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RESEARCH ARTICLE

Occurrence of heavy metals and radionuclides in sediments and seawater in mangrove ecosystems in Pattani Bay, Thailand

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Abstract Mangrove ecosystems in Pattani Bay, Thailand are considered representatives for monitoring the occurrence of anthropogenic and natural pollution due to metal and radionuclide contamination. Sediments and seawater were collected from five locations to determine metal (Cd, Cr, Cu, Mn, Ni, Zn, and Pb) and radionuclide (^{226}Ra, ^{232}Th, and ^{40}K) concentrations. Spatial variations in metal and radionuclide concentrations were determined among the sampling sites. A geoaccumulation index (I_{geo}) and enrichment factor (EF) were used to classify the impacts of metals from anthropogenic point sources. Significant values for I_{geo} and EF were measured for Pb in site 4 (I_{geo} 0.65; EF 28.2) and Cd in site 1 (I_{geo} 1.48; EF 46.2). EF values in almost all sampling sites were >1 which indicates anthropogenic pollution. To assess the potential public hazard of radioactivity, the average radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), the internal hazard index (H_{in}), the absorbed dose rate in air (D), and the annual effective outdoor dose rate (E) were determined. Based on these measurements, it is concluded that the probability of human health risk from radionuclides is low. However, the absorbed dose in air (D) values in sites 4 and 5 were greater than the global average value of 55 nGy h^{-1}, indicating that sediments in these locations pose a radiological hazard. The data obtained in this study provides useful information on metal and radionuclide background levels in mangrove sediments and seawater, and can be applied toward human health risk assessment and metal and radionuclide mapping.

Keywords Heavy metals · Radionuclides · Mangrove coastal area · Sediment · Geoaccumulation index · Enrichment factor · Radiological risk assessment · Pattani Bay

Introduction

Sediment in mangrove ecosystems is a primary sink for contaminants from terrestrial sources (Mountouris et al. 2002). Contaminants become immobilized in sediments in part due to their anaerobic character combined with their enrichment in sulfides and organic matter (Peters et al. 1997). Contaminants such as lead (Pb) and cadmium (Cd) are adsorbed on ion exchange sites of fine silt or clay or are co-precipitated with other metals, i.e., manganese (Mn), aluminum (Al), and iron (Fe) (Harbison 1986). Furthermore, metals and radionuclides commonly occur in seawater in trace quantities (Yap et al. 2011; Yunus et al. 2015). However, anthropogenic and natural disturbances can increase the quantity of contaminants via redistribution from sediments to seawater. In particular, mining is considered a key source of radionuclides (Hu et al. 2014), while fertilizer runoff from agricultural lands serves as a source of ^{226}Ra, particularly in superphosphates. Levels as high as of 571.22 Bq kg^{-1} have been measured (Saleh et al. 2007).

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The original publication of this paper contains a mistake. The sentence in the caption of Table 4 "Sharp (#) indicates the difference of 40K concentration between sites 4 and 5" should be removed and retained in the caption of Table 3.

The original article was corrected.

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Pattani Bay is located along the southern Gulf of Thailand. Coastal zones are characterized by sand and muddy sand sediments (Swennen et al. 2001). The region consists of a diverse array of mangrove vegetation, and aquatic and terrestrial organisms. Industrial activities around Pattani Bay have, over the past few decades, improved the economic status of local communities (Cheewasedtham et al. 2003; Sowana et al. 2011). Unfortunately, however, massive quantities of wastes from these activities have been released into local ecosystems, particularly in waterways. Several facilities and sites are considered primary point sources of contaminants, which have led to bioaccumulation in food chains. Accurate and current data for metal and radionuclide concentrations in terrestrial and marine ecosystems of Pattani Bay, Thailand is essentially nonexistent (Kaewtubtim et al. 2015, 2016). Little data is available on the physicochemical status of mangrove ecosystems along Pattani Bay, as many occur in conflict zones or so-called 'red areas'.

A number of metals and radionuclides occurring in Pattani Bay sediments are believed to be enriched from industrial, agricultural and domestic activities, and from abandoned tin mines. It is known that several metals are linked with an increased incidence of acute and chronic diseases, including those affecting the nervous system, the eyes, skin, and lungs. The rates of certain types of cancers are also elevated (Singh et al. 2011; Haddad 2012). A number of chronic diseases are known to be associated with exposure to radionuclides (Frontasyeva et al. 2001). Hence, an accurate accounting of metal and radionuclide concentrations in both sediments and seawater is necessary for proactive environmental management since harvest of marine organisms (i.e., fish, macroalgae, crab, etc.) and aquaculture are common activities in coastal ecosystems of Pattani Bay.

In this first reported investigation, the distribution of metals and radionuclides in sediment and seawater from Pattani Bay, Thailand was documented. Heavy metals including Cd, Cr, Cu, Mn, Ni, Zn, and Pb were selected for determination as they comprise key contaminants of local mangrove ecosystems. Radionuclides such as ^{226}Ra, ^{232}Th, and ^{40}K are considered representative radionuclides that affect human health (Alamgir Miah et al. 2012). The Geoaccumulation Index (I_{geo}) and enrichment factor (*EF*) were used to screen the anthropogenic impacts for metals in the study sites and to determine ecological risk. This information will be used for decision making to ultimately establish an environmental management program in coastal areas of Thailand, with particular emphasis on mangrove ecosystems.

Materials and methods

Study areas and physicochemical analysis of the samples

Sediment and seawater samples were collected from five sites in the coastal area of Pattani Bay (site 1: N 6° 53′ 77.6″ E 101° 15′ 01.2″; site 2: N 6° 53′ 68.4″ E 101° 14′ 41.9″; site 3: N 6° 53′ 68.4″ E 101° 14′ 41.9″; site 4: N 6° 54′ 85.3″ E 101° 14′ 82.1″; and site 5: N 6° 54′ 87.8″ E 101° 14′ 98.6″) (Fig. 1). The average annual temperature and precipitation in 2014 were 27.5 °C and 5.62 mm y^{-1}, respectively. The distance between sampling sites is approximately 0.5 km. The sampling sites are located near mangrove forests, and are surrounded by residential dwellings and industrial estates.

Sediments were collected using a 2-cm diameter stainless steel hand corer, placed into plastic bags and transferred to an insulated box. All samples were subsequently transported to the laboratory. The sediments were allowed to air dry at room temperature following which they were grounded with an agate mortar and pestle, and sieved to pass a 2-mm mesh sieve.

Physicochemical properties of the sediments were determined following standard methods. One-half gram of sediment was placed into a Pyrex® test tube and digested with conc. HNO_3 in a heating block (APHA, AWWA and WEF 2005). Total Zn, Pb, Mn, Cr, Cu, Ni, and Cd concentrations were determined using a flame atomic absorption spectrophotometer (FAAS; AAnalyst™ 200, PerkinElmer) or a graphite furnace atomic absorption spectrophotometer (GF-AAS; AAnalyst™ 600, PerkinElmer) after HNO_3 digestion (APHA, AWWA and WEF 2005), depending on metal concentration. Extractable Zn, Pb, Mn, Mn, Cr, Cu, Ni, and Cd concentrations were determined by FAAS or GF-AAS after extraction with 0.05 M diethylene triamine pentaacetic acid (DTPA) (ICARDA 2001). Analytical accuracy and precision were determined by running standard solutions after every 20 samples. A method blank and certified reference materials (NIST SRM® 2710a Montana soil and HPS Certified Waste Water Trace Metals Lot#D532205 for sediment and seawater, respectively), were used for quality control. Percentage recovery was in the range of 100.2–118.0 and 100.2–116.0% for sediment and seawater, respectively.

Total nitrogen (N) was determined using the Kjeldahl method (Black 1965), extractable phosphorus (P) concentrations using the Bray II method (Bray and Kurtz 1945), and extractable potassium (K) concentrations using FAAS after extraction with neutral NH_4OAc (Sparks 1996). Soil cation exchange capacity was determined after leaching with 1 N NH_4OAc buffer (Sparks 1996). Organic matter content was measured using the Walkley–Black titration method (Walkley and Black 1934). Soil pH was measured on a 1:5 (*w*/*v*) suspension of soil deionized water using a glass electrode pH meter (Hanna instruments HI 221). Electrical conductivity (EC) was measured using an EC meter (Hanna instruments HI 993310). Soil texture was determined using the hydrometer method (Allen et al. 1974). Concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were determined by gamma-ray spectrometry with a high purity germanium (HPGe) detector. The energy calibration was performed using standard reference radionuclide sources ^{60}Co and ^{137}Cs, while the efficiency calibration was performed using reference samples ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K obtained from the Office of Atoms for Peace, Thailand.

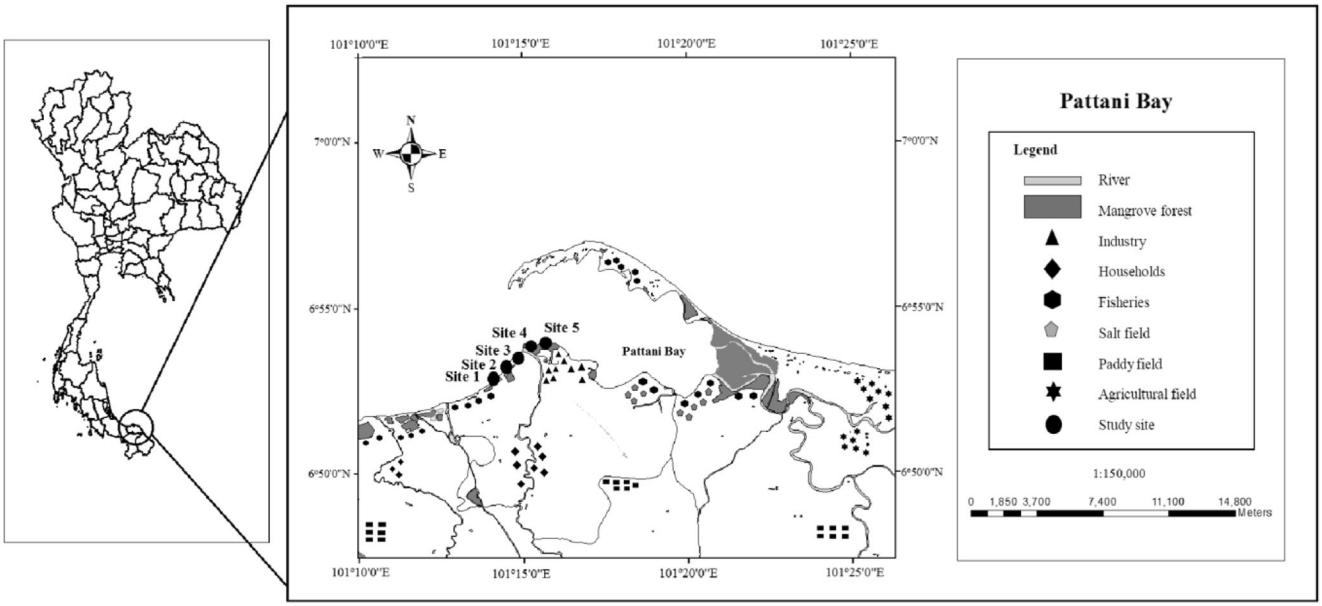

Fig. 1 Map of study sites in Pattani Bay, Thailand

Fifty mL of seawater was filtered through a 0.45-μm cellulose membrane filter and then acidified with 0.05 mL double-distilled HCl (Merck®) to attain pH < 2. Total metal concentrations were determined by FAAS or GF-AAS. Separate samples were analyzed for the selected radionuclides by gamma-ray spectrometry with a high purity germanium (HPGe) detector after filtering without acidified water.

The radioactivity of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using the following equation (IAEA 1989):

$$C_i = \mathrm{A}/(E \times T \times P \times W)$$

Where C_i is the specific activity of the radionuclide in the plant (Bq kg^{-1}), A is the count of the radionuclide, E is the detector efficiency of the specific gamma-ray, P is the absolute transition probability of the specific gamma-ray, T is the time (s), and W is the mass of the sample (kg).

Evaluation of sediment contamination

The Geoaccumulation Index (I_{geo}) is a tool for assessing possible enrichment of metals in sediments (Müller 1969). It is calculated as follows:

$$I_{geo} = \log_2\left[C_n/(1.5\ B_n)\right.$$

where C_n is the measured concentration of metal 'n', B_n is the metal concentration in 'average shale' (Turekian and Wedepohl 1961), and 1.5 is the background matrix correction factor due to lithogenic effects (Nowrouzi and Pourkhabbaz 2014).

I_{geo} is divided into seven classes, where the highest value (class 6) indicates a 100-fold enrichment above background values (Ghrefat et al. 2011). The classes for I_{geo} range from Class 0 (practically uncontaminated); $I_{geo} \leq 0$; to class 6 (extremely contaminated); $I_{geo} > 5$.

The enrichment factor (EF) has been widely used for assessing the degree of metal enrichment and is useful for comparing metal enrichment from anthropogenic and natural sources in surface sediments (Karageorgis et al. 2009). For a better understanding of anthropogenic contribution of the metal, an enrichment factor was calculated for each metal by dividing its ratio to the normalizing element by the same ratio found in the chosen baseline (Taylor 1964), and is described as follows:

$$EF = [C_n/C_{\mathrm{Mn}}]_{\mathrm{sediment}}/[C_n/C_{\mathrm{Mn}}]_{\mathrm{background}}$$

where C_n is the concentration of metal 'n'. The background value is the metal concentration in an 'average shale' (Turekian and Wedepohl 1961). Manganese was chosen as a normalizing, or reference element for determining EF values as it is one of the major components of the earth's crust and its concentration in sediment is associated mainly with the matrix (Uduma and Awagu 2013). As proposed by Simex and Helz (1981), EF is classified into five categories, ranging from $EF < 2$ depletion to minimal enrichment; to $ER > 40$ extremely to high enrichment.

The radium equivalent activity is used to assess hazards from ^{226}Ra, ^{232}Th, and ^{40}K in Bq kg^{-1} (Agbalagba and Onoja 2011; Tufail et al. 2011) and is calculated as follows:

$$\mathrm{Ra_{eq}} = A_{\mathrm{Ra}} + 1.43\ A_{\mathrm{Th}} + 0.077\ A_K$$

Where $A_{\mathrm{Ra}}, A_{\mathrm{Th}}, A_K$ are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The external hazard index (H_{ex}) and internal hazard index (H_{in}) were determined. The external hazard index evaluates the hazard of natural gamma radiation, while the internal exposure to radon and its daughter products are evaluated by the internal hazard index (Agbalagba and Onoja 2011; Tufail et al. 2011):

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810)$$

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

The absorbed dose rate in air is commonly employed to assess health risk index (Jahan et al. 2016). As regard to biological effects, radiological, and clinical effects are directly associated with absorbed dose rate. Measured activity concentrations of detected radionuclides are converted into doses as follows:

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Using the conversion coefficient from absorbed dose in air to effective dose of 0.7 Sv Gy^{-1}, as recommended by UNSCEAR (2000), the number of hours in a year of 365 days (8760 h) and the outdoor occupancy factor of 20% (Freitas and Alencar 2004; Jibiri and Okeyode 2012), the annual effective outdoor dose rate is calculated by the equation:

$$E = D\ (\text{nGy h}^{-1})\ \text{x 8760 h x 0.2 x 0.7 Sv Gy}^{-1}\text{x } 10^{-6}$$

Where D is the absorbed dose rate.

Statistical analysis

All data were analyzed using SPSS® version 18.0 (SPSS Inc. Chicago, IL). Analysis of variance was performed using least significant difference (LSD) post hoc comparisons, with a 95% confidence level ($p \leq 0.05$).

Results and discussion

Physicochemical properties of sediments in Pattani Bay mangrove forests are shown in Table 1. Sediment pH values ranged from neutral to slightly alkaline (7.1–7.5), indicating that mobility of metallic and radionuclide contaminants should be minimal (Hu et al. 2014). Mangrove sediments in many areas worldwide tend to be slightly basic in pH due to limited buffering capacity of the sediments (Middelburg et al. 1996). Sediment EC values ranged from 0.4–8.1 mS cm^{-1}, N from 0.02–0.18%, and extractable P from 89 to 986 mg kg^{-1} (Table 1). Maximum organic matter content was 2.5%, which is fairly typical for mangrove sediments of the humid tropics. The influences of organic matter accumulation in tropic sediments depend on sediment grain size, anthropogenic organic input, and decomposition rate of mangrove litterfall (Hossain et al. 2014).

The texture for all sediment samples was loam. In different mangrove forests of the tropics, the majority of sediments are reported as having high proportions of silt and clay; such colloids may trap radioisotopes on particles surfaces, resulting in elevated levels of radiation (Ibrahiem et al. 1993). Sediment texture in the study areas partly reflects parent material, but is also due to disturbance by anthropogenic activities including extensive construction as well as mining. The latter effects changed the mangrove sediment texture from a high clay proportion to loam. In addition, coastal waves affect sediment accumulation at the sampling sites which may affect sediment properties as well. Sites 1, 2, 4, and 5 possessed higher CEC and metal concentrations compared to site 3. These data demonstrate the importance of CEC for metal adsorption and/or precipitation in the sediment; however, CEC values for samples in this study may have lower levels of potentially bound metals on adsorption sites compared to those of other coastal mangrove sediments which have higher clay contents (Nematollahi and Ebrahimi 2015).

Total Cd concentrations in sediment ranged from 0.2 to 2.0 mg kg^{-1}, from 27.4 to 65.2 mg kg^{-1} for Cr, from 0.4 to 23.5 mg kg^{-1} for Cu, from 44.4 to 213.8 mg kg^{-1} for Mn, from 11.4 to 41.4 mg kg^{-1} for Ni, from 3.9 to 30.8 mg kg^{-1} for Zn, and from 9.7 to 57.4 mg kg^{-1} for Pb (Table 1). Concentrations of Cr, Mn, Ni, and Zn were the highest in site 1, while the highest Cd and Cu concentrations were measured in site 5 and Pb in site 1. The differences in metal contents in the sediments might be influenced by mineral and organic matter content that tend to immobilize metals (Saedeleer et al. 2010).

The I_{geo} values for sediment metals are rated as essentially unpolluted except for Cd in site 1 which is classified as moderately polluted (Table 2). This effect may be due to current circulation and sediment dynamics of large bodies of seawater. Cd can be found together with Pb and Zn via discharges from paint factories (Sekabira et al. 2010; Seshan et al. 2010). Furthermore, the high *EF* value for Cd in site 1 could indicate a high degree of anthropogenic impact (Table 2). *EF* values for Pb were >1, which indicate that anthropogenic pollution occurred in the sampling sites (Abdel Ghani 2015).

Metal concentrations of seawater samples were all significantly ($p < 0.05$) lower as compared to concentrations in sediments (Table 3). The lower metal concentrations may be explained by the precipitation–coprecipitation of particulates and sorbed metals during periods of limited water movement (Fonseca et al. 2011).

The activities of sediment radionuclides varied significantly with sampling site ($p < 0.05$). Radionuclide activities ranged from 3.1–5.8 Bq kg^{-1} for ^{226}Ra, 145.9–227.2 Bq kg^{-1} for ^{40}K, and 28.2–86.9 Bq kg^{-1} for ^{232}Th.

Table 1 Physicochemical properties of sediments along the coast of Pattani Bay, Thailand

Parameter	Site 1	Site 2	Site 3	Site 4	Site 5
pH	7.4	7.0	7.4	7.5	7.3
EC (mS cm^{-1})	4.1	0.4	2.2	7.7	8.1
CEC (%)	13.4	8.2	0.3	13.6	11.3
OM (%)	2.5	0.4	1.3	2.5	2.7
Total N (%)	0.17	0.05	0.07	0.18	0.02
Ext P (mg kg^{-1})	867.2	89.3	180.5	787.1	986.3
Ext K (mg kg^{-1})	846.2	65.5	178.8	806.1	1006.4
Soil texture	Loam	Loam	Loam	Loam	Loam
Cd (mg kg^{-1})	0.27 ± 0.03^{a}	0.15 ± 0.04^{a}	0.15 ± 0.02^{a}	0.36 ± 0.02^{a}	1.98 ± 2.70^{a}
Cr (mg kg^{-1})	59.53 ± 0.96^{c}	27.42 ± 1.46^{a}	47.65 ± 2.84^{b}	65.23 ± 2.69^{d}	60.94 ± 1.93^{cd}
Cu (mg kg^{-1})	23.45 ± 0.45^{c}	0.41 ± 0.03^{a}	20.72 ± 1.50^{b}	20.55 ± 0.12^{b}	23.52 ± 0.85^{c}
Mn (mg kg^{-1})	88.57 ± 12.31^{b}	44.44 ± 8.72^{a}	93.23 ± 11.60^{b}	213.80 ± 9.33^{c}	122.37 ± 5.04^{c}
Ni (mg kg^{-1})	12.79 ± 4.36^{a}	11.41 ± 4.33^{a}	13.58 ± 4.26^{a}	41.41 ± 0.59^{b}	29.90 ± 9.99^{b}
Zn (mg kg^{-1})	26.98 ± 0.36^{c}	3.94 ± 0.46^{a}	21.43 ± 0.57^{b}	30.75 ± 1.31^{d}	27.38 ± 0.89^{c}
Pb (mg kg^{-1})	57.40 ± 0.16^{c}	9.70 ± 0.54^{a}	40.94 ± 2.85^{b}	46.04 ± 2.00^{b}	44.73 ± 5.41^{b}
Ext Cd (mg kg^{-1})	BDL[e]	BDL	BDL	BDL	BDL
Ext Cr (mg kg^{-1})	0.56 ± 0.08^{a}	2.53 ± 0.26^{c}	1.80 ± 0.07^{a}	12.98 ± 2.38^{a}	0.93 ± 0.01^{d}
Ext Cu (mg kg^{-1})	7.25 ± 0.60^{d}	2.21 ± 0.25^{c}	2.13 ± 0.02^{b}	24.68 ± 0.71^{b}	0.74 ± 0.08^{c}
Ext Mn (mg kg^{-1})	6.35 ± 1.22^{c}	2.29 ± 0.01^{c}	2.07 ± 0.10^{b}	13.34 ± 2.31^{a}	0.54 ± 0.01^{a}
Ext Ni (mg kg^{-1})	1.18 ± 0.10^{b}	1.46 ± 0.11^{a}	3.81 ± 0.11^{c}	32.62 ± 1.27^{c}	1.56 ± 0.35^{a}
Ext Zn (mg kg^{-1})	6.71 ± 2.19^{c}	1.77 ± 0.13^{b}	3.59 ± 0.38^{c}	30.87 ± 0.59^{c}	0.61 ± 0.03^{b}
Ext Pb (mg kg^{-1})	0.56 ± 0.08^{a}	2.53 ± 0.26^{c}	1.80 ± 0.07^{a}	12.98 ± 2.38^{a}	0.61 ± 0.03^{b}
^{226}Ra (Bq kg^{-1})	4.78 ± 1.67^{b}	3.11 ± 0.22^{a}	4.95 ± 0.30^{b}	5.78 ± 0.52^{c}	5.84 ± 2.45^{c}
^{40}K (Bq kg^{-1})	145.88 ± 1.60^{a}	200.48 ± 1.92^{b}	227.22 ± 2.14^{c}	173.10 ± 1.94^{ab}	169.12 ± 1.84^{ab}
^{232}Th (Bq kg^{-1})	34.98 ± 1.22^{ab}	$28.18 \pm 1.18a^{a}$	49.71 ± 2.80^{b}	79.03 ± 3.74^{c}	86.91 ± 3.29^{d}

Values followed by the same letter are not significantly different (LSD, $p > 0.05$). Mean ± Standard deviation ($n = 3$)

[e] *BDL* indicates below detectable limits

Highest ^{226}Ra and ^{232}Th concentrations were noted at site 5, whereas the highest ^{40}K concentration was detected in site 3. Radionuclide abundance was as follows: $^{40}K > {}^{232}Th > {}^{226}Ra$. The lowest concentrations of radionuclides were from stations located farther from the river mouth (Fig. 1).

Radionuclide concentrations in seawater were generally below detection limits, except for ^{40}K at very low concentrations (3.1 and 2.3 Bq m^{-3} for sites 4 and 5, respectively). However, some reports state that even low doses of radionuclides may increase the frequency of mutations in chromosomes and genes in human somatic, bone marrow, and muscle cells (Cristaldi et al. 1991; Livingston et al. 1997); therefore, radionuclide concentrations must be monitored continuously in environments where bioaccumulation may occur in the food chain. Furthermore, there is little information of natural radioactivity available in both sediment and seawater of Pattani Bay. Concentrations of 175.5, 252.6, and 58.0 Bg kg^{-1} for ^{226}Ra, ^{40}K, and ^{232}Th,

Table 2 Geoaccumulation Index (I_{geo}) and Enrichment Factor (EF) of sediments in Pattani Bay, Thailand

Heavy metal	Site 1		Site 2		Site 3		Site 4		Site 5	
	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF	I_{geo}	EF
Cd	1.48	46.2	−1.10	9.6	−1.10	4.6	−0.51	8.7	−0.22	4.8
Cr	−0.80	4.7	−1.59	5.8	−1.04	4.8	−0.82	6.3	−0.73	2.9
Cu	−1.05	3.6	−5.10	0.2	−1.18	4.2	−1.06	5.0	−1.19	1.8
Ni	−1.22	3.1	−2.19	3.2	−2.02	1.8	−2.08	1.8	−0.90	2.4
Zn	−1.64	2.0	−3.59	0.8	−1.89	2.1	−1.66	2.7	−1.53	1.3
Pb	0.40	15.9	−1.13	9.5	0.31	19.1	0.65	28.2	0.43	9.4

Table 3 Physicochemical properties of the seawater along the Pattani Bay coastline, Thailand

Parameter	Site 1	Site 2	Site 3	Site 4	Site 5
Salanity	20	6	18	20	18
Cu (mg L^{-1})	0.48 ± 0.02^{c}	0.01 ± 0.00^{c}	0.42 ± 0.03^{b}	0.47 ± 0.01^{c}	0.42 ± 0.00^{b}
Zn (mg L^{-1})	0.55 ± 0.02^{c}	0.08 ± 0.01^{c}	0.44 ± 0.01^{b}	0.54 ± 0.01^{c}	0.62 ± 0.03^{d}
Ni (mg L^{-1})	1.21 ± 0.40^{d}	0.48 ± 0.20^{a}	0.56 ± 0.16^{b}	0.53 ± 0.17^{b}	0.84 ± 0.01^{cd}
Mn (mg L^{-1})	4.94 ± 0.22^{c}	1.85 ± 0.42^{a}	3.88 ± 0.54^{b}	3.69 ± 0.58^{b}	4.33 ± 0.19^{c}
Cr (mg L^{-1})	BDL[e]	BDL	BDL	BDL	BDL
Cd (mg L^{-1})	BDL	BDL	BDL	BDL	BDL
Pb (mg L^{-1})	BDL	BDL	BDL	BDL	BDL
^{226}Ra (Bq m^{-3})	BDL	BDL	BDL	BDL	BDL
^{40}K (Bq m^{-3})	BDL	BDL	BDL	3.05 ± 0.01#	2.33 ± 0.08
^{232}Th (Bq m^{-3})	BDL	BDL	BDL	BDL	BDL

Values followed by the same letter are not significantly different (LSD, $p > 0.05$). *Sharp* (#) indicates the difference of ^{40}K concentration between sites 4 and 5. Mean ± Standard deviation ($n = 3$)

[e] *BDL* indicates below detectable limits

respectively, were determined in sediments of Pattani Bay in a previous investigation (Kaewtubtim et al. 2015). In that study, ^{226}Ra concentrations were higher than those of the present study (30.1–56.4 x).

Solubility, partitioning and redox reactions are considered primary factors influencing radionuclide mobility. Furthermore, increases in radionuclide contents via water column transportation depend upon type of radionuclide, soil/sediment porosity, redox state, and types and amounts of humic materials (Payne and Edis 2012; Tchokosssa et al. 2012). The decomposition rate of organic matter must also be taken into account in mangrove sediments because radionuclides are transported in sediment via plant uptake. Sediments allow for limited distribution of radionuclides at different depths as a function of the root length (Yanagisawa et al. 2000; Uchida 2007).

In sediment samples, values for average radium equivalent activity (Ra_{eq}), external hazard index, internal hazard index, absorbed dose rate in air, and annual effective outdoor dose rate were in the ranges of 57.4–143.7 Bq kg^{-1}, 0.2–0.4, 0.02–0.40, 26.1–62.3 nGy h^{-1}, and 0.03–0.08 mSv y^{-1}, respectively (Table 4). Values of Ra_{eq} were lower than standard values of 370 Bq kg^{-1} in sampling sites worldwide (UNSCEAR 2000). Other radiological risk assessment indices showed no potential internal or external radiation hazards for humans. The absorbed dose in air (D) values for sediments were lower than the global average value of 55 nGy h^{-1}, except for the sediment in sites 4 and 5 (56.8 and 62.3 nGy h^{-1}, respectively). The annual effective outdoor dose rate (E) in the sediments was within the acceptable levels provided by The International Commission for Radiological Protection of 1.0 mSv y^{-1}. Sediment in site 5 had the highest E value, which implies that this site must be monitored for potential future environmental and public health impacts (Amekudzie et al. 2011).

Concentrations of metals in the present study varied among those of other mangrove ecosystems worldwide (Table 5). The highest values for Cu, Ni, and Mn were recorded for Vaitarna estuary (India), the highest Zn concentrations in Sungai Puloh (Malaysia), and the highest Cr in Alibag (India). Differences in sediment metal concentrations are a function of the point source involved and the quantity of metal released. As indicated in the map of the study area (Fig. 1), the influence of human activities on metal and radionuclide contaminations must be taken into account, as the affected areas are located near industrial facilities and domestic dwellings. Such activities have been cited as primary causes of metal contamination in coastal zones (Duman and Kar 2012; Sundaray et al. 2012). Furthermore, rubber, fruit, and oil palm plantations located in the Pattani watershed may be minor sources of metals via their presence in phosphate fertilizers and fungicides. In this investigation, Cd and Pb concentrations exceeded world average values for shale (Table 1), as sediments in sites 4 and 5 had Cd concentrations >0.3 mg kg^{-1} and Pb concentrations >20 mg kg^{-1} in all sampling sites except for site 2 (Turekian and Wedepohl 1961). The average Pb content from all sampling sites (47.3 mg kg^{-1}) exceeded world average values for shale. Considering the sediment quality guidelines as proposed by Luo et al. (2010), Pb concentrations were categorized as moderately-polluted for sediment. Lead and Cd, even in low concentrations, can be toxic to both aquatic plants and animals (Galas-Gorcher 1991). The remaining metal concentrations were within world average concentrations of shale as proposed by Turekian and Wedepohl (1961).

Recent research conducted by Paiva et al. (2016) reported higher ^{226}Ra and ^{40}K concentrations in mangrove sediments in Chico Science (410 and 24 Bq kg^{-1} for ^{40}K and ^{226}Ra, respectively) and Rio Formoso (851 and 21 Bq kg^{-1} for ^{40}K and ^{226}Ra, respectively) in Brazil, as compared to data for this

Table 4 Average radium equivalent activity (Ra_{eq}), external hazard index, internal hazard index, absorbed dose rate in air, and annual effective outdoor dose rate determined in Pattani Bay sediments

Index	Site 1	Site 2	Site 3	Site 4	Site 5
Ra_{eq} (Bq kg^{-1})	69.12 ± 2.70^{b}	57.40 ± 0.24^{a}	95.07 ± 3.55^{c}	130.14 ± 1.27^{d}	143.65 ± 3.29^{e}
H_{ex}	0.19 ± 0.01^{b}	0.15 ± 0.00^{a}	0.26 ± 0.01^{c}	0.35 ± 0.00^{d}	0.39 ± 0.01^{e}
H_{in}	0.02 ± 0.01^{b}	0.16 ± 0.00^{a}	0.27 ± 0.01^{c}	0.37 ± 0.00^{d}	0.40 ± 0.01^{e}
D (nGy h^{-1})	30.71 ± 1.14^{b}	26.06 ± 0.11^{a}	42.29 ± 1.50^{c}	56.78 ± 0.60^{d}	62.34 ± 1.42^{e}
E (mSv y^{-1})	0.04 ± 0.00^{b}	0.03 ± 0.00^{a}	0.05 ± 0.00^{c}	0.07 ± 0.00^{d}	0.08 ± 0.00^{e}

Values followed by the same letter are not significantly different (LSD, $p > 0.05$). Mean ± Standard deviation ($n = 3$)

study (Table 5). Furthermore, all radionuclides had lower concentrations compared with those of global soil values of 32 Bq kg^{-1} for ^{226}Ra, 420 Bq kg^{-1} for ^{40}K, and 45 Bq kg^{-1} for ^{232}Th (UNSCEAR 2008). However, the average ^{232}Th activity concentration in this study was higher than that found in Wadies mouth and Safaga along the Red Sea coast of Egypt. Radionuclide activity concentrations may differ due to the effects of industrial activities; however, other point sources must be considered as minor sources, including fertilizers for palm and rubber tree plantations, clam harvesting, excavation of sediments for a deep-water port, and runoff from abandoned and active mines in nearby locations. The

Table 5 Comparison of the average heavy metal content and radionuclide activity in mangrove sediment and sediment quality guidelines worldwide

Location	Country	Heavy metal (mg kg^{-1})							Radionuclide (Bq kg^{-1})			Reference
		Cu	Zn	Ni	Mn	Cr	Cd	Pb	^{226}Ra	^{40}K	^{232}Th	
Sungai Puloh	Malaysia	46.9	1023.7	35.5	–	–	0.9	78.8	–	–	–	Udechukwu et al. (2015)
Sungai Buloh	Singapore	7.1	51.2	7.4	–	16.6	0.2	12.3	–	–	–	Cuong et al. (2005)
Sungai Khatib Bongsu	Singapore	32.0	120.2	11.7	–	32.1	0.3	31.0	–	–	–	Cuong et al. (2005)
Safaga coast, Red Sea	Egypt	12.7	45.5	11.5	444.7	–	0.1	16.2	14.3	346.5	17.1	Uosif et al. (2016)
Chao Phraya River	Thailand	0.7	1.4	–	–	–	0.02	0.7	–	–	–	Chaiyarat et al. (2013)
Tha Chin River	Thailand	0.6	1.9	–	–	–	0.02	1.2	–	–	–	Chaiyarat et al. (2013)
Mae Khlong River	Thailand	0.6	1.9	–	–	–	BDL	1.5	–	–	–	Chaiyarat et al. (2013)
Chico Science	Brazil	–	–	–	–	–	–	–	24.0	–	414.0	Paiva et al. (2016)
Rio Formoso	Brazil	–	–	–	–	–	–	–	21.0	–	869.0	Paiva et al. (2016)
El-Esh	Egypt	3.7	12.6	2.1	49.6	–	0.1	1.2	–	–	–	Mansour et al. (2013)
Wadi El-Hamra	Egypt	56.4	23.9	4.8	403.6	–	0.2	32.6	18.5	380.0	31.1	El-Taher and Madkour (2011)
Wadi El-Esh	Egypt	25.7	31.6	28.5	363.0	–	1.2	51.2	20.4	357.0	36.3	El-Taher and Madkour (2011)
Wadi Abu-Shaar	Egypt	14.0	19.7	26.2	319.2	–	1.2	26.7	24.2	418.0	35.6	El-Taher and Madkour (2011)
Wadi El-Gemal	Egypt	51.3	93.5	58.0	526.0	–	0.1	54.0	38.8	526.0	47.5	El-Taher and Madkour (2011)
Wadi Hamata	Egypt	12.9	24.3	6.5	454.0	–	0.3	31.9	35.1	491.0	42.5	El-Taher and Madkour (2011)
Cochin estuary	India	33.8	184.2	55.1	253.1	77.6	0.2	30.2	–	–	–	Ratheesh Kumar et al. (2010)
Gazi	Kenya	–	3.8	0.8	4.6	1.6	0.03	–	–	–	–	Okuku and Peter (2012)
Mikindani	Kenya	23.1	94.4	21.5	248.9	38.0	0.1	–	–	–	–	Okuku and Peter (2012)
Mtwapa	Kenya	20.7	80.5	14.6	150.5	37.2	0.1	–	–	–	–	Okuku and Peter (2012)
Chale	Kenya	2.2	11.4	6.3	20.7	9.6	BDL	–	–	–	–	Okuku and Peter (2012)
Msimbazi	Tanzania	20.5	106.9	15.0	185.6	17.7	0.2	–	–	–	–	Okuku and Peter (2012)
Vaitarna estuary	India	118.3	107.7	122.3	1614.3	–	–	–	–	–	–	Volvoikar and Nayak (2014)
Alibag	India	92.6	78.2	63.3	1020.1	545.2	5.3	–	–	–	–	Pahalawattaarachchi et al. (2009)
Nansha	China	113.0	159.0	48.4	880.0	155.0	0.8	–	–	–	–	Wu et al. (2014)
Santa Cruz	Brazil	3.5	BDL	7.7	70.2	28.6	BDL	6.3	–	–	–	Souza et al. (2015)
Serra	Brazil	5.4	BDL	14.3	59.1	52.6	BDL	7.9	–	–	–	Souza et al. (2015)
Santa Maria	Brazil	4.3	BDL	6.9	49.4	26.0	BDL	12.5	–	–	–	Souza et al. (2015)
Lameirão	Brazil	3.9	BDL	5.8	50.3	23.7	BDL	10.8	–	–	–	Souza et al. (2015)
Pattani Bay	Thailand	22.1	26.6	16.9	101.4	58.3	0.2	47.3	4.9	183.2	55.8	Present study
Average shale (metal)		45.0	95.0	68.0	850.0	90.0	0.3	20.0	–	–	–	Turekian and Wedepohl (1961)
Worldwide (radionuclide)		–	–	–	–	–	–	–	32.0	412.0	45.0	UNSCEAR (2008)
SQG non-polluted (metal)		<25.0	<90.0	<20.0	<300	<25.0	–	<40.0	–	–	–	Luo et al. (2010)
SQG moderately-polluted (metal)		25–75	90–200	20–50	300–500	25–75	–	40–60	–	–	–	Luo et al. (2010)
SQG heavily polluted (metal)		>50.0	>200.0	>50.0	>500	>75.0	>6.0	>60.0	–	–	–	Luo et al. (2010)

BDL below detection limits, "-"no data, *SQG* sediment quality guidelines

mangrove areas addressed in this study are considered as having low radionuclide-contaminated sediments.

Conclusion

Metal and radionuclide contamination in sediment and seawater pose major hazards for coastal biota and the coastal environment. In this study, the quantities of heavy metals and radionuclides in mangrove sediment ecosystem were relatively low; thus, there is a minimal risk of food web bioaccumulation. The geoaccumulation index (I_{geo}) and enrichment factor (EF) were used to assess the effect of metal pollution in surface sediments due to anthropogenic inputs. Concentrations of several metals, especially Cd and Pb should be monitored often, as they are toxic to biota even at low accumulation rates in tissue. In addition, metal concentrations found in the sampling sites, particularly Pb and Cd in sites 1 and 5, respectively, were considered to result from anthropogenic activities (presumably mining, industrial, and agricultural activities). The higher levels of radionuclides may be due to movement of fine and particulate sediments in the water column; this is particularly likely in sites 4 and 5 which are located near the river mouth. It is believed that mining, industrial, and agricultural inputs served as key point sources of radionuclide contaminants in those sites. Furthermore, natural phenomena such as river flow, intense rainfall events, and sediment resuspension may increase disturbances on suspended sediments, and dissolved materials in the water column thereby increasing radionuclide concentrations. Most of the radiological risk assessment indices had relatively low values, except for the D value in sites 4 and 5 that exceeded the standard recommended value of 55 nGy h^{-1}. It is important, therefore, that metal and radionuclide monitoring continues in these locations.

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