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ORIGINAL ARTICLE

Change in carbon flux (1960–2015) of the Red River (Vietnam)

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Abstract

Global riverine carbon concentrations and fluxes have been impacted by climate and human-induced changes for many decades. This paper aims to reconstruct the longterm carbon concentrations and carbon fluxes of the Red River, a system under the coupled pressures of environmental change and human activity. Based on (1) the relationships between particulate and dissolved organic carbon (POC, DOC) or dissolved inorganic carbon (DIC), and suspended sediments (TSS) or river water discharge and on (2) the available detailed historical records of river discharge and TSS concentration, the variations of the Red River carbon concentration and flux were estimated for the period 1960–2015. The results show that total carbon flux of the Red River averaged 2555 ± 639 kton C year^{-1}. DIC fluxes dominated total carbon fluxes, representing 64% of total, reflecting a strong weathering process from carbonate rocks in the upstream basin. Total carbon fluxes significantly decreased from 2816 kton C year^{-1} during the 1960s to 1372 kton C year^{-1} during the 2010s and showed clear seasonal and spatial variations. Organic carbon flux decreased in both quantity and proportion of the total carbon flux from 40.9% in 1960s to 14.9% in 2010s, reflecting the important impact of dam impoundment. DIC flux was also reduced over this period potentially as a consequence of carbonate precipitation in the irrigated, agricultural land and the reduction of the Red River water discharge toward the sea. These decreases in TSS and carbon fluxes are probably partially responsible for different negatives impacts observed in the coastal zone.

Keywords Carbon fluxes · Dam impoundment · POC · Red river · Vietnam

Introduction

A major component of global biogeochemical cycles, rivers play an important role in connecting terrestrial, oceanic and atmospheric carbon reservoirs (Meybeck and Vorosmatry 1999). Tropical rivers are associated with the highest carbon flux, thus play a critical role in the total global fluvial carbon flux (Huang et al. 2012).

Tropical riverine carbon flux is closely associated with suspended solid flux. The major Asian rivers contribute a large proportion (50%) of sediment to global sediment load (Ludwig et al. 1996), however, many Asian rivers have seen dramatic changes in sediment fluxes over the past decades

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due to environmental changes (climate and human-induced changes). Climate, notably cyclones or storms, plays an important role in the erosion of soils and organic carbon fluxes (Dawson et al. 2008; Lloret et al. 2011). In addition, human activities have altered riverine suspended solid and carbon fluxes in this region. Rapid environmental changes such as those resulting from forest conversion or those driven by rapid economic development have resulted in an increasing magnitude and extent of soil erosion and sediment loads. On the other hand, reservoir construction and the reduction in rainfall associated with climate change have led to dramatically reduced sediment loads in some of the large Asian rivers, e.g., the Yellow River (Wang et al. 2007; Cai et al. 2008; Miao et al. 2011), the Yangtze River (Yang et al. 2006; Zhang et al. 2006) and the Pearl River (Zhang et al. 2007; Sun et al. 2010). Such dramatic reductions in sediment load also have a significant effect on the associated elements such as carbon, phosphorus and iron, affecting the ecological and environmental status of downstream estuaries, coastal regions and continental shelf areas (Chen 2000).

Several studies have discussed long-term observations of riverine inorganic and/or organic carbon in Europe such as the Tees, Wear and Coquet Rivers (Worrall et al. 2003) or in the Americas such as the St. Lawrence River (Helie et al. 2002), the Amazon River (Botta et al. 2002), and the Mississippi River (Raymond et al. 2008). However, less is known on the large rivers in Asia, except for some rivers originating in China such as the Pearl River (Cai et al. 2004), the Xijiang River (the largest tributary of the Pearl River) (Sun et al. 2010), the Yellow River (Ran et al. 2015) and the Mekong River (Li and Bush 2015).

The Red River (Vietnam and China) is characteristic of a South-East Asian river in that it is strongly impacted by both natural conditions and human activities in its basin. Previous studies have examined the long-term impacts of human activities on hydrology and suspended sediment and associated nutrients (N, P) loads of the Red River (Dang et al. 2010; Vinh et al. 2014; Le et al. 2015a) and short-term organic carbon variation (Le et al. 2017). Previously, the Red River was ranked 15th of the World's rivers in terms of suspended sediment load (Syvitski and Milliman 1992). However, due to the recent man-made changes in the Red River watershed, like damming, suspended sediment load of this river has dramatically decreased (Dang et al. 2010; Vinh et al. 2014), as has been observed for many Asian rivers (Huang et al. 2012). This large decrease in suspended solids has also probably led to a reduction in the amount of associated carbon in the rivers. The large Asian rivers originating in the Himalayan Mountains and on the Tibetan Plateau play an important role in the global carbon cycle through carbon consumption via chemical weathering and organic carbon burial in ocean environment. However, little information is available on longterm carbon fluxes in this region, except some studies on rivers in China (Cai et al. 2004; Sun et al. 2010; Ran et al. 2015; Li and Bush 2015). Thus, studies of longterm variations of suspended solids and carbon fluxes from this region are a priority to improve the estimates of global riverine carbon cycle.

The present study aims to: (1) reconstruct the longterm variation of carbon concentrations (DOC, POC, DIC) and carbon fluxes of the Red River system over the period 1960–2015; (2) analyse the influence of some factors including natural conditions and human activities in the Red River basin on the riverine carbon fluxes. To attain this objective, we applied the previously demonstrated linear relationship between POC, DOC and DIC with water discharge and TSS concentration (Le et al. 2015a, 2017) to the available daily, historical records of river water discharge and suspended solids concentrations.

Study site

Lithological and geomorphological characteristics

The Red River basin (Fig. 1) is located in South-East Asia (from 20°00–25°30 N; from 100°00 to 107°10 E). It is the second largest river in Vietnam and the basin covers three countries, including China, Vietnam and Laos, with a total area of 156,448 km^2. The river rises in a mountainous region with a mean elevation of about 2000 m (IMRR 2010) in Yunnan province, China where its name is Yuan River and when entering into Vietnam, it is renamed Cai, Thao, or Hong River. The Thao River has two major tributaries, the Da and Lo rivers, downstream of which the main branch is named Hong (Red) River which flows eastward into the Tonkin Gulf (South Asian Sea) though four branches named Day, Lach Gia, Ba Lat, and Tra Ly. The main branch is about 1140 km length. In the Vietnamese section, a half of the basin is the Red River delta, which covers a flat and lowland area with elevation ranging from 0.4 to 12 m (Dang 2001).

The upstream part of the Red River basin is tectonically active and has high erosion rates (Fullen et al. 1998). Le et al. (2007) indicated that the geologic substratum of the upper basin is dominated by consolidated paleozoic sedimentary rocks of complex lithology, with variable contributions of mesozoic silicic or carbonate rocks (Fig. 2). The three main tributaries of the Red River (Thao, Da and Lo) drain their watersheds that have considerable differences in their lithology. The Red River main channel basin is dominated by metamorphic rocks, except in the upper reaches where Mesozoic sedimentary deposits are found. The Da drainage basin is composed of sedimentary rocks from the Mesozoic and Paleozoic period with minor felsic intrusions. The Lo drainage has low-grade metamorphic rocks and Proterozoic to Paleozoic sedimentary rocks with some granitic

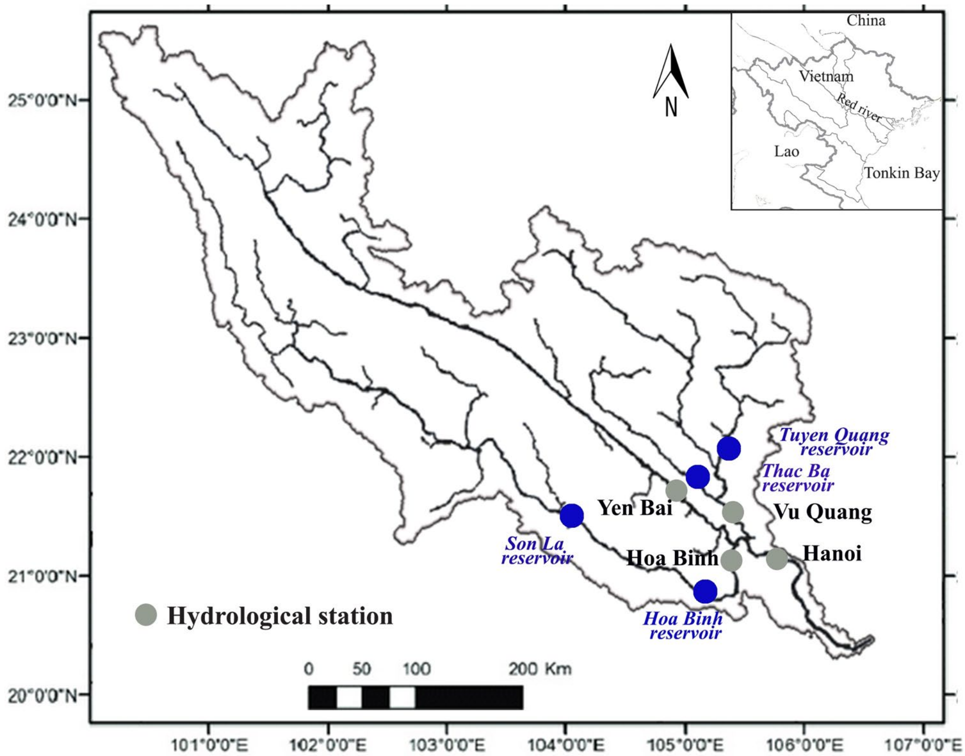

Fig. 1 The Red River basin and four main hydrological stations. Insert provides the regional location of the study site. Blue points indicate the location of the four main reservoirs in the Vietnamese section of the system and grey points indicate the location of the four hydrological stations used in this work

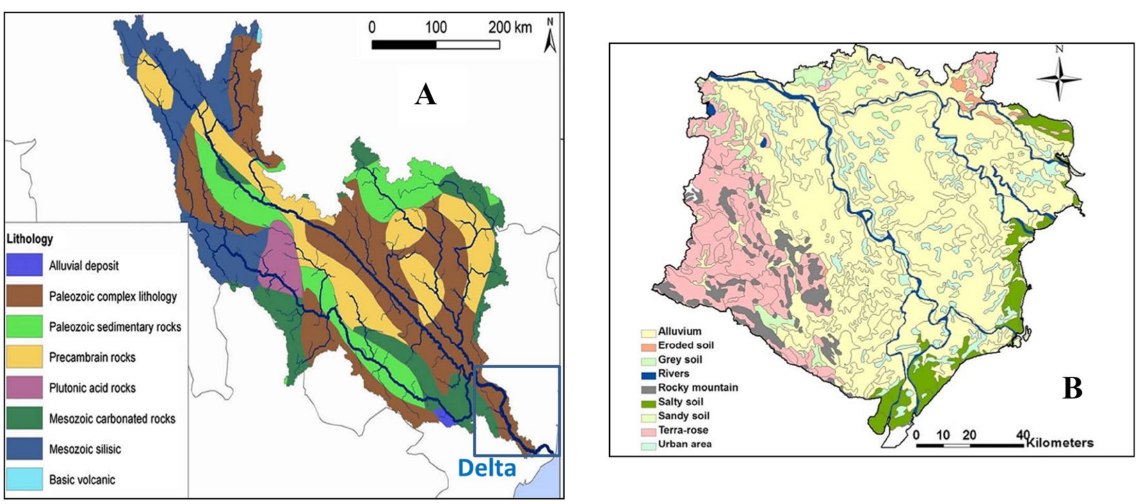

Fig. 2 Lithological characteristics of the Red River: **a** in the upstream of the Red River (Le et al. 2007); **b** in the Delta of the Red River (Luu 2010). Noted that the maps for the upstream part and the Delta area are in different scales

intrusions (Moon et al. 2007) (Fig. 2a). This is in contrast with the lithological characteristics of the Red River delta area where alluvial deposits dominate (MOSTE 1997; Luu 2010) (Fig. 2b). Moon et al. 2007 indicated that silt and clay are the most abundant sediments while fine sand is present in small amounts from the Holocene succession in this area (Moon et al. 2007) (Fig. 2). This indicates that the supply source is weathered and that transport from far depositional centers or from the reworking of Pleistocene silty, clayey layers were dominant. Main colour of the Holocene sediments is reddish brown.

Soils in the upper basins are typically Ultisols (by US classification) or "red soil" (by Chinese soil classification), while in the delta area alluvial soils dominate (MOSTE, 1997). Luu (2010) indicated that alluvium soils with high alluvium content occupy nearly 80% of total area of the delta

(Fig. 2b). Indeed, alluvium comprises a fine, muddy, river-bornesediment which is slightly acidic. Beside alluvium, terra-rose soils associated with rocky mountain soils are found in the west, sandy soils in the north-west and salty soils are found along the coastline of the Red River Delta.

Climate

The climate in the Red River basin is sub-tropical East Asia monsoonal type, where the South West monsoon from May to October brings warmer and wetter weather during the rainy season and the North East monsoon from November to the following April brings cooler and dryer weather during the dry season. Annual rainfall varies from 700 to 4800 mm year^{-1} across the basin with about 80% of rainfall occurring during the rainy season (Nguyen et al. 2007). This climate results in a hydrologic regime that is characterized by high runoff during the rainy season and low runoff during the dry season. The average daily temperature varies from 14 to 27 °C and relative humidity is high throughout the year, averaging 82–84%.

Hydrology

Daily river discharges at the outlets of the three tributaries Thao (at Yen Bai station), Da (at Hoa Binh station) and Lo (at Vu Quang station) and of the downstream main axe of the Red River (at Hanoi station) were collected for the period 1960–2015 by the Ministry of Natural Resources and Environment of Vietnam (MONRE) (1960–2015) (Fig. 1). They averaged 720 ± 159 m^3 s^{-1}, 1670 ± 284 m^3 s^{-1}, 1020 ± 227 m^3 s^{-1} and 2500 ± 477 m^3 s^{-1}, respectively. Of the three upstream tributaries, the Da River accounts for a half of the total Red River discharge. Long-term data on the decadal averages of water discharge showed clear decreases in the Thao, Da tributaries and in the main downstream Red River in the 1960–2015 period (Table 1).

Gao et al. (2015) revealed that El Niño and La Niña events induced respectively, extreme low and extreme high discharge from the Red River into the Tonkin Gulf over the long period 1960s–2010s. Indeed, the minimum water level in the Red River at Hanoi station was 1.46 m and 0.4 m in 2006 and 2010, respectively. Similarly, extreme low river discharges of the main axe Red River at Lao Cai (130 m^3 s^{-1} in March 2010) and Son Tay (368 m^3 s^{-1} in May 1960 or 380 m^3 s^{-1} in March 2002) hydrological stations were observed during El Nino years (Quach 2011a, b; Pham 2015). In contrast, extreme high river discharge (37,800 m^3 s^{-1}) of the Red River at Son Tay station was observed in August 1971. Serious floods causing dam failures were noted in 1913, 1915, 1945 and 1971 when the water level in Hanoi reached, respectively, 11.35 m, 11.2 m, 11.45 m, and 13.3 m (To et al. 2000).

Reservoir impoundment

Reservoirs play an important role in the socio-economic development of both China and Vietnam. Reservoirs are used for flood control, irrigation, hydropower, water supply and flow management. In both Chinese and Vietnamese sectors of the Red River basin, a series of reservoirs and dams are in operation.

Since 2007, a series of small and medium size reservoirs/dams has been impounded for hydropower in the upstream Chinese sector of the Red River. Some dams have been constructed on the upstream Thao River. The two main intercepting dams, named Namsha and Madushan (located 140 km from the Vietnamese border to China), have a capacity of 130 and 300 MW, respectively (IMRR 2010). Eleven small hydrological dams are located on the upstream Da River and at least eight hydropower reservoirs are located

Table 1 Decade average of river discharge and total suspended solids concentrations of the three tributaries Thao, Da, Lo Rivers and the main branch Red River during the period 1960–2015

Hydrological station	River	Variables	1960	1970	1980	1990	2000	2010
Yen Bai	Thao	River discharge, m^3 s^{-1}	774 ± 126	818 ± 208	674 ± 103	752 ± 137	708 ± 140	517 ± 34
		TSS concentration, mg L^{-1}	883 ± 274	965 ± 79	1054 ± 454	1479 ± 344	928 ± 297	278 ± 62
Hoa Binh	Da	River discharge, m^3 s^{-1}	1720 ± 219	1706 ± 213	1544 ± 236	1840 ± 301	1798 ± 230	1276 ± 216
		TSS concentration, mg L^{-1}	475 ± 144	477 ± 75	354 ± 169	74 ± 15	56 ± 26	15 ± 6
Vu Quang	Lo	River discharge, m^3 s^{-1}	955 ± 144	1077 ± 227	1085 ± 165	1098 ± 158	897 ± 213	990 ± 439
		TSS concentration, mg L^{-1}	158 ± 28	167 ± 54	163 ± 56	207 ± 51	140 ± 53	56 ± 22
Hanoi	Main downstream Red River branch	River discharge, m^3 s^{-1}	2610 ± 317	2821 ± 380	2607 ± 439	2660 ± 390	2310 ± 310	1673 ± 262
		TSS concentration, mg L^{-1}	505 ± 112	554 ± 105	434 ± 116	389 ± 74	343 ± 172	77 ± 8

on the upstream Lo River (Ha and Vu 2012) (Supplementary Materials Table S1).

In Vietnam, there are four large dams/reservoirs along the Red River including: the Hoa Binh (in operation since 1989) and Son La (in operation since 2010) reservoirs on the main axe of the Da River, the Thac Ba (in operation since 1975) and the Tuyen Quang (in operation since 2010) reservoirs on the Lo River. Several other reservoirs are presently under construction such as the Huoi Quang and Lai Chau reservoirs (projected to be in operation in end 2017) on the Da River.

Population

Population density is distributed unevenly throughout the whole basin with a low density (100 inhabitants km^{-2}) in the upstream mountainous region and a very high density (1300 inhabitants km^{-2}) in the delta region. Presently, about 70% of the total basin population lives in rural areas although both total population and urban population have increased rapidly over the last decades (General Statistics Office of Vietnam 2010; Le et al. 2015a).

Land use

Land use differs across the three upstream sub-basins Thao, Da, Lo and in the Delta. Whereas, forest and industrial cash crops mainly dominate in the upstream river basin, the delta area is characterised by paddy rice fields (66.3%) (Le et al. 2017). Over the long-term, changes in land use have been observed for the three upstream sub-basins and the Delta: the forest area in the delta had been quite stable (about 3%) for several centuries but the deforestation in the plateau and mountainous regions dramatically accelerated during the war years (1970s). This continued during the following period of rapid population growth and economic development (1990s), notably for the Da basin (from 62% in 1960s to 14% in 2010s) and the Thao basin (66.6% in 1960s to 24% in 2010s) (Table 2). Agricultural land including rice field increased from 32.2–52.4%; 15.3–24.8%; 56.5–74.8% to 55.6–72.1% for the three basins Thao, Da and Lo and the Delta area, respectively, during the period 1960s–2010s (Table 2) (Le et al. 2015a).

Agricultural practices

Agricultural activity in the Red River basin in the Vietnamese section has considerably changed over the last 50 years, accompanying the demographic development after the war (Le et al. 2015a). Considerable increases in cereal production have been made possible through the use of chemical fertilizers and manures from livestock; however, the excessive fertilizer utilization has also led to increase soil organic matters loss through the leaching and erosion processes.

Methods

Data collection

Data on daily and monthly river flows and suspended solid concentrations in the Red River at four hydrological stations at the outlets of the Thao (Yen Bai station), Da (Hoa Binh station), Lo (Vu Quang station) rivers and main axe of the downstream Red River (Hanoi station) were

Table 2 Change in land use in the three tributary sub-basins and the Delta area of the Red River during the period 1960–2010

	Basin area, km^2	% Land cover	1960	1970	1980	1990	2000	2010
Thao	61,169	Forest	66.6	52.4	38.2	24.0	24.0	24.0
		Bare land	0.5	13.7	25.7	39.3	26.2	22.2
		Agricultural land	32.2	33.0	35.1	35.5	48.4	52.4
		Urban	0.7	0.8	1.0	1.3	1.4	1.4
Da	51,285	Forest	62.6	46.4	30.2	14.0	14.0	14.0
		Bare land	22.0	37.8	53.0	68.9	62.8	60.9
		Agricultural land	15.3	15.7	16.6	16.8	22.9	24.8
		Urban	0.2	0.2	0.2	0.3	0.3	0.3
Lo	34,559	Forest	38.0	32.0	26.0	20.0	20.0	20.0
		Bare land	5.2	9.7	12.1	17.3	4.7	4.6
		Agricultural land	56.5	57.9	61.4	62.1	74.7	74.8
		Urban	0.3	0.4	0.4	0.5	0.6	0.6
Delta	9435	Forest	3.0	3.0	3.0	3.0	3.0	3.0
		Bare land	39.4	36.9	32.5	22.2	16.0	13.6
		Agricultural land	55.6	57.0	60.5	69.5	73.0	72.0
		Urban	2.0	3.1	4.0	5.3	8.0	11.4

collected for the period from 1960 to 2015 as presented in "Hydrology" section (Fig. 1) (MONRE 1960–2015).

From 2008 to 2015, monthly sample collection and analysis was conducted to obtain DOC, DIC and POC concentrations in river surface water at the four main hydrological stations, except in 2011. The detail of sampling, analysis methods and a part of the organic carbon content results (2008–2010) are presented in Le et al. (2017). The relationships between POC–TSS and POC/DOC and TSS are also presented in that article. Similarly, the monthly measured DIC concentrations for the period 2008–2015 at the same four stations of the Red River as were used in Le et al. (2015b) were used for determining DIC concentrations over the period 1960–2007.

Calculation of carbon concentrations and fluxes

Calculation of carbon concentrations

First, the missing daily POC concentrations for each of the four stations (Yen Bai, Hoa Binh, Vu Quang and Hanoi) for the period 1960–2015 were calculated using the linear relationship between POC concentrations and suspended solids as given by Le et al. (2017) and the daily suspended solids concentrations given by MONRE (1960–2015). After obtaining daily POC concentrations, missing daily DOC concentrations were determined based on the relationship between suspended solids and the DOC:POC ratio as defined in Le et al. (2017). In parallel, the missing daily DIC concentrations in the studied period were obtained from the linear relationships between DIC and the river discharge, which were presented in Le et al. (2015b), and the data of daily river discharge given by MONRE (1960–2015) for each station. All equations used for calculations of POC, DOC and DIC concentrations are presented in Table 3.

Calculation of carbon fluxes

Annual fluxes of POC, DOC and DIC were calculated for each hydrological station as the sum of all daily POC or DOC or DIC flux during a year (365 days) where the daily POC or DOC or DIC flux at each station was calculated by multiplying the daily (POC or DOC or DIC) concentrations and daily river discharge for the studied period. The total annual carbon flux of each year is the sum of the annual fluxes of POC, DOC and DIC for each station.

The calculation methods for DOC, DIC and POC concentrations based on TSS and water discharge rely upon the POC/TSS, DOC/POC and DIC/discharge relationships derived over a short period have also been applied in other research, such as in the Xijiang River (Sun et al. 2010). Recently, the relationship between river discharge/suspended solids and river discharge/carbon concentration combined with daily river discharge has been used to calculate longterm carbon fluxes of the tropical Tana river (Geeraert et al. 2018).

Results

Suspended solids

Long-term data on the decadal average of total suspended solids (TSS) concentrations showed a clear decrease for the three tributaries and the main branch of the Red River over the period 1960–2010s (Table 1). TSS of the three tributaries and of the main branch of the Red River varied over three orders of magnitude from 8 to 2144 mg L^{-1}. The highest concentration of TSS occurred at Yen Bai station in the Thao River in 1986, whereas, the lowest was observed at Hoa Binh station in the Da River in 2015 after the complete impoundment of the Son La dam. Within the three tributaries, over the 1960–2015 period, mean TSS concentrations

Table 3 Equations of POC–TSS, POC/DOC–TSS and DIC–Q relationships for calculations of missing POC, DOC and DIC concentrations of the Red River in the period 1960–2015

Relationship	Station	Equations	R^2	Refs.
POC and TSS	Yen Bai	$Y=0.01x+0.37$	0.87	Le et al. (2017)
	Hoa Binh	$Y=0.01x+0.29$	0.44	
	Vu Quang	$Y=0.01x+0.22$	0.90	
	Hanoi	$Y=0.01x+0.32$	0.86	
DOC:POC and TSS	Yen Bai	$Y=16.59x-0.58$	0.68	Le et al. (2017)
	Hoa Binh	$Y=3.5x-0.03$	<0.1	
	Vu Quang	$Y=22.9x-0.66$	0.6	
	Hanoi	$Y=20.5x-0.58$	0.45	
DIC and river discharge	Yen Bai	$Y=-0.01x+20.98$	0.41	Le et al. (2015b)
	Hoa Binh	$Y=-0.0005x+17.16$	<0.1	
	Vu Quang	$Y=-0.001x+21.55$	0.42	
	Hanoi	$Y=-0.0007x+21.86$	<0.1	

were the highest at the Thao station (966 ± 432 mg L^{-1}) where less dams are present along its main course. TSS concentrations in the Lo and Da Rivers were much lower and averaged 157 ± 59 and 297 ± 554 mg L^{-1}, respectively. In the main branch of the Red River at Hanoi station, TSS averaged 411 ± 170 mg L^{-1} over the period 1960–2015.

Additionally, TSS concentrations varied with seasons, being about 5 times higher during the rainy season for the Thao, Lo and Da Rivers. Mean TSS concentrations in the Thao, Lo and Da Rivers during the rainy season were 1495 ± 744 mg L^{-1}, 271 ± 112 mg L^{-1} and 83 ± 262 mg L^{-1}, respectively, whereas, the concentrations in dry season were only 301 ± 159 mg L^{-1}, 35 ± 18 mg L^{-1} and 24 ± 48 mg L^{-1}, respectively. In the main branch of the Red River, TSS varied by only a factor of 3 between rainy and dry seasons (655 ± 276 mg L^{-1} and 171 ± 90 mg L^{-1}, respectively).

Little information exists concerning the characteristics of suspended materials in the Red River. A previous study (Borges et al. 2007) reported sands (63–500 µm) and silts (2–63 µm) were present in varying proportions: 99.5% and 3.5%, 69.8% and 27.3%, 97.6% and 0.9% at the Yen Bai (Thao river); at Vu Quang (Lo river) and at Lai Chau (upstream of the Hoa Binh station, Da River), respectively. The median diameter D50 of surface sediment averaged 0.35, 0.16 and 0.175 mm in the Da, Thao and Lo rivers, respectively (Ministry of Agriculture and Rural Development 2009) and it varied from 5 to 195 µm in the downstream, in the estuaries and coastal zones (Do et al. 2007). The red colour of the Thao river is due to the suspended solids that are rich in iron oxides and weathering products from lava and crystalline schist. The yellow colour of the Lo river is due to the presence of products of weathering from limestone and sandstone.

Organic and inorganic carbon concentrations

Organic carbon concentrations

Over the 55-year period, the calculated yearly DOC concentrations varied from 0.9 to 15.9 mg C L^{-1}, with a mean value of 3.5 ± 2.7 mg C L^{-1} for the whole Red River system. The mean yearly concentrations were highest at Hoa Binh on the Da River (6.2 ± 4.4 mg C L^{-1}) and lowest at Vu Quang on the Lo River (2.2 ± 0.4 mg C L^{-1}) (Fig. 3).

During the 1960–2015 period, the yearly average calculated POC concentrations ranged from 0.4 to 24.8 mg C L^{-1}, with an average of 4.9 ± 4.6 mg C L^{-1} for the whole Red River system. Within the three tributaries POC concentrations were the lowest at Vu Quang and Hoa Binh stations (2.2 ± 0.9 and 2.1 ± 1.5 mg C L^{-1}, respectively) and the highest (10.8 ± 5.2 mg C L^{-1}) at Yen Bai station (Fig. 3). The highest yearly value (24.8 mg C L^{-1}) was recorded at Yen Bai when the river discharge in the Thao River during the major flood of 1986 reached 901 m^3 s^{-1} and the TSS concentration peaked at 2144 mg L^{-1}.

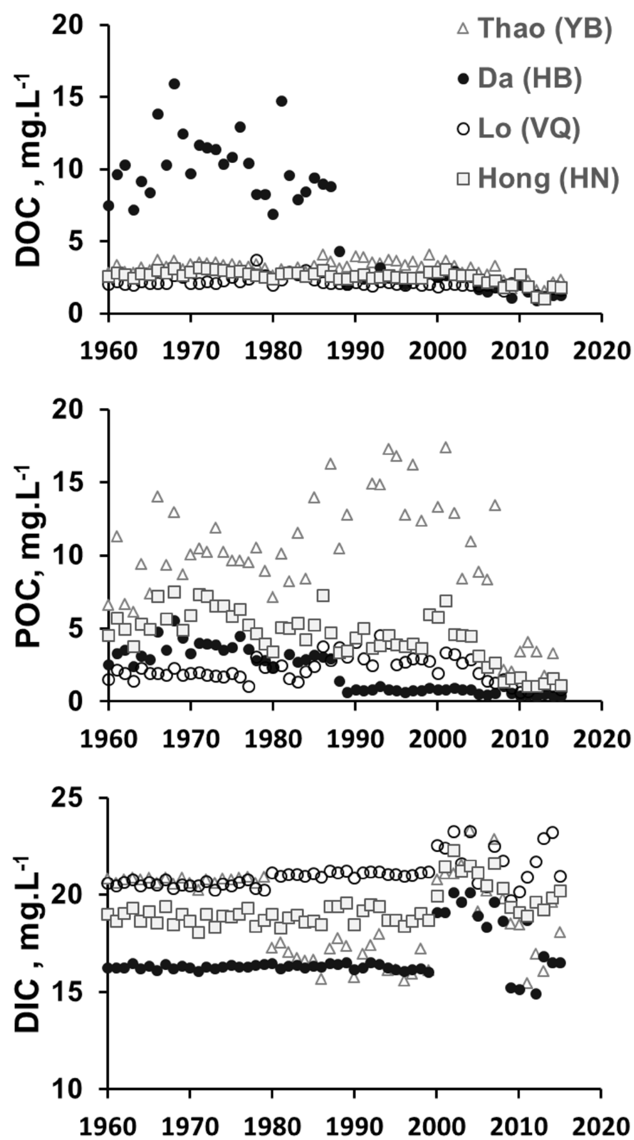

Fig. 3 Calculated DOC, POC and DIC concentrations at the 4 gauging stations

Inorganic carbon concentrations

The calculated yearly DIC concentrations varied by a factor of almost 2 (14.9–23.3 mg C L^{-1}) and the average value for the whole Red River was 19.1 ± 2.0 mg C L^{-1} during the 1960–2015 period. The lowest yearly average DIC concentration was observed at Hoa Binh station (16.8 ± 1.2 mg C L^{-1}) and the highest yearly average DIC value (21.0 ± 0.7 mg C L^{-1}) was measured at the Vu Quang station on the Lo River (Fig. 3).

Riverine carbon fluxes

Over the studied 55-year period, the calculated DOC fluxes varied over a large range in all three river tributaries (16–1884 kton C year^{-1}) (Fig. 4). Except for the Da River, which experienced a dramatic decrease from 1493 kton C year^{-1} (in 1981) to 94 kton C year^{-1} (in 1989), the DOC fluxes of the Thao and Lo Rivers were moderately stable as was the flux of the whole Red River (Fig. 4). For the whole Red River, the DOC fluxes averaged 282 ± 90 kton C year^{-1}.

Similar to DOC fluxes, POC fluxes of the three main tributaries fluctuated markedly from 7 to 1282 kton year^{-1} during the studied period. The highest mean value was in the main axe of the Red River at Hanoi station (546 ± 280 kton C year^{-1}) and the lowest at Vu Quang station (127 ± 66 kton C year^{-1}) (Fig. 4). For the whole Red River, POC fluxes averaged 637 ± 326 kton C year^{-1} and tended to decrease with time. Indeed, in the Da River, the mean POC flux during the period from 1960 to 1989 was 351 ± 138 kton C year^{-1}, this value decreased by a factor of 5 after the complete impoundment of the Hoa Binh reservoir (1991) and by a factor of over 20 after the construction of Son La reservoir (2010). For the Lo River, the POC fluxes decreased from 128 kton C year^{-1} in the period 1960–1970 to 35 kton year^{-1} in 2010–2015.

The DIC fluxes varied between 185 and 1843 kton C year^{-1} in the tributaries over the 55-year period, resulting a mean value of 1639 ± 261 kton C year^{-1} for the DIC flux of the whole Red River over this period. The DIC fluxes decreased slightly in the Thao and Da Rivers, however, in the Lo River, a slight upward trend was observed from 1960 to 2015. Overall, the DIC fluxes of the whole Red River displayed a decreasing trend, from 1663 ± 165 kton C year^{-1} in 1960s to 1168 ± 205 kton C year^{-1} in 2010s (Fig. 4).

Eventually, for the whole Red River system, total carbon fluxes (DOC + POC + DIC) over the studied period showed a downward tendency. The mean value of the total carbon flux of the Red River was 2555 ± 639 kton C year^{-1} (equivalent to 16.3 ± 4.1 ton C km^{-2} year^{-1}) with the range of 993– 3903 kton year^{-1} (from 6.4 to 24.9 ton C km^{-2} year^{-1}). Of which, the mean annual DIC flux accounted for the highest proportion in the mean annual total carbon flux of the Red River (1639 ± 261 kton C year^{-1}) (64%), whereas, the smallest one was the DOC flux (282 ± 90 kton C year^{-1}) (11%) during the whole study period.

Discussion

Influence of dams on temporal variation of the Red River carbon concentrations and fluxes

Vorosmarty et al. (2003) emphasised the prominent role of large reservoirs in trapping suspended materials of numerous rivers, especially in Asia. Our results suggest that it is also the case for the Red River, considering the development of a series of hydroelectric dams in the upstream Red River both in China and Vietnam over the last few decades

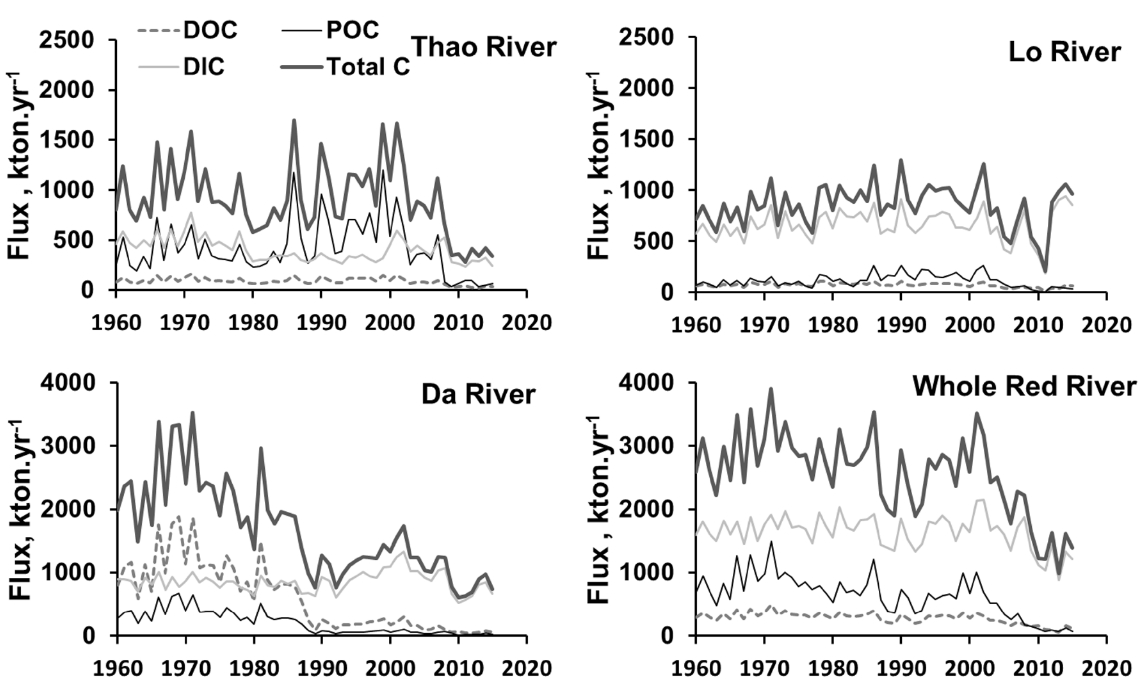

Fig. 4 Carbon fluxes of 3 upstream tributaries (at stations Yen Bai, Hoa Binh, Vu Quang) and of the whole Red River

(Pham 1998; Le et al. 2007). Prior to 1990, only two large reservoirs (Thac Ba, Hoa Binh) with a total storage capacity of about 24 Mm^3 were present, however, they were considered as significant sediment traps for the Da and Lo rivers (Ngo and Tran 1998; Le et al. 2007). From 2008 onwards, a series of reservoirs have been constructed in both the Chinese (such as the Nanshan dam in the upper reach of the Thao River, the Zhongaiqiao reservoir in the upper of the Da River, etc.) and in the Vietnamese (Lai Chau, Son La and Tuyen Quang dams) sections of the Red River system. These dams have impacted water and sediment discharges downstream (Ha and Vu 2012; Ngo et al. 2014; Lu et al. 2015) with significant sediment deposition being observed in the reservoirs (Dang et al. 2010; Vinh et al. 2014; Lu et al. 2015). Our calculations show that the TSS flux of the whole Red River decreased dramatically during the period from 1960 to 2015, from 822 kton C $year^{-1}$ (in 1968) to 94 kton C $year^{-1}$ (in 2015) corresponding to the increase of the reservoir numbers along the river. The present specific TSS fluxes of the Red River (598 ton km^{-2} $year^{-1}$ in 2015) are relatively low compared to most Asian river systems (Liu et al. 2009), as a consequence of dam impoundments; however, it is still higher than the mean global annual yield (190 ton C km^{-2} $year^{-1}$) (Milliman and Farnsworth 2011).

Previous studies emphasised that riverine POC fluxes are tightly correlated to riverine TSS fluxes (Ludwig et al. 1996; Ni et al. 2008; Zhang et al. 2009; Sun et al. 2010). It is also the case of the Red River (Le et al. 2017). During the period 1960–2015, the significant reduction in TSS flux of the Red River resulted in a clear decrease in POC fluxes for all tributaries and the whole Red River (Table 4). POC fluxes decrease together with the reduced SS fluxes have also been observed in other dammed rivers such as the Yellow River (Wang et al. 2012) and the Changjiang (Yangtze) River (Wang et al. 2012; Ji et al. 2016) and the Amazon River (Latrubesse et al. 2017). Using a spatially explicit modelling approach to predict global in-reservoir primary production, mineralization and burial of organic carbon, Maavara et al. (2017) found that dams decreased organic carbon delivery to the oceans by 13% at global scale in 2000.

For the whole Red River system, the DOC fluxes fluctuated moderately and showed a slight decrease over the studied period, except for the Da River. Of the three tributaries (Thao, Lo and Da), the DOC fluxes of the Da River at the Hoa Binh station underwent a more dramatic decrease than other three stations (see Fig. 4). In our calculations, the DOC concentrations were obtained based on the relationship DOC/POC vs TSS (Le et al. 2017). Thus, any decrease of TSS flux, from, for example dam impoundment, translates into a decrease in POC and DOC fluxes, especially for the Da River where TSS flux decreased the most strongly. This is consistent with what has been observed in the Yellow River, where dam impoundment has caused decreases in TSS and DOC loads due to the DOC adsorption onto large particles (Moreira-Turcq et al. 2003; Zhang et al. 2013). However, other studies have revealed contrasting effects of dams on riverine TSS and DOC loads. Work from the Xijiang River reported that reservoir impoundments reduced TSS loads which stimulated photosynthesis and thus contributed to increase DOC concentrations (Spitzy and Leenheer 1991; Sun et al. 2010).

Concerning inorganic carbon, the decreased riverine DIC fluxes after dam impoundment have been observed elsewhere in Asia. For example, in the Yellow River during

Table 4 Carbon fluxes at the four examined hydrological stations and for the entire Red River during different periods

Stations	Carbon fluxes (kton C $year^{-1}$)	Period					
		1960–1969	1970–1979	1980–1989	1990–1999	2000–2009	2010–2015
Yen Bai (Thao R.)	DOC	101 ± 30	104 ± 27	83 ± 26	111 ± 26	83 ± 32	34 ± 8
	POC	399 ± 195	392 ± 122	418 ± 285	683 ± 258	405 ± 267	67 ± 23
	DIC	500 ± 78	521 ± 117	319 ± 29	317 ± 33	442 ± 92	274 ± 35
Hoa Binh (Da R.)	DOC	1229 ± 472	1103 ± 314	629 ± 384	202 ± 45	157 ± 71	58 ± 15
	POC	430 ± 169	384 ± 111	238 ± 135	66 ± 15	56 ± 20	23 ± 12
	DIC	849 ± 101	848 ± 83	770 ± 111	893 ± 133	1026 ± 187	675 ± 131
Vu Quang (Lo R.)	DOC	70 ± 18	83 ± 22	81 ± 15	82 ± 12	60 ± 20	47 ± 20
	POC	128 ± 41	140 ± 63	134 ± 56	182 ± 45	112 ± 75	35 ± 20
	DIC	642 ± 93	699 ± 135	708 ± 103	712 ± 100	615 ± 162	675 ± 319
Hanoi (Red River main branch)	DOC	281 ± 56	305 ± 52	256 ± 58	251 ± 45	204 ± 57	97 ± 35
	POC	730 ± 238	782 ± 198	589 ± 210	532 ± 150	401 ± 231	77 ± 23
	DIC	1425 ± 141	1509 ± 131	1433 ± 204	1429 ± 169	1474 ± 237	1001 ± 176
Whole Red River	DOC	328 ± 66	356 ± 61	298 ± 67	294 ± 53	238 ± 67	114 ± 41
	POC	852 ± 278	913 ± 231	678 ± 245	621 ± 176	468 ± 270	90 ± 26
	DIC	1663 ± 165	1761 ± 153	1672 ± 238	1668 ± 197	1721 ± 277	1168 ± 205

the period 1950–2012 following the construction of the Qingtongxia Dam in 1968 (Ran et al. 2015); in the Xijiang River, more than 200 reservoirs have been constructed over the past 50 years (1957–2004) leading to increasing DOC loads and decreasing POC and DIC loads (Sun et al. 2010). In our study, the DIC fluxes of the Thao and Da Rivers and the whole Red River slightly decreased whereas those of the Lo River slightly increased during the period 1960–2015 (Fig. 4). However, no clear pattern between the DIC and TSS fluxes was observed ($R^2 < 0.5$).

Over the past 55 years, the proportion of organic carbon fluxes to total carbon fluxes of the three tributaries Thao, Da and Lo has decreased (Fig. 5). Moreover, these declines were strongly linked to dam impoundment. For example, in the Da river, organic fluxes counted for 66.1% of total carbon flux in the period 1960s, this then decreased to 23% in 1990s when the first dam (Hoa Binh) was impounded and then to 17.2% when a series of small dams constructed in the Red River in the Chinese part in 2008, to the present value of 10% of the total carbon fluxes in the Da River following the Son La dam impoundment in 2010. The same trend was observed for the Thao and the Lo Rivers when different dams were constructed.

Ludwig et al. (2011) proposed that globally, around 44% of the riverine carbon flux was transported as organic carbon, in roughly equal quantities as POC and DOC. These authors emphasized that in tropical systems, riverine carbon fluxes were thought to be dominated by DOC (DOC > POC > DIC). However, our results for the Red River for the present period indicated that this system differs from this general trend, with 85.1% of the total carbon transported as DIC and the rest of 14.9% as organic carbon fluxes, including 8.3% DOC and 6.6% POC. This result is consistent with other works from the Tana River (Africa) (Tamooh et al. 2014), the Xijiang River (Sun et al. 2010), the Yellow River (Wang et al. 2012; Ran et al. 2013) where many dams and reservoirs have affected river discharge, TSS and organic carbon fluxes leading to a dominance of DIC in riverine carbon concentrations and loads.

Influence of climate and hydrological characteristics on the Red River carbon flux

The influence of climate (precipitation and temperature) or hydrology (river discharge) on riverine DIC, POC and DOC fluxes has been previously discussed by several authors. Shi et al. (2016) reported significant positive correlations between precipitation and both DOC concentrations and fluxes in the Pearl and the Yangtze Rivers. Cai et al. (2008) found a strong correlation between DIC fluxes and river discharge for four major World rivers [Changjiang, Huanghe, Zhujiang (Pearl) and Mississippi Rivers]. This has been observed in numerous tropical rivers such as the Xijiang River (Sun et al. 2010), the Tana River (Tamooh et al. 2014), the Mekong river (Li and Brush 2015), and the Yellow river (Liu et al. 2015). The same trend was observed for

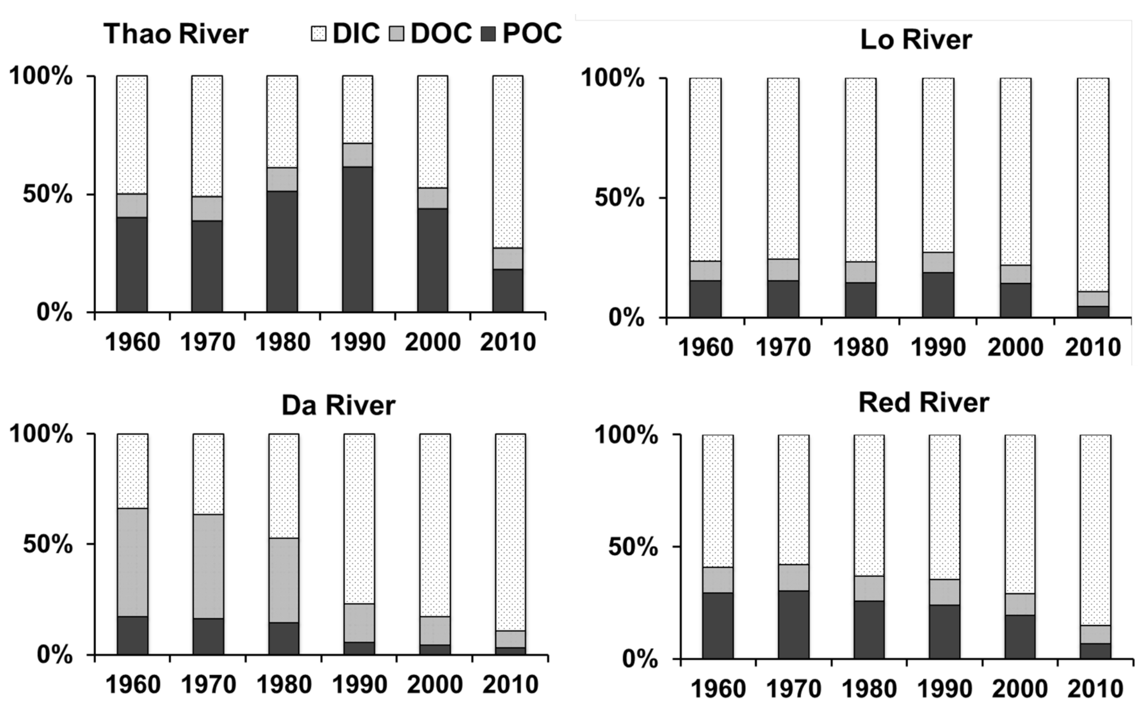

Fig. 5 Decadal change in carbon proportions (1960–1969, 1970–1979, 1980–1989, 1990–1999, 2000–2009, 2010–2015) at four gauging stations: Yen Bai station (Thao River); Hoa Binh station (Da River); Yen Bai station (Thao River) and Hanoi station (Red River)

the tropical Red River when both the river discharge and suspended matter concentrations are higher during the rainy summer and during which more than 90% of the annual POC fluxes and 80% of the annual DOC and DIC fluxes were transferred to the estuary (Fig. 6).

Regarding longterm annual variation in 1960–2015, the decreased DIC fluxes for the Thao and Da tributaries and the main branch Red River at Hanoi station were related with the reduced river discharges: the Thao (774 ± 126 $m^3 s^{-1}$ in 1960s to 517 ± 34 $m^3 s^{-1}$ in 2010s); the Da (1720 ± 219 in 1960s to 1276 ± 216 $m^3 s^{-1}$ in 2010s) and the Red River at Hanoi (2610 ± 317 in 1960s to 1673 ± 262 $m^3 s^{-1}$ in 2010s) (Table 1). Similarly, the increased DIC flux of the Lo river over the studied period was correlated with its slight higher river discharge in 2010s (990 ± 439 $m^3 s^{-1}$) vs 1960s (955 ± 144 $m^3 s^{-1}$ in 1960s) (Table 1). The link between river discharge and carbon flux also meant that during high peak flood years, highest annual total carbon fluxes were observed. Over the 55 years of the study, the highest DOC and POC fluxes of the whole Red River system (498 and 1497 kton C $year^{-1}$, respectively) were observed in 1971, when extreme flood event was recorded in the Red River basin. The highest DIC flux at the Vu Quang station was 947 kton C $year^{-1}$ in 2014, corresponding to the typhoon Rammasun that strongly affected the Lo river sub-basin. Consequently, during cyclones or storms, soil erosion and the riverine organic carbon flux were accelerated, suggesting that rainfall–river discharge was one of key drivers of carbon fluxes in the Red River.

Furthermore, DOC fluxes in the Red River main channel were lower than POC fluxes (Table 4). Consequently, the mean DOC/TOC ratio of the Red River was lower than 0.5, which is typical for the Asian rivers in monsoon areas, as opposed to the higher DOC/TOC ratios (> 0.5) found in European and American rivers (Meybeck et al. 2005). This ratio may be due to the strong leaching processes of organic carbon from soils and rocks, namely from diffuse source in the drainage basin, especially during the rising stage (Coynel et al. 2005; Zhang et al. 2009). During low river discharge, leaching processes are reduced; consequently, DOC in the Red River mainly originates from point sources (population, urbanization, industrial activities).

Temperature can also have an impact on carbon concentrations and fluxes. Shi et al. (2016) observed a significant positive correlation between temperature and DOC concentrations for the mainstream Yangtze, whereas no significant correlation between temperature and DOC flux for the Pearl River. Dessert et al. (2003) suggested that higher

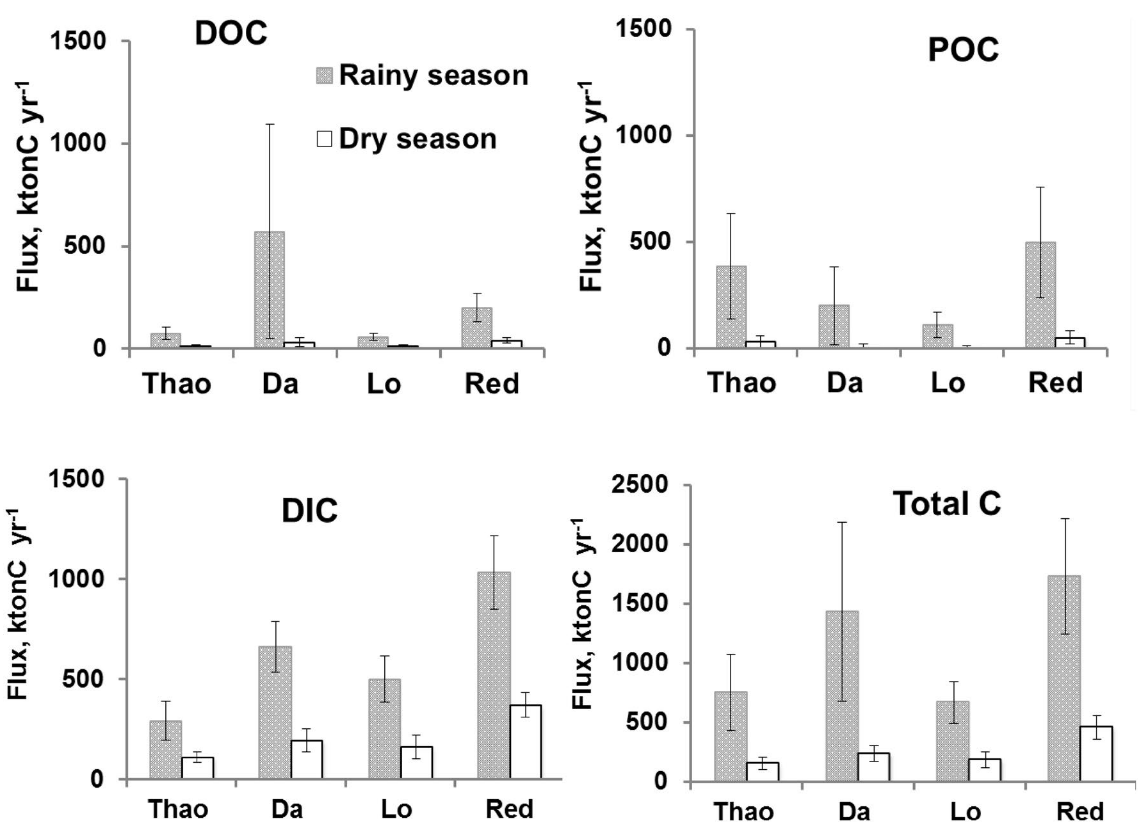

Fig. 6 Mean value of carbon (DOC, POC, DIC and total C) fluxes (and standards deviation) of the Red River system in the rainy and dry seasons during the period 1960–2015. Noted that Total C is at different scales

temperature should induce higher weathering rates and thus higher DIC transfer. This is in contrast to our results for the whole Red River where the DOC and DIC fluxes tended to decreases over the past 50 years whereas annual average temperature in this area has increased by 0.7–1 °C (Lai 2016).

The long residence time exposes river water to evaporation, which affects solute chemistry, combined with dense aquatic and wetland vegetation allows for extensive river water–vegetation–atmosphere interactions which can affect carbon cycling. Some previous studies suggested that the balance of evaporation plays a central role in modulating the seasonal variation of HCO_3^- concentrations in some large rivers (Changjiang, Huanghe, Pearl, and Mississippi rivers) (Cai et al. 2008) or the spatial variation of the DIC concentrations in the Okavango River in Botswana (Akoko et al. 2013). In the case of the Red River, unfortunately, no information available for longterm variation of evaporation in the Red River basin; however, we may suggest the influence of evaporation was less than the dam impacts on the riverine carbon fluxes.

Influence of lithological and geomorphological characteristics on the Red River carbon flux

Lithology is responsible for most of the variation of riverine carbon export to ocean (Huang et al. 2012). Indeed, the percentage of carbonate rock, which has the highest weathering rate is closely linked to riverine inorganic carbon concentration and export. As known, half of the riverine HCO_3^- concentrations come from carbonate minerals and the rest is from the atmosphere and soil CO_2 which is related with silicate weathering (Meybeck and Vorosmarty 1999; Cai et al. 2008). The percentage of carbonate rock is known to be highest in Asia and lowest in tropical Africa (Suchet et al. 2003), strongly impacted riverine carbon concentrations on these two continents. Brunet et al. (2009) found in the Nyong basin (Cameroon, tropical Africa) where lacks carbonates rocks in its basin, the mean DIC concentration was quite low, 3.6 ± 0.3 mg C L^{-1}. Low DIC concentrations have also been found in other rivers with few carbonates rocks in their basins such as the Amazon River: 4 mg C L^{-1}; the Congo River: 3 mg C L^{-1}; the Niger River: 6.6 mg C L^{-1} (Prosbt et al. 1994). In contrast, the high DIC concentrations have been observed in Asian rivers such as the Pear river: 18 mg C L^{-1}; the Changjiang river: 21 mg C L^{-1}; the Huanghe: 31 mg C L^{-1}, the Longchuanjiang: 39 mg C L^{-1} (Cai et al. 2008; Li et al. 2011) as compared to the mean DIC value of the World river: 9.6 mg C L^{-1} (Meybeck et al. 2005) (Table 5). In this study, the mean DIC concentration of the Red River was 19.1 ± 2.0 mg C L^{-1} over the 1960–2015 period was close to that of other Asian rivers. In parallel, the specific DIC flux of the whole Red River system (10.5 ton C km^{-2} $year^{-1}$) was close to the mean value for

Table 5 Carbon concentrations and fluxes of some World rivers

River	Concentration (mg C.L^{-1})			Refs.	Fluxes (ton km^{-2} $year^{-1}$)			Refs.
	DOC	POC	DIC		DOC	POC	DIC	
Amazon River	4.46	2.83	8–10	Moreira-Turcq et al. (2003)	5.8	5.45	33.4	Moreira-Turcq et al. (2003)
Brahmaputra River			23.0	Singh et al. (2005)		3.6	48.9	Galy and France-Lanord (1999) and Aucour et al. (2006)
Congo River	10.6	1.6–2.3	3.0	Coynel et al. (2005) and Wang et al. (2013)	2.4	0.3	1	Wang et al. (2013)
Irrawaddy River			24.0	Bird et al. (2008)		7.7–12.6		Bird et al. (2008)
Mississippi River	5.87	16.9	30–32	Bianchi et al. (2007) and Raymond et al. (2008)	0.95	1.3	4.1	Bianchi et al. (2007) and Raymond et al. (2008)
Pearl River	1.9–3.5	1–3.8	12–15	Ni et al. (2008) and Wu et al. (2007)	1.5	2.0	64.4	Zhang et al. (2007) and Wu et al. (2007)
Xijiang River		0.03–2.4		Sun et al. (2010)		2.5		Sun et al. (2010)
Yangtze River	1.6–3.3	0.7	17.8	Wang et al. (2012) and Wu et al. (2007)	0.8	1.6	7.5	Wang et al. (2012) and Wu et al. (2007)
Yellow River	1.0–8.8	4.7–92.4	9.9–55.1	Liu et al. (2015)	0.09	0.8	7.3	Liu et al. (2015) and Chen et al. (2005)
Mean value Asian rivers	1.16	–	30.7	Qu et al. (2017)	3.97	5.32	9.79	Huang et al. (2012)
Mean value for tropical rivers	5.38	–	8.3	Huang et al. (2012)	2.1	2.05	3.29	Huang et al. (2012)
Mean value World rivers	5.75	5	9.55	Meybeck, (1982) and Meybeck et al. (2005)	1.9	1.6		Ludwig et al. (1996)

the Asian rivers (9.8 ton C km^{-2} $year^{-1}$) (Huang et al. 2012) (Table 5). The high DIC concentration in the Red River can be explained by wide distribution of carbonate–silicate rocks in the upper Red River drainage area (Suchet 1995; Moon and Huh 2006; Dang et al. 2010). Moon et al. (2007) noted that bi-carbonate accounts for more than 55% of the TDS in the Red River, of which more than 70% of this is derived from carbonate weathering. Thus, the high export DIC of the Red River is linked to the abundance of carbonate minerals in the watershed, especially during high water discharge.

Lithology and mechanical erosion also play a considerable role in the export of organic matter. The high DOC concentration (averaged 16 ± 1.3 mg C L^{-1}) in the Nyong River results mostly from plants and kaolinite that are found in its basin and that are rich in old organic matter (Brunet et al. 2009). In tropical Asia, the very high DOC concentrations (up to 46 mg C L^{-1}) found in the black-water Sebangau River (Indonesia) are due to the large areas of peat found in their basins (Moore et al. 2011). In the Red River, the low organic carbon (DOC: 3.5 ± 2.7 mg C L^{-1}; POC $= 4.9 \pm 4.6$ mg C L^{-1}) concentrations are probably related to several factors including the strong mechanical erosion present that has led to the export of lower soil horizons with lower organic matter contents, as has been suggested by Hu et al. (2015) for the Yellow river.

Influence of population and land use change on the Red River carbon flux

The riverine carbon fluxes of many rivers have undergone significant changes due to rapidly increasing urbanization and the transition of land cover (Aldrian et al. 2008; Loh et al. 2012). Mayorga et al. (2005) suggested that deforestation in the Amazon led to immediate changes to the organic matter fuelling riverine heterotrophic energy requirements and that such an impact was consistent with apparent lag times observed in bulk organic carbon composition. Coynel et al. (2005) proposed that the contribution of the forested areas to total DOC fluxes in the Congo River varied between 74–78% in the savannah system and up to 81% in the evergreen forest. Huang et al. (2012) also noted that the enhancement of soil erosion by agricultural processes has increased POC input for rivers. Li and Bush (2015) demonstrated the human-induced land use change, including deforestation and agricultural expansion primarily accelerated chemical and physical weathering rate inducing changes in riverine carbon flux for the Mekong river basin. In addition, impacts of population on riverine organic carbon fluxes cannot be ignored. Ran et al. (2013) pointed out that the DOC flux nearly doubled and the POC flux was 3.7 folds higher at Tongguan station in the middle basin than the ones at the Toudaoguai station in upstream of the Yellow River maybe due to the severe human influences affecting the area (with 100 million people) and the agricultural pollution (1.2×10^5 km^2 of farmland).

In the Red River basin, total population has rapidly increased in both rural and urban areas over the last decades. In parallel, deforestation has occurred in the Thao, Da and Lo basins (Table 2) (Le et al. 2015a). Deforestation and more intensive cultivation practices increase soil degradation and erosion, leading to the increase in riverine organic and inorganic carbon export (Regnier et al. 2013; Janeau et al. 2014a, b). However, surprisingly, our results show a decreasing tendency of both organic and inorganic carbon fluxes of the Red River despite large increase in population, agricultural activities and deforestation during the period 1960–2015 (Fig. 4). The decreased organic carbon fluxes were strongly related with the dam impoundment, whereas the decrease of the Red River DIC flux may related to (1) the reduced river discharge in the Thao, Da tributaries and in the main branch Red River; or (2) to high rates of carbonate precipitation in the irrigated agricultural land, the surface of which has dramatically increased over the period 1960–2015 (Table 2; Fig. 7). The precipitation of carbonate in irrigated agricultural land leading to clear decrease of the riverine DIC flux has been observed in the Mekong river (Li and Bush 2015).

Consequences of riverine carbon flux changes

Shifts in the transport in riverine suspended solids, dissolved inorganic carbon (DIC) and organic carbon (OC) loads towards the ocean can have strong impacts on beach erosion, coastal sediment dynamics, algal photosynthesis, fisheries production and on coral reef health (Meyer et al. 2016; Kuntz et al. 2005; Haas et al. 2011). Furthermore, elevated DOC concentrations stimulate microbial respiration and growth (Cole 1982) leading to oxygen depletion and to the potential accumulation of toxic substances (Kline et al. 2006; Haas et al. 2011). In Asia, the construction of reservoirs in the upstream rivers has caused dramatic reductions in discharge and in sediment, carbon and nutrient loads into the estuaries and coastal areas. The decreases of suspended solids and associated substances (C, N, P and Si) loads have serious consequences, such as increasing coastal erosion, reducing nutrient elements for phytoplankton and aquaculture, decreasing aquaculture production, loss of shelter and breeding grounds in coastal zones (Chen 2000). Some major river systems in Asia, such as the Yellow River, the Changjiang River, the Mekong River have seen the emergence of these problems in recent decades (Lu and Siew 2006; Kummu and Varis 2007; Wang et al. 2012; Lu et al. 2015). Actually, the Red River is experiencing a clear decrease in TSS fluxes that have led to increased coastal bank erosion, salinization of aqua-cultivated land and damage to ecosystems in the coastal zone. While some of the consequences

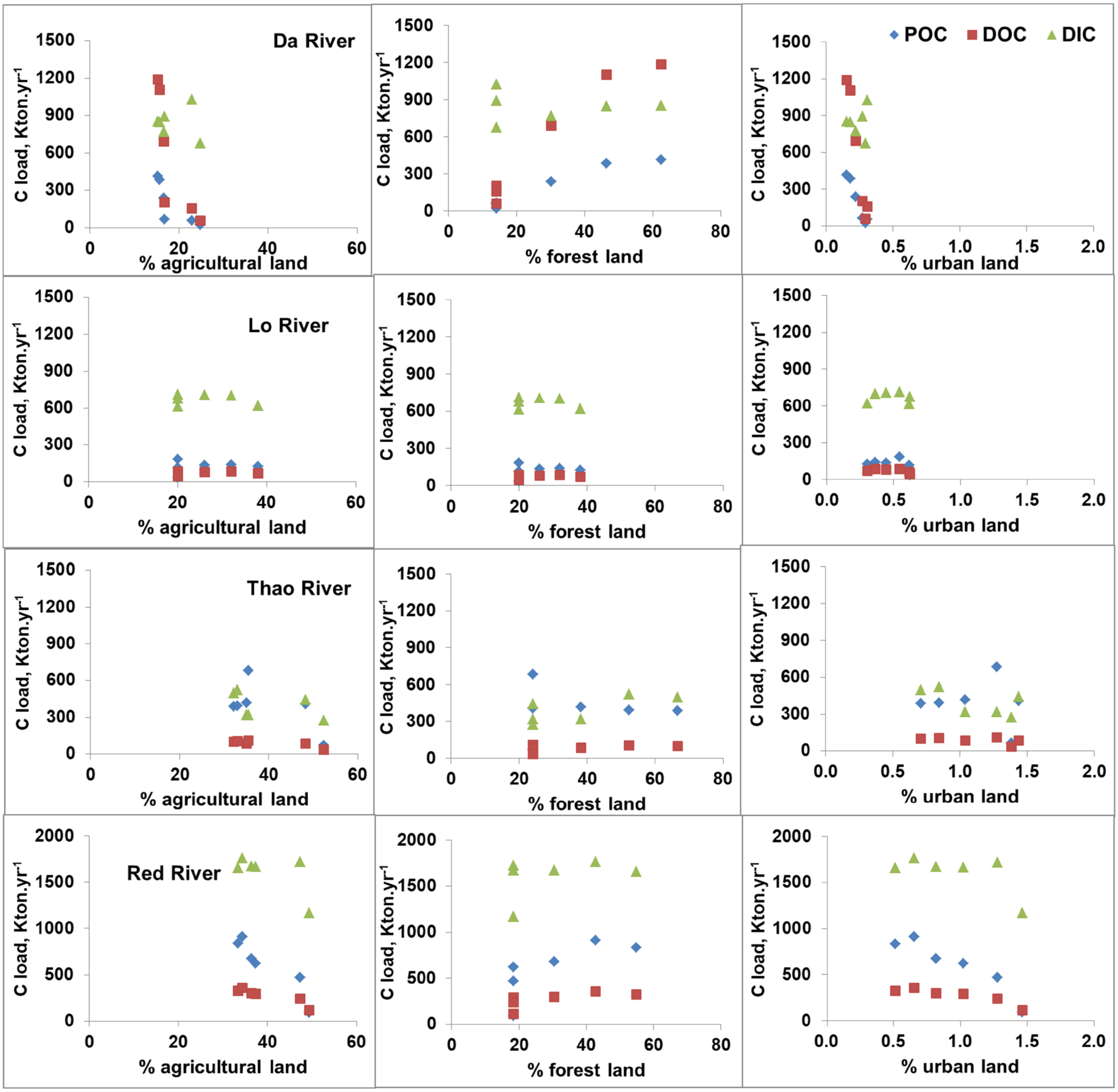

Fig. 7 Relationship between the average decadal fluxes of POC, DOC and DIC in six periods (1960s, 1970s, 1980s, 1990s, 2000s and 2010s) with the evolution of agricultural, forest and urban lands for the tributaries Da, Lo and Thao rivers and of the whole Red River

have been identified, an assessment of the full impact of the change in carbon and nutrient loading from the Red River towards the sea remains to be done.

Bias in our calculation

The calculations presented here are subject to a number of possible biases. Firstly, the Red River carbon fluxes were calculated from the DOC, DIC and POC concentrations, which were extrapolated from the different linear relationships between POC and TSS concentrations, between DOC/POC ratio with daily suspended sediment concentrations, and between DIC concentrations and river water discharge determined on the observations obtained over the period 2008–2015 (Le et al. 2015b, 2017). However, such calculation methods for longterm DOC, DIC and POC concentrations based on TSS and water discharge and upon the POC/TSS, DOC/POC and DIC/discharge relationships derived over a short period have also been used for the Xijiang River in China (Sun et al. 2010) or the Tana River in

Kenya (Geeraert et al. 2018). The use of equations calculated on a recent dataset may not be suitable for the entire longterm period (about 55 years for the Red River) as these ratios may have a certain spatial and temporal variability, however, without contemporaneous data, it is difficult to evaluate this bias.

Secondly, carbon transformation and lost towards the atmosphere as CO_2 was not considered in our calculation of the total carbon fluxes of the whole Red River system transported to the coastal zone. As known, carbon transport in fluvial systems is not passive and significant transformations can occur during the transit to the ocean, particularly in reservoirs and floodplains that act as hot spots in the regulation of riverine carbon dynamics (Cole et al. 2007). Nevertheless, despite these potential errors, the results presented here show the importance of the long-term variations in the Red River carbon flux, both in term of quantity and its organic/inorganic carbon flux proportion.

Conclusions

In the Red River, POC, DOC and DIC flux deliveries are highly seasonal, with about 90% and 82% of their respective loads transported during the high-flow seasons (May to October). POC and DOC concentrations are strongly linked with diffuse sources such as strong leaching process of soil and rocks during the high river discharge and with point sources in the watershed at low river discharge. In contrast, the high riverine DIC contents are strongly linked to the large proportion of carbonate rocks in this basin. Spatially, among the three main tributaries of the upstream Red River, the highest carbon load was transported through the Da River to its higher river discharge. The Thao and Lo Rivers were equal in terms of total carbon transported (906 ± 354 kton C year^{-1} and 870 ± 211 kton C year^{-1}, respectively) during the 1960–2015 period.

Overall, the total carbon fluxes of the Red River transferred to estuary significantly decreased from 2816 kton C year^{-1} in the period 1960s to 1372 kton C year^{-1} in the period 2010s (a 48% reduction). This reduction of organic carbon fluxes was observed together with a significant decrease of organic proportion from 40.9% in 1960s to 14.9% in 2010s, reflecting the important impact of dam and reservoir impoundment in the upstream tributaries. In parallel, the decrease in DIC flux may related to both decreasing river discharge and to increased carbonate precipitation in the irrigated, agricultural land.

The impoundment of the Lai Chau and the Huoi Quang (at the end of 2017) will probably further reduce TSS and, organic carbon fluxes and water discharge in the downstream Red River. This, combine with the anticipated effects of climate change and changes in land use will all interact further altering riverine carbon fluxes in this system and in other systems subject similar pressures.

The longterm carbon flux of the Red River may contribute to the database of global riverine carbon fluxes. Moreover, the reconstruction of longterm POC concentrations may allow the estimation of longterm partial pressure CO_2 (pCO_2) and then CO_2 emission rates once the relationship between POC–pCO_2 is established for the Red River. Note that the Red River is typical of many South-East Asian rivers in that it has been strongly impacted by environmental change and human activity for many decades, but studies on carbon transfer and CO_2 emission, especially in longterm period is still limited.

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