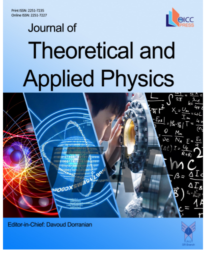

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Radiometric analysis of the meat and skin of shrimps and some farmed and marine fish from the Persian Gulf

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Original Research

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Abstract:

Nuclear radiation from radioactive elements may cause genetic abnormalities or causes dangerous diseases such as cancer. For this reason, it is essential to measure food contamination with radionuclides, especially seafood prepared from areas contaminated with industrial pollution. In this case, it is necessary to calculate the risk of developing cancer and determine the permissible amount of their consumption. In this work, the specific activity of 4 radioactive elements ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs was measured in 18 meat and skin samples, including 3 species of farmed fish, such as Cyprinus carpio, Pomadasys kaakan and Epinephelus multinotatus from the Persian Gulf, and 2 species of shrimp, including the marine Metapenaeus ensis and the farmed Litopenaeus vannamei. The results indicate that the specific activities of radionuclides in seafood, skins were higher than in meat, and the maximum concentration of ^{226}Ra, ^{232}Th, and ^{40}K was observed in the skins of Cyprinus carpio (8.34 Bq/kg), Metapenaeus ensis (31.61 Bq/kg) and Epinephelus multinotatus (404.26 Bq/kg). For all samples, the amount of ^{137}Cs was below the detectable limit.

Keywords: Gamma spectroscopy; Shrimp; Fish; Radioactive elements

1. Introduction

The Persian Gulf is a semi-closed marine environment due to the extensive exploitation of huge oil reserves and the route of many transfers of petroleum materials and oil tankers, the pollution load per square kilometer of the surface of the Persian Gulf is more than the global. Among the environmentally important pollutants, heavy metals and radioactive elements can be mentioned [1]. As a result of advances in the application of nuclear physics, the increase in the number of active reactors, the development of the nuclear industry, nuclear accidents such as Chernobyl and Fukushima, and nuclear weapons testing, especially in the oceans, the amount of radioactive contamination on the earth's surface such as soil, rocks, and water has increased and the water in the oceans has increased significantly. And the entry of these substances into the human body through the digestive cycle can lead to skin damage, internal bleeding, genetic diseases, and cancer, so it is very important to measure the activity of radioactive elements in food, especially in seafood. Today, man-made radioactive elements have been added to natural radioactive sources [2]. In recent years, many efforts have been made to measure natural and artificial radioactive substances in food, demonstrating their importance for the health of people in society. From the past to the present, seafood has always had a special place in the human food basket, and its consumption has increased due to the increase in population, the development of technology, and the increase in the human need for food. The per capita consumption of fish in Iran in 2018 reached about 10.6 kg per year [3]. And the per capita consumption of shrimp in Iran this year is 0.487, which is much lower than the world average [4]. People are exposed to natural and artificial radioactive sources, which can be exposed both internally and externally. Internal sources are much more dangerous than external sources. Consuming contaminated food causes radioactive substances to enter the body and deposit in various tissues of the body.

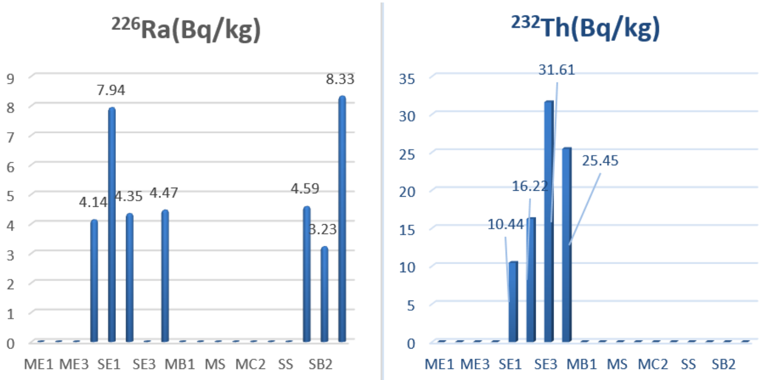

Figure 1. Graph of ^{226}Ra and ^{232}Th concentration in samples in Bq/kg for dry weight.

2. Material and methods

In this study were collected and measured radionuclides in 18 meat and skin samples, including 3 species of farmed fish, such as Cyprinus carpio, Pomadasys kaakan and Epinephelus multinotatus from the west of Persian Gulf, and 2 species of shrimp, including the marine Metapenaeus ensis and the farmed Litopenaeus vannamei from Bushehr region. After collecting the samples, it is time to prepare them. For this purpose, edible (meat) and non-edible parts, including skin, scales, head, tail, fins, etc., are separated and then washed with distilled water to remove external contamination on the samples. After accurate measurement of the mass of each part, to dry the samples quickly, pieces of fish meat and shrimp samples were cut into pieces and dried using a freeze dryer for 24 hours so that the samples are free of moisture. Due to the high-fat content of the non-edible parts, the fish were dried using a thermal oven at a constant temperature of 80 degrees Celsius for a week [5]. The homogeneous powder was prepared using a sterilized mill. Then 200 grams of each powder sample was packed and coded in special containers. All codes of meat samples and skin samples were prefixed with *M* and *S*, respectively. To prevent leakage of radon gas, the containers were sealed with silicone glue. And they were kept in the lab for at least 60 days until the elements in the uranium chain reached in full equilibrium [6]. To determine the specific activity of radioactive nuclei in the samples, the gamma-ray spectrum of each sample was recorded. Spectrometry was performed using an HPGe coaxial detector model GCD30195 manufactured by Baltic Scientific Instruments (BSI) company with a relative efficiency of 30% and an energy resolution of 1.95 keV for the cobalt gamma line of 1332 keV belonging to Cobalt 60 [7]. Sample containers were placed in two protective layers of lead and copper with a thickness of 10 cm and 2 mm. Cosmic rays were reduced to a very low level

Table 1. Measurement results of radioactive elements in samples (Bq/kg).

sample code	specific activity (Bq/kg)			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
ME1(Litopenaeus vannamei) long	< 2.35	< 4.21	288.46 ± 8.45	< 0.92
ME2(Metapenaeus ensis) short	< 1.91	-	182.64 ± 7.53	< 0.37
ME3(Metapenaeus ensis) long	< 2.33	-	199.82 ± 7.79	< 0.93
ME4(Litopenaeus vannamei) short	4.14 ± 0.27	< 4.36	245.76 ± 7.64	< 0.67
MB1(Epinephelus multinotatus)	< 2.48	< 4.34	12.10 ± 404.26	< 0.98
MB2(Epinephelus multinotatus)	< 2.34	< 4.24	10.84 ± 376.21	< 0.80
MS(Pomadasys kaakan)	< 2.40	< 3.82	8.44 ± 279.11	< 0.81
MC1(Cyprinus carpio)	< 1.19	< 3.11	6.06 ± 140.61	< 0.44
MC2(Cyprinus carpio)	< 2.41	< 4.15	7.04 ± 140.94	< 0.67
MC3(Cyprinus carpio)	< 0.86	< 1.34	6.05 ± 141.71	< 0.22
SE1(Litopenaeus vannamei) long	7.94 ± 0.87	10.44 ± 1.16	244.48 ± 7.08	< 0.82
SE2(Metapenaeus ensis) long	4.35 ± 0.16	16.22 ± 1.94	81.82 ± 4.66	< 0.51
SE3(Metapenaeus ensis) short	< 2.25	31.61 ± 2.48	84.48 ± 4.71	< 0.76
SE4(Litopenaeus vannamei) short	4.47 ± 0.24	25.45 ± 2.62	135.74 ± 5.61	< 0.83
SS(Pomadasys kaakan)	< 3.30	< 6.19	7.43 ± 153.50	< 0.82
SB1(Epinephelus multinotatus)	0.33 ± 4.59	< 4.10	4.44 ± 67.66	< 0.76
SB2(Epinephelus multinotatus)	0.31 ± 3.23	< 6.85	10.37 ± 185.06	< 0.66
SC(Cyprinus carpio)	0.26 ± 8.33	< 7.42	6.69 ± 76.22	< 1.27

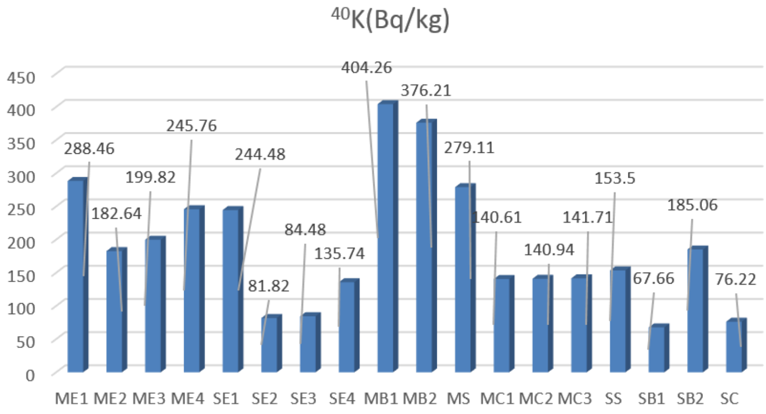

Figure 2. Graph of ^{40}K concentration in samples in Bq/kg for dry weight.

using a 100 mm thick lead shield [8]. To reduce the effect of radiation scattering, the detector was placed in the center of the shield, and the gamma ray spectra was registered using LsrmBSI manufactured by Baltic Scientific Instrument company (00-5- Latvia) software for 24 hours (86400 seconds). It is a program that distinguishes and collects pulses according to the height of the pulse in special channels. All recorded spectra were analyzed using Gamma Vision32 Master II software, a product of EG&G Ortec. Energy and efficiency calibration of the detector system was performed using sources known to contain ^{241}Am, ^{137}Cs, and ^{152}Eu radionuclides with known activities using Equation 1 [9].

$$\varepsilon(\%) = \frac{N_i}{Act \times P_n(E_i) \times t} \times 100 \tag{1}$$

where N_i represents the net number under the full energy peak corresponding to the energy of E_i, Act is the activity of the radioisotope at the time of measurement, $P_n(E_i)$ is the probability of emission of the photon E_i and t is the counting time [7]. To determine the specific activity of ^{226}Ra in the samples, gamma rays with an energy of 351.93 keV belonging to ^{214}Pb and 609.31 keV gamma line belonging to ^{214}Bi were used. The specific activity of ^{232}Th was determined using the ^{228}Ac gamma lines with energies of 911.21 keV with an intensity of 26.6% and an energy of 968.97 keV with an emission percentage of 17.4%. The specific activity of ^{40}K and ^{137}Cs was assessed using their gamma lines of 1460.70 keV and 661.66 keV respectively [10]. According to the spectra analysis results, the specific activity was determined using Gamma Vision Master II manufactured by EG&G Ortec company using Equation 2.

$$Act = \frac{Net\ \ Area}{\varepsilon \times BR(\%) \times T \times m} \times 100 \tag{2}$$

where $Net\ \ Area$ denotes the net counts under the peak, Act (Bq/kg) is the activity concentration, ε represents the energy efficiency for the gamma-ray by the detector, BR signifies the branching ratio of gamma-ray intensity (%), T (s) is the time of spectra, and m (kg) indicates sample mass [8].

3. Results and discussion

The measurement results are summarized in Table 1. The concentration of ^{226}Ra in meat samples ranged from < 0.86 to 4.14 Bq/kg detected only in small-sized Litopenaeus vannamei meat. For other meat samples, the amount of ^{226}Ra was below the minimum detectable activity (MDA), the amount of MDA marked with $<$ is also given in Table 1. In skin samples, the amount of ^{226}Ra ranged from $<$ 0.2.25 to 8.33 Bq/kg, which was below the MDA in only

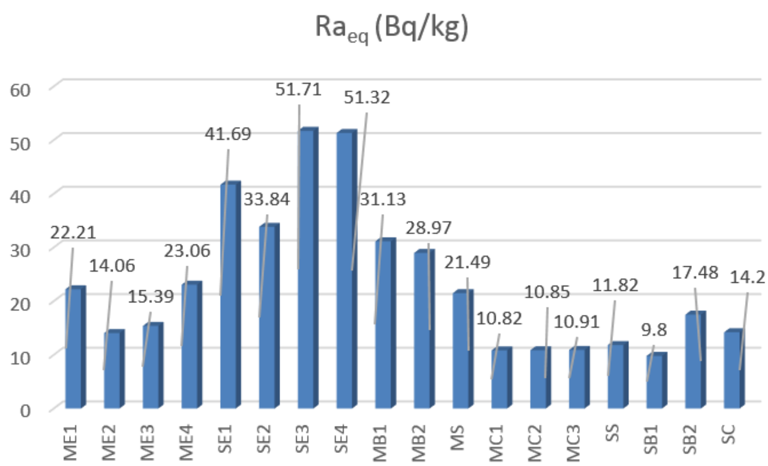

Figure 3. histogram of Ra_{eq} value for different samples.

Table 2. Comparison of the amount of natural radionuclides in shrimp samples meat (Bq/kg).

Name of Species	^{226}Ra	^{232}Th	^{40}K	References
Penaeus Monodon (Bay of Bengal)	1.15 ± 0.20	1.48 ± 0.28	12.62 ± 2.55	[11]
Metapenaeus Monoceros (Bay of Bengal)	0.80 ± 0.16	1.02 ± 0.15	7.27 ± 1.70	[11]
Panulirus Versicolor (Bay of Bengal)	1.00 ± 0.17	0.95 ± 0.79	12.13 ± 2.48	[11]
ME1(Litopenaeus vannamei) long (Iran)	< 2.35	< 4.21	288.46 ± 8.45	This work
ME2(Metapenaeus ensis) short (Iran)	< 1.91	-	182.64 ± 7.53	This work
ME3(Metapenaeus ensis) long (Iran)	< 2.33	-	199.82 ± 7.79	This work
ME4(Litopenaeus vannamei) short (Iran)	4.14 ± 0.27	< 4.36	245.76 ± 7.64	This work

two samples. Almost all skin samples detected ^{226}Ra, the maximum of which is in the long-sized fish Litopenaeus vannamei (7.94 Bq/kg) and the shrimp sample Cyprinus carpio (8.33 Bq/kg). These results show that ^{226}Ra was more absorbed in the skin of fish and shrimp. Comparing the amount of ^{226}Ra in skin samples of Litopenaeus vannamei long and short size showed that long life in this zone caused more absorption of radium in the skin. Thorium in all meat samples was below the MDA, but in some skin samples, it was high. The amount of ^{232}Th in the skins ranged from < 4.10 to 31.61 Bq/kg. These results showed that thorium and radium accumulate more in the skin of fish and shrimp samples.

^{40}K radionuclides ranged from 140.94 to 404.26 Bq/kg in meat and from 76.22 to 244.48 in skin samples. The average of ^{40}K in the meat samples was 239.95, while the skin average was 128.58 Bq/kg, indicating that the absorption of potassium was almost twice as high in the meat samples as in the skin samples. In all meat and skin samples, the concentration of ^{137}Cs was below the MDA, and the amount of MDA is shown in Table 1. For a better understanding of the changes in the radioactivity of the samples, the histogram of the concentration of ^{226}Ra, ^{232}Th, and ^{40}K is shown in Figures 1 and 2. Tables 2 and 3 compare the results of this work with tested samples from the Bay of Bengal and Nigeria, which, in addition to their particular location, are connected to open waters and are therefore exposed to regional and global pollution.

3.1 Radium equivalent

Since the distribution of natural radionuclides in the analyzed samples is not uniform, a radiological index called radium equivalent activity (Ra_{eq}) has been defined to estimate the radiation risk associated with these radionuclides. This parameter represents the total radioactivity of the sample expressed in ^{226}Ra equivalent, which allows comparing the total radioactivity. This indicator was calculated using the Equation 3 [14].

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{3}$$

where A_{Ra}, A_{Th} and A_K are the specific activities (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K in the studied samples. The results of this calculation presented in Figure 3.

3.2 Average annual effective dose

Due to the ingestion of naturally occurring radioactive materials (NORM) in edible foods, the average annual effective dose (AAED) was estimated using Equation 4 [11].

$$\text{AAED} = \sum_i Cr \times \text{DCF}_i \times A_i \tag{4}$$

Table 3. Comparison of the amount of ^{40}K radionuclides in fish samples meat (Bq/kg).

Name of species	^{40}K	References
MB1(Epinephelus multinotatus) (Iran)	12.10 ± 404.26	This work
MB2(Epinephelus multinotatus) (Iran)	10.84 ± 376.21	This work
MS(Pomadasys kaakan) (Iran)	8.44 ± 279.11	This work
MC1(Cyprinus carpio) (Iran)	6.06 ± 140.61	This work
MC2(Cyprinus carpio) (Iran)	7.04 ± 140.94	This work
MC3(Cyprinus carpio) (Iran)	6.05 ± 141.71	This work
T.fuscatus var.radula (Nigeria)	91.7 ± 5.6	[12]
Ergeria radiata (Nigeria)	132.7 ± 8.6	[12]
Ethmalosa fimbriata (Nigeria)	37.4 ± 5.2	[12]
Penaeus notialis (Nigeria)	71.8 ± 6.6	[12]
EuthnnusAffnis (Bay of Bengal)	27.35 ± 6.23	[13]
OtolihesArgentues (Bay of Bengal)	54.64 ± 9.05	[13]
StromateusSinensis (Bay of Bengal)	37.34 ± 7.24	[13]
Hilish llisha (Bay of Bengal)	43.32 ± 7.27	[13]

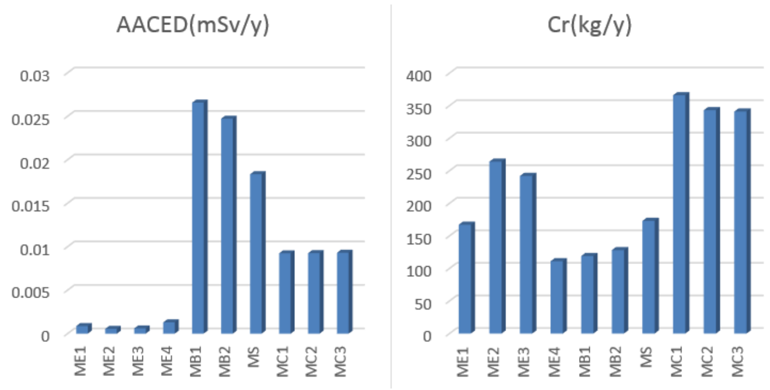

Figure 4. The histogram of AACED and Cr values for samples.

where Cr is the consumption rate of food in year fish (10.6 kg/y), shrimp (0.487 kg/y), DCF_i represents the dose conversion factor for each radionuclide (2.8×10^{-7}, 2.3×10^{-7}, 6.2×10^{-9}, and 1.3×10^{-8} Sv/Bq for ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs, respectively), and A_i signifies the activity concentration of each radionuclide.

3.3 Consumption rate

This is the amount of food a person eats in a year. If this type of food contains radionuclides, they cause an absorption dose as an internal source. UNSCARE estimated the global average annual effective dose rate from food to be 0.3 mSv/y [15]. In this sense, it is possible to calculate the amount of specific food that can cause this amount of dose absorbed by the body. The average annual consumption can be calculated by considering the average yearly absorbed dose through food (0.3 mSv/y) using Equation 5 [16].

$$Cr = \frac{E_{ave}}{\sum_{i=1}^{4} DCF_i \times A_i} \tag{5}$$

where E_{ave} (0.3 mSv/y) is the average annual effective dose, A_i is the specific activity of radioactive nuclei and DCF_i is the same quantity mentioned in Equation 4 [16].

3.4 Excess lifetime cancer risk assessment

The estimated excess lifetime cancer risk (ELCR) for consuming foods containing radioactive elements was calculated using Equation 6 [12].

$$ELCR = Aing.DL.RF \tag{6}$$

Where $Aing$ denotes the annual consumption rate of radionuclides (Bq/kg), DL is the mean lifetime (year), and RF represents the risk factors of radionuclide ingestion (1/Bq). RF values of radionuclides ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs were determined to be 4.80×10^{-8}, 2.30×10^{-7}, 5.90×10^{-9}, and 1.3×10^{-8}, respectively [12]. The amount of ELCR, AAED, and Cr is given in Table 4 and shown in Figure 4 for better visibility. In Figure 4, the AAED value for fish samples including MB1, MB2, Ms, MC1, MC2, and MC3 samples was much higher than shrimp meat, indicating that shrimp meat was safer and better quality. The maximum allowable value for the ELCR value is 10^{-3} and is exceeded for samples MB1, MB2 and MS, which means that the probability of developing cancer for these samples is estimated at 1 in 568 to 819 people. It is expected that the number of illnesses for shrimp consumption will be approximately one person per 170,000 inhabitants.

4. Conclusion

This study measured the specific activity of some common fish and shrimp in the Persian Gulf region. The results showed that the skin of fish and shrimp is a good protector, and most of the radionuclides, except the potassium radionuclide, are concentrated in the skin. Meat consumption

Table 4. Radiological parameters of fishes meat and maximum annual consumption in kg.

sample code	AAED (μSv/y)	Cr (kg/y)	ELCR ($\times 10^{-3}$)
ME1	0.87	167	0.058
ME2	0.55	264	0.036
ME3	0.60	242	0.04
ME4	1.30	111	0.056
MB1	26.57	119	1.76
MB2	24.72	128	1.64
MS	18.34	173	1.22
MC1	9.24	366	0.62
MC2	9.26	343	0.62
MC3	9.31	341	0.62

at this level poses no risk to human health, but it would be better to measure the radionuclides concentration in the meat of other fish that do not have scales.

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Ethical approval
This manuscript does not report on or involve the use of any animal or human data or tissue. So the ethical approval is not applicable.

Authors Contributions
Reza Pourimani: Conceptualization, Methodology, Investigation, Data curation, Software, Formal analysis, Review & editing, Validation; Erfan Hatamabadi Farahani: Data curation, preparing the manuscript, Visualization, Writing-Original draft preparation, data analysis, investigation of references, and editing; Mohammad Reza Amiri Siavashan: Sampling and sample preparation, Data curation, Software, Formal analysis.

Availability of data and materials
Data presented in the manuscript are available via request.

Conflict of Interests
The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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