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ORIGINAL ARTICLE

Environmental Radioactivity in the Marine Environment of Ha Long Bay, North Vietnam, and Biomagnification of Polonium

Fernando P. Carvalho[1] · Dang Duc Nhan[2] · João M. Oliveira[1] · Nguyen Quang Long[2] · Dao Dinh Thuan[3] · Margarida Malta[1] · Marta Santos[1]

Received: 13 July 2021 / Accepted: 29 September 2021

Abstract

Environmental preservation of Ha Long Bay, a UNESCO World Heritage Site, is a national priority for Vietnam. Coal mining is growing in the region with interim coal stockpiling on the shore by the maritime harbour in Ha Long Bay. Due to the presence of radionuclides in coal, radioactivity analyses were carried out for the first time in coastal samples to assess the radionuclide levels in Ha Long Bay. Samples of coal, sediments, molluscs, shrimp, and fish were collected from the Bay site and analyzed for U, Th, ^{226}Ra, ^{210}Pb and ^{210}Po by alpha spectrometry, and for ^{40}K, ^{228}Ra and ^{137}Cs by gamma spectrometry. Radionuclide concentrations in sediments ranged from 21 to 43 Bq kg^{-1} dry weight (dw) for ^{238}U, from 14 to 35 Bq kg^{-1} dw for ^{226}Ra, and from 25 to 122 Bq kg^{-1} dw for ^{210}Pb and ^{210}Po. Radionuclide concentrations in marine biota were comparable to data reported for other coastal areas. It was concluded that despite discharges of the long-lasting coal mining industry in the area, no significant enhancement of radioactivity occurred in biota of Ha Long Bay. Although at naturally-occurring levels, ^{210}Po concentrations in biota were remarkably higher than concentrations of other radionuclides and showed a clear trend of biomagnification with increasing trophic levels, contrasting with the discrimination of U, Th and other radionuclides in the marine food chain. Radionuclides of artificial origin, namely ^{137}Cs, were present at very low level. Current radionuclide levels in marine biota do not represent a radiation hazard for sea food consumers.

Highlights

- **Coal mines discharges into the Bay enhanced U and Th levels in sediments**
- **Radioactivity in marine biota from the Bay was comparable to biota from other seas.**

✉ Fernando P. Carvalho
carvalho@ctn.tecnico.ulisboa.pt

[1] Laboratório de Protecção e Segurança Radiológica/Instituto Superior Técnico, Universidade de Lisboa, 2695-066 Bobadela LRS, Portugal

[2] Institute for Nuclear Sciences and Technologies, 179 Hoang Quocs Viet, Ha Noi, Viet Nam

[3] Ha Noi University of Mining and Geology, Dong Ngac, Tu Liem, Ha Noi, Viet Nam

Published online: 11 October 2021

- **Po was biomagnified in the marine food chain while U, Th, Ra, Pb were discriminated.**

Keywords Coal · Naturally occurring radionuclides · Marine biota

1 Introduction

Coastal seas are the most productive zones of the marine environment and support the development of large biomasses, including those of the species most contributing for the fishery catches. Coastal areas are also the most affected by anthropogenic activities and often receive waste discharges from cities and land-based industries while, at the same time, are intensively used for tourism, recreation, and fisheries (Sheppard 2019).

Ha Long Bay in the Quang Ninh province, North of Vietnam, is an example of such a coastal area with multiple uses and impacts. The Bay area is vast (1,553 km^2) and provides for wealthy fisheries, booming shrimp farms, several developing industries, intensive tourism activities, and a growing human population living around the Bay (Giuliani et al. 2019; Le Tuan Anh 2015). Owing to the scenic value of the near 2000 limestone islands and islet formations existing at the sea, this Bay is a UNESCO World Heritage Site attracting a growing number of tourists every year. For all these reasons the assessment of the status of the environment for protection of the bay is a concern of the authorities and a priority for the country (Le Tuan Anh 2015; Mustafin et al. 2019).

Around the bay, land-based industries such as coal mining and cement manufacturing, both using sea harbors for shipping their production, may potentially increase radioactivity levels through discharges of naturally occurring radioactive materials (NORM) into the Bay.

Coal mining at Hon Gai, near Cam Pha city, started about one hundred years ago. Due to the dependence of Vietnam on coal for internal energy production and for fossil fuel export, coal mining in this area duplicated in the last 30 years and continues to grow (Vu Dinh-Hieu et al. 2012). Currently, there are 30 active coal mines in the Quang Ninh province that bear about 95 % of the coal production of Vietnam, of about 42.5 Mtons per year. The landscape around the Bay is nowadays deeply marked by the mining tailings and by the nearshore coal stockpiles waiting to be transferred to the ships. The mines and coal piles are sources of mine water discharges and surface runoff into the marine environment, respectively. Every year, Cam Pha mines consume water estimated over 2.2 million m^3 and discharge about 9 million m^3 of mining water into the sea. Cam Pha city stores the biggest quantity of sludge from the mines, and receives further 60 to 70 million cubic meters of mine sludge each year (EcoWatch 2015). In some countries, reports have shown that water discharges and sludge from coal mines are a source of enhanced concentrations of naturally-occurring radionuclides, especially radium (^{226}Ra), as observed in the coal mines of Poland and in their surrounding environment (Pociask-Karteczka et al. 1997; Wysocka et al. 2019).

Cement production near Cam Pha is based on local extraction of limestone. Although limestone generally displays lower levels of radioactivity than other rocks, the production of clinker in high temperature furnaces can be a mechanism of volatilization and reconcentration of some natural radionuclides, in particular ^{210}Po and ^{226}Ra. This was reported, for example, in ceramic furnaces (Zampieri et al. 2008), and these radionuclides may enter the

marine environment if particulate materials from the kilns and furnaces have been included in process water discharges.

In Quang Ninh province, other mining activities are carried out, such as gold and uranium mining, although located in more inland locations and with no direct coastal discharges (Vu Dinh Hieu et al. 2012). However, occasionally, intense rainfall has flooded mines and made tailing dams to collapse allowing sludge to flow into the bay, as happened several times in Cam Pha area, in 2015 and before (ABC News 2015).

The presence of radionuclides of U and Th radioactive decay series in coal has been known for decades and in the past U was recuperated from some coal deposits (Dai and Finkelman 2018). Therefore, coal production and coal burning may contribute to the enhancement of environmental radioactivity levels through discharges of effluents. The IAEA and other international organizations such as the European Union, recommend the evaluation of radioactivity levels from industries processing NORM in order to assess the need for radiation protection measures (EU 2013; IAEA 2014; Michalik 2017). In the region around the Bay, there are no nuclear facilities, and therefore, no environmental discharges of artificial radionuclides. However, global environmental radioactive contamination originated by nuclear tests and nuclear accidents, through atmospheric transport pathways may have delivered to this area long-lived artificial radionuclides, such as ^{137}Cs, in non-reported amounts.

The assessment of radioactivity levels in the marine environment of Ha Long Bay has not been attempted before. Because of long lasting coal mining and discharges into the bay, there were concerns with potential impacts of NORM on the quality of sea food from the bay and on tourism development in the region. Therefore, this investigation was considered timely.

2 Materials and Methods

Sampling of the north part of Ha Long Bay, along the coast in the Quang Ninh province, was carried out in 2010. Figure 1 shows the location of sediment and biota sampling stations, along with the main features of the region.

A large sample (5 kg) of freshly extracted coal was collected at Hon Gai mine through combining coal portions from several piles, in order to obtain a more representative sample. The coal was analyzed to assess this fossil fuel as a potential source of natural radionuclides discharged into the coastal environment.

Sediment sampling was carried out along the shore during low tide, and in the bay with the use of a local fishing boat. Samples were collected with a small hand operated sediment sampler, taking bulk samples of the upper 15 cm of the seafloor. Samples were transferred into plastic bags and duly identified for later processing in the laboratory.

Biota samples were obtained with the help of local fishermen. Species collected included: the Peanut worm *(*Family *Sipunculidae, Sipunculus nudus)*, a local delicacy that is collected from soft muddy bottoms during low tide; bivalve molluscs, comprising the Blood clam (Family *Arcidae, Anadara granosa*) and the Asiatic hard clam (Family *Veneridae*, *Meretrix meretrix*), oysters (Family *Ostreidae*, *Cassostrea gigas*), and the shrimp or Tiger prawn (Family *Penaeidea*, *Penaeus monodon*); and a common pelagic fish, the round Scad (Family *Carangidae, Decapterus punctata*), a planktivorous fish.

In the laboratory, all sediment samples were sieved through a 1 mm stainless steel sieve and dried in the oven at 95 ^{o}C. Later, following disaggregation of samples and

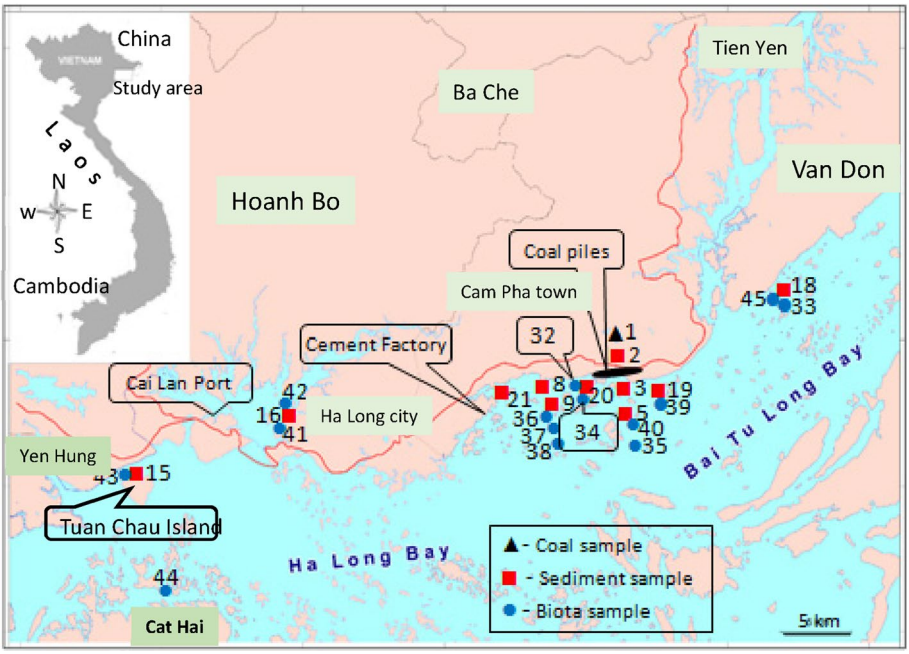

Fig. 1 Ha Long Bay, in North Vietnam, and location of sampling stations. Station 1 corresponds to the Hong Cai coal mine

homogenization, portions of sieved sediment samples were used for gamma spectrometry. Sediments were packed in dense polyethylene 100 mL cylindrical boxes leaving no head space, sealed and stored for over a month to allow formation of radioactive equilibrium between ^{226}Ra and ^{226}Ra daughters before measurements.

Portions of the 1 mm sieved sediment samples were dry sieved again to remove sand and the grain size fraction less than 63 μm was used for the determination of radionuclides by alpha spectrometry. As the sample size for alpha spectrometry is of around 1 g, removal of sand is needed to ensure representativeness of samples and to allow intersample comparisons (Carvalho 1995).

Aliquots of the 1 mm sieved sediment samples, after determination of sediment dry weight, were used to determine the weight loss on ignition at 500 °C. This determination was performed in triplicate aliquots and the relative standard error of replicates was <1.2 % of the average value.

Biota samples were dissected carefully, washed to remove all sediment materials, and freeze dried. The edible parts were analyzed only, as follows: the peanut worm was analyzed *in toto*; from bivalve molluscs were analyzed the soft tissues; shrimp was analyzed for meat (muscle plus hepatopancreas); and from fish, only the fish muscle (filet) was analyzed.

Radionuclides were determined by alpha spectrometry and by gamma spectrometry. Analyses of alpha particle emitting radionuclides were performed by radiochemistry and alpha spectrometry using well established procedures. In brief, aliquots of the homogenized < 63 μm fraction of samples were used for the determination of ^{238}U, ^{235}U, ^{234}U, ^{232}Th, ^{230}Th, ^{226}Ra, ^{210}Po and ^{210}Pb. Pb-210 in sediments was determined through analysis of ^{210}Po made more than 1 year after the sample collection, therefore, in ensured secular radioactive equilibrium.

In biota, ^{210}Po and ^{210}Pb were determined through a first ^{210}Po plating soon after sample collection, and a second ^{210}Po plating more than 6 months later for ^{210}Po ingrowth from ^{210}Pb radioactive decay. Appropriate decay and ingrowth corrections were applied in the calculations (Carvalho and Oliveira 2007, 2009; Oliveira and Carvalho 2006). All the radionuclides were determined applying a sequential radiochemical separation of radionuclides, followed by their electrodeposition on metal planchets and radiation measurement by alpha spectrometry using OCTETEplus spectrometers from ORTEC, with 450 mm^2 active surface ULTRA AS Mark detectors. The Maestro software from ORTEC was used in spectrum analysis. The procedures followed, including the analytical quality control, were validated and are described in detail in published reports (Carvalho and Oliveira 2007, 2009; Oliveira and Carvalho 2006). The analytical quality control included the use of certified reference materials produced by the International Atomic Energy Agency (IAEA), such as the IAEA-384 (Lagoon sediment), IAEA-412 (Pacific Ocean Sediment), IAEA-446 (Baltic Sea Seaweed), and the IAEA-414 (Mixed fish from the Irish Sea and North Sea).

Gamma-ray spectrometry was applied for the determination of activity concentrations of radionuclides ^{40} K, ^{226}Ra, ^{228}Ra and ^{137}Cs. The acquisition time was set to 15 h and the photo peaks used for the activity determination were: 1460.82 keV for ^{40} K; 295.2, 351.9 and 609.3 keV for ^{226}Ra; 238.63, 583.19 and 911.20 keV for ^{228}Ra; and 661.70 keV for^{137}Cs. A 50% relative efficiency broad energy HPGe detector (Canberra BEGe model BE5030), with an active volume of 150 cm^3 and a carbon window, was used for the gamma spectrometry measurements. A thick lead shield, with internal lining of copper and tin protects the detector from the environmental radiation background. Standard nuclear electronics was used for signal treatment and the software Genie 2000 (version 3.4) was employed for the spectrometric data acquisition and spectral analysis. The detection efficiency was determined using NIST-traceable multi-gamma radioactive standards (POLATOM Laboratory of Radioactivity Standards) with an energy range from 46.5 to 1836 keV and customized in a water-equivalent epoxy resin matrix (density of 1.15 g cm^{-3}) to exactly reproduce the geometries of samples in the sample containers used. GESPECOR software (version 4.2) was used to correct for matrix (self-attenuation) and coincidence summing effects, as well as to calculate the efficiency transfer factors from the calibration geometry to the measurement geometry (whenever needed). The stability of the system (activity, FWHM, centroid) was checked at least once a week with a ^{152}Eu certified point source. This gamma spectrometry technique is accredited according to the ISO/IEC 17,025:2005 standards.

Internal Analytical Quality control was applied through the analysis of IAEA Certified Reference Materials (CRM) with each batch of samples, such as the IAEA-384 (Lagoon sediment), IAEA-412 (Pacific Ocean Sediment), IAEA-446 (Baltic Sea Seaweed), and the IAEA-414 (Mixed fish from the Irish Sea and North Sea). External Analytical Quality control was ensured through periodic participation in International Atomic Energy Agency (IAEA) inter comparison exercises (blind tests), with good results (e.g., Povinec et al. 2007; Pham et al. 2014, 2016).

The activity concentrations of radionuclides are expressed in Bq kg^{-1} (dw) with the calculated expanded uncertainty.

3 Results and Discussion

3.1 Radionuclides in Coal and Sediments

The activity concentrations of naturally occurring radionuclides determined by alpha spectrometry in sediments are shown in Table 1. Table 1 includes the results of analysis of coal (sample #1) freshly extracted from the Hong Gai mine. Radionuclide concentrations in the coal showed, within the experimental uncertainty, a secular radioactive equilibrium between uranium (^{238}U) and uranium daughters. This radioactive equilibrium indicates long-term geochemical stability of the coal deposit and minor water perfusion through the coal.

The ^{238}U activity concentration in the Hong Gai coal, 41.0±2.3 Bq kg^{-1}, corresponds to 84 Bq kg^{-1} of natural uranium (Unat) or 3.3 ppm (mg kg^{-1}) of U which is close to the U average concentration reported for world coals, i.e., 2.4 ppm (equivalent to 60 Bq kg^{-1} Unat) (Dai and Finkelman 2018). Notwithstanding, many coal deposits display much higher U concentrations up to 7200 ppm, as for example the coal from Ghizou province, PR China (Dai and Finkelman 2018; Prachiti et al. 2011; USGS 1997; Wang et al. 2015).

The ^{226}Ra activity concentration in the Hong Gai coal sample, 57.4±11.9 Bq kg^{-1}, was in the range of ^{226}Ra concentrations reported for hard coal worldwide (0 - 121 Bq kg^{-1}). However, it was higher than the world average ^{226}Ra concentration in coal (20 Bq kg^{-1}), and also higher than the average ^{226}Ra concentration in the Earth's crust (25 Bq kg^{-1}) (Wysocka et al. 2019). For comparison also, ^{226}Ra concentration in coal from Hong Gai mine was much higher than the average ^{226}Ra concentration in coal from Poland (17 Bq kg^{-1}). In Poland, despite the relatively low concentration of ^{226}Ra in coal, some mines are known for the discharge of high ^{226}Ra activity dissolved from coal layers by groundwater (hundreds of MBq per day in mine water discharges). These discharges increased radium levels in the Vistula river basin and required discharge control and radiation protection measures (Pociask-Karteczka et al. 1997; Wysocka et al. 2019).

A similar comment can be made about ^{228}Ra, daughter from ^{232}Th, whose activity concentration in worldwide hard coal averages 20 Bq kg^{-1} (range 0-105 Bq kg^{-1}), while in coal from Poland the average concentration is 11 Bq kg^{-1} and in the Earth crust is 25 Bq kg^{-1} (Pociask-Karteczka et al. 1997). In the coal from Hong Gai mine, assuming radioactive equilibrium with ^{232}Th, the ^{228}Ra concentration was 45.6±2.3 Bq kg^{-1} (Table 1).

Because of the transport of coal particles by surface runoff and river discharges into Ha Long Bay, the whitish sediments from the bay, especially those collected near the shore displayed a visible contamination with black coal. Table 1 shows the results of % weight loss on ignition for coal and sediment samples. The weight loss of coal, 66.5 %, corresponds to the combustible organic matter in the coal; hence, the mineral ash residue was 33.5 % of coal dry weigh. The weight loss of sample #2 was 35.6 % of the dry weight, meaning that a large fraction of this sediment sample was indeed coal. In contrast to this sediment, in most offshore samples the % weight loss on ignition was lower and fluctuated from about 4–16 %, being the combustible material probably a mixture of some coal particles and organic matter from marine origin in undetermined proportions. A map of coal distribution in the Bay sediments is to be made yet.

The Hong Gai coal sample contained higher concentration of ^{238}U, 41.0±4.6 Bq kg^{-1}, than most sediment samples from the bay. These sediments contained uranium (^{238}U) with activity concentrations averaging 33±7 Bq kg^{-1} (range 21-43 Bq kg^{-1}), i.e., in average the sediments contained 20 % less ^{238}U than the coal (Fig. 2). In sediments,

Table 1 Activity concentration of radionuclides (Bq kg^{-1} dry weight ± 2σ) in coal and surface sediments of Ha Long Bay, determined by alpha spectrometry

Sample number	Sample material	% Weight loss on ignition	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{232}Th
#1	Coal	66.5	41.0 ± 2.3	2.0 ± 0.4	40.8 ± 2.3	46.7 ± 2.4	57.4 ± 11.9	36.7 ± 2.4	45.6 ± 2.3
#2	Sediment	35.6	36.7 ± 1.4	1.9 ± 0.3	38.0 ± 1.5	101 ± 21	24.4 ± 4.8	24.9 ± 3.4	52.6 ± 15.3
#3	Sediment	6.6	33.4 ± 1.3	1.9 ± 0.3	31.5 ± 1.2	39.5 ± 2.2	31.0 ± 8.0	76.1 ± 4.3	47.0 ± 2.5
#5	Sediment	9.6	43.1 ± 1.6	2.1 ± 0.3	43.6 ± 1.6	85.9 ± 5.2	25.8 ± 17.4	122 ± 5	79.9 ± 5.1
#8	Sediment	9.7	34.8 ± 1.1	1.4 ± 0.2	33.1 ± 1.1	44.2 ± 2.4	22.3 ± 8.3	74.0 ± 2.8	62.2 ± 2.8
#9	Sediment	4.7	24.9 ± 0.9	1.0 ± 0.2	23.2 ± 0.8	32.9 ± 1.7	13.5 ± 4.0	55.7 ± 3.0	32.9 ± 1.7
#15	Sediment	7.4	33.7 ± 1.8	1.6 ± 0.3	32.4 ± 1.8	147 ± 9	22.8 ± 3.1	59.4 ± 3.0	47.8 ± 4.9
#16	Sediment	16.0	41.8 ± 1.5	1.7 ± 0.3	39.7 ± 1.4	157 ± 7	28.7 ± 4.5	51.5 ± 2.1	74.8 ± 4.1
#18	Sediment	5.2	21.5 ± 0.9	1.0 ± 0.2	22.1 ± 0.9	111 ± 3	15.5 ± 2.6	44.2 ± 1.8	31.6 ± 1.2
#19	Sediment	11.1	40.0 ± 1.5	1.8 ± 0.3	39.9 ± 1.5	43.5 ± 2.6	35.2 ± 8.8	55.9 ± 2.7	60.3 ± 3.1
#20	Sediment	4.2	20.7 ± 1.0	0.8 ± 0.2	21.4 ± 1.0	63.3 ± 2.5	23.4 ± 5.3	32.9 ± 1.9	23.7 ± 1.4
#21	Sediment	4.0	34.6 ± 1.2	1.2 ± 0.2	31.7 ± 1.1	63.4 ± 3.6	32.6 ± 6.1	46.7 ± 2.1	55.0 ± 3.2
Sediments:	Average		33	1.5	32	81	25	58	52
	SD		7	0.4	7	41	6	25	17
	Min	4.0	21	0.8	21	33	14	25	24
	Max	35.6	43	2.1	44	157	35	122	80

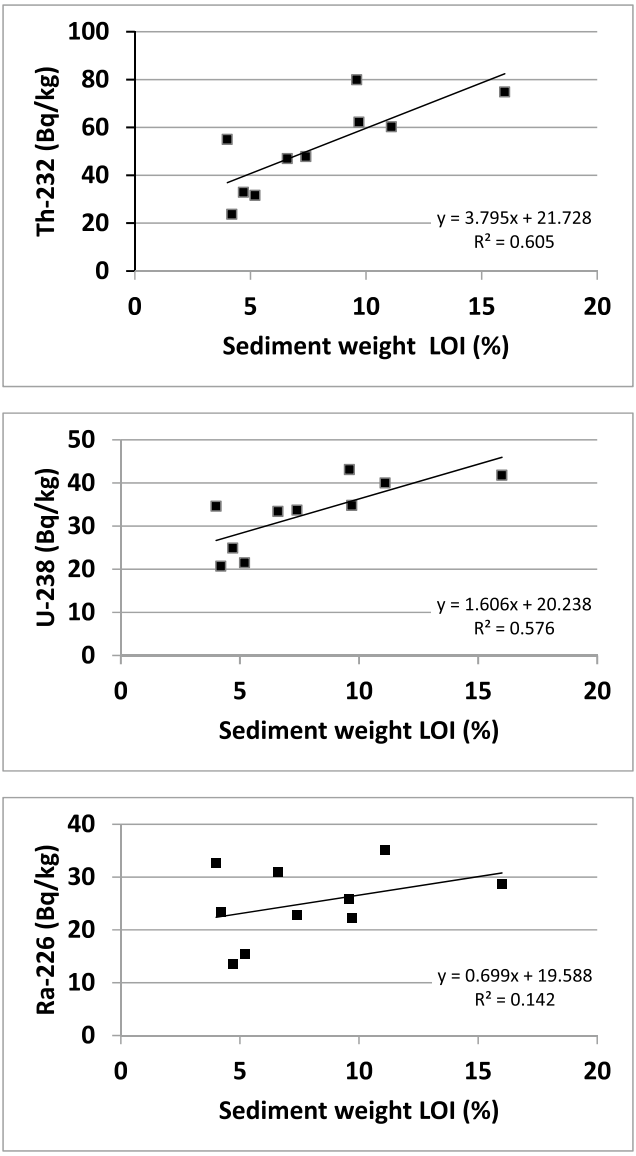

Fig. 2 Correlation between radionuclide concentrations (Bq kg^{-1} dw) in Ha Long Bay sediments and sediment weight loss on ignition (%). As per data in Table 1 (n=10; one outlier excluded)

the average concentrations of the ^{238}U descendants ^{230}Th, ^{226}Ra, and ^{210}Pb were at 81±41 Bq kg^{-1}, 25±6 Bq kg^{-1}, and 58±25 Bq kg^{-1}, respectively. These values indicate departure from radioactive secular equilibrium with reduction of ^{226}Ra concentration in sediments because of the high radium solubility in seawater, while ^{230}Th and ^{210}Pb radionuclides, both much less water-soluble than radium, remained in the solid phase and accumulated in sediments (Atwood 2010).

It is relevant to note also that ^{232}Th, the precursor of thorium radioactive decay series, was present in sediments in activity concentrations averaging 52±17 Bq kg^{-1} (24-80 Bq kg^{-1}), thus in activity concentrations slightly higher than in the coal, 45.6±2.3 Bq kg^{-1}, and at a level unusually high for carbonate sediments (Ivanovich and Harmon 1992; Carvalho et al. 2017). As the geology of the bay region is a karstic formation, most marine sediments are derived from limestone (calcium carbonate). Radioactivity levels in limestone are known to be generally much lower than in other rocks, such as shale, schist and granite (Ivanovich and Harmon 1992; Carvalho et al. 2017) and the source of this enhanced ^{232}Th concentration in the Bay is attributed to sedimentation of coal particles.

The sediment samples from offshore, such as samples #18 and #20, contained a minimal contamination with coal (approximately 4-5 % weight loss on ignition), and simultaneously displayed the lowest concentrations of thorium, uranium and radium. Sediments with higher % weight loss on ignition displayed slightly elevated concentrations of these radionuclides, suggesting that coal was the source of the additional radioactivity to sediments.

Attempts were made to establish correlations between radionuclide concentrations and % sediment weight loss on ignition (LOI). These correlations were statistically significant for ^{232}Th ($p<0.01$) and also for ^{238}U ($p<0.01$), while ^{226}Ra, ^{210}Pb and ^{230}Th were poorly correlated with % weight LOI ($p<0.20$) (Fig. 2). Equations of the best-fit correlation lines indicate a y-axis intercept at approximately 20 Bq kg^{-1}, which would be the likely natural radionuclide content of carbonate sediment without organic matter and coal contamination (Fig. 2). Notwithstanding, because of the hydraulic sorting of coal particles in the Bay, complex chemical behavior and water solubility of radionuclides from coal, and presence of variable amounts of organic matter from other origins than coal, the correlations of radionuclide concentrations with LOI are of limited significance. Furthermore, in coal ores the U generally is associated with the organic matter, while Th is mostly associated with refractory minerals which may render more complex the redistribution of radionuclides in the marine environment (Dai and Finkelman 2018; Prachiti et al. 2011).

Both ^{232}Th and ^{238}U concentrations in sediments indicated a minor, but still detectable enhancement of these primordial radionuclides likely related with coal particle and mine water discharges into the Bay (Fig. 2). Thorium-232 (and ^{230}Th from uranium series) present in coal discharges remained in the solid phase because thorium is water insoluble, while a fraction of uranium and a larger fraction of radium from coal gradually dissolved in seawater. Such as in the coal, the ^{234}U activity concentrations in sediments were nearly identical to those of ^{238}U (Table 1). The ^{235}U, precursor of actinium series, was represented in sediments with a much lower activity concentration than the uranium isotope ^{238}U, as expected, and averaging 1.5 Bq kg^{-1} (range 0.8-2.1 Bq kg^{-1}) (Table 1). The average activity concentration ratios of uranium isotopes in Bay sediments were ^{234}U/^{238}U=0.970 (≃1, within the experimental uncertainty) and ^{235}U/^{238}U= 0.045, corresponding to the isotopic ratios of natural uranium (Magill and Galy 2005).

On land, around the Ha Long Bay area, results of a radon (^{222}Rn) survey made in Quang Ninh buildings did show relatively low radon concentrations indoors (Nhan et al. 2012). Based on those results, relatively low levels of uranium series radionuclides could be expected in soils and rocks in this area. However, this expectation could not be extended to the sea floor because of the coal mine discharges into the bay. Sampling and analysis performed in this study allowed to confirm now the low radioactivity levels inside the bay.

Although we consider the discharge of coal particles in surface runoff and mine water discharges as the primary pathway for potential radioactivity enhancement in the Bay, an atmospheric transport pathway may also exist. The Tonkin Gulf, in which Ha Long Bay is located, is a hot spot area for black carbon (BC) particulates in the atmosphere, >3 μg BC

m^{-3}, derived from coal and biomass burning in the region (Mari et al. 2019). These BC particles enter the sea with wet and dry atmospheric depositions and, although their mass is minimal, they may carry contaminants and contribute to enhance their concentrations in sediments. Black carbon and soot from coal burning, and fly ashes from biomass combustion usually carry enhanced concentrations of natural radionuclides, especially ^{210}Po and ^{210}Pb which add up to the ^{210}Pb deposition flux from atmospheric radon decay (Carvalho et al. 2017). These atmospheric depositions, particularly the ^{210}Pb deposition flux from decay of atmospheric radon, could explain the excess of ^{210}Pb in bay sediments whose average concentration was approximately twofold the ^{238}U and ^{234}U levels (Table 1).

Several radionuclides were determined by gamma spectrometry in a selection of sediment samples (Table 2). The most abundant radionuclide was potassium-40 (^{40} K), whose concentration in sediments averaged 424±22 Bq kg^{-1}. Ra-226 from uranium series and ^{228}Ra from thorium series matched the range of concentrations determined by alpha spectrometry (Table 1) and ^{228}Ra confirmed the higher activity concentration of thorium series radionuclides in comparison with uranium series radionuclides in sediments. Despite the good agreement between alpha and gamma spectrometry results, it must be noted that results for radionuclide concentrations strictly cannot be directly compared because of the difference in sediment grain size cut off used in sample preparation. Cesium -137 in sediments was below detection limits at very low concentrations (<1.3 Bq kg^{-1}). Other measurements of ^{137}Cs in marine sediments from the entire coast of Vietnam, including this study area, confirmed these low levels for the year 2010, with concentrations ranging from 0.51 to 2.62 Bq/kg (dw) (Quang Long et al. 2011). These values were cesium-137 levels prior to the Fukushima nuclear accident and, since then, might have increased following the radioactivity releases into the environment originated by that nuclear accident in March 2011.

3.2 Radionuclides in Biota

Results of biota analysis are shown in Table 3. In the Bay, there was no ubiquitous mollusc species, such as mussels or oysters for example, that could be used as a bioindicator for synoptic assessment of radionuclide levels in the Bay area. The distribution of bivalve mollusc species was uneven and different species were available in mollusc banks.

Table 2 Activity concentrations of radionuclides (Bq kg^{-1} dry weight ± 2σ) determined by gamma spectrometry in samples from Ha Long Bay

Sample number	Sample material	^{40}K	^{137}Cs	^{226}Ra	^{228}Ra
#3	Sediment	455 ±55	<1.1	28.3±4.4	53.9±7.9
#15	Sediment	403±51	<1.0	31±4.3	47.7±6.5
#18	Sediment	414±54	<1.3	25.5±3.9	48.6±7.1
#45	Peanut worm	219±31	<7.3	<16	5.0±0.6
#43+#44	Oysters	249±81	<5.0	<15	<10
#41+#42	Blood clam	113±58	<4.3	<12	<7.0
#36+#37+#38	Asiatic hard clam	255±38	<2.2	<3.2	4.5±2.3
#39	Tiger prawn	227±105	<8.8	<21	<13
#40	Fish (filet)	343±84	<5.8	<12	<7.5

< x, lower than the detection limit x

Table 3 Activity concentration of radionuclides (Bq kg^{-1} dry weight ± 2σ) in biota samples from Ha Long Bay determined by alpha spectrometry

Sample	Species	Site	D/W	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
#45	Peanut worm	Van Don	0.15	90.5 ± 2.5	4.0 ± 0.2	103 ± 3	3.7 ± 0.2	18.6 ± 2.5	15.9±0.9	21.6±0.8	5.6 ± 0.4
#39	Tiger prawn	Bai tu long Bay	0.23	0.45 ± 0.05	0.02 ± 0.01	0.48 ± 0.05	0.21 ± 0.02	1.2 ± 0.1	9.1±0.6	421.6±14.4	0.33 ± 0.02
#40	Fish (filet)	Bai tu long Bay	0.25	0.044±0.006	0.006±0.004	0.08 ± 0.01	1.0 ± 0.3	0.84 ± 0.08	2.0±0.2	17.7±0.8	0.5 ± 0.2
Bivalve molluscs:											
#32	Blood clam	Bem Do	0.17	4.2 ± 0.2	0.24 ± 0.03	4.9 ± 0.2	1.5 ± 0.1	1.4 ± 0.2	14.3±1.0	177.3±7.4	3.0 ± 0.2
#33	Blood clam	Van Don	0.15	5.2 ± 0.2	0.21 ± 0.03	5.5 ± 0.2	1.2 ± 0.1	0.9 ± 0.1	8.7±0.8	274.8±10.5	2.1 ± 0.2
#34	Asiatic hard clam	Ben Do	0.13	6.4 ± 0.2	0.30 ± 0.04	7.6 ± 0.2	1.3 ± 0.1	4.7 ± 1.1	28.4±1.2	669.8±26.4	2.0 ± 0.1
#35	Blood clam	Bai tu long Bay	0.17	2.30 ± 0.08	0.12 ± 0.02	2.46 ± 0.09	0.38 ± 0.05	1.2 ± 0.2	1.2±0.1	84.7±2.7	0.52 ± 0.06
#36	Asiatic hard clam	Bai tu long Bay	0.15	2.38 ± 0.07	0.12 ± 0.01	3.00 ± 0.08	0.90 ± 0.06	2.1 ± 0.3	15.9±1.0	184.3±10.6	1.51 ± 0.09
#37	Asiatic hard clam	Bai tu long Bay	0.15	1.55 ± 0.06	0.07 ± 0.01	1.79 ± 0.06	0.5 ± 0.1	0.6 ± 0.1	2.0±0.1	54.2±1.6	0.31 ± 0.08
#38	Asiatic hard clam	Bai tu long Bay	0.13	1.69 ± 0.04	0.082±0.008	1.94 ± 0.05	0.6 ± 0.2	0.16 ± 0.02	196.8±8.5	326.5±12.8	0.31 ± 0.15
#41	Blood clam	Port Cai Lan	0.17	4.4 ± 0.1	0.22 ± 0.02	5.3 ± 0.1	0.42 ± 0.02	2.9 ± 0.5	4.3±0.4	138.2±4.4	0.87 ± 0.04
#42	Blood clam	Cai Lan	0.16	5.3 ± 0.1	0.26 ± 0.02	5.8 ± 0.1	1.47 ± 0.06	3.1 ± 0.5	8.2±0.8	186.8±6.7	2.63 ± 0.09
#43	Oyster	Tuan Chan		10.1 ± 0.3	0.48 ± 0.05	11.2 ± 0.3	0.35 ± 0.03	0.36 ± 0.04	3.3±0.4	384.5±15.8	0.62 ± 0.04

Table 3 (continued)

Sample	Species	Site	D/W	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
#44	Oyster	Cat Hai	0.13	1.96 ± 0.07	0.08 ± 0.01	2.21 ± 0.07	0.7 ± 0.2	0.028 ± 0.005	2.8±0.2	242.3±4.4	< 0.5
All bivalves:		Average		4.10	0.20	4.70	0.80	1.60	9.9	248	1.3
		SD		2.50	0.10	2.70	0.40	1.40	8.3	163	0.9
		Min	0.13	1.55	0.07	1.79	0.35	0.03	1.2	54	0.3
		Max	0.25	10.10	0.48	11.20	1.50	4.70	28.4	670	3

D/W= Dry /wet weight ratio

Notwithstanding, as all were sediment dwelling and deposit feeders, the activity concentrations turned out to be similar in different species (Table 3). Soft tissues of these molluscs contained low concentrations of uranium, averaging 4.1±2.5 Bq kg^{-1}, followed by ^{226}Ra with 1.6±1.4 Bq kg^{-1}, and ^{232}Th with 1.3±0.9 Bq kg^{-1}. These values contrast with the most accumulated radionuclides, namely ^{210}Pb, with average activity concentration of 9.9±8.3 Bq kg^{-1}, and ^{210}Po, with 248±163 Bq kg^{-1}. Likewise, the activity concentration of polonium was similarly high in shrimp and molluscs while in the fish muscle and peanut worm it was very low (Table 3).

Radionuclide concentrations in biota from Ha Long Bay, namely those of bivalve molluscs, crustacean and fish, were in the range of concentrations reported for similar species from other coastal seas and are not exceptional (Table 4).

The main source of uranium, radium, and radioactive lead for marine biota generally is sea water, and bioaccumulation takes place by radionuclide absorption through the gut and gills. For polonium, the main source is the food and the transfer pathway is food ingestion (Carvalho 2011, 2018; Carvalho et al. 2017). Generally, for biota such as the species sampled from Ha Long Bay, radionuclides bound to sediment are not readily available for bioaccumulation. However, sediments are the larger reservoir of natural radionuclides in the ecosystem and the one that maintains radionuclide concentrations in seawater nearly constant through a dynamic equilibrium of ion exchanges between sediment and water (IAEA 2010).

It is interesting to note that the soft tissues of sediment-dwelling bivalve molluscs from Ha Long Bay, displayed activity concentrations of U (both ^{238}U and ^{234}U) higher than ^{226}Ra, and these ones were nearly similar to Th concentrations. This is not the case in other coastal seas, where Ra is more bioconcentrated than U and Th (IAEA 2004). This low bioconcentration of Ra in Ha Long Bay biota is likely an effect of the high concentration of Ca^{++} ions in water resulting from dissolution of calcium carbonate of the abundant limestone rocks (Cao Thi Thu Trang et al. 2020; Cerrano et al. 2006; Drogue et al. 2000). Because the divalent Ca^{++} ion has similar chemical properties to Ra^{++} ion, they compete in the absorption by marine biota, and high Ca^{++} concentrations in water dilute and reduce the bioaccumulation of radium ion (IAEA 2004; Carvalho 2018).

Attempts were made to determine radionuclides by gamma spectrometry in a selection of biota samples (Table 2). The concentrations of ^{40} K averaged 234 Bq kg^{-1} (113-343 Bq kg^{-1}). Cs-137 was not detected and ^{226}Ra from uranium series and ^{228}Ra from thorium series were generally below detection limits (Table 2).

3.3 Food Chain Transfer of Radionuclides

In the marine environment, the partitioning of radionuclides between sediment, water and biota is controlled by several parameters, but the chemical properties of each element are those playing the central role in their concentration in biota (IAEA 2004; Carvalho 2018). Results reported herein (Table 3) show that some chemical elements, such as polonium, are greatly accumulated in organisms and transferred in marine food chains while others, such as uranium, thorium, radium, and lead are not bioaccumulated significantly and are even discriminated along food chains. A graphic display of this is given in Fig. 3.

The average concentration of ^{238}U in sediment - the main radionuclide reservoir - is used as a reference line, and organisms from sediment worm to fish are displayed in a sequence, according to the increasing trophic level. To render data on biota comparable on a whole body basis, the ^{210}Po activity concentration in whole body fish was estimated

Table 4 Activity concentration of radionuclides (Bq kg^{-1} dw) in marine biota from several coastal areas

Biota	Region	D/W	^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th	^{228}Ra	^{40}K	^{137}Cs	Reference
Bivalve molluscs													
Four species	Ha Long Bay	0.16	4.1	4.7	0.8	1.6	9.9	248	1.3				This study
Four species	NE Atlantic	0.20	3.3	3.9			15.5	500					Carvalho and Oliveira, 2008
Four species	Mediterranean	0.20	2.4	2.9			4.4	387					Fonollosa et al. 2017
Mussels	Marmara sea	0.21				2.6	11.4	92			388	1.02	Kilic et al. 2014
Oysters	English Channel	0.13					5.9	188					Connan et al., 2007
Four species	Arabian Gulf	0.22 0.26						45-215					Uddin et al., 2015
Clam	Adriatic Sea	0.16						81±12					Desideri et al., 2010
Crustaceans													
Tiger prawn	Ha Long Bay	0.23	0.45	0.48	0.21	1.2	9.1	421.6	0.33				This study
Shrimp	NE Atlantic	0.28					1.43	90					Carvalho, 2011
Shrimp (Whole Body)	Several seas						0.37-38	15-1669					Cherry and Heyraud, 1981
Fish													
Scad (muscle)	Ha Long Bay	0.25	0.04	0.08	1.00	0.84	2.00	17.70	0.50				This study
Sardine (muscle)	NE Atlantic	0.23	0.10	0.11	0.08	1.74	4.35	130				2.17	Carvalho and Oliveira, 2008
Anchovy (muscle)	Mediterranean Sea	0.30					17.9	64.53	0.20		279-477		Kiliç et al. 2018
Anchovy (muscle)	Adriatic Sea	0.25						110±48					Desideri et al., 2010
Six benthic species (Whole body)	Arabian Gulf	0.24 0.40				0.7-7.3		0.88-5.34		1.2-15.8			Uddin et al., 2015

Whenever needed, results from original reports were converted to the units used in this table

D/W, dry/wet weight ratio

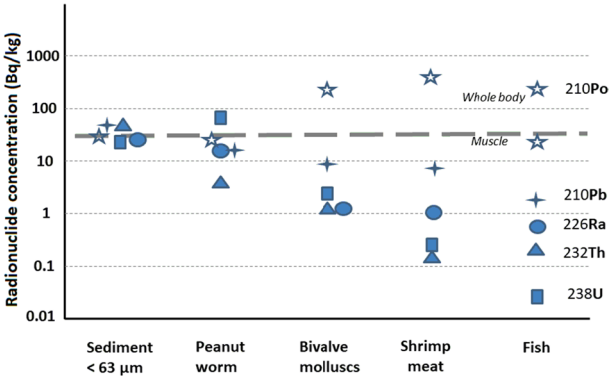

Fig. 3 Bioconcentration trends of radionuclides in biota from Ha Long Bay in comparison with radionuclide concentrations in sediments (dashed line)

(based on proportional radionuclide distribution data on sardines and other fish species). In the marine species analyzed, uranium clearly was not bioconcentrated and, actually, the uranium concentrations in biota decreased with increasing trophic level. The same decreasing trend in higher trophic levels can also be seen for thorium (both ^{232}Th and ^{230}Th), radium, and lead. In contrast to U, Th, Ra, and Pb, the increase of polonium concentrations in higher trophic levels illustrates the distinct behavior of this radioelement and shows its biomagnification in marine biological systems.

In biota from Ha Long Bay, the polonium concentrations were always much higher than the concentrations of other radionuclides. Notwithstanding, ^{210}Po concentrations were similar, or at least were in the range of ^{210}Po concentrations reported for biota from other coastal seas. The remaining naturally occurring radionuclides also showed concentrations similar to marine biota from other regions (Table 4). The trends of polonium biomagnification and uranium discrimination illustrated above with results from Ha Long Bay are supported also by data from other marine regions and are not related to coal discharges into Ha Long Bay (Carvalho 2011). Instead, these trends seem to be universal to all marine regions.

This study on Ha Long Bay showed that the long-lasting industrial activities and discharges of NORM containing effluents into Ha Long Bay, in particular from coal mining, resulted in a minor enhancement of radioactivity in nearshore sediments and no radioactivity enhancement in the marine biota.

The consumption of seafood from the bay will transfer radionuclides to human beings through ingestion. Due to coal mine discharges into the bay, the consumption of sea food should be a major pathway to take into account in the radiological risk assessment for members of the public through exposure to environmental radioactivity. The radionuclide concentrations in sea food from the Bay and the activity-to-dose conversion factors applicable to the calculation of radiation dose to consumers through food ingestion (IAEA 2014) were used to assess the relative contribution of radionuclides to the effective dose. The contributions of ^{210}Po and ^{210}Pb to the effective dose become highlighted as the main ones among the set of radionuclides analyzed, i.e., they are identified as the *critical radionuclides* to the dose (Fig. 4). As the activity levels in biota from Ha Long Bay were comparable to levels reported for other coastal seas, these critical radionuclides will not bring about enhanced radiation exposure of seafood consumers, at least as far as their levels in the Bay remain comparable to background levels.

A recent study performed in Vietnam on the effect of cooking procedures on ^{210}Po concentrations in food revealed that cooking eliminates a large fraction of ^{210}Po present in the

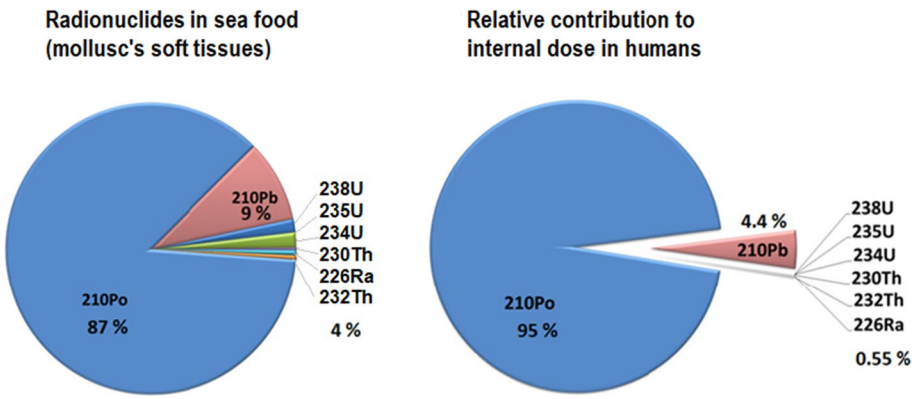

Fig. 4 Left, relative contribution (%) of radionuclides to radioactivity in seafood represented by mollusc soft tissues. Right, relative contribution (%) of radionuclides in sea food to the internal effective dose received by seafood consumers

raw food (Van et al. 2019). Therefore, not only the radionuclide levels in sea food from Ha Long bay are comparable to radionuclide levels in sea food from other coastal seas, as well as the radiation dose to consumers probably will be even reduced by cooking procedures.

4 Conclusions

Along the shoreline of Ha Long Bay, and in particular by the coal harbour, U and Th levels in sediments were slightly increased in comparison with the cleaner calcium carbonate sediments from open sea in the bay. The increase in U and Th concentrations in sediments along the shoreline was due to accumulation of coal particles on the seafloor following coal transport by surface runoff and mine water discharges into the bay.

The relatively small increment of U and Th concentrations in the near shore sediments was not reflected in radioactivity levels in marine biota. Globally, radionuclide concentrations determined in marine biota from Ha Long Bay were comparable to data reported for similar biota from other coastal seas worldwide.

At such naturally-occurring radionuclide levels in biota, the ^{210}Po concentrations stand out in comparison with other radionuclides but this reflected solely the well-known bioaccumulation of polonium in marine organisms. It was noticed that radionuclide concentrations in marine organisms displayed an interesting feature: a polonium bioaccumulation trend increasing with trophic levels, and contrasting with the decreasing bioaccumulation trend of uranium and other radionuclides in higher trophic levels. This feature supports our interpretation that there is biomagnification of polonium in marine food chains.

Although anthropogenic activities around Ha Long Bay might have some impacts on environmental quality, they have not significantly changed the radioactivity levels in the marine environment. Therefore, at the present, the use of Ha Long Bay as a leisure area and the consumption of local seafood do not enhance the radiation exposure of members of the public.

This study focused mainly on radionuclides of natural origin and potential enhancement of their levels in the Bay caused by coal discharges. Other issues related to coal discharges should also be assessed, such as environmental contamination by metals from the coal.

Authors' Contributions All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by FPC, DDN, JMO, NQL, DDT, MM, MS. FPC wrote the first draft of the manuscript and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Funding Supported by the participating institutes, identified as author's affiliations and addresses.

Data Availability All relevant data sets are included in the manuscript.

Code Availability Not applicable.

Declarations

Conflicts of Interest/Competing Interests None.

Ethics Approval Not applicable (no experiments with animals or humans).

Consent to Participate Not applicable.

Consent for Publication Not applicable.

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