Age-dependent dose and carcinogenic risk assessment for radionuclide ^{210}Po in 5 species of shellfish, Thailand

P Nochit*, W Kulsawat and J Khunsamut

Thailand Institute of Nuclear Technology (Public Organization),
9/9 Moo 7, Ongkharak District, Nakhon-Nayok 26120, Thailand

*E-mail: phatchada@tint.or.th

Abstract. Seafood is a significant nutrition dishes of Thai citizen because it is tasty and good for health of all ages. This paper aimed to determine the ^{210}Po concentrations in 5 species of shellfish, i.e., green mussel (*Perna viridis*), oyster *(Saccostrea cucullata)*, enamel venus clam *(Meretrix meretrix)*, radiated scallop (*Amuseum pleuronectes)* and cockle *(Anadara granosa)* caught off the coast of Thailand. The estimation of age dependent effective dose and lifetime cancer risk were performed. The activity concentration of ^{210}Po in shellfish sample was analysed by radiochemical method followed by alpha spectrometric measurement. The average concentrations of ^{210}Po revealed that cockle (6.06 ± 0.93) > green mussel (3.31 ± 0.20) > oyster (2.54 ± 0.36) > radiated scallop (1.73 ± 0.37) > enamel venus clam (1.07 ± 0.24) Bq.kg^{-1} wet basis. The variation activities of ^{210}Po among species were resulting from feeding habit and the transfer factor along the trophic level. Annual effective dose for each species as well as for four age groups (children, juvenile, adult, and senior) of 0.76 to 21.18 μSv.y^{-1} were well below the ICRP permissible limit of 1000 μSv.y^{-1}. The calculated cancer risk of mortality ranged from 0.47 x 10^{-5} to 2.63 x 10^{-5} and also the calculated cancer risk of morbidity ranged from 0.65 x 10^{-5} to 3.64 x 10^{-5}. As stated by the ICRP and US.EPA, the carcinogenic risks in the study area were well below the recommended safe level for radiological risk. Therefore, the investigated ^{210}Po radionuclide in 5 species of shellfish in the coast of Trad province were considered a safe level for human consumption.

1. Introduction

Marine shellfishes settle in a wide variety of environments, and they inhabit in a wide variety of preferences for different conditions; many live-in clean sands, others are mud dwellers, still others prefer to attach themselves to rocks exposed at low tide. Shellfishes consumed in Thailand are clams, mussels, oysters, cockles, and scallops which available in supermarket and fresh food market. Shellfishes are sources of lean protein, vitamins, minerals and healthy fat in the form of omega–3 and omega–6 [1].

It has been documented that marine organism accumulated radionuclides in their tissues through various physiological processes by thousandfold than those in their surrounding environment. Molluscs or shellfishes have been recognized as an organism of excellent bio-accumulators, non–detoxification, stationary, filter feeding, and ubiquitous. Therefore, shellfishes are appropriate for the environmental monitoring of both heavy metals and radionuclides [1–3].

Polonium–210 (^{210}Po) is an almost pure alpha emitter with energy 5.3 MeV and half-life of 138.4 days which is considered to be a highly radiotoxic member of ^{238}U decay chain. ^{210}Po is relatively high values in the marine organisms since it has strong affinity for binding with certain internal tissues and enhances ability of marine organisms on accumulation. As many species of marine biota accumulate ^{210}Po, therefore, ^{210}Po is a major contributor (90%) to the natural radiation dose received by the marine biota and by the human related to intake of seafood [2–4]. The intake of Po^{210} presented in shellfish tissues depends on the Po^{210} contents in the organisms and the consumption rate of the people [2].

The present study was aims at (i) determining ^{210}Po activity concentrations in 5 species of shellfish, i.e., green mussel, oyster, enamel venus clam, radiated scallop and cockle collected from Trad Bay, a coast of Trad province locates at the upper Gulf of Thailand (GOT) during 2017–2018; (ii) calculating the age dependent dose due to ^{210}Po consumption of these different shellfish groups and (iii) assessing the lifetime cancer risk for the Thai population of four age groups.

2. Method

2.1. Study area

Trad Bay is located at the coastal zone of Trad province, a famous coastal tourist city and the easternmost province of Thailand. Trad Bay is part of the eastern upper Gulf of Thailand (GOT). The GOT is a semi-enclosed sea surrounded by Malaysia, Thailand, Cambodia and Vietnam and connected to the South China Sea (Pacific Ocean). The Trad Bay, where this study was performed, is recognized in its rich biodiversity and healthy marine ecosystems. The majority citizen in the Trad Bay is marine fishing household who depend on coastal and fisheries resources. Trad Bay is significantly productive area due to its high nutrient coastal waters. Seafloor is slightly flat with low slope; mud flat is the majority of its coastal topography. The general characteristics are 0 -19 m of water depth, 28^{o}C - 33^{o}C of water temperature, and 8 - 32 psu of salinity with good water quality [5].

2.2. Sample collection and preparation

The different 5 shellfish species were. collected at the same location in the upper GOT and during the same period of time. Green mussel, oyster, enamel venus clam, radiated scallop and cockle were collected from Trad province, for a period of two year from 2017 to 2018. All of shellfish samples were bivalve. The bivalve samples were cleaned and rinsed with deionized water, fresh tissues were separated and weighed. For each species, fresh tissues were freeze dried and were homogenized. Afterward, dried tissues were kept in a sealed bag until analysis.

2.3. Analytical method

The radiochemical process was carried out to determine ^{210}Po in the dried samples matrix [6]. Briefly, ^{209}Po tracer with known amount activity was spiked to 5 g of dried tissues. Then, the sample was dissolution and digestion sequentially using concentrated mineral acids i.e., HNO_3, $HClO_4$ and HCl. Sample solution was raised to a moderate temperature using hot plate until dryness. At this step, caution was the loss of polonium should be limited by controlling temperature levels. The residue was dissolved in 50 ml of 0.3 M HCl and was added 0.5 g of ascorbic acid in order to lower Fe (III). ^{210} Po was spontaneously plated onto silver disc of 99.99% purity at room temperature for overnight. The measurement was performed in passivated implanted silicon detector with a surface area of 450 mm^2 attached to EG&G ORTEC (Octete Plus) Alpha spectrometry and the MAESTRO ORTEC® MCA Emulation software.

The known activity of ^{209}Po spiked tracer is crucial on calculation step. It is assumed that the chemical behavior of ^{209}Po is equivalent to the ^{210}Po in the sample throughout the radiochemical procedure. Based on the peak areas of ^{210}Po and ^{209}Po tracer, the rule of three in mathematics was performed to achieve the ^{210}Po activity concentration in the sample.

The quality control of the Po^{210} analytical method was conducted using certified reference material (SRM) obtained from the International Atomic Energy Agency (IAEA); SRM-IAEA-437 (mussel flesh). The obtained results of 5 replicate analysis (mean 4.1 $Bq.kg^{-1}$) and the certified value (4.2 $Bq.kg^{-1}$) were in good agreement and under the 95% confidence interval.

2.4. The annual committed effective dose (AED)

The annual committed effective dose (AED) due to the intake of ^{210}Po presented in certain shellfishes was calculated to assess the radiological health risk on Thai population of four age groups (children 5–12 y, juvenile 13–18 y, adult 19–59 y and senior 60+ y). The AED was estimated using the equation (1) [7, 8]:

$$AED = DCF \times M_F \times \Sigma(Ai \times Ci \times Fi) \quad (1)$$

where AED is the annual committed effective dose ($\mu Sv.y^{-1}$); DCF is the age dependent committed effective dose conversion factor recommended by the Environmental Protection Agency (EPA) ($\mu Sv.Bq^{-1}$) [9]; M_F is the modifying correction factor (0.6) due to decay of ^{210}Po between catch and consumption [10]; Ai is the ^{210}Po radioactivity concentration ($Bq.kg^{-1}$ fresh weight); Ci is shellfish ingestion rate per year for precisely on a consumer-only basis ($kg.y^{-1}$), [11] and Fi is the real fraction consumed (30% for mollusc) [9].

The committed effective dose conversion factor (DCF) and shellfish consumption rate for eater only (Ci) are shown in table 1.

Table 1. Dose conversion factor and shellfish consumption rate per year in four age groups

Age dependent group		DCF	Consumption rate ($kg.y^{-1}$) fresh weight				
(year)		($\mu Sv.Bq^{-1}$)	Mussel	Oyster	Clam	Scallop	Cockle
children	5 – 12	4.4	3.20	5.73	4.16	4.16	4.42
juvenile	13 – 18	2.6	3.45	6.55	4.71	4.71	4.62
adult	19 – 60	1.2	3.84	6.66	5.07	5.07	4.98
senior	65+	1.2	2.79	4.40	3.26	3.26	3.54

2.5. Evaluation of lifetime cancer risks to humans

The lifetime cancer risk (LTCR) was also calculated using the equation (2) [7, 8]:

$$LTCR = Ai \times Ci \times L_e \times C_{coe} \quad (2)$$

where LTCR is the lifetime cancer risk, Ai is the ^{210}Po concentration, Ci is the annual shellfish ingestion rate ($kg.y^{-1}$), L_e is the exposure duration (70 years for children, 60 for juvenile, 50 for adult and 20 for senior while the average life expectancy at birth for Thai people is 75 y) [12], C_{coe} is the ^{210}Po cancer risk coefficients of 4.4 $x10^{-8}$ $Risk.Bq^{-1}$ for mortality and 6.09 $x10^{-8}$ $Risk.Bq^{-1}$ for morbidity [13].

3. Results and discussion

3.1. Radioactivity concentrations of ^{210}Po in 5 species of shellfish

Measured activity concentrations of ^{210}Po in mussel, oyster, enamel venus clam, scallop, and cockle samples of the upper GOT are presented in table 2.

Table 2. Radioactivity concentrations of ^{210}Po in 5 species of shellfish

Parameter	Species of shellfish				
	Mussel	Oyster	Clam	Scallop	Cockle
^{210}Po activity	3.31 ± 0.20	2.54 ± 0.36	1.08 ± 0.25	1.73 ± 0.37	6.06 ± 0.93
Habitat	Mid–depth	Mid–depth	Shallow	Shallow	Sediment

Among the five species of bivalves investigated, the cockle recorded a higher range of ^{210}Po (6.06 ± 0.93 Bq.kg^{-1} wet weight) than green mussel (3.31 ± 0.20 Bq.kg^{-1} ww), oyster (2.54 ± 0.36 Bq.kg^{-1} ww), radiated scallop (1.73 ± 0.37 Bq.kg^{-1} ww) and enamel venus clam (1.07 ± 0.24 Bq.kg^{-1} ww). However, the recorded values are higher when compare to the work of other investigators [2, 4, 6–8, 14–16].

Although bivalve are a filter feeder organisms that takes phytoplankton, zooplankton, and detritus in the sea water as a food source. Normally, bivalve uptakes food using either filtration or suspension method. Suspension feeders feed on materials that are found suspended in water whereas filter feeders consume materials that are so large that technically they are not "suspended" in water. Filter Feeding indicates that they are actively pumping water through their gills while suspension feeding indicates they are letting water flow through, and they are not actively pumping [17]. The ^{210}Po levels in bivalve internal tissues are depend on the feeding system. It was documented that bivalve species using filtration system accumulated ^{210}Po in their tissues more than the suspension food intake [18].

In addition, the concentration of ^{210}Po in bivalve tissues was influenced by the habitat environment of bivalves. The finding results revealed that cockle had higher average ^{210}Po level compared to green mussel, oyster, enamel venus clam, and radiated scallop. These results suggested that cockle uptake ^{210}Po from its living environment since cockles' preference habitat is the soft mud and fine sand in subtidal areas whilst mussel, oyster, enamel venus clam, and radiated scallop lives in inter–tidal zone attached with abyssal threads to rocks or firm substrate near to the water surface. The content of ^{210}Po mainly enlarges with rising content of silt, clay and organic matter. The sedimentation of organic debris is gradually enhancing ^{210}Po concentration in the sediment. Moreover, cockle is filter feeders that feed on organic detritus whereas mussel, venus clam, and scallop are filter feeders that feed on phytoplankton and zooplankton. Hence, ^{210}Po rich sediment is the main medium for the transfer of ^{210}Po from seawater at different depth to marine organisms [18]. The finding results of this study are in agreement with those given in the literatures [14–16, 19–21].

3.2. The age-dependent annual committed effective dose of ^{210}Po

The total annual effective dose of the four age groups 5–12 y children, 13–18 y juvenile, 19–60 y adult, and >65 y senior are shown in table 3.

Table 3. Age dependent annual committed dose and cancer risk

Age dependent group		AED	Lifetime Cancer Risk (risk x 10^{-5})	
(year)		(μSv.y^{-1})	Mortality	Morbidity
children	5 – 12	3.55 – 21.18	0.47 – 2.63	0.65 – 3.84
juvenile	13 – 18	2.37 – 13.08	0.40 – 2.25	0.56 – 3.12
adult	19 – 60	1.18 – 6.51	0.34 – 1.88	0.46 – 2.60
senior	65+	0.76 – 1.63	0.13 – 0.54	0.19 – 1.04

The annual committed effective dose (AED) of four age groups (children, juvenile, adult, and senior) from the consumption of shellfishes varied widely, ranging from 0.76 to 21.18 $\mu Sv.y^{-1}$. According to the ICRP permissible exposure limit of 1000 $\mu Sv.y^{-1}$; therefore, the estimated dose values for ^{210}Po in 5 species of shellfish collected from Trad province were not exceed the ICRP permissible exposure limit as well as within the mean global shellfish concentration of ^{210}Po [13-16, 19–21]. Consequently, it could be concluded that investigated shellfishes of Trad province were considered a safe level for consumers related to ^{210}Po radionuclide.

3.3. Carcinogenic risk assessment (lifetime cancer risks)

The lifetime cancer risks associated with the ingestion of ^{210}Po in the 5 species of shellfish of the upper GOT were evaluated in terms of mortality and morbidity risks. The cancer mortality risk is a number of fatal in a specified resident that have occurred due to cancer in the course of one's lifespan. While morbidity risk is concerned with the state of having cancer within the certain age group of population.

The calculated cancer mortality risk and cancer morbidity risk owing to the intake of ^{210}Po are displayed in table 3, and cancer mortality risk for children ranged from 0.47×10^{-5} to 2.63×10^{-5} whereas cancer morbidity risk for children ranged from 0.65×10^{-5} to 3.64×10^{-5}. As per the ICRP and US.EPA, the carcinogenic risks in the study area were well below the recommended safe level for radiological risk [9, 13]. In addition, the total lifetime risk of cancer from ingestion of ^{210}Po observed herein, all were below the world mean value of 5.3×10^{-3} [16]. Therefore, there is no risk to consumer's health related to the intake of ^{210}Po radionuclide from the five species of shellfish caught off the coast of Trad province.

4. Conclusion

Polonium-210 activities measured in 5 species of shellfish were presented. In this study, cockle revealed the highest mean activity concentration for ^{210}Po (6.06 ± 0.93 $Bq.kg^{-1}$ wet weight) while Enamel venus clam exhibits its lowest activity concentration (1.07 ± 0.24 $Bq.kg^{-1}$ wet weight). As a result of the variation in ^{210}Po content may be because of the bivalve feeding process, i.e., food intake filtration process and suspension process including species habitat and the surrounding environmental conditions.

The findings of the study revealed the activity concentrations of ^{210}Po in shellfish from the upper GOT were consistent with the values reported in previous studies. The radiation doses and carcinogenic risk of ^{210}Po in all samples were presented with levels much lower than the ICRP permissible limits and were within the world mean values. The health risks resulting from a radiation dose of ^{210}Po to humans from ingestion of green mussel, oyster, enamel venus clam, radiated scallop and cockle were below levels that should cause concern. Therefore, the five species of shellfish investigated were safe and presenting a low health hazard risk to the Thai consumers.

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