HEAVY METAL(LOID)S IN THE SURFACE SEDIMENT IN COASTAL AREAS OF SOUTH VIET NAM (2016-2021)

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Abstract. The coastal areas' sediments are basins of metal deposition from natural and anthropogenic sources. In this study, sediment samples were collected at 07 sites, to estimate spatial variation, and the contamination levels of heavy metal(loid)s (As, Cd, Cu, Cr, Pb, Zn), as well as to assess the ecological risks. Contents of Zn, Cr, Cu, Pb and As in sediments have ranges of 16.0 - 69.7 (52.1 ± 19.6), 11.9 - 48.1 (34.6 ± 11.5), 7.3 - 29.4 (21.0 ± 8.1), 6.9 - 24.1 (19.4 ± 6.4), and 3.2 - 5.4 (4.0 ± 0.7) µg/g. The lowest contents of 05 heavy metals were found in Phan Thiet site. Meanwhile, the highest levels of 03 metals (Cr, Cu, Zn) occurred in Ganh Rai. Besides, As and Pb had the highest values in Dinh An and Rach Gia sites, respectively. The contents of As, Cd, and Zn were lower than threshold effect levels (TEL) in Canadian Sediment Quality Guidelines for the Protection of Aquatic Life, while contents of Cr, Cu, and Pb were exceeded by 1.1 - 1.2 times, 1.0 - 2.5 times, and 1.0 - 1.3 times, respectively. The contamination factor (CF), and the Geo-accumulation index (I_{geo}) were computed to evaluate the pollution degrees of heavy metals in sediments. Besides, the ecological risk factor (E_r), and the potential ecological risk index (RI) were applied to assess the ecological risk of heavy metals. The values of contamination factor (CF) showed that As and Cd existed in the surface sediments at moderate or considerable pollution levels, while Cr, Cu, Pb, and Zn occurred at low or moderate pollution grades. In addition, the spatial distribution of CF values for 05 heavy metals showed that the heavy metal contamination of Dinh An and Ganh Rai sites was greater than other sites. According to the ecological risk factor (E_r) and Geo-accumulation index (I_{geo}), Cd pollution grade in sediments was at zero or moderate levels, with moderate to appreciable potential ecological risks. The potential ecological risk index (RI) values proposed that moderate ecological risk were presented at Nha Trang, Dinh An, Ganh Rai, and Rach Gia sites.

Keywords: Heavy metals, sediments, contamination degrees, ecological risk, South Vietnam.

1. Introduction

Heavy metals are serious environmental contaminants, because of their persistence, accumulation, and toxicity in aquatic ecosystems. They occur widely in soils, water, and sediments. Heavy metals are entered into the environment from natural and anthropogenic sources, such as soil erosion, industrial discharges, agricultural activities, and urban sewage. Heavy metals tend to accumulate in the surface sediments through adsorption and precipitation (Xiao et al., 2019). Therefore, sediments are considered the final sink of heavy metals. However, heavy metals in sediments might be also released into the overlying water, depending on the changes in environmental conditions such as pH,

oxidation potential (Eh), and salinity, posing potential ecological risks. For that reason, heavy metal levels in sediments describe the pollution status of the nearby areas, as well as evaluate the potential ecological risks. Various indexes have been applied to assess the pollution levels, and the potential ecological risks in sediment (Niu et al., 2020), such as contamination factor (CF), geo-accumulation index (I_{geo}), and the ecological risk index (E_r), the potential ecological risk index (RI). In the present paper, these indexes are computed and combined to bring the scientifical information.

The Vietnam coast has a rich ecosystem, such as coral reefs, mangroves, sand dunes, seagrass beds, estuaries, and coastal forests (Sekhar, 2005). For this reason, these areas might create high economic values, related to tourism and fisheries. The rapid growth of tourism is associated with the increase of urbanization in the coastal areas. The coastal population might constitute 35 % of the total population of Vietnam (Sekhar, 2005). It may cause pressure on the coastal environment, such as organic compounds, nutrients, and heavy metals pollution. In this studied area, Nha Trang, Phan Thiet, Ha Tien, and Song Doc sites are affected by coastal urbanization, port activities, and tourism. Besides, Ganh Rai site is mainly influenced by industrialization. Meanwhile, Dinh An site is represented by the Mekong river effects. In Vietnam, the heavy metal pollution in sediment has been concerned in previous studies (Trinh et al., 2021; Le et al., 2019; Nguyen et al., 2016; Ho et al., 2010; Ngo et al., 2009). However, these studies focused on heavy metal pollution on a local scale. In this present study, the heavy metal levels in sediment on a large scale - coastal southern Vietnam were provided, as well as examined the ecological risk.

The aims of this study were: (1) to determine spatial variations of As, Cd, Cr, Cu, Pb, and Zn in sediments; (2) to assess contamination status and potential environmental risks of sediments.

2. Materials and methods

2.1. Study area, sampling, and analysis

In the present study, 7 sites in coastal waters of south-central Vietnam were observed, 76 sediment samples were collected in two seasons during 2016 - 2021. Trace element levels (As, Cr, Cd, Cu, Pb, Zn) in sediment samples were measured. In this work, the sampling sites are located in South Central Vietnam, including the coastal bays (Nha Trang, Phan Thiet, Ganh Rai, Rach Gia) and the estuarine areas (Dinh An, Song Doc, Ha Tien) (Figure 1).

Sediment samples were collected by the grab sampler, contained in the zip bags, preserved in 4 ^{o}C for later analysis. Sediment samples were dried in an oven, and dissolved in the mixture of HNO_3 and HCl, with a volumetric ratio of 1:3. These mixtures were placed in the Teflon tubes, and digested with a microwave digestion (Mars 6). The solutions of digestion were analysed for heavy metal contents.

Figure 1. Map shows the studied areas

2.2. Contamination evaluation

2.2.1. Contamination factor (CF) and degree of contamination (CD)

The contamination factor (CF) represent the pollution level of an observed metalin sediments. CF is obtained by dividing the individual heavy metal concentration in samples (C_{sample}) by the background concentrations ($C_{background}$). The background concentrations of As, Cd, Cr, Cu, Pb, and Zn were 2, 0.102, 35, 14.3, 17, and 52 µg/g (Hakanson, 1980). The CF can be classified into four levels: CF < 1 represents low pollution, 1 ≤ CF < 3 represents moderate pollution, 3 ≤ CF < 6 represents considerable pollution, and CF ≥ 6 represents very high pollution.

$$CF = \frac{Csample}{Cbackground}$$

The contamination degree (C_d) was computed to identify the pollution level of samples from each site. The pollution levels were classified six classes, according to the values of C_d: C_d < 6, low; 6 < C_d < 12, moderate; 12 < C_d < 24, considerable; C_d > 24, very high contamination (Hakanson, 1980).

$$C_d = \sum_{i=1}^{n} CFi$$

2.2.2. Geo-accumulation index (I_{geo})

The geo-accumulation index (I_{geo}) was applied to evaluate the contamination levels of heavy metals in sediments.

$$I_{geo} = \log_2 \frac{Csample}{1.5\ x\ Cbackground}$$

The heavy metal pollution levels can be interpreted as follows: $I_{geo} \leq 0$, uncontaminated; $0 < I_{geo} \leq 1$, uncontaminated to moderately contaminated; $1 < I_{geo} \leq 2$, moderately contaminated; $2 < I_{geo} \leq 3$, moderately to heavily contaminated; $3 < I_{geo} \leq 4$, heavily contaminated; $4 < I_{geo} \leq 5$, heavily to extremely contaminated; $I_{geo} \geq 5$, extremely contaminated (Muller, 1969).

2.3. Potential ecological risk index

The potential ecological risk index (RI) was introduced by Hakanson (1980). The ecological risk factor of an observed metal (E_r) is defined as: $E_r = T_r \times CF$. E_r and T_r are the ecological risk index and the toxicity coefficient of heavy metal, respectively. The toxicity coefficients of As, Cd, Cr, Cu, Pb, and Zn are 10, 30, 2, 5, 5, and 1, respectively (Hakanson, 1980). According to E_r values, the potential ecological risk factor of a target metal is categorized as follows: $E_r < 40$, low; $40 < E_r < 80$, moderate; $80 < E_r < 160$, considerable; $160 < E_r < 320$, high; $E_r \geq 320$, very high risk.

The risk index of sammpling sites as follows: $RI = \sum_{n}^{i=1} E_r^i$. The risk index classified as follows: $RI < 150$, low; $150 \leq RI \leq 300$, moderate; $300 \leq RI \leq 600$, considerable; $E_r \geq 600$, high risk.

2.4. Data analysis

Spatial differences of heavy metal concentration were detected with one-way ANOVA. Pearson correlation analysis were used to explore the correlations between metals. The significant level was set at $p < 0.05$. The statistical analysis were performed with Minitab 18.

3. Results and discussions

3.1. Contents of heavy metals in surface sediments

Heavy metal contents in sediments were showed in Table 1. Element contents in sediments were followed the order: Zn > Cr > Cu > Pb > As > Cd. Contents of Zn, Cr, Cu, Pb, As and Cd in sediments have ranges of 8.9 - 89.1 (51.2 ± 20.3), 6.2 - 64.4 (34.6 ± 13.4), 4.1 - 46.9 (21.3 ± 9.6), 5.2 - 39.1 (19.0 ± 7.3), 1.6 - 7.9 (4.0 ± 1.3), and 0.1 - 2.0 (0.4 ± 0.3) µg/g. The significant spatial variations of heavy metal contents were exhibited. All of elements had lowest values in Phan Thiet bay. The highest values of 03 metals (Cr, Cu, Zn) occurred in Ganh Rai bay. Meanwhile, As and Pb had highest values in Dinh An estuary and Rach Gia bay, respectively. According the results, the heavy metal contents in sediments in this study were lower the limits of Vietnam National Technical Regulation on Sediment Quality (QCVN 43:2017/BTNMT). However, the contents of Cr, Cu and Pb were exceeded the threshold effect levels (TEL) in Canadian Sediment Quality Guidelines for the Protection of Aquatic Life, by 1.1 - 1.2 times, 1.0 - 2.5 times, and 1.0 - 1.3 times, respectively.

Table 1. Descriptive statistics for the heavy metal concentrations in sediments (μg/g) of monitoring sites. S.D: standard deviation

Elements	Monitoring sites	Means	Ranges	S.D
As	Nha Trang	3.9	1.7 - 6.4	1.4
	Phan Thiet	3.1	1.6 - 6.8	1.4
	Ganh Rai	4.3	2.1 - 6.4	1.3
	Dinh An	5.3	4.0 - 6.6	0.8
	Rach Gia	4.2	2.6 - 7.9	1.3
	Song Doc	3.8	3.1 - 4.6	0.5
	Ha Tien	3.4	2.4 - 4.5	0.7
Cd	Nha Trang	0.6	0.2 - 1.4	0.4
	Phan Thiet	0.3	0.1 - 0.7	0.2
	Ganh Rai	0.5	0.2 - 2.0	0.5
	Dinh An	0.5	0.1 - 1.2	0.3
	Rach Gia	0.4	0.1 - 1.3	0.3
	Song Doc	0.2	0.2 - 0.3	0.04
	Ha Tien	0.2	0.1 - 0.5	0.1
Cr	Nha Trang	41.5	23.2 - 55.8	10.9
	Phan Thiet	12.5	6.2 - 21.4	5.2
	Ganh Rai	47.8	36.5 - 64.4	8.6
	Dinh An	38.9	23.7 - 54.3	9.0
	Rach Gia	36.4	26.3 - 48.3	6.7
	Song Doc	33.2	22.9 - 39.4	6.4
	Ha Tien	29.4	18.3 - 38.3	7.2
Cu	Nha Trang	26.0	15.7 - 35.8	6.8
	Phan Thiet	7.2	4.1 - 12.5	2.7
	Ganh Rai	29.2	20.3 - 43.1	7.2
	Dinh An	26.3	19.5 - 46.9	7.3
	Rach Gia	24.4	16.1 - 39.6	7.1
	Song Doc	19.1	14.2 - 31.9	5.8
	Ha Tien	13.3	10.3 - 15.8	2.2
Pb	Nha Trang	22.7	16.7 - 39.1	6.7
	Phan Thiet	6.8	5.2 - 8.8	1.1
	Ganh Rai	22.7	16.2 - 31.1	4.9
	Dinh An	22.0	17.4 - 31.1	4.1
	Rach Gia	23.2	14.7 - 28.4	4.2
	Song Doc	19.7	14.2 - 27.6	4.4
	Ha Tien	14.4	10.4 - 17.2	2.2
Zn	Nha Trang	65.3	45.6 - 89.1	14.1
	Phan Thiet	16.0	8.9 - 21.0	3.0
	Ganh Rai	68.8	58.1 - 88.7	9.9
	Dinh An	60.9	50.9 - 75.6	8.2
	Rach Gia	54.7	30.9 - 74.1	12.6
	Song Doc	52.5	41.2 - 63.1	7.6
	Ha Tien	34.8	30.5 - 44.5	5.2

Most of element contents in sediments of studied areas were lower than those of the other regions (Table 2). All of element contents in sediments in this study were less than in the subaqueous delta of Red River, Tra Vinh coast and Yangtze estuary. However, Cu contents were greater in comparison with sediments of North Central coast and Tra Vinh coast. In general, the contamination level of heavy metals in sediments in the studied areas were less than the other regions.

Table 2. Heavy metal contents (μg/g) in sediments in period 2016-2021 in comparison with those in some the other regions and the threshold levels

Regions	As	Cr	Cu	Pb	Zn	References
Red River, Vietnam	-	59.9	26	32	64	(Nguyen et al., 2016)
Duyen Hai port, Tra Vinh coast, Vietnam	18.5 (16.5-22.8)	-	5.1 (3.1-9.2)	72.6 (65.3-85.2)	149 (82.9-212)	(Tham et al., 2021)
Yangtze estuary	8.6 (4.5-12.7)	-	22.8 (10.6-34.9)	63.2 (17.4-109)	82.5 (57.9-107)	(Zhao et al., 2012)
South Central coastal areas of Vietnam	4.1 (1.6-7.9)	34.9 (6.2-64.4)	21.6 (4.1-46.9)	19.6 (5.2-39.1)	53.0 (8.9-89.1)	This paper
% values exceeded TEL	0%	5%	60%	6%	0%	
TEL[1]	7.24	52.3	18.7	30.2	124	
QCVN 43:2017[2]	41.6	160	108	112	271	

[1]Canadian Sediment Quality Guidelines.
[2]Vietnam National Technical Regulation on Sediment Quality (QCVN 43:2017/BTNMT).

3.2. Pollution assessment of heavy metals

The mean values of element contents in sediments in period 2016 - 2021 were used to calculated CFs (Table 3). The contamination factor (CF) is computed to evaluate the contamination status of single metals in sediments. The average of CF values of all monitored sites was Cd (4.05) > As (2.02) > Cu (1.47)> Pb (1.14) > Zn (1.00) > Cr (0.99). According to results, the average CF values were highest for Cd, and lowest for Cr. Based on the classification of CF, the monitoring sites has considerable level for Cd, moderate for As, Cu, Pb, Zn, and low for Cr. The results showed that Nha Trang, Dinh An, Ganh Rai, and Rach Gia sites were considerable Cd pollution, with highest value in Nha Trang. As, Cu, Pb, Zn contents in sediments exhibited as moderate contamination, due to average CF values ranged from 1.00 to 2.02. Meanwhile, As contents in sediments of 07 sites presented as moderate pollution. For Cu, Pb, Zn, CF values of Phan Thiet and Ha Tien showed as low contamination. For Cr, CF values of Nha Trang, Dinh An, Ganh Rai, Rach Gia sites were higher than 1, as moderate pollution.

The estimated CD values of the monitoring sites were given in Table 3 by summing the CF values, ranged from 6.09 to 13.84. According to the categorise of CD values, it indicated that Nha Trang, Dinh An, and Ganh Rai were a considerable degree of contamination. Meanwhile, the other sites were a moderate level of contamination. The maximum of contamination degree was found in Ganh Rai. Additionally, this site was recorded the highest CF values for Cd, Cr, Cu, and Zn. In general, CF and CD values displayed that the high accumulation of Cd in sediments may give the considerable contaminated degree for Nha Trang, Dinh An, Ganh Rai sites.

Table 3. Contamination factor of individual heavy metal and contamination degree for multi-metals

Locations	CF values of each heavy metal						CD
	As	Cd	Cr	Cu	Pb	Zn	
Nha Trang	1.97	5.98	1.16	1.84	1.40	1.33	13.67
Phan Thiet	1.58	2.94	0.34	0.51	0.41	0.31	6.09
Dinh An	2.70	5.59	1.15	1.87	1.34	1.20	13.83
Ganh Rai	2.15	5.59	1.37	2.06	1.33	1.34	13.84
Rach Gia	2.12	3.82	1.06	1.69	1.42	1.10	11.21
Song Doc	1.90	2.25	0.97	1.36	1.25	1.06	8.80
Ha Tien	1.72	2.16	0.86	0.93	0.85	0.68	7.20

3.3. Ecological risk of heavy metals

For the environmental assessment, the geo-accumulation index (I_{geo}) was computed and given in Table 4. The observed I_{geo} for metals in sediments were (0.08 ~ 0.85) for As, (0.52 ~ 2.00) for Cd, (-2.1 ~ -0.1) for Cr, (-1.6 ~ 0.5) for Cu, (-1.9 ~ -0.1) for Pb, (-2.3 ~ -0.2) for Zn. Based on the average I_{geo} values, the pollution grades of metals in sediments was as follows: Cd (1.32) > As (0.41) > Cu (-0.16)> Pb (-0.50) > Cr (-0.71) ~ Zn (-0.72). Among the observed metals, only two metals As and Cd had average $I_{geo} > 0$, categorized as uncontaminated to moderately contamination. Meanwhile, the negative I_{geo} values of Cr, Cu, Pb, Zn suggested that the monitoring sites were not polluted by these metals.

Table 4. The estimated I_{geo} of heavy metals in sediments

Locations	I_{geo}					
	As	**Cd**	**Cr**	**Cu**	**Pb**	**Zn**
Nha Trang	0.39	2.00	-0.38	0.29	-0.10	-0.18
Phan Thiet	0.08	0.97	-2.14	-1.55	-1.89	-2.29
Dinh An	0.85	1.90	-0.39	0.32	-0.17	-0.33
Ganh Rai	0.52	1.90	-0.13	0.46	-0.17	-0.16
Rach Gia	0.50	1.35	-0.50	0.17	-0.08	-0.45
Song Doc	0.34	0.59	-0.63	-0.14	-0.26	-0.49
Ha Tien	0.19	0.52	-0.80	-0.69	-0.82	-1.13

The RI and Er values for the heavy metals in sediments are shown in Table 5. The calculated Er values for As, Cr, Cu, Pb, Zn were less than 40, meaning that the risk posed

by these metals was low. Meanwhile, the average Er for Cd was 121.43 (64.71 - 179.41), indicating that Cd was the main contaminant, posed the considerable ecological risk. Moreover, The Er indices of Cd in Nha Trang, Dinh An, Ganh Rai were relatively high (> 160), indicating that Cd might pose a high ecological risk. Overall, according to the average Er values, the potential risk in sediments followed an order of Cd (121.43) > As (20.19) > Cu (7.33) > Pb (5.71) > Cr (1.97) > Zn (1.00).

The RI index presents the potential ecological risk posed by contaminants. Sediments of Phan Thiet, Song Doc, Ha Tien sites caused a low ecological risk (RI < 150). Meanwhile, RI values indicated a moderately polluted state in Nha Trang, Dinh An, Ganh Rai, Rach Gia (150 < RI < 300). Besides, these sites also contaminated As and Cd, so that, the As and Cd contamination in sediments might cause the potential ecological risk, and should be considered in the environmental management policy for these sites.

Table 5. The estimated Er of heavy metals in sediments

	Er values of each heavy metal						RI
	As	Cd	Cr	Cu	Pb	Zn	
Nha Trang	19.70	179.41	2.31	9.20	6.99	1.33	218.93
Phan Thiet	15.83	88.24	0.68	2.56	2.03	0.31	109.63
Dinh An	26.96	167.65	2.29	9.36	6.68	1.20	214.13
Ganh Rai	21.46	167.65	2.75	10.30	6.67	1.34	210.16
Rach Gia	21.24	114.71	2.12	8.45	7.08	1.10	154.69
Song Doc	19.00	67.65	1.94	6.80	6.25	1.06	102.71
Ha Tien	17.17	64.71	1.73	4.63	4.25	0.68	93.17

Conclusions

This study estimated levels of six heavy metals (As, Cd, Cr, Cu, Pb, Zn) in sediments at seven monitoring sites, southern Vietnam. The results proposed that Zn content was highest, while Cd content was lowest level. In comparision with other areas, the heavy metal contamination in sediments of the monitoring sites were less than the other regions. According to CD values, heavy metal contamination in sediments was occurred in Nha Trang, Dinh An, Ganh Rai, and moderately level of contamination was existed in Phan Thiet, Rach Gia, Song Doc, and Ha Tien. The measurement of I_{geo} indicated that the heavy metal pollutions were mainly caused by As and Cd. The Er results showed that the potential ecological risk of Cd was higher than the other metals. Based on the estimated RI, Nha Trang, Dinh An, Ganh Rai, and Rach Gia demonstrated higher potential ecological risks than the other sites. This study might provide the scientific information for the heavy metal contamination levels in sediments and the potential ecological risks.

Acknowledgments: This work was supported by the National Monitoring Program. This paper is a devotement to celebrate the 100th anniversary of the Institute of Oceanography, Vietnam Academy of Science and Technology.

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HÀM LƯỢNG KIM LOẠI NẶNG TRONG TRẦM TÍCH BỀ MẶT VÙNG BIỂN VEN BỜ NAM VIỆT NAM (2016-2021)

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Tóm tắt. Trầm tích tại các khu vực ven biển là nơi lắng đọng kim loại được phát tán từ các nguồn tự nhiên và hoạt động dân sinh. Trong nghiên cứu này, các mẫu trầm tích được thu thập tại 07 trạm, để đánh giá phân bố không gian, mức độ ô nhiễm của kim loại nặng (As, Cd, Cu, Cr, Pb, Zn), cũng như để xem xét khả năng ảnh hưởng đối với hệ sinh thái. Hàm lượng Zn, Cr, Cu, Pb và As trong trầm tích dao động trong khoảng 16,0 - 69,7 (52,1 ± 19,6), 11,9 - 48,1 (34,6 ± 11,5), 7,3 - 29,4 (21,0 ± 8,1), 6,9 - 24,1 (19,4 ± 6,4), và 3,2 - 5,4 (4,0 ± 0,7) µg/g. Trạm Phan Thiết có mức độ các kim loại trong trầm tích thấp nhất. Trong khi đó, giá trị cực đại đều ghi nhận tại các trạm phía Nam, cụ thể tại các trạm Gành Rái (Cr, Cu, Zn), Định An (As), và Rạch Giá (Pb). Hàm lượng As, Cd và Zn thấp hơn ngưỡng TEL (threshold effect levels) theo Chất lượng trầm tích Canada với mục đích Bảo vệ Sinh vật Thủy sinh, trong khi hàm lượng Cr, Cu, và Pb vượt ngưỡng này lần lượt từ 1,1 - 1,2 lần, 1,0 - 2,5, và 1,0 - 1,3 lần. Hệ số ô nhiễm (CF) và chỉ số tích lũy địa lý (I_{geo}) được tính toán để đánh giá mức độ ô nhiễm của kim loại nặng trong trầm tích. Bên cạnh đó, hệ số rủi ro sinh thái (E_r) và chỉ số rủi ro sinh thái tiềm ẩn (RI) đã được áp dụng để đánh giá nguy cơ đối với hệ sinh thái. Các giá trị CF cho thấy mức độ ô nhiễm As và Cd trong trầm tích ở mức độ trung bình hoặc đáng kể, trong khi Cr, Cu, Pb và Zn ở mức ô nhiễm thấp hoặc trung bình. Bên cạnh đó, giá trị CF cho thấy mức độ ô nhiễm kim loại nặng của Định An và Gành Rái lớn hơn so với các địa điểm khác. Theo hệ số rủi ro sinh thái (E_r) và chỉ số tích lũy địa lý (I_{geo}), mức độ ô nhiễm Cd trong trầm tích ở mức không ô nhiễm hoặc trung bình, với rủi ro sinh thái từ mức trung bình đến đáng kể. Các giá trị chỉ số rủi ro sinh thái tiềm ẩn (RI) cho thấy Nha Trang, Định An, Gành Rái và Rạch Giá có mức độ rủi ro trung bình.

Từ khóa: Kim loại nặng, trầm tích, mức độ nhiễm bẩn, mức độ rủi ro sinh thái, Nam Việt Nam.