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CALCULATION METHOD FOR DERIVED EMISSION LIMITS VALUES FROM "HORIA HULUBEI" NATIONAL INSTITUTE FOR R&D IN PHYSICS AND NUCLEAR ENGINEERING AREA

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Abstract. The paper presents studies devoted to the assessment of derived emission limits (DELs) for potential radionuclides emitted as gaseous and liquid effluents resulting from the nuclear activities carried out in *"Horia Hulubei" National Institute of Physics and Nuclear Engineering, IFIN-HH*.

The radionuclides in gaseous effluents identified by qualitative and quantitative specific measurements are: Co-60, Cs-134, Cs-137, Ag-108m, Eu-152, Eu-154, Eu-155, and H-3 total. The maximum concentration was found for Co-60 in the quantum of 10^9 Bq·a^{-1}. In the case of liquid effluents, a number of radionuclides, mainly C-14 in a concentration of 10^9 Bq·a^{-1}, I-123 with 10^8 Bq·a^{-1} concentration, and Tc-99m and Mo-99, in concentrations of up to 10^{10} Bq·a^{-1} were found. The calculations are based on these data and an approved annual effective dose constraint for the critical group from the Magurele town of $E_{\mathrm{constr}} = 100$ μSv·a^{-1}.

Key words: derived emission limits (DELs), dose constraints, liquid/gaseous effluents.

1. INTRODUCTION

IFIN-HH is situated in the Măgurele town, at a distance of 0.5 km from the circle line of the Bucharest city and at 8 km from the downtown. Some departments, considered as potential sources of radioactive effluents, as liquid or gas, are operated within the Institute: i) station for the treatment of radwastes (STDR); ii) radioisotope and radiation metrology department, operating the radioisotope center building (DRMR) and the new center for radiopharmaceutical research, owing a cyclotron type T19 (CCR). Another special source of radioactive effluents is the nuclear reactor (RN) VVR-S type, under the last phase of decommissioning, which will cease to release radioactive effluents after the finalization of the operations. For RN, the derived emission limits, evaluated for the radionuclides of interest, were established and published in the paper [1].

This paper is intended to establish derived emission limits for both types of effluents, liquid and gaseous, considering the whole list of radionuclides involved in all installations of IFIN-HH and taking into account the activity concentrations of the entire inventory of radioisotopes as to be found at the present time and as planned to be implied in the future, according to the present and future research plans.

The main interest points, where the influence of effluents released from IFIN-HH units must be quantified, are: the population of the Magurele town with the downtown at about 1 km south-west of IFIN-HH, Jilava village in south-east and rivers Ciorogârla, Sabar, and Argeș, situated at respectively 0.8 km, 1.2 km, and 5 km in south-west. All these geographical considerations must be taken into account according to the Romanian Norm on the limits for radioactive effluent's release [2].

The qualitative and quantitative determinations of activity concentrations for both types of effluents, liquid and gas, were performed in the IFIN-HH laboratories, designated by the National Commission for the Control of Nuclear Activities (CNCAN) as notified laboratories for testing in the nuclear field, or accredited by the national accreditation body (RENAR).

The used equipment and the applied measurement methods are gamma-ray spectrometry, tritium (^{3}H), gross alpha/beta/gamma activity measurement, and iodine (^{131}I) monitors.

CNCAN approved for IFIN-HH a dose constraint for a person belonging to the critical group of $E_{\text{constr}} = 100\ \mu\text{Sv}\cdot\text{a}^{-1}$. The critical group, defined in [3], represents a group of the population members, reasonably homogeneous as regarding its exposure to a radiation source, whose individuals receive the highest effective or equivalent dose, if applicable, for the given pathway of exposure.

2. EQUIPMENT AND ANALYSIS METHODS

Monitoring of each source of effluents is carried out as follows:

The DRMR, a suspect installation of releasing tritium and iodine gaseous effluents, is monitored permanently with tritium and iodine monitors, placed on the top of a chimney, at a height of 40 m; a mean concentration is calculated from the measured values, for a given monitoring period. The measurements are performed by the gamma-ray spectrometry method, for each individual gamma-ray emitter.

For monitoring H-3 in gaseous effluents, the tritium monitor OS1700 type is used and the LSC spectrometer model TRICARB 2800 TR is used for measurement.

The liquid effluents, consisting of certain and suspect radioactive waters, are measured both by the gamma-ray spectrometry and as gross alpha/beta/gamma activity.

The complex decommissioning of RN involves both types of effluents [4 JER] monitoring: i) gaseous effluents containing dust and solid radioactive powders resulting from the dismantling of RN structure; ii) liquid effluents resulted from the primary circuit, the spent fuel cooling pond, the spent fuel storage pools, as well as from the operations of dismantling of the metals and concrete – sludge – and operations of decontamination of personnel and working tools.

The gaseous effluents are retained on the ventilation's filters, at the place of production, at chimney evacuation, and on the filters attached to the local monitors.

The air contamination is performed in two ways: i) by using the Fixed Radiometric System, which registers the gross volumetric activity; ii) from the evacuation chimney, at a height of 40 m.

Both types of liquid effluents produced by DRMR and decommissioning of RN are measured in advance and then are transferred to the 300 cubic meter pools belonging to DNDR, where they are treated and finally released in the Ciorogârla river, the receiver in agreement with CNCAN approved derived limits for IFIN-HH. The final activity concentration of the liquid effluents is measured by gamma-ray spectrometry.

3. EXPERIMENTAL SECTION

3.1. RADIONUCLIDES TO BE FOUND IN THE EFFLUENTS

From the analyses and summarizing of all effluents, it was found that the two ways of discharge can have the following radionuclides, present and planned to be implied:

– Gaseous effluents: ^{60}Co, ^{134}Cs, ^{137}Cs, ^{108m}Ag, ^{152}Eu, ^{154}Eu, ^{155}Eu, and ^{3}H;

– Liquid effluents, according to the Table 4, among which the following radionuclides are to be found in considerable concentrations:^{14}C, ^{123} I, ^{3}H, ^{99m}Tc, and ^{99}Mo.

In order to establish the annual derived limits, it is necessary to determine the annual doses for the critical group, corresponding to the intended discharges for all the emitted radionuclides, after the evaluation of the relative weights of the individual doses due to each radionuclide and each pathway of release, as to meet the constraint for the effective annual dose, according to the relation [2, 4]

$$\sum_i \sum_k (f_{ik}) \cdot Q^*_{ik} \leq \frac{E_{constr}}{\Gamma}, \tag{1}$$

where:

– f_{ik} is the maximum annual effective dose for a person from the critical group, due to the release of an activity of 1 Bq from the radionuclide i on the evacuation way k.

– Q^*_{ik} is the derived limit for the annual evacuation, expressed in Bq/a, for the radionuclide i, or the group of radionuclides i, on the evacuation way k.

– E_{constr} is the constraint of effective annual dose, expressed in $Sv \cdot a^{-1}$, for the persons from the critical group due to the exposition to ionising radiations due to the radioactive effluents. CNCAN approved the value of $E_{constr} = 100\ \mu Sv \cdot a^{-1}$ for IFIN-HH.

– Γ is a safety factor, taking into account the uncertainties due to the used model for the calculation of doses. From conservatism reasons, $\Gamma = 1$ was proposed to assure the dose constraints.

The methodology used for the calculation of the DELs for IFIN-HH contains the transport model in the environment using the compartment concept, according to Fig. 1.

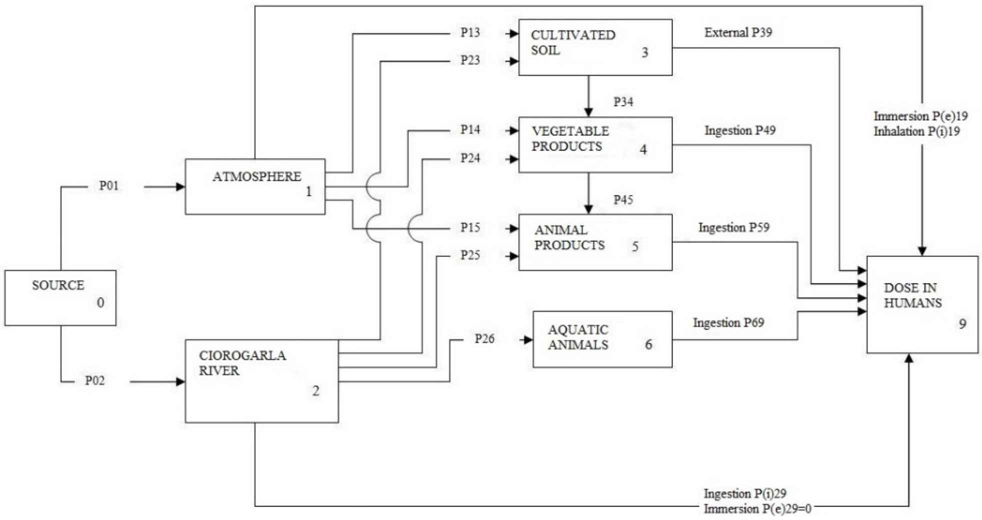

Fig. 1– Model for the transfer of effluents from IFIN-HH (source) in the environment.

Each compartment is numbered and the quantity of activity in the compartment “i-th” is denoted by X_i.

The activity transfer from the compartment i-th to the compartment j-th is characterized by the transfer parameter P_{ij} in such a way that the transferred activity in conditions of stability from the compartment i-th to the compartment j-th is $P_{ij}X_i$.

The quantity of activity represented by each compartment j-th is

$$X_j = \sum_i P_{ij} X_i . \tag{2}$$

The summation is done for all the compartments i, transferring activity to the compartment j-th.

For the calculation of the Derived Emission Limit (DEL) for a specified radionuclide and a critical group, the applicable limit dose must be divided by the quantity X_9/X_0.

X_9 is the dose rate received by an individual (Compartment 9); X_0 is the release rate from the source (Compartment 0). The relation for the calculation of DEL is then,

$$\mathrm{DEL} = \frac{E_{constr}\,[\mathrm{Sv}\cdot\mathrm{a}^{-1}]}{X_9/X_0\,[\mathrm{Sv}\cdot\mathrm{a}^{-1}\cdot\mathrm{Bq}^{-1}\cdot\mathrm{s}]} \tag{3}$$

DELs, in $\mathrm{Bq\cdot s^{-1}}$, are calculated in several steps:

– Identification of the critical group and of the exposition ways. In our case, the critical group is identified as the populated zone of the Magurele town, placed at a distance of 1 km from the source X_0 (IFIN-HH) and consists of the infants of 0–1 years old and adults. It is supposed that the fruits, vegetables, and animal products are produced in their own households.

– Development of the expressions between the evacuation rates for air, X_0 (air) and water X_0 (water) and the dose rate of an exposed individual, X_9, according to the previous equations.

– The DELs are calculated from the limit dose for the whole body, separately for infants (0–1 years) and adults.

– The choice of the adequate values of the relevant transfer parameters for the considered critical group; the values of these parameters can be found in various publications [2].

– For each radionuclide, the minimum value from DELs, as calculated in the previous step is chosen.

The repartition of the emissions for the potential radionuclides on each evacuation way was done taking into account the evaluation of the research activities in the nuclear field planned to be deployed in the future and a real situation [3–6]. In Table 1, the repartition was 10% for aerial release and 90% *via* liquid discharge. The individual contributions for each radionuclide were established from this choice [7, 8].

Taking into account the relative weights of each radionuclide, on each release way, the derived limits for annual emissions are established in such a way, as to satisfy globally the effective annual dose constraint, according to the formula:

$$\sum_i \sum_k f_{ik} \cdot Q_{ik} \le \left(\frac{100\ \mu\mathrm{Sv}\cdot\mathrm{a}^{-1}}{1} \right), \tag{4}$$

where *i* represents the radionuclide and *k* is the discharge way.

Table 1

Emission weights and annual effective dose constraints for the radionuclides *i* and discharge *k*

Discharge way (*k*)	**Weights [%]**	**Radionuclide (*i*)**	**Weights [%]**	$E_{constr\ i}$ [μSv·a^{-1}]
Gas	10% (10 μSv·a^{-1})	Co-60	73.91	7.391
		Cs-134	0.15	0.015
		Cs-137	12.00	1.200
		Ag-108m	0.60	0.060
		Eu-152	0.40	0.040
		Eu-154	8.64	0.864
		Eu-155	2.30	0.230
		H-3 total	2.00	0.200
Liquid	90% (90 μSv·a^{-1})	I-123	1	0.900
		I-125	1	0.900
		I-131	1	0.900
		Cs-134	1	0.900
		Cs-137	8.4	7.560
		Am-241	14	12.600
		Co-60	8	7.200
		Ir-192	2	1.800
		Eu-152	1	0.900
		H-3 total	47	42.300
		Lu-177	2	1.800
		Tc-99m	1.5	1.350
		Re-188	2	1.800
		C-14	2	1.800
		Ga-68	1	0.900
		U-235	0.1	0.090
		U-238	2	1.800
		Mo-99	2	1.800
		Zn-65	1	0.900
		Na-24	1	0.900
		Co-58	1	0.900

Transfer parameters:

- P_{01} – the atmospheric dispersion parameter, which considers the activity dispersion/dilution in the air from the source up to the critical group. For a mean wind speed of 2 m/s in the IFIN-HH zone has resulted: $P_{01} = 5 \times 10^{-6}$ s×m^{-3} [9];
- P_{12} – the transfer from air to surface water in Măgurele zone, where no lakes exist, so it was not considered;
- P_{13} – the transfer of radionuclides from atmosphere to soil;
- P_{14} – the transfer of radionuclides on vegetal products [m^3·kg^{-1}];
- P_{15} – the transfer of radionuclides from atmosphere to the animal product [m^3·kg^{-1}];
- $P(i)_{19}$ – the transfer from atmosphere to the human being, parameter connecting the dose produced by inhalation of the radioactive material [Sv·a^{-1}] with the activity concentration in air [Bq·m^{-3}], expressed in [Sv·a^{-1}·Bq^{-1}·m^3];

– $P(e)_{19}$ – the transfer factor immersion in the air in [$Sv\cdot a^{-1}\cdot Bq^{-1}\cdot m^3$];
– P_{23} – the contamination of the soil through irrigation water – considered only for liquid evacuations and not considered for gaseous releases;
– P_{24} – the contamination of the vegetal products *via* irrigation waters, considered only for liquid discharges;
– P_{25} – the water ingestion of the terrestrial animals, considered only for liquid evacuations;
– P_{34} – the absorption of plants from the soil; it connects the specified radionuclide concentration in vegetation [$Bq\cdot kg^{-1}$] with the superficial activity of the soil surface [$Bq\cdot m^{-2}$], in [$m^2\cdot kg^{-1}$];
– P_{39} – the external dose due to the atmospheric fallout on the soil [$Sv\cdot a^{-1}\cdot Bq^{-1}\cdot m^2$];
– P_{45} – the transfer parameter from vegetation to the animal product;
– P_{49} – the human ingestion from vegetal products [$Sv\cdot a^{-1}\cdot Bq^{-1}\cdot kg$];
– P_{59} – the human ingestion from animal products [$Sv\cdot a^{-1}\cdot Bq^{-1}\cdot kg$];

Both P_{49} and P_{59} connect the ingestion dose with vegetal and animal products consumption;

– P_{02} – the dispersion of the liquid effluents, connecting the concentration in water, X_2, in a given place, with the evacuation rate, $X_0(w)$, [$s\cdot L^{-1}$]; for the calculation of DEL, a value of $P_{02} = 0.091\ s\cdot L^{-1}$ was used;
– P_{26} and P_{27} – the transfer from the Ciorogârla river to aquatic animals, known as bioaccumulation;
– P_{69} – the human ingestion of aquatic animals – parameter connecting the ingestion dose due to fish consumption [$Sv\cdot a^{-1}\cdot Bq^{-1}\cdot kg$];
– $P(i)_{29}$ – the ingestion dose due to drinking water consumption [$Sv\cdot a^{-1}\cdot Bq^{-1}\cdot kg$].

4. DERIVED EMISSION LIMITS AND DISCUSSIONS

The ratio $X_9/X_0 = f_{ik}$ is used for the calculation of DEL – (Q_{ik}) for a certain radionuclide and critical group.

$f_{i,k}$ – is the maximum annual dose for a person from the critical group, due to the release of one Becquerel from the radionuclide *i*-th on the evacuation way (atmosphere/liquid);

A. The transfer parameters for gaseous releases are used for the calculation of the f_{ik} according to the formula:

$$f_{ik} = \frac{X_9}{X_0} = P_{01}\left(P_{e19} + P_{i19} + P_{13}P_{39} + P_{14}P_{49} + P_{15}P_{59} + P_{13}P_{34}P_{49} + P_{14}P_{45}P_{59} + P_{13}P_{34}P_{45}P_{59}\right) \quad [\mathrm{Sv\cdot a^{-1}Bq^{-1}\cdot s}] \tag{5}$$

where: X_9 is the dose rate for an individual (Compartment 9); X_0 is the evacuation rate from the source (Compartment 0); DEL for gaseous releases and respectively liquid discharges is:

$$Q_{ik}[\mathrm{Bq}\cdot\mathrm{a}^{-1}] = \frac{E_{constr\ ik}[\mathrm{Sv}\cdot\mathrm{a}^{-1}]}{f_{ik}[\mathrm{Sv}\cdot\mathrm{a}^{-1}\cdot\mathrm{Bq}^{-1}\cdot\mathrm{s}]}\cdot 365\cdot 24\cdot 3600, \tag{6}$$

where: $Q_{i,k}$ is the DEL, in Bq·a^{-1} for radionuclide *i*-th and gaseous release way; $E_{constr\ ik}$ is the value of annual dose constraint, for radionuclide *i*-th on the evacuation way *k*-th (atmosphere/liquid).

Table 2

The values of the derived emission limits – Q_{ik} – for the effective dose constraint corresponding to the radionuclides released in atmosphere

Emission way (*k*)	**Radionuclide**	**$E_{constr\ ik}$ [Sv·a^{-1}]**	**f_{ik} [Sv·a^{-1}·Bq^{-1}·s]**		**$Q_{i,k}$ [Bq·a^{-1}]**	
			infant	**adult**	**infant**	**adult**
Atmosphere	Co-60	7.391E-06	3.45E-07	3.17E-07	6.75E+08	7.36E+08
	Cs-134	0.015E-06	3.37E-07	3.74E-07	1.40E+06	1.27E+06
	Cs-137	1.200E-06	5.25E-07	7.51E-07	7.21E+07	5.04E+07
	Ag-108m	0.060E-06	7.40E-08	1.07E-07	2.56E+07	1.77E+07
	Eu-152	0.040E-06	6.25E-08	1.20E-07	2.02E+07	1.05E+07
	Eu-154	0.864E-06	9.05E-08	5.86E-08	3.01E+08	4.65E+08
	Eu-155	0.230E-06	9.05E-08	5.86E-08	8.02E+07	1.24E+08
	H-3	0.200E-06	8.41E-12	7.21E-10	7.50E+11	8.74E+09

Taking into account the different values of limits for the same radionuclide released in atmosphere corresponding to different age categories, the lowest (conservative) values of Q_{ik} were proposed Table 3.

Table 3

DEL values for the radionuclides released in atmosphere

Emission way (*k*)	**Radionuclide**	**Q_{ik} [Bq·a^{-1}]**
Atmosphere	Co-60	6.75E+08
	Cs-134	1.27E+06
	Cs-137	5.04E+07
	Ag-108m	1.77E+07
	Eu-152	1.05E+07
	Eu-154	3.01E+08
	Eu-155	8.02E+07
	H-3	8.74E+09

B. For the calculation of the DEL – Q_{ik} – for the liquid effluents, radionuclide i-th and the critical group, one uses the ratio X_9/X_0 =(f_{ik}), where f_{ik} is calculated using the transfer parameters for the liquid discharges, according to the formula:

$$\frac{X_9}{X_0} = f_{ik} = P_{02}(P_{e29} + P_{i29} + P_{28}P_{89} + P_{27}P_{79} + P_{26}P_{69} + P_{25}P_{59} + \\ + P_{24}P_{49} + P_{23}P_{39} + P_{24}P_{45}P_{59} + P_{23}P_{34}P_{49} + P_{23}P_{34}P_{45}P_{59}) \quad (7) \\ [\mathrm{Sv \cdot a^{-1} \cdot Bq^{-1} \cdot s}]$$

DEL = (Q_{ik}) is calculated according to (6).

Table 4

The values of DELs – Q_{ik} – for the effective dose constraints corresponding to radionuclides i-th from the liquid effluents

Emission way (k)	**Radionuclide**	$E_{constr\ ik}$ **[Sv·a^{-1}]**	f_{ik} **[Sv·a^{-1}·Bq^{-1}·s]**		Q_{ik} **[Bq·a^{-1}]**	
			infant	**adult**	**infant**	**adult**
Liquid	I-123	9.00E-07	2.68E-07	3.22E-08	1.06E+08	8.82E+08
	I-125	9.00E-07	1.56E-06	9.56E-07	1.82E+07	2.97E+07
	I-131	9.00E-07	3.57E-05	1.07E-05	7.95E+05	2.66E+06
	Cs-134	9.00E-07	4.60E-05	2.20E-04	6.16E+05	1.29E+05
	Cs-137	7.56E-06	1.08E-04	2.38E-04	2.21E+06	1.00E+06
	Am-241	1.26E-05	8.74E-05	1.37E-04	4.55E+06	2.91E+06
	Co-60	7.20E-06	6.72E-05	4.64E-05	3.38E+06	4.89E+06
	Ir-192	1.80E-06	2.78E-07	5.98E-07	2.04E+08	9.49E+07
	Eu-152	9.00E-07	1.07E-04	1.04E-04	2.66E+05	2.73E+05
	H-3 total	4.23E-05	2.49E-08	5.42E-08	5.36E+10	2.46E+10
	Lu-177	1.80E-06	1.06E-07	5.20E-08	5.33E+08	1.09E+09
	Tc-99m	1.35E-06	3.55E-09	1.43E-09	1.20E+10	2.98E+10
	Re-188	1.80E-06	3.00E-07	8.92E-08	1.89E+08	6.37E+08
	C-14	1.80E-06	4.37E-08	3.69E-08	1.30E+09	1.54E+09
	Ga-68	9.00E-07	1.83E-08	6.37E-09	1.55E+09	4.46E+09
	U-235	9.00E-08	1.25E-04	1.66E-04	2.27E+04	1.71E+04
	U-238	1.80E-06	8.59E-05	1.23E-04	6.61E+05	4.63E+05
	Mo-99	1.80E-06	6.25E-09	4.52E-08	9.09E+09	1.26E+09
	Zn-65	9.00E-07	9.36E-07	1.01E-06	3.03E+07	2.81E+07
	Na-24	9.00E-07	1.56E-08	6.03E-07	1.82E+09	4.70E+07
	Co-58	9.00E-07	4.91E-07	6.15E-07	5.78E+07	4.61E+07

Table 5

Proposed DEL values, corresponding to the radionuclides *i*-th, discharged in the Ciorogârla river

Radionuclide	$Q_{i\ water}$**(Bq/a)**	**DEL(Bq/week)**
0	**1**	**2**
I-123	1.06E+08	1.92E+06
I-125	1.82E+07	3.31E+05
I-131	7.95E+05	1.44E+04
Cs-134	1.29E+05	2.35E+03
Cs-137	1.00E+06	1.82E+04
Am-241	2.91E+06	5.29E+04
Co-60	3.38E+06	6.13E+04
Ir-192	9.49E+07	1.73E+06
Eu-152	2.66E+05	4.83E+03
H-3 total	2.46E+10	4.46E+08
Lu-177	5.33E+08	9.69E+06
Tc-99m	1.20E+10	2.17E+08
Re-188	1.89E+08	3.44E+06
C-14	1.30E+09	2.37E+07
Ga-68	1.55E+09	2.83E+07
U-235	1.71E+04	3.12E+02
U-238	4.63E+05	8.40E+03
Mo-99	1.26E+09	2.29E+07
Zn-65	2.81E+07	5.10E+05
Na-24	4.70E+07	8.54E+05
Co-58	4.61E+07	8.38E+05

5. CONCLUSIONS

Studies on emission derived limits for potential radionuclides contained in gaseous and liquid effluents were made in order to assess the impact of IFIN-HH nuclear activities on the environment and the population from the critical group.

A value of E_{constr} = 100 μSv·a^{-1}, approved by CNCAN was taken as a basis for the calculations.

Based on the experimental data on the emissions produced as a result of the activities carried out in IFIN-HH, the effective exposition dose for the persons in the critical group was much lower than the values approved by CNCAN for the last five years [10–12].

The economic activities carried out by IFIN-HH and the other institutes and/or companies situated within the Măgurele town and its surroundings, as well as the agricultural and industrial activities upstream of the sampling points, have comparative and very low annual contributions to the radiological impact. Their radiological influence is comparable to the level of radioactivity for environmental factors in the IFN-HH influence area.

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