

CONCENTRATION ACTIVITIES OF NATURAL RADIONUCLIDES IN THREE FISH SPECIES IN BRAZILIAN COAST AND THEIR CONTRIBUTIONS TO THE ABSORBED DOSES

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ABSTRACT

Activity concentrations of U-238, Ra-226, Pb-210, Th-232 e Ra-228 were analysed in three fish species at the Brazilian Coast. The fish “Cubera snapper” (*Lutjanus cyanopterus*, Cuvier, 1828), in the region of Ceará and “Whitemouth croaker” (*Micropogonias furnieri*, Desmarest, 1823) and “Lebranche mullet” (*Mugil liza*, Valenciennes, 1836) in the region of Rio de Janeiro. These concentrations were transformed in absorbed dose rate using a dose conversion factor in unit of gray per year ($\mu\text{Gy y}^{-1}$), per becquerel per kilogram (Bq kg^{-1}). Only the absorbed dose due to intake of radionuclides was examined, and the contributions due to radionuclides present in water and sediment were disregarded. The radionuclides were considered to be uniformly distributed in the fish body. The limit of the dose rate used, proposed by the Department of Energy of the USA, is equal to $3.65 \cdot 10^3 \text{ mGy y}^{-1}$. The average dose rate due to the studied radionuclides is equal to $6.09 \cdot 10^0 \mu\text{Gy y}^{-1}$, a value menor que 0.1% than the limits indicated by DOE, and quite similar to that found in the literature for “benthic” fish. The most important radionuclides were the alpha emitters Ra-226 having 61 % of absorbed dose rate. U-238 and Th-232, each contributes with aproximadamente 20 % of the absorbed dose rate. These three radionuclides are responsible for almost 100% of the dose rate received by the studied organisms. The beta emitters Ra-228 and Pb-210 account for aproximadamente 1 % of the absorbed dose rate.

1. INTRODUCTION

Radiation is an inherent fact of life that arose and evolved in a constantly irradiated environment. Some authors point to ionizing radiation as one of the factors that participated of biological evolution, allowing for greater gene flow [1-4]. Human activities can expose biota to radiation levels that can be harmful, causing undesirable effects. These activities are called ‘practices’ and are subjected to norms in Brazil [5].

The concept of practice was established in 1991 by the ICRP [6]. Practice is defined in the Brazilian Norm, as a human activity that alters the exposure of humans to radiation, either by changing the routes of exposure, either by introducing new routes or new radionuclides, or

even new exposed groups. This concept makes it possible to legislate on the change of exposure, setting up limits for the increase in dose caused by practice [5]. This concept is also used by ICRP in its recommendation on environmental protection of non-human biota [7] and also in other recommendations [6] and [8].

Aiming protection of the environment against the unwanted effects of radiation caused by practices, a science was developed called Environmental Radioprotection. This science is a fairly recent concern, which had its standardization in the 70s of last century [5-6], [9-11]. This science takes care of all the measures to protect the biota of the unwanted effects of ionizing radiation, but unlike human radioprotection, environmental radioprotection does not have a global consensus [6], [11-12]. In the current state of environmental radioprotection, there are no explicit criteria for environmental protection to prevent the unwanted biological effects of radiation on biota [6], [11-13].

Currently, Brazilian law uses the concept proposed by the ICRP that states: *"The commission therefore believes that if man is adequately protected then other living things are also likely to be sufficiently protected"* [8]. This paradigm has been broken in Recommendation Number 103 of ICRP [8], after having been put in doubt by several authors [14-19], inclusive by ICRP itself [6]. Indeed, ICRP now states that the radioprotection of the environment must be made explicit and not based on the anthropocentric concept used so far. This new approach follows proposals made at the Eco-92, which systematized the concept of sustainable development [20].

The anthropocentric approach based on the vision proposed by the ICRP [9] is not the only approach used. The Ecological Risk Assessment (ERA) has been used as a form of environmental radioprotection [21-23] as well as the use of a flora and fauna reference series of organisms [7], [15], [17], [24-28], to simulate the man reference, proposed by ICRP for human radioprotection [6], [8-9].

Another concept that has been used to achieve environmental radioprotection is the concept of absorbed dose (unit: Gray, symbol Gy, and size of J kg⁻¹). This concept, suitable for human radioprotection, focuses on the deposition of energy by radiation in the medium and tries to correlate this with possible biological effects. The concept of absorbed dose allowed the creation of the standard dose limit. Such approach is used by the USA [29-30], the UK [28] and Canada [31].

Organizations also have made their propositions using the concept of standard dose limit, for example, the project "Protection of the Environment from Ionizing Radiation in a Regulatory Context", bringing together 15 institutions from seven European countries [25] and IAEA [24]. Individual proposals also appeared [12].

To achieve radioprotection based on the concept of standards dose limit, a dosimetric model is needed to transform the exposition to radiation and contamination with radionuclides into absorbed dose rate. There is also the need to estimate the biological effects of absorbed dose rates, aiming to establish limits for the impact caused by human activity [12].

The first step to calculate the increase in dose rate in biota is to establish the background of absorbed dose rate to which are exposed members of the biota that is being evaluated.

This study aims to establish the values the activity concentration of natural radionuclides and their contribution to the dose rate at which fish from two regions of the Brazilian coast are subjected. The radionuclides considered are those belonging to the uranium series (isotope 238 of uranium, U-238, isotope 226 of radium, Ra-226 and isotope 210 of lead, Pb-210) and of the thorium series (isotope 232 of thorium, Th-232 and isotope 228 of radium, Ra-228). Analyzed fish were: “cubera snapper” (*Lutjanus cyanopterus*, Cuvier, 1828), from Ceará and the “whitemouth croaker” (*Micropogonias furnieri*, Desmarest, 1823) and the “lebranche mullet” (*Mugil liza*, Valenciennes, 1836) from the region of Rio de Janeiro.

2. METHODOLOGY

2.1. Framework For Radioprotection Of Non-Human Biota

As a framework for calculating the absorbed dose of non-human biota, we used the approach proposed by [12] and [32], and as a proposal for environmental radioprotection we used the concept of standard dose limit proposed by IAEA [24] and sanctioned by ICRP [7], USDOE [30], NCRP [29] and Environmental Canada [31].

To calculate the dose rates, this work made some statements. Thus, the dose rate due to radionuclides present in water and sediment has not been taken into consideration. Only internal dose rate has been considered. The alpha, beta and gamma emitter radionuclides were considered to be homogeneously distributed, all over the organism. The alpha radiation is totally absorbed by it. Moreover, using the conservative approach, it is considered that all beta and gamma energy was absorbed.

The internal dose rate is calculated, in $\mu\text{Gy y}^{-1}$ from the activity concentrations of the radionuclide, obtained in Bq kg^{-1} fresh weight, as follows:

For alpha-emitter:

$$D_{\alpha} = D_{CF} E_{\alpha} N_{\alpha} C_o \quad \mu\text{Gy y}^{-1} \quad (1)$$

For beta-emitter:

$$D_{\beta} = D_{CF} E_{\beta} N_{\beta} C_o \quad \mu\text{Gy y}^{-1} \quad (2)$$

For gamma-emitter:

$$D_{\gamma} = D_{CF} E_{\gamma} N_{\gamma} \Phi_{\gamma} C_o \quad \mu\text{Gy y}^{-1} \quad (3)$$

where:

- D_{CF} is the dose conversion factor [$5,05 (\mu\text{Gy y}^{-1}) (\text{Bq kg}^{-1})^{-1}$];
- E_{α} , E_{β} and E_{γ} are the energies of the alpha, beta and gamma radiations respectively (MeV);
- N_{α} , N_{β} and N_{γ} are the amount of transitions that produces an alpha particle, beta particle or a gamma ray (dimensionless);
- Φ_{γ} is the amount of absorbed gamma energy (dimensionless) and;
- C_o is the activity concentration of the radionuclide in the organism (Bq kg^{-1} , fresh weight).

For the calculation of the total internal dose (D_i), the sum of the contributions of alpha, beta and gamma radiations was considered, as follows:

$$D_i = D_\alpha + D_\beta + D_\gamma \quad \mu\text{Gy y}^{-1} \quad (4)$$

2.2. Biological Material: Collection and Preparation

The fish cubera snapper (*Lutjanus cyanopterus*, Cuvier, 1828) was collected along the coast of Ceará, and the whitemouth croaker (*Micropogonias furnieri*, Desmarest, 1823) and lebranche mullet (*Mugil liza*, Valenciennes, 1836) in the region of Rio de Janeiro. Six specimens of the cubera snapper fish were caught in January of 2007, eight specimens in June of 2007 and seven additional specimens in November of 2007. Nine specimens of the whitemouth croaker were collected in June of 2007 and five specimens of the lebranche mullet in June of 2007. The fish was put into plastic bags and carried to the Laboratory. The cubera snapper was processed at the Laboratory of Analytical Chemistry of the Federal University of Ceará (UFC), and the whitemouth croaker and the lebranche mullet at the Laboratory of Environmental Monitoring of the ore treatment unit UTM of the Brazilian Nuclear Industries (INB). Fish was weighted and dried at 80°C, until constant weight. The biological material was then reduced to ashes at 450°C to yield a pale residue from which aliquots were taken for analysis.

2.3. Radionuclides

Analyzed radionuclides belong to the U-238 and Th-232 series: U-238, Ra-226 and Pb-210, from the U-238 family, and Th-232 and Ra-228, from the Th-232 family.

Uranium and thorium were measured by spectrophotometry using arsenazo [33], modified by [34]. Ra-226 activity was determined by radiochemistry and total alpha radiometry, and Ra-228 and Pb-210 by radiochemistry and total beta radiometry, as described by [35].

The values of the radiations energies E_α , E_β and E_γ , and the de N_α , N_β and N_γ values for all studied radionuclides are shown in Table 1. The value of Φ for alpha, beta and gamma radiations has been stated equal to 1.

Table 1 – Amounts of transitions that produce particles and radiation, and energies of particles and radiation emitted by analyzed radionuclides (in MeV) following [36].

Radionuclide	N_α	E_α	N_β	E_β	N_γ	E_γ
U-238	1	4,26	0	N.A.	1	$1.36 \cdot 10^{-3}$
Ra-226	1	4,86	0	N.A.	1	$6.47 \cdot 10^{-3}$
Pb-210	0	N.A.	1	$3.80 \cdot 10^{-2}$	1	$4.81 \cdot 10^{-3}$
Th-232	1	4.07	0	N.A.	1	$1.33 \cdot 10^{-3}$
Ra-228	0	N.A.	1	$1.69 \cdot 10^{-2}$	1	$4.14 \cdot 10^{-9}$

N.A. = does not apply

2.4. Limits of Absorbed Dose Rate

In Brazil, no limits have been established for the absorbed dose rate by biota. In some countries, in turn, the model proposed by the International Atomic Energy Agency (IAEA)

has already been adopted. This is the case of the Department of Energy (DoE) of the United States that uses the value proposed by IAEA, which is stated as 10 mGy d^{-1} [30]. In Brazil, the system of radioprotection is done on an annual basis [5]. Therefore the value of the limit for annual rate was fixed as $3.65 \cdot 10^3 \text{ mGy y}^{-1}$, and will be used in this work as dose rate limit.

3. RESULTS AND DISCUSSION

The activity concentrations (in Bq kg^{-1}) of U-238, Ra-226, Pb-210, Th-232 and Ra-228 in the fish cubera snapper, whitemouth croaker and lebranche mullet are shown in Table 2 and are compared with world averages values for U-238, Ra-226 and Th-232 reported in the literature [37] e [27].

For U-238, the mean values found in this work were an order of magnitude higher than the world average one [37] and [27]. For Ra-226, the values found were at the same level of literature [37] and [27], and the values for Th-232 were three orders of magnitude higher than reported by Brown et al. [27]. No comparison could be made for Pb-210 and Ra-228, since no world average values could be found in the literature. This might be due to the fact that both radionuclides are not alpha emitters [37] and since they are less studied [27].

Table 2 – Average of activity concentrations (in Bq kg^{-1}) of the radionuclides analyzed in the fish of Brazilian coast.

Local	Date	Fish	U-238	Th-232	Ra-226	Ra-228	Pb-210	N
Ceará	02/07	cubera snapper	0.06	0.03	0.28	0.65	0.04	6
Ceará	06/07	cubera snapper	0.08	0.02	0.10	0.19	0.10	8
Ceará	09/07	cubera snapper	0.03	0.03	0.17	0.55	0.17	7
Rio de janeiro	06/07	lebranche mullet	0.04	0.07	0.24	0.13	0.36	5
Rio de Janeiro	06/07	whitemouth croaker	0.02	0.14	0.17	0.82	0.16	9
Average this work			0.05	0.06	0.19	0.47	0.17	
Average of [37]			0.003	0.12	N.D.	N.D.	N.D.	
Average of [27]			0.008	0.20	N.D.	0.001	N.D.	

N.D. = not determined, N = number of samples

For evaluations of internal absorbed dose rate, the average concentration of activity of the three studied species were used; observed values are outlined in Table 3, that also brings the world average values reported by Brown and colleagues [27]. The values of absorbed dose rate (in $\mu\text{Gy y}^{-1}$) were analyzed in terms of the radionuclide and the kind of radiation emitted.

The absorbed dose rate found was $6.08 \mu\text{Gy y}^{-1}$, a value less than 0.1% of the limit of absorbed dose rate used in this work ($3.65 \cdot 10^3 \text{ mGy y}^{-1}$). Observed value corresponds to 16% of the value reported in the literature as world average, which is $38.4 \mu\text{Gy y}^{-1}$ [27].

Considering the contribution to the absorbed dose rate, the most important radionuclide was the alpha emitter Ra-226, with 61% of the absorbed dose rate. The second in importance was

Th-232 with 20%, and close to it, U-238 with 18 %; both with approximately 1/3 of the absorbed dose rate of Ra-226. The other analyzed radionuclides (Pb-210 and Ra-228) had their contributions considered negligible, with a total contribution of almost 1% (Figure 1).

Table 3 - The internal absorbed dose rate in Brazilian fish and world average values proposed by [27] ($\mu\text{Gy y}^{-1}$) by radionuclides and radiation type.

Radionuclide	Mean of absorbed dose rate [27]	Absorbed dose			
		Alpha	Beta	Gamma	Total
U-238	4.2	1.07	N.A.	0.00	1.07
Ra-226	34.2	3.70	N.A.	0.01	3.71
Pb-210	N.D.	N.A.	0.03	0.00	0.03
Th-232	0.02	1.23	N.A.	0.00	1.23
Ra-228	0.0001	N.A.	0.04	0.00	0.04
Total		6.00	0.07	0.01	6.08

Differences in the dosimetric models used in this work and by Brown et al. [27] are reflected in the assessment of the radiological environmental impacts (REI). Thus, similar results may lead to different assessments, depending only on the model used. For example, for Ra-226 similar values 0.19 Bq kg^{-1} (this work) and 0.20 Bq kg^{-1} [27] generated impacts assessments with an order of magnitude of difference (3.70 in this work against 34.2 in [27]). The differences in values, in this case, were not important in deciding on the estimation of the Radiological Environmental Impact since, in both approaches, the REI was negligible (less than 1% of the limit of absorbed dose rate).

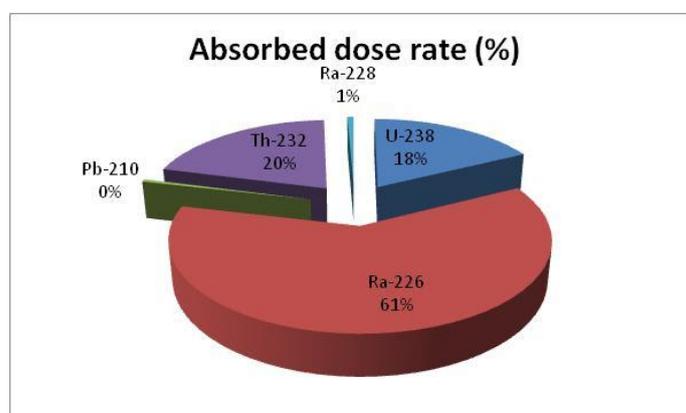


Figure 1 – Contribution of analyzed radionuclides to the absorbed dose rate.

4. CONCLUSIONS

The methodology of standard dose limit was appropriate to assess the REI of natural radionuclides and was sensible enough to assess the impact of radionuclides in the marine environment. It was concluded that these radionuclides, in these fish, are not a concern in terms of environmental radioprotection. Additional relevant information obtained in this work was about the composition of the absorbed dose rate. It indicated Ra-226 as the critical natural radionuclide, *i.e.* the radionuclide that is responsible for most of the absorbed dose rate among the here considered ones. This result is in agreement with the literature [27].

A first important limitation for assessments in terms of radioecology and environmental radioprotection is the lack of data on natural radionuclides in fish from the Brazilian coast, which prevents radioecological comparisons with other regions of Brazil. Another limitation is the study of only few species of fish, which, again, prevents radioecological characterization. The analysis of other members of the ecosystem, such as fish from different habitats or different positions in the food chain, etc., and other members of the biota is of utmost importance for such characterization. In addition, the evaluation of other natural radionuclides, primarily the alpha emitters Po-210, U-235, U-234, Th-230 and Th-228 can alter to some extent the framework outlined in this work.

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