

# <sup>210</sup>Po IN MARINE FISH: ESTIMATION OF THE EFFECTIVE DOSE TO CONSUMERS IN PENINSULAR MALAYSIA

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#### ABSTRACT

The activity concentrations of <sup>210</sup>Po and its annual committed effective dose to consumers were quantified in some species of marine fishes from the east coast of Peninsular Malaysia. The study aimed to evaluate <sup>210</sup>Po in marine fish i.e. Indian mackerel and Delegoa thread fin bream and to estimate the annual committed effective dose of this radionuclide to humans via fish consumption in the region. The activity concentrations of <sup>210</sup>Po in the whole body of marine fishes and its concentration ratio were ranged of  $8.34 \pm 0.38 - 25.38 \pm 1.14$  Bq/kg fw. and  $4.32 \times 10^3 - 8.12 \times 10^4$  L/kg, respectively. The annual effective doses of <sup>210</sup>Po per adult consumer, who consumed 44.78 kg of marine fish a year, were estimated to be ranged from 161.34 µSv/yr to 490.98 µSv/yr. Thus, the biological variables between species, geographical characteristics and other factors are the main causes for the elevation of <sup>210</sup>Po concentrations that accumulated in both fishes and their annual effective dose levels. However, these values are considered radiological safe for human consumption in the region with is to be taken account on preparation, storage and cooking methods before ingestion.

Keywords: <sup>210</sup>Po, marine fishes, activity concentration, effective dose, consumption

#### 1. Introduction

Polonium-210 (Po-210, <sup>210</sup>Po) is one of the most interesting and important a pure alpha emitters naturally occurring radionuclides due to its relatively high activity concentrations in certain foods and its relatively high ingestion dose coefficient (IAEA, 2017). It has a half-life of 138.377 + 0.002 days which is occurred in the environment through the decay products of <sup>238</sup>U radioactive series. Additionally, <sup>210</sup>Po is considered has a great significance in the ocean in particular to marine organism due to it has a highly radiotoxic properties and radiation exposure (Khan and Wesley, 2011). Its radio-toxicity through internal contamination which is related to a relatively high energy of about 5.3 MeV (Harisson et al., 2007; Jefferson et al., 2009) and that it is concentrated in the soft tissues, such as muscle, liver, kidney, and haemoglobin (IAEA, 2004; Khan & Wesley, 2011; Matthews et al. 2007; Ŝtrok & Smodiš, 2010; UNSCEAR, 2000)

Polonium-210 enters to the marine environment via the natural radioactive decay of Rn-222 gas, Ra-226 in solution and through wet and dry atmospheric deposition of Bi-210 and Pb-210 (Turekian et al., 1977). It can accumulate in various marine environmental materials such as sediments, etc. which is strongly accumulated in a variety of marine organisms (Carvalho, 2011; Cherry and Shannon, 1974). It is also strongly adsorbed onto the surfaces of the living marine organisms and is mostly accumulated in the organic structures or in the edible portions of marine organisms. Due to those properties, <sup>210</sup>Po is known to be a major contributor (90%) to the natural radiation dose received by most marine organisms (Cherry et al., 1989; Cherry and Shannon, 1974) and critical group doses in particular seafood consumption (CEC, 1990). Its activity concentrations in marine fish vary widely from 0.08 to 12 Bq/kg (w.w.) (UNSCEAR, 2000) and concentration factors (CR) varying from  $10^3$  to  $10^6$  (Fowler, 2011). While, the radiation exposure of adult populations from ingestion of natural radionuclides is 0.11 mSv/yr, this large portion is contributed by <sup>210</sup>Pb/<sup>210</sup>Po (UNSCEAR, 2000). The concentration of Po-210 in edibles of marine organisms is very much higher than that of seawater because of biological re-concentration processes (Musthafa & Krishnamoorthy, 2012). Due to the even activity concentration in ocean water, the concentrations of Po-210 in marine biota are related to species rather than to geographical regions. Other possible factors affected this variation are taxonomic groups, habitat, species, type of tissue, environmental condition, feeding habit, transfer factor in the trophic level, physical size, etc. It is clearly tend to contribute critical doses to seafood consumer. Ingestion of <sup>210</sup>Po through seafood consumption varies from place to place and depends on the concentration of <sup>210</sup>Po in seafood organisms, as well as on the consumption rate of seafood (Alam & Mohamed, 2011a). Thus, the knowledge of the concentration, distribution effective dose of this radionuclide is interesting since it provides important information for the monitoring of environmental contamination by natural radioactivity (Alam et al., 2011).

Marine fish products are consumed around the world but Malaysia is among the countries in the Southeast Asian region with the relatively highest fish and seafood consumption as a main source of protein. It is known to be the highest consumer of seafood, both in terms of per capita intake and percentage of protein. Therefore, such study regarding the <sup>210</sup>Po concentration in edible seafood and its dose is very important for this country in order to assess the level of radiological safety to human in this region. The aims of this study were to evaluate <sup>210</sup>Po in marine fishes i.e. Indian mackerel and Delegoa thread fin bream from the east coast of Peninsular Malaysia and to estimate the annual committed effective dose of this radionuclide to humans via fish consumption in the region.

#### 2. Materials and methods

#### 2.1 Study area

A total of 6 points are a part of area for the study of level, trends and effects of natural and anthropogenic radionuclides in the Malaysian marine environment under IAEA CRP K41017 project (Behaviour and effects of natural and anthropogenic radionuclides in the marine environment and their use as tracers for oceanography studies) were selected for sampling stations. These sampling stations with respectively coordinates i.e.  $04^{\circ}$  36.672' -  $05^{\circ}$  58.067'

N Latitude and  $102^{\circ} 35.657' - 103^{\circ} 37.582'$  E longitude are located on the east coast of Peninsular Malaysia which is facing to South China Sea (Table 1, Figure 1). The stations are relatively shallow with water depth between 23.4 - 41.8 m and situated about 20 km from mainland. These stations were selected by presuming the mainly sources of naturally radionuclides from most of the mainland of the neighbouring countries are able to accumulate or transfer into Malaysian waters in particular in the east coast of Peninsular Malaysia. Moreover, many residential villages are situated along the same coast which are active as a fisherman with their fishing heritage, aquaculture and tourism activities that there have greater intakes of seafood and potential to expose to radiological risk.

Location	Station Id	Latitude, N	Longitude, E	Date	Water Depth (m)
Tok Bali	TB03	05° 58.067′	102° 35.657′	4 May 2018	23.4
Kuala Terengganu	KT01	05° 33.467′	103° 08.905′	4 July 2018	30.7
Marang	MG03	05° 08.557′	103° 26.703′	5 July 2018	41.8
Redang-Bidong	<b>RB</b> 04	05° 40.234′	102° 59.982′	9 August 2018	30.0
Dungun	DG03	04° 47.751′	103° 36.619′	3 Oct. 2018	40.5
Paka-Kerteh	PK01	04° 36.672′	103° 37.582′	4 Oct. 2018	39.6

Table 1 – Metadata of sampling collection localities, time and water depth



Fig 1 – Location of the sampling station in the east coast of Peninsular Malaysia

### 2.2 Sample collection and pre-treatment

2 kg of fish samples i.e. Indian mackerel and Delegoa thread fin bream were collected throughout 2018 using a fishing rod or purchased from local fisherman. All the samples were transferred into a zipped plastic bag, stored in cold storage boxes which are covered by ice cube and transported to the laboratory. These fish are selected in this study due to both are the most commonly eaten by Malaysian and consisting of pelagic and demersal species. 20 L of surface seawater sample (0.5 m depth) was collected at each sampling station using in-house modified water bottle sampler and filled in water plastic bottle to bring back to the laboratory.

In the laboratory, the fish samples were removed from ice and let them for a while until no water or ice. All the whole body fish samples were weighed and record a total fresh weight in a preparation logbook. Then, the samples were dried at  $60^{\circ}$ C in an electric oven until a constant weight, weighed it again after cooling and record their dry weight to estimate the water content for conversion factor calculation from dry weight to fresh weight. All the dried samples were ground to powder form to homogeneity and then, samples were kept into air tight container.

### 2.3 Sample preparation and digestion

For biota samples: 0.5 g of sample was spiked with 0.5 mL of a known activity concentration of Po-209 tracer and was digested by adding 10 mL of concentrated HNO<sub>3</sub> and was evaporated on a hot plate to dryness. Then, 10 mL of concentrated HNO<sub>3</sub> and 1 mL of  $H_2O_2$  were added and evaporate to almost dryness. Finally, 10 mL of concentrated HCl was added into the samples, then evaporated until almost dryness and the sample was let to cool. The dry residues were dissolved in 10 mL of 2 M HCl and proceed to radiochemical separation using Sr resin for the purpose of Pb-210 analysis (Zal U'yun et al., 2019).

For seawater samples: 1 L of unfiltered seawater was used for the analysis. The sample was acidified with 2 mL of concentrated HCl and 0.1 mL of a known activity concentration of Po-209 tracer was added into the sample. 0.5 mL of saturated KMnO<sub>4</sub> was added and the pH of the solution was adjusted to pH 8 - 9 by adding 10 M NaOH. After that, 10 mL of 0.5 M MnCl<sub>2</sub> was added into the beaker and the solution was stirred for 3 hours and left overnight. The supernatant was decanted, centrifuged and the precipitate was rinsed until neutral pH obtained. Then, the precipitate was dissolved in a small volume of concentrated HCl and H<sub>2</sub>O<sub>2</sub> and evaporated until dryness. The dry residues were dissolved in 10 mL of 2 M HCl and proceed to radiochemical separation using Sr resin for the purpose of Pb-210 analysis (Zal U'yun et al., 2019).

#### 2.4 Radiochemistry separation

The dissolved precipitate of seawater and biota in 2 M HCl was performed radiochemical separation using Sr resin. 0.7 g of Sr resin was loaded on a separation column with a ball of glass wool on top of the resin. Firstly, the resin was pre-conditioned by adding 10 mL of distilled water and 15 mL of 2 M HCl. Then, the dissolved precipitate was loaded into the Sr resin and was rinsed twice with 5 mL of 2 M HCl. Sr resin was further rinsed with 15 mL of

2 M HCl and the eluent was discarded. Po-210 was eluted by passing 25 mL of 6 M  $HNO_3$  into the resin and the collected solution which contains polonium isotope was then evaporated to dryness (Zal U'yun et al., 2019).

# 2.5 Spontaneous auto-deposition

3.3 mL of concentrated HCl was added to the dried sample until totally dissolved and dissolution. Then, the solution was transferred into the plating jar and added with distilled water until mark up of 80 mL. One (1) mL of stable Bi carrier (10 mg/g) solution and 1 g of hydroxylammoniumchloride were added into the solution and was heated gently until all precipitate dissolve. Polished silver disks were mounted in the plating holder and were put slowly and carefully into plating jar to avoid the solutions are splashed out. The plating jars were placed on a magnetic stirrer hot plate, stirred and heated at a temperature of 85°C for four hours. After the plating process is completed, the plating holders were taken out, washed with distilled water and rinsed with ethanol. Lastly, the discs were air dried (Zal U'yun et al., 2019).

# 2.6 Counting by alpha spectrometry system

The measurement of <sup>210</sup>Po particles was carried out by using alpha spectrometry system (Ortec, Ortate Plus) for 24 hours. Polonium-210 activity concentrations were corrected to the time of collection.

### 3. Results and discussion

Activity concentrations, concentration factors (CFs) and annual effective dose of <sup>210</sup>Po in the marine fish collected from six sampling stations situated along the east coast of Peninsular Malaysia are summarized in Table 2. The <sup>210</sup>Po concentration found in the whole-body of Indian mackerel and Delegoa thread fin bream were ranged from  $10.21 \pm 0.46$  Bg/kg fw. to  $25.38 \pm 1.14$  Bq/kg fw. and  $8.34 \pm 0.38$  Bq/kg fw to  $23.19 \pm 1.04$  Bq/kg fw, respectively. The finding of this study showed that activity concentrations of <sup>210</sup>Po in marine fish were high compared to worldwide ranges of 0.08 - 12 Bq/kg w.w. reported by UNSCEAR (2000). This indicated Malaysia is one of the country has a high content of mineral consisting ilmenite, zircon, rutile, and monazite in the soil and beach sands which progressively enhances natural radionuclides such as <sup>210</sup>Po into the marine water. Furthermore, the east coast regions are mostly enclosed by the landmasses due to received large input of soil which are known as sources of naturally occurring radioisotopes including <sup>210</sup>Po. This supply of radionuclides to the marine environment through soil erosion by rain water flow from mainland of neighbouring countries, Peninsular Malaysia and surrounding islands in particular during monsoon season. Additionally, high concentrations of <sup>210</sup>Po were also due to continuously regenerated from the oil and gas exploration activity along the offshore of this zone and released to the surrounding area as a main supply source for this radionuclide at the study site.

Station Id	Fish Local Name/English Name/Scientific Name	Habitat	<sup>210</sup> Po in Fish (Bq/kg fw.)	<sup>210</sup> Po in Seawater (Bq/m <sup>3</sup> )	Concentration Factor (CF) (L/kg)	Annual Committed Effective Dose (AED) (µSv/yr)
TB03	Ikan Kembong / Indian Mackerel / Scombridae rastrelliger Kanagurta	Pelagic	$25.38 \pm 1.14$	$2.16\pm0.09$	1.18E+04	490.98
TB03	Ikan Kerisi / Delegoa Thread Fin Bream / <i>Nimipterus delegoa Smith</i>	Demersal	9.33 ± 0.42	$2.16\pm0.09$	4.32E+03	180.49
KT01	Ikan Kerisi / Delegoa Thread Fin Bream / <i>Nimipterus delegoa Smith</i>	Demersal	$23.19 \pm 1.04$	$0.53\pm0.02$	4.38E+04	448.61
MG03	Ikan Kembong / Indian Mackerel / Scombridae rastrelliger Kanagurta	Pelagic	$10.21\pm0.46$	$1.61\pm0.07$	6.34E+03	197.51
MG03	Ikan Kerisi / Delegoa Thread Fin Bream / <i>Nimipterus delegoa Smith</i>	Demersal	$18.70\pm0.84$	$1.61\pm0.07$	1.16E+04	361.75
RB04	Ikan Kerisi / Delegoa Thread Fin Bream / <i>Nimipterus delegoa Smith</i>	Demersal	$8.34\pm0.38$	$0.15\pm0.01$	5.56E+04	161.34
RB04	Ikan Kembong / Indian Mackerel / Scombridae rastrelliger Kanagurta	Pelagic	$12.18\pm0.55$	$0.15\pm0.01$	8.12E+04	235.62
DG03	Ikan Kerisi / Delegoa Thread Fin Bream / <i>Nimipterus delegoa Smith</i>	Demersal	$12.40\pm0.56$	$0.84\pm0.04$	1.48E+04	239.88
PK01	Ikan Kerisi / Delegoa Thread Fin Bream / <i>Nimipterus delegoa Smith</i>	Demersal	$12.95 \pm 0.58$	$1.27 \pm 0.06$	1.02E+04	250.52

# Table 2 – Activity concentration of $^{210}$ Po in the whole-body of fish and its annual effective dose to consumers

Ideally, it is found the concentration of  $^{210}$ Po in fishes is negative strongly correlated to the water depth with r = 0.81 (Fig. 2), indicated there are many suspended matter and food particles containing the  $^{210}$ Po in shallow water as a source of food for fishes are living there. Moreover,  $^{210}$ Po is one a strong particle reactive radionuclide and tendency to associate with suspended particle resulted easily and rapid to remove into bottom water column by scavenging process at shallow water and lastly accumulated in the fish body.

Referring to the ranges of <sup>210</sup>Po concentration showed a significant different between both species, whereas Indian mackerel was slightly higher <sup>210</sup>Po concentrations compared to Delegoa thread fin bream. The differences in the level of <sup>210</sup>Po accumulation in different species of fish could be due to the different living or geographical characteristics habitats, location and environmental condition, feeding metabolism, habit and pattern of the species, biological processes, size and seasonal changes (Alam & Mohamed, 2011b). Aközcan and Uğur (2013) agreed with us that mechanism of uptake of <sup>210</sup>Po by fish depends on those factors which are discussed above as biological variables. The higher concentration of <sup>210</sup>Po in Indian mackerel which is a pelagic species who lives in the water column or the pelagic zone of the ocean was fed a suspended matter and food particles from the water. Since polonium is absorbed from water and incorporated into the suspended particles, it is suggested that the high concentration in the body tissues of Indian mackerel might be due to the feeding habits of the marine organisms and its transfer factor to the higher trophic level (Kulsawat & Porntepkasemsan, 2016). Ŝtrok and Smodiš, (2011) was strictly supported that the pelagic environment contribute significantly to  $^{210}$ Po accumulation that is aligned with Indian mackerel which is accumulated high concentration of  $^{210}$ Po. The content of protein in fish also play important role in accumulation of  $^{210}$ Po, it is suggested  $^{210}$ Po is primarily associated with proteins in living organisms (Alam & Mohamed, 2011b). Since the content of protein in the body of Indian mackerel (19.32 g) is relatively higher than Delegoa thread fin bream (17.76 g) (http://www. Fatsecret.co.id, 2020), therefore, Indian mackerel obtained the highest concentrations of  $^{210}$ Po.



Fig 2 – Relationship between the activity concentration of <sup>210</sup>Po in fish and water depth

While Delegoa thread fin bream is demersal fish that lives on the bottom of the sea and feed on invertebrates and small fishes. Thus, this related to the depth profile indicated an increase of <sup>210</sup>Po to a mid-water (Fowler, 2011) which almost pelagic fishes live in this zone and the decreasing again of Po-210 to lower levels at greater depths (Fowler, 2011) which demersal fishes live and feed. This finding indicates that Po-210 in fishes is most probably contributed by seawater and less from the sediments. It is strictly proven by Carvalho and Fowler (1993) that the ingestion of food also plays a major role in the accumulation of Po-210. Additionally, the result revealed a clear relationship between <sup>210</sup>Po accumulation in fishes and the ecological niche of fishes where the accumulation decreases with depth (Alam & Mohamed, 2011a). In other context, low concentration of <sup>210</sup>Po in Delegoa thread fin bream fishes is also due to their body size are relatively larger compared to Indian mackerel fish. Thus, this can be related to the factor of smaller surface area per volume for this species of fish to accumulate the <sup>210</sup>Po. In other hand, Ŝtrok and Smodiš (2011) was reported that the larger animals (eg. fish) have the lower <sup>210</sup>Po activity concentration. According to them, this could be related to the slower metabolism for the larger, older, and heavier animals.

Concentration Factor (CF) is understood as the concentration ratio of a contaminant substance in biota and the concentration of the same substance in surrounding water were

first introduced in aquatic toxicology (Rand, 1995). This factor is usually represented in terms of an activity concentration in biota relative to that in ambient seawater (IAEA, 2004). Then, CF (L/kg) = Concentration per unit mass of the organism (Bq/kg fresh weight)/Concentration per unit mass of unfiltered seawater (Bq/L). The activities of <sup>210</sup>Po in seawater were ranged of  $0.15 \pm 0.01 - 2.16 \pm 0.09$  are presented in Table 2. The <sup>210</sup>Po CFs in the whole-body fish varied widely from  $4.32 \times 10^3$  L/kg to  $8.12 \times 10^4$  L/kg. These values are relatively higher than the recommended value of  $2 \times 10^3$  in the fraction of fish intended for human consumption (IAEA, 2004). This is also higher than published by IAEA (1985), Alam et al. (2011) and Mohamed et al. (2006). This may probably depend on the factors of the physical and chemical properties of the ambient water, behaviour of fishes and related to their species; and geographical regions. This elevation of CFs is also associated with <sup>210</sup>Po level in fish that their influence factors were more detail discussed in above section.

The annual committed effective dose of <sup>210</sup>Po from fish consumption by an adult was estimated from the <sup>210</sup>Po concentrations measured in Indian mackerel and Delegoa thread fin bream in this study. The average yearly consumption of marine fish per adult was set by Ministry of Health Malaysia for consumer in Peninsular Malaysia is 44.78 kg/yr (FCSM, 2014). The annual committed effective dose for an individual from seafood consumption was calculated as follows equation (Connan et al., 2007)

$$AED = DF \times MF \times \Sigma (A_i \times C_i \times f_i)$$

AED is the annual committed effective dose ( $\mu$ Sv/yr); DF is the ingestion dose coefficient,  $1.2 \times 10^{-6}$  Sv/Bq for an adult (EPA, 1988); MF is a weighted factor of 0.6 to account for the time elapsed between catch and consumption, owing to the 138 d half-life of <sup>210</sup>Po (Aarkrog et al., 1997; UNSCEAR, 2000); A<sub>i</sub> is the <sup>210</sup>Po concentration (Bq/kg fresh weight); C<sub>i</sub> is seafood consumption (kg/yr), per capita of marine fish consumption by adult consumer who stayed in Peninsular Malaysia was 44.78 kg/yr (FCSM, 2014) and f<sub>i</sub> is the assumption for 60% real fraction consumed of fish.

The ingestion dose of <sup>210</sup>Po from the consumption of fish was estimated to be in range from 161.34  $\mu$ Sv/yr to 490.98  $\mu$ Sv/yr (Table 2). This indicated the committed effective doses of <sup>210</sup>Po for Indian mackerel (197.51 – 490.98  $\mu$ Sv/yr) were comparatively higher than in Delegoa thread fin bream (161.34 – 448.61  $\mu$ Sv/yr) owing to the relatively high <sup>210</sup>Po activity concentration in Indian mackerel. The dose for <sup>210</sup>Po from the ingestion of two common species of fish estimated in this study found to be slightly higher compared to the radiation exposure of adult populations from ingestion of natural radionuclides of 0.11 mSv/yr. This estimation is large portion contributed by <sup>210</sup>Pb/<sup>210</sup>Po (UNSCEAR, 2000). However, the results of this study could not be compared to other findings because this study is based on the radionuclide concentration in the whole-body of fish and different species with other studies. Most other study elsewhere were reported the total doses for seafood consumption which consisting of fish, crustaceans, molluscs etc., while this study was specific dose for individual fish. Furthermore, <sup>210</sup>Po from these fishes consumption may depend on when the fish are caught, prepared, storage and cooking methods before ingestion (Kim et al., 2017). Since <sup>210</sup>Po has a short half-life and it is easily volatile at high temperature, so it may be minimally present in this two of fishes as typically prepared and cooked in Malaysian cuisine. Thus, in the execution to estimate the dose of <sup>210</sup>Po from fish needs to be more precisely assessed for the next time. The <sup>210</sup>Po concentrations in the edible portions of fish or other marine organisms were used to assess the effect of natural variations of <sup>210</sup>Po on the estimation of effective dose to seafood consumers.

### 4. Conclusions

Activity concentrations of <sup>210</sup>Po accumulated in the whole-body of Indian mackerel and Delegoa thread fin bream were ranged from  $10.21 \pm 0.46$  Bq/kg fw. to  $25.38 \pm 1.14$  Bq/kg fw. and  $8.34 \pm 0.38$  Bq/kg fw to  $23.19 \pm 1.04$  Bq/kg fw, respectively. This showed a significant different between both species, resulted <sup>210</sup>Po concentrations in Indian mackerel was slightly higher compared to Delegoa thread fin bream. While, the annual effective doses of <sup>210</sup>Po per adult individual from the ingestion of those fishes in Peninsular Malaysia were estimated to be ranged from161.34 µSv/yr to 490.98 µSv/yr. Thus, these all values were higher compared to other reported elsewhere. In this context, the biological variables between species, geographical characteristics and other factors are main causes the elevation of <sup>210</sup>Po concentration that accumulated in both fishes and their annual effective dose levels. However, these values are considered radiological safe for human consumption in the region with is to be taken account on preparation, storage and cooking methods before ingestion.

### 5. Aknowledgement

The authors are grateful to International Atomic Energy Agency (IAEA) for funding the CRP 41017 project (IAEA Research Contract No.: 22192). The authors would like to thank to Malaysian Nuclear Agency (Nuklear Malaysia) for continuous supporting and encouraging as well as all personnel and project members involved in the implementation of the research project, sampling and sample analysis.

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