



Seasonal, spatial variation, and pollution sources of heavy metals in the sediment of the Saigon River, Vietnam[☆]

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ABSTRACT

The current study was conducted to (1) examine seasonal and spatial distribution of heavy metals and metalloid in sediment from the Saigon River and (2) apportion and quantify their pollution sources. Ninety-six sediment samples were taken in the rainy and dry season on 13 sampling sites, distributed over the lower reaches of the River, to analyze for exchangeable concentration of 11 heavy metals and metalloid (Al, B, Cd, Co, Fe, In, Mn, Ni, Pb, Sr, and Zn), pH, EC, organic carbon content, and particle-size distribution. Generally, the concentration of 11 elements was ranked in the order Mn > Al > Fe > Zn > Sr > In > B > Ni > Co > Pb > Cd. Hierarchical cluster analysis grouped 13 sampling sites into two parts based on the similar concentration of the 11 elements. Three-way analysis of variance showed that the total exchangeable concentration of 11 elements was significantly higher in the rainy season than in the dry season and in the upper part than in the lower part of the river. Principal component analysis/factor analysis and correlation analysis revealed that three pollution sources (PS) may contribute to enriching the 11 examined elements in the sediment. These sources included (PS1) from catchment through water erosion over natural areas, explaining 83%, (PS2) mixed sources from catchment through water erosion over agricultural fields and inside Ho Chi Minh City, accounting for 6%, and (PS3) mixed sources from lowland areas, explaining 7.8% of the total variance of the elements. In brief, the sediment concentration of 11 metals and metalloid varied with season and space and three major pollution sources from river catchment, inside Ho Chi Minh City, and lowland contributively enriched the elements in the sediment of the River.

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1. Introduction

Heavy metal status in aquatic ecosystems is critically important because the metals can affect the biodiversity of the ecosystems through providing nutrients, if their concentration within certain

levels, or inducing toxicity, if exceeding specific high thresholds. Therefore, their pollution status in aquatic ecosystems is an increasingly concerning issue widely, also because of their persistence, bioaccumulation, and non-biodegradability (Jaishankar et al., 2014; Bawuro et al., 2018; Ayangbenro and Babalola, 2017). Aquatic sediment, a component of aquatic ecosystems, is much susceptible to heavy metal pollution because it can serve as a sink of deposited materials, including heavy metals (Duncan et al., 2018; Ugbomeh et al., 2019). The sediment is also a source of pollutants such as heavy metals to the ecosystems upon re-suspension of the deposited metals back to surface water (Tang et al., 2014). With a relatively long residence time, sediment pollution of heavy metals is

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important to study to identify and quantify their sources for better management.

Heavy metals in the river sediment could come from two main sources of the nature and anthropogenic activities. In nature, heavy metals are constituents of soil parent materials, and thus they are commonly present in the soil at various magnitudes, after a pedogenetic process of weathering of the parent materials (Lenart-Boroń and Boroń, 2014). Because soil erosion is a selective process of preferentially detaching and transporting fine materials (Quinton et al., 2001), which are commonly bound with heavy metals (Qu et al., 2019), soil erosion, followed by transport and deposit of the eroded materials to rivers, could increase the concentration of heavy metals in the aquatic sediment. Similarly, the erosion was noticed as an important process transporting some heavy metals from their source to downstream sediment in Tunisia (Othmani et al., 2015). The anthropogenic sources of metals in sediment could come from various origins, including industry, agriculture, and residential areas (Masindi and Muedi, 2018). Consequently, positions close to or surrounded with residential, plant, or industrial centers could be more polluted with heavy metals than the others.

In general, heavy metals and metalloids in sediment exist in two forms of unavailable and available, together making the total content. Naturally, heavy metals and metalloids in sediment are much less mobile, present in a low available concentration because they tend to bind to sediment components strongly (O'Geen et al., 2010). The natural sources, especially native soil metals and metalloids, could have a low available concentration due to their existing in highly immobile forms (Orroño and Lavado, 2009). Meanwhile, Ayangbenro and Babalola (2017) indicated that the anthropogenic sources of metals tended to be more mobile and thus having relatively high available concentration. In some environments, sediment was contaminated with some metals, but the exchangeable form, which could directly affect living organisms typically native flora, was lowest of the five examined forms (residual, reducible, oxidizable, carbonate, and exchangeable) of metals in sediment (Pradhanang, 2015). Therefore, the total content of metals in sediment may be high but the extractable or exchangeable portion could be low, making the information about the total content less meaningful in term of potential risks to environment (Asmoay, 2019). Consequently, the current study was conducted, based on the exchangeable form of 11 metals and metalloid of sediment in the Saigon River to more focus on apportioning and quantifying the anthropogenic sources of the elements.

The Saigon River is located in southern Vietnam, originating from southeastern Cambodia, crossing three provinces (Binh Phuoc, Binh Duong, and Tay Ninh) before reaching Ho Chi Minh City, Vietnam. The River is important because it provides fresh supply water for a part of Ho Chi Minh City. In addition to many small canals surrounding the River, there are 5 large canal systems, including Thamluong-Bencat, Tanhoa-Logom, Tauhu-Benghe, Nhieuoc-Thinghe, and Doi-Te originating from inside Ho Chi Minh City, together adding water to the river. These large canal systems could be the main anthropogenic sources of heavy metals and metalloid in the river sediment. A study by Hoang et al. (2007) found that these canals were more polluted with heavy metals than the mainstream of the Saigon River, indicating that one of the main sources of sediment heavy metals of the River could originate from inside Ho Chi Minh City. Moreover, the heavy metals and metalloid in the sediment of the River could come from three main pathways, including (1) water erosion and runoff of dissolved/suspended materials, and agricultural, industrial, and domestic wastes from upper catchment area, (2) runoff from inside Ho Chi Minh City, and (3) high tide bringing suspended materials back to the River. There

have been a few studies conducted to examine the heavy metal status in the sediment of the Saigon River or related (Huy et al., 2003; Hoang et al., 2007; Phuong et al., 1998). A general common argument/or assumption from these studies was the pollutant sources of heavy metals possibly originating from inside Ho Chi Minh City through runoff of agricultural, industrial, and domestic wastes.

However, no study was conducted to analyze and quantify the various sources (from river catchment, inside Ho Chi Minh City, and outside the river estuary) of heavy metals and metalloid in the sediment of the Saigon River. In addition, our up-to-date literature search revealed that there are limited studies conducted to both apportion and quantify pollution sources of heavy metals and metalloid in the river and its sediment. While principal component analysis/factor analysis (PCA/FA) was applied to apportion the pollution sources of heavy metals and metalloid in river sediment in many studies (Islam et al., 2018; Shen et al., 2019) no study was carried out to quantify the contributive percentage of individual pollution sources identified through PCA/FA.

Therefore, the current study was conducted to (1) examine seasonal and spatial variation of the exchangeable concentration of heavy metals and metalloids in surface sediment from the Saigon River and to (2) apportion and quantify their pollution sources. Heavy metals and metalloids accumulated in the surface sediment of a river could vary with season, spatial position, and be affected by various pollution sources. It was hypothesized that (1) the effect of rainy season was greater in the upper part than in the lower part of the River while the effect of dry season was lesser different between the two parts, and (2) pollution sources of the metals could be derived from the river catchment and from inside Ho Chi Minh City through runoff of various wastes. To apportion and quantify the pollution sources of heavy metals and metalloids, sequential steps of multivariate analyses, including PCA/FA, the Pearson correlation matrix, and PCA factors-based multiple regression analysis were carried out in the current study.

2. Materials and methods

2.1. The studied area

The current study was conducted on the lower reaches of the Saigon River. The river originates from southeastern Cambodia with a length of about 250 km and a catchment area of 4717 km² (Lahens et al., 2018) (Fig. 1). The river flows through Ho Chi Minh City in the lower reaches before adding water to the Dong Nai River. Ho Chi Minh City is an economic and political center in southern Vietnam, with a fast growth rate recently. The population of the City was around 8 859 688 (updated in January 2019) with an increasing rate of 2.15%/year for the last 10 years (Tran, 2019). Ho Chi Minh City is situated on a relatively flat topography with a dense hydrological waterway network (700 km length), being affected by the semidiurnal-tide regime from the East Sea of Vietnam and varying rainfall in the rainy and dry seasons. The tropical climate regime of the studied river is characterized with two distinct seasons, the rainy season from May to October and the dry season from December to April (van Emmerik et al., 2018). The annual rainfall of Ho Chi Minh City is around 1868 mm, more concentrated on the rainy season, and the average temperature is 27.4 °C. More information about the Saigon River and Ho Chi Minh City can be seen in the study of Lahens et al. (2018).

2.2. Experimental factors and setup

Two experimental factors were examined, including season and spatial position, which could induce varying effects on the heavy

metal status of sediment. Thirteen sampling sites were examined and positioned to take sediment samples in four sampling series. The first two series were conducted in the rainy season in August and September 2018 and the last two were in the dry season in March and April 2019. For each of sampling sites from site 2 to site 12, two sediment samples per site were taken on the confluence of the Saigon River and its tributaries (urban canals), one on the mainstream of the River (sample a) and one on the mouth (70–150 m from the River, sample b) of the urban canal (Fig. 1). This was to differentiate the heavy metal concentration of the river

position and canal position connected to the river position, in order to examine spatial variation (hereafter called river position). In addition, the 13 sampling sites were further divided into two parts (lower and upper part, hereafter called River part) based on hierarchical cluster analysis (see below for more details of the method). For the site 1 and site 13, one sediment sample per site (sample a) was taken to make comparisons of input and output sediment. The total sediment samples taken in one sampling series were 24 (1 (site 1) +11 (sites 2 to 12) *2 (samples a and b) +1 (site 13)), and total sediment samples for the current study was 96 samples.

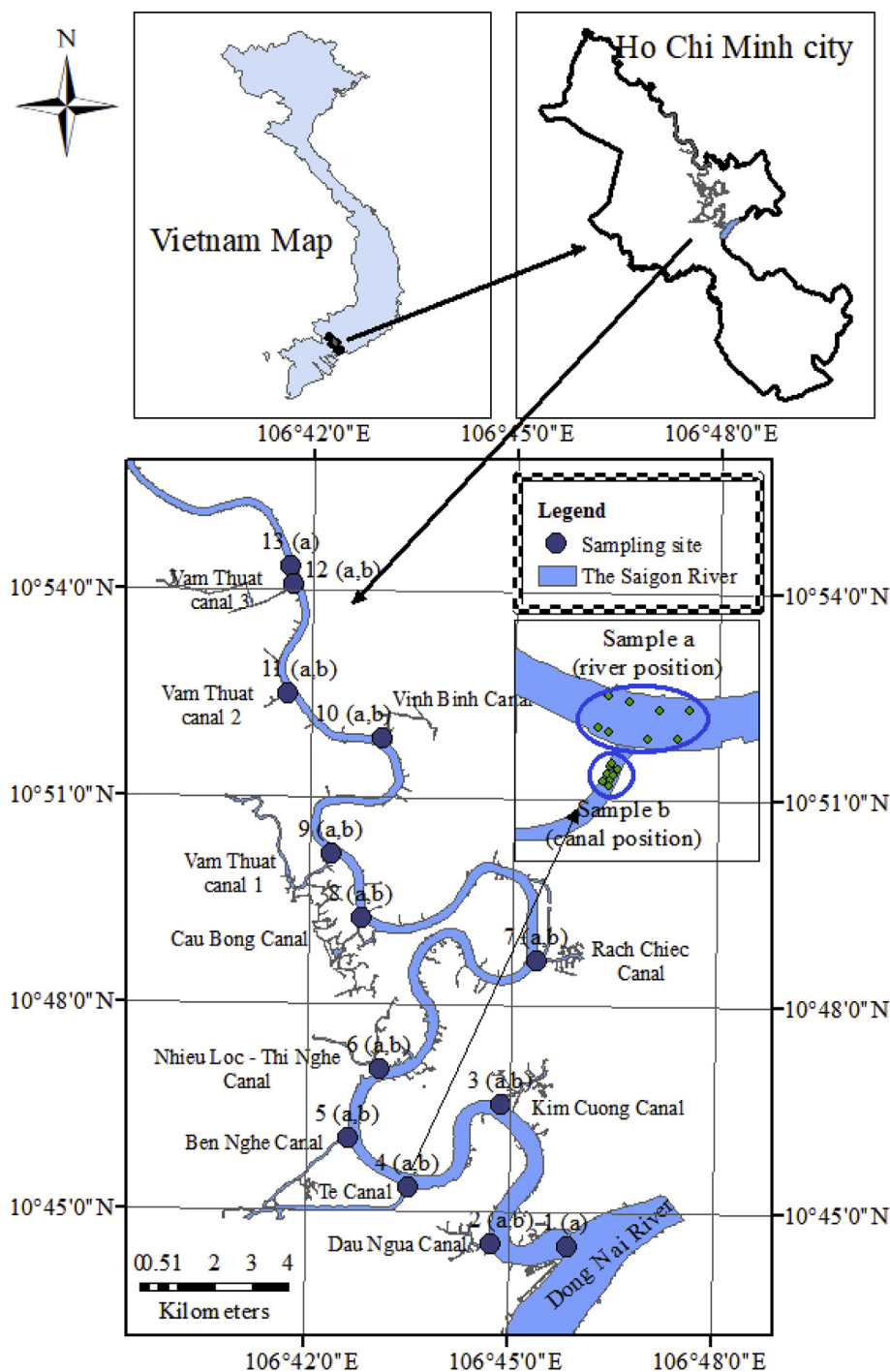


Fig. 1. Sampling sites and map of the Saigon River in Ho Chi Minh City, Vietnam. (a) is the sample on the mainstream of the River, and (b) is the sample on the mouth of the urban canal.

2.3. Sampling and chemical analysis

On the day of sampling, a boat was used to travel from the mouth of the River to the other end to take sediment samples, using a Petersen grab. For one sediment sample, 8 grabs distributed over the two sides of the River or canal mouth were conducted and sediment from these grasps for the top 0–10 cm layer was carefully taken with a stainless steel knife into a plastic bucket. After well mixing around 5 kg of the sediment from the bucket was taken into a polybag, which was immediately stored in an ice-chest at 4 °C and transported to a laboratory for analyses. A global positioning system (GPS) device was used to locate the 13 selected sites for four sampling series.

The sediment samples were air-dried in a laboratory and ground to pass through a 2-mm sieve before doing chemical analyses. All sediment samples (24 * 4 = 96) were analyzed for pH, electrical conductivity (EC), organic carbon (OC) concentration, and exchangeable concentration of 11 metals and metalloid. In addition, 48 sediment samples taken in the first two series were analyzed for particle-size distribution (Carter and Gregorich, 2008). The materials were added with distilled water at 1:2 (w/w) ratio and the extract was measured for pH and EC. TOC was measured using the Walkley – Black method. The exchangeable concentration of heavy metals (Al, Cd, Co, Fe, In, Mn, Ni, Pb, Sr, and Zn) and metalloid (B) were measured using the barium chloride method (Carter and Gregorich, 2008). The method was initiated by weighing 0.5 g of air-dried and ground sediment into a 50 mL centrifuge tube, which was added with 30.0 mL of 0.1 M BaCl₂ and shaking the tube on an end-over-end for 2 h. The filtrate after centrifuging the tube for 15 min and filtering the supernatant with Whatman No. 41 filter paper was measured for 11 heavy metals and metalloid using an Inductively coupled plasma - optical emission spectrometry (ICP-OES).

2.4. Statistical analyses

Multivariate analyses (hierarchical cluster analysis, principal component analysis/factor analysis) (Jin et al., 2019; Phung et al., 2015) were applied to group the 13 sampling sites based on the exchangeable concentration of 11 examined metals and metalloid and to identify as well as quantify potential pollution sources of the metals. Hierarchical cluster analysis (CA) was conducted on the entire data to classify 13 sampling sites into two groups (lower part and upper part) having similar heavy metal concentrations. Principal component analysis/factor analysis (PCA/FA) to apportion potential pollution sources and identify important heavy metals associated with the pollution sources was applied on the whole data, following the procedure described by (Jin et al., 2019) and (Phung et al., 2015). Prior to PCA/FA, Kaiser-Meyer-Olkin (value = 0.698) and Bartlett's Test (p = 0.000) were conducted to determine the data suitability for PCA/FA (Giri et al., 2019). Multiple regression analysis to quantify the contributive percentage of individual pollution sources (varimax factors) identified from PCA/FA to the total variance of exchangeable concentration of 11 examined metals and metalloid (TCM). The stepwise method was used to eliminate any uncorrelated factors and thus establish a final significant regression model (Putri et al., 2018). The Pearson correlation coefficients were calculated to examine the relationship among the heavy metals to help determine similar pollution sources of the metals within a varimax factor. Analysis of Variance (ANOVA) was conducted to compare the exchangeable concentration of the metals, being affected by the experimental factors (season and river position, as well as river part from the cluster analysis). A full statistical model applied to examine three-way interaction of season, river position, and river part was $\gamma_{ijke} =$

$\mu + \beta_i + \alpha_j + \beta_{ij} + \tau_e + \beta\tau_{ie} + \alpha\tau_{je} + \alpha\beta\tau_{ije} + \varepsilon_{ijek}$, where γ_{ijke} is the response of individual combination; μ is overall mean; β_i is a fixed effect of the *i*th season; α_j is the fixed effect of the *j*th river position; $\beta\alpha_{ij}$ is the interactive effect of season and river position; τ_e is the fixed effect of *e*th river part; $\beta\tau_{ie}$ is the interactive effect of season and river part; $\alpha\tau_{je}$ is the interactive effect of river part and river position; $\alpha\beta\tau_{ije}$ is the interactive effect of season, river position, and river part, and ε_{ijek} is the random error with mean Zero and having a normal distribution (Akhtar and Memon, 2009). When the ANOVA result indicated a significant effect at $P \leq 0.05$, Tukey's Honest Significant Difference test was used to classify treatment means. Spatial variation of the heavy metals was examined using scatter plot method. All statistical analyses were carried out using JMP pro 13 (SAS Institute Inc, NC, USA). Figures were established using Sigmaplot 12 (Systat Software Inc.).

3. Results

3.1. General status of the exchangeable concentration of metals in sediment

Cluster analysis was conducted on 11 measured metals and metalloid to group 13 sampling sites into two main clusters (Fig. 2), based on metal concentration similarity. It was interesting that sampling sites from 1 to 6, located in the lower part of the Saigon river, were grouped into cluster 1, hereafter called lower part, and sampling sites from 7 to 13, located in the upper part of the studied river, were combined into cluster 2, hereafter called upper part.

The mean exchangeable concentration of 11 metals and metalloid, organic carbon, values of pH, EC and particle-size distribution of sediment were shown in Table 1. Of the 11 measured metals and metalloid, Al and Mn were highest in exchangeable concentration, 105 and 107 mg kg⁻¹, respectively, and Cr and Pb were lowest in exchangeable concentration, 0.06 and 1.3 mg kg⁻¹, respectively. The mean total exchangeable concentration of 11 metals and metalloid varied from 187 (in the dry season, lower part, and canal position) to 659 mg kg⁻¹ (in the rainy season, upper part, and canal position). The averaged value of pH varied from 3.98 to 5.36 and that of EC was from 1363 to 3017 μS cm⁻¹. Mean concentration of organic carbon was from 3.62 to 5.28%. Particle-size distribution of sediment was analyzed with the samples collected in the rainy season and its values were also shown in Table 1.

The total exchangeable concentration of all 11 metals and metalloid (TCM) was significantly affected by two single experimental factors, the rainy season (Fig. 3a) and the river part (Fig. 3b). The rainy season had significantly higher TCM (489) than the dry season had (227, mg kg⁻¹) and the upper part had significantly higher TCM (427) than the lower part had (289, mg kg⁻¹). Fig. 3c showed that the TCM in the rainy season was located above the TCM in the dry season. In both seasons, TCM tended to increase with sampling site, from site 1 in the river mouth toward site 13 in the other side.

3.2. Results of statistical analyses

The results of principal component analysis/factor analysis conducted on the 11 metals and metalloid of sediment samples collected from two seasons, in two river parts, and two river positions were shown in Table 2. Three significant varimax factors (having eigenvalue greater than 1) were extracted that together explained 75.46% of the total variance of all exchangeable concentrations of the 11 measured metals and metalloid. The most important factor (VF1), explaining 40.8% of the total variance, had high loading values (greater than 0.5) with four metals, Al, Co, Fe,

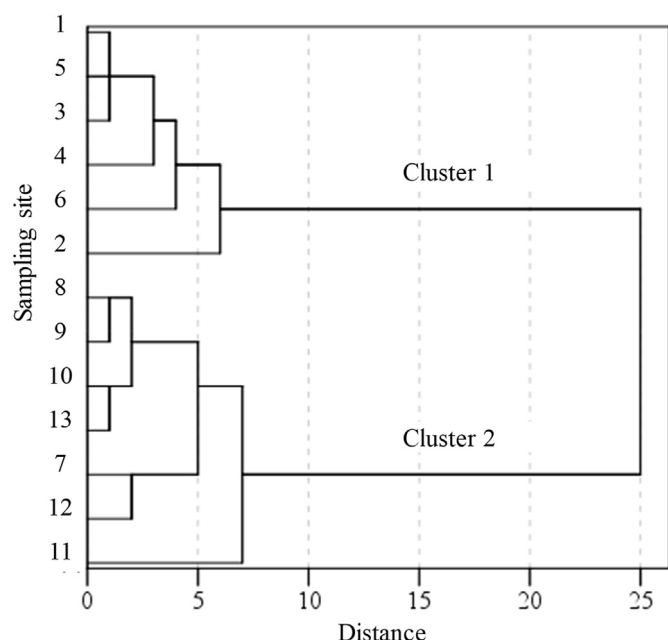


Fig. 2. Dendrogram from cluster analysis showing two clusters of the 13 sampling sites.

and Ni. The second most important factor (VF2), explaining 22.6% of the total variance, had high correlation coefficients with five metals, including Cd, Co, Ni, Pb, and Zn. Similarly, factor 3, explaining 12.07% of the total variance, had a strong relationship with three metals, In, Mn, and Sr.

Correlation analysis was conducted to investigate the relationship among the 11 metals and metalloid and the Pearson correlation coefficient was calculated by using simple linear regression analysis (Table 3). In general, the coefficients greater than 0.2 was statistically significant with probability <0.05 . Al was significantly correlated with B, Co, Fe, Ni, Pb, Sr, and Zn and the highest coefficients were with Co (0.79), Fe (0.66), and Ni (0.72). Boron was statistically significantly correlated with 6 metals. Similarly, Cd was correlated with 4 metals; Co was with the other metals, Fe with 5 metals, In with 3 metals, Mn with 3 metals, Ni with 8 metals, Pb with 6 metals, Sr with 8 metals, and Zn with 8 metals.

Multiple regression analysis was conducted to quantify the dependency of TCM on three varimax factors extracted from PCA/FA (Table 4). TCM was significantly correlated with three extracted factors, of which factor 1 explained 83.2%; factor 2 explained 5%, and factor 3 explained 7.8% of the total variance of TCM. Together, the three factors explained 96% of the total variance of TCM.

3.3. The exchangeable concentration of metals in three varimax factors

Because Al, Co, Fe, and Ni had high correlation coefficients with

Table 1

Mean concentration (mg kg^{-1}) and standard deviation of the mean (SE) of 11 metals and metalloid, pH, EC, organic carbon (OC), and particle distribution.

Parameter	Statistics	Rainy season				Dry season			
		Lower part		Upper part		Lower part		Upper part	
		Canal	River	Canal	River	Canal	River	Canal	River
Al	Mean	119.5	47.4	250.6	223.7	19.6	23.6	59.9	38.8
	SE	45.4	43.1	63.1	37.8	1.1	2.3	22.7	3.9
B	Mean	4.33	3.90	3.98	3.95	5.03	5.82	4.55	4.60
	SE	0.17	0.13	0.10	0.10	0.09	0.50	0.08	0.15
Cd	Mean	0.07	0.04	0.10	0.07	0.03	0.03	0.07	0.07
	SE	0.02	0.02	0.01	0.01	0.00	0.00	0.01	0.01
Co	Mean	3.36	2.11	4.95	4.50	0.82	0.72	2.19	2.16
	SE	0.62	0.60	0.44	0.26	0.15	0.11	0.31	0.14
Fe	Mean	94.6	40.4	163.2	99.0	19.9	27.7	78.3	65.7
	SE	35.2	34.8	80.0	22.5	4.7	9.1	19.1	5.8
In	Mean	12.05	11.46	9.02	8.55	8.53	8.94	7.50	7.14
	SE	1.37	1.12	0.58	0.53	0.50	0.30	0.20	0.23
Mn	Mean	172.5	162.1	106.3	95.6	99.2	108.7	71.1	62.8
	SE	31.3	25.2	13.6	11.5	13.4	8.2	4.8	6.1
Ni	Mean	4.83	2.27	8.30	6.93	2.37	0.87	3.34	3.23
	SE	1.02	0.92	0.89	0.43	0.62	0.26	0.60	0.26
Pb	Mean	1.04	1.10	2.08	2.00	0.88	0.88	1.06	1.11
	SE	0.22	0.40	0.42	0.56	0.03	0.01	0.06	0.03
Sr	Mean	17.1	14.3	11.6	11.1	20.8	20.5	12.9	12.3
	SE	2.6	2.1	1.5	1.1	1.2	0.9	0.7	0.7
Zn	Mean	36.0	15.5	99.0	75.0	10.2	4.5	41.2	39.6
	SE	8.7	8.0	12.9	7.0	2.6	0.6	7.2	4.3
Total concentration	Mean	465.3	300.5	659.2	530.5	187.4	202.3	282.2	237.6
	SE	94.2	94.9	127.6	59.1	16.1	15.2	48.6	13.8
pH	Mean	4.75	4.89	3.98	4.18	5.25	5.36	4.92	5.21
	SE	0.32	0.33	0.30	0.19	0.10	0.06	0.14	0.07
EC ($\mu\text{S cm}^{-1}$)	Mean	1627	1460	1733	1362	3017	2938	1802	1447
	SE	135	129	159	74	339	280	249	154
OC (%)	Mean	3.62	3.95	4.26	5.28	3.88	3.58	5.25	4.84
	SE	0.28	0.28	0.35	0.31	0.18	0.25	0.37	0.37
Sand (%)	Mean	19.5	23.4	25.8	26.2				
	SE	2.7	2.9	2.4	2.5				
Silt (%)	Mean	70.8	65.6	65.5	64.1				
	SE	1.8	2.1	1.8	2.1				
Clay (%)	Mean	9.8	10.9	8.8	9.7				
	SE	1.2	1.2	1.0	0.8				

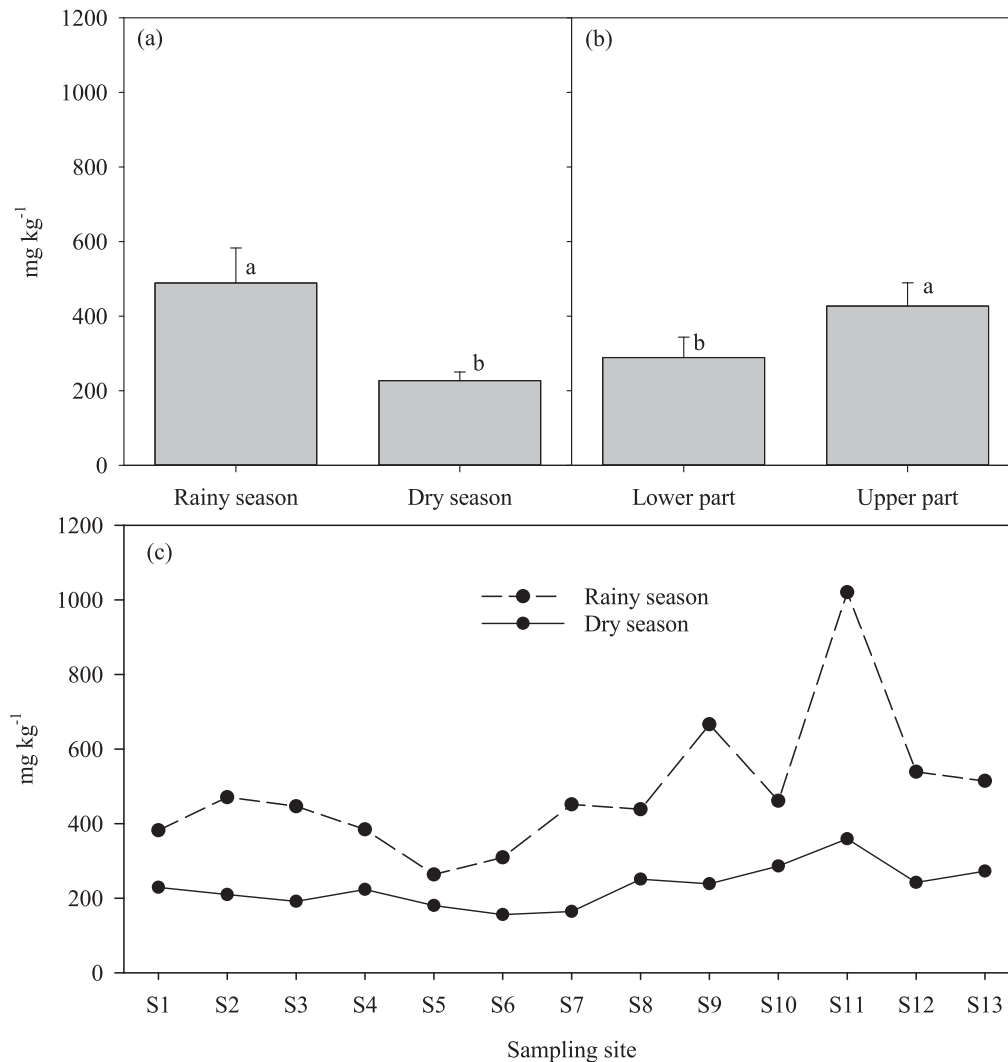


Fig. 3. The total exchangeable concentration of 11 examined metals and metalloid in the sediment as affected by season (a) and river part (b) as well as varying with the sampling site (c). Within a panel, a and b, bars attached with the same letters are not significantly different from the other. Error bars indicate standard errors.

Table 2

Loading values of 11 examined heavy metals and metalloid from principal component analysis/Factor Analysis. Bold numbers are those greater than 0.75, and underlined numbers are those greater than 0.5 and smaller than 0.75.

Parameters	Varimax factor (VF)		
	VF1	VF2	VF3
Al	0.87	0.24	0.04
B	-0.37	-0.33	0.18
Cd	0.02	0.86	0.14
Co	0.79	<u>0.55</u>	0.08
Fe	0.88	-0.10	0.00
In	0.13	0.06	0.96
Mn	0.12	0.05	0.96
Ni	<u>0.64</u>	<u>0.69</u>	0.06
Pb	0.05	<u>0.55</u>	-0.07
Sr	-0.38	-0.22	0.75
Zn	0.36	0.86	-0.11
Eigenvalue	4.49	2.48	1.33
% total variance	40.80	22.59	12.07
Cumulative percentage variance	40.80	63.39	75.46

factor 1, the exchangeable concentration of these metals was summed up and analyzed to examine the effects of experimental factors. The summative exchangeable concentration of the 4 metals in varimax factor 1 (SCM1) was significantly higher in the rainy season (269) than in the dry season (87 mg kg^{-1}) (Fig. 4a). The SCM1 was significantly higher in the upper part (254) than in the lower part (102, mg kg^{-1}) (Fig. 4b). The SCM1 tended to increase with sampling site, from site 1 toward site 13 in both seasons (Fig. 4c).

The exchangeable concentration of 5 metals (Cd, Co, Ni, Pb, and Zn) having high loading values with varimax factor 2 was summed up to examine the effect of experimental factors. The summative exchangeable concentration of the 5 metals (SCM2) was significantly higher in the canal position (55) than in the river position (41, mg kg^{-1}) (Fig. 5a). The SCM2 was significantly affected by the interaction of season and river part (Fig. 5b). In the rainy season, the upper part had a much significantly higher SCM2 (101) than the lower part (33 mg kg^{-1}). Meanwhile, in the dry season, the upper part had a significantly higher SCM2 (47) than the lower part (11 mg kg^{-1}). Spatial variation of the SCM2 in two river positions

Table 3

The Pearson correlation coefficients (r) of 11 measured metals and metalloid. (*) indicated the correlation coefficient was statistically significant.

Metal	Al	B	Cd	Co	Fe	In	Mn	Ni	Pb	Sr	Zn
Al	1.00										
B	-0.29*	1.00									
Cd	0.17	-0.19	1.00								
Co	0.79*	-0.43*	0.51*	1.00							
Fe	0.66*	-0.17	0.07	0.63*	1.00						
In	0.13	-0.01	0.15	0.22*	0.07	1.00					
Mn	0.10	0.00	0.14	0.20*	0.06	0.99*	1.00				
Ni	0.72*	-0.34*	0.58*	0.87*	0.48*	0.15	0.13	1.00			
Pb	0.24*	-0.21*	0.23*	0.33*	0.03	0.00	-0.02	0.31*	1.00		
Sr	-0.27*	0.46*	-0.08	-0.40*	-0.24*	0.56*	0.54*	-0.29*	-0.15	1.00	
Zn	0.55*	-0.32*	0.74*	0.73*	0.22*	-0.04	-0.06	0.86*	0.36*	-0.36*	1.00

(Fig. 5c) and in two seasons (Fig. 5d) was an increasing pattern from site 1 to site 13. Especially, from site 7 to site 13, SCM2 in the rainy season was much higher than that in the dry season.

Quite different from the above trend, the summative exchangeable concentration of 3 metals (In, Mn, and Sr) in factor 3 (SCM3) was a decreasing pattern from site 1 to site 13 (Fig. 6c). The SCM3 was significantly higher in the rainy season (158) than in the dry season (110 mg kg⁻¹) (Fig. 6a) and was significantly higher in the lower part (164) than in the upper part (104 mg kg⁻¹) (Fig. 6b) of the studied reaches of the Saigon River.

4. Discussion

An interesting finding from the current study was a general increase in the exchangeable concentration of 11 examined heavy metals and metalloid in surface sediment from site 1 located in the mouth of the Saigon River to site 13 located on the entrance of the studied reaches of the River. Nevertheless, some metals, including In, Mn, and Sr had exchangeable concentrations decreasing from the mouth (site 1) to the entrance (site 13) of the studied reaches. Rainy season increased the total exchangeable concentration of the 11 examined metals and metalloid in the surface sediment, relative to the dry season, but the effect of the rainy and dry season depended on the pollution sources (river part) that was in line with our initial hypothesis.

4.1. Seasonal variation of heavy metals in the sediment

Two distinct rainfall-based seasons in Vietnam were normally identified, including rainy season and dry season, which partly determine the hydrological regime of river systems. The rainy season may cause soil more eroded and thus bring eroded solid materials such as soil parent materials from the catchment to rivers and deposit them in the downstream of a river. Meanwhile, the dry season may cause river water lost through evaporation, increasing concentrations of suspended materials and dissolved elements including heavy metals. In general, the current study found that rainy season had significantly higher total exchangeable

concentration of 11 examined metals and metalloid (TCM) in sediment than the dry season had. This indicated that water erosion and runoff of heavy metals-containing materials from the upper catchment and industrial and municipal wastes from residential and industrial centers surrounding the Saigon River in Ho Chi Minh City could happen predominance. This finding was similar to the other studies, such as that of Ramalingam et al. (2004). The authors attributed the higher concentration of some metals such as Hg, Cd, Cu, and Zn during the monsoon than the summer season to higher river runoff due to greater rainfall to transport and deposit the heavy metal-containing materials such as industrial, municipal, and agricultural wastes and soil material to the studied sites. Similarly, Kumar Gaur et al. (2005) concluded that higher concentration of Cd, Cr, Cu, Ni, Pb, and Zn of sediment collected from Gomti river in the rainy season than in the other seasons could be due to greater runoff of untreated industrial, agricultural, domestic wastes.

Nevertheless, the current study found that B and Sr had higher exchangeable concentrations in the dry season than in the rainy season. Coulibaly et al. (2012) found that the concentration of some metals such as Hg, Cd, Pb, and Zn of sediment collected from Bie'tri Bay in Ebrie' Lagoon, Ivory Coast was higher in the dry season than in the rainy season, which was attributed to "reduced water volume during the dry season". Sarasiab et al. (2014) reported that the concentrations of some metals such as Ni, Cu, Co, Pb, Cd, and Hg were higher in the summer season the others, and the authors attributed that to the variation in temperature, salinity, pH and summer discharge.

The concentration of the exchangeable metals and metalloid in the current study could additionally be affected by sediment pH. We additionally did a correlation analysis between sediment pH and the concentration of all exchangeable metals and metalloid and the result showed that pH was significantly and positively correlated with B and Sr, while negatively correlated with Al, Cd, Co, Fe, Ni, Pb, and Zn. In general, it was reported that the increased concentration of exchangeable heavy metals such as Al, Fe, Mn, and Zn could be observed when pH below 7 (Brady and Weil, 2017). Sediment pH of the current study varied from 3.2 to 6.2, with a mean of 4.8, which could be considered as acidic condition, resulting in higher exchangeable concentrations of the 7 metals. The decreased exchangeable concentration of 7 metals in sediment upon increased sediment pH in the current study could be due to metal retention to sediment surface as a result of adsorption, surface complexation, and precipitation, the important natural processes strengthened on the alkaline pH direction (Orhue and Frank, 2011).

4.2. Spatial variation of heavy metals in sediment

Two contrastive patterns of spatial distribution of 11 metals and

Table 4

Percentage of individual varimax factors (VF) from PCA/FA in explaining the total variance of TCM (total exchangeable concentration of 11 examined metals and metalloid). Note: stepwise elimination was applied first and the multiple regression model describing the dependency of HPI and TCM on three VFs was finalized.

Source	Sum of Squares	Percentage	Important parameters
Factor 1	5735157	83.2	Al, Co, Fe, Ni
Factor 2	344390	5.0	Cd, Co, Ni, Pb, Zn
Factor 3	537426	7.8	In, Mn, Sr
Error	278490	4.0	
Total variance	6895462	100.0	

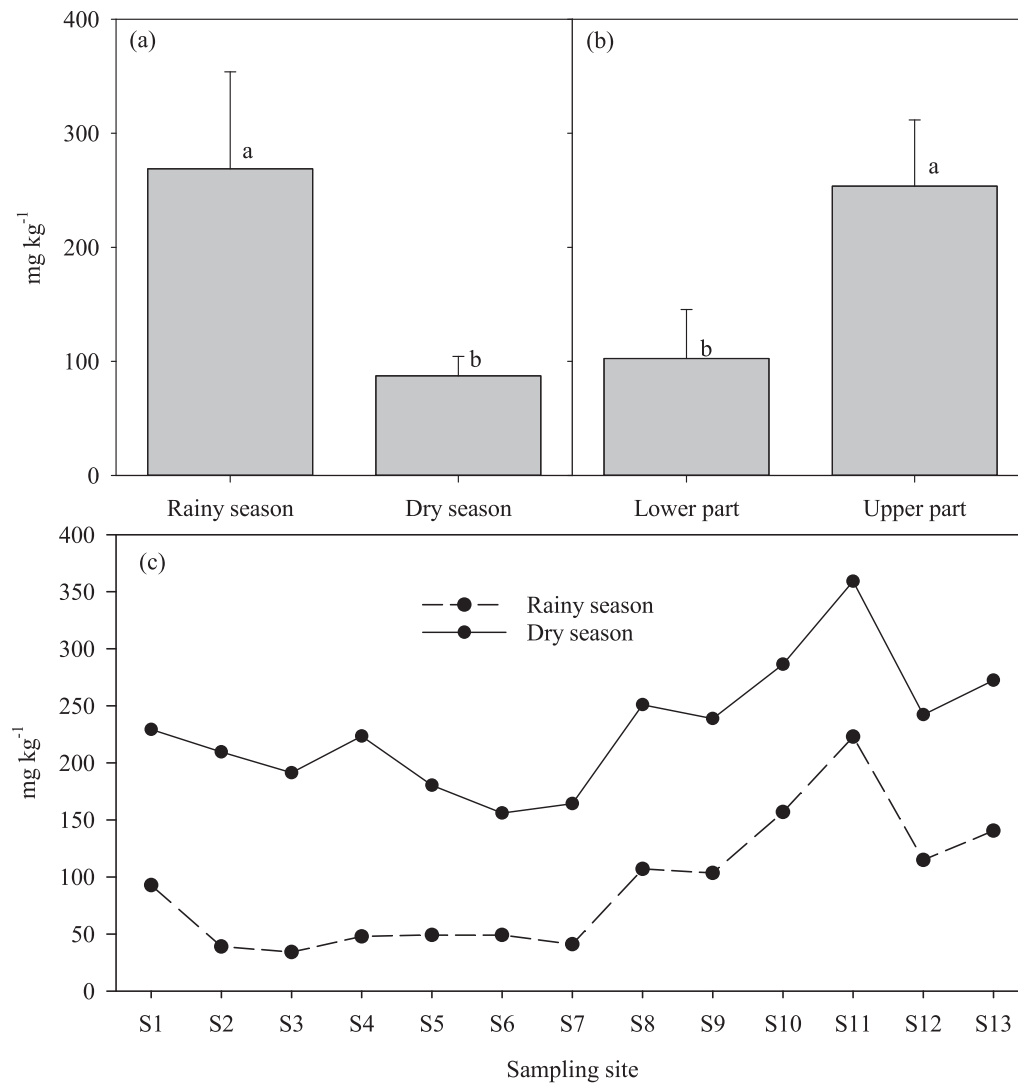


Fig. 4. The summative exchangeable concentration of 4 metals having high loading values within varimax factor 1 from PCA/FA as affected by season (a) and river part (b) as well as varying with sampling site (c). Within a panel a and b, bars attached with the same letter are not significantly different from the other. Error bars indicate standard errors.

metalloid in sediment were an increasing trend in exchangeable concentration of Al, Co, Fe, Ni, (varimax factor 1) and Cd, Co, Ni, Pb, and Zn (varimax factor 2) and a decreasing trend in exchangeable concentration of In, Mn, and Sr (varimax factor 3) with sampling sites, from site 1 (located in the mouth of the studied river) to the site 13 (the entrance of the studied reaches of the river) (Figs. 4c, 5c and 5d, and 6c). This is an interesting and important finding from the current study. With such the spatial distributions of measured metals, hierarchical cluster analysis separated the 13 sites into two main clusters of lower and upper parts of the studied river (Fig. 2). The above discussion indicated that transport of eroded materials and or dissolved elements including measured metals from the river catchment and deposition of these materials on the Saigon River was one of the main mechanisms happening strongly during the rainy season. The deposition of the transported materials through riverine water could happen more strongly on the upper part than the lower part, resulting in different heavy metal concentration in sediment between the two parts. Nevertheless, some metals such as In, Mn, and Sr had significantly higher exchangeable

concentration in the lower part than in the upper part, or a decreasing pattern from upstream to downstream. Possibly the main sources of these metals could be derived from inside Ho Chi Minh City or from the area outside the estuary, which would be discussed in the following section. Hoang et al. (2007) found that sediment collected from canals located inside Ho Chi Minh City was higher in the concentration of heavy metals such as Cd, Cr, Cu and Zn than that collected on the Saigon River. The current study found that of the 11 measured metals and metalloid, the summative exchangeable concentration of Cd, Co, Ni, Pb, and Zn (SCM2, Fig. 5a) was significantly higher on the canal position (located at the mouth of canals originated from inside Ho Chi Minh City) than on the river position.

4.3. Pollution sources of the examined metals

Principal component analysis/factor analysis showed that there could be three main sources enriching the heavy metals in sediment of the Saigon River. Significantly higher SCM1 in the upper

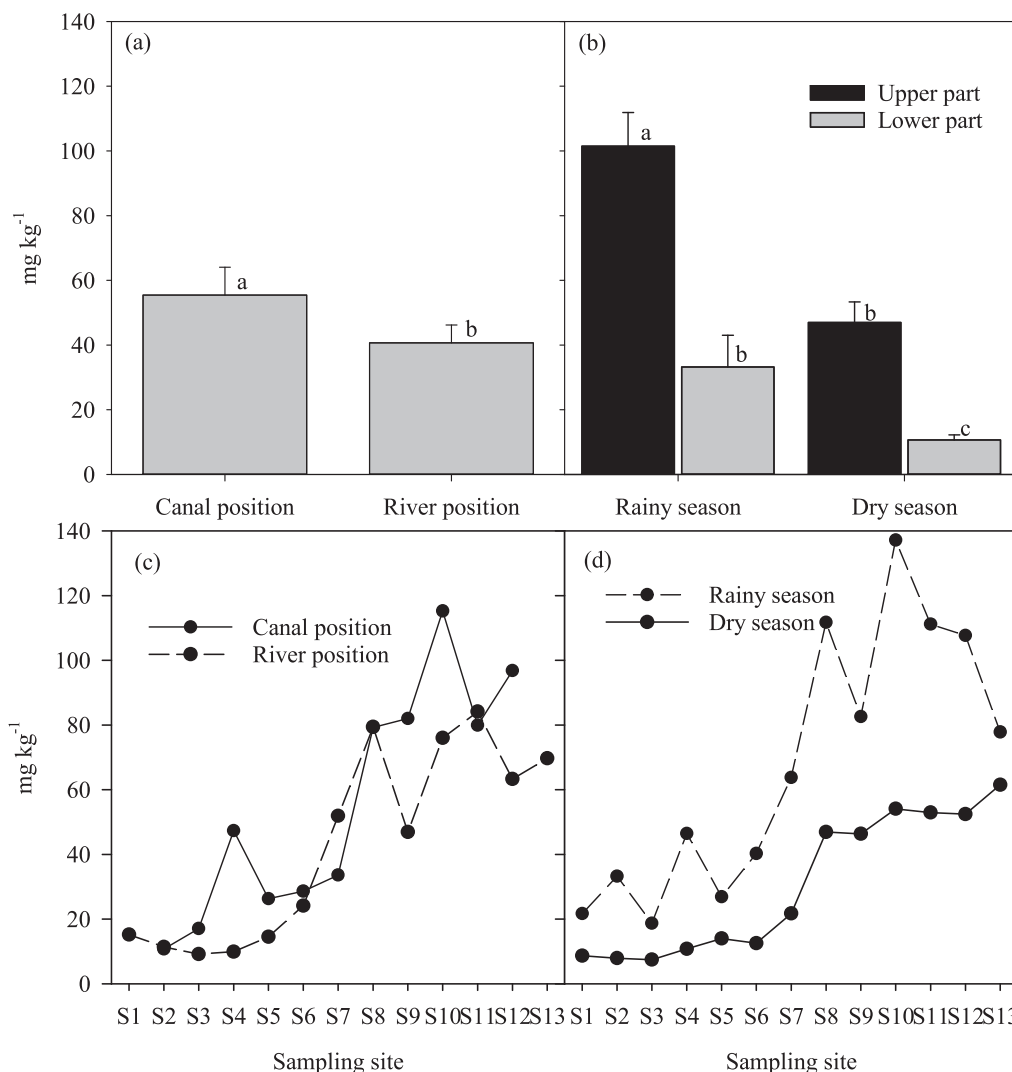


Fig. 5. The summative exchangeable concentration of 5 metals having high loading values within varimax factor 2 from PCA/FA as affected by river position (a), the interaction of season and river part (b), and varying with sampling site (c and d). Within a panel a and b, bars attached with the same letter are not significantly different from the other. Error bars indicate standard errors.

part than the lower part, and in the rainy season than in the dry season (Fig. 4a and b) indicated that the first and most important source for Al, Co, Fe, and Ni in sediment could come from the river catchment through erosion/runoff of natural soil parent materials. Quinton and Catt (2007) demonstrated that water erosion over the agricultural fields increased the metal concentration of sediment of the receiving river. This is because water erosion is a selective process, preferentially detaching and transporting clay and silt (Quinton et al., 2001), which was regularly bound with heavy metals. The metal concentration of the catchment soil over agricultural and natural fields could be derived from various sources, including original soil parent materials, and agronomic and atmospheric additions (Atafar et al., 2009; Alkhader, 2015). The Saigon River has a large catchment area of about 4717 km² (Lahens et al., 2018), spreading over three agriculture-based provinces, Binh Duong, Binh Phuoc, and Tay Ninh. Two main soil types of the three provinces are Ferralsols and Acrisols, which are characterized with low pH (Pham, 2010). These are highly weathered acid upland soils with high concentrations of Al and Fe (Rahman et al., 2018).

Because the Saigon river catchment is located on a hilly and sloppy area with high rainfall annually, water erosion over the catchment may bring a great amount of the Al and Fe-rich soil materials to the Saigon River, enriching these two metals in the upper part of the studied river. In addition, there were many industrial parks, residential villages located within the Saigon River catchment that could add wastes to the River, contributing to this pollution source.

The second most important source for Cd, Co, Ni, Pb, and Zn in the sediment of the Saigon River identified through PCA/FA (Table 2) could come from (1) the river catchment through agricultural fields and (2) associated areas inside Ho Chi Minh City through runoff of municipal, agricultural, and industrial wastes. Similar origins of these metals could additionally be confirmed through correlation analysis with significant Pearson correlation coefficients among these elements (Table 3). Phosphorous (P) fertilizers used on agricultural fields for better crop production were a major source of heavy metals, such as Cd, Ni, Pb, and Zn (O'Geen et al., 2010). In particular, because the soil in catchment area of the Saigon River was characterized with low pH (Pham,

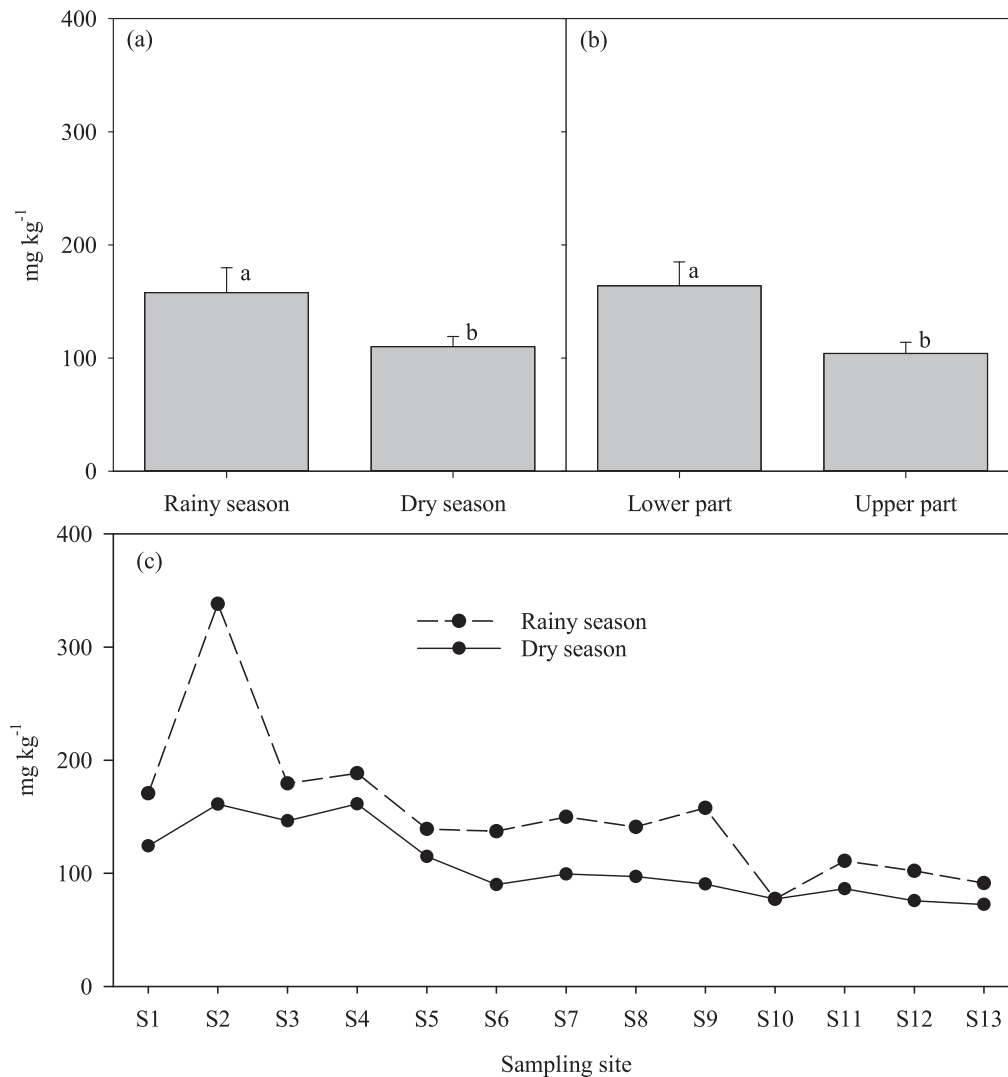


Fig. 6. The summative exchangeable concentration of 3 metals having high loading values within varimax factor 3 from PCA/FA as affected by season (a) and river part (b) as well as varying with sampling site (c). Within a panel a and b, bars attached with the same letter are not significantly different from the other. Error bars indicate standard errors.

2010), resulting in a low concentration of available P due to fixation (Penn and Camberato, 2019), more addition of P fertilizers could be expected (Cong, 2017), resulting in more accumulation of these metals in the catchment soils and subsequently in the sediment of the Saigon River due to selective process of water erosion (Quinton et al., 2001). In addition, significantly higher SCM2 in the canal position than in the river position (Fig. 5a) may suggest that additional source of Cd, Co, Ni, Pb, and Zn could come from inside Ho Chi Minh City through runoff of municipal, agricultural, and industrial wastes. These wastes contained a large amount of metals, including Cd, Co, Ni, Pb, and Zn (Barakat, 2011; Wang et al., 2005). Likewise, anthropogenic discharge of uncontrolled and untreated industrial runoff was assumed to add metals to the sediment of the studied canals (Hoang et al., 2007). Similarly, Phuong et al. (1998) found that the concentration of some heavy metals such as Cr, Cu, Pb, and Zn was higher in the inside area than in the outside area of Ho Chi Minh city. This indicated that one possibly main source of these metals could be derived from the activities within Ho Chi Minh City, which released solid and liquid wastes into the studied river and

canals.

The last important source of metals, including In, Mn, and Sr, identified with PCA/FA could be derived from either inside Ho Chi Minh City or the lowland surrounding the confluence of the Saigon River and the Dong Nai River. As these metals had significant relationships with each other (Table 3), they could originate from similar sources. The significantly higher SCM3 in the lower part than in the upper part of the Saigon River (Fig. 6b) may suggest that the source could be derived from the lower reaches of the studied river. Although specific sources of these elements was unclear, some possible sources such as mangrove forest soil (Almahasheer et al., 2018), lowland paddy fields (Nguyen et al., 2019), and some municipal and industrial wastes (Seyoum and Adeleju, 2007) could contribute to more enriching these metals in the lower part than the upper part of the Saigon River.

Comparative analysis of 13 sampling sites showed that sites 11 and 9 were more contaminated with total heavy metals (690 and 453 mg kg⁻¹, respectively) than the others and sites 5 and 6 was lowest in TCM (222 and 233 mg kg⁻¹, respectively). The

proportional analysis showed that Al and Fe were the two metals occupying the largest proportion of TCM of site 11 (41 and 32%) and site 9 (30 and 27%, respectively). Different from sites 11 and 9, site 5 had the highest percentage of Mn and Al (47 and 21%, respectively) and site 6 had highest percentages (37 and 21%) for Mn and Al, respectively. Because Mn concentration was higher in the sites 1 and 2 located at the mouth of the studied river, these sites were mostly contaminated with Mn, and thus were not in the lowest position, compared to the other sites. These indicated that the studied river was mostly contaminated with Al and Fe in the upper part, while with Mn and Al in the lower part.

Because Al and Fe contributed the greatest proportion of total concentration of 11 measured metals and metalloid, the most important main source for heavy metals (Factor 1) explained as high as 83% of the total variance of 11 heavy metals and metalloid in the sediment of the Saigon River (Table 4). Water erosion from natural fields and runoff of municipal and industrial wastes from the river catchment contributed a large percentage to contaminating sediment of the lower reaches of the Saigon River with Al, Fe, Co, and Ni. The second most important sources of metals in the sediment could come from the river catchment and from inside Ho Chi Minh City through runoff of municipal, agricultural, and industrial wastes. The two sources together explained around 5% of the total variance of the total metal concentration. The last sources explaining 7.8% were still unknown, but possible could come from various origins such as mangrove soil, lowland paddy soil, and industrial and municipal wastes.

5. Conclusions

The total exchangeable concentration of heavy metals and metalloid (TCM) in the sediment collected from the Saigon River was significantly higher in the rainy season than in the dry season and in the upper part than in the lower part. Principal component analysis/factor analysis revealed that three most important pollution sources may contribute to enriching the 11 examined metals and metalloid in the sediment of the studied river, including (pollution source 1) water erosion of natural land and runoff of municipal and industrial wastes over the river catchment, (pollution source 2) mixed sources of agricultural activities from catchment and runoff of agricultural, industrial, and domestic wastes from inside Ho Chi Minh City, and (pollution source 3) mixed sources of lowland areas. The metals, having high relationship coefficient with the pollution sources 1 and 2, including Al, Co, Fe, and Ni, Cd, Co, Ni, Pb, and Zn tended to decrease in summative exchangeable concentration (SCM) from the upstream to downstream of the River. Meantime, other metals such as Mn, Sr, and In having high loading values with pollution source 3 tended to increase in SCM from upstream to downstream. Together, three pollution sources could explain 96% of the total variance of total exchangeable concentration of 11 examined metals and metalloid in the sediment of the Saigon River.

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