

## Po-210 as an Indicator of Safe Radiation Levels in Cockles (*Anadara granosa*) in Peninsular Malaysia

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### Abstract

Activities of Po-210 in cockles *Anadara granosa* around Peninsular Malaysia have been determined by Alpha Spectrometry using a radiochemical separation technique. The results showed that Po-210 activity distribution varies according to sampling sites. The range of activity was  $2.61 \pm 1.50$  to  $309 \pm 26$  Bq/kg. The lowest mean was recorded from Kuala Selangor and the highest mean was from Mersing. The activity pattern shown by this radionuclide was a decrease with an increase in cockles mass. The activity was higher in cockles of smaller sizes than the larger sized cockles, a pattern attributed to their digestion and metabolism system. The activity intake and effective dose was found to be less than international guidelines and seafood intake was considered safe for human consumption. These findings suggest that regular monitoring should be carried out to help reduce morbidity and mortality among humans as a result of occasional seafood consumption.

**Keywords:** Polonium-210; *Anadara granosa*; cockles; Peninsular Malaysia

### 1. Introduction

Po-210 is a naturally occurring  $\alpha$ -emitter with a half-life of 138 days which is also used in industrial applications, such as in static elimination and neutron generation. Po-210 exists in the environment as a result of decay within the uranium-238 decay chain. It is distributed in rocks and soil that make up the Earth's crust, as well as in the atmosphere and in natural water bodies as a result of the decay of radon-222. Other natural sources of Po-210 include volcanic eruptions, fires, fossil-fuel burning, migration of sea-salt and resuspension of soil dust (Carvalho, 2011; Jia *et al.*, 2001). These widespread distributions enter the food chain through direct uptake, ingestion or inhalation (e.g., Zakaria *et al.*, 2013).

Historically, scientific interest in Po-210 focused on its application as a tracer of environmental processes and its impact on human health through radiation exposure. The possibility of health effects on humans and other organisms associated with Po-210 arises from its  $\alpha$ -particle emission coupled with its relatively high radiation exposure (UNSCEAR, 2000). Po-210 is considered one of the most toxic naturally occurring radionuclides (Al-Masri *et al.*, 2004), and one of the most important environmental radionuclides due to its wide distribution and potential for human radiation exposure through ingestion and inhalation (Momoshima *et al.*, 2002). Po-210 has received greater attention in the marine scientific community due to their hyper

accumulation in edible portions and because they contribute a higher radiation dose and toxicity (>90%) to humans via seafood consumption (Connan *et al.*, 2007). Understanding the biological interactions of polonium is thus important in risk assessments that consider the health of the ecosystem and public health. The affinity of Po-210 for protein enables it to pass through the food chain and increased body burdens of Po-210 are found where diets include protein meat and seafood (Skwarzec *et al.*, 2001).

Rapid development, urbanization and industrialization in coastal areas contribute to pollutant discharges which directly and indirectly affect aquaculture activities and the marine ecosystem. The determination of radioactivity in marine food supplies assumes great importance since seafood and their products form one of the major sources of protein for the coastal public with a great value in Malaysia and other countries. In this study, cockles; *Anadara granosa* commonly known in Malaysia as "kerang" was chosen because bivalve molluscs have been recognized as first-order biological indicators for contamination as they ingest detritus which has a high degree of radionuclide association (Khan and Wesley, 2012) and their life cycle which connects seawater to sea sediment. It is also largely consumed by the local population as it can be found everywhere in Malaysia. The knowledge of natural isotope of polonium in the edible tissues of cockle or marine organism at Malaysia waters are still lacking and not well published by previous researchers (e.g.,

Zakaria et al., 2013; Zakaria et al., 2013b; Alam and Mohamed 2011; Mohamed et al., 2006; Mohamed and Kuan, 2005). Then an information on the intake of natural radionuclides e.g., Po-210 through seafoods is important to assess its risk and safety to humans. Thus, the objectives of this study were: (i) to find out the distribution of Po-210 in the soft tissues of *Anadara granosa* in Malaysia and (ii) to find out the daily intake and radiation dose due to cockle consumption.

## 2. Materials and Methods

### 2.1. Sampling

Sampling was carried out between March 2012 and December 2012 at various locations along west and south coasts of Peninsular Malaysia (Fig. 1). The samples were collected from the fresh catch sold in the local markets and fishermen. All of the sampling locations; Kuala Perlis, Kuala Kedah, Kuala Gula, Kuala Juru, Bayan Baru, Kuala Selangor, Tanjung Karang, Sebatu and Mersing are situated in Peninsular Malaysia. The samples were transported to the laboratory for further analysis.

### 2.2. Analysis of Po-210

The radiochemical separation method was used to estimate Po-210 in the samples (Alam and Mohamed, 2011). The cockles were dissected to obtain the soft tissue and oven dried at 60°C. About 0.5 g of dried sample was taken and a known activity of Po-209 was added as a yield tracer. Then the samples were digested with nitric acid and peroxide acid. The solution was filtered and gently evaporated to dryness. The sample was dissolved in 0.5 M hydrochloric acid and a pinch of ascorbic acid to reduce Fe (III) and Po-210 was spontaneously deposited on brightly polished silver discs (2 cm diameter) for a period of 3 to 4 hours at a temperature of 70-90°C. The discs were counted for Po-210 activities with an Alpha Spectrometry with a silicon-surface barrier detector by Canberra immediately after deposition process. The recovery yield varied from 65% to 90%. Depositions of Po-210 were carried out within 2 months of sampling and the activities were calculated involving the date of sampling. To ensure the quality of the methodology, Po-210 was estimated in a certified reference material IAEA-134 (Cockle flesh).

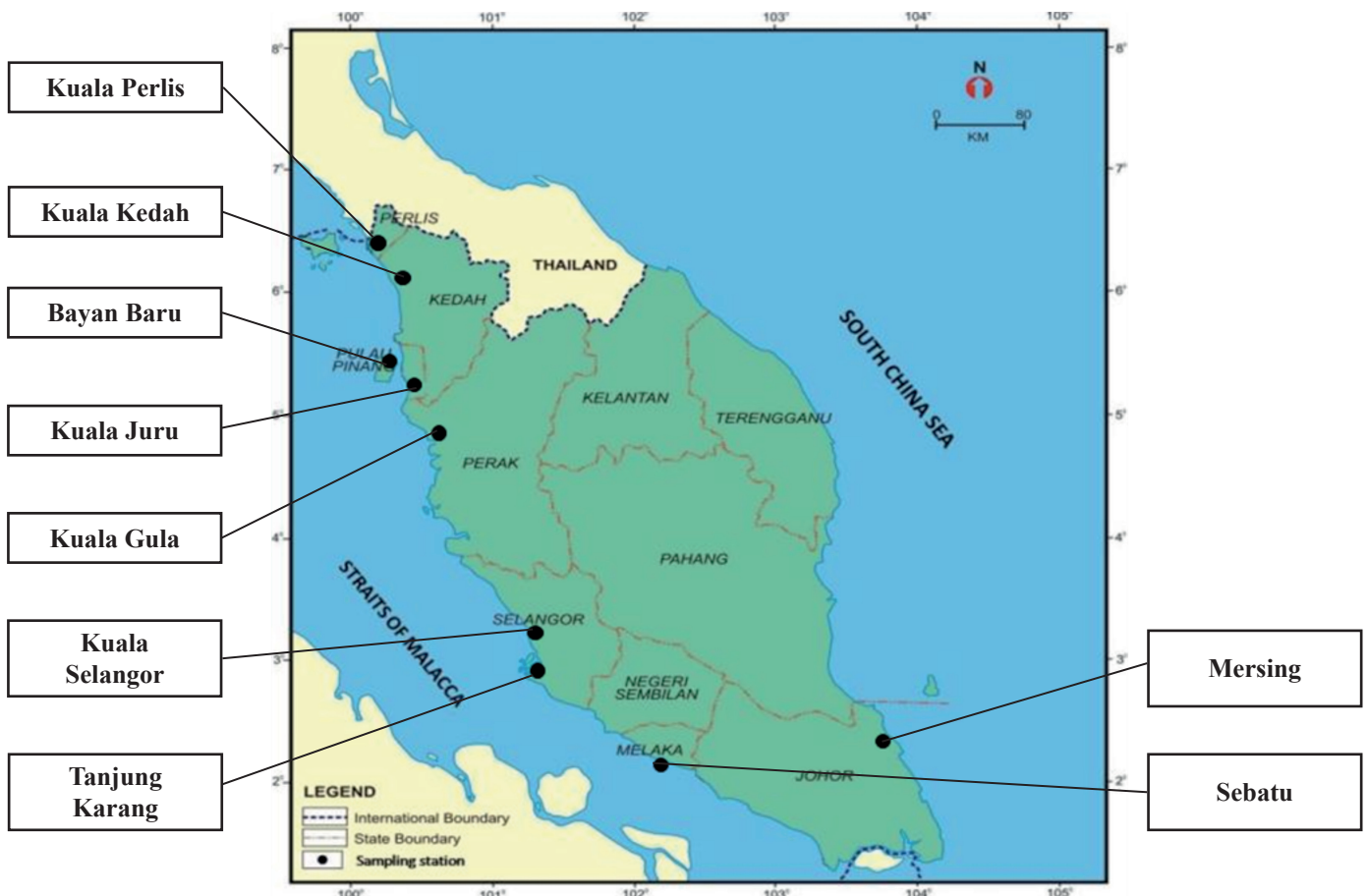


Figure1. Map of sampling locations in Peninsular Malaysia

### 3. Results and Discussion

#### 3.1. Po-210 activity concentration in the soft tissue of cockles

The Po-210 activities measured in *Anadara granosa* ranged from  $2.61 \pm 1.50$  Bq/kg to  $308.54 \pm 25.90$  Bq/kg (Table 1). The mean Po-210 activity in *Anadara granosa* found in Peninsular Malaysia in various sizes and weight were  $82.85 \pm 10.53$  Bq/kg (Table 1), respectively.

Through the statistical correlation test ( $r^2 = 0.38$ ;  $y = -55.78 \ln(x) + 31.474$ ) shows that cockles with smaller weights had a tendency to show higher Po-210 activity than the larger ones (Fig. 2). This pattern was similar to previous research which showed that Po-210 activity in cockles depends on the content of food, food ingestion rate, the degree of assimilation of Po-210 from food into cockles and metabolic rates; all of these variables depend on cockle weights and sizes (Ryan *et al.*, 1999; Jia *et al.*, 2001; Desideri *et al.*, 2009).

To study the influence of size on the whole body accumulation of Po-210, they were categorized into different groups and the recorded activity is shown in Table 2. The activities of Po-210 decreased with increasing size. Higher activities of Po-210 were noticed in the smaller-sized group (1-2 cm) and lower activity occurred in the bigger-sized group ( $>2.7$  cm). This pattern was reported in previous research that showed that radionuclide activity in mollusks significantly decreased with size, shell length or weight (Ryan *et al.*, 1999; Bustamante *et al.*, 2002; Carvalho and Oliveira, 2008; Carvalho *et al.*, 2010; Khan and Wesley, 2012).

The food consumption pathway is important in Po-210 accumulation, and cockles are filter-feeders; they feed on phytoplankton and other suspended matter in near-bottom water. Organic particles in sea water are filtered and ingested by cockles. This is thus their main organic carbon source and this also seems to be the major pathways for radionuclide accumulation by cockles. The authors e.g., Carvalho *et al.*, (2010); Khan and Wesley (2012) suggested that higher Po-210

Table 1. Concentration levels of polonium at various location and sizes found during this study

Location	Shell Length (mm)	Dry weight (g)	Po-210 (Bq/kg)
Kuala Perlis (n = 10) Date: April 2012	22	0.2640	93.05 ± 6.96
	21	0.1874	119.12 ± 11.88
	21	0.2070	161.69 ± 14.29
	22	0.2120	87.84 ± 6.94
	22	0.2580	100.04 ± 8.94
	24	0.3460	98.02 ± 7.14
	23	0.4520	68.77 ± 6.17
	26	0.4730	80.75 ± 5.77
	27	0.4620	83.89 ± 6.46
	28	0.5010	87.72 ± 9.75
Kuala Kedah (n = 10) Date: April 2012	21	0.3227	155.33 ± 12.02
	24	0.4616	119.42 ± 9.84
	24	0.4160	128.14 ± 10.84
	24	0.3600	116.31 ± 9.71
	25	0.4080	158.43 ± 12.56
	25	0.3731	137.65 ± 11.63
	28	0.4720	132.66 ± 10.27
	29	0.6260	110.26 ± 9.40
	28	0.4930	88.29 ± 7.48
	30	0.7220	101.16 ± 12.02
Kuala Gula (n = 10) Date: April 2012	24	0.6197	37.06 ± 9.66
	25	0.6838	81.92 ± 17.15
	25	0.6796	74.02 ± 17.93
	25	0.6453	112.04 ± 26.10
	25	0.6587	83.09 ± 20.56
	26	0.6551	38.39 ± 10.18
	28	0.7105	85.58 ± 21.72
	28	0.7353	92.62 ± 23.06
	30	0.8367	19.39 ± 5.72
	32	0.9874	103.64 ± 14.58

Table 1. Concentration levels of polonium at various location and sizes found during this study (continued)

Location	Shell Length (mm)	Dry weight (g)	Po-210 (Bq/kg)
Juru (n = 10) Date: April 2012	15	0.5046	19.78 ± 7.91
	19	1.0716	4.01 ± 2.28
	20	1.4402	11.92 ± 2.97
	20	2.0816	10.93 ± 1.12
	22	0.2160	38.54 ± 11.48
	25	0.7728	16.65 ± 4.62
	24	1.5828	10.13 ± 2.25
	25	0.1250	151.80 ± 31.88
	28	0.0966	280.09 ± 24.13
	29	1.8171	14.87 ± 2.35
Bayan Baru (n = 10) Date: July 2012	21	0.0688	308.54 ± 25.90
	20	0.1012	16.66 ± 17.42
	22	0.1566	144.04 ± 10.15
	24	0.0965	245.62 ± 19.66
	24	0.1046	21.83 ± 18.35
	24	0.1255	145.47 ± 25.19
	24	0.1655	144.29 ± 38.28
	25	0.1678	171.95 ± 15.55
	25	0.1550	15.34 ± 9.60
	27	0.1337	98.19 ± 26.61
Kuala Selangor (n = 10) Date: September 2012	22	0.2400	109.82 ± 10.84
	22	0.2590	101.81 ± 10.05
	22	0.3448	86.04 ± 14.60
	24	0.2875	107.35 ± 8.52
	25	0.3707	65.23 ± 5.83
	26	0.4066	69.50 ± 5.49
	26	0.3036	78.84 ± 9.47
	26	0.2992	114.50 ± 8.81
	27	0.3328	104.44 ± 8.16
	29	0.4747	70.59 ± 5.37
Tanjung Karang (n = 8) Date: September 2012	25	0.3533	71.72 ± 8.89
	26	0.4929	44.86 ± 6.76
	28	0.8045	52.46 ± 5.93
	30	0.8965	45.09 ± 6.42
	31	0.8070	66.32 ± 7.20
	32	0.8800	41.41 ± 5.25
	31	0.9107	26.00 ± 4.93
	32	1.0783	12.03 ± 1.88
Sebatu (n = 9) Date: December 2012	22	0.3934	22.89 ± 10.13
	24	0.5763	42.65 ± 8.94
	24	0.5526	9.60 ± 5.15
	24	0.4259	28.82 ± 8.91
	23	0.4324	26.18 ± 9.07
	25	0.4718	6.44 ± 1.06
	25	0.4723	5.66 ± 1.22
	25	0.5346	3.49 ± 0.65
Mersing (n = 6) Date: March 2012	28	0.6123	2.61 ± 1.50
	21	0.2270	200.27 ± 14.23
	23	0.2734	178.82 ± 17.01
	24	0.3226	146.88 ± 15.85
	24	0.3609	36.15 ± 3.86
	26	0.4438	36.58 ± 5.51
27	0.4034	35.15 ± 6.43	
Mean value			82.85 ± 10.53

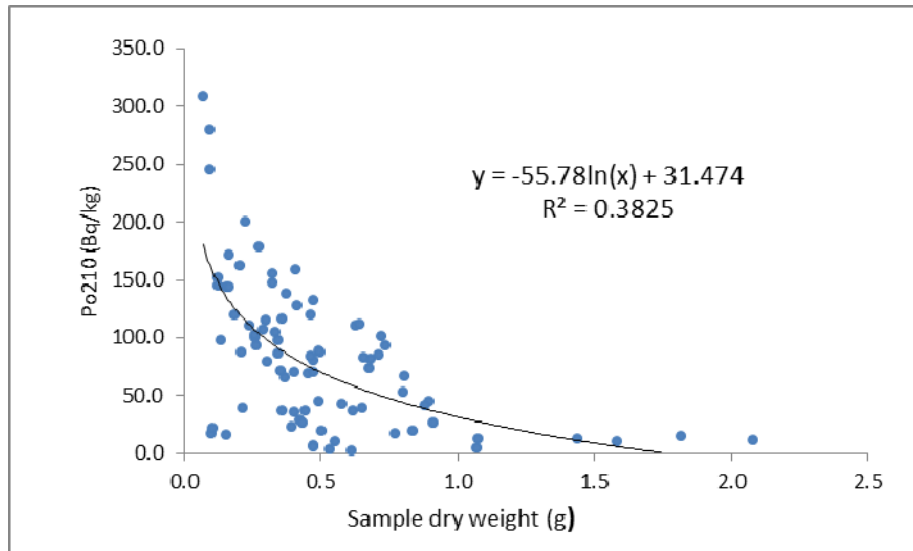


Figure 2. Po-210 activity in *Anadara granosa* based on weight

activity in smaller sized cockles is probably due to their rates of ingestion and metabolism. Higher ingestion and metabolic rates allow them to grow into adult cockles. Lower Po-210 in larger sized cockles may be due to the presence of mature gonads, which indicate a period of gametogenesis. During gametogenesis, the animal does not feed and all stored nutrients will be utilized for the development of gametes. Therefore a lower Po-210 was present in larger-sized cockles as showed in Fig. 2.

Fig. 3 illustrate the highest Po-210 activity, which was noticed in cockles from Mersing ( $131.19 \pm 20.67$  Bq/kg), followed by cockles from Kuala Perlis ( $124.76 \pm 10.24$  Bg/kg) and Tanjung Karang ( $105.64 \pm 20.67$  Bg/kg). The difference in Po-210 activity in cockles from different locations may be due to anthropogenic impacts on the location. Higher Po-210 activities was noticed in organisms from locations with high developed industries due to anthropogenic impact, and were exposed to variation contamination either from a land or sea based source (McDonald *et al.*, 1991; Theng and Mohamed, 2005).

As shown in Table 3, a comparison with other locations in previous studies showed that the level of Po-210 activity in this mollusk is at a moderate level.

### 3.2. Daily intake of Po-210 via consumption of cockles

The consumption of cockles plays a major role in the accumulation of Po-210 in the human body. The daily intake of Po-210 is considered to be an accumulation of Po-210 in the human body through the consumption of seafood. The daily intake of Po-210 by the Malaysian adult population of 20.6 million (Malaysian Demographic Profile, 2013) was calculated with the following equation;

$$\text{daily intake, (Bq/day/person)} = [A \times B \times C] / [D \times E] \dots\dots\dots (1)$$

Where A is the activity of radionuclides in the soft tissue of cockles (Bqkg<sup>-1</sup>), B is the annual production of aquaculture production by culture system reported by Department of Fisheries Malaysia (2011), which was 57,526 tonnes, C is rate of edible part is the constant value (0.6), D is Malaysian Population (20.6 million) and E is the time (365 days). A calculation of daily intake of Po-210 was carried out to be 379 mBq/day/person. It was observed that the intake value in the present study is relatively low compared to a previous

Table 2. Mean concentration levels of Po-210 in edible tissues of cockles at different sizes

Size class	Po-210 (Bq/kg)
class 1 (1 – 20 mm)	89.8 ± 10.6 (n=10)
class 2 (21 – 26 mm)	82.3 ± 11.2 (n=49)
class 3 (> 27 mm)	76.3 ± 9.7 (n=22)

The *n* is the number of samples

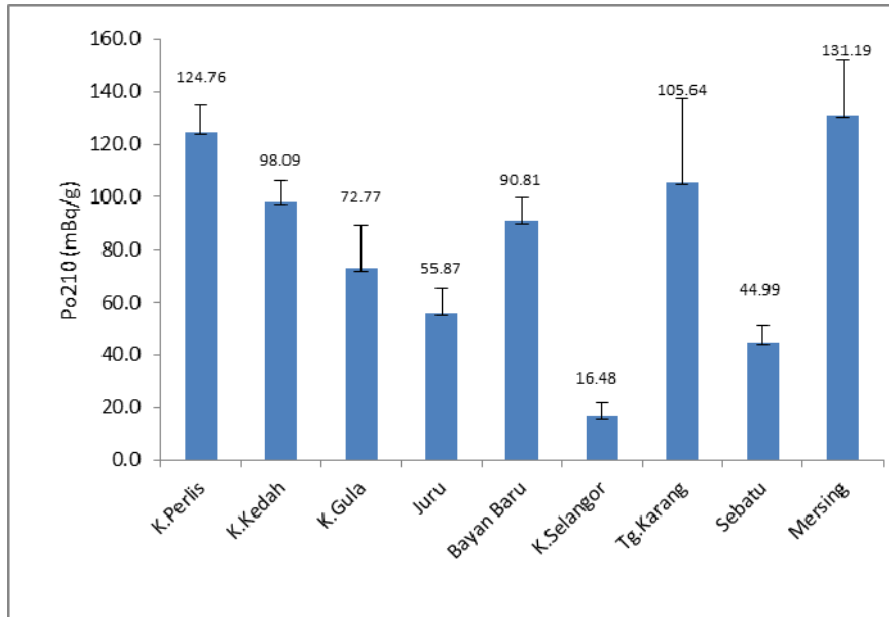


Figure 3. Mean activity of Po-210 in *Anadara granosa* from different sampling locations

study; *Anadara granosa* at Kapar coastal, Malaysia in which the intake value was 5400 mBq/day/person by Alam and Mohamed (2011). In fact, the daily intake value of the Malaysian population was lower than the world reference value, which is 58000 Bq/day/person (ICRP, 1994)

3.3. Committed Effective Dose (CED) calculation

When radioactive compounds enter a human body through consumption, the effects are different from exposure to an external radiation source. This is especially so in the case of alpha radiation, which usually does not penetrate the skin; exposure can be much more damaging after ingestion or inhalation. Radiation exposure is normally expressed as a committed effective dose equivalent (CEDE). There are two methods that can be used to estimate radiation dose deposition in the human body (IAEA, 2003). The estimate can be based on

Po-210 activity in seawater or on Po-210 activity in cockle tissue. Internal dose received through a concentration of Po-210 in the soft tissue of cockles was measured here. The committed effective dose was calculated as follows:

$$\text{committed effective dose, ced (mSv/year)} = A \times B \times C \dots\dots\dots (2)$$

Where A is the daily intake (379 mBq/day/person) and B is the ingestion dose coefficient (dose equivalent per intake of unit activity, Sv/Bq) reported by the International Commission on Radiological Protection (ICRP, 1994) is  $2.4 \times 10^{-7}$  for Po-210 and C is the exposure frequency (365 days/year). By applying the above dose conversion factors, the value of CED was calculated to be 0.03 mSv/year. This value is within the safety limit recommended by ICRP (1 mSv/year) for Po-210 (ICRP, 2007).

Table 3. Po-210 concentrations in mollusk tissues from different regions of the world

Po-210 (Bg/kg)	Locations	References
2.6-309	Peninsular Malaysia	Present study
5.8-132	Portugal	Carvalho (2011)
305.4-596.6	India	Suriyanarayanan <i>et al.</i> (2008)
21-30	Cuba	Alonso-Hernandez <i>et al.</i> (2002)
16-36	England	Young <i>et al.</i> (2002)
4.61-239	Malaysia	Lubna Alam (2012)
5.4-248	Kudankulam coast, India	Khan and Wesley (2012)
1939	Gulf of Mannar, India	Masilamani (2001)
35.19	Kalpakkam, India	Iyengar <i>et al.</i> (1981)
57-106	Tiruchirappalli, India	Shaheed <i>et al.</i> (1997)
51.2-124.6	Adriatic Sea	Štok and Smodiš (2011)

#### 4. Conclusions

IAEA estimates that the portion of the background dose resulting from the ingestion of natural radionuclides in food is about 0.25-0.4 mSv/year. Consuming contaminated food will increase the amount of radioactivity inside a person and therefore increase their exposure to radiation, thereby possibly increasing health risks associated with radiation exposure. The exact health effects will depend on which radionuclides have been ingested and the amount being ingested (UNSCEAR, 2000). On the basis of the findings in this study, it was concluded that Po-210 activity in cockles *Anadara granosa* would pose no health hazards for consumers. As calculated, the daily intake and internal dose received was found to be within international guidelines dictated by the IAEA and ICRP and the seafood intake was considered safe for human consumption. Results presented here suggest that low radiation doses are expected for the Malaysian population through the consumption of Po-210 in analyzed cockles.

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