



Determination of Natural Radionuclides (Ra-226, Po-210, Ra-228 and K-40) and Cs-137 in Fish Consumed in the City of São Paulo

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ABSTRACT

The aim of the study was to determine the activity concentration of the natural radionuclides (^{226}Ra , ^{210}Po , ^{228}Ra and ^{40}K) and artificial radionuclide ^{137}Cs in the muscle of the marine fish species most consumed in the city of São Paulo and to evaluate the annual effective dose due to the consumption of the fish. Samples were collected in the supermarket chain Extra and Carrefour and in the distribution center CEAGESP. Six fish species were selected among the most available and consumed in São Paulo city: anchovy, tuna, dogfish, croaker, hake and sardine. After the sample preparation procedures, the determination of ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs was carried out by gamma spectrometry and the determination of ^{210}Po by alpha spectrometry. The results obtained for all the species studied are below the limits adopted by the Brazilian Standards for ^{137}Cs and, therefore, their consumption offers no risk due to the ingestion of this radionuclide. The concentration obtained for the radionuclides ^{226}Ra , ^{210}Po , ^{228}Ra and ^{137}Cs in the fish samples analyzed are low and of the same order of magnitude as data from the literature. The results obtained for the doses, for all the species studied, showed that their consumption offers no risk of exposure due to the ingestion of the analyzed radionuclides.

Keywords: alpha spectrometry, gamma spectrometry, natural and artificial radionuclides, marine fish.



1. INTRODUCTION

The intake of safe and sufficient food to meet people's nutritional needs is not always a reality accessible to all, and such conditions can still worsen if the food presents contamination. Such a concern with the supply of unsafe or nutritionally inadequate food has become a concern for the entire chain involved in the process, from the commercial, economic and political dimensions to governments and the entire food industry, triggering an alert to the need for monitoring and control of population diets and the possible intake of contaminants [1].

Although the monitoring of chemical substances is done because of their nutritional importance and their health benefits, it is also essential that possible chemical contaminants are detected so that the concentration levels of toxic elements that can be consumed by the population are known. Such monitoring can be vital since inadequate consumption can generate serious health consequences such as cancer, deficiencies, and other damage to various organs. Thus, several studies and institutions have been concerned with this topic, reinforcing the importance of this evaluation [2, 3, 4].

Another institution that reinforces the importance and need for monitoring chemicals in food is the World Health Organization (WHO), highlighting that the population needs access to information about the food they consume, raising awareness regarding the possible toxic effects that some elements can cause to health. In addition, it is important to know which functions are the key elements for people's health [5].

Studies that identified and evaluated the biological effects of people's exposure to natural radionuclides, especially from the ^{238}U and ^{232}Th decay series [6, 7] reinforce the basis that natural and artificial radionuclides should be included in the list of contaminants to be monitored [8, 9].

1.1. Behavior of natural and artificial radionuclides in the marine environment

Like pesticides and heavy metals, marine organisms can also accumulate and concentrate radionuclides in their tissues. In the oceans, algae can accumulate radionuclides from the surrounding water, such as ^{40}K and ^{87}Rb by a factor of 1,000 to 5,000 times. Animals that feed on these algae tend to concentrate these elements even more and, in turn, be ingested by man and can

cause contamination of the human body by radionuclides considered harmful when ingested on a large scale [10].

Man and his environment have always been subject to ionizing radiation, being continuously exposed to radiation from space (cosmic radiation), radionuclides present in the soil (terrestrial radiation or primordial radionuclides) and in the consumption of food, air and water. The largest source of naturally occurring radioactive material (NORM) in the environment is the earth's crust, with radionuclides belonging to the decay chains of ^{235}U , ^{238}U and ^{232}Th and ^{40}K [11].

Natural radiation accounts for most of the radiation exposure (about 70%) to which the general population is subject. The average annual dose from natural sources is estimated to be 2.4 mSv. These include external sources, such as cosmic radiation and radioactive substances existing in the Earth's crust and building materials, and internal sources resulting from inhalation and ingestion of radioactive substances naturally existing in the air and the diet. It is important to make this distinction because terrestrial radiation is the largest source of natural irradiation, contributing about 85% to the average annual dose received by the population, of which 14% is due to ^{40}K , 17% from the natural series of ^{238}U and ^{232}Th and 53% because of radon [11].

The release of radioactive materials, resulting from anthropogenic activities such as the use of nuclear reactors and nuclear explosion tests, can eventually contribute by increasing radioactivity levels in the environment and increasing the radiation dose of exposed individuals, reaching values that can be considered significant.

Among the elements that are present in food and that can reach levels of contamination, we have natural radioactive isotopes or radionuclides. Such elements are evenly distributed in all compartments of the ecosystem (soil, air and water) [12] and can eventually be enriched on food. Fish, for example, can concentrate radionuclides, reaching levels considered toxic, depending on local factors such as land use and occupation, agriculture and industries [13].

Natural radionuclides can be found in the marine environment due to activities related to the oil, phosphate and fertilizer industries [14, 15]. An example of this is the discharges of ^{226}Ra and ^{228}Ra into the ocean in large volumes of water from the oil exploration industry [16].

Due to its high toxicity and physical, chemical and biological characteristics in the metabolism of humans, the elements ^{226}Ra and ^{228}Ra has great importance because, when incorporated into the

organism of humans, it has a chemical behavior similar to calcium, accumulating in bones and muscles [4, 17, 18].

The ^{210}Po is a natural radionuclide that can be found in nature as a result of the decay of the natural series of the ^{238}U . The main source of this radionuclide in the environment is due to the emanation of ^{222}Rn , which is released from the earth's crust into the atmosphere resulting in the deposition of ^{210}Pb and ^{210}Po through fall-out and rain-out.

^{210}Po accumulate in sediments, marine organisms, and fish, entering the aquatic food chain, which is an important source of food for man.

Marine biota, in particular fish and shellfish, may contain high concentrations of ^{210}Po , which is considered the largest contributor to the radiation dose of individuals [19]. Several studies conducted in several countries point out that the ^{210}Po can also accumulate in the estuary and aquatic system due to discharges from the phosphate, oil and gas industries, as well as the burning of fossil fuels [20].

The ^{137}Cs is one of the main artificial radionuclides launched into the marine environment as a result of nuclear weapons testing, nuclear reprocessing, nuclear accidents, marine discharges and river runoff [21]. The estimated atmospheric and marine discharge of this radionuclide into the environment in the accident at the Fukushima Daiichi Nuclear Power Plant was 7 to 20 PBq and 1 to 6 PBq, respectively [22].

The ^{137}Cs is present in the water column as simple ions, suffering a dispersal action due to dilution in the sea, and may participate in exchange reactions with suspended solids, especially clays. This radionuclide can be incorporated into muscle tissues of various marine organisms, such as fish, which can be ingested by man [23]. According to Carvalho [24], due to radioactive discharges in the marine biota, this radionuclide accumulates in fish and other species existing in the sea.

1.2. Fish importance in the diet in Brazil

In the marine environment, several natural and artificial radionuclides can be discharged with a variety of concentration levels and, with increased fish consumption in Brazil [25], may also present risks if there is any type of contamination. Thus, fish, which are a food rich in proteins and nutrients

necessary for human health and of excellent nutritional quality, are monitored for possible contamination.

One of the possible ways to monitor data on food consumption is to use research studies and technical reports that provide information about the consumption of this type of food. This research used the report published by the SOS Mata Atlântica Foundation, which analyzed the fish species most available in different types of supermarkets in the cities of São Paulo and Rio de Janeiro [26].

The average annual consumption of fish in Brazil, 9 kg/inhabitant /year, was taken from the report of “Ministério da Agricultura, Pecuária e Abastecimento” [27].

Taking into account the information available in Estrella et al. [26] six fish species were selected for analysis in this study: bluefish (*Pomatomus saltatrix*), tuna (*Thunnus spp.*), Smooth-hounds nei (*Mustelus spp.*), croaker (*Argyrosomus regius*), hake (*Merluccius merluccius*) and sardine (*Sardina pilchardus*). All the species selected are from saltwater, salmon and tilapia were excluded because they are species cultivated in captivity, as well as all kinds of seafood and other species that have low consumption by the population.

The places chosen for this study were the two larger chains of supermarket distribution in São Paulo (Extra and Carrefour Supermarket) and also the “Companhia de Entrepostos e Armazéns Gerais de São Paulo” (CEAGESP), which is a major distributor for restaurants and open markets.

In this context, the objective of this study was to determine the activity concentration of natural radionuclides (^{226}Ra , ^{210}Po , ^{228}Ra and ^{40}K) and artificial radionuclide ^{137}Cs in the muscle of the most consumed saltwater marine fish species in the city of São Paulo and evaluate the annual effective dose due to the consumption of the fish.

These objectives were chosen due to the importance of analyzing the concentration of these elements in the food chain, particularly in fish, and evaluating the exposure of the population resulting from their consumption.

2. MATERIALS AND METHODS

Twenty-four samples were collected from the Extra and Carrefour supermarkets and in the CEAGESP distribution center, located in the northern area of the city of São Paulo. In all of them, the request was to clean the fish, taking to the laboratory only the edible part of the fish chosen.

2.1. Determination of ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs by gamma spectrometry

Approximately 1 kg of muscle samples was dried by calcination at 450°C resulting in about 45g of ash, the calcination process took 48 hours per sample.

The activity concentration of radionuclides ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs in fish samples was measured by gamma spectrometry with the Hyper Pure Germanium HPGe detector, GX4020 with 47% relative efficiency with associated electronics and coupled to a microcomputer.

For the determination of ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs , the fish ash was packed in a polyethylene bottle and sealed for about four weeks before the measurement, to ensure that the equilibrium was achieved between the ^{226}Ra (half-life 1600 years) and its short half-life decay products. In the determination of ^{226}Ra (alpha emitters), it is assumed that it is in equilibrium with its decay products ^{214}Pb and ^{214}Bi , gamma emitters. The activity of the ^{226}Ra was determined by the activity of the peaks of ^{214}Pb , which emits gamma energies of 295.2 keV and 351.9 keV, and the peak of ^{214}Bi , which emits gamma energies of 609.3 keV and 1120.3 keV, respectively. The ^{228}Ra was determined by measuring the gamma energies of 911.07 keV and 969.11 keV of the ^{228}Ac , the ^{40}K by measuring the peak of 1460 keV and the ^{137}Cs by measuring the peak of 661.6 keV.

The Maestro multichannel emulator program [28] was used to obtain gamma spectra. The spectra analysis was made with the Winner Gamma program on the InterWinner software [29].

Background radiation was estimated by ultrapure water measurement. The determination of the minimum detectable concentration (MDC) was made using the model proposed by Currie [30]. The performance of gamma ray spectrometry measurements was evaluated through participation in Proficiency Tests (PTs) offered by the Instituto de Radioproteção e Dosimetria (IRD), available 3 times a year.

For the calculation of the activity concentration of ^{226}Ra , ^{228}Ra , ^{40}K and ^{137}Cs the following equation was used:

$$C = \frac{\text{Area} - BG}{m \cdot t \cdot Ef \cdot I\gamma}$$

(1)

Where:

C: concentration of radionuclide in the sample (Bq.kg^{-1});

Area: net peak area of the gamma transition (counts);

BG: net area of the background radiation for the gamma transition considered (counts);

m: sample mass (kg);

t: counting time (s);

Ef: counting efficiency for the energy of the gamma transition considered (cps.dps^{-1});

I γ : absolute intensity of the gamma transition considered (%).

2.2. Determination of ^{210}Po by alpha spectrometry

For the determination of ^{210}Po by alpha spectrometry, the samples were not dried by calcination to avoid ^{210}Po losses by volatilization. The samples were dried in a stove with controlled air circulation of 80°C , so that the loss of polonium isotopes did not occur.

Approximately 1g of each sample (dry weight) was used. Before the analysis, $100\mu\text{L}$ of ^{209}Po tracer (0.6483 Bq.g^{-1}) was added. For acid dissolution of the sample, 10 mL of concentrated HNO_3 and hydrogen peroxide were added under heating at 80°C . The solution was carefully evaporated until almost dry. This procedure was repeated until the sample was completely dissolved. Concentrated HCl was added to the final solution, to eliminate nitrates. The final residue was dissolved in HCl 0.5M and filtered in Millipore $0.1 \mu\text{m}$; 20% hydroxylamine hydrochloride was added to the solution. The pH was adjusted to 1.5 and Po was spontaneously deposited in the silver disc at 80°C for 4h, with the agitation of the solution. The prepared sources were counted in a Canberra Alpha Analyst surface barrier detector for 150,000 seconds.

The equation used to determine the concentration of ^{210}Po was:

$$C = \frac{Rn - Rb}{Ef \cdot Rq \cdot m}$$

(2)

Where:

C: concentration (Bq.kg^{-1});

Rn: counting rate in the isotope region (cps);

Rb: blank count rate in the region considered (cps);

Ef: detector counting efficiency (cps.dps⁻¹);

Rq: chemical yield;

m: sample mass (kg).

2.3. Internal exposure assessment due to ingestion of fish

Radioactive materials remain in the body for a certain period after being taken into the body. In the meantime, the body will be continuously exposed to radiation. Thus, the total amount of radiation that a person will be exposed to in the future is calculated as dose due to internal exposure based on a single intake of radioactive materials. This is called a committed dose (in sieverts).

Any radioactive materials taken into the body will decrease over time. One contributing factor is the decay of radioactive materials. Another is excretion as urine and feces. The rate of excretion from the body varies according to the types of elements, their chemical forms, and the age of the person. With these differences taken into account, the cumulative amount of radiation that the human body will receive in a lifetime from radioactive materials is assumed as the amount received in the year of the intake, and a committed dose is calculated.

In particular, the lifetime cumulative dose based on the effective dose is called “committed effective dose”. The lifetime here is 50 years for adults, and for children it is the number of years up to reaching age 70.

The concept of ‘effective dose’ (E) was developed by ICRP Publications 119 [31] as a risk-adjusted dosimetric quantity for the management of protection against stochastic effects, principally cancer. Dosimetric models to estimate internal exposures in humans, subsequent to incorporation of radionuclides via ingestion and inhalation, was developed by the International Commission on Radiological Protection (ICRP). The most recent models for ingestion are described in ICRP Publications 119 [31]. The endpoints of those models are dose coefficients for ingestion, which are calculated for six age groups (3-month-old children; 1, 5, 10 and 15-year-old children; and adults).

The committed effective dose to fish consumers was evaluated according to the following equation:

$$E = (CixQxDC)$$

(3)

Where:

E: is the annual effective dose (Sv.y^{-1}),

C_i : is the activity concentration of radionuclide i in fish (Bq.kg^{-1}),

Q: annual intake of fish (kg.y^{-1}),

DC: is the dose coefficient (Sv.Bq^{-1}).

The annual intake of fish was assumed as 9 kg.y^{-1} from “Ministério da Agricultura, Pecuária e Abastecimento” [27].

3. RESULTS AND DISCUSSION

The mean activity concentration obtained for radionuclides ^{226}Ra , ^{210}Po , ^{228}Ra and ^{137}Cs in the fish samples and the annual effective dose are presented in Table 1.

Table 1: Mean activity concentration (Bq.kg^{-1} wet weight) for ^{226}Ra , ^{210}Po , ^{228}Ra , ^{40}K and ^{137}Cs , and annual effective dose (E) ($\mu\text{Sv.y}^{-1}$)

Fish sample	^{226}Ra	^{210}Po	^{228}Ra	^{40}K	^{137}Cs	E (total)
Bluefish	0.08 ± 0.01	3.87 ± 0.19	0.18 ± 0.04	89 ± 12	< 0.04	
E	0.20	8.36	1.09	4.97	-	14.6
Tuna	0.09 ± 0.02	5.34 ± 0.29	< 0.16	116 ± 19	0.12 ± 0.02	
E	0.23	11.5	-	6.47	0.01	18.2
Smooth-hounds nei	0.09 ± 0.03	2.02 ± 0.09	0.19 ± 0.05	94 ± 4	0.06 ± 0.01	
E	0.23	4.36	1.15	5.25	0.01	10.1
Croaker	0.12 ± 0.03	1.86 ± 0.08	0.21 ± 0.06	93 ± 6	0.06 ± 0.01	
E	0.30	4.02	1.27	5.19	0.01	10.8
Hake	0.10 ± 0.03	1.42 ± 0.06	0.18 ± 0.04	77 ± 3	0.17 ± 0.05	
E	0.25	3.07	1.09	4.30	0.02	8.7
Sardine	0.11 ± 0.03	4.12 ± 0.23	0.20 ± 0.05	102 ± 4	0.05 ± 0.01	
E	0.28	8.90	1.21	5.69	0.01	16.1

All the species analyzed presented ^{226}Ra , ^{228}Ra and ^{137}Cs concentrations low or close to the detection limits of the equipment used for the measurements. The ^{40}K presented the higher concentrations ranging from $77 \pm 3 \text{ Bq.kg}^{-1}$ wet weight (ww) to $116 \pm 19 \text{ Bq.kg}^{-1}$ (ww), followed by Po-210, ranging from $1.42 \pm 0.06 \text{ Bq.kg}^{-1}$ (ww) to $5.34 \pm 0.29 \text{ Bq.kg}^{-1}$ (ww).

The Brazilian regulatory guide “Posição Regulatória 3.01/006” of 2011 entitled “Protective Measures and Intervention Criteria in Emergency Situations” established the recommended action levels for food control [32]. According to this standard, the concentration of ^{137}Cs in food cannot exceed the value of $1 \text{ kBq}\cdot\text{kg}^{-1}$. The results obtained for this radionuclide are below this limit.

The highest doses were found for ^{210}Po , ranging from $3.07 \text{ }\mu\text{Sv}\cdot\text{y}^{-1}$ to $11.5 \text{ }\mu\text{Sv}\cdot\text{y}^{-1}$ (50% of the total dose), which was considered the most relevant radionuclide as far as internal exposition due to ingestion of fish; followed by ^{40}K , $4.30 \text{ }\mu\text{Sv}\cdot\text{y}^{-1}$ to $6.47 \text{ }\mu\text{Sv}\cdot\text{y}^{-1}$, (42% of the total dose); and ^{228}Ra , $1.09 \text{ }\mu\text{Sv}\cdot\text{y}^{-1}$ to $1.27 \text{ }\mu\text{Sv}\cdot\text{y}^{-1}$ (8% of the total dose). All components of the diet contain the radionuclide of natural origin ^{40}K . Potassium is a key element in regulating many body functions such as digestion and heart rate and the potassium content of the body is kept constant by metabolic processes. Potassium naturally contains 0.12% by weight of ^{40}K , and so the content of ^{40}K in the body is also regulated naturally [33]. UNSCEAR has estimated that the annual effective dose due to the presence of ^{40}K in the body is typically about $165 \times 10^{-3} \text{ mSv}$ for adults. Therefore, no control can reasonably be exercised over the dose from ^{40}K in the diet [11].

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has reviewed the available scientific data on the doses from ingestion of radionuclides of natural origin in food. The “reference values” of activity concentrations of ^{226}Ra and ^{210}Po in fish for use in the assessment of dose are 0.1 and $2 \text{ Bq}\cdot\text{kg}^{-1}$ dry weight (dw). When the values of all radionuclides of natural origin are combined with typical consumption rates for each food group, the annual individual doses from radionuclides in the uranium and thorium series in the diet (considering only food and drinking water together) is 0.11 mSv for adults. The bulk of this dose comes from the food component of the diet, and the radionuclides which contribute the bulk of this dose are ^{210}Po , ^{210}Pb and, to a lesser extent, ^{228}Ra . [11].

The results obtained in this study for the activity concentration in fish were compared with data from the literature (Table 2). In general, the radionuclide’s concentration in this study is in good agreement with values from literature, except for the concentration of ^{210}Po found in sardine by Carvalho (1995), $66 \pm 2 \text{ Bq}\cdot\text{kg}^{-1}$ [34], which is one order of magnitude higher than our results. Our results of ^{226}Ra activity concentration ($0.63 \text{ Bq}\cdot\text{kg}^{-1}$ to $1.90 \text{ Bq}\cdot\text{kg}^{-1}\text{dw}$) and of ^{210}Po ($9.42 \text{ Bq}\cdot\text{kg}^{-1}$ to $19.20 \text{ Bq}\cdot\text{kg}^{-1}\text{dw}$) on a dry basis are higher than the corresponding UNSCEAR “reference values” 0.1 and $2 \text{ Bq}\cdot\text{kg}^{-1}$ (dw).

Table 2: Range of concentration (Bq.kg⁻¹ wet weight) of ²²⁶Ra, ²¹⁰Po, ²²⁸Ra, ⁴⁰K and ¹³⁷Cs in fish obtained in this study and literature data

Reference	²²⁶ Ra	²¹⁰ Po	²²⁸ Ra	⁴⁰ K	¹³⁷ Cs
This paper	0.08 - 0.12	1.42 - 5.34	0.18 - 0.21	77 - 116	<0.05 - 0.17
UNSCEAR, 2000 [11]	0.1*	2*			
Nisti et al., 2019 [35]	<0.7 - 0.58		<0.31 - 0.71		<0.08 - 0.13
Godoy et al., 2003[36]					0.03 - 1.7
Cunha et al., 1993 [37]					0.1 - 0.3
Görür et al., 2012 [38]	0.06 - 0.96			35.04 - 127.41	0.06 - 1.53
Carvalho, 1995 [34]		6.7 - 66			
Giri et al., 2010 [39]	0.052 - 0.83	<0.2 - 3.19			

*Concentration in Bq.kg⁻¹ dry weight.

The results obtained for the doses were compared with literature values and are depicted in Table 3. The values of committed effective doses for the radionuclides analyzed are in agreement with the literature values and do not compromise the health of the population.

Table 3: Total annual effective dose (E) (μSv.y⁻¹) due to ingestion of ²²⁶Ra, ²¹⁰Po, ²²⁸Ra, ⁴⁰K and ¹³⁷Cs in fish obtained in this study and literature data

Reference	Radionuclides	E
This paper	²²⁶ Ra, ²¹⁰ Po, ²²⁸ Ra, ⁴⁰ K and ¹³⁷ Cs	8.7 – 18.2
Cunha et al., 1993 [37]	¹³⁷ Cs	0.013
Görür et al., 2012 [38]	²²⁶ Ra, ²³² Th, ⁴⁰ K and ¹³⁷ Cs	5.67 - 19.21
Giri et al., 2010 [39]	U ^(nat) , ²²⁶ Ra, ²³⁰ Th and ²¹⁰ Po	0.01 - 3.26

4. CONCLUSION

Monitoring of natural and artificial radionuclides in food is important for the knowledge of possible chemical contaminants that can be consumed by the population and to verify the quality of fish consumed in the city of São Paulo. The concentration obtained for the radionuclides ²²⁶Ra, ²¹⁰Po, ²²⁸Ra and ¹³⁷Cs in the fish samples analyzed are of the same order of magnitude as data from

the literature. The results obtained for the doses for all the species studied showed that their consumption offers no risk of exposure due to the ingestion of the analyzed radionuclides.

Furthermore, the values obtained in this study, both for activity concentration and committed effective dose, serve as a database for future research and monitoring regarding the most consumed marine fish in the city of São Paulo.

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