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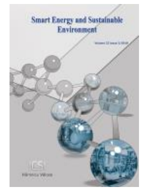

DETERMINATION AND EVOLUTION IN TIME OF RADIOACTIVITY LEVELS IN FRESHWATER FISH (*CARASSIUSGIBELIO*) SAMPLED FROM OLT RIVER, ROMANIA

Mihaela Roxana Ene[1], Narcisa Ghimis[2*], Gheorghe Bulubasa[2], Alina Miu[2]

[1]National Agency for State Reserves and Special Problems - Territorial Unit 135, 1 Depozitului Street, 247125, Dăești, Vâlcea, Romania
[2]National Research and Development Institute for Cryogenics and Isotopic Technologies – ICSI Rm. Valcea, 4 Uzinei Street, P.O. Box Râureni 7, 240050, Rm. Valcea, Romania

Abstract:

The paper presents a monitoring activity of the Freshwater fish (*Carassius gibelio*) samples consumed in Olt River, Romania. A monitoring study lasting 9 years (2014-2022) was realized. Two sampling points for the Olt River were chosen, the first one upstream of the discharge point of the magistrate channel "Deversor Olt" (background location) and respectively the second sampling point downstream of the liquid effluents discharge point (indicator location). A maximum activity concentration for ^{40}K of 95.75 Bq·kg^{-1} fw, is observed. The activity of interest radionuclides (^{7}Be, ^{51}Cr, ^{60}Co, ^{137}Cs, ^{226}Ra, ^{235}U) is below the detection limit. The method used was high resolution gamma ray spectrometry performed with an HPGe detector.
It was observed that the values recorded for the background location had a slight difference compared with the values recorded in the indicator locations. The nuclear activity in the pre-operational stage of the Experimental Pilot Plant for Tritium and Deuterium Separation did not have any impact on the environment so far.
This study creates a unique database that could be relevant for further Operational Program for Environmental Monitoring of the Experimental Pilot Plant and other economical agents in the area.

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1. INTRODUCTION

Fish is among the most consumed animal product by the population in the interest area, which is Valcea County, Romania. This product is obtained by fishing in the Olt River. In this river, the predominant species is crucian carp (*Carassius gibelio*). The yearly

Corresponding author: Narcisa Ghimis, e-mail: narcisa.ghimis@icsi.ro

consumption of fish in this area is estimated at 13 kg/year per individual, because fishing is a well spread habit among the people in the area. [UNSCEAR, 2000]

The liquid effluents resulted from economic activities on the Chemical Industrial Platform are effused into Olt River. This platform produces hundreds of thousands tons per year of salt, soda ash and organic derivatives. The water in the industrial platform is used for chemical processing, discharge of wastes and electricity production. The industrial waste water is continuously monitored, when overflows of inorganic nitrogen contaminants are registered at high production. [Popescu et al., 2015]

The exhaust systems for used water spill into the Olt River are as follows:

A. The open Channel «DeversorOlt» - used for the evacuation of the locally pre-treated and treated water that is chemically not-biodegradable, and also the conventionally clean water and rain water.
B. The Biological Purifying Unit which exhausts in Govora River, affluent of Olt River on the right side. This unit chemically and biologically treats the water from production units of Oltchim S.A, and the sewage from Chemical Industrial Platform Ramnicu Valcea

In this exhaust system is also found the industrial and process water from the Experimental Pilot Plant for Tritium and Deuterium Separation (PESTD), from National Research and Development Institute for Cryogenics and Isotopic Technologies-ICSI Rm. Valcea.

The main objective of the PESTD is to demonstrate the viability an efficiency of the detritiation technology developed for the extraction of tritium from the moderator heavy water of CNE Cernavoda nuclear power plant. The adopted technological solution for detritiation is based on the tritium transfer from the liquid phase (tritiated water) to the gaseous phase (tritiated deuterium gas) through a catalysed isotopic exchange process followed by a final concentration of tritium using cryogenic distillation. As a final step, the obtained pure tritium gas is safely stored in getter beds as a metal hydride.

In the tritiated moderator heavy water from CANDU reactors, tritium is the most abundant radioisotope, but others radionuclides are also present, created by activation of corrosion products. Before being processed in the detritiation plant, the heavy water will be purified in the purification systems of the CNE Cernavoda nuclear power plant. Most of the beta-gamma emitters will be eventually retained in these purification systems, but a small amount remains in the water. Radioactive substances can be introduced in Olt River in small quantities from this Experimental Pilot Plant for Tritium and Deuterium Separation.

2. SHORT REVIEW

In this paper, we consider a critical group hypothetically exposed through all the significant contamination paths: fish ingestion from the Olt River, vegetal products ingestion from areas irrigated with Olt River water, animal products from animals contaminated with water from Olt River or animal food from areas irrigated with water from Olt River. This paper aims to study the radioactive pollution degree of the fish intended for human consumption.

So, we realised a monitoring study during 9 years (2014-2022), in which we sampled several fish samples *(Carassius gibelio*) along the Olt River in order to determine the activity concentration. The samples were collected from two locations, upstream (reference location) and downstream (indicator location) from the Deversor Olt Channel. These locations are shown in figure 1. Also, in Table 1 some details about these sampling locations are shown.

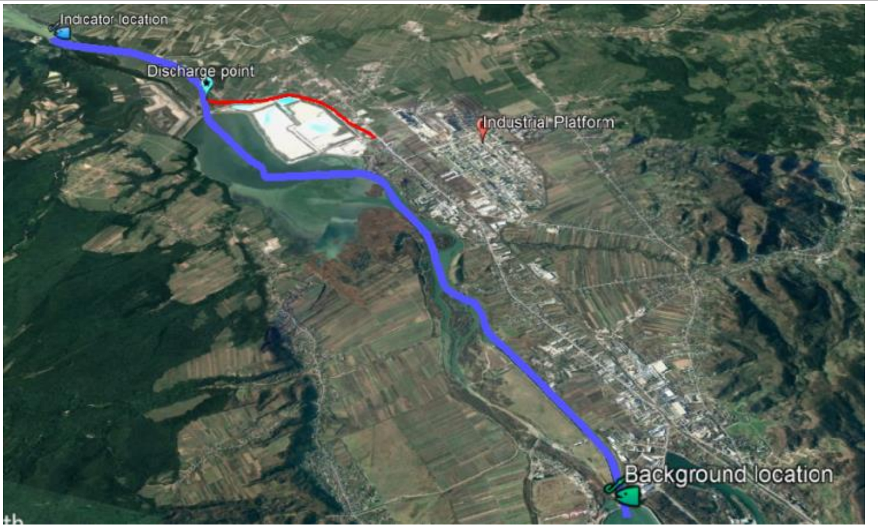

Figure 1. Map of the study area showing: Olt River, Deversor Channel, Sampling Locations (Background and Indicator), Discharge point of Deversor Channel in Olt River, Industrial Platform Ramnicu Valcea

Table 1. Summary of sampling locations, and one-line distance from discharge point.

Site/ Sampling locations		**Sampling location**		Distance from discharge point of Deversor Olt (km)
		Latitude ^{0}(N)	Longitude ^{0}(E)	
Upstream from discharge point of Deversor Olt	Background location (UP)	45^{0}04’16”	24^{0}21’46”	9.17
Downstream from discharge point of Deversor Olt	Indicator location (DW)	44^{0}57’32”	24^{0}15’36”	5.93
Discharge point of Deversor Olt	Deversor Olt	45^{0}00’28”	24^{0}17’35”	-
Chemical Industrial Platform	Industrial Platform	45^{0}02’54”	24^{0}18’12”	4

This study is also part of the Pre-operational Program for Environment Monitoring from the PESTD, from National Research and Development Institute for Cryogenics and Isotopic Technologies-ICSI Rm. Valcea. This program’s goal is to determine the “zero” level before the commissioning of the heavy water detritiation pilot plant.

The issue of radioactivity in fish samples from the Olt River has not been studied before, because, probably, it was not considered necessary, taking into account that the activity from Chemical Industrial Platform did not include any nuclear practice. Nevertheless, the study is necessary in the idea that a reference level should be determined, to be able to identify a radioactive contamination.

Other studies have focused on radioactivity levels in fish species but not in freshwater fish from Olt River. There are studies about fish from seawater as anchovy, sediments or seawater. [Baltas Hasan et al., 2017]

3. MATERIAL AND METHODS

3.1 Sampling methodology and preparation

The radionuclides followed in our study are: ^{7}Be, ^{40}K, ^{51}Cr, ^{60}Co, ^{137}Cs, ^{226}Ra and ^{235}U. Approximately 5 kg of fresh fish were sampled from each location. Sampling was effectuated with adequate fishing equipment. Excessively large or excessively small fish was avoided. The ideal fish is 0.5 kg heavy. The sampling was performed in accordance with the regulation for the practice of recreational / sport fishing and the applicable laws.

The raw fish was eviscerated, removing the inedible parts. The sample was washed with distilled water, was dried in drying stove at 110°C until its mass stopped decreasing. Afterwards, the dried fish was burned in an electric oven at 420°C, until the ash became light grey. After cooling, the resulted ash was grinded with a pestle in a mortar. Then, the grinded ash was sieved through a 240 µm mesh sieve. The ash samples were sealed in cylindrical polyethylene plastic recipients of 150 ml volume which respect the geometry of the calibration source, and were weighted precisely (uncertainty of $\pm 2\cdot10^{-4}$ g). In order to avoid the humidification, the dried samples were kept in a desiccator between the preparation steps.

3.2 Gamma spectrometry measurement

Gamma-ray spectra were measured by a coaxial HPGe detector with 65% relative efficiency and 1.85 keV energy resolution at 1332.5 keV, equipped with a lead shielding, and multichannel digital analyser DSPEC Plus. The spectra were analysed by the GammaVision software were an interactive analytical mode was applied to optimize peak fitting. Each sample was analysed for 25 200 s.

Activity concentration C $(Bq\cdot kg^{-1})$ of each radionuclide present in sample was obtained from the area of the individual peak at energy E using formula (1):

$$C\ (Bq\cdot kg^{-1}) = \frac{N/t}{P_E\cdot\varepsilon_E\cdot m\cdot f_d\cdot f_{cc}} \tag{1}$$

where t is the sample spectrum counting tine given in seconds time of measurement; P_E is the probability of the emission of a gamma ray with energy E of each radionuclide, per decay; ε_E is the efficiency of the detector at energy E at actual measurement geometry; m is the mass of the fresh fish given in kilograms; f_d is the correction factor for decay for a reference data and f_{cc} is summing coincidence factor.

When a radionuclide emits gamma radiation at more than one energy, the reported activity concentration is the one calculated for the energy with the biggest emission probability, and the other ones are watched only qualitatively, to confirm/infirm the presence of the radionuclide in the measured sample. As an exception, for the radionuclides that have two or more energies with emission probabilities with values close one to the other, (e.g. ^{60}Co), the reported activity concentration is a pondered average of the calculated activity concentration for each of the energies.

Minimum Detectable Activities (MDA's) are calculated and reported according to the latest ISO 11929 standard. MDA's are calculated using a coverage factor of 1.645, corresponding to a 95% confidence level, and reported as <VALUE. [ISO 11929-1]

Efficiency calibration curve established for the same geometry used for test samples using standard mixed gamma Certified Reference Material which is traceable to SI. The calibration source contains gamma emitters with energies covering the desired energy range, namely, ^{60}Co (1173.2 keV and 1332.5 keV), ^{133}Ba (81 keV, 302.8 keV, 356 keV), ^{137}Cs (661.66 keV) and ^{241}Am (26.34 keV, 59.54 keV). The activity concentration of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{241}Am is given as the certified value for certified reference material.

^{40}K, ^{137}Cs and ^{226}Ra activity concentrations were determined using the gamma lines from 1460.8 keV, 661.6 keV and 186.2 keV respectively. [Al-Qaradawi et al., 2015] The activity values were given as Bq·kg^{-1}in fresh weight.

3.3 Dose estimation

In the Pre-operational Program for Environment Monitoring from the Experimental Pilot Plant for Tritium and Deuterium Separation, it's important to determine the local factors that might influence the values of radiation doses received by the population (meteorological and hydrological, hydro-biological conditions of the aquatic environment, population distributions, rates of food consumption, etc.). According to ICRP [ICRP, 1996], the ingested dose is given by:

$$H_{T,r} = U \sum \left(C^r g_{T,r} \right) \qquad (2)$$

where U is the consumptions rate (kg·y^{-1}), C^r is the activity concentration of the radionuclide r of interest (Bq·kg^{-1}) and $g_{T,r}$ is the dose conversion coefficient for the ingestion of radionuclide r (Sv·kg^{-1}) in tissue T. For adults, the recommended dose conversion coefficient $g_{T,r}$ for ^{40}K and ^{137}Cs are 6.29 x 10^{-9} Sv·Bq^{-1} and 1.3 x 10^{-8} Sv·Bq^{-1}, respectively.

4. RESULTS

The activity concentration and ingested dose was raported in Table 2.

Table 2. The activity concentration ± standard error at 95% confidence, in freshwater fish (*Carassius gibelio*) sampled from Olt River

Year	**Location**	**Radionuclides**							**H**
		^{7}Be	**^{40}K**	**^{51}Cr**	**^{60}Co**	**^{137}Cs**	**^{226}Ra**	**^{235}U**	
		Bq·kg^{-1} fw							µSv.y^{-1}
2014	UP	<0.55	86.07 ± 8.95	<0.56	<0.04	<0.02	<2.09	<0.99	7.04
	DW	<0.23	89.53 ± 9.37	<0.28	<0.02	0.25±0.06	<1.77	<1.15	7.36
2015	UP	<0.22	38.02 ± 4.02	<0.24	<0.04	0.04±0.02	<1.17	<0.47	3.12
	DW	<0.46	72.06 ± 7.55	<0.33	<0.07	<0.06	<1.85	<0.82	5.89
2016	UP	<0.56	66.97 ± 7.04	<0.34	<0.07	0.07±0.04	<1.92	<0.99	5.49
	DW	<0.68	70.94 ± 7.45	<0.40	<0.09	0.10±0.05	<2.00	<1.18	5.81
2017	UP	<0.58	65.75 ± 3.85	<0.43	<0.08	<0.12	<1.77	<1.45	5.38
	DW	<0.61	64.63 ± 3.77	<0.25	<0.10	<0.08	<1.88	<1.52	5.28
2018	UP	<0.64	38.33 ± 4.73	<0.57	<0.12	<0.11	<2.29	<1.03	3.13
	DW	<0.46	59.65 ± 6.76	<0.41	<0.13	<0.07	<3.73	<1.68	4.88

2019	UP	<0.78	85.61 ± 10.9	<0.86	<0.15	<0.12	<2.35	<1.31	7.00
	DW	<1.03	95.75 ± 11.9	<0.81	<0.12	<0.11	<3.59	<1.73	7.83
2020	UP	<0.40	73.52 ± 3.91	<0.21	<0.04	<0.05	<1.69	<1.07	6.01
	DW	<0.35	94.96 ± 5.04	<0.42	<0.05	<0.04	<2.31	<1.23	7.76
2021	UP	<0.27	93.95 ± 5.77	<0.30	<0.06	<0.04	<0.97	<0.14	7.68
	DW	<0.87	93.62 ± 5.73	<0.26	<0.05	<0.04	<1.09	<0.26	7.66
2022	UP	<0.16	76.20 ± 4.34	<0.16	<0.04	<0.02	<1.28	<0.29	6.23
	DW	<0.46	84.12 ± 4.80	<0.15	<0.03	<0.02	<0.86	<0.16	6.88

In the general case, doses from the ingestion of natural radionuclides in foods and drinking water have been estimated from measured concentrations of the radionuclides in body tissues or organs. For ^{40}K, metabolic balance mentains body levels irrespective of intake amounts.

The activity concentration of the primordial radionuclid of ^{40}K in freshwater fish *(Carassius gibelio*) sampled from Olt River presents an uniform evolution (Figure 2). The highest concentration 95.75±11.9 Bq·kg^{-1} fw was recorded in 2019, in the indicator location.

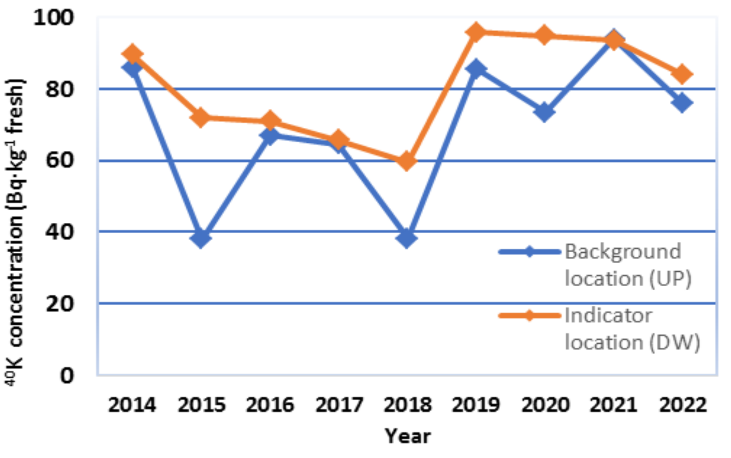

Figure 2. ^{40}K activity concentration in freshwater fish samples collected from Olt River

In all samples, ^{235}Uwas below the detection limit, also, ^{60}Co,^{7}Be and ^{51}Cr. ^{137}Cs is found in low concentrations, seldom below the detection limit. The mean value recorded in freshwater fish collected from Fukushima Prefecture, in 2016, where the average value of ^{137}Cs is 1.84 x 10^{3}Bq·kg^{-1}- wet, is significantly higher than the maximum recorded value of 0.25 ± 0.06 Bq·kg^{-1}fw in samples from the Olt River.

Figure 3 shows the radioactivity levels of interest radionuclides in freshwater fish (*Carassius gibelio*) sampled from Olt River, Romania, collected in 2014 (the first year of study), 2022 (the last year of study), 2018 (the year with the lowest concentration of activity) and 2019 (the year with the highest concentration of activity).

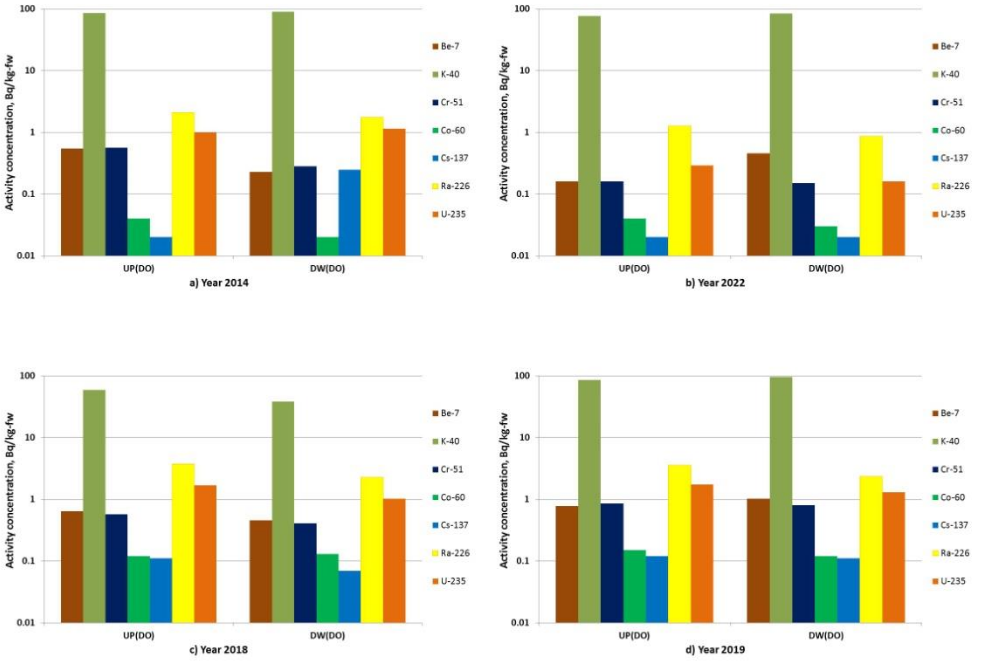

Figure 3. The radioactivity levels of interest radionuclides in freshwater fish (*Carassius gibelio*) sampled from Olt River, Romania, collected in: a) 2014, b) 2022, c) 2018 and d) 2019. Values below detection limit are included

Activity concentration of ^{137}Cs in freshwater fish samples, in year 2016, was 0.07 ± 0.04 Bq·kg^{-1}fw in indicator location and 0.10 ± 0.05 Bq·kg^{-1}fw in background location. These values indicate that there are no significant modifications in the values recorded upstream and downstream from the Deversor-Olt Channel.

The high value of the detection limit for ^{226}Ra is due to the interference of the peak from 186.2 keV with Uranium-235's peak from 185.7 keV.

In Table 3 are presented the average values of the activity concentration of some of the interest radionuclides from different fish species from random locations.

Table 3. The average values of the activity concentration (Bq·kg^{-1}fw) from different fish species, collected from different locations

Location	**Species**	**^{137}Cs**	**^{40}K**	**^{226}Ra**	**References**
		Bq·kg^{-1}fw			
Black Seain Rize, Turkey	Anchovy	-	77.50 ±1.77	1.16 ± 0.15	[Baltas Hasan et al., 2017]
Black Sea Region of Turkey	Anchovy, trout, bluefin, whiting	0.58 ± 0.04	86.37 ± 1.22	0.35 ± 0.08	[Korkmaz Görür et al., 2012]
Ottawa River	Bass	4.7 ± 0.3	71.5 ± 4.2	< 0.07	[J. Chen et al., 2015]
	Walleye	2.9 ± 0.2	88.0 ± 5.1	-	
	Catfish	2.0 ± 0.1	52.3 ± 3.4	< 0.05	
Mississippi Lake, White Lake	Pike	3.7 ± 0.2	73.3 ± 4.4	< 0.06	

Lake 305 Ontario, Canada	Northern Spike	14.93±0.78	-	< 0.06	[J. Chen et al., 2016]
	Lake Whitefish	4.25 ± 0.25		< 0.08	
Lake 302 Ontario, Canada	Lake Whitefish	5.73 ± 0.33	-	< 0.07	
	White Sucker	4.0 ± 0.2		< 0.06	
Lake 226 Ontario, Canada	Lake Whitefish	5.65 ± 0.3	-	< 0.07	
New Zealand	Seafood	0.05÷0.22	-	-	[Pearson et al., 2016]
Forest river (Takase, Ogaki-shita, Akougi, Amado)	-	1.847 x 10^3	-	-	[Wada et al., 2019]
Irrigation pond (Inkyozaka, Funasawa, Kashiramoni)	-	4.556 x 10^3	-	-	
Olt River, Romania	Carassius gibelio	0.12 ± 0.04	74.98 ± 6.45	<1.92	Present Study

The average activity level of ^{137}Cs, ^{40}K and ^{226}Ra in Anchovy, trout, bluefin, whiting samples from Black Sea Region of Turkey have been investigated reported by Görür F. Korkmaz et al. [F. Korkmaz et al., 2012]. The ranges of activity concentrations in different species of fish were: ^{137}Cs: 0.18±0.02 Bq·kg^{-1} for Anchovy, 0.17±0.03 Bq·kg^{-1} for Trout, 1.34± 0.06 Bq·kg^{-1} for Bluefin, respectively 1.15 ± 0.06 Bq·kg^{-1} for Whiting; ^{40}K: 51.72 ± 0.52 Bq·kg^{-1} for Anchovy, 102.29 ± 1.51 Bq·kg^{-1} for Trout, 101.07 ± 1.49 Bq·kg^{-1} for Bluefin, respectively 108.58 ± 1.78 Bq·kg^{-1} for Whiting. The rangers of average activity concentrations are generally similar to in our study.

Considering that people of the critical group eat fish captured from downstream of the discharge point of the magistrate channel "Deversor Olt", indicator location respectively, the ingested dose by an adult during the study years 2014-2022, is ranged from 3.13 µSv.y^{-1} to 7.83 µSv.y^{-1}. If they eat fish captured from background location, the ingested dose by an adult during the study years 2014-2022, is ranged from 3.12 µSv.y^{-1} to 7.68 µSv.y^{-1} (Figure 4). The dose from ingestion of radionuclides is considered to be very low compared with the total exposure resulting from ingestion of 2400 µSv.y^{-1} in the world [UNSCEAR, 2000].

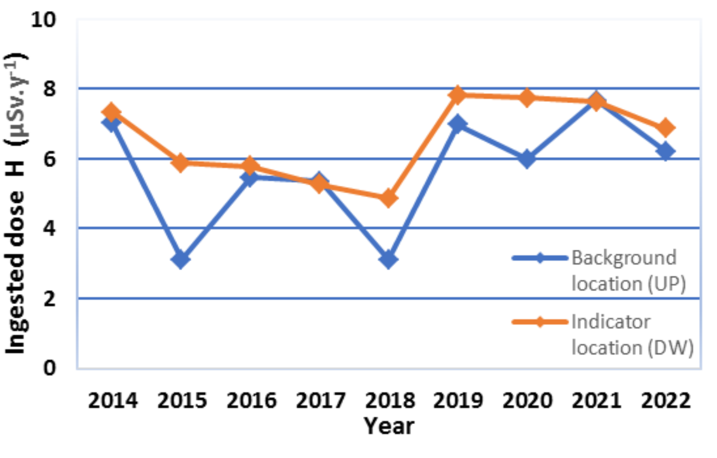

Figure 4. Ingested dose as a result of fish consumption by an adult

5. CONCLUSIONS AND DISCUSSION

Following the distribution of the activity concentration for each interest radionuclide, upstream and downstream from the discharge point Deversor-Olt Channel, no significant modification can be observed, neither in time (along the years), nor in space (the two sampling locations).

We can affirm that our recorded values constitute normal background level, comparable with other values from literature. Also, the dose ingestion of radionuclides as a results of eating fish captured from Olt River is insignificant compared to the total exposure in the world.

In conclusion, we can state that the entire activity of the Chemical Industrial Platform, and implicitly of PESTD, from National Research and Development Institute for Cryogenics and Isotopic Technologies - ICSI Rm. Valcea, does not influence the activity concentration of the radionuclides in freshwater fish (*Carassius gibelio)* sampled from Olt River.

This study creates a unique database that could be relevant for further Operational Program for Environmental Monitoring of PESTD facility and other economical agents in the area.

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Conflict of Interest Statement: The authors declare no conflicts of interest associated with this manuscript.

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