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Monitoring ^{210}Po and ^{210}Pb in Some Pelagic Fishes Collected from Malabar Coast of India

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ABSTRACT

Marine fish are included as potential bioindicators as they strive to incorporate the target radionuclide into their tissues from the surrounding medium. They contribute significantly to the natural radiation dose to human beings through intake. Marine fishes are also sensitive to radiation and regular monitoring of radionuclides in fish is essential. Pelagic fishes of economic importance are collected along the south-west coast (Malabar Coast) of India and analysed for ^{210}Po and ^{210}Pb in muscle tissues using alpha spectrometry The range of ^{210}Po concentration ranged between 29.9 ± 2.2 and 42.5 ± 3.6 Bq kg^{-1} wet, while, the concentration of ^{210}Pb varied between 10.2 ± 3.2 to 16.9 ± 5.2 Bq kg^{-1}wet. Statistically significant variation was observed between the radionuclides between species ($p \leq 0.05$). The ^{210}Po daily intake of each fish muscle tissue ranged from 6.0 to 8.5 Bq d^{-1}. The range of the predicted dose caused by ingestion of both ^{210}Po and ^{210}Pb are comparable with the higher end values reported internationally.

Key words *: Pelagic, Polonium, Radiation, Malabar Coast, Ingestion Dose*

Introduction

Areas of high background radiation (HBRA) offer a unique opportunity to study potential biological effects of long-term exposure to the local population continuously exposed to radiation. The higher radiation is due to uranium, thorium and phosphate rock deposits and thus considered as natural laboratories for radioecological and epidemiological investigations (IAEA, 1995). Populations in these regions are exposed to radiation doses that are significantly higher than the estimated global average background human dose of 2.4 mSv y^{-1} (UNSCEAR, 2000). Additionally, these regions can be classified into four categories based on the annual radiation dose rates at each location: low (up to 5 mSv y^{-1}), moderate (5-20 mSv y^{-1}), high (20-50 mSv y^{-1}), and very high (>50 mSv y^{-1}) (Sohrabi, 1998; ICRP, 1993). Two prominent HBRA regions are located along the southwest coast of Kerala, in Kollam (Chavara and Karunagapally) and many epidemiological investigations were already conducted (Nair *et al*., 1999). Although there are some studies on external exposure to water, air and soil, there is a lack of information on internal exposure to natural radionuclides for the local population around these areas (Sreejith, 2017).

Fish are animals known for their high nutritional value as they contain nutrients that are healthy for humans such as proteins, fats and omega-3 fatty acids. Smaller (like the sardine) and larger (like the tuna) species of marine pelagics inhabit the area on the surface or in the water column. Pelagic fish found on the west coast of India make up the majority (55%) of the country's marine fish production. Over the past seven decades, the production of pelagic marine fish in India has increased significantly, in line with the long-term trend (Nair, 2015). Pelagic fishes ere the first-hand receivers of any pollution from the land and consuming these species leads to

hidden risk to human beings due to the presence of contaminated substances such as heavy metals and radionuclides. Among various radionuclides in the environment, ^{210}Po and ^{210}Pb require special attention due to its radiotoxic properties and hyper accumulation potential in marine organisms (Rani *et al*., 2014; Komperod *et al*., 2020; Hansen *et al*., 2022). ^{210}Po is easily accumulated in fish tissues due to its affinity with proteins when compared to terrestrial foods (Cho *et al*., 2016). Numerous studies have reported the concentration of ^{210}Po and ^{210}Pb in various marine biota around the World (Gouvea *et al*., 1992; Lubna*et al*., 2011; Uddin *et al*., 2012; Ahmed *et al*., 2021). In India, several authors have reported ^{210}Po and ^{210}Pb concentrations in shellfishes and fin fishes from south-east coast, Gulf of Mannar and Manavalakuruchi (Saiyad and Krishnamoorthy, 2012; Rani *et al*., 2014; Khan and Wesley, 2011, 2016; Khot *et al*., 2018; Pillai *et al*., 2019; Sivaperumal *et al*., 2020; Sabu *et al*.,2022). Due to the enhanced bioaccumulation of ^{210}Po in the edible flesh of marine life, especially fish, humans are exposed to it through daily ingestion over a long period of time. (Kong *et al*., 2021: Real *et al*., 2004). ^{210}Po is transferred to fish tissues by plankton and organic waste particles after being absorbed from the sea water column (Tateda *et al*., 2003). Numerous marine fish can therefore be used as bioindicators of environmental radionuclide exposure (IAEA, 2004; Van, 2020). Monitoring radionuclide levels in fish is now more critical than ever because ^{210}Po and ^{210}Pb greatly increases the natural dose of radiation received by the fish itself (Connan *et al*., 2007).

Although studies on the accumulation of ^{210}Po and ^{210}Pb in marine fish have been conducted in a number of countries, the coastal regions of Kerala are rarely the site of these studies. This study aims to identify the accumulation of ^{210}Po and ^{210}Pb in a range of economically valuable fish and to determine the effective dose for the general public.

Materials and Methods

Fish samples were collected between June and October 2021 directly from fishermen at fish markets in Thiruvananthapuram (Vizhinjam), Kollam and Alappuzha (Fig. 1). The fish samples were then taken straight to the laboratory. In the lab, the fish samples were sliced to separate the flesh from the bones. The fish meat samples were then ground after being dried in an oven at 60°C until dry. A dry sample weighing approximately 0.5 gm was added along with ^{209}Po as a tracer and wet digested in microwave digestor using nitric acid and perchloric acid. The final sample was then redissolved in 0.5 M HCl and a small amount of ascorbic acid was added to reduce Fe(III) and ^{210}Po was allowed to plated on a silver disk at a temperature of 70 to 80 °C. After 5 hrs, the activity level of ^{210}Po was determined using the silver disc counted by alpha spectrophotometry with a limit of detection of 0.3 Bqkg^{-1}. This technique was used with minor modifications from Sabu *et al*. (2022) and the IAEA-MEL method (IAEA, 1995). The remaining solution was kept for 6 months for the ingrowth of ^{210}Po from ^{210}Pb. The ^{210}Po was once again counted to back calculate the activity of ^{210}Pb. The quality of the methodology was checked using a reference material IAEA 352. The results were reported on a wet weight basis.

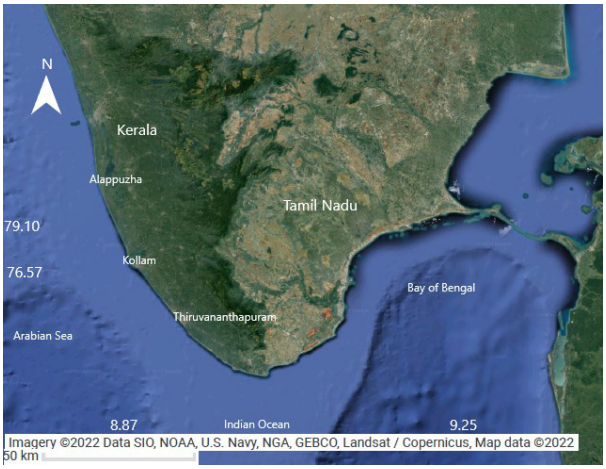

Fig. 1. Map showing the study area

Results and Discussion

Fish muscle tissue had ^{210}Po concentrations ranging from 29.9 ± 2.2 to 42.5 ± 3.6 Bq kg^{-1} (Table 1). The range of the ^{210}Pb was 10.2 ±3.2 to 16.9 ±5.2 Bq kg^{-1}. the activity concentration of both radionuclides was higher in small pelagic fishes compared to large pelagic fishes ($p \leq 0.05$). Species that eat algae, phytoplankton, zooplankton and other debris had the highest concentrations of ^{210}Po and ^{210}Pb, while carnivorous fish such as seerfish and others had the lowest concentrations. The plankton food web, which is controlled by current environmental conditions, is closely linked to marine pelagic fish. Fish that are planktivorous occupy niches that meet their

dietary needs and consume more plankton, which in tern accumulate more radionuclides from the medium. Populations of small pelagic fish can be adversely affected by a number of factors including food competition, predation at different levels and pollution of coastal waters (James, 2010). Among the different fish species, clupidae have been found to accumulate the most radionuclides which is concordant with the literature values (Sabu *et al*., 2022). Morwoto *et al*., (2022) reported a range of 46.4 to 251.6 Bq kg^{-1} for ^{210}Po in different pelagic fishes recently from Indonesia which is found higher than our findings. Due to the binding of ^{210}Po to fish meat, the activity value differed in different fish species which could be due to the difference in habitat, feeding patterns, biological processes, size, season, and other physicochemical elements in the sea. Certain authors correlated the level of ^{210}Po and ^{210}Pb in relation to the amount of amino acids, protein and sulfur (Cherrier *et al*., 1995; Cho *et al*., 2016; Carvalho *et al*., 2017).

The concentration of ^{210}Po in the waters along the coast ranges from 3.6 to 5.7 mBq l^{-1}. These values were used to calculate the biological concentration factor (BCF; Fig. 2). Based on the activity concentration in muscle tissue and seawater, the biological concentration factor (BCF) for ^{210}Po and ^{210}Pb is found highest in fish eating phytoplankton, slightly lower in fish eating worms, molluscs and very small fish and lowest in fish that primarily eat other fish. The raw activity data showed a normal distribution with no outliers at a 95% confidence level andour results indicate that ^{210}Po muscle concentration varied between species ($P \leq 0.001$). Scientific literature revealed that about 90% of ^{210}Po enter into fish tissues through diet, therefore dietary changes, prey ^{210}Po content, and prey intake rate could lead to interspecies differences (Carvalho *et al*., 2017; Kim *et al*., 2017). According to Connan *et al.* (2019) differences in radionuclide concentration in fish could be

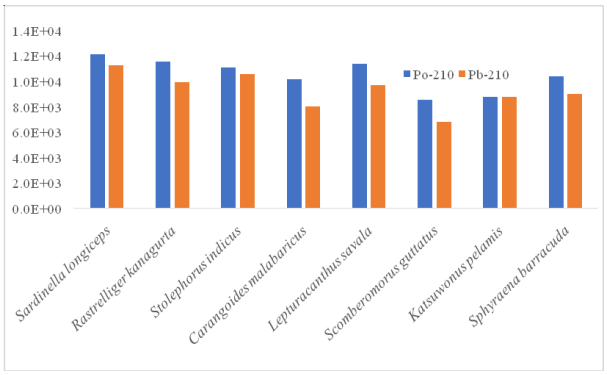

Fig. 2. Biological concentration factors calculated for different fishes

caused by higher or lower levels of protein, calcium, iron, omega-3 fatty acids and zinc in these fish. According to Morwoto *et al*. (2022) there is a significant correlation between protein content and ^{210}Po, and other factors as explained before. differences are the reproductive cycle, trophic levels, and dietary preferences (Kulsawat and Porntepkasemsan, 2016).

The consumption of marine fish containing radionuclides increases the population's internal dose. According to numerous authors, ^{210}Po alone accounts for >80% dosage to human beings via shellfish and finfish consumption (Musthafa *et al*., 2019; Guy *et al*., 2020). On the other hand, due to the high catch, low price, high landing and diversity of finfish, finfish consumption is expected to be higher than shellfish consumption. Most of the fish's muscle tissue is consumed by humans, which should require a lower ^{210}Po activity concentration. The annual committed effective dose (CED) was calculated according to the formula published by Sabu *et al.* (2022). The daily intake of radionuclides was determined by multiplying the corresponding element content by the typical daily consumption of each fish species (determined using the household expenditure method; IAEA, 1989).

The daily ^{210}Po consumption of each fish muscle

Table 1. Concentration of ^{210}Po and ^{210}Pb in different pelagic fishes

S.No.	Species	Common name	^{210}Po	^{210}Pb
1	*Sardinella longiceps*	Indian Oil Sardine	42.5± 3.6	16.9 ± 5.2
2	*Rastrelliger kanagurta*	Indian Mackerel	40.5± 2.6	14.9 ± 3.2
3	*Stolephorus indicus*	Engraulid	38.9 ± 1.8	15.9 ± 2.3
4	*Carangoides malabaricus*	Malabar trevally	35.7 ± 7.8	12.1 ± 1.8
5	*Lepturacan thussavala*	Small-headed hairtail	39.8 ± 5.3	14.6 ± 1.8
6	*Scomberomorus guttatus*	Indo-pacific King Mackerel	29.9± 2.2	10.2 ± 3.2
7	*Katsuwonus pelamis*	Skip-jack tuna	30.8 ± 3.8	13.2 ± 4.2
8	*Sphyraena barracuda*	Barracuda	36.5 ± 2.2	13.5 ± 1.8

Table 2. Radiological dose calculation for adults consuming pelagic fishes

Species	^{210}Po activity (Bq kg^{-1})	Intake (Bq)		CED (µSv)		^{210}Pb activity (Bq kg^{-1})	Intake (Bq)		CED (µSv)	
		Daily	Annual	Daily	Annual		Daily	Annual	Daily	Annual
Sardinella longiceps	42.5	8.5	3102.5	4.3	1563.7	16.9	3.4	1233.7	1.6	595.9
Rastrelliger kanagurta	40.5	8.1	2956.5	4.1	1490.1	14.9	3.0	1087.7	1.4	525.4
Stolephorus indicus	38.9	7.8	2839.7	3.9	1431.2	15.9	3.2	1160.7	1.5	560.6
Carangoides malabaricus	35.7	7.1	2606.1	3.6	1313.5	12.1	2.4	883.3	1.2	426.6
Lepturacan thussavala	39.8	8.0	2905.4	4.0	1464.3	14.6	2.9	1065.8	1.4	514.8
Scomberomorus guttatus	29.9	6.0	2182.7	3.0	1100.1	10.2	2.0	744.6	1.0	359.6
Katsuwonus pelamis	30.8	6.2	2248.4	3.1	1133.2	13.2	2.6	963.6	1.3	465.4
Sphyraena barracuda	36.5	7.3	2664.5	3.7	1342.9	13.5	2.7	985.5	1.3	476.0

tissue ranged from 6.0 to 8.5 Bq d^{-1} (Table 2). The CED is based on rate of activity consumption, and marine fish consumption varies widely around the world. In Slovenia, only 3.7 kg of fish was consumed annually, but in Korea, 42.8 kg was consumed annually along with other seafood delicacies. In Kerala, the annual consumption of fishes is reported to be 19.4 percapita kg y^{-1} (IFS, 2020). According to Moroz and Parfenou (1972), exposure to ^{210}Po and ^{210}Pb in small doses can lead to cancer and other diseases in animals, while larger amounts are immediately dangerous (Harrison *et al*., 2007). Animals exposed to low doses of ^{210}Po activity develop subacute and chronic activity in soft tissue and bone malignancies and lymphomas (Seiler, 2016). Therefore, it is important to estimate the internal dose administered to fish consumers. The CED for this study was estimated to range from 2182.7-3102.5 µSv y^{-1}. the predicted dose was higher to the range of concentrations reported from Indian waters (0.38 to 208 µSv y^{-1}) and comparable with the higher end value reported from various regions of the world (0.01 to 2802 µSv y^{-1}) Pillai *et al.* (2019).

Conclusion

Values of ^{210}Po and ^{210}Pb in various edible pelagic fish from Kerala's coastal waters were estimated. Compared to larger carnivorous fish, smaller plankton-eating pelagic fish have higher levels of radionuclides. The overall CED of the fish consuming population in the study region was determined and found to be higher. A detailed study on the primordial radionuclides and calculation of internal exposure is recommended.

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Conflict of Interest

The author (s) certify that they have **No Conflict** of **Interest** in the subject matter or materials discussed in this manuscript.

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