Contamination of Hydrographic Bassins in Uranium Mining Areas of Portugal

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Abstract. The uranium-radium mining industry generated about six million tons of radioactive by-products composed mainly by the tailings of uranium milling facilities and by fine mud deposits resulting from treatment of acid mine waters. Most of these radioactive materials are deposited on the ground in the village of Urgeiriça, near Viseu, and are currently exposed to weathering. Following rains, leaching and surface runoff of these tailings drain into a stream, Ribeira da Pantanha that joins the river Mondego. In this river a wide artificial lake, Barragem da Aguieira, is the main water reservoir for the centre region of the country. Other rivers flowing through the mining region, namely the rivers Dão, Vouga and Távora, may also receive drainage from the areas of old uranium mines. The radioactive contamination that uranium mining and milling wastes may have originated in rivers and water reservoirs is assessed.

Introduction

The mining of radioactive ores in Portugal begun in 1909 for the production of radium salts exported to France. At that time no recuperation of uranium was made. After 1945, the production of uranium was profitable and became the main aim of radioactive ores mining industry while the production of radium was abandoned. The exploitation of uranium mines and production of U_3O_8 concentrates continued until 2001. During the entire radium-uranium mining cycle that lasted 90 years, about 60 sites with deposits of uraniferous mineralisations were exploited in Hercynian granites. These sites are located in the uranium bearing region of the North-centre of the country in the departments of Guarda, Viseu and Coimbra

(JEN, 1964). The mines were of different size, ranging from vary small to relatively large exploitations, and in total the production of U_3O_8 amounted to about $4x10^6$ tons. Most of the mines were exploited as open pits, and only a few as underground works. In most of the mine sites only barren rocks were left as mining waste. The ore from small mines was transported to one of the main facilities of the mining company and it was milled and extracted there. The main facilities for chemical treatment of radioactive ores were installed at Urgeiriça/Canas de Senhorim, Quinta do Bispo, Senhora dos Remédios and Bica. In a few other places, sulphuric acid was used for *in situ* leaching of uranium and did originate acid mine waters (JEN, 1964; Carvalho et al. 2005).

In the ore processing facilities with milling and chemical treatment plants, there are tailings with solid waste disposed off on the ground. In Urgeiriça there are two main tailing areas: one, known as Barragem Velha (BV), received the sand like materials remaining after the ore milling and uranium extraction, and another, known as Barragem Nova (BN), receiving since years ago the precipitate (radioactive mud) from neutralization of acid mine waters with barium chloride and calcium hydroxide. Two other spoil heaps containing radioactive materials are the "tailings" of Santa Bárbara, actually a heap of low grade ore disposed near the administration buildings of Urgeiriça mine, and the spoil heap at the milling station in the site of the former discharge of uranium ore transported by lorries. The amount of solid mining and milling waste in Urgeiriça is estimated at about 3 million tons.

In the past, seepage and surface runoff from these tailings may have drained directly into the streams. In the present, those are barely collected into a pond for acid neutralization and, after co precipitation of radionuclides and pH adjustment, the supernatant water is released into the Ribeira da Pantanha, a stream tributary to the River Mondego. During the last few years, acid water from the galleries of the underground mine of Urgeiriça, was pumped into this pond for pH neutralization also. In the past, process water from the uranium extraction procedure was released into the Ribeira da Pantanha at this same point. This area of Urgeiriça was selected as a case study (Fig. 1).

The three administrative Departments where the 60 old mining sites are located encompass the catchments areas of several main rivers. These rivers might have received the discharges of mine wastes and, today, may still receive surface runoff and discharges of small tributary creeks flowing across the mine sites. As dispersal of radionuclides from radioactive ores might have occurred in these hydrographical systems, the basins of the rivers Mondego, Dão, Vouga and Távora were selected to investigate enhanced levels of natural radioactivity (Fig. 2).

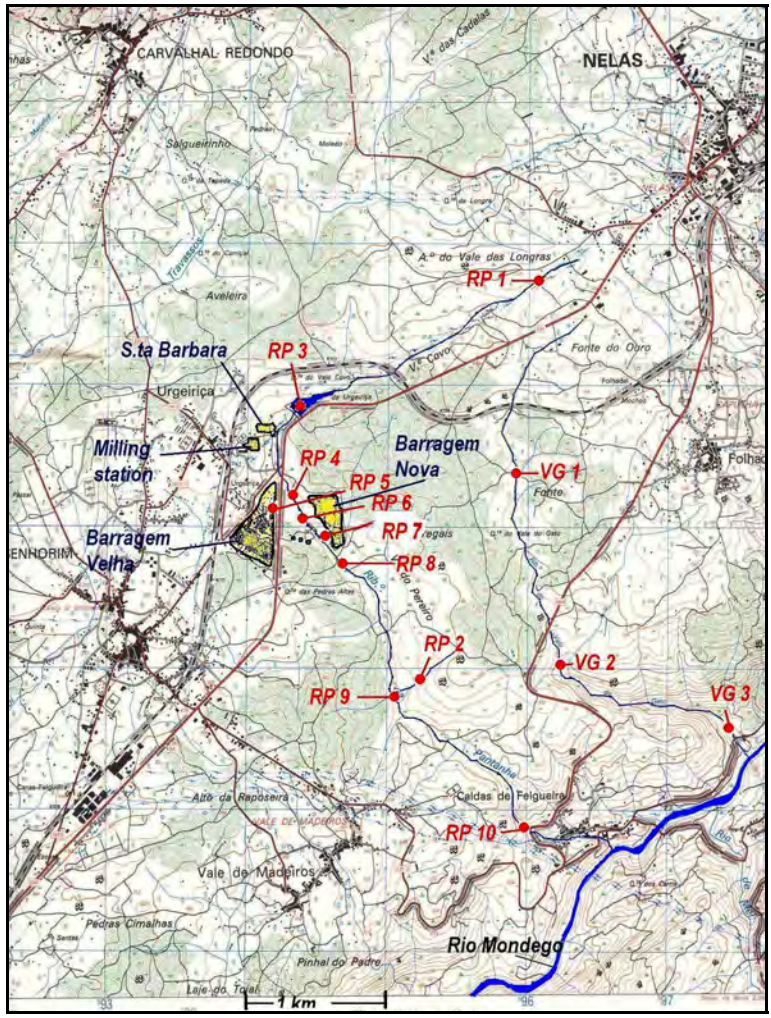

Fig. 1. Map of the region Urgeiriça/Canas de Senhorim/Nelas. The mining and milling facilities and radioactive tailings in Urgeiriça are indicated. Tagged points show sampling sites.

Materials and Methods

Samples of the top layer from soils and tailings were collected with a metal tube sampler. Several samples of the tailings materials were combined for analysis in order to obtain results better representing radionuclide concentrations in these wastes.

Samples of river bed sediments and river water were collected in four to seven stations in each river, from the source in the mountains to the river section in the coastal plain (Fig. 2). The water samples were filtered in the same day, near the collection site, using large diameter (142 mm) 0.45 μm pore size Millipore membrane filters. Sediments and folded filters were transported in plastic bags. In the laboratory, sediments were sieved through a stainless steel metallic screen of 63 μm pore size and only the fraction <63 μm was analysed to allow for inter sample comparisons (Carvalho, 1995). Sediment samples and filters with the suspended matter were dried in the oven, at 60º C, before analysis.

The filtered waters were acidified with HNO_3 to pH<2 and known activities of isotopic tracers ^{232}U, ^{229}Th, ^{209}Po and stable lead were added, to be used as internal tracers for the yield of the radiochemical procedure. Dissolved radionuclides were co precipitated with MnO_2. Solid samples were weighted with analytical precision and tracers added to the sample in a beaker along with mineral acids for complete dissolution of the sample (Carvalho 1997).

Radionuclides were separated and purified through ion chromatography columns UTeVa following a procedure described in detail elsewhere (Carvalho, 1997; Oliveira and Carvalho, 2005). Purified radioelements were electroplated on stainless steel discs using a power supply source and a platinum wire as anode. Polonium isotopes were plated on a silver disc from HCl 0.5 M solution in the presence of ascorbic acid. Lead carrier was precipitated to remove ^{210}Pb from solution as lead chromate. After several months of storage, this precipitate was dissolved to determine the ^{210}Po formed from the radioactive decay of ^{210}Pb. All these alpha emitting radionuclides were measured using silicium surface barrier detec-

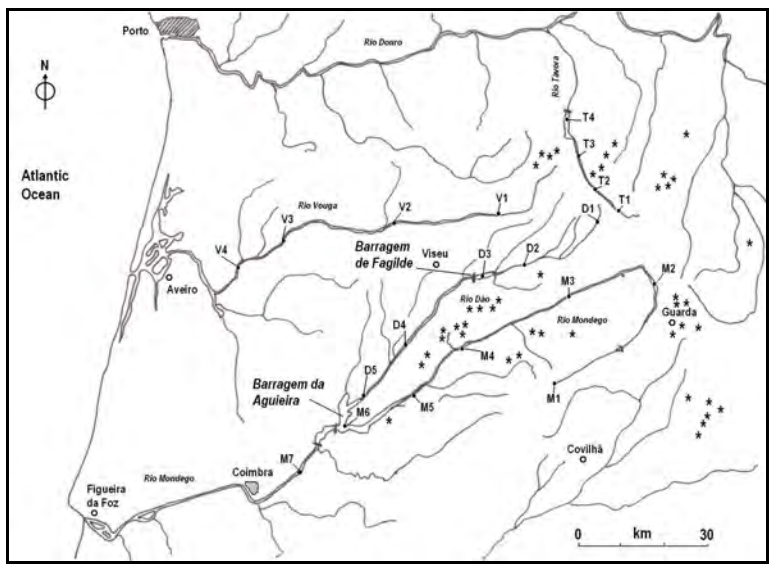

Fig. 2. The Centre-north of Portugal and hydrographical basins investigated. Stars indicate old uranium mines and tagged points indicate sampling stations.

tors connected to an Octete system from Ortec Eg&G and spectra analyzed with the software Maestro.

Radon in water was measured in samples directly collected into glass vials containing scintillation cocktail, tightly closed and measured in the laboratory in a Tri Carb liquid scintillation spectrometer (Lopes et al. 2004). In the field, the main physical-chemical parameters of waters were determined with a portable multiparameter probe, Horiba Instruments. Ions in solution were determined in separate samples, collected in glass vials and kept in the refrigerator until analysis performed with a liquid chromatograph Dionex DX-500.

Results and Discussion

Solid residues from mining operations

The concentrations of radionuclides measured in samples from the uranium mining and milling facilities at Urgeiriça, are shown in Table 1. Concentrations of alpha emitting radionuclides in the heap of low grade ore in the tailings of Santa Barbara (SB), can be compared with concentrations in sand like materials from the tailings of rejected materials (BV) and with the concentrations of radionuclides in the top soil layer of the spoil heap near the ore milling station (MS). Concentrations of radionuclides differ widely amongst sites. Results of the analysis of agriculture soils from the region, collected several km apart of Urgeiriça are shown in Table 1 to provide a comparative near background value. One may observe that the soil at the milling station (MS) is highly contaminated, with concentrations similar to uranium ore, and much higher than those in the pile of untreated low grade ore (SB). In both materials there is no radioactive secular equilibrium of radionuclides belonging to uranium series. The weathering and, likely, the leaching by rain water did nearly halve the radioactivity concentration of ^{226}Ra relative to ^{230}Th and ^{238}U concentrations, and did remove also a fraction of ^{210}Pb that might have existed in the original ore.

In the tailings of Barragem Velha (BV), there is an obvious reduction of uranium concentration, with ^{238}U accounting for about 6% of the ^{238}U concentration measured in the spoil heap at the milling station (MS). However, ^{226}Ra, ^{230}Th and ^{210}Pb concentrations are high in the mill tailings and, clearly, these radionuclides have not been co-extracted with uranium in the chemical treatment plant but, instead, followed the rejected solid waste disposed of in Barragem Velha (Table 1).

The solid wastes disposed of in Barragem Nova (BN) originate from the neutralization of acid mine water and acid runoff from the BV tailings. These wastes are the mud from co precipitation of radionuclides with $BaCl_2$ and $Ca(OH)_2$ in a pond. It is interesting to compare radionuclide concentrations in this mud (BN) with concentrations in mill tailings (BV; Table 1).

Table 1. Residues from mining operations in Urgeiriça. Radionuclide concentrations ($Bq.kg^{-1} \pm 1\sigma$) in solid materials from mining and milling wastes, in soils, in mine water and surface runoff from tailings.

	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Po	^{232}Th
Tailings and soils							
BV-Tailings Bar. Velha	2530± 94	118 ± 12	2876 ± 105	10337 ± 598	24717 ± 2039	20354±681	412 ±32
BN-Tailings Bar.Nova	41598 ±1228	1959 ± 67	40182 ± 1187	13390 ± 613	1690 ±150	1176±43	386 ± 22
SB-Tailings Sta Barbara	6108 ± 173	276 ± 14	6175 ± 175	8052 ± 282	3608 ± 133	3501±112	112 ± 7
MS- Tailings Milling station	38316 ± 1154	1717 ± 67	38247 ± 1152	30115 ± 1123	15569 ± 707	30824±1147	426 ± 21
Soil - Póvoa Espinho #13	230±10	10±2	236±10	301±24	619±96	287±11	226±19
Soil - Espinho #37	386±18	13±2	412±19	301±48	1242±365	321±12	301±48
Urgeiriça mine water							
Filtered water (-20 m)	2.17±0.07	0.106±0.005	2.18±0.07	0.015±0.003	1.48±0.06	0.146±0.004	0.0007±0.0006
Filtered water (-70 m)	2.24±0.07	0.105±0.005	2.24±0.07	0.010±0.005	1.44±0.06	0.129±0.004	0.0006±0.0005
Suspended matter(-20 m)	$(35.1\pm1.7)\times10^3$	$(1.6\pm0.1)\times10^3$	$(35.0\pm1.7)\times10^3$	$(1.8\pm0.1)\times10^3$	$(12.1\pm0.5)\times10^3$	$(52.5\pm2.8)\times\times10^3$	$(0.69\pm0.05)\times10^3$
Suspended matter(-70 m)	$(61.1\pm2.0)\times10^3$	$(2,7\pm0.1)\times10^3$	$(60.4\pm0.2)\times10^3$	$(5.4\pm0.2)\times10^3$	$(14.4\pm0.5)\times10^3$	$(92.2\pm4.7)\times10^3$	$(0.034\pm0.004)\times10^3$
Runoff from tailings							
BV1(solution)-surface runoff	13.2±0.6	0.66±0.06	12.7±0.6	1.17±0.08	0.672±0.06	0.69±0.03	0.024±0.010
BV2(solution)-percolation water	35.7±1.1	1.64±0.07	34.8±1.1	1.06±0.06	1.84±0.03	0.70±0.04	0.044±0.009
BV4(solution)- pond discharge	5.8±0.2	0.28±0.01	5.4±0.2	nd	0.035±0.003	0.020±0.001	nd
BV1(susp.mat)	$(1.96\pm0.08)\times10^3$	$(0.10\pm0.02)\times10^3$	$(2.08\pm0.08)\times10^3$	$(4.8\pm0.3)\times10^3$	$(3.6\pm0.2)\times10^3$	$(7.2\pm0.3)\times10^3$	$<(0.006)\times10^3$
BV2(susp.mat)	$(5.0\pm0.2)\times10^3$	$(0.28\pm0.04)\times10^3$	$(5,4\pm0.2)\times10^3$	$(2.8\pm0.3)\times10^3$	$(0.92\pm0.18)\times10^3$	$(9.2\pm0.5)\times10^3$	$(0.57\pm0.12)\times10^3$
BV4(susp.mat)	$(10.6\pm0.3)\times10^3$	$(0.50\pm0.03)\times10^3$	$(10,7\pm0.3)\times10^3$	$(1.4\pm0.3)\times10^3$	$(4.0\pm0.3)\times10^3$	$(4.4\pm0.2)\times10^3$	$(0.20\pm0.11)\times10^3$

The mud in BN display high concentrations of uranium and very low concentrations of ^{226}Ra and ^{210}Po (= ^{210}Pb), indicating that the precipitation method used to remove radionuclides from liquid wastes may has been effective with uranium but was not very effective with radium and lead.

Mine water and liquid waste

From the sulphuric acid used in the Urgeiriça mine and partial flooding of the underground works, resulted a large volume of acid mine water. During several years after cessing the exploitation of the Urgeiriça mine in 1991, the acid mine water was pumped and neutralized at an external facility near the ponds of BN. In 2003, the pumping of mine water was stopped and the mine was rapidly flooded. The acid mine water, now at pH near 5, contains dissolved uranium series radionuclides (Table 1). This water may come up in surface wells of the region. The mine water was sampled through the main vertical well of the Urgeiriça mine at two different depths, -20m and -70 m below the water surface, and analyzed. The radionuclides in solution and in the particulate phase display very high concentrations (Table 2). This water, after treatment at the neutralization pond and pH adjustment to near neutrality, is released into the Ribeira da Pantanha.

The seepage of rain water and surface runoff from the tailings, loaded with radionuclides leached from the rejected waste materials, drain into the Ribeira da Pantanha (Table 2). These waters are acid and contain also elevated sulphate ion concentrations that are residues of the chemical treatment of uranium ore. Supernatant water released from the neutralisation pond (sample BV4) still contains 5.8 Bq L^{-1} of ^{238}U, *i.e.*, more uranium than water from the Urgeiriça mine, 2.2 BqL^{-1}, although much less ^{238}U than percolation water from BV tailings, 35.7 Bq L^{-1}.

Radionuclides in the percolation water and surface runoff from the mill tailings, currently displaying a pH<3, are mostly in the dissolved phase. Therefore, an effective precipitation would have been important for their removal from waste water before release into the Ribeira da Pantanha.

Ribeira da Pantanha

The water of Ribeira da Pantanha near the source of this stream and upstream the tailings, contains relatively low concentrations of naturally occurring radionuclides. These concentrations are similar to those measured in Ribeira de Vale do Gato, a small stream nearby, and in a tributary to the Ribeira da Pantanha, ranging from 7 to 9 mBq L^{-1}, and can be considered as the non-modified radioactive background (Table 2, RP1 through to RP2). However, drainage from the tailings, containing $42x10^3$ mBq L^{-1} of ^{238}U, and water discharges from the pH neutralization pond containing $5.5x10^3$ mBq L^{-1} of ^{238}U, increase the uranium (^{238}U) concentration in the stream to $4.4x10^3$ mBq L^{-1}. The same happens with ^{226}Ra. With increased distance from the tailings area, this small river contains decreasing con-

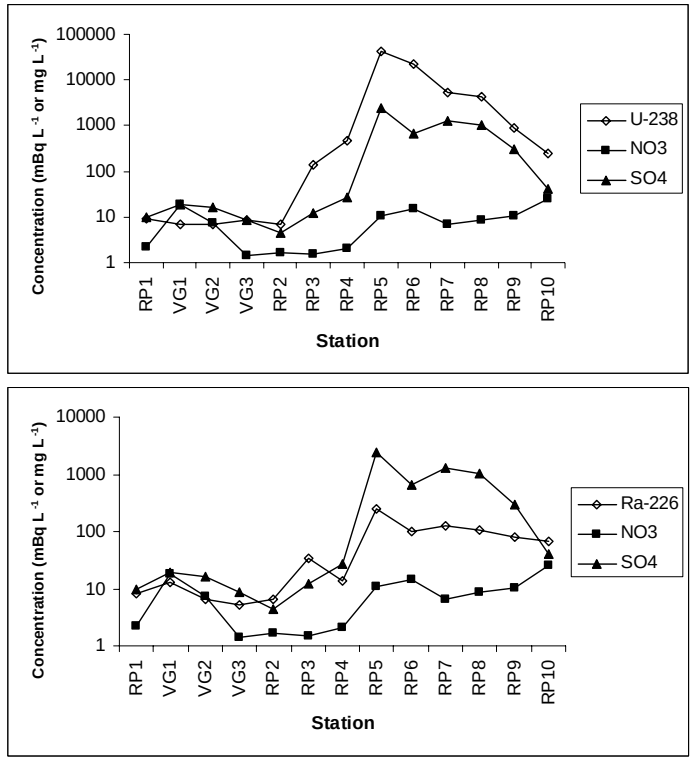

Fig. 3. Concentration of dissolved radionuclides (mBq L^{-1}) and sulphate and nitrate ions (mg L^{-1}) in the streams Ribeira da Pantanha and Vale do Gato.

centrations of radionuclides because of dilution with water from other sources and sorption of dissolved radionuclides onto sediments (Fig. 1 and Table 1). Near the discharge into the river Mondego, ^{238}U in the water of Ribeira da Pantanha decreases to 253 mBq L^{-1}. The radionuclide discharges of Pantanha into the river Mondego are still noticed a few km downstream but are rapidly diluted in the much higher volume of Mondego waters.

In this set of water samples (Table 2), the concentrations of dissolved uranium (^{238}U) and radium (^{226}Ra) display statistically significant positive correlations with concentrations of sulphate ion, R^2= 0.72 and R^2=0.92 respectively (Fig. 3). This correlation reflects the common origin of the dissolved radionuclides and sulphate in the BV tailings. No significant correlations exist between the same radionuclides and the nitrate ion in the water, R^2= 0.04 and R^2=0.09 respectively, revealing that nitrate does not come from the same source (Fig. 3). Instead, nitrate likely comes from agricultural fields mainly located after the tailings. ^{210}Po in the filtered water does not show concentration peaks after the tailings due to its very low water solubility and high distribution coefficient (Kd).

Radioactivity concentrations in the suspended matter of Ribeira da Pantanha were analyzed also. Upstream the tailings, ^{238}U in suspended matter varies between 0.6 and 1.5 kBq kg^{-1}. After the discharge of contaminated water, ^{238}U concentration raises to about 23 kBq kg^{-1} and it is traceable downstream to the discharge into the river Mondego. The signal of the uranium waste discharges from facilities at Urgeiriça is detected also in the bottom sediments of the Ribeira da Pantanha (not shown) with maximum concentrations, about 18 kBq kg^{-1}, at the stations RP8 and RP9 (Fig. 1).

Still today, in the water of Pantanha there is a clear signal of the radioactive wastes from past mining operations. The current levels are likely lower than during the years of ore processing in Urgeiriça, where at the station RP10 ^{238}U concentrations around 6.5 Bq L^{-1} have been reported (Severo at al. 1995). Nevertheless, current levels of radionuclides could still enhance levels in soils and plants if the water from this stream is used for irrigation purposes.

Rivers of the region

The ranges of radionuclide concentrations dissolved in the water of the four rivers are shown in Table 3. Sampling stations were selected, as much as possible, upstream and downstream the uranium mining areas (Fig. 2). The results show that the River Mondego displays at the source, in the granite mountain of Serra da Estrela, very high concentration of radon but negligible concentrations of uranium and uranium daughters in the soluble phase. Down river concentrations are slightly increased in the mining region (M4 and M5) near the discharge of Ribeira da Pantanha at Caldas da Felgueira, with a maximum of 11 mBq L^{-1} of ^{238}U measured there. Nevertheless, radionuclide concentrations decrease again with distance and in the artificial lake of Aguieira (M6), the radioactivity concentrations are reasonably low. Also in the reservoir of Fagilde in the river Dão (D3), radionuclide concentrations in the water are reasonably low.

Concentrations of radionuclides in the waters of rivers Dão, Távora and Vouga can be compared with those of Mondego (Table 3). One could notice that near the source, the water of river Dão contains comparatively high concentrations of radionuclides including uranium, radium and dissolved radon. This is likely due to the mineralogy composition of the surface soils and rocks in the respective river basin. Due to the existence of several unexploited and exploited uranium deposits near the source of Dão, these elevated concentrations are not entirely surprising.

The range of concentrations measured in these rivers is much lower than in the water of Ribeira da Pantanha subject to discharges from the mill tailings, nevertheless, they are comparable to the background values in Ribeira da Pantanha e Ribeira de Vale do Gato.

Table 2. Concentration of dissolved radionuclides (mBq L^{-1} ± 1σ) in water of Ribeira da Pantanha (RP) and Ribeira de Vale do Gato (VG) in the area of Urgeiriça.

	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{222}Rn	^{210}Po	^{232}Th
RP1	8.9 ± 0.3	0.43 ± 0.05	9.4 ± 0.3	0.78 ± 0.08	8.0 ± 0.7	(16.1±1.1)x10^3	9.2 ± 0.2	0.36 ± 0.05
VG1	6.7 ± 0.3	0.31 ± 0.05	6.8 ± 0.3	0.74 ± 0.09	12.9 ± 1.2	(9.9± 0.8)x10^3	7.8 ± 0.2	0.74 ± 0.09
VG2	7.0 ± 0.2	0.34 ± 0.04	7.7 ± 0.2	0.91 ± 0.18	6.7 ± 0.4	____	9.5 ± 0.3	0.39 ± 0.15
VG3	8.6 ± 0.3	0.38 ± 0.03	9.0 ± 0.3	1.5 ± 0.1	5.2 ± 0.3	(2.5± 0.3)x10^3	10.9 ± 0.3	0.35 ± 0.07
RP2	7.1 ± 0.4	0.37 ± 0.08	7.8 ± 0.4	0.42 ± 0.11	6.7 ± 0.9	(72.8± 3.0)x10^3	22.6 ± 0.9	0.13 ± 0.09
RP3	135 ± 5	6.2 ± 0.3	129 ± 4	2.6 ± 0.2	33.4 ± 1.8	(11.0± 0.7)x10^3	11.7 ± 0.3	0.10 ± 0.06
RP4	464 ± 15	22.0 ± 0.9	472 ± 15	4.7 ± 0.3	13.8 ± 0.9	(3.8± 0.3)x10^3	6.6 ± 0.2	0.30 ± 0.08
RP5	(42±1)x10^3	(1.84±0.09)x10^3	(41±1)x10^3	(1.3±0.1)x10^3	247 ± 16	(73.6±12.9)x10^3	134 ± 7	44.8 ± 6.3
RP6	(22.6±0.8)x10^3	992 ± 39	(22.2±0.8)x10^3	691 ± 31	99.8 ± 3.5	(116.8±4.0)x10^3	176 ± 9	14.3 ± 1.7
RP7	(5.5±0.6)x10^3	235 ± 30	(5.3±0.6)x10^3	2.7 ± 0.3	125 ± 14	(1.9± 0.4)x10^3	19.0 ± 0.8	0.46 ± 0.14
RP8	(4.4±0.2)x10^3	202 ± 13	(4.4±0.2)x10^3	2.5 ± 0.4	103 ± 6	(1.9± 0.4)x10^3	20.6 ± 0.8	0.51 ± 0.30
RP9	874 ± 48	38.0 ± 3.5	873 ± 48	5.6 ± 0.6	78.5 ± 3.4	____	21.5 ± 0.9	0.63 ± 0.27
RP10	253 ± 8	11.2 ± 0.7	262 ± 8	4.6 ± 0.5	66.2 ± 3.5	____	22.7 ± 0.9	0.41 ± 0.21

Table 3. Range of radionuclide concentrations (mBq L^{-1}) in filtered water of the rivers Dão, Vouga, Távora, and Mondego.

	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{222}Rn	^{210}Po	^{232}Th
River Dão (D1-D5)	8.0 - 23	0.36 – 1.2	8.4 –25.6	1.9 –13.1	5.3 – 23.5	(1.1 - 81) x10^3	7.6 – 26.5	0.10 – 0.60
River Vouga (V1-V4)	3.7 – 6.6	0.14 - 0.30	3.8 –6.6	0.86 –1.4	4.4 –7.5	(1.7 – 2.5)x10^3	6.9 – 13.0	0.19 – 0.30
River Távora (T1-T4)	3.6 – 7.6	0.14 - 0.36	3.7 –7.8	0.88 –1.6	1.3 –5.2	(0.9 – 15.1)x10^3	9.0 – 17.7	0.14 – 0.21
River Mondego (M1-M7)	3.4 – 11.0	0.17 - 0.60	3.8 –11.5	0.52 – 2.1	2.5 –8.9	(<LLD - 103)x10^3	5.3 – 12.2	0.15 – 0.30

Conclusions

The concentrations of radionuclides of the uranium series in the mill tailings and mud from neutralization of acid mine waters in Urgeiriça, are 10 to 180 times higher than agriculture soils of the region. The disposal of those spoil materials in the open air, without coverage and confinement, facilitates the dispersal of radioactive waste in the environment. Such dispersal may cause an unacceptable exposure of the population to external radiation doses and may lead to internal contamination. Therefore, would be advisable to take appropriate measures to prevent such a radiation exposure (IAEA, 2002). Furthermore, the drainage from mill tailings and liquid discharges from acid water treatment ponds, increase the concentrations of radionuclides in the Ribeira da Pantanha. These radionuclide levels are above the recommended limits and, if water is used for irrigation or to feed cattle, may transfer contamination to the plants and to livestock.

The analysis of waters and sediments in the main rivers of this region indicate that the dispersal of radionuclides did not spread far from the mine sites and mill tailings. Although Ribeira da Pantanha displays a strong enhacement of radioactivity levels, no significant enhancement of radioactivity levels were detected in the main river basins of the region. In particular, the two main water reservoirs in the region, Aguieira and Fagilde dams, in the rivers Mondego and Dão, respectively, do not show enhanced radioactivity levels. Furthermore, radionuclide concentrations in water are currently below the legal limits applicable to water for human consumption. Nevertheless, measures should be implemented to keep that milling waste properly confined near the mines and avoid waste dispersal and transport to the hydrographical basins and to agriculture soils.

The proper management of the radioactive mining and milling waste would advise to confine the uranium mining wastes, especially those in the mill tailings, and monitor the environmental contamination (IAEA, 2002). Furthermore, this would go along with precaution measures to prevent consequences of natural catastrophes that may turn into radiological emergences. Actually, the total amount of radioactive mining and milling waste is meaningful and it is exposed to weathering. Exceptional storms or even average rainy weather conditions may flush these solid wastes from the tailings to the river basins, and eventually this would cause a meaningful radiological impact in water reservoirs of the region.

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