



# Surface sediment quality of the Red River (Vietnam): impacted by anthropogenic and natural factors

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## Abstract

Riverine surface sediments are known to be important sources and sinks for different variables like metals, organic carbon and nutrients. This paper presents the surface sediment quality of the Red River which was monitored for nine trace metal elements, total organic carbon and nutrients (nitrogen, phosphorus and silica) during four sampling campaigns in 2019. The results showed that Fe and As concentrations were higher than the allowed values of the Vietnam National technical regulation on the surface sediment quality QCVN 43:2017/BTNMT. Trace metal element concentrations were, from highest to lowest, Fe > Mn > Cu > Zn > Pb > Cr > As > Cd > Hg. The geoaccumulation index and the enrichment factor values revealed that the Red River sediments were polluted by As; moderately polluted by Cu, Pb, Cd, and Hg; and uncontaminated by other metals (Mn, Cr and Zn). For other variables, total organic carbon ( $0.43 \pm 0.31\%$ ), total nitrogen ( $0.09 \pm 0.04\%$ ), and total phosphorus ( $0.060 \pm 0.018\%$ ) were low in comparison with other urban rivers, whereas total silica ( $29.93 \pm 4.52\%$ ) seemed to be high. Spatially, the lowest values of most variables were observed at the Hoa Binh site, where human population and activities are lowest and when discharge was highest. In contrast, most variables were highest at the Ba Lat site where the human activities clearly impacted on the Red River. These results indicate that the Red River sediment is affected by a combination of natural characteristics and human activities in the basin.

**Keywords** Sediment · Nutrients · Total organic carbon · Trace metal elements · Human impacts · Natural factors

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## Introduction

Riverine sediment quality is of great concern to scientists due to its important impact on aquatic ecosystem functioning and, indirectly, on human health. Islam et al. (2015) pointed out that the riverine sediment is an essential and dynamic compartment composed of various organisms and environments. Many important biogeochemical processes occur in the riverine sediment surface such as organic matter storage and degradation (Marmonier et al. 1995; Rulik et al. 2001), phosphorus adsorption and transformation (Butturini and Sabater 1999; Vervier et al. 2009), nitrification or denitrification (Dahm et al. 1998; Lefebvre et al. 2005) and absorption and release of trace metal elements (TME) with riverine water column (Li et al. 2020; Schmitt et al. 2003). Accumulation of different variables (TME, total organic carbon (TOC) and nutrients) in riverine surface sediment has been observed worldwide (Liu et al. 2009; Li et al. 2013). It has been emphasized that organic matter can be decomposed during the natural process, whereas TME in riverine water might be enhanced by the presence of organisms, transformed into organic compounds or be absorbed into particles, and then subsequently accumulated at high levels in riverbed sediments (Li et al. 2020). When changes in environmental circumstances cause TME in sediment to be released back into the water column, they have the potential to impact water environmental safety and pose a health risk to aquatic life. Sedimentary structure, mineral composition and physical–chemical transport are factors that affect the distribution and accumulation of pollutants in sediments (Zhang et al. 2014). Changes in land covers and human activities in the basins have led to increases in concentrations of different variables in sediment such as nutrients, TME and TOC (Karageorgis et al. 2003).

The major Asian rivers contribute a large proportion (50%) of sediment to global sediment load (Ludwig et al. 1996), and many Asian rivers have seen dramatic changes in sediment fluxes over the past decades due to climate and human-induced change. Although some studies have focused on the surface sediment quality of different rivers in Asia, they have tended to separately study TME (Ramesh et al. 2000; Islam et al. 2015; Strady et al. 2017; Li et al. 2020) or organic matter (Trinh et al. 2012; Li et al. 2017) or nutrients (Maazouzi et al. 2013). Riverine sediment quality is of increasing concern to researchers in Asia in recent years (Strady et al. 2017; Wei et al. 2019a).

The Red River (Vietnam and China) with a total basin area of about 156,450 km<sup>2</sup> is characteristic of a Southeast Asian river system. Previous studies have shown that human activities in the basin have induced significant changes in the hydrology and suspended sediment and

associated element (N, P) loads of the Red River (Dang et al. 2010; Le et al. 2015). Information concerning TME in surface sediment of the Red River (Nguyen et al. 2012, 2016) or of its distribution in urban rivers (To Lich and Kim Nguu) (Nguyen et al. 2007; Nguyen et al. 2015b) or organic carbon, nitrogen and phosphorus concentrations at some sites of the Day River in the Red River Delta (Trinh et al. 2012) have been published, however, knowledge on the sediment quality of the whole Red River system is still limited.

This study focuses on (i) assessing the surface sediment quality of the Red River from the upstream to the estuary in 2019; (ii) identifying some factors that may control sediment quality of the Red River. The results may contribute to the assessment of sediment quality and provide a scientific basis for environmental protection and management for the Red River basin as well as for Vietnam. The results also provide a dataset for river sediment quality in Southeast Asia.

## Materials and methods

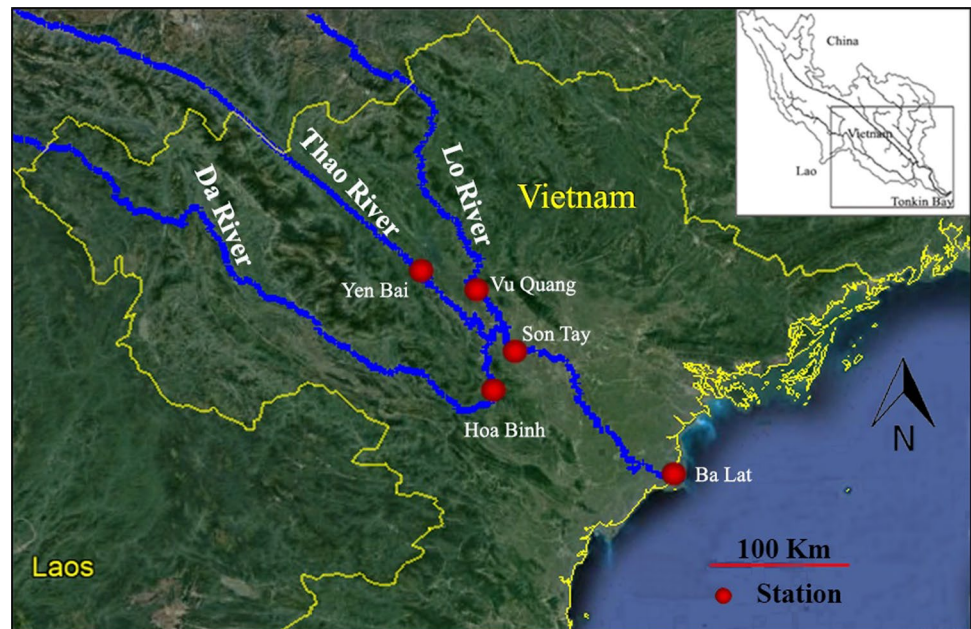
### Study site

The Red River (156,450 km<sup>2</sup>) originates in the mountainous Yunnan province in China. The upper basin is covered by sandstones or mudstones (Chinadata 1998), whereas grey and alluvial soils (70%) dominate in the lower part (Le et al. 2015). High soil erosion and red laterite soils going into rivers in the upstream Red River system give the river its characteristic red color (van Maren and Hoekstra 2004). The main axis, Thao River, receives two main tributaries, Da and Lo at Viet Tri city before forming a large delta area (Fig. 1).

The climate is monsoonal with two distinct seasons: rainy (May–October) and dry (November–next April) seasons. The climate results in a hydrologic regime that is characterized by the high runoff during the wet season and the low runoff during the dry season. During the last 50 years, the mean annual river discharges of the Thao, Da and Lo rivers and in the main axis of the Red River at the Hanoi station were  $720 \pm 159$ ,  $1670 \pm 284$ ;  $1020 \pm 227$ , and  $2500 \pm 477$  m<sup>3</sup> s<sup>-1</sup>, respectively (MONRE 1960–2015). Several dams have recently been installed in this river system in both China and Vietnam (Wei et al. 2019b).

Land use cover differs in the three upstream river basins and the Delta area. Forests and bare land (74%) dominate in the Da sub-basin, whereas industrial crops (58%) and paddy rice fields (66%) mainly cover the Lo and Delta areas, respectively. The Thao sub-basin is diversely characterized by forest, paddy rice fields, and industrial crops (85%) (Le et al. 2015).

Population density is much lower upstream (< 300 inhabitants.km<sup>-2</sup>) than downstream (> 1000 inhabitants.km<sup>-2</sup>) (Le

**Fig. 1** Sampling sites of the Red River in 2019

et al. 2015), with about a total of 42 million inhabitants in the whole basin (Wei et al. 2019b).

### Sampling campaigns and laboratory analysis

Surface sediment samples (about 0–30 cm) were collected in four campaigns in 2019 (March and April, the dry season; June and July, the rainy season) at five gauging stations: Hoa Binh (Da River), Vu Quang (Lo River), Yen Bai (Thao River), Son Tay (main axe of the Red River after the conjunction of the Thao, Da, and Lo) and Ba Lat (the Red River estuary) (Table 1, Fig. 1). At each site, three sediment sub-samples (banks and middle stream of the river) were collected with a Van Veen grab sampler and then well mixed to obtain a site sample for analysis. Samples were stored in polyethylene bags during transportation to the laboratory. In the laboratory, the samples were air-dried at room

temperature and then ground before being sieved through a 1-mm sieve.

### Laboratory analysis

#### pH

The liquid mixture of sediment and distilled water at ratios 1:2.5 was well stirred for 1 h to obtain soil slurry (Pansu and Gautheyrou 1998). The pH of the solution was measured electrometrically with a pH meter that was calibrated before using.

#### Trace metal elements

For Pb, Cu, Cd, Zn analysis: 2 g of each dried soil sample was added to 15 mL HCl 37% and 5 mL HNO<sub>3</sub> 65%. The

**Table 1** Location of five sampling sites of surface sediment of the Red River in 2019

No.	Site name	River	Position 1	Position 2	Position 3	Mean water depth (m)	
						In wet season	In dry season
1	Yen Bai (YB)	Thao	21° 42' 3.557" 104° 52' 5.826"	21° 42' 3.142" 104° 52' 1.194"	21° 42' 3.156" 104° 52' 1.244"	4.8	3.7
2	Hoa Binh (HB)	Da	20° 50' 49.857" 105° 21' 13.092"	20° 50' 48.256" 105° 21' 9.430"	20° 50' 35.805" 105° 21' 25.999"	11.5	9.9
3	Vu Quang (VQ)	Lo	21° 34' 59.430" 105° 14' 54.613"	21° 34' 58.795" 105° 14' 54.529"	21° 35' 0.426" 105° 14' 57.174"	12.3	9.2
4	Son Tay (ST)	Hong/Red	20° 09' 23.134" 105° 30' 42.485"	20° 9' 20.656" 105° 30' 41.075"	20° 9' 22.801" 105° 30' 42.379"	9.5	7.5
5	Ba Lat (BL)	Hong/Red	20° 18' 57.700" 106° 31' 19.700"	20° 18' 52.500" 106° 31' 19.900"	20° 18' 52.500" 106° 31' 19.900"	2.1	1.4



mixture was digested on a hot plate at 95 °C until the supernatant became clear and a brownish-coloured fume was no longer generated. The samples were then left to cool to room temperature before being analysed (Rashid et al. 2016).

For Hg and As analysis: 0.2 g of each dried soil sample was added 5 mL HNO<sub>3</sub> 65% and then was digested for 4 min in a Microwave Digestion system (Niessen et al. 1999).

After the soil sample treatment procedure, all TME were in the form of dissolved salts. TME were then measured by atomic absorption spectrophotometer (Varian 280FS, Australia), proposed by APHA (2017) (3114, 3112 and 3110 methods for As, Hg, and the other metals, respectively).

### Total organic carbon

Organic matter and total organic carbon (TOC) in sediment samples were determined by the Walkley Black method (Walkley and Black 1934): the dried samples were added to 20 mL concentrated H<sub>2</sub>SO<sub>4</sub> and then digested with 10 mL K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> 1 N at 150 °C for 30 min and then titrated with a standard ferrous ammonium sulphate (Mohr's salt) solution.

### Nutrients (TN, TP and TSi)

Total phosphorus (TP) and total silica (TSi) were determined by the following steps: the dried samples were digested and mineralized by sodium persulfate at 110 °C for 2 h. Phosphate concentration then was determined spectrophotometrically (Jasco V630, Japan) according to the method 4500-P proposed by (APHA, 2017). Total silica concentration was analysed by the method 4500-SiO<sub>2</sub> (APHA, 2017) after sample digestion.

Total nitrogen (TN) was determined by the Total Kjeldahl Nitrogen method (4500-N nitrogen; APHA 2017). Dried samples were digested by K<sub>2</sub>SO<sub>4</sub> in an acidic phase (H<sub>2</sub>SO<sub>4</sub> 2 N) for 1 h at a temperature of less than 350 °C. The sample was then distilled and titrated with H<sub>2</sub>SO<sub>4</sub> 2 N in the presence of boric acid (4%).

All reagents and solvents were of analytical grade (Merck-GR). The recoveries of standard reference metals were 90–112%. Triplicate measurements were taken on all

samples, standards, and blanks. The data shown below were the average values of the triplicates.

### Calculation of different pollution indexes (geoaccumulation index ( $I_{geo}$ ) and enrichment factor (EF))

$I_{geo}$

$I_{geo}$  which is the enrichment relative to the geological substrate and calculated in Eq. (1) (Muller 1969) was applied to quantify the degree of anthropogenic contamination in the sediment:

$$I_{geo} = \log(C_n/1.5B_n) \quad (1)$$

where  $C_n$  is the measured concentration of the examined TME (n) in the sediment (µg/g); 1.5 is the factor used for a lithological variation of metals;  $B_n$  is the geochemical background concentration of the same TME. In this study,  $B_n$  values (ppm) for different TME are given in Table 2.

The  $I_{geo}$  value of each trace metal element was classified as uncontaminated with  $I_{geo} \leq 0$ ; uncontaminated to moderately contaminated ( $0 < I_{geo} < 1$ ); moderately contaminated ( $1 < I_{geo} < 2$ ); moderately to heavily contaminated ( $2 < I_{geo} < 3$ ); heavily contaminated ( $3 < I_{geo} < 4$ ); heavily to extremely contaminated ( $4 < I_{geo} < 5$ ); extremely contaminated ( $I_{geo} \geq 5$ ) (Muller 1969).

### EF

Enrichment factor (EF) is known as a normalization technique widely used to categorize the TME fractions that are associated with sediments (Manno et al. 2006). To assess the level of TME contamination, EF of all studied TME was calculated for each sample following Eq. (2) and presented in Table 2. The calculation of EF is based on analysed TME concentrations and standard threshold values.

$$EF = [C_x/C_{ref}]_{sample} / [B_x/B_{ref}]_{background} \quad (2)$$

where  $C_x$  and  $C_{ref}$ : Concentration of the examined element in the examined environment and the reference environment,

**Table 2** Reference/background concentrations of different TME used for  $I_{geo}$  and EF calculation for the Red River sediment

Metals	Pb	Zn	Cu	Cd	Fe	Cr	Mn	As	Hg
Reference concentrations, ppm	17	67	28	0.09	35,000	92	600	4.8	0.05
References	Rudnick and Gao (2003)	Rudnick and Gao (2003)	Rudnick and Gao (2003)	Rudnick and Gao (2003)	Taylor and McLennan (1995)	Rudnick and Gao (2003)	Taylor and McLennan (1995)	Rudnick and Gao (2003)	Rudnick and Gao (2003)

respectively.  $B_x$  and  $B_{ref}$ : Concentration of the reference element in the examined environment and the reference environment, respectively.

The EF value was used to classify the level of trace metal element contamination as follows: The EF of each trace metal element is classified as: deficiency to minimal enrichment (non-pollution) with  $EF < 2$ , moderate enrichment with  $2 \leq EF < 5$ , severe enrichment with  $5 \leq EF < 20$ , very high enrichment ( $20 \leq EF < 40$ ), very strong level of pollution ( $EF \geq 40$ ) (Manno et al. 2006).

## Statistical analysis

All statistical and analytical results were performed with Student t-tests to verify the difference of variables (pH, nutrients, TME, and TOC) values between two seasons (rainy and dry) and at different sites. Probabilities ( $p$ ) were calculated and a  $p$ -value of  $< 0.05$  was deemed significant.

To detect the correlation between different environmental variables, statistical software XLSTAT (XLSTAT Addinsoft, 2019) was applied to calculate the Pearson correlation coefficients. Some environmental variables were evaluated by “core” to compare the correlation and selected representative variables.

## Results and discussion

### Results

#### Different variables of the Red River sediment quality

##### Trace metal elements

The concentrations of nine TME (Pb; Zn; Cu; Cd; Fe; Cr; Mn; As; and Hg, in ppm) in surface sediments of the Red River at 5 sampling sites during 4 sampling campaigns were presented in Table 3. The highest values were observed at the Ba Lat site (in the estuary), whereas the lowest were found at the Hoa Binh site, in the Da River. The average TME concentrations in all studied sediments were in the following decreasing order:  $Fe > Mn > Cu > Zn > Pb > Cr > As > Cd > Hg$ . The mean values of some TME (Pb, Zn, Cu, Cd, Cr, and Hg) of all sediment samples were lower than the allowed values of the QCVN 43:2017/BTNMT (MONRE 2017) and of the US EPA sediment standards (US EPA 2000). No value for Mn concentration is presented in the sediment standards. The mean values of Fe and As concentrations were higher than the QCVN 43:2017/BTNMT, especially Fe concentration

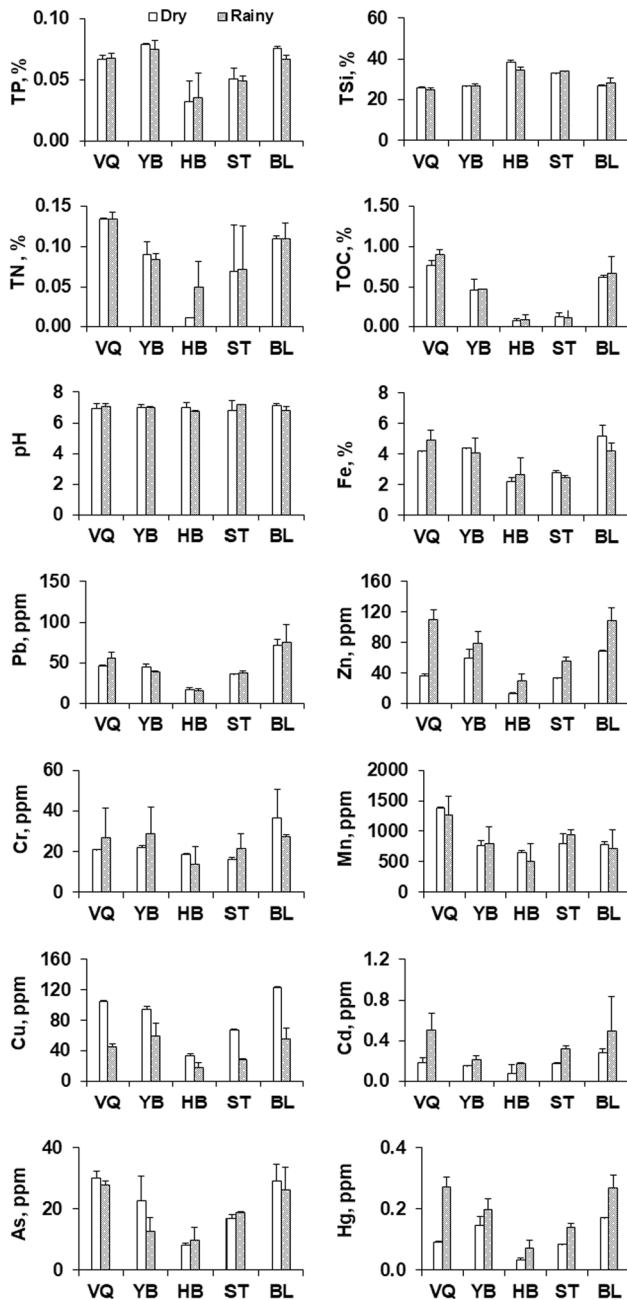
**Table 3** Trace metal element concentrations in surface sediments at 5 sites of the Red River in 2019

Sample name	Pb (ppm)	Zn (ppm)	Cu (ppm)	Cd (ppm)	Fe (ppm)	Cr (ppm)	Mn (ppm)	As (ppm)	Hg (ppm)
VQ	50.72 ± 7.23	73.03 ± 42.86	75.3 ± 34.81	0.34 ± 0.21	45,551.6 ± 5560.8	23.83 ± 9.14	1318.5 ± 192.5	28.93 ± 20	0.18 ± 0.11
YB	41.91 ± 4.48	69.31 ± 15.91	77.27 ± 22.14	0.18 ± 0.04	42,368.8 ± 5800.2	25.23 ± 8.68	787.4 ± 163.0	17.63 ± 7.78	0.17 ± 0.04
HB	16.42 ± 2.05	21.23 ± 11.10	25.63 ± 10.50	0.12 ± 0.08	24,443.8 ± 7058.5	16.12 ± 5.80	579.4 ± 186.3	8.91 ± 2.54	0.05 ± 0.03
ST	37.39 ± 1.55	44.84 ± 13.34	47.42 ± 22.58	0.25 ± 0.08	25,984.4 ± 2274.9	19.05 ± 5.09	869.8 ± 138.8	17.8 ± 1.25	0.11 ± 0.03
BL	73.37 ± 13.58	88.73 ± 24.60	89.5 ± 39.80	0.38 ± 0.23	46,915.6 ± 7405.3	31.92 ± 9.73	754.1 ± 178.0	27.61 ± 5.51	0.22 ± 0.06
Average	43.96 ± 20.07	59.43 ± 32.64	63.02 ± 34.41	0.26 ± 0.17	37,052.8 ± 11,332.9	23.23 ± 8.98	861.8 ± 296.5	20.18 ± 8.56	0.15 ± 0.08
min	14.55	11.43	12.65	0.012	18,800.0	7.48	307.0	6.86	0.03
max	90.63	120.35	123.85	0.73	56,662.5	46.48	1486.5	32.96	0.30
QCVN 43:2017/BTNMT	91.3	315	197	3.5	20,000	90	–	17	0.5
USEPA (2000) sediment standards <sup>a</sup>	91.3	315	197	3.53	–	90	–	17	0.486
Sediment quality guidelines, NOAA (1999)									
Effects range low/median (ERL/ERM)	46–218	150–410	34–270	1.2–9.6	–	81–370	–	8.2–70	0.15–0.71
Threshold/probable effect level (TEL/PEL)	30.2–112	124–271	18.7–108	0.68–.21	–	52.3–160	–	5.9–17	0.13–0.7

<sup>a</sup>Sediment quality guidelines that reflect probable effect concentrations (PEC)

was 1.85 fold higher than the allowed value (20,000 ppm). In comparison with the values proposed in sediment quality guidelines (NOAA, 1999), some TME such as Pb, Cu, Hg were classified at the threshold/probable effect level (TEL/PEL) and some TME (Pb, Cu, As, Hg) were at effects range low/median (ERL/ERM) (Table 3).

The mean values in the rainy season were higher than those of the dry season for Zn, Cd, and Hg, whereas the inverse was observed for Cu (Fig. 2). No clear seasonal variation was found for Fe, As, Pb, Mn, and Cr (Fig. 2).



**Fig. 2** Mean values in dry and rainy seasons of different variables of surface sediment quality at five stations in the Red River in 2019

## pH

pH values of all observed sediment samples of the Red River varied from 6.37 (at Hoa Binh) to 7.24 (at Vu Quang site) with an average value of  $6.98 \pm 0.24$  (Table 4), which was within the allowed value of QCVN 43:2017/BTNMT. No clear seasonal variation of pH values was detected ( $p < 0.05$ ).

## Total organic carbon TOC

The average TOC concentrations were from 0.08% at Hoa Binh site to 0.83% at Vu Quang site (Table 4). TOC concentration of all Red River sediment samples averaged  $0.43 \pm 0.31\%$ . However, no limit for TOC concentration is presented in the Vietnam national technical regulation QCVN 43:2017/BTNMT. The average TOC concentration in the Red River's sediment in the wet season was higher than during the dry season (Fig. 2) ( $p < 0.05$ ).

## Nutrients

TN and TP concentrations fluctuated from 0.01 to 0.14% (mean value:  $0.09 \pm 0.04\%$ ) and from 0.02% to 0.08% (mean value:  $0.060 \pm 0.018\%$ ), respectively for all Red River sediment samples. Within 5 sites, the highest nutrient values were detected at Vu Quang (for TN) or Yen Bai (for TP) and the lowest values were found at Hoa Binh site (for TN, TP). For silica, the mean TSi concentrations were  $29.9 \pm 4.5\%$  with the highest value at Hoa Binh site and the lowest value at Vu Quang site. We noted that no limits for nutrients (TN, TP, and TSi) are assigned in the Vietnam standard QCVN 43:2017/BTNMT.

No clear seasonal variation was found for the nutrient variables (TN, TP and TSi concentrations) (Fig. 2).

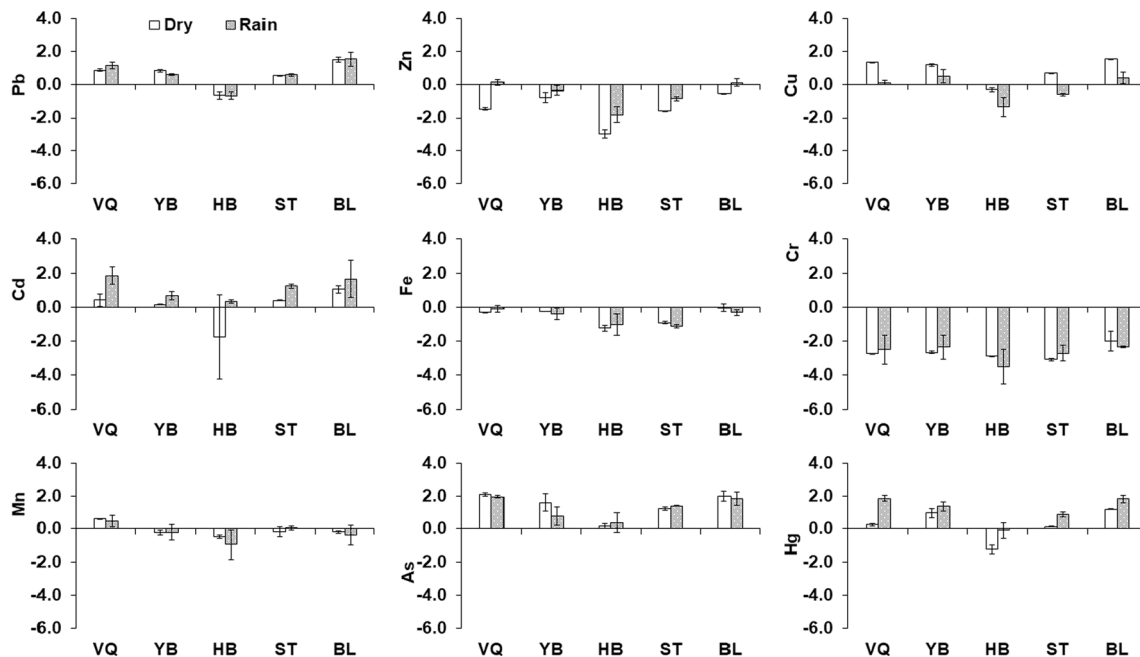
## Pollution index of the Red River sediment quality

The calculated  $I_{geo}$  values of different metals in the Red River sediment samples averaged  $1.34 \pm 0.72$  (As);  $0.71 \pm 0.95$  (Hg);  $0.62 \pm 0.75$  (Pb);  $0.60 \pm 1.17$  (Cd);  $0.35 \pm 0.90$  (Cu);  $-0.15 \pm 0.53$  (Mn);  $-2.67 \pm 0.56$  (Cr);  $-1.01 \pm 0.95$  (Zn) for all 5 studied sites. Notably, at Hoa Binh site (located in the Da River outlet, after the Hoa Binh reservoir),  $I_{geo}$  values showed the loss of most TME except for As (Fig. 3). The  $I_{geo}$  results indicated the decreasing order of  $As > Hg > Pb > Cd > Cu > Mn > Cr > Zn$  among the investigated metals. Based on the calculated  $I_{geo}$  values and the classification method of Muller (1969), metals such as Mn, Cr and Zn (with  $I_{geo}$  values less than 0), were classed as uncontaminated, whereas the metals such as Cu, Cd, Pb and Hg were classed as uncontaminated to moderately contaminated. Only As ( $I_{geo} > 1$ ) was classified in moderately contaminated level in the Red River sediments.

The mean EF values of As, Hg, Cd, Pb, Cu, Mn, Zn and Cr for all sites of the Red River were  $4.20 \pm 1.78$ ;  $2.93 \pm 1.61$ ;

**Table 4** pH, total organic carbon, and nutrients concentrations of surface sediment at 5 sites of the Red River in 2019

Site	pH	TOC (%)	TN (%)	TP (%)	TSi (%)
VQ	7.02 ± 0.21	0.83 ± 0.09	0.13 ± 0.01	0.067 ± 0.003	25.35 ± 0.80
YB	7.01 ± 0.11	0.47 ± 0.07	0.09 ± 0.01	0.077 ± 0.005	26.90 ± 0.39
HB	6.89 ± 0.23	0.08 ± 0.04	0.03 ± 0.03	0.034 ± 0.015	36.41 ± 2.37
ST	6.99 ± 0.42	0.11 ± 0.06	0.07 ± 0.05	0.050 ± 0.006	33.43 ± 0.68
BL	6.97 ± 0.24	0.64 ± 0.13	0.11 ± 0.01	0.071 ± 0.005	27.56 ± 1.55
Average	6.98 ± 0.24	0.43 ± 0.31	0.09 ± 0.04	0.060 ± 0.018	29.93 ± 4.52
Min	6.37	0.04	0.01	0.080	24.22
Max	7.24	0.95	0.14	0.020	39.15

**Fig. 3** Seasonal variation of  $I_{geo}$  values for nine trace metallic elements at five sites in the Red River

2.84 ± 1.85; 2.59 ± 1.18; 2.25 ± 1.23; 1.44 ± 0.49; 0.89 ± 0.49 and 0.25 ± 0.10, respectively (Table 7). The mean EF values of almost all studied metals, except for Mn, Zn and Cr were in the range from 2 to 5, reflecting the anthropogenic origin of metal enrichment in the surface sediments of the Red River. The analysis of seasonal fluctuation suggested that EF levels of Cu were greater in the dry season than in the wet season, whereas other metals (e.g. Zn, Cd, Hg) were higher in the rainy season (Fig. 4).

### Pearson relation

Within different variables observed, good positive correlations existed between Fe-Pb; Fe-As; Cd-Pb, Cd-Zn; Zn-Hg; Pb-As; and Pb-Hg ( $R^2 > 0.7$ ,  $p < 0.01$ ) (Table 5). TOC was significantly positively correlated with TN, TP and some TME (Fe, Zn, Pb, As, Hg) (Pearson correlation coefficients  $> 0.6$ ) but a strongly negatively correlated with TSi (Table 5).

Spatially, of the different variables observed, only TSi concentration did not vary across the Red River sediment samples, whereas almost all of the other variables were lowest ( $p < 0.05$ ) at the Hoa Binh site in the Da River where the forest land dominates and population density is lowest.

## Discussion

### Sediment quality of the Red River

#### Comparison with the sediment quality of other rivers in the world

The TME values of the Red River sediment were close to those observed in the Lanzhou Reach of the Yellow River, China (Liu et al. 2009). The values were higher than in the Koshi River which is mainly influenced by watershed

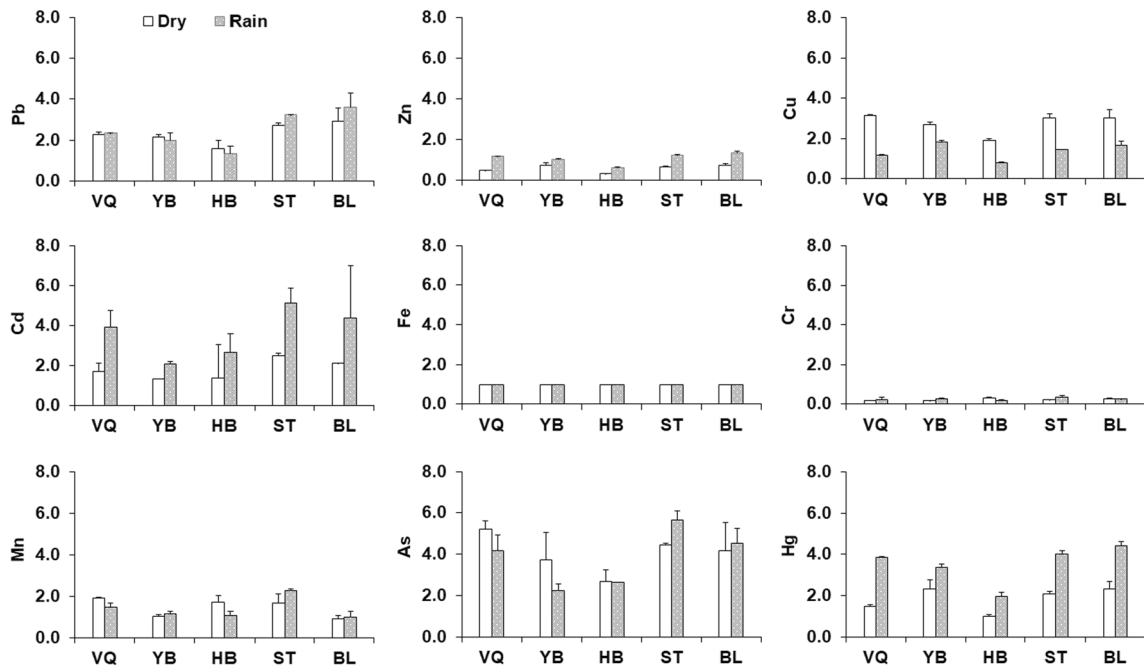


Fig. 4 Seasonal variation of EF values for nine trace metallic elements at five sites in the Red River

Table 5 Pearson correlation coefficients of different variables of surface sediment quality of the Red River

	TP	TSi	TOC	TN	pH	Pb	Zn	Cu	Cd	Fe	Cr	Mn	As	Hg
TP	1													
TSi	<b>-0.884**</b>	1												
TOC	<b>0.690**</b>	<b>-0.865**</b>	1											
TN	<b>0.728**</b>	<b>-0.796**</b>	<b>0.768**</b>	1										
pH	0.287	-0.163	0.098	0.281	1									
Pb	<b>0.665**</b>	<b>-0.720**</b>	<b>0.665**</b>	<b>0.620**</b>	0.133	1								
Zn	<b>0.628**</b>	<b>-0.701**</b>	<b>0.665**</b>	<i>0.559*</i>	0.107	<b>0.772**</b>	1							
Cu	<b>0.691**</b>	<b>-0.650**</b>	<i>0.503*</i>	<i>0.478*</i>	0.135	<b>0.584**</b>	0.124	1						
Cd	0.266	<b>-0.456*</b>	<i>0.434</i>	<i>0.436</i>	0.135	<b>0.713**</b>	<b>0.810**</b>	-0.025	1					
Fe	<b>0.863**</b>	<b>-0.906**</b>	<b>0.810**</b>	<b>0.709**</b>	0.185	<b>0.749**</b>	<b>0.697**</b>	<b>0.683**</b>	<i>0.467*</i>	1				
Cr	<i>0.566**</i>	<b>-0.482*</b>	<i>0.446*</i>	0.319	0.311	<i>0.521*</i>	<i>0.508*</i>	<i>0.428</i>	0.262	<b>0.650**</b>	1			
Mn	0.376	<b>-0.568**</b>	<i>0.538*</i>	<i>0.513*</i>	0.101	0.364	0.330	0.343	<i>0.431</i>	<i>0.485*</i>	0.154	1		
As	<b>0.632**</b>	<b>-0.750**</b>	<b>0.732**</b>	<b>0.762**</b>	0.210	<b>0.817**</b>	<i>0.531*</i>	<b>0.661**</b>	<i>0.529*</i>	<b>0.718**</b>	<i>0.419</i>	<b>0.622**</b>	1	
Hg	<b>0.629**</b>	<b>-0.701**</b>	<b>0.665**</b>	<i>0.559*</i>	0.108	<b>0.773**</b>	1.000**	0.126	<b>0.810**</b>	<b>0.697**</b>	<i>0.509*</i>	0.331	<i>0.532*</i>	1

Bold: very strong correlation (>0.8); bold and italic: strong correlation (between 0.6 and 0.8); italic: good correlation (between 0.4 and 0.6); and Regular: weak correlation (<0.4)

\*\*Correlation coefficient is significant at the 0.01 level (2-tailed)

\*Correlation coefficient is significant at the 0.05 level (2-tailed)

lithology with only a minor contribution from the local human activities (Li et al. 2020) (Table 6). The mean EF and  $I_{geo}$  values of sediment TME of the Red River were higher also than the Koshi River but were much lower than that has been observed in the urban To Lich River (Table 7).

The average pH value of the Red River sediment ( $6.98 \pm 0.24$ ) was close to values observed in other tropical river sediments such as the Sai Gon–Dong Nai (Nguyen et al. 2018b), the Mekong River (Phung and Huynh 2015); the Tirumalairajan (India) (Venkatraman et al. 2015) or



the urban Korotoa River (Bangladesh) (Islam et al. 2015) (Table 8).

The mean TOC concentration of all Red River sediment samples ( $0.43 \pm 0.31\%$ ) was similar to that of other large Asian rivers such as the Lanzhou Reach of the Yellow River, China (Liu et al. 2009) or the Yangtze River (estuary) in China (Li et al. 2013) (Table 8). This value, however, was much lower than that of the urban rivers in Hanoi city, Vietnam: the To Lich (1.2–5.3%), the Kim Nguu (1.8–10.6%) where most of the untreated domestic and industrial wastewater from Hanoi city discharged (Nguyen et al. 2007; Marcussen et al. 2008) or of the peri-urban river (Day river) in North Vietnam (3.9–5.2%) (Trinh et al. 2012) or of the Sai Gon–Dong Nai river estuary which runs through Ho Chi Minh City in South Vietnam (3.28–3.48%) (Nguyen et al. 2018). Such high TOC values were also found for other rivers in the World, for example, those of the Trirumalajian River (in India) in the rainy season (2.05–3.44%) (Venkatraman et al. 2015) or in the urban river like the Korotoa (Bangladesh) with the sediment TOC concentrations from 0.74 to 2.5% during winter and from 0.98 to 1.80% during summer (Islam et al. 2015) (Table 8).

TN ( $0.09 \pm 0.04\%$ ) and TP ( $0.060 \pm 0.018\%$ ) concentrations of the Red River sediment were much lower than that of the peri-urban Day River which receives a large amount of domestic and industrial wastewater and agricultural runoff (0.2–3.9% for TN and 0.12–0.17% for TP (Trinh et al. 2012)) (Table 8). The mean TSi concentration ( $29.9 \pm 4.5\%$ ) in this study was close to the values observed in the Ganges River, India (31.2%) (Ramesh et al. 2000) but still higher than the average value of the World river sediment (28.5%) (Martin and Meybeck 1979) and some other Asian rivers (the Brahmaputra River (28.4%); the Indian average (24.5%)) (Ramesh et al. 2000).

### Relationship between different variables of sediment quality

Sediment pH exerts a strong influence on the solubility of TME and on other sediment processes. High pH values accelerate adsorption and precipitation processes, whereas low pH weakens the strength of metal bonds with sediments and reduces TME retention in sediments (Zhang et al. 2014). Phung and Huynh (2015) found that TME concentrations in the Mekong River's sediment were proportional to the pH value (6.6–8.0). However, in our study, pH was near neutral at all sites, and no significant relationship between pH and TME was detected (Pearson correlation coefficients  $< 0.3$ ) (Table 5).

The relationship between sediment TME concentrations and salinity was also reported for some river cases, e.g. in the Mekong River (Phung and Huynh 2015). It's noted that an increase of tidal water salinity may accelerate the release

capacity of TME from sediments in the absence of sulfur leading to an increase in total TME concentrations in the riverine water column (Du Laing et al. 2008). In the Red River, sediment TME concentrations (except for Mn and As) tended to increase toward the downstream section where salinity increased (Ba Lat estuary) (Table 3).

Organic matter in river sediments plays an important role in trapping and bonding TME. Martinez and McBride (1999) demonstrated that organic matter can strongly absorb TME but the decomposition process could create soluble organic ligands (DOC) altering the state of the TME and affecting its adsorption, mobility, toxicity, and bioavailability. Sediment rich in organic matter is more likely to be contaminated with heavy metals. The sediment TOC concentrations of the urban To Lich and Kim Nguu rivers where its low river discharges ( $30 \text{ m}^3 \text{ s}^{-1}$ ) were observed, showed a remarkable impact on sediment TME concentrations (As, Cd, Cr, Cu, Pb, Zn) due to the important retention through the sorption processes (Marcussen et al. 2008). In our study, TOC concentrations were low but showed a clear positive correlation with some TME (Fe, Zn, Pb, As, and Hg) which may reflect the less important retention through sorption processes in the Red River surface sediments (Table 5).

### Factors impacted on sediment quality

#### Influence of the urban, industrial production and mineral mining

Various studies have demonstrated that point sources are an important contributor to surface sediments of the World Rivers (Islam et al. 2014, 2015; Huang et al. 2018). Many factories and urban domestic wastewater contaminated by TME, nutrients, and organic pollutants discharge directly into the river, causing spatial variation of sediment quality. This has been observed in the Lanzhou reach of the Yellow River (China) (Liu et al. 2009), the Korotoa River, Bangladesh (Islam et al. 2015); the Hindon River, India (Chabukdhara and Nema 2012) or rivers in the vicinity of the Thessaloniki city, Greece (Karageorgis et al. 2003; Dhamodharan et al., 2019), or in South Korea (Pandey et al. 2019), showing the importance of industrial and urban waste for riverine sediment quality. In Vietnam, an investigation into the sediments of the Pô Kô River (in Kon Tum province, Vietnam) revealed that high sediment Pb (11.36–116.94 ppm) and Zn (averaged 201.93 ppm) concentrations were due to the mineral mining/exploitation and domestic wastewater from Kon Tum city, respectively (Nguyen et al. 2018a).

The total population of the Red River basin has rapidly increased in both rural and urban areas over the last decades (Le et al. 2015), producing a large quantity of domestic wastewater in some cities/provinces such as Hanoi ( $75.000 \text{ m}^3 \cdot \text{day}^{-1}$ ); Bac Giang ( $51.000 \text{ m}^3 \cdot \text{day}^{-1}$ ); Bac Ninh ( $65.000$



**Table 6** Trace metal element concentrations (in  $\text{mg}\cdot\text{kg}^{-1}$  or ppm) in surface sediment of different rivers in the World

River name, country	Pb	Zn	Cu	Cd	Fe	Cr	Mn	As	Hg	References
Amazon Mouth, Brazil	83	110	37.5	–	–	65	–	–	–	Siqueira (2003)
Koshi River, China	21.36 (2.96–36.17)	49.46 (33.34–93.36)	21.06 (1.21–43.63)	0.20 (0.07–0.59)	–	47.18 (4.89–123.0)	–	–	–	Li et al. (2020)
Lanzhou Reach of Yellow River, China	26.39–77.66	89.80–201.88	29.72–102.22	–	–	41.49–128.30	773.2–1459.7	13.68–48.11	–	Liu et al. (2009)
Yangtze River estuary, China	14.5–38.6	57.0–108.9	18.1–61.1	1.26–2.21	23,000–55,000	67.1–113.1	–	–	–	Yuan et al. (2014)
Raohe Basin, China	14.3±0.5–222.2±2.8	11.2±0.9–72.1±2.4	15.6±1.2–793.5±12.6	0.2±0.03–1.6±0.1	–	13.1±0.6–97.1±2.4	–	12.9±0.8–318.1±4.9	–	Wei et al. (2019a)
Ghaghara River, India	10.71–14.26	13.26–17.59	2.76–11.74	0.21–0.28	–	61.25–87.68	–	–	–	Singh et al. (2017)
Ganges River, India	22.8±8.2	203.8±58	–	0.36±0.13	–	134±31.5	–	–	–	Ramesh et al. (2000)
Ganga River, India	148.8–214.8	127.5–201.2	12.7–84.0	9.5–79.0	7175.5–9385	126.8–196.1	409.4–350.4	–	–	Pandey et al. (2014)
Gomti River, India	40.33	–	5	2.42	–	8.15	–	–	–	Singh et al. (2005)
Hindon River, India	27.56–313.57	22.50–288.29	21.70–280.33	0.29–6.29	4151.8–17,318.8	17.48–33.70	49.55–516.97	–	–	Chabukdhara and Nema (2012)
Korotoa River, Bangladesh	58 (36–83)	–	76 (35–118)	1.2 (0.26–2.8)	–	109 (55–183)	–	25 (2.6–52)	–	Islam et al. (2015)
Old Brahmaputra River, Bangladesh	7.6	52.7	6.2	0.48	–	6.6	126.2	–	0.001	Bhuyan et al. (2019)
Suspended sediment Da River, Vietnam	129	179	94	0.43	54,600	154	1161	–	–	Gaillardet et al. (1999)
Mekong River, Vietnam	–	–	–	–	–	–	–	2–35	–	Cenci and Martin (2004)
Mekong River, Vietnam	25.05	31.93	15.88	1.48	–	10.47	–	–	–	Strady et al. (2017)
Poko River, Vietnam	11.36–116.94	201.9 (100.2–389.2)	55.3	1.82–4.3	–	–	–	–	–	Nguyen et al. (2018a)



Table 6 (continued)

River name, country	Pb	Zn	Cu	Cd	Fe	Cr	Mn	As	Hg	References
Cam River, Hai Phong, Vietnam	92 ± 15	178 ± 31	82 ± 16	–	36,200 ± 3500	90 ± 11	827 ± 94	–	–	Ho et al. (2013)
Phan River, Vinh Phuc, Viet Nam	0.08–27.57	22.0–57.2	3.44–6.32	0.02–0.20	–	–	–	–	–	Tran (2016)
To Lich River (urban river in the Red River Delta), Vietnam	93.5 ± 33.0 to 168 ± 64	1240 ± 779 to 517 ± 297	97.2 ± 31.2 to 150 ± 107	10.4 ± 8.0 to 427 ± 306	55,114 ± 20,029 to 72,681 ± 21,282	179 ± 67 to 262 ± 123	595 ± 131 to 1201 ± 696	35 ± 16 to 73 ± 57	–	Marcussen et al. (2008)
Soil, Red River delta, Vietnam	45.89	109.14	58.14	0.21	–	137.22	–	–	–	Nguyen et al. (2010)
Mangrove sediment, Ba Lat estuary, Vietnam	187 ± 9	393 ± 18	323 ± 13	2.35 ± 0.21	–	81.0 ± 4.0	417 ± 20.0	–	–	Nguyen et al. (2012)
Red River, Vietnam	66 ± 28	127 ± 50	83 ± 55	0.35 ± 0.27	37,600 ± 9900	85.71 ± 23.75	806 ± 236	–	–	Nguyen et al. (2016)
Red River, Vietnam	43.96 ± 20.07	59.43 ± 32.64	63.02 ± 34.41	0.26 ± 0.17	37,052.81 ± 11,332.88	23.23 ± 8.98	745.85 ± 374	20.18 ± 8.56	0.15 ± 0.08	This study
World average	17	67	28	0.09	35,000	92	600	4.8	0.05	Rudnick and Gao (2003) and Taylor and McLennan (1995)



**Table 7** EF and  $I_{geo}$  values of different TME in sediment of some Rivers in the World

River name, country	River description	Pb	Zn	Cu	Cd	Fe	Cr	Mn	As	Hg	Reference
<i>EF value</i>											
Koshi River, (China, Nepal and India)	560 km of mainstream and major tributaries in China (upstream) to Nepal (downstream) was studied.	0.83	0.67	0.85	1.38	-	0.66	-	-	-	Li et al. (2020)
To Lich, Vietnam	Length of 13.5 km, river discharge of $30 \text{ m}^3 \text{ s}^{-1}$ , basin area of $77.5 \text{ km}^2$	8.5	17	6.6	56.7	1	3.2	1.6	4.3	-	Nguyen et al. (2015b)
Red River (China, Laos, Vietnam)	Basin area of $156,450 \text{ km}^2$ , length of 1149 km	$2.41 \pm 0.75$	$0.81 \pm 0.34$	$2.07 \pm 0.83$	$2.71 \pm 1.51$	$1 \pm 0$	$0.24 \pm 0.07$	$1.42 \pm 0.48$	$3.95 \pm 1.22$	$2.69 \pm 1.13$	In this study
<i>I<sub>geo</sub> value</i>											
Koshi River, (China, Nepal and India)	560 km of mainstream and major tributaries in China (upstream) to Nepal (downstream) was studied	-0.58	-0.85	-0.6	0.16	-	-0.92	-	-	-	Li et al. (2020)
Chaghara River, India	1080 km long; basin area of $127,950 \text{ km}^2$ , water discharge of $2990 \text{ m}^3 \text{ s}^{-1}$	-1.5 to -1.1	-3.4 to -3.0	-4.6 to -2.5	-1.1 to -0.7	-	-1.4 to 0.7	-	-	-	Singh et al. (2017)
Beijiang, China	633 km long, located in northern Guangdong province, main tributary of the Pearl River Delta	0.85	0.31	-0.36	2.12	-	-1.75	-	1.17	0.04	Zhao et al. (2018)
Xijiang, China	Basin area of $409,480 \text{ km}^2$ with discharge of $6965 \text{ m}^3 \text{ s}^{-1}$ ; the western tributary of the Pearl River in China	0.33	-0.02	-0.75	2.42	-	-1.6	-	1.15	-0.13	Zhao et al. (2018)
Tanjiang, China	North China	0.98	0.42	0.3	2.81	-	-0.8	-	1.18	0.66	Zhao et al. (2018)
Dongjiang, China	River discharge $808 \text{ m}^3 \text{ s}^{-1}$ ; eastern tributary of the PRD in Guangdong province, major source of water for Hong Kong	0.41	-1.03	-1.52	0.23	-	-2.93	-	0.28	-1.56	Zhao et al. (2018)



Table 7 (continued)

River name, country	River description	Pb	Zn	Cu	Cd	Fe	Cr	Mn	As	Hg	Reference
Pearl River delta, China	Formed by three major rivers: Beijiang, Xijiang and Dongjiang river, in South China	0.56	-0.13	-0.74	1.8	-	-1.92	-	0.94	-0.38	Zhao et al. (2018)
To Lich River, Vietnam	Length of 13.5 km, river discharge of 30 m <sup>3</sup> s <sup>-1</sup> , basin area of 77.5 km <sup>2</sup>	1.1	2.1	0.7	3.8	-2	-0.3	-	0.1	-	Nguyen et al. (2015b)
Red River (China, Laos, Vietnam)	Basin area of 156,450 km <sup>2</sup> , length of 1149 km	0.62±0.75	-1.01±0.95	0.35±0.90	0.60±1.17	1±0	-2.67±0.56	-0.15±0.53	1.34±0.72	0.71±0.95	In this study

m<sup>3</sup>.day<sup>-1</sup>); and Ninh Binh: (13.000 m<sup>3</sup>.day<sup>-1</sup>) (MONRE 2019). Most of the domestic wastewater is directly discharged into the hydrosystem, e.g. only 26.2% of the total domestic waste was treated in Hanoi city in 2019 (MONRE 2019). Moreover, dense industrial parks with diverse industrial production (paint and pigments, tanning, textile and dyeing, paper production, batteries, chemicals and metallurgical, chemical fertilizer, food processing, etc.,) also discharge large volumes of wastewater into the hydrosystem (Nguyen et al. 2015a; MONRE 2019). These industrial parks are present in several provinces of the Red River basin such as Thai Nguyen, Viet Tri, Lao Cai, Bac Giang, Hai Phong and Hanoi. The Viet Tri Park (located next to the Thao-Da-Lo conjunction) has a total industrial wastewater production of 102,000 m<sup>3</sup>.day<sup>-1</sup> and seven provinces (Hanoi, Vinh Phuc, Hai Phong, Hai Duong, Hung Yen, Quang Ninh, and Bac Ninh) in the Red River Delta region release about 155,055 m<sup>3</sup>.day<sup>-1</sup> (MONRE 2009). In addition, in the upstream Red River, the exploitation of numerous mineral mines (carbon, TME, phosphorus, etc.,) in both China and Vietnam has been expanding (Le 2008; Nguyen et al. 2015a).

Comparing the spatial variation of the variables observed in this study, the lowest values were found in the Da and Lo sediments where population densities and industrial production were much lower (Le et al. 2017). In contrast, the highest values were found at the seaward Ba Lat site, at the outlet of the Delta area. Finally, the higher values of Cu were observed during the dry season may reflect the important point sources for this element.

### Influence of agricultural activities

Agricultural activities contribute pollutants to river systems (Goel 2006; Laws 2017). Numerous authors have demonstrated that fertilizers and pesticides used for different crops have strong impacts on the water column and sediment chemistry and ecosystem functioning (Mali et al. 2015; Islam et al. 2015; Li et al. 2020). In the Red River basin, the amount of chemical fertilizer used per hectare can attain 120–180 kg/year, an increase of 3–4 times compared to two decades ago (Nguyen 2013), however, more than 55% of applied chemical fertilizers are not being used by the crops (Truong 2009). As concentrations over the permitted value of the Vietnamese standards in the Red River sediment may partly reflect the application of fertilizers and arsenical pesticides on agricultural lands. This was also observed for the downstream section of the Axios River, Greece where the higher As concentrations were related to the wide use of arsenical pesticides, herbicides, and crop desiccants in the extensive rice fields (Karageorgis et al. 2003). In Bangladesh, the doubling of agricultural phosphate fertilizer use from 1961 to 2000 considerably accelerated As contamination (Boreysza et al. 2006).

**Table 8** Nutrients, total organic carbon and pH values in surface sediment of different rivers in the World

River name, country	TN (%)	TP (%)	TSi (%)	TOC (%)	pH	References
Dagu River, China	–	–	–	0.056–0.192	–	Dong et al. (2020)
Têt River, Gulf of Lions, France	0.03 ± 0.01	–	–	0.24 ± 0.05	–	Pruski et al. (2019)
Cross River, Nigeria	0.11–0.25	–	–	0.88–3.99	–	Dan et al. (2019)
Shenzhen River, China	0.19 ± 0.086	0.16 ± 0.058	–	1.7 ± 0.70	–	Wijesiri et al. (2019)
Ibrahim River, Lebanon	0.03–0.12	–	–	0.54–2.1	–	Ghsoub et al. (2020)
Conwy catchment, North Wales, UK (upland)	0.00084 ± 0.00013	0.00021 ± 0.00005	–	0.0094 ± 0.0016	4.75 ± 0.05	Brailsford et al. (2019)
Conwy catchment, North Wales, UK (lowland)	0.00011 ± 0.00008	0.00021 ± 0.00021	–	0.00072 ± 0.00012	6.87 ± 0.06	Brailsford et al. (2019)
Betwa river, India	0.04–0.19	–	–	0.64–1.05	8.5–8.7	Venkatesh and Anshumali (2020)
Ganges River, India	–	–	31.2	–	–	Ramesh et al. (2000)
Yangtze River estuary, China	–	–	24.6–32.1	0.9–1.7	6.6–8.5	Yuan et al. (2014)
To Lich River, (Red River Delta), Vietnam	–	–	–	1.2–5.3	7.4–8.1	Marcussen et al. (2008)
Day River (Red River Delta), Vietnam	0.21–0.39	0.12–0.17	–	3.94–5.21	–	Trinh et al. (2012)
Phan River, Vietnam	0.028–0.084	0.05–0.08	–	0.034–0.796	6.07–7.83	Tran (2016)
Red River, Vietnam	0.08 ± 0.05	0.06 ± 0.02	29.9 ± 4.6	0.45 ± 0.3	6.98 ± 0.24	This study
World sediment average	–	–	28.5	–	–	Martin and Meybeck (1979)

### Influence of climate and hydrological characteristics on the sediment quality

Previous studies have pointed out that natural factors such as air temperature, rain events and storms, river discharge, and turbulence affect the exchange of pollutants between the water and sediment compartments altering water and sediment quality (Deborde et al. 2007). In tropical river systems, most of the suspended sediment load is transported during the rainy season and high river discharge due to erosion and weathering within the river basins (Li et al. 2020; van Maren and Hoekstra 2004). Suspended sediments are known to be associated with numerous elements. Park et al. (2011) examined the effect of climate variability on the surface water quality of the Mekong River and found that the dissolved ion concentrations tended to be higher during the dry periods, whereas suspended sediment, dissolved organic matter and poorly soluble metals were significantly higher during the wet season. These authors also highlighted that rainfall variability is strongly linked to surface water quality in East Asia.

Our study showed the higher concentrations of sediment TOC and some TME (Zn, Cd, and Hg) in the rainy season that may be related to the strong erosion and weathering in the basin. According to the latest IPCC report (2021), severe daily precipitation events are predicted to grow by around 7% for every 1 °C increase in global warming. Air temperatures are predicted to increase by 2 °C by the middle of this

century and by 4 °C by the end of this century in Southeast Asia (Amato and Hein 2014). An increase in air temperature and rain events in the upcoming decades in Southeast Asia may accelerate erosion and weathering processes, exacerbating pollutant discharge into hydrosystems (Zhang et al., 2018).

In contrast, studies from some urban rivers, for example, the Korotoa River in Bangladesh, revealed that the Cd and Pb concentrations in sediment were greater during the dry season due to their significant accumulation in sediment during periods of low river discharge (Islam et al. 2014, 2015). Similarly, the higher Cu concentrations observed during the dry season in our work may be related to Cu precipitation in the surface sediment during low river flow in the dry season.

Spatially, we found the highest TOC and some TME concentrations at the Ba Lat site in the estuary and the lowest values at the Hoa Binh site on the Da River ( $p < 0.05$ ). This may also reflect the impact of the variable hydrological regime. Low river flow in the flat deltaic river may provide favorable conditions for the precipitation of suspended solids to the sediments, resulting in organic-rich sediments, as has been reported for the Day River in the Red River Delta (Trinh et al. 2012). In contrast, the lower concentrations of most variables (except for Si) at the Hoa Binh site could be explained by the higher river slope and higher river discharge at this site, which may accelerate riverbed erosion, leading to less accumulation of substances.



In addition, as presented above, the surface sediment concentrations of TOC, nutrients (TN, TP), and some TME of the Red River were much lower than in several Vietnamese urban rivers or in other rivers in the world. We assume that these lower concentrations in the Red River surface sediment were due to the high river discharge causing strong erosion and absorption/desorption process at the interface of riverine water-surface sediment.

### Influence of dams impoundment on spatial variation of sediment quality

The dams are known to strongly impact downstream water and sediment as a consequence of significant sediment and associated elements deposition in the dams. In the Red River, a series of hydroelectric dams in the upstream Red River both in China and Vietnam have been constructed over the last few decades (Wei et al. 2019b) significantly reducing riverine sediment concentration in the downstream river (Dang et al. 2010; Vu et al. 2014; Le et al. 2020). Within the three tributaries Da, Lo and Thao, our results show that the TME, TOC and nutrients concentrations were always lowest at the Hoa Binh site on the Da River downstream of numerous large and medium-sized reservoirs in the mainstream of the upstream part, whereas higher concentrations of almost all elements were found in the Lo and Thao Rivers where fewer reservoirs are constructed were observed. Similar observations on the decrease of sediment TME concentrations have also been reported for other rivers in the world. For example, a significant amount of TME, especially Zn, Pb, Cd, and Cr, associated with suspended sediment, was retained by the Prochoma dam located on the Axios River in Greece (Karageorgis et al. 2003).

### Influence of lithological and geomorphological characteristics on the sediment quality

Lithology and mechanical erosion also play a considerable role in the export of different variables to riverine water and sediment. Studies on the sediment quality of the Aliakmon River (Greece) have reported that natural weathering of abundant ophiolites was the primary source of certain TME (Cr and Ni) enrichment in the sediment, whereas human activities had a little influence on the river system (Economou-Eliopoulos and Vacondios 1995; Karageorgis et al. 2003). Similar results were demonstrated for the trans-Himalayan Koshi River where Cr, Co, Ni, and Zn were primarily derived from the parent rock (Li et al. 2020).

In Asia, the high concentrations of some elements, notably Si, Fe and As are due to the geological formations rich in these metals, particularly for the rivers situated in the

Himalayan region (Meybeck 1987; Barton et al. 2004; Berg et al. 2007).

Previous studies demonstrated that the upstream Red River was characterized by Paleozoic sedimentary 55.5–72.7%, by Mesozoic silicic (18.0–21.5%), and Mesozoic carbonated (14.7–16.7%) rocks, whereas alluvial deposits covered the delta area (Moon et al. 2007). The Si concentrations found in the Red River sediment in this study were higher than the average value for the river sediment of the world., This is probably due to its derivation from natural rocks (silicate and carbonate) as well as being a reflection of the high physical/chemical erosion rates in the upstream Red River basin (Barton et al. 2004; van Maren and Hoekstra 2004).

High Fe concentrations (39,110 ppm) have also been detected in the Red River Delta soils (Trinh et al. 2012) (Table 6). This is also the case in the sediments of urban rivers in the Red River Delta such as the To Lich–Kim Nguu rivers ( $55,114 \pm 20,029$  to  $72,681 \pm 221,282$  ppm) (Marcusen et al. 2008) and Day River (28,060–47,430 ppm). The high sediment Fe concentrations observed along in the Red River in this study, partly suggest a natural source probably from the weathering processes of red laterite soils. This is similar to what was observed for the high sediment Fe concentrations (4151.8–17,318.8 ppm) of the Hindon River, India (Chabukdhara and Nema 2012).

Similarly, very high soil As concentrations, especially in carboniferous rocks of the Himalayan region have been previously reported (Polizzotto et al., 2006; Dang 2011). The elevated rates of soil leaching and rock weathering in this region have led to high As contamination in the riverine sediment and water. Thus, the abundance of Si, Fe and As in the Red River sediment also supports the conclusion that the geochemistry of large river sediments could provide insights into the erosional processes (Ramesh et al. 2000; Li et al. 2020).

The low  $I_{geo}$  and EF values in some hydrosystems may provide insight into the natural inputs of some metals. For example, in the Xiangjiang River, South China, some metals (Cr, Ni, and Co) with low  $I_{geo}$  values were found to be derived from natural processes (soil erosion and rock weathering) (Huang et al. 2020). In our study, the low values of both  $I_{geo}$  ( $< 1$ ) and EF ( $< 1.5$ ) of some sediment metals (e.g. Cr, Zn, and Mn) may also reflect their dominant natural sources. Indeed, a previous study observed an increase in some sediment metal (Cr, Fe and Al) concentrations together with the increasing clay fraction from the upstream towards the coast and highlighted the control of particle size on metal concentrations as well as the role of natural weathering processes in the Red River (Nguyen et al. 2016).



## Influence of a combination of natural characteristics and human activities

In many cases, both anthropogenic and natural inputs were responsible for exacerbating some metal concentrations in riverine sediment. Some authors indicated that the negative relationship between observed variables was due to the combination of geochemical support and mobility of the sources, whereas the positive correlation may suggest common sources, mutual dependence, and identical behavior during transportation (Liu et al. 2009; Singh et al. 2017). In our study, sediment TSi showed clear negative correlations with all other variables (TME, TOC, TN, and TP) concentrations, whereas high positive correlations between some variables (TOC with TN, TP, Zn, Pb, As, and Hg) (Pearson correlation coefficients > 0.6) were observed (Table 5). It is probable that silica in the Red River sediment was mainly derived from natural sources, whereas other variables were probably originated from both natural and anthropogenic sources.

Regarding the seasonal variation of metal concentrations, our results show a clear increase of some metals (Zn, Cd, and Hg) in the rainy season in 2019 (Fig. 2), suggesting the important contribution of the diffuse sources from the parent rocks and agricultural land. The combination of complex sources for Zn was also reported for the Pô Kô River in Vietnam where high sediment Zn concentration (mean: 201.93 ppm) was due to the erosion/leaching of natural rocks containing high Zn metal, together with the domestic wastewater source (Nguyen et al. 2018a). For some other variables (e.g. TN, TP, Mn, Cr, and Pb), no significant variation between their concentrations in the rainy and dry seasons was observed (Fig. 2), probably reflecting their complex sources. A clear positive correlation between some TME and TOC and nutrients (TN and TP) (Table 5) suggests the building of geochemical associations during their transportation and their similar anthropogenic sources (e.g. agricultural soils, industrial and domestic wastewater), as has been observed in the Yunnan-Guizhou Plateau, China (Zhang et al. 2018). Some authors observed high Fe concentrations in the Red River water and suggested that its pollution source was due to a combination of natural characteristics and human activities (e.g. mining exploitation, industrial wastewater release) in the basin (Nguyen et al. 2015a; DONRE-Lao Cai 2011). This is similar to the situation for As where both natural input and anthropogenic origins (the excess of fertilizers use in agricultural land, untreated domestic and industrial wastewater...) have contributed to As contamination, notably in the urban rivers in the Red River delta (Nguyen et al. 2015a, b). This is similar to the case in Bangladesh where high As concentrations were due to both lithological characteristics and a doubling of phosphate fertilizer use for agriculture (Boreysza et al. 2006).

The accumulation of metals and organic matter in sediment is known to affect benthic organisms and human health through accumulation up the food web. Our results showed that some TME (Pb, Cu, As, Hg) were at the effects range low/median (ERL/ERM) proposed by NOAA (1999), thus effective measures should be taken to minimize these metal loads entering the Red River. In addition, our previous study revealed the extent of human-induced impacts in the Red River basin including increased population, industrial production, and land-use change with the deforestation and the modification of intensive cultivation practices over the past decades (Le et al. 2015; 2018). With such trends of intensifying human activities together with climate change, the water and sediment quality of the Red River may be more strongly impacted in the next decades. Thus, preventive measures of environmental management, including wastewater treatment, are needed to control the pollution sources in this basin.

## Conclusion

The TME average concentrations in the Red River sediments in this study were in the decreasing order of  $Fe > Mn > Cu > Zn > Pb > Cr > As > Cd > Hg$ . The concentrations of Fe and As were higher than the allowed values of the Vietnam National technical regulation on the surface sediment quality QCVN 43:2017/BTNMT. Additionally, the geoaccumulation index ( $I_{geo}$ ) and enrichment factor (EF) data also indicated that the Red River sediments were polluted by As; moderately contaminated with Cu, Pb, Cd, and Hg; and uncontaminated by other metals (Mn, Cr and Zn).

TOC ( $0.43 \pm 0.31\%$ ) TN ( $0.09 \pm 0.04\%$ ) and TP ( $0.060 \pm 0.018\%$ ) concentrations in Red River sediments were low in comparison to some urban rivers, whereas the TSi concentration ( $29.93 \pm 4.52\%$ ) was high. The low concentrations of TOC, TN and TP in the Red River surface sediment are probably explained by the high river discharge which strongly impacts erosion and absorption/desorption processes at the water–sediment surface interface, whereas high TSi may reflect the basin lithological characteristics.

The lowest values of almost all variables were found at the Hoa Binh site where the lowest population densities, human activities, and highest river discharge were observed, whereas most variables were highest at Ba Lat site, the outlet of the Red River Delta where human activities clearly impacted on Red River system. For the whole Red River basin, the results show that the Red River surface sediment was impacted by different sources from both natural processes and human activities. Some variables (TSi, Zn, Cd, and Hg) were dominated by diffuse sources; Cu was mainly impacted by point sources, whereas other variables (TOC, TN, TP, Pb, As, Fe, Mn, and Cr) were affected by both diffuse





and point sources in the basin. These results may help guide better management decisions necessary for the sustainable development of the Red River basin.

Our study did not investigate the speciation of metals or the toxic organic substances of sediments which would have provided a more accurate appraisal of the risk of the pollutants in the aquatic ecosystems. In addition, long-term observations of surface sediment quality are needed for the understanding of the influence of intensified climate change and human activities on the Red River system and on other rivers in Asia.

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**Code availability** Not applicable.

## Declarations

**Conflict of interest** The authors declare that they have no conflict of interests that are relevant to the article content.

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