (see Table S2).

In the SRB, the DOC and POC fluxes varied from 0.12 to 1.57 t·km⁻²·year⁻¹ and from 0.03 to 0.43 t·km⁻²·year⁻¹, respectively. The highest DOC flux (1.57 t·km⁻²·year⁻¹) occurred in the Tangwang River, which drains a typical forested basin, whereas the highest POC flux (0.43 t·km⁻²·year⁻¹) occurred in the Ersong River, which is a subbasin with a moderate proportion of cropland. With the exception of the POC flux in the Woken River, both the DOC and POC fluxes in the cropland-dominated subbasins of Hulan, Nen, and Woken were observed to be the lowest. The annual fluxes of DOC and POC estimated at the JMS station, a representative of the entire SRB located at the outlet of the SR, were 0.30 and 0.14 t·km⁻²·year⁻¹, respectively (Table 2).

4.3 | Stable carbon isotopes of POC ($\delta^{13}\text{C}_{\text{POC}}$) and the C/N ratio

The $\delta^{13}\text{C}_{\text{POC}}$ in the SR system ranged from -27.1‰ to -23.6‰, with a mean value of -25.2‰ (Table 2); These values exhibited no significant seasonal or spatial trends. The atomic C/N ratio of the POC varied between 6.3 and 10.9 and averaged 8.3 (Table 2). The average C/N ratio in the main stem was lower than that in the tributaries, except for the Ersong and Hulan Rivers. Temporally, the C/N ratio exhibited higher values in August and December than in May and September (Table S1).

4.4 | PCA of environmental factors

The correlation coefficients between the environmental factors were calculated as the first step in the PCA (Table 3) to determine the spatial relationships between the environmental factors in the SRB. For example, the relatively significant relationships between the cropland area and altitude and population density reflect that in the SRB, farmland and inhabitants are mainly distributed in low-altitude plain areas, whereas forests are concentrated in the mountainous areas with low

population density. By calculating the eigenvalue of the correlation coefficient matrix, we obtained three principal components corresponding to the three largest eigenvalues, which together contributed to 92.5% of the total influence.

The coefficients, which are also called loadings, of the principal components (Table 4) represent the correlation between the principal components and the environmental variables. The first principal component (Z1) shows a strong positive correlation with the forest area and strong negative correlations with both the cropland area and the population density. Thus, Z1 is an index of forest conservation (or the opposite of agricultural activity). The second principal component (Z2) shows strong positive correlations with precipitation, altitude, and slope, and it can be regarded as an index of hydrological characteristics (runoff volume and velocity). The third principal component (Z3) is negatively correlated with land relief and the soil erosion modulus; therefore, it is an index of soil erosion resistance. On the basis of the coefficients (Table 4), the score of each principal component was calculated, and the following two multiple regression equations between the riverine organic carbon fluxes (F_{DOC} and F_{POC}) and the three principal components were obtained.

$$F_{\text{DOC}} = 0.3051 \cdot Z1 + 0.4304 \cdot Z2 + 0.2616 \cdot Z3 + 0.0000, \quad (5)$$

$$F_{\text{POC}} = -0.2879 \cdot Z1 + 0.4936 \cdot Z2 - 0.0973 \cdot Z3 + 0.0000. \quad (6)$$

The above two regression equations successfully passed the *F* test ($\alpha < 0.01$). According to Equations 5 and 6, the hydrological characteristic index (Z2) is the first control, and it is positively correlated with both the DOC and POC fluxes, as was reported in previous studies (Lauerwald et al., 2012; Ludwig et al., 1996; Meybeck, 1982). The forest conservation index (Z1) is the second control, and it is positively correlated with the DOC flux but negatively correlated with the POC flux. The weight coefficient of the soil erosion resistance (Z3) ranks last, indicating that its influence on riverine organic carbon is smallest among the indexes.

5 | DISCUSSION

5.1 | Spatial and seasonal variations in the concentrations of riverine organic carbon

The seasonal patterns of the DOC concentrations observed in this study (Figure 3a) can be well explained by the flow conditions.

TABLE 3 Correlation coefficient matrix of the environmental factors in the Songhua basin

	Rainfall	Altitude	Slope	Relief	Cropland	Forest	Erosion M	Population
Rainfall	1.000							
Altitude	0.329	1.000						
Slope	0.659	0.420	1.000					
Relief	0.339	0.721	0.230	1.000				
Cropland	0.162	-0.566	-0.079	-0.021	1.000			
Forest	-0.167	0.550	0.093	-0.022	-0.988	1.000		
Erosion M	-0.478	0.159	-0.366	0.522	0.231	-0.287	1.000	
Population	0.800	-0.207	0.438	0.122	0.527	-0.520	-0.452	1.000

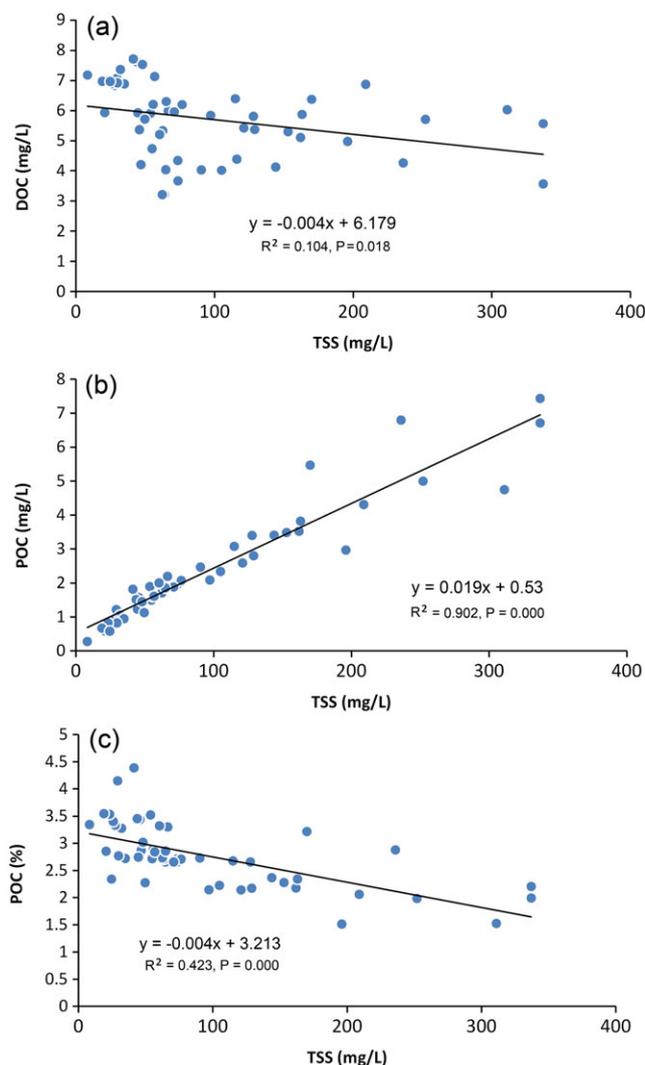
TABLE 4 The principal component coefficients (loadings)

	Z1	Z2	Z3
Rainfall	-0.3494	0.4673	0.0134
Altitude	0.2747	0.4685	-0.2979
Slope	-0.1629	0.4814	0.0583
Relief	0.0279	0.3062	-0.6111
Cropland	-0.4931	-0.2575	-0.2244
Forest	0.4869	0.2571	0.2616
Erosion M	0.1422	-0.2391	-0.6413
Population	-0.5241	0.2169	0.0644

Floods caused by ice melting in May flush out the DOC from the soil that accumulated during the cold season (Haei et al., 2010). Thus, the concentration of riverine DOC increases in the spring. In August (the flood season), the dilution effect caused by increased discharge decreases the DOC concentration to the lowest annual value. After the rainy season, discharge begins to decrease rapidly, and the riverine DOC concentration gradually increases to a normal level (Sun, Han, Zhang, & Lu, 2007). In addition, deep draining water became a more prominent flow component in autumn and early winter when precipitation or surface runoff decreased. This deep flow may have carried more DOC leached from soils and contributed to a higher riverine DOC level in these seasons. Apart from the influence of hydrology, other factors can influence the variations in DOC concentrations, as reflected by a weak relationship between the DOC concentration and water discharge (Figure 4a). For instance, the physical disruption of the soil, the mortality of fine roots, and the lysis of freeze-damaged soil organisms can increase the DOC concentration in the soil solution (Scott, Jones, Woof, & Tipping, 1998; Austnes & Vestgarden, 2008; Haei et al., 2010).

The POC concentration in the SR main stem showed a positive relationship with TSS (Figure 4b), whereas POC% was negatively related to the TSS (Figure 4c). A plausible explanation for such relationships is that the high TSS concentrations represent deep soil erosion caused by increasing precipitation/runoff, whereas the organic carbon content decreases with soil depth (Coynel, Seyler, Etcheber, Meybeck, & Orange, 2005; Li, Li, & Xu, 2006). In addition, these relationships may be partly associated with decreased phytoplankton production caused by the reduced availability of light as the TSS concentration increases (Balakrishna & Probst, 2005).

The riverine DOC and POC concentrations showed clear differences among the tributaries (Table 2); both increased with the proportion of cropland area, except for the extraordinarily high value of DOC in the Tangwang River (Figure 5). In the cropland-dominated Woken basin, both the DOC and POC concentrations ranked the highest among those in all subbasins and were approximately 2.5 and 10 times higher than the lowest values observed in the Mayi and Tangwang basins, respectively. This result is consistent with previous observations, which suggested that agricultural activities can facilitate mechanical soil erosion and DOC leaching from soils (Chantigny, 2003).

**FIGURE 4** Relationships of TSS with (a) DOC, (b) POC, and (c) POC% for the Songhua River at the JMS hydrological station. DOC = dissolved organic carbon; POC = particulate organic carbon; TSS = total suspended sediment

5.2 | Sources of riverine organic carbon

The strong correlation between POC and TSS, as shown in Figure 4b, demonstrates the dominance of terrestrial sources of POC in the SR. This trend is also reflected by the $\delta^{13}\text{C}_{\text{POC}}$ value and C/N ratio. The $\delta^{13}\text{C}$ of SOC in the SRB varies between -18.4‰ and -23.8‰ , with a mean value of -21.8‰ (Cheng et al., 2010; Fang, Yang, Zhang, & Liang, 2005; Liu, Liu, Wang, Yu, & Wei, 2004), whereas the C/N ratio of SOC is generally within the range of 9–15 (mean 12; Tian, Chen, Zhang, Melillo, & Hall, 2010). Therefore, for most of the samples, the $\delta^{13}\text{C}_{\text{POC}}$ value ($-25.2 \pm 2.2\text{‰}$ in average, Table 2) was slightly lower than that of the SOC, indicating some input of in-river primary production, which has more depleted $\delta^{13}\text{C}$ values than the land-sourced riverine organic carbon. The C/N ratios of POC (8.3 ± 1.8) were lower than those of SOC. The effect of the aquatic biomass on the riverine POC might contribute to decreasing the C/N ratio because aquatic biomass has a notably lower C/N ratio than terrestrial plants. Another reason for this observation may be the selective microbial degradation of labile organic matter with high carbon and low nitrogen

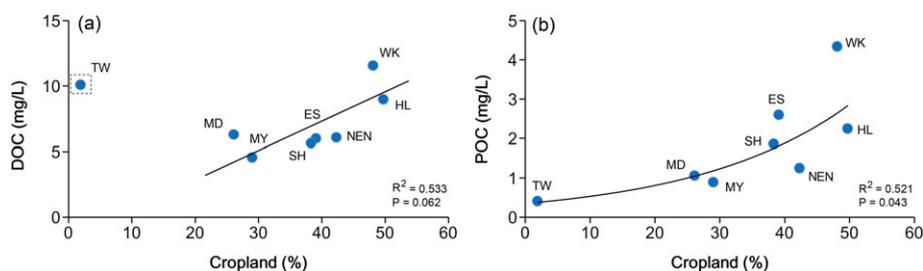


FIGURE 5 Relationships of cropland percentage with concentrations of (a) DOC and (b) POC. The extraordinarily high value of DOC in the Tangwang River draining a forested region is excluded for correlation analysis. The capital letters in the graph are the abbreviated basin names: TW = Tangwang, MD = Mudan, MY = Mayi, SH = Songhua, ES = Ersong, NEN = Nen, WK = Woken, HL = Hulan. DOC = dissolved organic carbon; POC = particulate organic carbon

contents, such as hydrolytic amino acids, carbohydrates, and lipids (Haynes, 2000; Oakes, Eyre, Middelburg, & Boschker, 2010). This process can result in a marked decrease in the C/N ratio of residual organic matter without significant change in carbon isotopic composition.

In general, land-sourced carbon dominates the organic carbon in rivers with high TSS concentrations (Ludwig et al., 1996), but the aquatic component (in-river primary production) may be dominant in some clean, slow-flowing waterways that facilitate phytoplankton photosynthesis. This mechanism can be observed in the Tangwang River, where the yearly minimum $\delta^{13}\text{C}_{\text{POC}}$ value and C/N ratio in autumn (September; Table S1) correspond to the optimum conditions (slow and clear water with a favorable temperature) for phytoplankton growth. The C/N ratios of some tributaries were relatively high in winter (December), likely due to restricted growth of aquatic organisms in soils and rivers at low or freezing temperatures in the basin (Haei et al., 2010).

5.3 | Influences of agricultural activities on riverine organic carbon export

The PCA, as mentioned previously, suggested that the hydrological characteristic (i.e., runoff volume and velocity) and land-use (cropland and forest) indexes were the most important factors that controlled the riverine organic carbon flux in the SRB. The former is expected because flow serves as the only mechanism for exporting riverine material. To better understand the cropland effect (agricultural activities) on the carbon flux, the relationships between cropland coverage, the DOC and POC concentrations and fluxes, runoff, and sediment loads are discussed below.

As discussed previously, both the DOC and POC concentrations exhibited increasing trends as the percentage of cropland increased (Figure 5). However, the POC flux displayed no obvious trend with increasing cropland, and the DOC flux exhibited a decreasing trend (Figure 6a and 6b). As an integrated result, the total carbon flux of the DOC plus POC showed a decreasing trend (Figure 6c). When the carbon fluxes were divided by the normalized precipitation, such relationships became more prominent in all the subcatchments (Figure 6d and 6f). Indeed, the TSS concentration was positively correlated with cropland area in the sampling year and based on the long-term records from 1956 to 2002 (Figure 7a). This result confirms the effect of cropland on increased riverine DOC and POC

concentrations. Nevertheless, these long-term data also showed a consistent negative relationship between cropland area and the annual TSS flux (Figure 7b), which could provide some insight regarding the overall reduction effect of cropland on the export of riverine organic carbon.

For a given amount of precipitation, the concurrence of high concentrations and low fluxes of riverine carbon in cropland-dominated basins could be best explained by the decreased water discharge due to agricultural activities. To test such a hypothesis, we analysed the runoff coefficients (i.e., area-specific runoff divided by rainfall) of the catchments in both the sampling year and historical period (Figure 8). It is clear and consistent that the runoff coefficients are negatively correlated with the percentage of cropland area in the catchment. Therefore, agricultural land use played a persistent role in reducing the runoff and riverine carbon flux in the SRB.

Annual water consumption accounts for approximately 35% of the water resources (the sum of groundwater and surface water) in the SRB on average (Figure 9), of which more than 70% is used for irrigation. Thus, cropland was the dominant factor responsible for the marked decrease in the runoff yield (SRLRC, 2004; Yang, 1994). In addition, the proportion of the reservoir impoundment volume to the total volume of water resources is much lower (less than 12%) than that of water consumption, mainly for irrigation (approximately 35%), in all the studied years (Figure 8). Therefore, reservoir or dam construction should have little effect on the total volume of river water discharge at an annual scale.

Sedimentation caused by reservoir or dam construction may generally lead to a large decrease in the POC flux of the river. Since the 1950s, the deposition of sediment in the small-sized reservoirs (with drainage area of 565 km²) in the Woken basin has been estimated to be 5.3 million tons (Tian & Li, 2008). The percentage contribution of annual discharge in 2008 to the total discharge from 1956 to 2008 is approximately 1.0%. On the basis of this weighed percent contribution of sediment deposition, 53,000 tons or 93.8 t/km² of sediment was trapped in these reservoirs in 2008. Applying this sediment accumulation rate to all the reservoirs (with a total drainage area of 2,608 km²) in the Woken basin, the amount of trapped sediment would total 244,630 tons. If POC% in the sediment ranges from 1.5–2.5%, the trapped POC would reach 3,669.5–6,115.7 tons, which is equivalent to 0.33–0.56 t·km⁻²·year⁻¹. Therefore, even if this trapped POC is considered, the export rate of total organic carbon from the cropland-dominated Woken basin

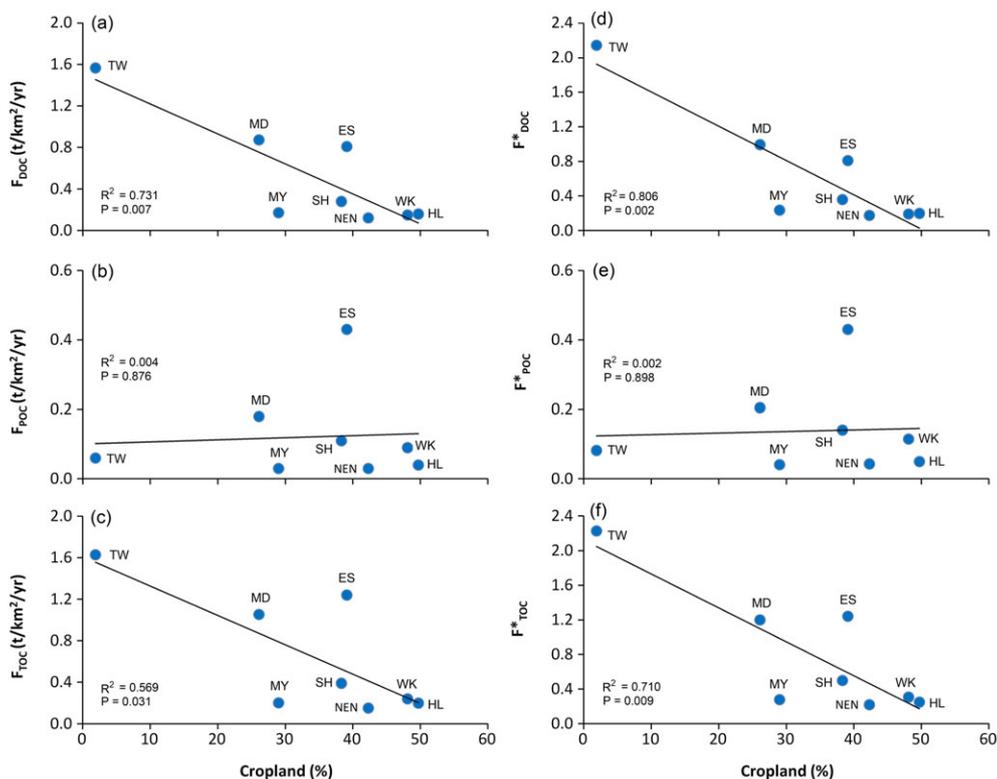


FIGURE 6 Relationships of cropland percentage with fluxes of riverine (a) DOC, (b) POC, and (c) TOC. F^* in (d), (e), and (f) denotes that the riverine organic carbon flux is divided by the normalized precipitation. The capital letters in the graph denote the basin names, see Figure 5 for explanation. DOC = dissolved organic carbon; POC = particulate organic carbon

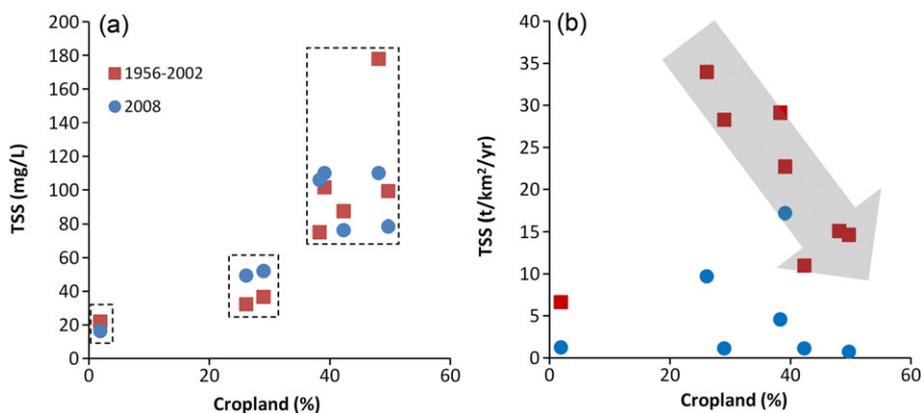


FIGURE 7 Relationships of cropland with TSS (a) concentrations and (b) loads in the subbasins for the long-term (1956–2002) average and for the sampling year 2008. Subbasins with a similar cropland percentage are grouped in a dashed line box. TSS = total suspended sediment

would be $0.57\text{--}0.80\text{ t}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$. This rate is still much lower than those from the forest-dominated Tangwang basin ($1.63\text{ t}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$) and the Mudan basin ($1.05\text{ t}\cdot\text{km}^{-2}\cdot\text{year}^{-1}$), which have small cropland areas (Table 2). In another cropland-dominated basin, the Hulan basin, which has not experienced significant reservoir construction, the riverine organic carbon flux is much lower than those from the Tangwang and Mudan basins (Table 2).

Note that the POC volume trapped in reservoirs in the Woken basin was estimated using the sedimentation rate from reservoirs in a typical cropland area, and the rate was much higher than that in

noncropland areas. In addition, the POC% in trapped sediment was assigned an upper-limit value (1.5–2.5%) to calculate the trapped POC. Hence, the estimated values of trapped sediment and POC can be considered upper bounds.

Cropland may cause more soil erosion when deposited in colluvial form on some slopes. This contribution is not directly estimated due to the lack of an obvious geomorphic indication. However, we argue that no matter how much eroded soil remains in colluvium, its final fate is river transport followed by reservoir or floodplain deposition or export from the river basin. All these processes are

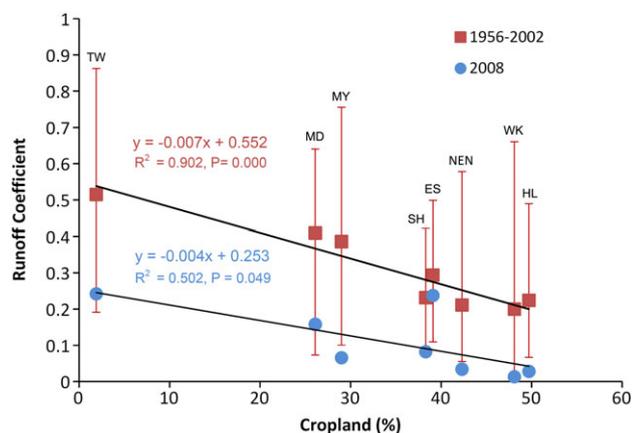


FIGURE 8 Relationship between cropland percentage and annual runoff coefficient in the Songhua River basin (runoff coefficient = runoff/basin area/precipitation). The two endpoints of each vertical line in the graph represent the maximum and minimum values in the period of 1956–2002, respectively. The rectangle represents the average value. The capital letters in the graph denote the basin names, same as in Figure 5. Note that the lower precipitation results in a lower level of runoff coefficient in 2008 but the relationship displayed between runoff coefficient and cropland percentage is rather similar to the long-term record (see the linear regression lines)

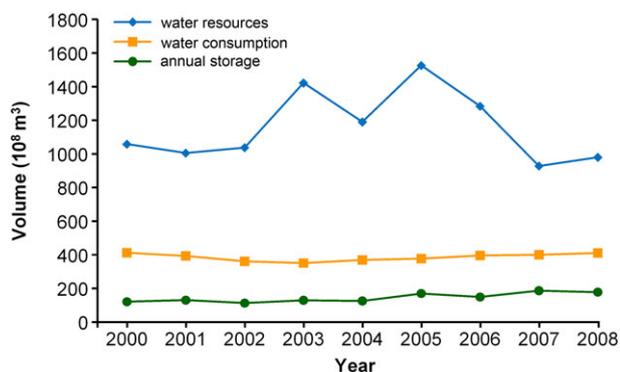


FIGURE 9 Water consumption, annual storage and water resources in the Songhua River basin (2000–2008). Data used from the annual report of China water resources by the Ministry of Water Resources of China

reflected in the 52-year historic record of riverine fluxes and sediment deposition in the reservoirs of the study area; therefore, these processes and fluxes are all included in the above estimation.

It is important to note that less severe heavy rainfall and flatter topography might be partially responsible for the reduction in the riverine organic carbon flux in the SRB. As has been observed, the soil erosion intensity and the sedimentation rate in the SRB are the lowest among those in the large river basins in China, such as the Pearl River basin (Tian, Zhang, Li, & Meng, 2006), where intense rainfall occurs frequently. In summary, massive withdraws of river water for cropland use and less severe rainfall in the region led to a reduction in the riverine carbon flux that suppressed the soil erosion or carbon export from cropland.

5.4 | Uncertainties in riverine organic carbon flux estimates and a subbasin comparison

At the JMS station, continuous records of daily discharge and TSS concentrations provided an opportunity to more accurately estimate the riverine organic carbon flux, which could be used as a reference flux (Table 2). For those sites with low-frequency sampling, flux estimation based on an interpolation method (Equation 1) may potentially result in large uncertainties depending on how close the measured concentrations in the water samples are to the average value in the river. At the JMS station, two sampling schemes (i.e., four-season sampling and high-frequency sampling) were independently performed in the same hydrological year. The DOC and POC fluxes derived from seasonal sampling were 6.7% and 21.4% lower than the reference fluxes (Table 2), respectively. These deviations in flux estimates can be considered satisfactory according to common practice. Thus, our seasonal samplings have indeed captured the representative ranges of the hydrologic conditions.

The SRB exhibited large annual variations (32.6% *rsd* from 1956 to 2002) in precipitation or discharge (SRLRC, 2004). Both annual precipitation and the annual flux of riverine carbon export were lower than average during the sampling year. Nevertheless, the spatial and seasonal distributions of precipitation, discharge, and sediment loading were similar to those based on historic records (Lu et al., 2013). Furthermore, the variations in the riverine organic carbon content in response to discharge or TSS were similar in the SR main stem and the tributaries, and they were comparable to those in other large rivers, such as the Xijiang River (Sun et al., 2007). This finding suggests that the relationship between the riverine organic carbon concentration and precipitation observed in this study should be widely applicable in both high- and low-precipitation years in the SR. Therefore, the low carbon flux in the sampling year should have no significant influence on subbasin comparisons, and the land-use effect on the riverine carbon flux revealed by this study can be considered reliable.

5.5 | Effects of agricultural activities on regional and global carbon budgets

Numerous studies based on local and plot or site scales have found that tillage may destroy the soil aggregate and intensify the mobilization and mineralization of soil organic matter, which can result in greater SOC export to aquatic systems (e.g., Chantigny, 2003; Ding et al., 2013; Don, Scholten, & Schulze, 2009; Liu et al., 2006; Xu et al., 2010). However, this conclusion may not be applicable at the basin scale in cases where irrigation significantly reduces the total discharge, as observed in this study. Here, we argue that the reduction effect of cropland or irrigation on riverine organic carbon should receive additional attention because it has not been considered in the global carbon cycle (Battin et al., 2009; Regnier et al., 2013; Tranvik et al., 2009). Currently, agriculture accounts for approximately 38% of the total terrestrial surface of the Earth (Foley et al., 2011), and more than 10% of global runoff is used for irrigation, which accounts for 70% of global water withdrawals (Hanasaki, Inuzuka, Kanae, & Oki, 2010). With the increasing demand for food

and growing economies at the global scale, the cropland area and irrigation are expected to increase in the future.

6 | CONCLUSIONS

The average concentrations of DOC and POC in the SR were 5.87 and 2.36 mg/L, which were higher than those reported in most rivers worldwide due to widely distributed organic-rich black soil in the basin. Both the DOC and POC concentrations exhibited increasing trends as the proportion of cropland increased, but the area-specific carbon fluxes displayed inverse trends. The strong and positive correlation between POC and TSS suggested that the organic carbon in the SR was derived mainly from the soil. This finding is consistent with the results of the stable carbon isotope and C/N ratio analyses, which indicated the dominance of terrestrial vegetation inputs, as well as some aquatic biomass inputs. The PCA of multiple environmental parameters revealed that hydrology and land use are the most important controls of riverine carbon transport. Through subbasin comparisons and analyses of historical hydrological data, we found that agricultural activities may intensify the soil erosion modulus, but irrigation can considerably reduce river discharge and lead to an overall decrease in riverine carbon export in the SRB. This observation suggests that agricultural activities may not necessarily lead to an increase in riverine carbon export, as previously reported, and caution must be taken in future studies.

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SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

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