

Analyses of radionuclides in soil, water, and agriculture products near the Urgeiriça uranium mine in Portugal

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Abstract Analyses of soils, irrigation waters, agriculture products (lettuce), green pasture, and cheese were performed in samples collected in the area of the old Urgeiriça uranium mine and milling facilities, Centre-North of Portugal, in order to assess the transfer of uranium series radionuclides in the environment and to man. Soils close to milling tailings display an enhancement of radioactivity. In the drainage basin of the stream Ribeira da Pantanha, receiving drainage from the tailings piles and discharges from the acid mine water treatment plant, there was enhancement of uranium series radionuclide concentrations in water and suspended matter. Agriculture products from kitchen gardens irrigated with water from the Ribeira da Pantanha show an increase of radioactivity, mainly due to uranium isotopes. Agriculture products from other kitchen gardens in this area, irrigated with groundwater, as well pasture and cheese produced locally from sheep milk did not show enhanced radionuclide concentrations. In the Urgeiriça area, some soils display radionuclide concentrations higher than soils in reference areas and, in agriculture products grown there, ^{226}Ra was the radionuclide more concentrated by vegetables. Through ingestion of these products ^{226}Ra may be the main contributor to the increment of radiation dose received by local population.

Keywords Uranium mining residues · Radium · Thorium · Polonium · Water · Plants · Pasture · Cheese

Introduction

The legacy of historic uranium mining and milling sites in Portugal, often located near villages and in agriculture areas, raised concerns with the potential enhancement of radioactivity in agriculture products and with the radiation exposure of populations through food chain transfer of uranium series radionuclides.

Extraction of radioactive ores in the Urgeiriça mine started in 1913 and lasted until 1992. Near this mine, located in the County of Canas de Senhorim, Viseu, Centre-North of Portugal, the uranium ore extracted from the Urgeiriça mine and from other mines in the region was chemically processed. Milling tailings (residues of acid extracted ores) were dumped nearby and, by the closure of Urgeiriça facilities in the year 2001, tailings piles amounted to about 3 million tonnes of sandy materials. To these tailings materials were added during the last two decades several tonnes of sludge containing the precipitate of barium chloride and radionuclides resulting from neutralization of acid mine waters [1]. Near the Urgeiriça mine, with the main shaft at Santa Barbara, there is the shaft of another abandoned underground uranium mine, Valinhos (Fig. 1).

Radioactivity of the uranium mining and milling wastes disposed off at Canas de Senhorim were measured before and these materials are known to contain high concentrations of radionuclides of the uranium radioactive decay series [2]. Surface runoff from these tailings materials transport dissolved and particulate radionuclides into the Pantanha stream, a small river draining into the Mondego River [3]. Reports of previous studies carried out in this area concluded that there was an enhancement of radioactivity in surface waters and water from wells in the vicinity of the milling tailings [4, 5]. In the year 2006, environmental remediation work was started by the mine

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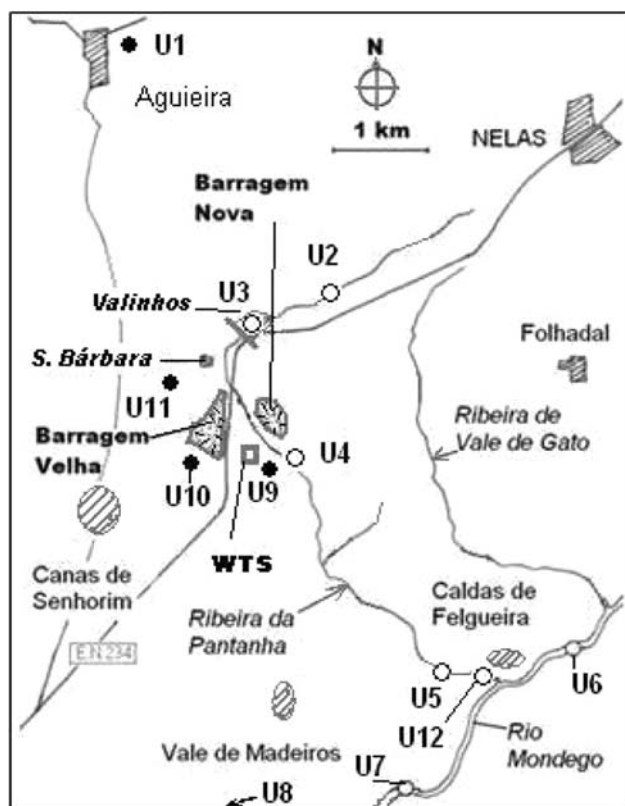


Fig. 1 Region of Urgeiriça mine with location of the sampling stations and other features of the region. *Solid circles*: wells; *open circles*: surface waters. WTS, mine water treatment station

holding company in the Urgeiriça area. Engineering works were undertaken to cap the main milling tailings of Barragem Velha. These works are nearly completed and are expected to control erosion and surface runoff from tailings piles, and to facilitate the chemical treatment of contaminated drainage. In order to follow up and assess the effect of engineering works on radioactivity levels in the region, an environmental survey was carried out in the Urgeiriça area during the summer of 2007.

Results of analyses of uranium series radionuclides in surface waters, water from wells, soils and agriculture products from the county of Canas de Senhorim are presented herein. Implications of these preliminary results for environmental radioactivity in this region are discussed.

Materials and methods

Sampling

Soil, water and agriculture product samples were collected in May and July 2007 in the county of Canas de Senhorim, district of Viseu, North of Portugal. Location of sampling

stations and other features of the region are identified in Fig. 1.

Soil samples of the 20 cm upper layer were collected in horticulture plots pooling in one sample equal soil portions taken at four points in a 3×3 m square in order to ensure more representative samples. In the laboratory, soils were dry at 60°C in the oven and sieved through a metal screen. Soil fraction of grain size $<63\ \mu\text{m}$ was retained for analysis in order to allow comparison of analytical results amongst soil samples.

Water samples were collected in rivers and wells directly into polyethylene drums and filtered on site. Filtration was performed through membrane acetate filters, with $0.45\ \mu\text{m}$ pore size and 142 mm in diameter, in stainless steel filter holders and using pressure (<1.5 bar). Filtered water was acidified with HNO_3 to pH below 2, and filters with suspended matter were stored for analysis in the laboratory. Physical chemical parameters of water were measured in situ with a portable multiparameter probe Horiba U-22. Accurate location of sampling stations and water parameters is shown in Table 1.

Vegetables (lettuce) were collected from the kitchen gardens and lettuce leaves only, after thorough washes with tap water and distilled water, were freeze dried, reduced to powder, homogenized and used for radionuclide analysis. A sample of green pasture from the watershed of the stream Ribeira da Pantanha, downstream the tailings piles, and locally produced cheese were collected and freeze dried for radionuclide analyses as well.

Radionuclide analysis

Soil, water and suspended matter and biological samples were analyzed by radiochemical methods and alpha spectrometry according to methods described in detail elsewhere [4, 6]. In brief, known amounts of isotopic tracers (^{232}U , ^{229}Th , ^{209}Po , ^{224}Ra , and stable Pb^{2+}) were added to the samples in the beginning of the chemical processing. In the case of water samples, radionuclides were co precipitated with MnO_2 and the redissolved precipitate was passed through ion exchange columns in order to separate radionuclide fractions. These were brought to electrodeposition and discs with radionuclides were measured for alpha radiation. Samples of suspended matter and foods were spiked with the isotopic tracers and completely dissolved with acids before chemical extraction of radioelements by ion exchange column chromatography. Solutions of purified radioelements were then transferred to electroplating cells and radionuclides plated onto stainless steel or silver discs. Radionuclides were measured with $450\ \text{mm}^2$, silicon surface low background detectors, connected to a multi-channel analyser OctetePlus® (Ortec EG&G). These methods have been thoroughly tested and accuracy and

Table 1 Radionuclide concentrations (Bq kg⁻¹ dry weight \pm 1 σ) in solid materials (<63 μ m size fraction) and water (mBq L⁻¹ \pm 1SD) from Urgeiriça mine and soils in the region of Canas de Senhorim and other counties

	²³⁸ U	²³⁵ U	²³⁴ U	²³⁰ Th	²²⁶ Ra	²¹⁰ Pb = ²¹⁰ Po	²³² Th
Uranium mining waste							
Milling pile “Barragem Velha”	2,530 \pm 94	118 \pm 12	2,876 \pm 105	10,337 \pm 598	24,717 \pm 2039	20,354 \pm 681	412 \pm 32
Sludge pond “Barragem Nova”	41,598 \pm 1,228	1,959 \pm 67	40,182 \pm 1,187	13,390 \pm 613	1,690 \pm 150	1,176 \pm 43	386 \pm 22
Acid mine water							
Filtered water (pH = 4.8)	(2.17 \pm 0.07) \times 10 ³	106 \pm 5	(2.18 \pm 0.07) \times 10 ³	14.9 \pm 2.9	(1.48 \pm 0.06) \times 10 ³	146 \pm 4	0.7 \pm 0.6
Suspended matter	35.1 \pm 1.7	1.6 \pm 0.1	35.0 \pm 1.7	1.8 \pm 0.1	12.1 \pm 0.5	52.5 \pm 2.8	0.69 \pm 0.05
Soils Canas de Senhorim							
U9	392 \pm 11	17.8 \pm 1.5	383 \pm 11	398 \pm 27	263 \pm 23	252 \pm 12	232 \pm 16
U10	392 \pm 12	18.6 \pm 1.8	440 \pm 13	1,155 \pm 82	1,253 \pm 102	1,233 \pm 51	310 \pm 24
Soils of other counties							
Seia/Arrifana	210 \pm 7	10.5 \pm 1.1	216 \pm 7	186 \pm 11	232 \pm 16	222 \pm 11	161 \pm 10
Celorico da Beira/S. Pedro	256 \pm 9	13.5 \pm 1.6	259 \pm 9	293 \pm 16	313 \pm 23	287 \pm 15	236 \pm 13

precision checked in international intercomparison exercises organized by the IAEA [7].

Results and discussion

Concentrations of radionuclides in agriculture soils from the county of Canas de Senhorim, area of the Urgeiriça mine, and soils from the counties of Seia and Celorico da Beira, located about 30–40 km NE of Canas de Senhorim, are shown in Table 2. Soils from Seia and Celorico da Beira counties are from the same geological province but these sampling locations were not close to any old uranium mine, while soils from Canas de Senhorim were sampled in horticulture plots near the Urgeiriça uranium mine facilities, at stations U9 and U10. Materials from the tailings piles Barragem Velha and Barragem Nova (Fig. 1) are included in the Table for comparison. Materials dumped in Barragem Velha are milling tailings (extracted ore residues) and materials dumped in Barragem Nova are the sludge from acid mine water treatment, i.e., precipitates of barium chloride and mud obtained with the addition of hydroxide to rise pH and neutralize the acid. Radionuclide composition of materials dumped into these two tailings piles is different, but both contain high concentrations of uranium series radionuclides and, if dispersed, these residues may elevate concentrations of radionuclides in the environment.

Radionuclide concentrations in soils, U9 and U10, from two kitchen gardens in the vicinity of tailings were slightly higher than in other soils of the region away of uranium mining areas. Those radionuclide concentrations in U9 and U10 soils were likely elevated by mixing with materials originated in the tailings. Particularly one of them, U9, displays a strong increase in ²³⁰Th, ²²⁶Ra and ²¹⁰Pb without significant increase of uranium concentrations which is the typical radionuclide disequilibrium found in milling tailings [2].

The stream Ribeira da Pantanha flowing by the Urgeiriça mining facilities is the main drainage basin collecting surface runoff, leaching from tailings, and mine water discharges. The water treatment station (WTS) located near the Barragem Nova, after neutralization of acid waters and decantation of precipitate in polyethylene coated ponds, discharges the overlaying water into the Ribeira da Pantanha and pumps the sludge as slurry into the Barragem Nova dump. Results of radionuclide analyses in water samples from Ribeira da Pantanha and Mondego River are shown in Table 3. Following the stations along the stream, there is a noticeable increase of radionuclide concentrations from the upstream area (U2) to the small dam of Valinhos (U3). This dam was built near the entrance and with mining materials from the underground uranium mine of Valinhos.

Table 2 Sampling stations in the Urgeiriça mine area, county of Canas de Senhorim

Sampling station	GPS coordinates		Physical chemical parameters of water samples			
	Latitude (N)	Longitude (W)	pH	Conductivity (mS m ⁻¹)	Eh (mV)	Suspended load (mg L ⁻¹)
U1 (well)	40°31'39.6"	07°54'44.8"	5.70	28	231	2.2
U2 (Pantanha)	40°31'01.2"	07°53'01.9"	6.31	10	154	1.3
U3 (Pantanha)	40°30'57.9"	07°53'13.3"	6.32	11	137	2.5
U4 (Pantanha)	40°30'23.5"	07°53'04.6"	5.63	140	74	27.4
U5 (Pantanha)	40°29'19.6"	07°52'07.2"	6.52	75	135	4.2
U6 (Mondego)	40°29'28.4"	07°51'05.9"	6.74	8	136	4.0
U7 (Mondego)	40°28'45.5"	07°52'05.2"	6.30	8	203	6.2
U8 (Mondego)	40°20'13.2"	08°09'25.9"	8.28	10	177	3.8
U9 (well)	40°30'22.0"	07°53'05.9"	5.65	13	211	3.2
U10 (well)	40°30'26.2"	07°53'37.7"	5.17	15	258	1.8
U11 (well)	40°30'38.8"	07°53'50.4"	4.77	36	257	1.4
U12 (Pantanha)	–	–	4.17	57	255	12.1

Downstream Valinhos, radionuclide concentrations were especially high after the point of discharge of the acid mine water treatment station, U4. Concentrations of dissolved radionuclides decreased downstream and are very low in the Aguieira dam in the Mondego River (U 8), about 30 km SW of Canas de Senhorim. These trends in concentrations of soluble radionuclides in Ribeira da Pantanha and Mondego River were noticed before, although absolute concentrations vary with the years [5].

Radionuclide concentrations in the water of Ribeira da Pantanha were measured also in 26 July 2007 (U12) several days after heavy rains that partially destroyed the cap laid down on tailings piles and enhanced surface runoff and transport of materials from Barragem Velha into the Mondego River. Radionuclide dissolved concentrations measured at U12 in July were higher in comparison with

measurements made in May, at station U5, although the reverse was observed in particulate matter which showed lower specific activity due to materials used in the capping (Tables 3 and 4).

Radionuclide concentrations in the soluble phase of water from the superficial aquifer in the area of Canas de Senhorim are shown in Table 3 also. This water is extracted through wells, used for irrigation of kitchen gardens and occasionally as a drinking water supply. U1 is a reference well in the village of Aguieira, 5 km NW of Canas de Senhorim, in an area not impacted by uranium mining activities. Other wells sampled in the region (U9, U10, U11) were located much closer to the Urgeiriça mine than U1. Nevertheless, radionuclide concentrations in their waters were not much different in comparison with U1 (reference well).

Table 3 Radionuclide concentrations (mBq L⁻¹ ± 1SD) in the soluble phase of waters in Canas de Senhorim region

Station	²³⁸ U	²³⁵ U	²³⁴ U	²³⁰ Th	²²⁶ Ra	²¹⁰ Pb	²¹⁰ Po	²³² Th
U1 (well)	80.4 ± 2.5	3.9 ± 0.3	73.3 ± 2.3	0.12 ± 0.02	97.6 ± 11.3	61.6 ± 1.8	6.6 ± 0.2	0.02 ± 0.009
U2 (Pantanha)	37.4 ± 1.3	2.0 ± 0.2	40.6 ± 1.4	1.06 ± 0.09	7.6 ± 2.8	11.4 ± 0.5	1.98 ± 0.08	0.03 ± 0.01
U3 (Pantanha)	136 ± 4	6.4 ± 0.4	135 ± 4	2.3 ± 0.2	13 ± 1.2	19.3 ± 0.7	7.4 ± 0.3	0.05 ± 0.01
U4 (Pantanha)	1035 ± 29	48.3 ± 1.7	1026 ± 29	1.0 ± 0.1	45.8 ± 4	6.0 ± 0.3	0.96 ± 0.06	<1.2 × 10 ⁻³
U5 (Pantanha)	205 ± 6.0	9.4 ± 0.5	208 ± 6	1.0 ± 0.1	108 ± 10	5.3 ± 0.3	2.1 ± 0.3	0.06 ± 0.02
U6 (Mondego)	5.0 ± 0.2	0.21 ± 0.03	5.2 ± 0.2	0.9 ± 0.1	3.5 ± 0.4	6.7 ± 0.3	1.5 ± 0.1	0.1 ± 0.03
U7 (Mondego)	9.3 ± 0.3	0.46 ± 0.04	9.8 ± 0.3	0.8 ± 0.2	7.5 ± 0.5	6.3 ± 0.3	1.88 ± 0.09	0.06 ± 0.06
U8 (Mondego)	3.8 ± 0.2	0.18 ± 0.03	3.8 ± 0.2	1.5 ± 0.2	16.4 ± 1.8	8.5 ± 0.5	4.2 ± 0.1	0.24 ± 0.06
U9 (well)	61.9 ± 1.5	2.7 ± 0.1	68.1 ± 1.6	1.7 ± 0.1	26.3 ± 1.6	37.9 ± 1.3	8.6 ± 0.2	0.20 ± 0.04
U10 (well)	9.8 ± 0.3	0.48 ± 0.05	10.9 ± 0.3	1.8 ± 0.1	29.4 ± 2.1	23.4 ± 0.8	12.7 ± 0.4	0.32 ± 0.05
U11 (well)	74.2 ± 1.7	3.5 ± 0.2	79.4 ± 1.8	1.9 ± 0.2	111 ± 8	93.1 ± 2.4	1.64 ± 0.03	0.38 ± 0.06
U12 (Pantanha 26.07.07)	373 ± 10	16.8 ± 0.7	371 ± 10	4.6 ± 0.2	153 ± 8	NA	8.1 ± 0.4	0.17 ± 0.03

Sampling performed on 21–23 May 2007

NA Not analyzed

Table 4 Radionuclide concentrations ($\text{Bq kg}^{-1} \pm 1\text{SD}$ dry weight) in the particulate phase of waters in Canas de Senhorim region

Station	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
U1 (well)	$28,241 \pm 801$	$1,284 \pm 69$	$28,301 \pm 803$	$1,781 \pm 91$	$4,427 \pm 289$	NA	$17,070 \pm 555$	103 ± 13
U2 (Pantanha)	$6,359 \pm 241$	260 ± 35	$6,454 \pm 244$	$1,171 \pm 74$	$4,934 \pm 387$	NA	$7,527 \pm 339$	267 ± 29
U3 (Pantanha)	$12,389 \pm 471$	825 ± 73	$11,954 \pm 457$	$1,208 \pm 80$	$2,658 \pm 267$	NA	$6,169 \pm 257$	59 ± 12
U4 (Pantanha)	$69,559 \pm 3535$	$3,438 \pm 189$	$66,319 \pm 3,371$	$2,387 \pm 119$	379 ± 61	NA	726 ± 25	16 ± 2
U5 (Pantanha)	$37,108 \pm 1027$	$1,759 \pm 73$	$37,050 \pm 1,026$	$3,923 \pm 205$	3147 ± 276	NA	$6,171 \pm 213$	201 ± 18
U6 (Mondego)	761 ± 44	36 ± 8	731 ± 43	440 ± 38	$3,600 \pm 348$	NA	$1,148 \pm 54$	52 ± 12
U7 (Mondego)	999 ± 30	47 ± 4	990 ± 30	364 ± 18	$3,304 \pm 290$	NA	768 ± 34	40 ± 4
U8 (Mondego)	524 ± 28	32 ± 6	551 ± 28	213 ± 16	$4,826 \pm 482$	NA	$1,345 \pm 60$	84 ± 9
U9 (well)	$3,338 \pm 101$	135 ± 13	3636 ± 108	414 ± 22	$5,989 \pm 349$	NA	$3,914 \pm 151$	65 ± 6
U10 (well)	$1,619 \pm 66$	68 ± 11	$1,673 \pm 67$	907 ± 46	$15,186 \pm 1,290$	NA	$4,150 \pm 144$	124 ± 12
U11 (well)	$10,326 \pm 254$	516 ± 29	$10,171 \pm 251$	$1,128 \pm 62$	$16,325 \pm 1,077$	NA	$3,425 \pm 130$	121 ± 15
U12 (Pantanha 26.07.07)	$4,884 \pm 107$	233 ± 8	$4,916 \pm 107$	$1,805 \pm 74$	$1,704 \pm 138$	NA	$3,310 \pm 110$	21 ± 2

Sampling performed on 21–23 May 2007

NA Not analyzed

Radionuclide concentrations in the particulate phase of water from rivers and water from wells are shown in Table 4. It may be noticed (computation can be made using suspended matter load given in Table 1) that radionuclide concentrations in the soluble phase account for 35–86% of total uranium, 20–91% of total ^{226}Ra , 6–64% of total ^{210}Po and 0.3–69% of ^{232}Th . Variation largely depends on the sampling station and particle-water partitioning of radionuclides is modified by the mine water treatment plant.

Results of the analysis of some agriculture products of the Urgeirica mine area are shown in Table 5. The most abundant vegetable on the periods of sampling was the lettuce.

Concentrations of radionuclides in lettuces from various kitchen gardens can be compared with the lettuce sample from the reference station U1. Lettuce was sampled at U1 station in May and July and an increase in radionuclide

concentrations was observed in July, in older vegetables. The same increase of concentrations in older vegetables was observed in other stations (Table 5). The highest uranium concentrations were measured in lettuce grown at station U12, irrigated with water from Ribeira da Pantanha containing enhanced radionuclide concentrations (Table 3). Lettuce from other stations irrigated with water from wells contained less uranium than U12.

Amongst radionuclides analyzed in lettuce, on one hand, uranium series (^{238}U) radionuclides are present in higher activity than thorium (^{232}Th) series radionuclides, as found in soils and waters. On the other hand, in the uranium series ^{226}Ra is consistently more concentrated by lettuce than other radionuclides. Concentration of ^{226}Ra to levels comparatively higher than other radionuclides was noticed before in potatoes (Carvalho et al., in press).

Table 5 Radionuclide concentrations ($\text{Bq kg}^{-1} \pm 1\text{SD}$ dry weight) in vegetables (lettuce), pasture and cheese from the region of Canas de Senhorim

Station	Dry : wet weight	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{232}Th
21–23.5.2007, Canas de Senhorim							
Lettuce U1 (Reference)	0.032	0.95 ± 0.04	0.049 ± 0.009	0.92 ± 0.04	0.88 ± 0.05	11.2 ± 0.9	0.18 ± 0.02
Lettuce U9, C	0.042	1.29 ± 0.05	0.060 ± 0.009	1.42 ± 0.05	0.89 ± 0.09	5.1 ± 0.4	0.30 ± 0.04
Lettuce U11	0.036	0.34 ± 0.02	0.010 ± 0.003	0.35 ± 0.02	0.31 ± 0.05	1.3 ± 0.2	0.17 ± 0.03
25–27.7.2007, Canas de Senhorim							
Lettuce U1 (Reference)	0.11	2.36 ± 0.08	0.16 ± 0.02	2.35 ± 0.08	2.2 ± 0.1	8.9 ± 0.8	0.80 ± 0.07
Lettuce U9	0.09	7.0 ± 0.2	0.34 ± 0.04	7.0 ± 0.2	6.7 ± 0.3	7.2 ± 0.5	3.2 ± 0.2
Lettuce U10	0.19	5.6 ± 0.2	0.24 ± 0.04	6.1 ± 0.3	10.8 ± 0.5	23.9 ± 2.2	1.80 ± 0.1
Lettuce U12	0.15	123 ± 4	6.0 ± 0.3	121 ± 4	28.5 ± 1.6	29.7 ± 3.5	4.3 ± 0.3
Pasture	0.26	1.79 ± 0.07	0.09 ± 0.02	1.74 ± 0.07	2.4 ± 0.1	25.2 ± 2.4	0.76 ± 0.06
Milk (Bq L^{-1}) ^a	0.22	0.88 ± 0.04	0.048 ± 0.009	0.91 ± 0.04	1.3 ± 0.1	5.9 ± 0.3	0.23 ± 0.05
Cheese	0.79	0.138 ± 0.008	0.003 ± 0.001	0.123 ± 0.008	0.42 ± 0.04	1.15 ± 0.07	0.25 ± 0.03

^a Density = 1.05

Green pasture grown in the watershed of Pantanha stream is used as a grazing field by lambs. Results of the analysis of a large pasture sample indicate that radionuclide concentrations in the pasture were similar to those determined in lettuce (Table 5). Cheese locally produced with milk from those sheep, displays concentrations that are roughly one order of magnitude lower than in the pasture. Furthermore, these results suggest that food chain transfer from pasture to sheep, and through milk production to the cheese, might not pose a hazard from the radioecological point of view.

Conclusions

Results from this survey in the geographic area of the old uranium mine and milling facilities of Urgeiriça confirmed the elevated radionuclide concentrations in the mining and milling waste and indicated some dispersal of uranium series radionuclides in the nearby environment [8–10].

The stream Pantanha still receives discharges of acid effluents from the mine and leaching from the waste piles containing enhanced radionuclide concentrations. These relatively high concentrations were traced downstream to the Mondego River, but rapidly are diluted in this river and decrease to background levels in the artificial lake of Aguieira.

In Canas de Senhorim, analysis of water from wells showed concentrations similar to those measured in water from a reference well a few km apart. However, it is a common feature of ground waters to contain higher radionuclide concentrations from uranium series in comparison with surface waters.

Vegetables produced in kitchen gardens in Canas de Senhorim, including locations close to the milling tailings of Barragem Velha, did not show unbearable enhancement of radioactivity. The highest concentrations were measured in lettuces grown with water from Ribeira da Pantanha. In general, ^{226}Ra is the radionuclide more concentrated by vegetables and through ingestion may be the main contributor to the dose to local population. This aspect shall be investigated further and an effective dose assessment is advised.

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