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Variations of ^{210}Po activity in mussel *(Perna viridis)* of Samut Sakhon and its contribution to dose assessment

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Abstract. The activities of ^{210}Po and its effective dose in green mussel *(Perna viridis)* collected from a mussel farming area in Samut Sakhon province during the period of 2012-2013 are presented. Several parameters including maximum shell length and the physiological performance of mussels using condition index and physical properties of seawater (pH, salinity, conductivity, TDS, DO and cation-anion elements) were measured. Each individual mussel was measured for its maximum shell length which was adopted as size class. The activity concentration of ^{210}Po was determined spectroscopically through its 5.30 MeV alpha particle emission, using ^{209}Po as an internal tracer. The ^{210}Po activity concentration in mussels was found to vary between 1.044 and 6.951 Bq/kg wet weight. The ^{210}Po concentration was higher in smaller-sized ($\leq$35 mm) and lower in larger ones (40-70 mm). This confirmed that larger mussels have lower ^{210}Po activities on a weight basis. The ^{210}Po body burden (activity per mussel) ranged from 1.035 to 17.183 mBq. Contrary to the ^{210}Po concentrations, results of the body burden revealed the lower activities in smaller-sized mussels ($\leq$35 mm) and the higher in larger-sized ones (40-70 mm). The type of fluctuations observed with ^{210}Po concentrations were interpreted as a seasonal effect. Total annual effective ^{210}Po dose due to mussel consumption was calculated to be in the range of 3.081 to 16.401 µSv. Based on the international guideline, the average dose calculated due to ^{210}Po in mussels of Samut Sakhon would not pose any significant radiological impact on human health and the mussels are considered to be safe for consumption.

1. Introduction

Among the radionuclides, naturally radioactive series of ^{238}U are the main source of radioactivity in the marine environment [1]. Basically, ^{238}U and their decay products are not required for the metabolism of the organisms. However, some of them are bioconcentrated in both hard and soft tissues of marine organisms for they can act as chemical analogs of metabolically essential elements [1, 2]. ^{210}Po is a 99.99% alpha emitter ($T_{1/2}$ = 138.4 d) with alpha particle energy of 5.3 MeV, ^{210}Po occurs in the marine environment as the decay products of ^{238}U radioactive series and it attains great significance in marine biota due to its radiotoxic properties. The toxicity of ^{210}Po is connected with its relatively high energy and that it is concentrated in the soft tissues, such as muscle, liver, kidney, and hemoglobin. The main sources of ^{210}Po entering the marine environment are atmospheric precipitation on the surface and coastal waters [3]. ^{210}Po has a tendency to get accumulated in the edible portions of

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marine organisms, and is considered to be the most important contributors of radiation dose received by humans via fish and shellfish consumption [4, 5]

Marine molluscs such as the green mussel, *Perna virisdis* form an important component of the marine food web. They act as sentinels and considered one of the abundant primary consumers in biogenic habitats. In general, molluscs have the potential to accumulate significant amounts of radionuclides which may be biomagnified in the food chain to higher trophic levels including human consumers. They also serve as a potential bioindicator species in coastal areas by reflecting the effects of natural and anthropogenic stressors [6, 7]. In addition, mussels are considered to be an important protein source, a nutritious and delicious food among the Thai population.

As levels of ^{210}Po activity concentrations in these organisms vary throughout the year due to environmental conditions, variations in ^{210}Po input to the marine environment, and biological cycles of the species [8]. The purposes of this study were (i) to determine the seasonal variation in ^{210}Po activity concentrations in mussel *(Perna viridis)* collected from Samut Sakhon coast in 2 years' time period (2012-2013) as an extension to our previous research [9] and (ii) to determine the dose assessment of ^{210}Po in this organism.

2.Materials and methods

2.1. Sampling

Samples of mussel, *Perna viridis*, were sampling from green mussel farming area at Phanthai Norasing subdistrict, Samut Sakhon province. This region is located in the upper Gulf of Thailand where green mussel is cultured by local fishermen using bamboo poles along the coastal region.

Seawater was collected from 8 stations where 3 kilometers apart from each station and surrounding the mussel sampling poles. Seawater sample was kept in 250 mL polyethylene bottles for measurements of physical-chemical parameters such as pH, conductivity, salinity, dissolved oxygen (DO), and total dissolved solids (TDS) including cation-anion elements.

Three to four whole clusters of live green mussel samples were collected every month at about 5 meters depth and transport in an ice box to the laboratory where they were separated by size.

2.2. Sample preparation and condition index

Upon return to the laboratory, the mussel samples were rinsed with seawater and were immediately frozen at -30^{0}C until grouping. Based on shell length, several groups of mussels were made using Vernier caliper. Each size class was separated and then cleaned to remove fouling organisms such as algae, barnacles and spats. The mussels were dissected to remove the whole soft tissue from the shell (excluding byssus thread). The soft tissues were pooled from each size groups and their fresh weights were recorded. The soft tissue samples were freeze-dried to complete dryness and further homogenized and powdered after recording the dry weight.

The mussel condition index (CI), a measure of the mussel's physiological status, was calculated according to the following equation [10].

$$\mathrm{CI} = W \bullet SC^{-1} \times 100$$

where, W is the average soft tissues dry weight (g) and SC is the shell cavity calculated as SC = total wet weight of soft tissue - dry shell weight.

2.3. Analysis of ^{210}Po

^{210}Po radionuclide analysis was performed using 20 g of homogenized mussel samples together with 0.12 Bq ^{209}Po as internal isotopic tracer. The sample was digested with 200 mL concentrated HNO_3 and then slowly heated on hotplate to reach incipient dryness. The sample residue was subsequently digested with concentrated $HClO_4$ and then HCl. The dry residue was finally dissolved in 6 M HCl and brought up to the volume of 100 mL in 0.3 M HCl. The residue was separated from supernatant by centrifugation and the Po contained in the solution plated onto a silver disc and measured by alpha-spectrometer. Measurements of polonium alpha particles emitted by the Ag disc was performed with low background 450 mm^2 ion implant detector from ORTEC EG&G connected to an Octete Plus alpha spectrometer.

Quality assurance of analytical results was ensured by analysis of IAEA certified reference materials (IAEA-414 fish muscle) and participation in inter-comparison exercises with good results.

2.4. The annual effective ingestion dose

The estimated effective ingestion dose for individuals as a result of radionuclide intake was derived from measured concentrations in sample using the appropriate ingestion dose coefficient factors (DCC) for adults recommended by IAEA [11]. This dose was calculated as follows:

$$E_{dPo\text{-}210} = A_i \times D_F \times M_F \times AI$$

where $E_{dPo\text{-}210}$ is the annual effective ingestion dose in µSv.y $^{-1}$, A_i is the activity intake (Bq/kg wet weight (ww)), D_F is the dose coefficient factor for adults (1.2 µSv/y) M_F is the modifying factor due to decay of ^{210}Po between catch and consumption (0.6) and AI is annual intake of mussel estimation. The Southeast Asia consumption rate is 4.1 kg/y dry weight [12].

3. Results and Discussion

3.1. Seawater properties and condition index

During this study, the coastal site seawater temperature varied between 21.5 °C in winter and 33.4 °C in summer. Seawater temperature generally followed the seasonal fluctuation of average air temperature of the surface seawater, which showed a minimum in December and a maximum in April.

The salinity of sea water ranged from 8.9 to 27.2. Lower salinity was found in September, October and November (figure 1). A decrease in salinity during the 24-month study period was due to the influence of high rainfall during raining season or wet season (with Monsoon or typhoon events) and, possibly, also some regional effect of Tha-Chin River and Chao-Phraya River freshwater discharges into the coastal sea. In this region, the rainfall displays a clear seasonal pattern with heavier rains in raining season and nearly zero rainfall in summer.

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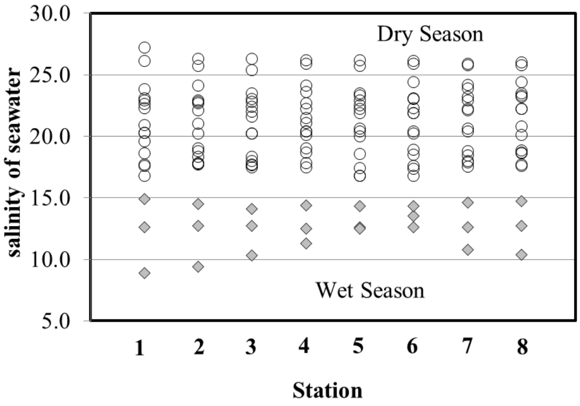

Figure 1. Salinity of seawater at 8 collecting stations.
Round symbols represent the dry season and diamond symbols represent the wet season.

Measurements of pH indicated peaks in March, April and May which was summer season or dry season. The rest of the study period were slightly constant (figure 2). The observed DO, TDS, conductivity and cation-anion elements fluctuated monthly and did not show any obvious pattern (data not shown).

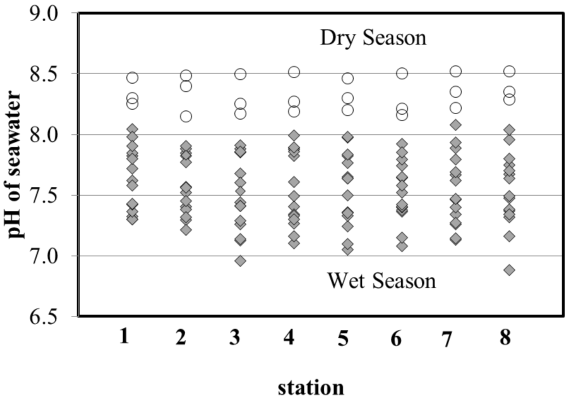

Figure 2. pH of seawater at 8 collecting stations
Round symbols represent the dry season and diamond symbols represent the wet season.

Figure 3 indicates that the mussel condition index changed from month to month. The value of condition index indicates physiological status of bivalves rather than its shell length. The higher value of condition index indicates the higher meat yield. The mussel condition index varied among size classes but was on average higher in August than in the other month. It was obviously separated into two groups: the higher was in the wet season (June – August) while the lower was in the dry season (September – December). This can be attributed to the phytoplankton bloom and spawning time. It is known that water temperature, food availability, cycle of lipid storage, and reproduction cycle (ripening of gonads and release of gametes) may influence the meat yield and biochemical compositions of mussels [13, 14]. The study area, as already mentioned, received significant drainage from the land. These abundant fresh water discharges provided more nutrients and generally higher availability of food in the study area (algae and phytoplankton). However, the continuous freshwater discharge during the wet season observed a dramatic mortality rate among the mussels and eventually deteriorated the mussel farming.

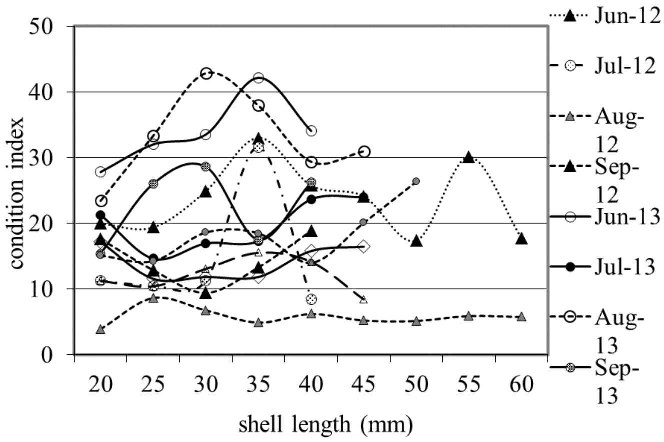

Figure 3. Condition index vs. shell length for mussels of all periods.

3.2. ^{210}*Po activity concentrations*

The activity of ^{210}Po in the soft tissue of green mussels based on size class or shell length are shown in figure 4. ^{210}Po activity concentration was markedly higher in the smaller sized groups (≤35 mm) and lower in the larger ones (40-70 mm). The graph showed a linear decrease when plotted against shell length (figure 4) and also showed the same trend when plotted against soft tissue dry weight (data not shown). This allometric relationship between ^{210}Po concentrations and mussel size is striking and certainly has implication to the interpretation of environmental monitoring data. The activity concentration of ^{210}Po in mussels was reported on the wet weight basis and ranged from 1.044 to 6.951 Bq/kg.

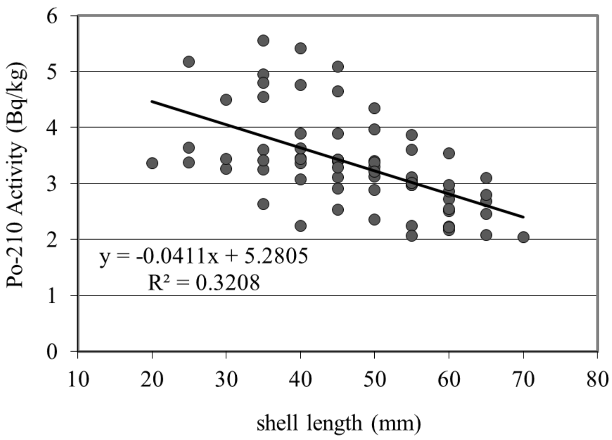

Figure 4. Relationship between Po-210 activity and shell length (size) *of Perna viridis*.

The decrease of ^{210}Po activity with increasing mussel size represented by linear functions of the type $Y = aX + b$ with $R^2 = 0.320$. The low value of R^2 found in this study was similar to the study of Calvaho et al [15] reporting $R^2 = 0.386$. The high ^{210}Po activity concentrations were found in June (3.261 – 5.178 Bq/kg), July (3.305 – 5.556 Bq/kg) and August (3.314 – 6.951 Bq/kg) which was raining season. The atmospheric total deposition of ^{210}Po is also seasonal and positively correlated with rainfall [13]. The possible source for enhanced ^{210}Po in the Upper Gulf of Thailand was the discharge from Tha Chin River and Chao Phraya River. The rivers flowed into the sampling stations carrying the drainage from agricultural land together with contaminants from industrial factories.

It is interesting to note that ^{210}Po activities in mussel soft tissues (radionuclide body burden) increased with increasing shell length (figure 5). This means that the activity of ^{210}Po significantly increased in proportion to the amount of soft tissues and thus to body size or age. However, in all size classes no meaningful variation was found between ^{210}Po body burden and different months of the year. The data on mussels of the same size class covering a 2-year period indicated that whole-body activity concentration remained constant ($R^2 = 0$) regardless of the mussel condition index (figure 6). This suggested that radionuclide body burden did not change significantly over the seasons of the year, despite changes in body weight in relation to the accumulation of lipids and the ripening of the gonad.

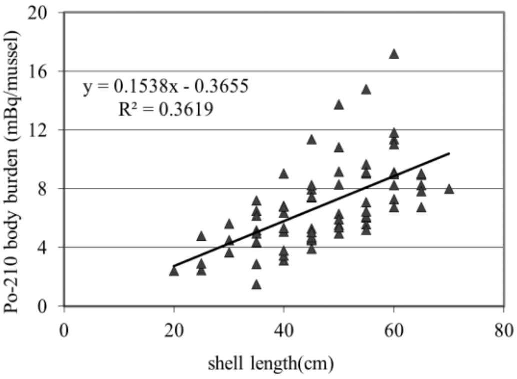

Figure 5. Relationship between ^{210}Po body burden and shell length.

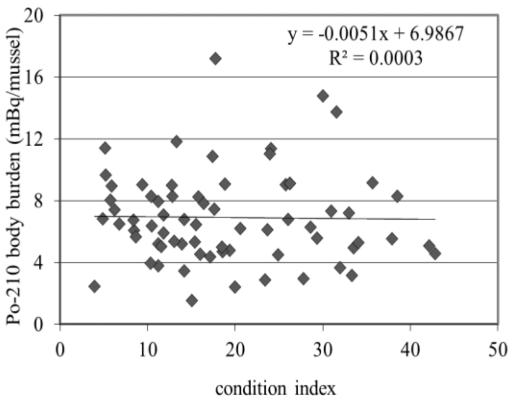

Figure 6. Relationship between ^{210}Po body burden and condition index.

3.3. The annual effective ingestion dose

The diet of the population living along the coast consisted primarily of fish and seafood, which could lead to a higher radiation exposure and subsequent relatively-high radiation doses due to ^{210}Po intake. Annual effective ingestion doses received by an individual were evaluated and found to be in the range of 3.081 to 16.401 µSv. The calculated dose values were compared with those published elsewhere. Aarkrog *et al* [16] estimated the individual dose to be between 51 and 91 µSv/y, with a value of 160 µSv/y for a critical group consuming large quantities of seafood products. Connan et al [17] reported a range of 78 – 129 µSv/y for the adult population from western English Channel, France. Pollard *et al* [18] calculated a dose of 19 µSv/y. Nielsen et al [19] estimated a dose of 700 µSv/y and Jia *et al* [20] gave exposure values between 50 and 200 µSv/y. In the southern coast of India, the annual ingestion dose was estimated to range from 5.1 and 34.9 µSv/y [21]. In this study, the annual effective doses were much lower than in the literatures which can be explained that the annual intake of mussel for Thai people are less than the other countries [12], the different species i.e., *Perna viridis*, *Perna perna*, *Mytilus galloprovincialis* and *Mytilus edulis* have the unique radionuclide accumulation capacities [13, 15] and the different regions have their own environmental conditions.

The International Commission on Radiological Protection [ICRP] has reported a maximum dose of 1000 µSv/y from practices in addition to natural radioactivity to the public, which was estimated to be 2400 µSv/y on average [22, 23]. The calculated doses for exposure to ^{210}Po from consuming green mussels collected from Samut Sakhon province were much less when compared to the ICRP and the global levels. Therefore, it could be concluded that in the mussel farming area of Samut Sakhon, the annual effective dose due to ^{210}Po via green mussel consumption would not cause any health hazards to the general public.

4. Conclusion

The rapid population growth coupled with a rising seafood demand has led aquaculture to become the fastest growing food sector in Thailand [12]. Phanthai Norasing subdistrict, Samut Sakhon province, is one of the promoted shellfish farming areas, and shellfish is one of the most important mariculture product within the country. ^{210}Po concentrations (Bq/kg wet weight) and ^{210}Po body burden (Bq/mussel) observed in the bio-indicator mussel samples from green mussel *(Perna viridis)* farming area in Phanthai Norasing subdistrict, Samut Sakhon province, was presented. The following conclusions could be derived:

1. Seawater properties including pH, conductivity, salinity, DO, TDS and cation-anion elements were measured. The values of pH and salinity were higher in dry season and lower in wet season. However, the conductivity, DO, TDS and cation-anion elements did not show any obvious patterns.

2. The ^{210}Po activity concentrations were found to vary between 1.044 and 6.951 Bq/kg ww and the fluctuation could be interpreted as a seasonal effect which peaked in the wet season and lower in the dry season. Also, ^{210}Po concentrations decreased with increasing mussel size.
3. The ^{210}Po activities in mussel soft tissues (radionuclide body burden) were 1.035 – 17.183 mBq/mussel. The total amount of radionuclide in mussel soft tissues did not fluctuate significantly through the year and did not correlate with the physiological condition (condition index) of mussels. The ^{210}Po body burden revealed the lower activities in smaller-sized mussels and the higher in the larger-sized group.
4. Green mussels *(Perna viridis)* from the study site displayed seasonal fluctuation of ^{210}Po concentrations which were caused by mussel body size fluctuation and were not due to changes in ^{210}Po body burden.
5. The estimated consequent annual effective ^{210}Po doses due to mussel consumption were calculated. They were found to be in the range of 3.081– 16.401 µSv. The obtained effective dose levels of mussels in the studied area were considered radiologically safe for human intake.

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