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Determination of Specific Natural Radionuclides in the Bones of Some Local Fish Commonly Consumed from the Eastern Libyan Coast

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Abstract: The concentrations of various radionuclides in fishbones of seven fish inhabiting nine different locations in the Eastern Coastline of Libya are investigated, a distance of about 500 km. These fish are: Epinephelus Marginatus (S_1), Pagellus Bogaraveo (S_2), Diplodus Vulgaris (S_3), Umbrina Cirrosa (S_4), Trachurus Mediterraneus (S_5), Balistes Carolinensis (S_6) and Seriola Dumerili (S_7). Assessment was made of the concentration activities of key indicator natural radionuclides, especially ^{40}K, ^{226}Ra, ^{234}Th and ^{235}U, using HPGe detector. The radioactivity concentration (in Bq/kg) ranged from 27.90±0.15 for S_3 to 255.00±0.02 for S_7 for ^{40}K; from <MDA for S_1 to 157.10±0.20 for S_6 for ^{226}Ra; from <MDA for S_2, S_3, S_4, S_5 to 53.30±1.16 for S_7 for ^{234}Th, while ^{235}U was detected only in S_1. The average concentrations (in Bq kg^{-1}) were 146.63, 86.40 and 16.84 for ^{40}K, ^{226}Ra and ^{234}Th, respectively for all samples. The obtained results for the levels of radionuclides were lower than the worldwide allowable limits. It is shown that the radiological hazard indices (I_γ, DR, $D_{outdoor}$, D_{indoor}, H_{in}, H_{ex} and Ra_{eq}) were determined and found to be less than the recommended safe limits given by UNSCEAR except H_{in} and I_γ for S_6 where the values were greater than one (>1). It is concluded that this increase may be of concern to consumers of this type fish.

Keywords: Concentration Activity, Eastern Coast of Libya, Fishbones, Natural Radioactivity, Radiological Hazard Indices.

1 Introduction

Radiological safety and assessment of any release of radioactivity to the environment are a prime concern and essential for the protection of public health. This concern is even more crucial when the released radioactivity may appear in the food chain. The detection of this radioactivity necessitates rapid, reliable and applicable procedures for determination of different radionuclides [1]. Naturally occurring radionuclides of terrestrial origin are found in the seawater of seas and in the marine biota in different proportions [2-5]. Seas act as sinks for the materials that flow across the aquatic chemical and biological cycles including radionuclide pollutants. Naturally occurring radionuclides, and associated external exposure due to gamma radiation, in environment contain uranium and thorium series radioisotopes and natural ^{40}K and are dominated primarily by the geological conditions and environmental formations of each specific location such as sea basins [6]. Some of the sources of such radioactive contamination of anthropogenic radionuclides to the marine environment are fallout atmospheric nuclear weapons testing, transport of radionuclides discharged from reprocessing plants and fallout from nuclear accidents such as Chernobyl and Fukushima accidents [7]. This represents the major input of these radionuclides in the seawaters and, in turn, to the fish both quantitatively and geographically. Therefore, it is difficult to estimate the impact on the eastern coast of Libya, the study area of the present work, from these sources both because the transport routes and the magnitude of transport are not sufficiently documented. To investigate radiological concerns about fish consumption in the eastern coast of Libya, particularly fishbones, this study is carried out. This is because previous studies concentrated only on the edible portions of fish [8-9]. The reason for this choice is that there are certain radionuclides which are the so called bone seekers. A bone seeker is an element, often a radioisotope, which tends to accumulate in the bones of humans and other animals when it is introduced into the body [10]. From the available literature, as far as the authors' best knowledge, there are no studies published on radionuclides in fishes in the study area and none on fishbones. Hence this paper is

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an attempt to fill this gap in the investigations and to determine the concentration activities of key indicator natural radionuclides, especially ^{40}K, ^{226}Ra, ^{234}Th and ^{235}U, as well as the relevant radiological hazard indices.

The present paper is organized as follows. In section 2, the materials and methods are presented where the study area, samples collection and preparation, the radioactivity determination and the evaluation of radiological hazard effects are demonstrated. In section 3, the results and discussion are presented. Finally, the results are summarized and concluded in section 4.

2 Materials and Methods

2.1 Study Area

The investigated area extended along the coast of eastern Libya (about 270 km along), from Benghazi (32˚12'92"N to 20˚09'16"E) in the west to Altememi (32˚33'35"N to 23˚07'69"E) in the east on the Mediterranean Sea. The study area is characterized mainly by a rocky shoreline and a border coastline plain with intermingled sandy beaches and tiny inlets (see Fig. 1).

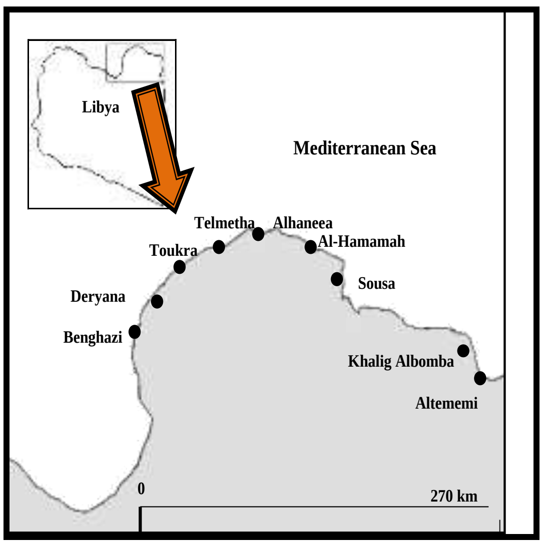

Fig. 1: Study area and locations of collected samples in the eastern coast Libya.

Seven samples of the commonly consumed fishes were collected from nine sites in the study area. Table 1 shows the locations of the collected edible fish samples. The locations are given the symbols L1 to L9 and the fish samples are indicated by the numbers 1 to 7, as shown in Table 1.

Table 1. Location of sampling sites in the study area.

Site	S. No.	East Longitude	North Latitude
Benghazi (L1)	2, 3	20˚09'16"E	32˚12'92" N
Deryana (L2)	2, 3	20˚32'86"E	32˚35'71" N
Toukra (L3)	4, 6	20˚60'07"E	32˚53'38" N
Telmetha (L4)	1, 3, 5	20˚14'13"E	32˚42'01" N
Alhaneaa (L5)	2	21˚47'44"E	32˚82'73" N
Alhamaa (L6)	4	21˚61'39"E	32˚90'73" N
Souse (L7)	7	21˚96'64"E	32˚90'69" N
Khalig Albomba (L8)	1	20˚94'88"E	32˚70'91" N
Altememi (L9)	6	23˚07'69"E	32˚33'35" N

2.2 Samples Collection and Preparation

Of the more than forty fish species recorded in the coastline waters of the eastern part of Libya seven fish types were selected in this work. Table 2 shows the seven sample fishes with their scientific, local and English names along with their collection locations. Samples were collected in different sizes and lengths to cover all fish ages. Then, the collected fishes were dried and washed several times with hot distilled water to remove the residue and soluble impurities. Fishes were left to dry in open air for enough time and then tissues were completely removed to end up with only fishbones. Next, the fishbones were dried in an oven at 105 ^{o}C, without a significant loss of any radionuclides except radioiodines, to be ashed. During ashing, low carbon nickel trays were used. The temperature for dry ashing varied but an upper recommended limit of 450°C was used [1]. Samples were then grinded into fine powder. The samples were packed in containers of specific geometry and were well sealed to prevent any loss of radium-isotope around the container walls. After packing, all samples were stored for a month to achieve secular equilibrium between radium and thorium and their progeny [11]. Table 3 shows the original fish weight, ashed weight and averaged ashed weight along with the average temperature at the time of collection. Gamma spectrometry system is chosen as analytical tool for use in this work. This procedure is considered reliable for sample types that particularly concern food intake.

Table 2. The seven analyzed fishes with their scientific, local and English names along with their sites and date of collection.

S. No.	Scientific name	Local name	English name	Site	Collection Date
1	Epinephelus Marginatus	AlFarooj	Dusky grouper	L4, L8	July, 2017 Dec., 2017
2	Pagellus Bogaraveo	Alhamria	red sea bream	L1, L2, L5	July, 2017
3	Diplodus Vulgaris	Algaragoz	two-banded sea bream	L3, L6	July, 2017 April, 2018
4	Umbrina Cirrosa	Albaghla	Shi drum (Corb)	L4	July, 2017 May, 2018
5	Trachurus Mediterran-eus	Alsauro	Mediterra-nean horse mackerel	L3, L9	July, 2017 May, 2018
6	Balistes Carolinensis	Alhalouf	Trigger-fish	L7	July, 2017
7	Seriola Dumerili	Alshoula or Alburiama	great amberjack	L3	July, 2017 May, 2018

Table 3. Original fish weights and ashed weights of the selected samples.

S. No.	Fish Wt (kg)	Ashed Wt (g)
1	4.94	105.01
2	3.51	98.62
3	2.45	89.15
4	2.09	109.48
5	3.20	99.26
6	2.26	85.57
7	2.44	96.72

2.3 Radioactivity Determination

To determine the activity concentration of U and Th series through their strong γ-emitting decay products (such as ^{226}Ra and ^{228}Th) and also that of ^{40}K, the following relation is used [12]:

$$A = \frac{C(E_\gamma)}{M\beta(E_\gamma)\epsilon(E_\gamma)} \quad (1)$$

where: $C(E_\gamma)$ is the count of net peak area per second at energy E_γ,
M is the mass fish sample,
β is the transition probability of gamma-decay at energy E_γ,
ε is the detector efficiency at energy E_γ.
To estimate the risk related to ^{226}Ra, ^{232}Th and ^{40}K nuclides, radium equivalent (Ra_{eq}) activity in (Bq/kg) is given by [13]:

$$Ra_{eq} = A_{Ra} + 1.43\,A_{Th} + 0.077\,A_K \quad (2)$$

Where: A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K. They are given as 10 Bq/kg for ^{226}Ra, 7 Bq/kg for ^{232}Th and 130 Bq/kg for ^{40}K, respectively.

2.4 Evaluation of Radiological Hazard Effects

These indices are the absorbed gamma dose rate (*DR*), annual effective dose equivalent (*AEDE*), internal and external radiation hazard indices (H_{in} and H_{ex}) and gamma radiation representative level index (I_γ), respectively. All radiation doses are evaluated using dose conversion coefficients and occupancy factors provided by UNSCEAR 2000 [14].
DR is calcul ated as follows:

$$DR\ (nGyh^{-1}) = 0.462\,A_U + 0.604\,A_{Th} + 0.042\,A_K \quad (3)$$

where A_U, A_{Th} and A_K are the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K in *Bq kg*$^{-1}$, respectively, where the numerical factors are the conversion factors from *Bq/kg* to *nGy/h*.
H_{ex} and H_{in} are given by:

$$H_{ex} = (A_U/370\ Bq\ kg^{-1}) + (A_{Th}/259\ Bqkg^{-1}) + (A_K/4810\ Bqkg^{-1}) \leq 1 \quad (4)$$

$$H_{in} = (A_U/185\ Bq\ kg^{-1}) + (A_{Th}/259\ Bq\ kg^{-1}) + (A_K/4810\ Bq\ kg^{-1}) \leq 1 \quad (5)$$

Each index value must be less than unity in order to keep the radiation hazard to be insignificant.
I_γ is given as:

$$I_\gamma = 1/150\,A_U + 1/100\,A_{Th} + 1/1500\,A_K \quad (6)$$

Values of the representative index (I_γ) less than unity correspond to annual effective dose of less or equal to 1 *mSv*.
To calculate the biological impact of such exposure, *DR* is converted to an annual effective dose equivalent (*AEDE*), outdoor and indoor, using the following two equations [15]:

$$D_{outdoor} = \left[DR\left(\frac{nGy}{h}\right) \times 8760\ h \times 0.2 \times 0.7\left(\frac{Sv}{Gy}\right) \times 10^{-6} \right]\left(\frac{mSv}{y}\right) \quad (7)$$

$$D_{indoor} = \left[DR \left(\frac{nGy}{h} \right) \times 8760\ h\ \times 0.8\ \times 0.7\ (\frac{Sv}{Gy}) \times 10^{-6} \right] \left(\frac{mSv}{y} \right) \quad (8)$$

The dose conversion coefficient from absorbed dose to effective dose is 0.7 $SvGy^{-1}$, 0.2 and 0.8 are the outdoor and indoor occupancy factors.

3 Results and Discussion

Table 4 shows the natural activity concentrations (in *Bq/kg*) of ^{40}K, ^{226}Ra, ^{234}Th, and ^{235}U in fish bone samples. The concentration values of these radionuclides are lower than world average values [14]. Thus, there is a fair amount of radioactivity in the fishbones, with ^{40}K being the dominant element. The highest ^{40}K activity concentration was found in Seriola Dumerili fish (255.00 *Bq/kg*) and the lowest was in Diplodus Vulgaris fish (27.90 *Bq/kg*), with an average of 146.63 Bq/kg. Table 4 also shows that 226Ra from 157.10 *Bq* kg^{-1} to less than < MDA, with an average of 86.40 *Bq* kg^{-1}; ^{234}Th ranged from 53.30 *Bq* kg^{-1} to less than < MDA, with an average of 16.84 *Bq* kg^{-1}; while the activity level of ^{235}U was recorded only in one sample with a value of 4.51 *Bq* kg^{-1}.

In the aquatic environment, ^{40}K is the highest abundant radionuclide due mainly to its solubility in water. Elements from other decay chains are found insignificant amounts in aquatic media since these elements are principally insoluble in water. Therefore and due to the importance of ^{40}K radionuclide, Fig. 2 shows the appropriate graph. Comparison of the mean activity concentration of ^{40}K (fishbones) obtained in this study with other countries (fish) is presented in Table 5.

Table 4. Natural activity concentrations (in Bq/kg) of ^{40}K, ^{226}Ra, ^{234}Th and ^{235}U in fishbones of samples.

S. No.	^{40}K	^{226}Ra	^{234}Th	^{235}U
1	91.5±0.03	< MDA	15.90±2.81	4.51±0.25
2	147.00±0.02	67.43±0.07	< MDA	< MDA
3	27.90±0.15	81.90±0.07	< MDA	< MDA
4	112.00±0.04	113.05±0.07	< MDA	< MDA
5	152.00±0.02	63.12±0.08	< MDA	< MDA
6	241.00±0.11	157.10±0.20	48.70±2.50	< MDA
7	255.00±0.02	122.20±6.40	53.30±1.16	< MDA
Average	146.63±0.05	86.40±0.98	16.84±0.92	4.51±0.25
World Average	420			

Table 5. Comparison of the mean activity concentration of ^{40}K (fishbones) obtained in this study with other countries (fish).

Country	^{40}K (Bq/kg)	Reference
Libya	147	Present work
Nigeria	618	[16]
Malaysia	31.2-42.6	[17]
India	64.3	[18]
India	13.36-41.27	[19]
Turkey	179	[20]
Turkey	319	[21]
Kuwait	412	[22]
USA	99	[23]
Pakistan	90	[24]
Iraq	243	[25]
Nigeria	533	[26]
Worldwide	**420**	[14]

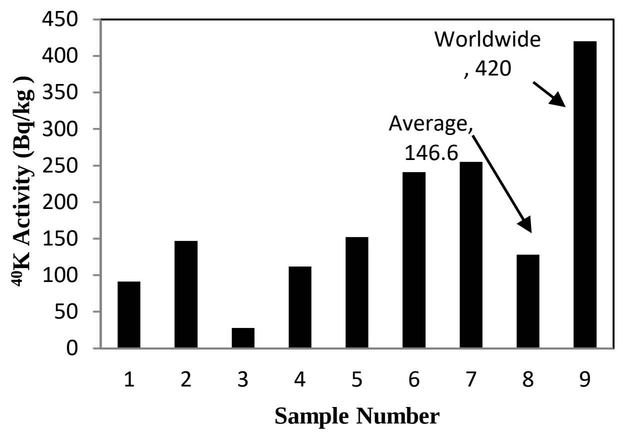

Fig. 2. Activity concentrations for ^{40}K radionuclide in fishbones of samples.

Table 6 presents the calculated radiological hazard indices H_{in}, H_{ex}, I_γ, *DR(nGy/h)*, $D_{outdoor}$ *(mSv/y)*, D_{indoor} *(mSv/y)* and Ra_{eq} *(Bq.kg^{-1})* in fishbone samples. Ra_{eq} values varied from 14.78x10^{-4} to 245.20 *Bq kg^{-1}* with an average of 90.48 *Bq kg^{-1}*. All the values of Ra_{eq} do not exceed the suggested maximum allowable value of 370 *Bq kg^{-1}* [13]. *DR* rate values ranged from 68.35x10^{-5} to 112.82, with a mean value of 42.46 *nGy h^{-1}*. Even though some of the fish samples have values larger than the world average, the estimated mean value of *DR* is lower than the world average (populated-weighted) absorbed gamma dose rate of 84 *nGy h^{-1}*. As can be from the values, some fish samples have the representative index of their samples exceeding unity. All external hazard indices are less than one as they should be except for H_{in} of one sample (no. 6). As for I_γ index all values were less than one except for samples numbers 6 and 7. This increase in their values should be of some concern to fish consumers. The results show that the *AEDE* values ($D_{outdoor}$ *and* D_{indoor}) are all lower than the recommended 1.0 *mSv/y* limit for members of the public. Thus, the radiation dose incurred from the ingestion of the studied samples pose no significant health risks as far as these indices are concerned.

Table 6. Radiological hazard indices H_{in}, H_{ex}, I_γ, DR (nGy/h), $D_{outdoor}$ (mSv/y), D_{indoor} (mSv/y) and Ra_{eq} (Bq.kg^{-1}) in fishbones of samples.

S. No.	H_{in}	H_{in}	I_γ	DR (nGy/h)	$D_{outdoor}$ (mSv/y)	D_{indoor} (mSv/y)	Ra_{eq} (Bq.kg^{-1})
1	0.08±0.01	0.08±0.01	0.22±0.04	13.70	0.02	0.07	29.78±2.84
2	0.30±3.52x10^{-4}	0.10±1.17x10^{-4}	0.24±2.82x10^{-4}	18.40	0.02	0.09	35.21±0.14
3	0.41±30.00x10^{-4}	0.19±12.00x10^{-4}	0.50±31.00x10^{-4}	33.20	0.04	0.16	70.60±5.95
4	0.60±0.03	0.26±0.01	0.70±0.04	46.30	0.60	0.23	97.22±3.49
5	0.25±4.00x10^{-4}	0.08±1.12x10^{-4}	0.19±2.66x10^{-4}	15.13	0.02	0.07	27.92±1.70
6	1.08±0.06	0.66±0.04	1.70±0.09	112.82	0.14	0.60	245.20±3.78
7	0.92±0.02	0.60±0.01	1.51±0.03	100.09	0.12	0.50	217.90±7.58
Ave.	0.45	0.25	0.63	42.46	0.12	0.22	103.40±3.64

4 Conclusions

In conclusion, this study was initiated to evaluate the level of radioactivity concentrations in fishbones from the eastern coast of Libya. Seven fish samples were collected from various sea shores of interest for commercial fishing industry from the study area. The study area extended from Benghazi city to Altememi city (a distance of about 500 km). The seven fish species, which are commonly consumed by the locals, were collected during different seasons of 2017-2018. These fish are: Epinephelus Marginatus, Pagellus Bogaraveo, Diplodus Vulgaris, Umbrina Cirrosa, Trachurus Mediterraneus, Balistes Carolinensis and Seriola Dumerili. Radioactivity measurements of radionuclides were performed on bones of these samples. The distributions of these radionuclides were determined using HPGe gamma ray spectrometry. Indices such as: gamma radiation representative level index (I_γ), absorbed gamma dose rate (*DR*), the annual effective dose equivalent (*AEDE:* $D_{outdoor}$ and D_{indoor}), internal (H_{in}) and external (H_{ex}) hazarded indices, radium equivalent activity (Ra_{eq}) index were determined. The radioactivity concentrations of ^{40}K, ^{234}Th, ^{226}Ra and ^{235}U were determined. The obtained results for the radionuclides were lower than the worldwide allowable limits. Hence, in general, the values of the obtained activity concentrations in the selected fish samples do not pose radiological risks at the time of this study. The radiometric indices were found to be less than the recommended safe and criterion limits given by UNSCEAR except H_{in} and I_γ for Balistes Carolinensis where the values were greater than one (>1). This increase may be of concern to consumers of this type fish. Therefore, the results of this study could serve as an important radio-metric baseline data, upon which future epidemiological studies and environmental monitoring initiatives could be based, as well as natural radiological mapping and a reference data in future in the study area. The data obtained represent an additional contribution in the study area, which are crucially lacking, where none was previously available.

Author Contributions: H. Hasan contributed to the conception of the study and analyses; R. Hamad contributed significantly to samples collection and preparation and analyses; F. Ikraiam performed the data analyses and wrote the manuscript. All authors have read and agreed to the published version of the manuscript.

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