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THE IMPACT ASSESSMENT OF THE ABANDONED URANIUM MINING EXPLOITATIONS ON ROCKS AND SOILS - ZIMBRU PERIMETER, ARAD COUNTY

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Abstract: The mining exploration and exploitation, especially the activity of uranium mineralization exploration and exploitation has a negative impact on the environment by the alterations of the landscape and the degradation of the environmental factors' quality. The principal environmental factors that could be affected by mining operations resulting from uranium exploitation are: water, air, soil, population, fauna, and flora. The aim of this study is, first, to identify the sources of pollution (natural radionuclides - natural radioactive series of uranium, radium, thorium, potassium and heavy metals that are accompanying the mineralizations) for two of the most important environmental factors: rocks and soils: and, second, to assess the pollution impact on those two environmental factors. In order to identify this pollutants and their impact assessment it was selected as a study case an abandoned uranium mining perimeter named the Zimbru perimeter located in Arad County, Romania.

Keywords: *environmental factors, heavy metals, natural radionuclides, pollution sources, rocks and soil pollution*

INTRODUCTION

Overall, the exploitation and exploration activities of the uranium mineralizations and heavy metals have a short and long term impact on the environment by the degradation of the landscape and especially by the deterioration of the environmental factors quality [1].
Inside all types of rocks and soils within the uranium mining perimeters can be found the radionuclides belonging, mainly, to the natural radioactive series of U^{238}, U^{235}, Ra^{226}, Th^{232} and K^{40}.
The radionuclides distribution is not uniformly, there are some areas on Terra, where there are in great concentrations, as well as some sort of rocks in which uranium and thorium are more abundant. In our country such areas are the perimeters of the mines for exploitation and exploration of uranium ores, and the nearby territories [2].

MATERIALS AND METHODS

The mining exploitation chosen, respectively the abandoned prospecting/exploration of uranium ore perimeter (Zimbru) is located at 15 km north of Gurahonț town, in the Codru Moma Mountains [3]. This area belongs to the Zimbru village, Buteni township, Arad County (Figure 1).

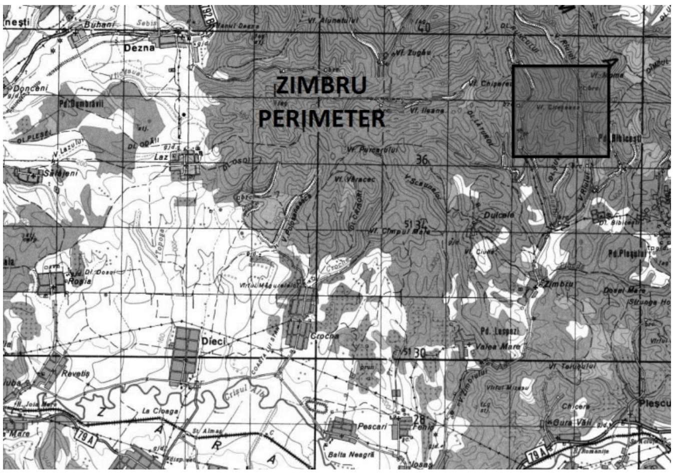

Figure 1. *The location of the Zimbru uranium mining area, Arad County*

In 1945, the first geological works were made: complex works of exploration and preliminary exploration aiming the minerals resources identification, continued in 1953 by the Sovrom - Kvartal Company for the radioactive ores identification [4].
Between 1960 - 1962 the copper mineralization was explored (G1 Gallery located on the Luştrilor Valley) and it was observed that the mineralization is sometimes accompanied by uranium [5].
Thus, during 1962 - 1967, the entire area was explored through underground drilling, through the execution of an inclined plane from G1 gallery, through the achievement of a new gallery G4 and through a 1:200 radiometric mapping, finally highlighting a new cooper and uranium mineralization located mineralization located on Socilor Valley. In

1969 a complex research begins through underground workings and drilling, most significant of them was G2 gallery located at 90 m North of G1 gallery.
The positive results have imposed the opening of the Shaft (401.8 m) on the right side of the Luştrilor Valley.
The exploration works performed within the Zimbru perimeter have lead to the shaping of some copper and uranium ore bodies.
In 1978 a part of the mining works entered the conservation (The Shaft and the G2 gallery), but, in the present, the entire activity is suspended [6].
The town located nearest the mining works is the Zimbru village, which is situated at a distance of only 3.8 km.
The most important of the Zimbru perimeter mining works (Figure 2) are represented by: The Shaft (+401.8 m), G2 gallery (+377.8 m), G1 gallery (+378.3 m) + Inclined Plane (+384 m); GIII gallery (+372 m) and G4 Soci gallery (+444.5 m) [3].
Inside the Zimbru perimeter the pollution sources are represented by the dumps, the gallery outputs and the mine waters.

- The Shaft is located on the Luştrilor creek at an altitude of 401.8 m, situated on the right slope. Currently the well is closed, but the mine water discharges an important flow, running down the slope and reaches the creek. Dump has the following characteristics: length 140 m; height 22 m; surface approx. 6700 m^2.
- The G2 Gallery is located on the Luştrilor creek at an altitude of 377.8 m, situated on the left slope. The gallery's output is collapsed and the mine water it flows from the output, running down the slope and reach the creek. Dump has the following characteristics: length 20 m; height 5 m, surface 150 m^2.
- The G1 Gallery (378.3 m) and the Inclined Plane (384 m) are located on the left slope of the Luştrilor stream, in this area five outputs were identified - A, B, C, D and E (all of them are collapsed). Dump has the following characteristics: length 50 m; height 2 - 6 m, surface approx. 800 m^2.
- The GIII Gallery is located at an altitude of 372 m, on the right slope of the Luştrilor stream, the dump base is at approx. 20 m from the forest road. Dump has the following characteristics: lenght 6 - 7 m; height max. 5 m: surface approx. 170 m^2.
- The G4 Soci Gallery is located at an altitude of 444.5 m, on the left slope of the Soci stream. Dump has the following characteristics: lenght 12 m; height max. 10 m; surface approx. 330 m^2 (Figure 3).

***Figure 2**. The Zimbru perimeter - the locations of major mining works*

***Figure 3.** G4 Soci Gallery dump*

This study focuses on the mining works' influence on the environmental factors such as soils and rocks from the Zimbru perimeter.
Thus, from the three listed pollution sources, the waste dumps are the main potential pollution source for soils and rocks through the processes of leaching and migration, due to the flowing of the rainwater and to the aeolian transport of the dust.
The pollutants of interest are represented by the natural uranium, radium and heavy metals.
The analysis methods that were used are:

- Gamma spectrometry, using the ORTEC multichannel analyzer - 2001 with the hyperpure Ge detector; calibrated and verified by the Laboratory of the Radiation Metrology, Tests and Dosimetry Department - IFIN HH, for the determination of radium, thorium, potassium.
- Beta-gamma global method with the 4-channel analyzer, for the determination of uranium
- Flame atomic absorption spectrometry VARIAN Spectra 220 - compact instrument of absorption and atomic emission, for the determination of heavy metals.

To quantify the sources of pollution, measurements of the gamma dose rate were conducted and also, samplings of the material were collected (Table 1). The samples were taken in order to analyze the radioactive elements and the metals that accompanying the mineralization.

Table 1. *The measurements of the gamma dose rate for quantify the sources of pollution*

Gallery	**Dump area [m^2]**	**Maximum concentrations**	**The dose rate surface [Sv·h^{-1}]**
The Shaft – 401.8 [m]	6700	- U = 23.15 [mg·kg^{-1}] - Ra^{226} = 0.082 [Bq·g^{-1}] - Th^{232} = 6.25 [mg·kg^{-1}] - K^{40} = 1.66 [%]	50
G2 – 378 [m]	150	- U = 20.34 [mg·kg^{-1}] - Ra^{226} = 0.031 [Bq·g^{-1}] - Th^{232} = 4.28 [mg·kg^{-1}] - K^{40} = 1.51 [%]	-
G1 + Inclined Plane (5 outputs)	800	- U = 22.42 [mg·kg^{-1}] - Ra^{226} = 0.174 [Bq·g^{-1}] - Th^{232} = 12.18 [mg·kg^{-1}] - K^{40} = 1.78 [%]	-
G III – 372 [m]	170	- U = 22.77 [mg·kg^{-1}] - Ra^{226} = 0.027 [Bq·g^{-1}] - Th^{232} = 13.96 [mg·kg^{-1}] - K^{40} = 3.58 [%]	-
G4 Soci – 444 [m]	330	- U = 27.63 [mg·kg^{-1}] - Ra^{226} = 0.064 [Bq·g^{-1}] - Th^{232} = 12.96 [mg·kg^{-1}] - K^{40} = 4.30 [%]	-
Regulations	-	- U = 16 [mg·kg^{-1}] - Ra = 0.2 [Bq·g^{-1}]	0.3 – Environmental Reconstruction Limit

The rock samples were collected from the waste dump (R6, R7, R8, R9, R10, R11, R12) in order to determine its content of heavy metals and radioactive material.
The soil samples were thus collected: seven samples from the waste dumps (S17, S19, S20, S22, S23, S26, S28), the place where they were constituted and are considered less sensitive areas and eight samples were also collected around the dumps (S16, S18, S21, S24, S25, S27, S29, S30), especially at their base (areas affected by rain water flowing) and which are considered sensitive use areas (Table 2).

Table 2. *The samples taken in order to analyze the radioactive elements and the metals that accompanying the mineralization*

Sample	**Sampling place**	**Sample**	**Sampling place**
R6	The Shaft Dump	S20	The G2 Dump
R7	The Shaft Dump	S21	The G2 Dump base
R8	The G2 Dump	S22	„5 outputs" Dump – A Output
R9	The G1 + Inclined Plane Dump	S23	„5 outputs" Dump – E Output
R10	The G1 + Inclined Plane Dump	S24	„5 outputs" Dump base – A Output
R11	The GIII Dump	S25	„5 outputs" Dump base – E Output
R12	The G4 Soci Dump	S26	The GIII Dump
S16	The Shaft Dump Slope (background)	S27	The GIII Dump base
S17	The Shaft Dump road	S28	The G4 Soci Dump
S18	The Shaft Dump base	S29	The G4 Soci Dump slope (background)
S19	The Shaft base Dump road	S30	Zimbru village – first house area

RESULTS AND DISCUSSIONS

For the 7 collected rock samples, the radioactive elements (U, Ra^{226}, Th^{232}, K^{40}) and the heavy metals accompanying the mineralizations (Cu, Co, Pb, Zn, Ni, Sn, Sb, V, As, Mo) were analyzed (Table 2).
The benchmarks used for the radioactive elements are represented by the Radiological Safety Fundamental Norms – 2002, which include the concept of exclusion level that limits the values of the materials covered by the Romanian National Commission for Nuclear Activities Control (U = 16 ppm; $\sum(Ra^{226} + Th^{232}) < 0.2\ Bq \cdot g^{-1}$) [7, 8].
The values higher than those presented in Table 3 illustrate that the material of the dump is framed as radioactive waste.
For heavy metals are not set limits.
For the 7 soil samples collected from the dumps and the 8 soil samples collected around the dumps, the radioactive elements (U, Ra^{226}, Th^{232}, K^{40}) and the heavy metals accompanying the mineralizations (Cu, Co, Pb, Zn Ni, Sn, Sb, V, As, Mo) were analyzed (Table 4 and Table 5).

Table 3. *The results of the rock samples analysis (Sn, Sb, V – under detection limit, detection limit = 0.0001 %)*

Sample	Collecting place	Radioactive elements – maximum concentrations				Metals [mg·kg^{-1}]						
		U [mg·kg^{-1}]	Ra^{226} [Bq·g^{-1}]	Th^{232} [mg·kg^{-1}]	K^{40} [%]	Cu	Co	Pb	Zn	Ni	As	Mo
R6	The Shaft Dump	19.76	0.082	6.25	1.66	126	3	6	79	36	11.9	1.51
R7	The Shaft Dump	23.15	0.076	5.75	1.22	95	5	10	195	42	12.5	1.2
R8	The G2 Dump	20.34	0.031	4.28	1.51	405	27	152	266	32	12.1	2.5
R9	The G1 + Inclined Plane Dump	17.96	0.174	12.18	1.78	4000	13	25	135	21	11.8	2
R10	The G1 + Inclined Plane Dump	22.42	0.075	10.14	1.72	570	19	18	152	27	9.5	1.85
R11	The GIII Dump	22.77	0.027	13.96	3.58	82	16	50	246	51	8.7	1.56
R12	The G4 Soci Dump	27.63	0.064	12.96	4.3	42	17	135	305	68	10	1.16
Exclusion level		16	$\sum(Ra^{226} + Th^{232}) < 0.2$ [Bq·g^{-1}]		-	-	-	-	-	-	-	-

Table 4. *The results of the soils samples collected from the dumps analysis (Sn, Sb, V – under detection limit, detection limit = 0.0001%)*

Sample	Collecting place	Radioactive elements –maximum concentrations				Metals [mg·kg^{-1}]						
		U [mg·kg^{-1}]	Ra^{226} [Bq·g^{-1}]	Th^{232} [mg·kg^{-1}]	K^{40} [%]	Cu	Co	Pb	Zn	Ni	As	Mo
S17	The Shaft Dump road	14.73	0.119	4.03	0.89	192	11	15	154	28	7.5	1.2
S19	The Shaft base Dump road	9.25	0.042	5.76	1.25	172	14	10	83	41	8.2	1.26
S20	The G2 Dump	12.14	0.05	8.25	2.12	450	18	20	302	27	10.12	2.02
S22	„5 outputs” Dump – A Output	13.82	0.031	6.18	2.25	826	6	25	172	21	7.25	2.14
S23	„5 outputs” Dump – E Output	12.2	0.062	4.08	1.72	720	14	14	142	55	9.42	1.75
S26	The GIII Dump	14.23	0.045	6.22	1.25	SLD	7	22	170	23	10.11	1.5
S28	The G4 Soci Dump	17.32	0.092	4.72	2.03	21	12	12	122	44	8.22	1.25
756/97 Order – Alert limit		-	-	-	-	250	100	250	700	200	25	15
756/97 Order– Intervention limit		-	-	-	-	500	250	1000	1500	500	50	40
Soil normal values		10	0.125	18	-							

Table 5. *The results of the soils samples collected around the dumps analysis (Sn, Sb, V – under detection limit, detection limit = 0.0001%)*

Sample	**Collecting place**	**Radioactive elements –maximum concentrations**				**Metals [mg·kg^{-1}]**						
		U [mg·kg^{-1}]	**Ra^{226} [Bq·g^{-1}]**	**Th^{232} [mg·kg^{-1}]**	**K^{40} [%]**	**Cu**	**Co**	**Pb**	**Zn**	**Ni**	**As**	**Mo**
S16	The Shaft Dump Slope (background)	9.5	0.045	5.2	1.7	10	10	8	30	26	SLD	0.75
S18	The Shaft Dump base	15.16	0.053	6.18	1.83	22	12	7	29	71	3.7	1.14
S21	The G2 Dump base	22.42	0.062	4.64	1.51	252	8	135	180	36	2.5	2.1
S24	„5 outputs" Dump base – A Output	12.12	0.042	3.75	2.17	438	7	95	63	38	4.5	1.1
S25	„5 outputs" Dump base – E Output	10.14	0.037	5.21	4.12	275	2	142	95	42	7.9	0.8
S27	The GIII Dump base	18.32	0.062	4.14	1.75	25	5	17	72	27	8.3	0.85
S29	The G4 Soci Dump slope (background)	13.2	0.042	7.2	2.02	17	3	22	48	30	SLD	1.28
S30	Zimbru village – first house area	11.25	0.035	6.21	1.21	11	7	12	50	35	SLD	1.14
756/97 Order – Alert limit		-	-	-	-	100	30	50	300	75	15	5
756/97 Order– Intervention limit		-	-	-	-	200	50	100	600	150	25	10
Soil normal values		10	0.125	18	-	-	-	-	-	-	-	-

For radioactive elements, the reference values of the area will be used: U = 4 - 6 ppm; Ra^{226} = 0.025 - 0.035 Bq·g^{-1}; Th^{232} = 2 - 5 ppm.
For heavy metals, the concentrations were compared to the values of the environmental legislation in force [9].
The soil samples collected from dumps were compared with "less sensitive" corresponding values and the soil samples collected around dumps were compared with "sensible use" corresponding values.
The uranium mining works can affect the environmental factors in a direct mode (water, air, soil, population - by stationing in contaminated areas, fauna) and in an indirectly mode (population, flora and fauna) [10].
The results of the analyzes and interpretations for the rock and soil samples collected from dumps and their surroundings, have highlighted the following issues:

- On all the dumps waste were recorded values for uranium at normal level or slightly above the exclusion limit;
- Non-radioactive elements from the analyzed rock samples have normal values of distribution (Table 3);
- The copper presence was found in high concentrations in the samples collected from the waste dumps of the G2 gallery and G1 gallery + inclined plane;

- Generally all soils formed on the dumps have the uranium concentration slightly above normal, but Ra^{226}, K^{40} and Th^{232} are within background limits (Table 4);
- On the dumps of the G2 gallery and the G1 gallery ("5 outputs" area) were recorded copper concentrations situated above the intervention limit;
- All metals analyzed: Cu, Cd, Pb, Zn, Ni, Sn, Sb, V, As, Mo have concentrations under the alert limit (756/97 Order - less sensitive use);
- The soils samples collected at the dumps' base of the Shaft, G2 gallery and GIII gallery, have uranium concentrations 1.5 - 2 more times than the area background (Table 5);
- The soils samples collected at the dumps' base of the G2 gallery and "5 outputs" have copper concentrations of up to 2 times above the intervention limit;
- Rest of metals analysed: Cu, Cd, Pb, Zn, Ni, Sn, Sb, V, As, Mo have concentrations - for all samples - under the alert limit (756/97 Order – sensitive use).

CONCLUSIONS

Although it is characterized by a small dose rate, the irradiation produced by the incorporated natural radionuclides can lead to a great irradiation dose and, consequently, to the appearance of some serious, irreversible lesions, due to the long period of exposure and the emitted alpha radiations. The clinical signs are often not evident long time after the human body contamination, because of the small dose rate, but the noxious effects are accumulated if the human organism is not able to repair rapidly the produced damage [2].

The main purpose of protective measures is to prevent the dispersion of uranium in the environment, thus preventing a further population irradiation [11, 12].

As noted, some existing mining works are sources of pollution of the environment.

Concluding, the analyzes, the measurements and the interpretations have found that:

Soils formed on the dumps have uranium concentrations slightly above normal in soil and copper concentrations above the intervention limit (G2 Gallery and "5 outputs")

Soils formed at the dumps' base have uranium concentrations of 1.5 - 2 more times than the area background (The Shaft, G2 Gallery and GIII Gallery) and concentrations of up to 2 times above intervention limit (G2 Zimbru Gallery and "5 outputs")

For environmental remediation and eliminating the people's irradiation risk it is recommended for the material of the dumps which exceeds the ecological restoration limit (respectively The Shaft dump) finding of remedial solutions (relocation, coverage) [13].

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