Concentrations of heavy metals in water from the Southern coast of Vietnam

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ABSTRACT

Concentrations of heavy metals (As, Cd, Pb and Hg) in water collected from seven coastal provinces between December 2012 and December 2015 were evaluated. The average total concentrations (μg/L) of As, Cd, Pb and Hg in water ranged from 2.90 to 6.38, $<$ 0.039 to 0.322, 4.26 to 10.5 and $<$ 0.01 to 0.118, respectively. The average concentrations (μg/L) of As, Pb and Hg in suspended particulate matters (SPM) ranged from 0.392 to 7.32, 0.365 to 18.7 and $<$ 0.01 to 0.038, respectively; whereas, Cd concentrations were not detected in most of SPM samples. There were positive linear relationships between concentrations of heavy metals in water and SPM, except for Cd. The results showed that the concentrations of metals analyzed in water remained below quality guidelines for the protection of aquatic life recommended by the international and Vietnamese organizations. However, As levels in 2/103 and 5/103 of water samples exceeded the QCVN 10:2015/BTNMT for maximum permitted level using for aquaculture and aquatic life protection (20 μg/L) and the Canadian water quality guidelines for the protection of aquatic life (12.5 μg/L), respectively.

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1. Introduction

Currently, nearly a half of Vietnam's population lives in the 28 coastal provinces, and there is an increasing migration into these regions where there are many large cities (e.g., Ho Chi Minh City, Da Nang, Hai Phong) and centralized industrial zones. These activities have created an increased pollution, most likely in hotspots such as the major estuaries and the coastline, which receive different kinds of wastes produced by inland industrial and population centers. The government findings estimate that 60% of the marine pollution originates from land-based sources, including nutrients, persistent organic pollutants and heavy metals (HMs) in water and sediments (MONRE, 2010).

The HMs, particularly As, Cd, Pb and Hg, are considered most toxic to biota and environment. HMs contamination can result in adverse effects including growth changes, metabolic process, and disease development (Morais et al., 2012). The fate and toxicity of HMs in the aquatic environment are significantly dependent on the distributing among the aqueous phase, suspended particulate matter (SPM) and sediments (Yang et al., 2016). Dissolved HMs existing in the pore waters are more bioavailable and toxic than particulate HMs (Chapman et al., 1998). While, SPM can play an important role in controlling the reactivity, transport, and biological impacts of HMs in the water (Yang et al., 2016). Understanding the main factors that influence the distribution of HMs in water would allow better prediction of

the changes in HMs toxicity to aquatic organisms (Atkinson et al., 2007). However, no study has comprehensively analyzed the multiphase partition of HMs for SPM-water in the Southern coast of Vietnam.

In the present study, the concentrations of four HMs (i.e. As, Cd, Pb and Hg) were analyzed in waters along the southern coast of Vietnam. Another objective of this study was to evaluate the relationships between the physico-chemical properties and HMs.

2. Materials and Methods

2.1. Sample collection and preparation

Water samples were collected from the extensive cockle culture along the Southern Vietnamese coast, including Ho Chi Minh City (HCM) and 6 provinces including Tien Giang (TGI), Ben Tre (BTR), Tra Vinh (TVI), Bac Lieu (BLI), Ca Mau (CMA), and Kien Giang (KGI) between December 2012 and December 2015 (Figure 1).

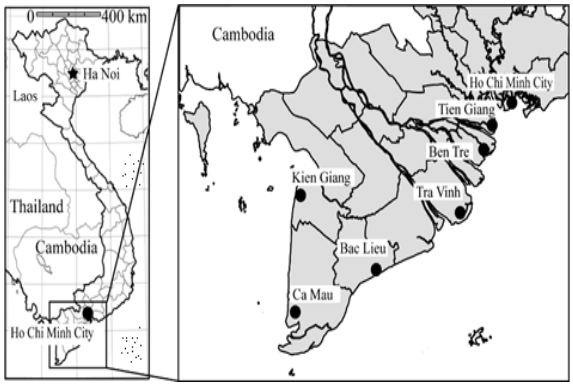

Figure 1. A map showing sampling locations in Southern coast of Vietnam.

Samples of 2 L of water were taken manually at a depth of approximately 20 cm below surface using acid-washed polyethylene bottles (VS, 1995a). Samples for total HMs determination were immediately acidified to pH 2 using 0.5 mL of concentrated HNO_3 (VS, 1995b). For analyzing HMs level in SPM fraction, approximately 500 mL of the water sample was filtered through 0.45-μm cellulose acetate membrane filters (Sartorius; cleaned with 5% v/v HNO_3 and washed in redistilled water) by vacuum pump. The membranes were kept in an ice box, transported to laboratory and stored in a deep freezer at – 20^0C before analysis.

At each sampling site, water quality parameters, including temperature, pH, electrical conductivity (EC and salinity), and dissolved oxygen (DO) were measured using a mercury thermometer, Denver pH meter UP-5, SevenGo pro^{TM} conductivity and MW600 standard portable DO meter, respectively.

2.2. Water sample analyses

Concentration of As in water was analyzed in accordance with the Vietnamese standard TCVN 6626-2000 (VS, 2000). For each sample, 50 mL of water was placed in a round-bottom flask with the addition of 5 mL of concentrated H_2SO_4 and 5 mL H_2O_2 30%, and then heated at 180^0C on a hot plate until white fume occurred. The solution was cooled, diluted to 25 mL and then transferred into a 40-mL glass screw-cap tube with a Teflon-faced silicone septum. Samples were prepared for hydride analysis by adding 0.5 mL of L-cysteine 5% and 4 mL of 3M HCl and heated in a water-bath at 90^0C for 45 min. Bring tube to 50 mL with 0.6 M HCl before analyzing.

Hg level in water was determined in accordance with TCVN 5991:1995 (VS, 1995c). Place a 50-mL portion of water in 50-mL glass screw-cap tube with a Teflon-faced silicone septum, add 0.4 mL of BrCl solution and kept in 3 hrs. To remove residue of BrCl, the solution was added some drops of 10% $NH_2OH.HCl$ before analyzing.

For analyzing Cd and Pb levels in water, 50 mL of samples were acidified by redistilled HNO3 and heated on hot plate at 90 – 95^0C until the volume was reduced to 15 – 20 mL. Samples were cooled, filtered and final volume was made up to 50 mL with double-distilled water.

Concentrations of Hg and As were determined by gold amalgamation cold vapour atomic absorption (CV-AAS) spectrometry and hydride generation atomic absorption spectrometry (HG-AAS), respectively. Levels of Cd and Pb were analyzed by an inductively coupled plasma mass spectrometry (ICP-MS).

2.3. SPM analyses

Whole membranes were placed in Teflon vials with the addition of 10 mL of concentrated

HNO_3, and then heated at 150^0C for 2 h. Digested solution was cooled, added 1 mL H_2O_2 and heated at 150^0C for 1 h. The whole procedure, including addition of H_2O_2 and heating was conducted in duplicate. The solution was then centrifuged, filtered, and then made up to 50 mL.

For As analysis, aliquot of 10 mL was placed in Erlenmeyer flask with addition of 0.5 mL of concentrated H_2SO_4 and heated on hot plate until white fume arose. And sample was treated and analyzed by the procedure used for total As analysis in water.

For Hg analysis, a portion of digested solution was added 1 ml of concentrated HCl and made up to 50 mL by double distilled water. Concentration of Hg was determined using a CV-AAS.

For Cd and Pb determination, the digested solution was directly analyzed by ICP-MS.

2.4. Quality assurance and quality control (QA/QC) procedure

Reagent grade chemicals were used in all analytical procedures. For each batch or 20 samples, at least one matrix spike, one matrix spike duplicate and two reagent blanks were used as the QA/QC procedure of the analysis. Recoveries of the HMs ranged from 80 to 120% of the spiked values. Detection limits for As, Cd, Pb and Hg were 0.13, 0.039, 0.14 and 0.01 μg/L, respectively. All data were expressed in μg/L.

2.5. Statistical analysis

One-half of the value of the respective limit of detection was substituted for those values below the limit of detection and used in statistical analysis (US EPA, 2000). All data were tested for goodness of fit to a normal distribution and homogeneity of variances with a Kolmogorov–Smirnov's one sample test and Levene test, respectively. Because most of the variables were not normally distributed, the data were logarithmically transformed and subjected to parametric statistics. Pearson correlation and simple linear regression were used to measure the relationships among concentrations of HMs in water, SPM and physicochemical parameters. For testing provincial differences, log transformed data were analyzed using one-way analysis of variance (ANOVA) with Duncan multiple range test as pairwise comparisons. A *P-value* < 0.05 was considered to indicate statistical significance. These statistical analyses were executed by the program SPSS (version 19, SPSS, Chicago, IL, USA).

3. Results and Discussion

3.1. Physicochemical parameters of water samples

Physicochemical parameters of water samples are shown in Table 1. Average temperature ranged from 27.8^0C in the BTR water to 32.1^0C in the HCM water. The lowest pH value (6.81) was found in water from HCM, and the highest value (7.93) occurred in KGI water, which was found to be within the prescribed limit of 6.5 – 8.5 (MONRE, 2015). pH values in water found in these regions were similar to those reported in the previous studies. Pham et al. (2007) reported that pH values in the coast of Can Gio, HCM City ranged from 7.8 to 8.1. During As accumulation study in surface water in some estuaries of the Mekong River Delta (MRD), Bui et al. (2011) observed that pH values remained same in sampling areas and were in range of 7.5 – 7.9. Similar results were reported by Nguyen (2007), the pH values ranged from 7.1 – 7.2 for the coastal regions of Ngoc Hien, Ca Mau. EC and salinity in water from the Southern coast found to be in the ranges of 20.9 – 35.9 mS/cm and 12.4 – 22.6‰, respectively. On comparison with other data by other works, it was found that the minimum and maximum values of EC and salinity were 0.29 – 43.1 mS/cm and 2.0 – 28.9‰, respectively (Bui et al., 2011), which are comparable to this study. From Table 1, it was clear that the range of the DO lies between 4.87 to 7.20 which follow the standards given by MONRE (2015).

3.2. Provincial differences in heavy metal concentrations

Concentrations of HMs in water (103 samples) and SPM (30 samples) collected from the Southern coast of Vietnam were exhibited in Table 2.

3.2.1. Arsenic

Mean As concentrations ranged from 2.90 μg/L in water from HCM to 6.38 μg/L in water from KGI (Table 2). No statistically significant difference was found among provinces (ANOVA, $P < 0.05$). Similar results were reported by Nguyen (2007), As concentrations in water ranged from

Table 1. Physicochemical parameters of water samples collected from the coasts of Vietnam[1]

Province	Temperature (^{0}C)	pH	EC (mS/cm)	Salinity (‰)	DO (mg/L)
KGI	31.1 ± 2.9	7.93 ± 0.42	21.1 ± 6.6	12.4 ± 4.3	7.20 ± 1.16
BLI	30.4 ± 3.3	7.84 ± 0.46	21.4 ± 9.6	12.9 ± 6.1	5.90 ± 1.66
CMA	30.0 ± 1.3	7.65 ± 0.26	32.0 ± 10.0	18.9 ± 5.9	6.08 ± 0.64
TRV	28.7 ± 1.7	7.40 ± 0.12	20.9 ± 7.6	12.6 ± 5.0	4.87 ± 0.47
BTR	27.8 ± 3.2	7.61 ± 0.93	24.0 ± 11.6	15.4 ± 8.1	6.40 ± 1.27
TGI	29.9 ± 2.1	7.10 ± 0.70	24.1 ± 14.9	14.6 ± 9.6	6.40 ± 1.32
HCM	32.1 ± 3.1	6.81 ± 0.65	35.9 ± 4.8	22.6 ± 3.4	6.67 ± 1.27

[1]Data are mean ± standard deviation.

Table 2. Heavy metal concentrations (μg/L) in water and SPM collected from the Southern coast of Vietnam[1]

Province	n	As	Cd	Pb	Hg
Water					
KGI	25	6.38 ± 5.44^{a}	< 0.039^{a}	10.2 ± 17.8^{a}	0.031 ± 0.023^{ab}
BLI	22	5.54 ± 3.99^{a}	0.167 ± 0.447^{a}	10.5 ± 16.4^{a}	0.064 ± 0.068^{bc}
CMA	26	3.41 ± 2.35^{a}	< 0.039^{a}	1.93 ± 2.29^{a}	0.118 ± 0.086^{c}
TRV	3	4.23 ± 2.24^{a}	< 0.039^{a}	8.81 ± 12.5^{a}	0.011 ± 0.010^{a}
BTR	15	5.09 ± 4.22^{a}	0.056 ± 0.082^{a}	5.87 ± 4.42^{a}	0.038 ± 0.064^{ab}
TGI	9	6.05 ± 7.40^{a}	0.322 ± 0.814^{a}	9.47 ± 12.0^{a}	0.049 ± 0.040^{bc}
HCM	3	2.90 ± 0.28^{a}	< 0.039^{a}	4.26 ± 5.11^{a}	0.022 ± 0.014^{ab}
SPM					
KGI	8	3.45 ± 3.39	ND	6.81 ± 7.05	0.019 ± 0.017
BLI	8	7.32 ± 5.63	ND	18.7 ± 19.8	0.038 ± 0.025
TRV	2	0.711 ± 0.419	ND	0.365 ± 0.132	< 0.01
BTR	6	2.61 ± 3.03	ND	4.32 ± 3.93	0.016 ± 0.012
TGI	5	6.76 ± 6.40	ND	9.32 ± 11.80	0.026 ± 0.028
HCM	1	0.392	ND	7.35	< 0.01

[1]Data are mean ± standard deviation. Values in the same column with different superscript letters are significantly different ($P < 0.05$, one-way ANOVA with Duncan test); ND: not detected.

0.40 – 23.3 μg/L for the coastal regions of Ngoc Hien, Ca Mau. In study of As accumulation in surface water in some estuaries of the Mekong River Delta (MRD), Bui et al. (2011) observed As levels in water between freshwater and brackish-water and saltwater were significantly different with mean levels of 1.48 ± 1.26 μg/L, 8.51 ± 7.79 μg/L and 49.47 ± 23.57 μg/L, respectively.

In this study, the general tendency in average As concentrations in SPM among different provinces were BLI > TGI > KGI > BTR > TVI > HCM. The positive relationship between As concentrations in water and SPM samples was relatively strong significant (Simple linear regression, $R^2 = 0.59$, $P < 0.001$) (Figure 2). This result was agreed with the work of Yang et al. (2016) at three large shallow lakes in China, e.g., Taihu, Chaohu, and Dianchi. These authors reported that total As in water had a significant positive relationship with the total As in SPM at all three lakes ($R^2 > 0.735$). As a result of fluctuations in physical, chemical, and biological factors occurred in natural water, As can be adsorbed onto SPM from aqueous dissolved phase, precipitated, and deposited to sediments. As reported by other works, the distribution of As between water and SPM can be influenced by concentrations and compositions of SPM (e.g., Fe and Mn oxides), salinity, pH, redox condition as well as biological effects (Balzer et al., 2013; Hong et al., 2016; Yang et al., 2016).

3.2.2. Cadmium

Average levels of Cd in our sample set ranged from < 0.039 μg/L to 0.322 ± 0.814 μg/L; the highest level was found in TGI, followed by BLI (0.167 ± 0.447 μg/L) and the lowest in KGI, CMA, TVI and HCM (< 0.039 μg/L). However, there was no statistically significant differences in

mean Cd concentration of water samples among provinces ($P > 0.05$). In contrast, concentrations of Cd were not detected in all SPM samples. This result was similar to that reported by Cenci and Martin (2004). In estuaries, Cd desorption should be expected due to chloride and sulfate complexation, and ionic strength effects. Mobilisation of Cd was recognized in many estuaries such as in the Amazon plume, Changjiang, Gironde, Mississippi, etc. (cited therein Cenci and Martin, 2004).

3.2.3. Lead

Lead concentrations in waters from these provinces varied widely, and extremely elevated levels were found in samples collected from BLI (Table 2). However, there were no statistically significant differences in mean Pb levels of samples among provinces ($P > 0.05$). While, Pb concentration in SPM samples ranged from 0.365 ± 0.132 μg/L in TRV to 18.7 ± 19.8 μg/L in BLI. The highly significant positive correlations found between total Pb concentrations in water and SPM samples ($R^2 = 0.71$, $P < 0.001$) (Figure 2).

3.2.4. Mercury

Average levels of Hg in water collected from the southern coast were highest in CMA (0.118 ± 0.086 μg/L), followed by BLI (0.064 ± 0.068 μg/L) and lowest in TVI (0.011 ± 0.010 μg/L). On the contrary, Hg concentrations were not detected in all SPM samples. There were statistically significant differences in mean Hg levels of water samples among provinces ($P < 0.05$) (Table 2). Log concentrations of Hg in water correlated positively with log concentrations of Hg in SPM ($R^2 = 0.84$, $P < 0.001$) (Figure 2).

Similar to As, in the estuarine waters, Pb and Hg distributed between the dissolved phase (water) and particulate (SPM). The dissolution of HMs between these phases depends on the physicochemical properties of the SPM as well as various ambient conditions such as salinity, pH, and the types and concentrations of dissolved organic matter (Wang et al., 2016). In this study, there were highly positive relationships between Pb and Hg concentrations in water and SPM samples. Yao et al. (2016) reported similar good linear regressions of total HMs concentrations between dissolved and particulate phase in estuarine waters.

3.3. Relationships between concentrations of total heavy metals in water and physicochemical properties

In the present study, relationships between physicochemical characteristics (temperature, pH, EC, salinity and DO) and HMs concentrations in water as well as among HMs were evaluated using Pearson correlations. The highly significant positive correlations discovered between EC and salinity ($r = 0.97$); therefore, just relationships between EC and other parameters were discussed. The weak positive correlations found between temperature and DO ($r = 0.31$), and concentrations of Cd and Pb ($r = 0.41$) in water; whereas the weak negative associations observed between EC and DO (r = -0.26), and concentrations of Pb and Hg (r = -0.29) in water. Moreover, EC and concentrations of Hg was weakly positively associated ($r = 0.25$). In addition, As levels were positively related with Cd ($r = 0.36$) and Pb ($r = 0.70$), but negatively related with Hg (r = -0.25). However, no correlations were found between pH and other parameters (Table 3).

3.4. Comparison with published data and guidelines

Analytical techniques have played an important role in assessing pollution of estuarine system and have been extensively used to determine various HMs levels in seawater, marine sediments and biota. However, the high salt content and a very low concentration for the HMs (ranging from < ng/L to μg/L) can be major obstacles to analyzing HMs in brackish and marine waters. Therefore, very little research has been done on concentrations of HMs in estuarine water.

Compared to other published works, average concentrations of As in water collected from the Vietnam southern coast were comparable with or lower. Levels of As in the coast water of Can Gio, HCM were 31 μg/L (Pham et al., 2007). While, accumulation levels of As in the coast water of Ngoc Hien, Ca Mau ranged 0.4 – 23,3 μg/L (Nguyen, 2007) (Table 4). Similarly, Bui et al. (2011) observed As levels in water between freshwater and brackish-water and saltwater were significantly different with mean levels of 1.48 ± 1.26 μg/L, 8.51 ± 7.79 μg/L and 49.47 ± 23.57 μg/L, respectively. Moreover, Phung and Huynh (2015) reported levels of As accumulated in water of Mekong River ranged 1.48 – 49.47

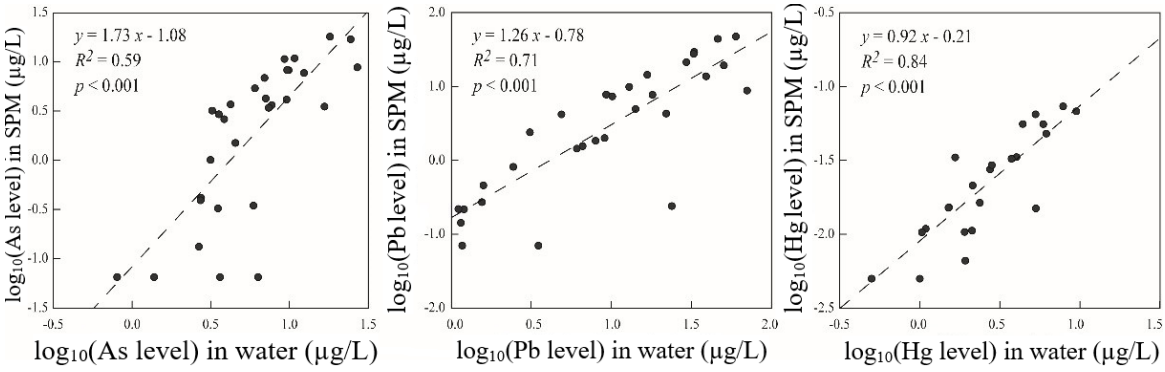

Figure 2. Relationships between heavy metal concentrations (μg/L, log-transformed) in water and SPM (n = 30) collected from the Southern coast of Vietnam.

Table 3. Correlation coefficients (r) for relationships between log-transformed concentrations (μg/L) of total heavy metals in water and physicochemical properties from the Southern coast of Vietnam[1]

	pH	EC	Salinity	DO	As	Cd	Pb	Hg
Temperature	-0.03	0.00	-0.10	**0.31**	-0.14	-0.17	-0.18	0.17
pH		0.00	-0.04	0.12	0.15	0.01	0.00	-0.17
EC			**0.97**	**-0.26**	0.14	-0.02	0.07	**0.25**
Salinity				**-0.26**	0.15	0.00	0.09	**0.21**
DO					-0.13	-0.20	-0.17	-0.12
As						**0.36**	**0.70**	**-0.25**
Cd							**0.41**	-0.19
Pb								**-0.29**

[1]Bold numbers in columns indicate significance at $P < 0.05$.

μg/L. However, concentrations of As in water in this study were lower than those collected in the Straits of Malacca, Malaysia (4.98 – 86.14 μg/L) (Looi et al., 2013). The concentrations of As analyzed in water remained below quality guidelines for the protection of aquatic life recommended by the Canadian Council of Ministers of the Environment (CCME, 2007) (12.5 μg/L) and the Vietnamese regulation (QCVN 10-MT : 2015/BTNMT, 20 μg/L) (CCME, 2007; MONRE, 2015) (Table 4). However, As levels in 2/103 and 5/103 of water samples exceeded the QCVN 10:2015/BTNMT and the CCME, respectively. The quality guidelines for As were developed from acute and chronic ecotoxicological data on marine species (CCME, 2007). Thus, the As contamination in the Southern coast of Vietnam did not exceed tolerable ecotoxicological risk levels.

Mean Cd levels in water collected from the estuary system were found to be similar to those reported from Mekong River (Cenci and Martin, 2004), but lower than those in water from the coast area of Can Gio, HCM (Pham et al., 2007), Ngoc Hien, Ca Mau (Nguyen, 2007) and the Malacca Straits (Looi et al., 2013). Concentrations of Cd detected in all water samples did not exceed the Vietnamese technical regulation on marine water quality (QCVN 10-MT: 2015/BTNMT, 5 μg/L), but, about 6% (6/103) of water samples had Cd levels exceeding the CCME guideline of 0.12 μg/L (Table 4).

Mean Pb concentrations in water collected from the estuary system were found to be higher than those reported from the coast area of Can Gio, HCM (Pham et al., 2007) and Ngoc Hien, Ca Mau (Nguyen, 2007), but comparable to samples from the Malacca Straits (Looi et al., 2013). Only 3/103 of water samples had Pb levels exceeding the Vietnamese technical regulation on marine water quality (QCVN 10-MT: 2015/BTNMT, 50 μg/L) (Table 4).

Generally, Hg is present in trace amounts in most of water samples. Compared with the

Vietnamese technical regulation on marine water quality (QCVN 10-MT : 2015/BTNMT, 1 μg/L) and quality guidelines for the protection of aquatic life recommended by the CCME (0.016 μg/L) (CCME, 2007; MONRE, 2015), Hg concentrations in all of samples were below the Vietnamese guideline, but 75/103 of samples had Hg levels exceeding the CCME guideline. According to the CCME (CCME, 2003), toxicity data on Hg for marine waters are much more limited. EC50s for inorganic Hg range from $<$ 5 - 55 μg/L for fish, from 1.2 - 20 μg/L for invertebrates and from 0.16 - 1002 μg/L for plants and algae. The LOAEL (lowest observed adverse effect level) of 0.16 μg/L was used to develop the guideline. The LOAEL was divided by a safety factor of 10 to give an interim Canadian water quality guideline of 0.016 μg/L or 16 ng/L.

4. Conclusions

Based on the results of this study, it may be concluded that the concentration of HMs in water collected from the Southern coast of Vietnam is quite uniform among provinces. The results indicated that the regions are in the range of 'unpolluted' water. These results could be useful for future relative studies of HMs contamination and monitoring programs to assess marine pollution originates from land-based sources. Further research on understanding the distribution and accumulation profiles of potential HMs in sediment and biota samples from these regions is clearly warranted.

Table 4. Comparison of concentrations of heavy metals (μg/L) in water with those from other sites and guidelines

Country	Region	As	Cd	Pb	Hg	Reference
Vietnam	South coast	5.07 ± 4.45	0.087 ± 0.320	7.28 ± 12.7	0.062 ± 0.070	This study
	Can Gio, HCM	31	1	3	< 0.5	Pham et al. (2007)
	Ngoc Hien, Ca Mau	0.4 - 23.3	0.18 - 2.63	0.06 - 8.96	Not detected	Nguyen (2007)
	Tien - Hau Rivers	1.48 - 49.47	-	-	-	Bui et al. (2011)
	Mekong river - dissolved	-	0.001 - 0.051	0.02 - 0.16	-	Cenci and Martin (2004)
	Mekong river - SPM	-	2 - 73	4 - 84	-	
Malaysia	Malacca Strait	4.98 - 86.14	Not detected - 5.66	Not detected - 28.6	-	Looi et al. (2013)
Vietnam guideline		20	5	50	1	MONRE (2015)
Canadian guideline		12.5	0.12	-	0.016	CCME (2007)

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