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GEOSCIENCES

Dose conversion coefficients to marine biota due to natural radionuclides in an oil spill accident using Monte Carlo simulation

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Abstract: The crude oil that reached the Brazilian coast in 2019 was the most significant environmental disaster ever recorded in Brazilian marine waters, with severe ecological and economic repercussions not fully dimensioned and understood. One consequence of this kind of oil spill is the absorbed dose delivered to marine organisms. The biota exposure to radiation can introduce consequences that range from fertility decrease to death of exposed population. Therefore, the current study aims to use the ICRP reference organisms (flatfish, crab, and brown seaweed) to assess marine biota exposure due to uranium and thorium series radiation. The oil spill scenario, as well as the reference biota, were simulated in MCNP. Thorium series stood out for presenting a significant contribution to the dose. Furthermore, it was noticed that a remedial action capable of removing Tl-208 would significantly decrease the effects of radiation on marine biota. Finally, dose conversion coefficients for both uranium and thorium series were obtained as oil activity concentration functions. The results obtained here can be used in an oil spill event along with other worldwide recognized models. In addition, the dose coefficients can be used strategically to assess the maximum exposure time for emergency oil control, removal, and mitigation.

Key words: brown seaweed, crab, flatfish, ICRP, MNCP, reference animal and plants.

INTRODUCTION

One of the most important energy resources for the global economy is petroleum. The industry growth of oil production and exploration has made it possible to explore these natural resources in places that are difficult to access, such as the deep ocean (Hall et al. 2003, Rademaekers et al. 2015). This growth is due to a constant global request for hydrocarbons use, either as a fuel or for the use of its derivatives (Ngene et al. 2016). However, the petroleum exploration industry is also responsible for a significant part of the hydrocarbons insertion in marine environments (Bollman et al. 2010).

The release of oil on the ocean's surface has been exhaustively investigated (Vasconcelos et al. 2020, Conceição et al. 2021). It can generate potential environmental impacts associated with heavy metal contamination (Osuji & Onojake 2004, Zhang et al. 2020), contamination by organic compounds (Ke et al. 2002, Celino et al. 2012), bioaccumulation in the food chain (Law & Hellou 1999, Gin et al. 2001, Boehm et al. 2005, Ingole et al. 2006, Martin-Skilton et al. 2008), habitat destruction (Peterson 2001), wildlife death (Mignucci-Giannoni 1999), and disturbances in the characteristic of the marine radiometric background (Al-Masri & Aba 2005, Barescut et al. 2005, Gazineu et al. 2005, Al-Saleh

& Al-Harshan 2008, Abo-Elmagd et al. 2010). Oil spills can also affect coastal communities' economies, reducing tourism or damaging commercial fishing areas (Chang et al. 2014).

In 2019, one of the biggest environmental tragedies ever registered occurred on the Brazilian coastline. Several Brazilian states in the northeast and southeast recorded crude oil's appearance on its beaches (Lourenço et al. 2020). However, this oil was not detected on the ocean surface due to its geochemical characteristics. Indeed, it was believed the oil floated about 1.5 meters below the surface. This behavior made it difficult to trace the oil origin by satellite images or non-commercial flights since it only became visible close to the coast. As a result, the spill's origin and how it occurred are still unknown nowadays.

Petroleum is constituted by a complex mixture formed by several compounds that may vary their composition depending on the area of geological formation. Petroleum's main elements are hydrocarbons (composed of hydrogen and carbon), but it also contains nitrogen, oxygen, sulfur, and some metals such as nickel, vanadium, and chromium (Fingas 2016, Philp 2019). Therefore, oil is considered toxic due to its chemical structure, where volatile organic compounds, polycyclic aromatic hydrocarbons (PAHs), hydrogen sulfide, and heavy metals are found (Pena et al. 2020, Philp 2019). Moreover, in its nuclear composition, radionuclides are also found, such as the elements of uranium and thorium series (Gregory et al. 2014).

The *Naturally Occurring Radioactive Materials* (NORM) are found in the environment. NORM primarily contains elements of uranium and thorium series and the K-40 (Attallah et al. 2012). Human activities can increase the natural radionuclides' activity concentration (AC) through industrial processes despite being found naturally in the environment. These materials with increased AC's are known by *Technologically Enhanced Naturally Occurring Radioactive Material* (TENORM) (Mazzilli et al. 2011). Exposure to ionizing radiation due to the release of oil into the environment has been a concern reported by other authors, such as Keum et al. (2013), Landsberger et al. (2017), and Ovuomarie-Kevin et al. (2018). Initially, environmental radioprotection was carried out indirectly with the human being as a reference baseline (ICRP 1977). Then, however, the need to evaluate the environment started to be a requirement. Today, there is a consensus that the environment needs to be analyzed as a whole (ICRP 2003, 2007, 2008).

The International Commission on Radiological Protection (ICRP) evaluated the effects of exposure to ionizing radiation in animals and plants (ICRP 2008). It prioritizes organisms that may undergo changes in population size or structure, early mortality (changes in age distribution, mortality rate, and density), morbidity (reduction in the capability of individuals to survive in the wildlife), reproductive capacity (birth rate, age distribution, number, and density), and induction of chromosomal damage (ICRP 2008).

Accordingly, Batlle et al. (2011) studied the absorbed dose rate (D_R) for 74 radionuclides in five ICRP Reference Animals and Plants with the approach turned to the non-human biota. These authors observed the occurrence of problems between the different approaches. Therefore, they suggested a new methodology based on the dose conversion coefficients used in some models available to evaluate biota's radiological effects. Brown et al. (2004) identified the importance of a methodology for D_R's assessment due to these natural radionuclides since the estimated D_R's were higher for freshwater organisms than marine organisms. These differences induced variability or uncertainty in the dose coefficient values.

Ulanovsky & Pröhl (2006) developed a methodology that allows the derivation of dose conversion coefficients (DCC's) for organisms with a determined density. These modeled organisms were simulated using a wide range of ellipsoidal formats, making it possible to derive theirs DCC's for any radionuclide present in the ICRP database.

Based on these methodologies, the current study aims to obtain DCC's for marine biota species due to uranium and thorium series radiation. DCC's can provide an estimate of the D_R in the biota from the AC of the oil. Furthermore, DCC's can be used as an initial assessment to estimate the time required for oil removal before the biota's damage occurs in an oil spill accident.

MATERIALS AND METHODS

Boundary conditions used

An oil spill's behavior and chemical composition in the ocean depends on several processes, such as evaporation, emulsification, dissolution, biodegradation, and photo-oxidation. In addition, there are interactions between oil, sediment, and water, which are sufficient to change crude oil's initial characteristics. The combination of all the processes above is known as oil weathering. All these events acting together reduce the concentration of various compound groups, modifying the oil's chemical and physical characteristics (Oudot et al. 1998, Souza & Triguis 2006).

It is known that sealing the environmental samples for gamma spectrometry analysis is necessary to avoid radon's escape. This methodology allows the elements of the radioactive series to reach secular radioactive equilibrium (Lopes et al. 2018a, Garcêz et al. 2019, Silva et al. 2020). Therefore, it is reasonable to imagine that the radioactive series lose its secular equilibrium during the weathering process. Currently, no mathematical model can faithfully represent the radioactive secular equilibrium breakdown in an oil spill accident.

Thus, the current study considered a conservative model where the uranium and thorium series are in secular radioactive equilibrium. Therefore, all elements of the series have the same AC. Furthermore, this procedure is considered reasonable since the radionuclides generally used to represent the series, Ra-226, and Ra-228, undergo oil weathering processes with no elements' enrichment, only depletion. However, it can be stated that considering the secular equilibrium may overestimate the AC values and, consequently, the D_R.

Computational simulation

MCNPX (Pelowitz 2011) is a radiation transport code that has been widely used in the medical physics survey (Thalhofer et al. 2018, Guimarães et al. 2018) to simulate ionizing radiation detectors (Salgado et al. 2012, Lopes et al. 2018b) and in the environmental science fields (Ulanovsky & Pröhl 2006, Park et al. 2018). The code runs using inputs that define the surfaces and cells to be irradiated and their chemical compositions, characteristics of the radiation source (decay, position, size), and a number of particle histories (following a particular emission from its origin until its extinction). The irradiation outline's geometry, the radioactive sources, and the desired magnitudes are defined in the input files. The statistical uncertainties express the precision of the results, and it is directly linked to the number of particle histories.

For the current study, an oil spill accident was simulated. The materials' chemical compositions were obtained from the Compendium of Material

Composition for Modeling Radiation Transport (McConn et al. 2011).

A cylinder with a height of 1 cm and a radius of 5 m was considered to simulate the oil spill on the ocean's surface. The height (1 cm) was arbitrarily chosen. On the other hand, the radius of a simulated oil spill was adjusted to ensure a negligible influence of the radiation from its edges. This assurance is based on depth dose profile simulations since at depths greater than 3 m the saltwater considerably attenuates the simulated gamma radiation. In other words, the radius was adjusted such that its emitted gamma radiation by the end (edge) of an oil spill would not influence the dose delivered to the organism significantly.

On the other hand, it is impossible to assess all individuals' D_R's in an ecosystem. Therefore, categorizing wildlife into various representative groups according to the biological and ecological level is a pragmatic approach to assessing the D_R for a non-human biota (Ulanovsky & Pröhl 2006, ICRP 2008).

The current study has chosen some reference species from the marine environment with a wide occurrence on the Brazilian coasts. The organism's geometry was approximated by a quadric surface and modeled according to the concept of reference species in the ICRP (ICRP 2008). The dimensions of each reference organism are shown in Table I.

The geometry details used in computational simulations are shown in Figure 1. The simulated organism was positioned on the central axis of a simulated oil spill at a depth of 100 cm.

The simulated energies are available in Table II (Leung et al. 1990). The choice of energies was based on two arbitrary criteria: higher than 150 keV and probability of emission higher than 3%. This is because energies below 150 keV are easily attenuated by water. The attenuation of low energies results in uncertainties above 5% for depths of 100 cm, thus losing reliability. Energies with a low probability of emission do not contribute significantly to the energy delivered to the organism.

The radiation transport was carried out until the energy was below 1 keV for photons and 10 keV for electrons (Ulanovsky & Pröhl 2006). Water was used as the organism's material tissue and 1.0 g.cm^{-3} as its density (Ulanovsky & Pröhl 2006, ICRP 2008, 2017). The tally **f8* was used to obtain the value of the energy delivered to the organism.

Dose Conversion Coefficients (DCC's)

The tally **f8* used in simulation calculates the energy delivered in an infinitesimal volume *dV* by subtracting the energy that leaves the volume from the incident energy. The result of the tally **f8* must be divided by the mass volume *dV* to obtain the absorbed dose, according to Equation 1:

$$D\ (Gy) = \frac{{}^{*}f8(MeV)}{m(kg)} \cdot 1.602\,x10^{-13} \quad (1)$$

Table I. Dimensions of the ICRP (2008) reference biota.

Organism	Major axis (cm)	1st minor axis (cm)	2nd minor axis (cm)	Body mass (kg)
Flatfish	40	25	2.5	1.31
Crab	20	12	6	0.754
Brown seaweed	50	50	0.5	0.654*

*** mass obtained from the MCNP output.**

Where: D is the absorbed dose

**f8* is the result of MCNP output file

m is the organism mass (ICRP 2008)

1.602×10^{-13} is the conversion factor from MeV to Joule

The energy delivered to the organism is normalized in the MCNP output file. It means that the energy delivered to the organisms is "per photon emitted by the source". The source is the simulated oil spilled in this case. Therefore, the total photons emitted by the radioactive source will be proportional to the AC ($Bq.kg^{-1}$) of the oil. The organism's exposure time is defined here as one day (86,400 s). This methodology standardizes the D_R ($Gy.d^{-1}$) with studies already available in the literature (Ulanovsky & Pröhl 2006, ICRP 2008, 2017).

The simulations were only interrupted when the uncertainties were low enough, i.e., < 5%. Then, all procedures were performed at the Laboratory of Environmental Analysis and Computational Simulation from the Federal University of Rio de Janeiro (LAASC/UFRJ).

RESULTS AND DISCUSSION

The D_R's contributions due to the gamma emissions of radionuclides from uranium and thorium series are shown in Figure 2. The contributions are presented as a function of the AC for each radionuclide. A linear relationship is expected because the radiation exposure is directly proportional to the AC. It should be noted the differences between the scales for each radioactive series. The difference is of one order of magnitude for Crab and Flatfish.

The emissions of each radionuclide contribute to the slope characteristic shown in Figure 2. For the uranium series, both Pb-214 and Ra-226 vary slightly in their D_R contributions with an AC increase compared with Bi -214. On the other hand, the Tl-208 stands out when compared to the delivered dose due to the Ac-228, Pb-212, and Bi-212 in the thorium series. These D_R differences are related to the gamma radiation energies emitted by each radionuclide added to each decay probability. It is expected

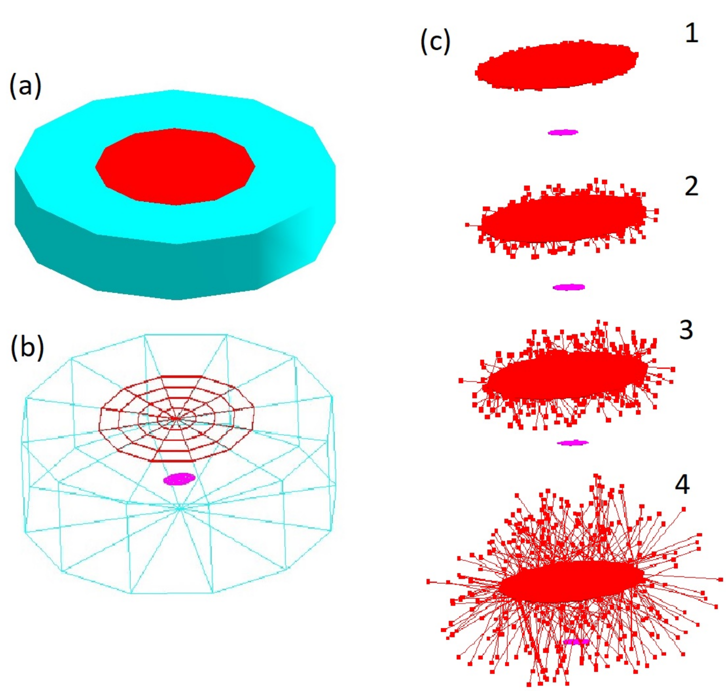

Figure 1. **Out-of-scale computational scenario set up for simulation: The ocean salt water (blue color) and the spilled oil (red color) in solid (a) and in frame (b) pictures. In (c) a sequence of four images, without simulated ocean salt water, showing the interaction of oil radiation with matter. The magenta color geometry is the simulated Flatfish.**

that the higher the emitted energy, the higher the probability of radiation interaction with a simulated organism because the seawater acts as a natural shield.

Table III shows the D_R's differences. The Bi-214 contributes for a D_R up to 97-fold compared with the contribution of the Pb-214 in the uranium series. These differences are more discreet for the thorium series. However, still large enough to make the differences in D_R's for some radionuclides imperceptible (Figure 2).

Ra-226 contributes discreetly in the uranium series to external D_R's. This element is characterized by being an alpha emitter with a reasonably long half-life, 1,600 years. In addition, Ra-226 has a considerable contribution to internal dose in case of ingestion or inhalation because it has a chemical affinity with the calcium element (Rowland et al. 1978, Silva et al. 2006). However, its principal gamma emission has discrete energy (186 keV) and a low probability of emission (3.6%).

The radionuclide that contributes less to D_R's in the thorium series is Pb-212. The leading element has high toxicity and can induce several intestinal, hepatic, or carcinogenic consequences if inhaled or ingested in excess (Moreira & Moreira 2004). However, D_R's due to external exposure is small compared to other elements of the series. Its prominent gamma energies are 238.6 and 300.1 keV, with an emission probability of 43.6% and 3.3%, respectively.

The D_R's delivered to each simulated organism are shown in Figure 3 and Figure 4 for the uranium and thorium series, respectively. Again, a linear relationship between these terms is expected because a D_R's dependence increases with the AC.

It was identified that there is a difference of up to four orders of magnitude in the scales of the figures for the uranium series only (Figure 3). However, the contribution of radionuclides to different organisms is in agreement, according to the literature. The most remarkable difference

Table II. Energies used in computational simulation.

Series	Radionuclide	Energies (keV)	Probability of Emission (%)
Uranium	Ra-226	186	3.6
	Pb-214	295	18.4
		352	35.6
	Bi-214	609	45.5
		1120	14.9
		1765	15.3
		2204	4.9
Thorium	Pb-212	239	43.6
	Bi-212	727	6.7
	Ac-228	338	11.3
		911	25.8
		969	15.8
	Tl-208	511	22.6
		583	85.0
		2615	99.7

was 16% in the D_R's due to the Ra-226 between Crab and Flatfish.

A difference of three orders of magnitude is observed on the D_R's due to the thorium series elements. It can also be stated that the D_R's do not behave similarly to the D_R's for uranium-series elements. The slight difference (2%) in D_R's due to Pb-212 for Crab and Flatfish stands out.

Figure 5 shows the total D_R's for each organism. For example, in an oil sample where the radioactive series elements have the same AC, the total D_R's will reach twelve times higher for crab, ten times higher for Flatfish, and five times higher for Brown Seaweed for the thorium series gamma emission.

In addition, Table IV shows the contribution percentage of total D_R for each gamma emitter. As expected, the Tl-208 and Bi-214 are the most significant contributors, with more than 96%.

The delivered dose to marine organisms is directly related to the spatial condition of the simulated animal. That is, the higher volume, the higher the absorbed dose for the reference species. Mainly for the simplified geometry proposed by ICRP (2007), the densities of an organism and the aquatic environment are practically the same. The densities are differentiated just because of the salinity of the sea. Under these conditions, a tiny organism has less exposure. However, an observation must be pointed out here. The planar structure of Brown Seaweed may contribute to a non-uniformity in

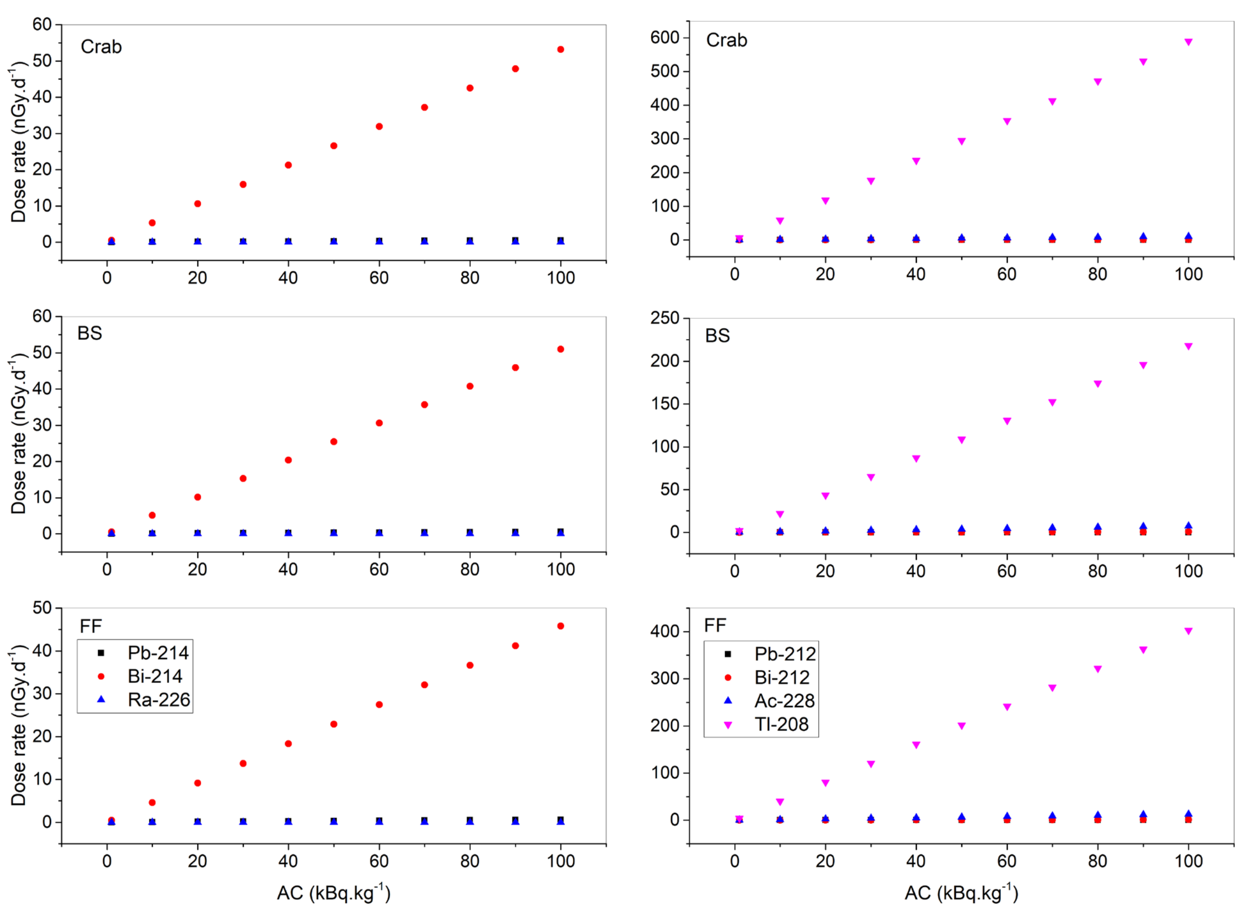

Figure 2. D_R **due to the elements of uranium and thorium series.**

Table III. Comparison between the D_R's of each radionuclide for each organism.

Series	Radionuclides	FF	BS	Crab
Uranium	Bi-214 > Pb-214	72x	85x	97x
	Pb-214 > Ra-226	125x	126x	93x
Thorium	Tl-208 > Ac-228	31x	29x	58x
	Ac-228 > Bi-212	12x	12x	14x
	Bi-212 > Pb-212	7x	25x	5x

the analyzes, inducing an unexpected difference in D_R's (Figure 4).

The D_R's obtained in the current study preserve a linear pattern in the AC function (Figure 3 and Figure 4), indicating the consistency of the results. It should be noted that these values are reasonable approximations since the order of magnitude is the factor that should be considered in cases of biota exposure to environmental radiation (Beresford et al. 2008, Batlle et al. 2011).

Table V shows some D_R's available in the literature. Considering an accident with oil spilled AC around 100 kBq.Kg^{-1}, an acceptable approximation for crude oil (Godoy & Cruz 2003, Attallah et al. 2012), the D_R obtained in the current study would not exceed 10^3 nGy.d^{-1} (Figure 5). This estimated D_R is higher than the D_R for environmental control situations (Pereira et al. 2020), higher than D_R for a high background radiation area (Pereira et al. 2008), and much lower than the D_R for severe nuclear accidents (Aliyu et al. 2015). Moreover, Pereira (2010) evaluated absorbed D_R due to natural radionuclides along the Brazilian coast. The study is focused on obtaining an absorbed dose pattern for biota using marine fish as a tool. The average value obtained by Pereira (2010) was 10^2 nGy.d^{-1}. Therefore, the consideration made at the beginning of the current paragraph (accident with oil spilled AC around 100 kBq.Kg^{-1}) exceeds the radioactive background level by one order of magnitude.

With the linear relationships obtained in Figure 5, it was possible to obtain dose conversion coefficients for each organism as an oil spill AC function (Table VI).

Thus, the current study's values aid in estimating the biota's exposure due to an oil spill accident. The analyses presented here must corroborate the already established techniques, such as Robles et al. (2007) and Brown et al. (2008). The total D_R can still verify the level of biota damage due to natural radiation in detail since more realistic reference animals are being implemented (Higley et al. 2015).

Current data acquisition is essential since the dose may induce harmful effects in biota for extreme conditions, such as reduced fertility and even mortality for hatchlings (ICRP 2008). Therefore, our study shows that exposure to seawater organisms should be considered an emergency criterion for local decontamination in an oil spill accident.

Finally, the unprecedented results obtained here show that in an accident where oil removal is impossible, one must develop technologies that reduce the radioactive effects of the radionuclides that contribute most to the D_R, like Tl-208 and Bi-214, if there are possibilities of developing immediate remediation techniques.

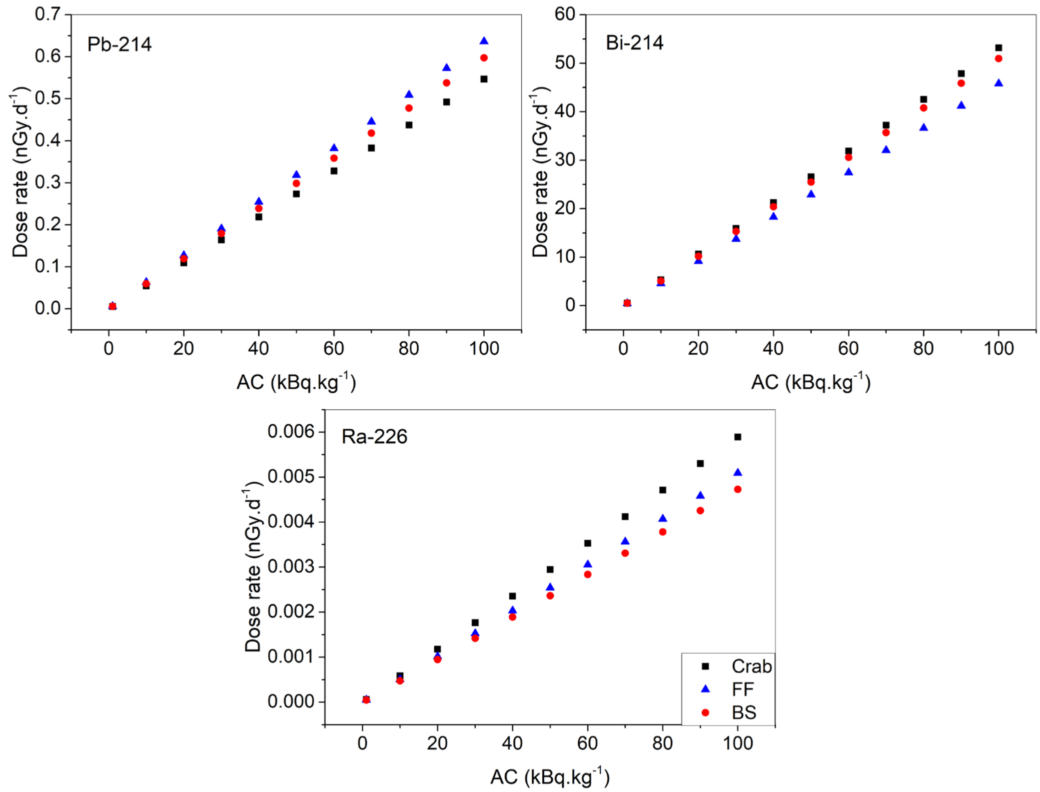

Figure 3. ***D**$_R$*’s contribution due to the uranium series for each organism.**

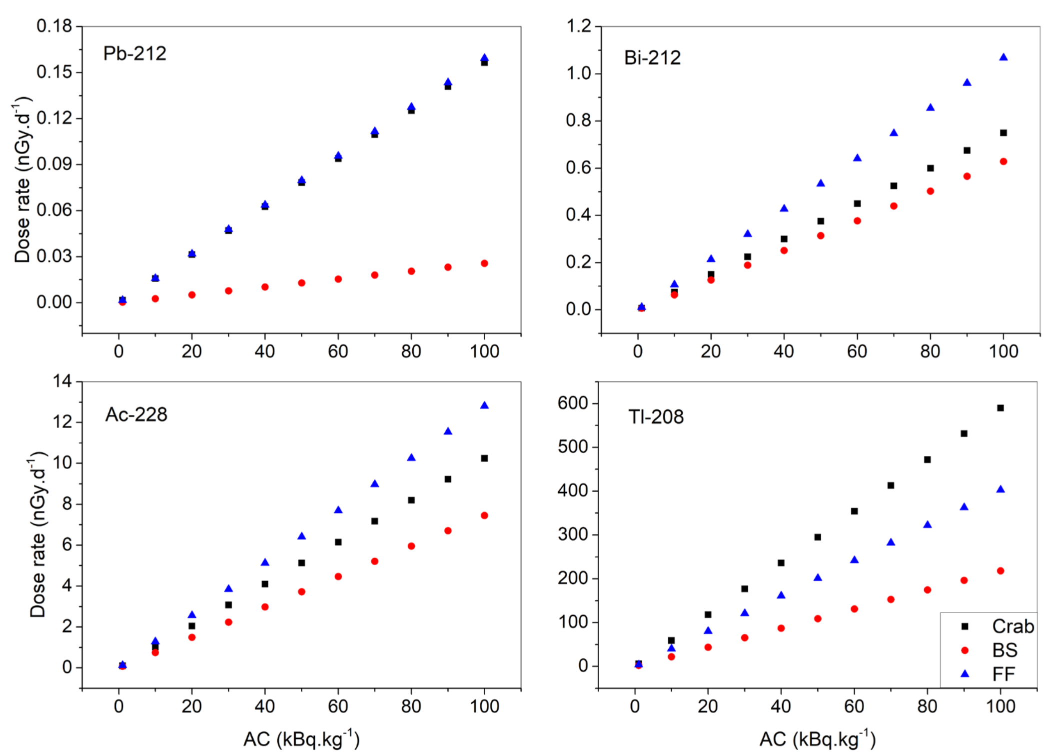

Figure 4. ***D**$_R$*’s contribution due to the thorium series for each organism.**

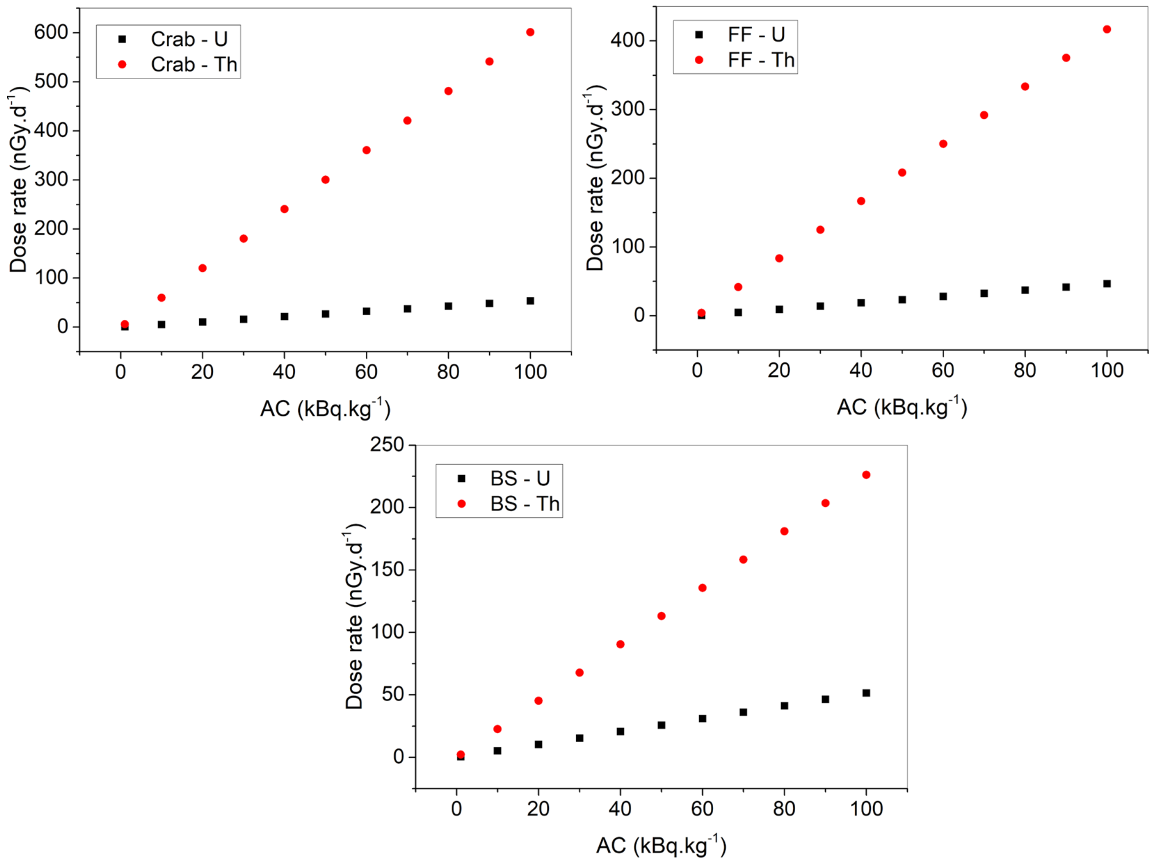

Figure 5. Total D_R due to uranium and thorium series for each organism in function of the AC.

Table IV. Percentage of D_R due to each radionuclide.

Series	Radionuclides	BS	FF	Crab
Uranium	Ra-226	0.01%	0.01%	0.01%
	Pb-214	1.16%	1.37%	1.02%
	Bi-214	98.83%	98.62%	98.97%
Thorium	Pb-212	0.01%	0.04%	0.03%
	Bi-212	0.28%	0.26%	0.13%
	Ac-228	3.29%	3.07%	1.71%
	Tl-208	96.42%	96.63%	98.14%

Table V. Estimated D_R obtained in the literature for different environmental situations.

Conditions	Organism	Dose ($nGy.d^{-1}$)	Radionuclides	Reference
Internal doses rate in fish that live in a dam that was built to store water for the Ore Treatment Unit.	*Hoplias spp* (fish)	$4 \cdot 10^1$	U-238, Th-232 and Ra-226	Pereira et al. 2020
	astyanax spp (fish)	$6.2 \cdot 10^2$		
Evaluation of absorbed D_R in fish from Brazilian coast.	Four (4) species of fish from three (3) Brazilian States	~10^2	U-238, U-235, U-234, Th-232, Th-230, Th-228, Ra-226, Ra-228, Pb-210	Pereira 2010
Assessment of D_R in fish from coast of Ceará, Brazil.	*Lutjanus cyanopterus* (cubera snapper)	14.7	U-238, Ra-226, Pb-210, Th-232 and Ra-228	Pereira et al. 2010
Assessment of D_R around uranium mining at Caetité, Brazil.	*Tilapia nilotica* (Tilapia Fish)	6.9	U-238, Ra-226, Pb-210, Th-232 and Ra-228	Pereira et al. 2008
Total dose rate in cooling pond of Chernobyl nuclear power plant (NPP) ten year after accident	Pelagic fish	$4 \cdot 10^4$	Several artificial radionuclides	Kryshev & Sazykina 2012
	Benthic fish	$2 \cdot 10^5$		
Total dose rate near the Fukushima NPP at the early stage of the accident	Benthic fish	~$1.7 \cdot 10^9$	Several artificial radionuclides	Aliyu et al. 2015
	Macroalgae	~ $3.1 \cdot 10^9$		
Total dose rate in the Republic of Korea 1 year after the Fukushima NPP accident	Fish and frog	~ $2.0 \cdot 10^1$	I-131, Cs-134 and Cs-137	Keum et al. 2013
Total dose due to discharge of liquid effluents from a nuclear medicine facility	Fish	$2.7 \cdot 10^2$	I-131	Carmo 2019
	Crustaceans	$2.8 \cdot 10^3$		

CONCLUSIONS

Encouraged by the oil spill accident along the Brazilian coastal waters in 2019, a fundamental and conservative parameter was developed to assess the marine biota's external dose. A detailed study was done considering the dose contributions in each organism with the Monte Carlo simulation aid and using the ICRP species' baseline reference. Additionally, dose conversion coefficients were obtained for Crab, Flatfish, and Brown seaweed, for both uranium and thorium series, for organisms located at a depth of 1 m (Table VI). The obtained results are not restricted to the accident that occurred on the Brazilian coastline. However, they can be used to assess the occurrences of oil spilled in the past or used in accidents that may occur in the future, as long as the oil spilled geometric conditions can be reasonably approximated to that presented in the current methodology.

It should be emphasized that the current study meets the ICRP guidelines for environmental radioprotection since the results obtained are helpful to prevent or reduce the

Table VI. **Dose conversion coefficient for external exposure due to gamma radiation, in (nGy.d^{-1})/(kBq. kg^{-1}).**

Serie	Crab	Flatfish	Brown seaweed
Uranium	0.54	0.46	0.52
Thorium	6.01	4.17	2.26

harmful effects of radiation on the environment to a level of negligible impact, aiming to the maintenance of biological diversity and ecosystems (ICRP 2008). It is also possible to use the dose coefficients to evaluate the maximum time that the biota can be exposed to oil radiation without damage (ICRP 2008). It is helpful for establishing protocols to suggest limits for removing oil from the water surface. This study is the first approximation to estimate the biota D_R's due to an oil spill accident. The next step of our research will be to study each radionuclide's contribution in terms of depth. It is also intended to implement mathematical models (Van Cleef 1994, Li et al. 2015, Wilson et al. 2019) to refine the values and increase this first approximation accuracy.

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