Radiation exposure and environmental remediation at the Urgeiriça mine site, Portugal

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Abstract. The Urgeiriça mine and milling facilities, Portugal, was in operation until 2001. Site remediation, including clean-up of milling plant facilities and terrains was implemented during 2006-8 and it was completed with the placement of a multilayer cap on milling tailings and construction of a drainage system for retrieval of mine water and waste piles seepage followed by neutralization in a mine water treatment plant. During this period the monitoring of environmental radioactivity was periodically carried out in the Urgeiriça area. Results of environmental surveys carried out during last 10 years on ambient radiation dose, radon in surface air, radionuclide concentrations in soils, agriculture products, and water from wells and from surface streams receiving mine effluents, are reviewed. Results of this environmental survey are presented and radiation exposure of the local population is discussed.

Introduction

The uranium mining and milling industry in Portugal was productive during most of the 20th century. Facilities at Urgeiriça mine, near Viseu, were the main milling site of radioactive ores and accumulated about 13 Mtonnes of tailings from chemical processing (Nero et al., 2005). The mining company ENU-SA ceased activities in 2001 and, following that decision, the government requested from national laboratories a major assessment of the environmental and public health risks posed by uranium mining and milling wastes in the region. The reports from such assessment (MinUrar project) were conclusive on the existence of a radioactive impact in the surrounding environment and public health and recommended the implementation of waste management and environmental remediation measures (Marinho Falcão et al., 2005, 2007). This work was entrusted to the EDM company under the Ministry of Economy, and the Director Plan for remediation of abandoned mine sites was submitted and approved by an inter-ministerial commission

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with representatives of Health, Economy, Environment, and Radiation Protection authorities (Santiago Baptista, 2005). The remediation work in the Urgeiriça area was implemented between 2006 and 2008, and consisted in cleanup of mine area and uranium chemical plant with relocation and grouping contaminated waste into the former milling tailings dump “Barragem Velha”.

The coverage of milling tailings and retrieval of drainage followed by treatment of contaminated water, restrained the dispersion routes of waste and radionuclides in surface air (radon and dust), of discharge of radioactive and acid drainage in surface streams, shielded the radioactive materials responsible for enhanced radiation doses in the area, and prevented the potential misuse of radioactive materials (sands, gravel, scrape metals, woods, etc) in the future (EDM, 2011).

Radiological surveys were made over the years in this and other old uranium mining areas, as part of the duties of LPSR/IST (Carvalho, 2014). An assessment of the environmental radioactivity was made in 2008 in Urgeiriça area, after the cover of milling tailings had been concluded. This synoptic monitoring of the status of the environment is described for 2008. Temporal trends of radioactive contamination over the years are presented also and discussed.

Materials and Methods

Ambient radiation doses were determined with calibrated portable radiation monitors (Thermos, FH-40). Samples of water from irrigation wells, soil from the agriculture plots, and plants from the available horticulture production were collected on June 2008. Water samples were filtered through 142 mm diameter, 0.45 µm pore size membrane filters, and filtered water and suspended particulate matter were analyzed separately. Soil bulk samples of the top 30 cm layer were collected and sieved in the laboratory. The size fraction less than 63 µm was used for radioanalysis. Vegetables were cleaned and washed with tap water in the laboratory and tubers and fruits peeled off, as for consumption. Samples were freeze dried and aliquots of homogenized material taken for determination of radionuclides. Radon in surface air was determined with continuous radon monitors (Sarad GmbH, Scout). Surface air aerosols were sampled with high volume aerosol samplers (Andersen).

Radionuclides were determined in all samples applying radiochemical separation of radionuclides, followed by electro deposition and radiation measurement by alpha spectrometry. The analytical procedures applied and their validation through the analysis of certified reference materials and participation in international inter comparison exercises organized by the IAEA, are described in detail elsewhere (Carvalho et al., 2005, 2009a, 2009b; Carvalho and Oliveira 2007, 2009; Oliveira and Carvalho 2006).

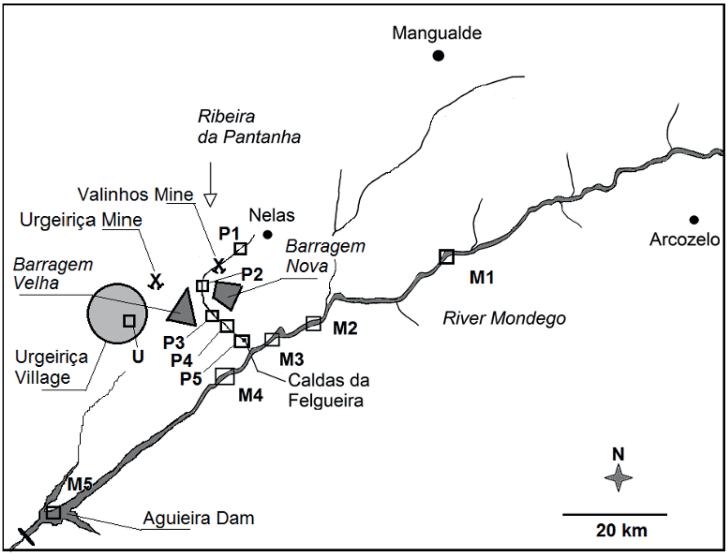

Fig.1. The region of Urgeiriça Mine and uranium milling tailings. Sampling stations are indicated with squares.

Results and Discussion

Ambient dose

Ambient radiation doses measured in 2002 on top of milling tailings "Barragem Velha" ranged from 2 to 12 μSv/h, averaging 7.5 μSv/h. These dose rates varied depending on the composition and grain size of materials disposed in the self-contained impoundments spread over a total area of about 10 hectares. The relocation of waste with transfer of contaminated materials from the mine shaft area and chemical plant area to the waste dump site "Barragem Velha", plus transfer of materials from the cleanup of industrial area onto the top and, finally, placement of additional layers of coverage materials (geotextile membrane, clay, sand, gravel, soil), allowed reducing ambient radiation dose on waste piles was to 0.35 μSv/h, i.e, about 5% of initial dose rate (EDM, 2011). Although this area remains fenced and not open to the public, the current external absorbed radiation dose in the air is of the same magnitude and even slightly lower than the regional background, 0.4-0.5 μSv/h.

Radionuclides in water

The survey of radionuclides in the stream Ribeira da Pantanha (performed in 2008), showed still high concentrations in the acidic seepage from waste piles particularly loaded with dissolved uranium, ^{230}Th and ^{226}Ra. Despite recuperation of tailings seepage and mine drainage into a water treatment plant for acid neutralization and co precipitation of radionuclides with barium sulphate, the enhancement of dissolved uranium and radium in stream water was still measurable downstream the area of waste piles. Concentrations of these radionuclides in stream water decreased downstream with increasing distance due to radionuclide sorption onto sediments and water dilution. Nevertheless, the discharge of Ribeira da Pantanha into the major River Mondego was still contributing to enhance radionuclide concentrations, particularly those of radium, above natural concentrations in this river (Table 1). Further downstream radionuclide concentrations decreased to natural levels with large dilution in the artificial lake of Aguieira Dam.

Bottom sediments of Ribeira da Pantanha stream and River Mondego showed higher radionuclide concentrations and stronger contamination downstream Urgeiriça and waste disposal areas. As sediments are more conservative of radionuclides, the concentrations in bottom sediments will remain above natural levels longer than in the flowing water (Table 2). Notwithstanding, determinations of radionuclides in suspended particulate matter and in bottom sediments showed a decrease in comparison with determinations made several years before remediation measures (Carvalho et al., 2007).

Radionuclides in vegetables

On the banks of stream Ribeira Pantanha as well as in the town of Canas de Senhorim-Urgeiriça area, there are small agriculture productions and kitchen gardens whose products are irrigated with water from wells and from Pantanha stream (Table 3). Agriculture plot # 22, in the Village Aguieira near Nelas, irrigated from a non-contaminated well is a reference area. Vegetables from plots #19-#22 located on the banks of Ribeira da Pantanha and regularly irrigated with stream water, generally showed higher radionuclide concentrations than at the reference area.

Atmospheric radioactivity

Radon in surface air was measured over the years in several locations around the Barragem Velha waste pile. Radon emanated from the uncovered waste piles was transported by the wind in any direction and rapidly diluted in the atmosphere but continuous recording averaged those effects. Measurements were made in three locations, by the nearest houses to tailings piles and at the center of the town Canas de Senhorim. Radon values in surface air at some distance from the waste piles before the placement of the tailings cover averaged 80-100 Bq/m^3, slightly higher than after placement of the cover averaging 55-65 Bq/m^3 on late 2008.

Long lived uranium series radionuclides attached to dust particles in suspension in surface air were measured also at several locations around the main waste pile Barragem Velha. U-238 concentrations in air dust were for example in the year 2006 from 200 to 400 Bq/kg, while in 2010 ranged from 70 to 190 Bq/kg. U-238 activity concentrations in atmospheric dust are an indicator of the resuspension of tailings, while ^{210}Pb and ^{210}Po in dust particles are the sum of radionuclide in minerals of tailings particles plus radon daughters scavenged in the atmosphere. Results over the years displayed fluctuations in specific activity of dust particles which indicates particles from different sources (soil particles, pollen, milling tailings particles, etc.), but a clear decrease of radioactivity in surface air occurred since the placement of the tailings cover, dropping to about half radioactivity levels of aerosols in surface air.

Compaction and coverage of waste materials reduced both the emanation of radon (^{222}Rn) into the surface air and dust re suspension, particularly during summer.

Conclusions

An abatement of ambient radiation dose from gamma radiation emitting radionuclides was obtained with the cleanup of the industrial area, relocation of residues into the “Barragem Velha” milling tailings, and placement of a multilayer cover. Cleaned areas, through removal of waste, debris, scrap materials, and top soil layer have now ambient radiation doses aligned to natural background radiation levels. These areas were opened to other uses such as construction of a new residential area, a public garden, and new roads. Some former industrial and administration buildings were released also by the company to other users and uses. The improvement of radiation protection in the area of these former uranium mining and milling activities was considerable, with radiation exposure of the members of the public being reduced, contaminated materials confined, erosion and leaching of radionuclides brought under control, and environment contamination risk and public health risk controlled, such as recommended by international standards (IAEA, 2006).

The challenge might be now the transfer of custody, maintenance, and monitoring of the remediated sites over future years in order to ensure continued radiation protection and confinement of residues.

Table 1. Activity concentrations of dissolved radionuclides in samples from surface waters (mBq/L).

Sample	Station Nr	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Stream Rib.ª da Pantanha	P1	81.7±2.9	3.4±0.3	82.2±3.0	2.3±0.2	33.3±2.6	11.8±0.7	3.6±0.2	0.15±0.05
Run off, B. Velha	P2	48867±2642	2387±157	47243±2556	565±41	364±49	44.1±2.8	87.7±3.6	13.1±0.3
Stream Rib.ª da Pantanha	P3	1885±100	87.2±5.65	1914±101	2.3±0.2	56.5±5.7	12.6±0.5	5.0±0.2	0.11±0.04
Stream Rib.ª da Pantanha	P4	1992±85	91.0±4.6	2009±86	8.7±0.5	59.3±3.7	7.7±0.3	4.3±0.2	0.34±0.07
Rib.ª da Pantanha, C. Felg	P5	298±9	13.8±0.7	310±9	-	69.4±7.7	12.9±0.7	8.6±0.04	-
R. Mondego, P. Cervães#9	M1	22.0±0.6	0.96±0.07	22.4±0.6	4.7±0.4	44.8±5.4	3.8±0.2	5.9±0.2	0.20±0.07
R. Mondego, C. Louça #8	M2	32.6±0.9	1.5±0.1	33.6±0.9	2.9±0.2	18.0±3.0	1.22±0.05	7.0±0.2	0.27±0.06
R. Mondego, upstream C. Felg #6	M3	28.8±0.8	1.4±0.1	29.9±0.8	2.8±0.2	1.4±0.2	3.1±0.1	5.3±0.2	0.21±0.04
R. Mondego, downstream C. Felg #7	M4	20.0±0.6	1.0±0.1	20.2±0.6	3.9±0.3	29.2±5.5	6.3±0.2	5.0±0.2	0.29±0.06
Agueira DAM, "Lagoa Azul" #10	M5	9.2±0.3	0.51±0.06	8.8±0.3	1.5±0.1	17.7±1.9	4.3±0.3	5.3±0.2	0.08±0.04

Table 2. Activity concentrations of radionuclides in riverbed sediments (Bq/kg dw).

Sample		Station Nr	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	$^{210}Pb = ^{210}Po$	^{232}Th
Stream Rib.ª da Pantanha	Sed.#1	P1	2107±47	99±3	2136±48	923±48	403±27	312±13	171±9
Run off B.Velha, Ribª Pantanha	Sed.#2	P2	1109±26	49±2	1200±27	2752±154	2211±140	3366±107	108±7
Stream Rib.ª da Pantanha	Sed.#3	P3	1951±44	91±3	1923±43	484±33	593±48	379±13	276±19
Stream Rib.ª da Pantanha	Sed.#4	P4	4106±102	178±6	4193±104	2892±189	1141±100	1165±37	132±10
Stream Rib.ª da Pantanha	Sed.#5	P5	5422±150	244±9	5584±154	2312±127	1733±77	3736±117	169±10
R. Mondego, P. Cervães	Sed.#9	M1	414±13	20±2	431±13	298±19	366±35	495±12	162±11
R. Mondego, C. Louça	Sed.#8	M2	453±12	19±1	463±12	411±19	674±50	443±11	407±19
R. Mondego. upstream C. Felg	Sed.#6	M3	508±13	25±1	511±13	470±28	534±44	493±18	593±35
R. Mondego. downstream C. Felg	Sed.#7	M4	827±20	39±2	842±20	1302±88	797±70	665±23	440±30
Aguieira Dam	Sed.#10	M5	95±5	3.2±0.7	98±5	96±6	85±8	96±3	50±3

Table 3. Activity concentrations of radionuclides in vegetables from local production (mBq/kg ww).

Sample	Station Nr	Dry/wet weight	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Lettuce, Aguieira Village #22	P1	0.07	197±9	8.1±1.7	164±8	147±15	674±45	470±11	612±22	100±11
Tomato, Aguieira Village #22	P1	0.06	7.8±1.2	< 2.5	5.9±1.2	19.8±4.1	66.4±6.5	17.3±1.2	0.06±0.003	0.42±0.38
Carrot, Aguieira Village #22	P1	0.12	25.0±2.8	< 5.4	27.4±3.0	29.7±4.1	906±59	116±6	57.8±2.7	9.3±2.3
Lettuce, Urgeiriça #21	U	0.04	100±6	5.2±1.2	100±6	113±9	360±24	343±20	157±6	48.6±4.8
Tomato, Urgeiriça #21	U	0.1	1.4±0.5	< 4.0	5.2±1.5	14.8±2.6	53.3±6.9	11.9±0.8	6.8±0.3	0.34±0.17
Cucumber, Urgeiriça #21	U	0.04	2.9±0.3	0.12±0.07	1.6±0.2	22.5±2.4	17.7±1.7	16.4±2.7	18.4±5.4	3.7±1.1
Carrot, Urgeiriça #21	U	0.11	54.9±8.2	< 5.5	65.3±9.5	80.2±8.4	631±46	99.1±3.8	35.2±1.6	30.4±5.1
Lettuce, Urgeiriça #19	P3	0.13	654±24	26.6±3.7	680±25	481±41	1840±107	1770±98	1113±26	227±23
Tomato, Urgeiriça #19	P3	0.09	5.6±1.2	1.1±0.7	3.6±0.9	3.9±0.7	82.1±5.9	20.0±1.4	17±1	0.30±0.12
Corn leaf, Urgeiriça #20	P4	0.16	555±29	25.9±5.1	574±30	355±19	1768±140	970±42	177±6	65.4±5.4
Corn cob, Urgeiriça #20	P4	0.14	4.4±0.4	0.22±0.10	2.0±0.2	3.2±1.0	274±23	32.1±4.4	10.5±0.9	2.4±0.9
Pasture, Urgeiriça #20	P4	0.1	1000±26	44.0±3.4	998±26	807±40	944±52	466±21	199±7	163±10
Turnip greens, C. Felgueiras #23	P5	0.06	1270±41	58.6±5.2	1239±40	430±23	623±32	652.9±34.5	537±17	136±8

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