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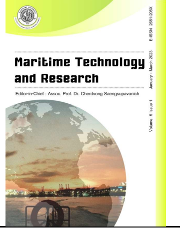

Research Article

Polonium content in some economically important fish in Semarang and Cirebon, Indonesia

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Abstract

As a marine country, Indonesia recognizes various sources of healthy food from the ocean, such as fish, shellfish, and seaweed, leading to the Indonesian government actively promoting a fish-eating campaign to improve health. Although fish consumption in Indonesia is increasing, research on the concentration of ^{210}Po in fish has not often been studied. Based on this condition, this study was needed to determine the activity concentration of ^{210}Po in several fish that have high economic value in Semarang and Cirebon, and their relationship with several parameters of oceanography. The ^{210}Po activity measurement method in fish is based on the IAEA-MEL (2005) method. The detected ^{210}Po activity concentrations were 207.3, 71.7, 130.9, 4.46, 112.6, and 251.6 Bq/kg (dw) for skipjack, four-finger, mullet, snapper, pomfret, and ponyfish, respectively. These values are still considered safe compared to the values allowed by the Indonesia Quality Standard Limit (Bappeten), which is 10×10^3 Bq/kg (dw). Compared to some fish and shellfish used in overseas research, the content of ^{210}Po in some fish in Indonesia is still lower. The activity value of ^{210}Po above 20 Bq/kg was detected in skipjack and ponyfish, which have higher protein, calcium, iron, and zinc content than other fish. However, the ^{210}Po value does not significantly correlate with other parameters, such as bathymetry, current, or sea surface temperature.

1. Introduction

Fish are animals known to have high nutrition because they contain protein, fat, and omega-3, which are beneficial for the human body. The Indonesian government conducted an eating fish campaign to increase fish consumption in Indonesian society, showing it as a healthy food full of nutrition. The results show that fish consumption in Indonesia is increasing every year. Behind the advantages and benefits of eating fish, there are some hidden threats; namely, the presence of polluted materials, such as heavy metals and radionuclides in the waters used as a living medium for the fish. Aquatic ecosystems in several places in Indonesia have been polluted by heavy metals and toxic radionuclides (Cahyani et al., 2016). One of the radionuclides that needs serious attention is ^{210}Po, which emits very toxic alpha radiation.

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^{210}Po is a critical natural radioactive element because it can pollute the marine environment and organisms (Macklin et al., 2014). The presence of ^{210}Po in soil, air, and seawater easily accumulates in fish bodies, which are then consumed by humans (Cho et al., 2016). Many researchers have studied the accumulation of ^{210}Po in marine organisms, such as the *Perna viridis* shellfish in Malaysia (Lubna et al., 2011), India (Saiyad & Krishnamoorthy, 2012), and the *Perna perna* shellfish in Brazil (Gouvea et al., 1992), and India (Rani et al., 2014). Ahmed et al. (2021) conducted research on several important commercial fish in Tadjoura bay, Djibouti, East Africa. Khan and Wesley (2016) also researched the content of ^{210}Po in various types of tuna found in Indian waters.

Although research on the accumulation of ^{210}Po in marine fish has been conducted in several countries, this research is rarely carried out in Indonesia. Semarang and Cirebon are two big cities on the island of Java Indonesia, located on the northern coast of Java, and both have big fish markets. Nuclear facility encourages the surrounding community to increase their fish consumption continuously. This phenomenon must be balanced with knowledge of the level of accumulation of pollutants in the fish.

The purpose of this research: based on this information, it is necessary to research the accumulation of ^{210}Po in several fish with significant economic values and the level of accumulation over several secondary parameters in Semarang and Cirebon, two big cities on the north coast of Java. This research will, hopefully, be the beginning of a furtherance of knowledge about the dangers of ^{210}Po in several economically important fish in Indonesia.

2. Materials and methods

2.1 Research materials

The material used in this research consists of primary data and secondary data. The primary data used is the activity concentrations of ^{210}Po contained in several economically important fish in Semarang and Cirebon, while the secondary data, in the form of bathymetry data, was obtained from BATNAS (Indonesia National Bathymetry Data), the current data in August 2020 was taken from LAS/AVISO images, the Sea Surface Temperature (SST) data was obtained from AQUA/MODIS images, and the biological and ecological data of fish, as well as data on protein and iron content in fish, was obtained from various references.

2.2 Sampling methods

Fish samples were taken directly from fishers who had just arrived at the fish market in Semarang and Cirebon, and were taken on 20 and 25 August 2020, respectively. Fish samples obtained in Semarang consisted of skipjack, four-finger, and mullet, and those in Cirebon were snapper, pomfret, and ponyfish. Then, the fish samples were immediately brought to the laboratory.

2.3 Preparation and analysis of ^{210}Po concentration in fish

The fish samples were cut to separate the bones and the flesh in the laboratory. The fish meat samples were then dried in an oven at 60 °C until dry and then grinded. About 0.5 grams of dry sample was taken and added with ^{209}Po as a tracer. Then, nitric acid and perchloric acid were added to bind ^{210}Po. The solution that was bound to ^{210}Po was filtered. The solution formed was evaporated by heating slowly until dry. Then, the sample was redissolved in 0.5 M HCl and added to with a pinch of ascorbic acid to reduce Fe(III). A silver disc with a diameter of 2cm was inserted into a beaker glass containing the solution. The silver disc was used to bind ^{210}Po within 3 - 4 h at a temperature of 70 - 90 °C. The silver disc that was attached to ^{210}Po was then taken and put into alpha spectrophotometry (which had a detection limit of 0.3 Bqkg-1) to determine the activity level of ^{210}Po. This method was adopted from the IAEA-MEL method (2005).

2.4 Bathymetry data analysis

The bathymetry images for Semarang and Cirebon waters were downloaded from the BATNAS website and were then processed using the ArcGIS application to obtain a clear bathymetry map, where the depth of the water could be indicated by green to red gradient colors and contour lines that showed the values of the water depth.

2.5 Current data analysis

The August 2020 current data downloaded from the LAS/AVISO website was then processed in the Microsoft Excel application. Current data processing in Excel aimed to eliminate unused data and calculate the data needed to be processed in the ArcGIS application. With the ArcGIS application, the current data was converted into a current flow map that showed the speed and direction of the current.

2.6 Sea surface temperature data analysis

SST images that had been downloaded from the AQUA/MODIS web were processed using the SeaDAS application to plot SST images in the study area. After that, the SST image data was processed with the ArcGIS application to obtain a color gradient that symbolized the sea surface temperature in the Semarang and Cirebon areas.

3. Results and discussion

The ^{210}Po activity concentrations detected in six different fish species showed varying results (**Table 1**), in the range of 46.4 Bq/kg to 251.6 Bq/kg. The activity value varied in various types of fish due to the binding of ^{210}Po to fish meat, depending on several factors such as the presence of amino acids, protein (Carvalho et al., 2017), and sulfur (Schwarz, 1976; Cherrier et al., 1995; Church & Sarin, 2007). Water conditions that affect the accumulation of ^{210}Po in fish include geographic characteristics of habitat, feeding habits, biological processes, fish size, seasonal changes, species, and physical chemistry variables in the sea (Cho et al., 2016).

Table 1 ^{210}Po activity concentrations in fish.

No	Fish species	Sampling location	Collection date	^{210}Po (Bq/kg)
1	Skipjack *(Katsuwonus pelamis)*	Semarang	20-Agt-20	207.3 ± 30
2	Four-finger *(Eleutheronema tetradactylu)*	Semarang	20-Agt-20	71.7 ± 4.50
3	Mullet *(Moolgarda seheli)*	Semarang	20-Agt-20	130.9 ± 3.50
4	Snapper *(Lates calcarifer)*	Cirebon	25-Agt-20	46.4 ± 5.40
5	Pomfret *(Pampus argenteus)*	Cirebon	20-Agt-20	112.6 ± 3.70
6	Ponyfish *(Leiognathus equulus)*	Cirebon	20-Agt-20	251.6 ± 2.80

Table 1 shows that the highest ^{210}Po value occurred in skipjack and ponyfish, with activity concentrations of 207.30 ± 30 Bq/kg (dry wt) and 251.60 ± 2.80 Bq/kg (dry wt), respectively. These values were the highest compared to other fish. This was possibly due to the high content of protein, calcium, iron, omega-3, and zinc contained in these fish (**Table 2**), because the nature of ^{210}Po is to be easily bound to protein (Connan et al., 2019) and some metals (Church & Sarin, 2007). However, after being tested for the regression relationship between the content of ^{210}Po with protein, calcium, iron, omega-3, and zinc in all fish, it did not show a significant relationship (**Table 3**), because these fish had differences in biological, ecological, and trophic levels, and eating habits (Kulsawat & Porntepkasemsan, 2016).

Table 2 Content of protein, calcium, iron, omega-3, and zinc in 100 grams of fish.

No	Fish species	Protein	Calcium	Iron	Omega-3	Zinc	References
1	Skipjack	22.4 g	145 mg	1.99 mg	0.471 g	1.52 mg	Mohanty et al. (2014)
2	Four-finger	18.8 g	39.6 mg	0.927 mg	0.354 g	1.12 mg	Bogard et al. (2015)
3	Mullet	18.1 g	50.5 mg	0.857 mg	0.308 g	1.73 mg	Hicks et al. (2019)
4	Snapper	19.3 g	147 mg	1.15 mg	0.407 g	1.03 mg	Mohanty et al. (2014)
5	Pomfret	18.1 g	36.5 mg	0.487 mg	0.289 g	0.526 mg	Bogard et al. (2015)
6	Ponyfish	18.2 g	282 mg	1.61 mg	0.466 g	1.6 mg	Hicks et al. (2019)

Table 3 Regression results ^{210}Po with protein, calcium, iron, omega-3, and zinc.

		Protein	Calcium	Iron	Omega-3	Zinc
Pearson Correlation	Polonium	0.245	0.677	0.670	0.589	0.323
Sig. (1-tailed)	Polonium	0.320	0.070	0.073	0.109	0.266
N	Polonium	6	6	6	6	6

Based on the limit of the allowable ^{210}Po value in the body of living organisms of ~749 Bq/kg (Pentreath & Allington, 1988), the conditions of the six economically important fish that were the subjects of this study (skipper, four-finger, mullet, snapper, pomfret, ponyfish) are safe for consumption. Likewise, they are safe based on the quality standard limit from the Indonesian government (Bapeten, 2017) of 1,000 (Bq/kg). These conditions are possible because the areas of Semarang and Cirebon have not had a lot of ^{210}Po activity generated from mining (UNSCEAR, 1988).

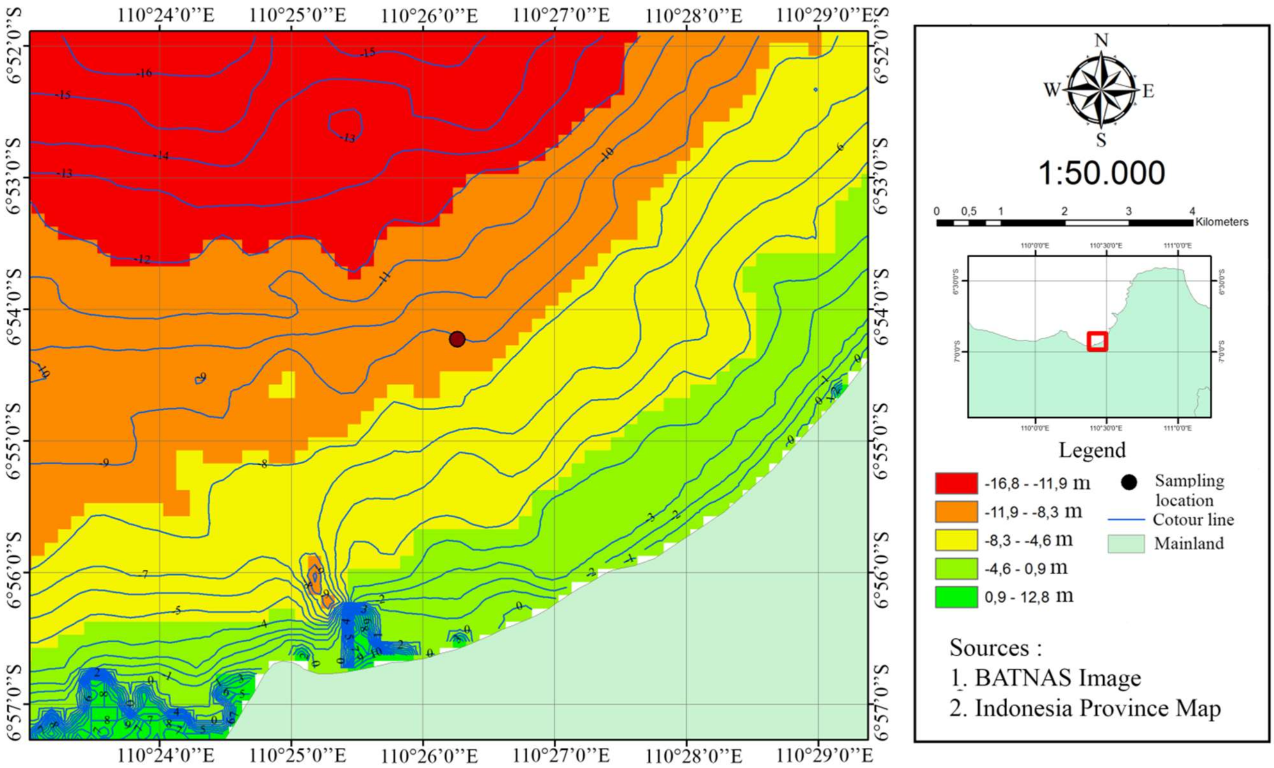

Figure 1 Bathymetry map of Semarang waters.

Although the geographical characteristics of the waters of Semarang and Cirebon (**Figures 1 - 4**) show relatively the same characteristics in the depth of the waters, the direction and speed of the currents, and the surface temperature, the fish studied have different living habits and eating patterns (**Table 4**), and are included in groups of fish with different migration patterns, so that the level of accumulation of polonium is also different (Khan & Wesley, 2011).

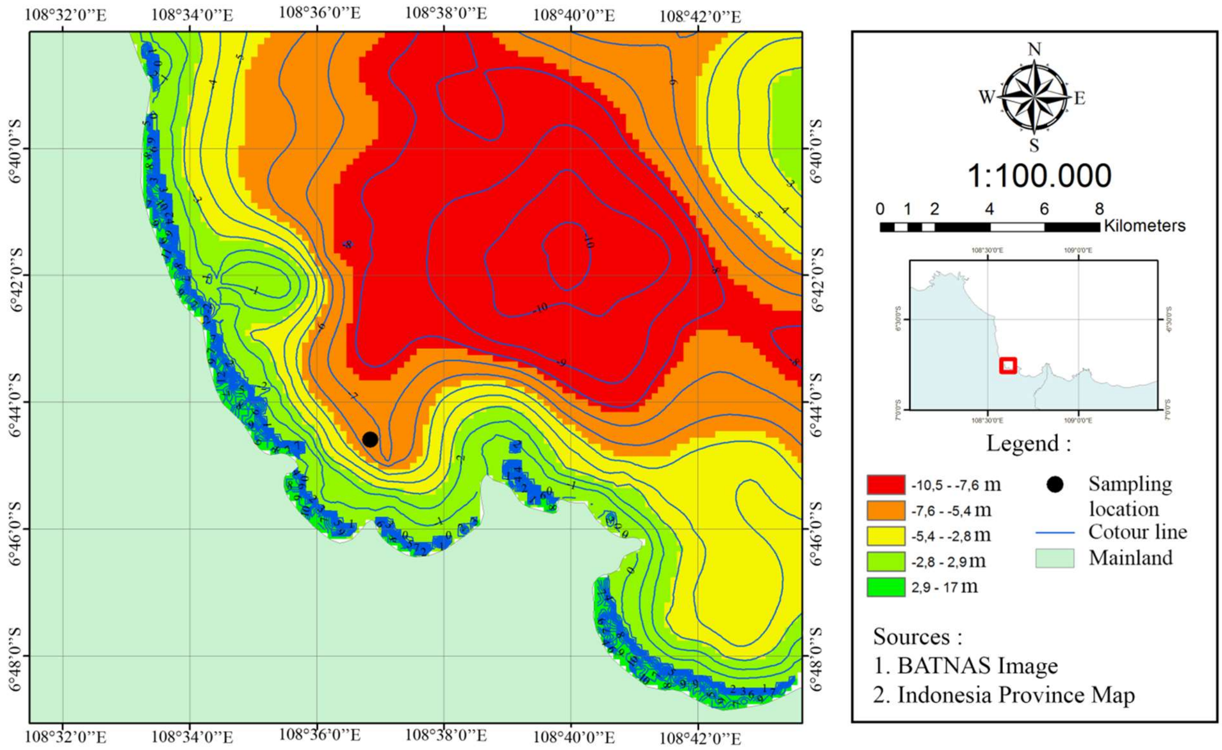

Figure 2 Bathymetry map of Cirebon waters.

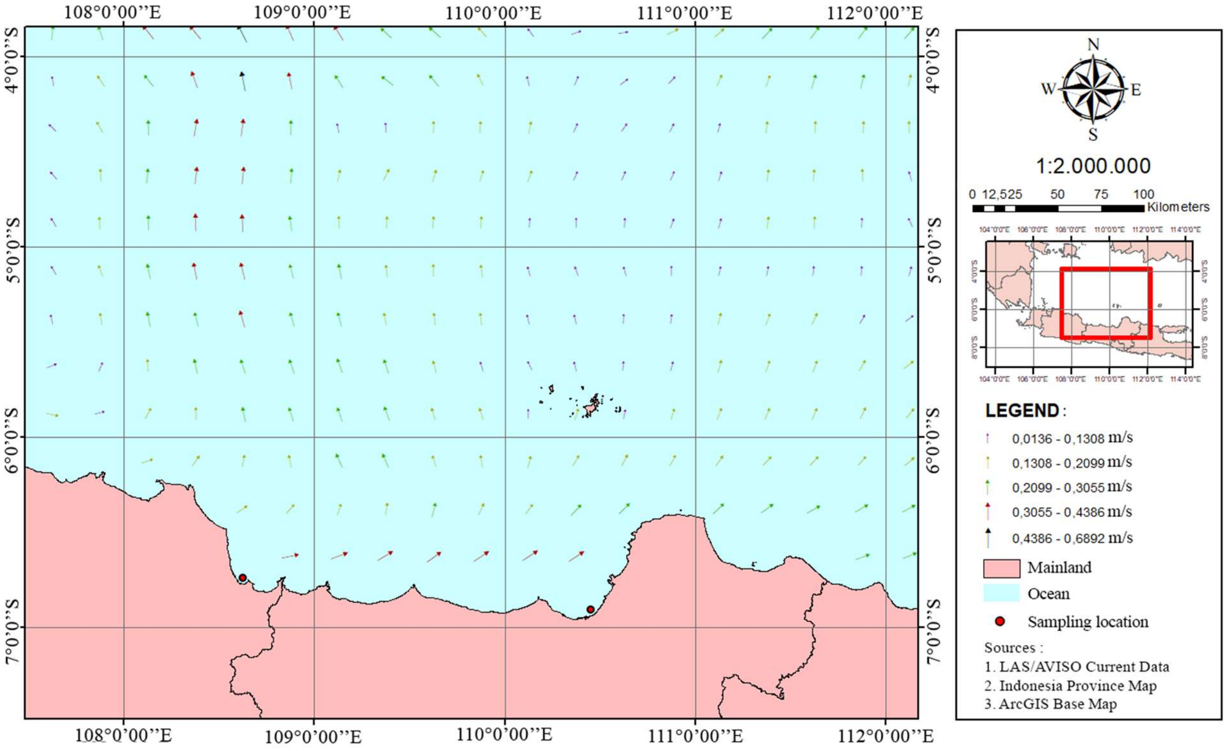

Figure 3 Current flow map of Semarang and Cirebon.

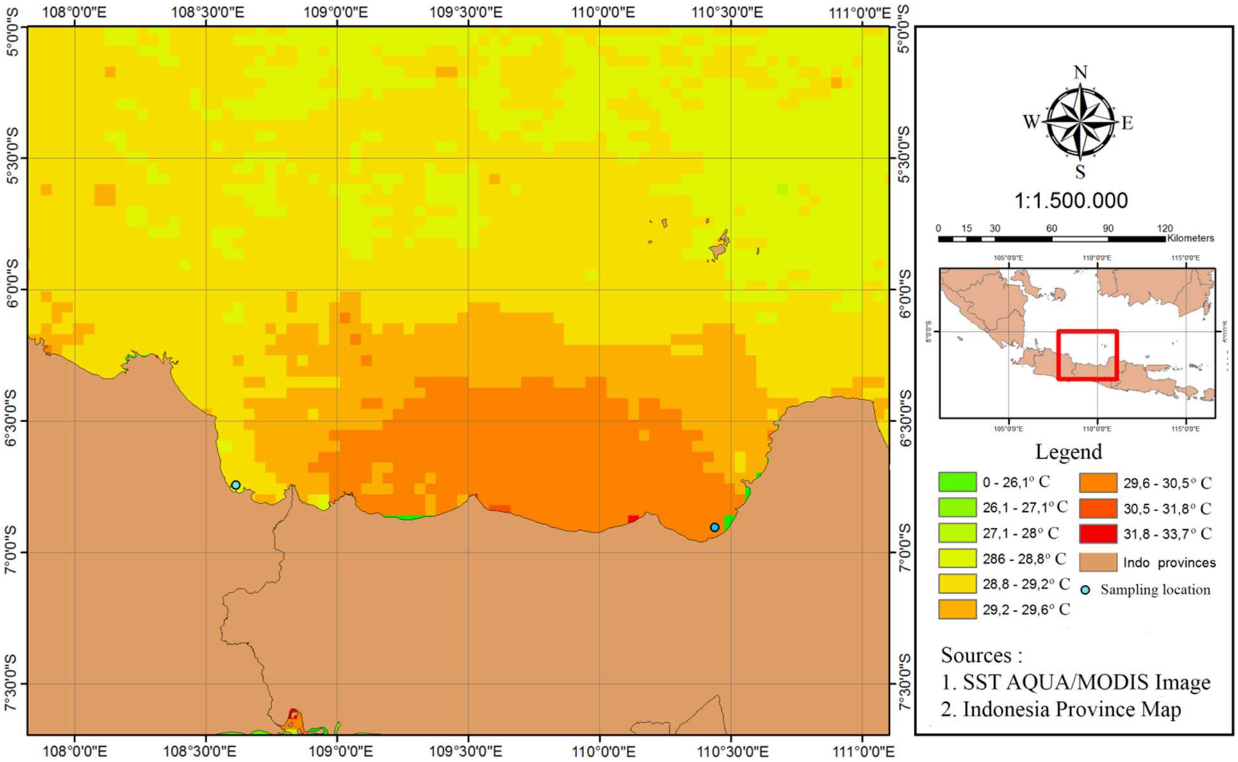

Figure 4 SST map of Semarang and Cirebon.

Table 4 Fish biology and ecology.

No	Fish	Habitat	Food and Habit	Literature
1	Skipjack	Marine; pelagic-oceanic; oceanodromous Depth : 0 - 260 m Tropical; 15 - 30 °C	Eggs and larvae live in pelagic areas. Food: small fish, crustaceans, cephalopods and mollusks;	Restiangsih and Amri (2018)
2	Four-finger	Marine; freshwater; brackish; pelagic-neritic; amphidromous Depth 0 - 23 m Tropical; 32°N - 26°S	Adult fish live on shallow muddy coastal bottoms, and are also found in rivers. Juveniles are found in estuaries. During the winter, fish enter the river. The fish eat shrimp and fish. Its larvae feed on copepods, mysids, and shrimp larvae.	Jaferian et al. (2010)
3	Mullet	Marine; freshwater; brackish; reef-associated; catadromous Depth 0 - 3 m Tropical; 35°N - 32°S	It inhabits coastal waters but enters estuaries and rivers where it feeds on microalgae, filamentous algae, diatoms, and detritus associated with sand and mud.	Harrison and Senou (1997)
4	Snapper	Marine; freshwater; brackish; demersal; catadromous Depth 10 - 40 m Tropical; 15 - 28 °C	Diadromous fish, inhabiting the river before returning to the sea to lay their eggs. Feed on small fish and crustaceans.	Mathew (2009)

Table 4 (continued) Fish biology and ecology.

No	Fish	Habitat	Food and Habit	Literature
5	Pomfret	Marine; benthopelagic; oceanodromous Depth 5 - 110 m Subtropical; 46°N - 10°S	Species commonly found on muddy bases. Adult fish feed on ctenophores, salps, medusae, and other zooplankton groups.	Kizhakudan et al. (2018)
6	Ponyfish	Marine; freshwater; brackish; demersal; amphidromous Depth 10 - 110 m Tropical; 26 - 29 °C	Adult fish are coastal fish found on a soft base, usually between 10 - 70 m deep. Frequently rising into fresh water reaches and rivers. Juveniles are commonly found in mangrove estuaries and tidal tributaries, sometimes entering the bottom of a river's water flow. Food: polychaetes, small crustaceans, small fish, and worms.	Soars and Leis (2010) Hendrayana et al. (2017)

Table 5 Content of ^{210}Po in some fish and shellfish in various countries.

Species	^{210}Po (Bq/kg)	Location	References
Perna viridis	45.69 ± 1.17 to 96.44 ± 2.19 (wwt)	Malaysia	Lubna et al. (2011)
Perna perna	1995 ± 363 (dw)	Brazil	Gouvea et al. (1992)
M.galloprovincialis	428 - 459 (dw)	Monaco	McDonald et al. (1986)
M edulis	149 ± 82 (dw)	Denmark	Dahlgaard (1996)
M trossudus	272 ± 28 (dw)	Baltic Sea	Stepnowski and Skwarsec (2000)
Perna indica	100.3 ± 2.8 (wwt)	Chinnamuttam and Kuthankuzhi, South India	Khan and Wesley (2011)
Perna viridis	113.32 ± 7.10 (wwt)	Ennore Creek, South India	Saiyad and Krishnamoorthy (2012)
Perna perna	78.09 ± 5.5 to 320.00 ± 18.1 (wwt)	Kanyakumari Beach, South India	Rani et al. (2014)
Euthynnus affinis	92.5 ± 7.9 (wwt)	India	Khan and Wesley (2016)
Thunnus albacares	72.3 ± 9.7 (wwt)	India	Khan and Wesley (2016)
Thunnus tonggol	48.5 ± 12.3 (wwt)	India	Khan and Wesley (2016)
Katsuwonus pelamis	82.5 ± 10.3 (wwt)	India	Khan and Wesley (2016)
Auxis rochei rochei	56.6 ± 6.5 (wwt)	India	Khan and Wesley (2016)
Auxis thazard thazard	40.9 ± 5.2(wwt)	India	Khan and Wesley (2016)
Jellyfish	5×10^{1}Bq.Kg^{-1} (wwt)	North Atlantic	Carvalho (2011)
Sardines	3×10^{4} Bq.Kg^{-1}(w wt)		
Mytilus galloprovincialis	10.9 ± 0.9 Bq.Kg^{-1}(dw)	Bosphorus Strait	Kılıç et al. (2014)
Sardines	0.7 ± 0.2 Bq kg^{-1} (dw)	İzmir Bay	Kül et al. (2020)
Fish samples	undetectable levels to 499 ± 44 Bq kg^{-1} (dw)	Turkish coast of Aegean Sea.	Mat Çatal et al. (2012)
Tuna fish	37.3 - 44.9, 451 - 548(wwt)	northern Gulf waters	Uddin et al. (2017)
Shrimp	79.2 to 477 Bq Kg^{-1} (wwt)	Northern Gulf waters	Uddin et al. (2019)
Mytilus galloprovincialis	53 ± 4 and 1960 ± 60 Bq kg^{-1} (dw)	Turkish coast of the Aegean Sea.	Uğur et al. (2011)

The concentration of ^{210}Po detected in fish in this study showed a lower value when compared to the concentration of ^{210}Po in shellfish and fish found in several other countries (**Table 5**). This is due to the fish species in this study having different habitats from the fish and shellfish listed in **Table 5**, including tuna and shellfish that live on the bottom of waters. The presence of radionuclides, such as ^{210}Po, in marine waters that have the potential to become bioaccumulated is found both in the water and the bottom of the sediment. According to Muslim et al. (2017), the concentration of radionuclides in bottom sediments is higher and more stable than in waters that are use to changes due to diffusion and advection.

4. Conclusions

Based on the study results, it can be concluded that the content of ^{210}Po in six economically important fish caught in the waters of Semarang and Cirebon had values of 46.4 to 251.6 Bq/kg. These values are still below the minimum limit allowed by BAPETEN (2017) of 10^3 Bq/kg, so they are still safe for consumption. Skipjack and ponyfish had the highest ^{210}Po content compared to the other four samples, with content exceeding 20 Bq/kg, in which both fish contained higher protein, calcium, iron, omega-3, and zinc content than other fish. However, the results of the regression test for all fish (six species) did not show a significant relationship between the value of the content of ^{210}Po and the content of protein, calcium, iron, omega-3, and zinc, even though the fish lived in waters with relatively the same conditions.

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