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Radioactivity in Soils and Vegetables from Uranium Mining Regions

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Abstract

Samples of soils, irrigation water, and vegetable products from three areas were analyzed for radionuclides. These areas are located in the basins of Ribeira da Pantanha, Ribeira do Castelo and streams tributaries to Ribeira de Inguias, all receiving drainage from old uranium mines. Results showed enhancement of radionuclide concentrations, especially ^{226}Ra, in vegetables from kitchen gardens in these areas when compared with reference areas upstream the mine's discharge points. The main source and transfer pathway of radionuclides to plants was the irrigation water used, rather than the soil. Non-contaminated products were obtained in these areas when irrigation was made with non-contaminated water from deep wells, instead of water from surface streams and irrigation wells contaminated with mine drainage. Guidelines for radionuclide concentrations in irrigation water and soil are needed.

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Keywords: Uranium; Radium; Polonium; Mine drainage; Soil to plant transfer.

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1. Introduction

Radiological exposure of humans to radionuclides from uranium mining and milling waste depends upon the environmental dispersion of uranium waste and radionuclide transfer pathways. These pathways include the transfer of radionuclides from uranium waste to soils and from soils to plants, and then radionuclide ingestion with the diet of local populations. This diet may include agriculture products from former uranium mining areas of the center of Portugal, especially those located along streams and rivers that receive U-mine drainage. In these hilly areas farms generally are of small area co-existing with forest and agro-forest farms and most horticultural garden farms are for family consumption. These garden soils are concentrated in alluvial deposits of rivers and generally are rich in organic matter and high in silica content. Irrigation is essential for the production and this is ensured either with water pumped from surface streams or from irrigation wells. These irrigation wells tap surface aquifers and generally were dug near the streams and water lines.

This paper reports results on radioactivity levels in soils, irrigation water, and vegetables grown in agriculture plots near former uranium mining in the center of Portugal and located in the catchment of streams and rivers flowing through those areas.

2. Materials and Methods

Three areas were investigated: one near the Urgeiriça and Valinho mines in the county of Nelas, other near the mines of Cunha Baixa and Quinta do Bispo in the Mangualde county, and a third one near the Vale de Arca, Carrasca, Pedreiros and Bica old uranium mines in the Sabugal county (Figure 1).

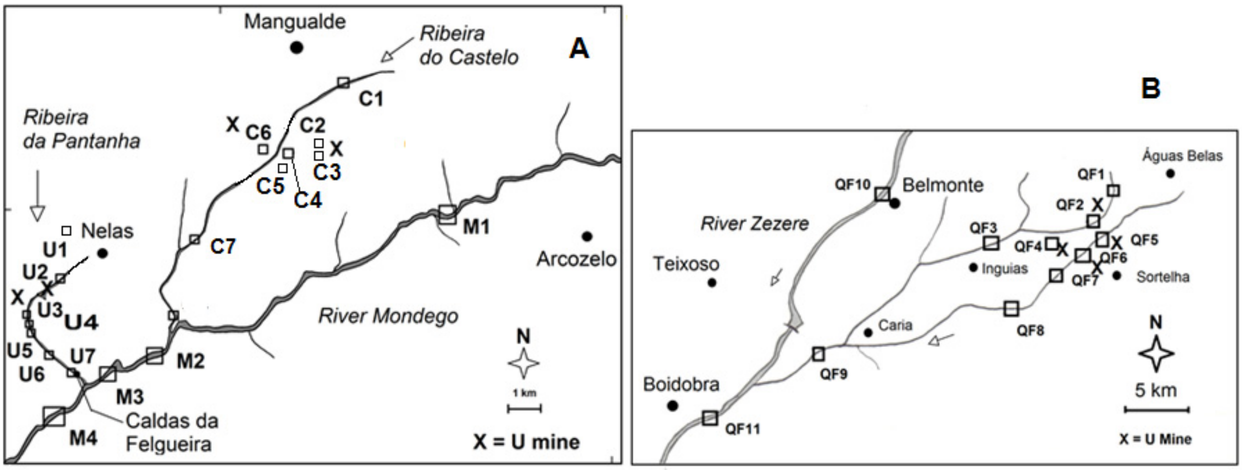

Figure 1 – Maps of the study areas with location of sampling stations. A, Mondego River basin; B, Zêzere River basin.

In the first area were located old uranium mines, mining waste, milling plant facilities, milling tailings plus unprocessed low grade ore and rubbles from the dismantling of the chemical plant. All these materials were concentrated in one single place, the milling tailings Barragem Velha, and the waste capped in 2005-6. The area, with about 10 ha, was reshaped, a multilayer cover put in place, and the area fenced. Nearby, at Barragem Nova, a waste water treatment plant (ETAM) was refurbished and automated to carry on neutralization and radionuclide co precipitation with barium chloride. Sludge from mine and waste water treatment is deposited at Barragem Nova. Acid mine drainage and acid seepage from the milling tailings are now collected and treated in this ETAM, and treated water released into Ribeira da Pantanha stream which flows through the area. After cleanup of the area of Valinhos mine, a small dam was build up on Ribeira da Pantanha and the artificial lake formed there is intended to became a leisure and picnic area. Stretching the Ribeira da Pantanha, there are several areas with agriculture and pasture for ovine and bovine cattle. An area of kitchen gardens does exist at the Caldas da Felgueira village and

Ribeira da Pantanha stream water is used for irrigation. The site U1, with similar characteristics but upstream the mine zones was used as a reference area (Figure 1).

In the second area, in the Mangualde county, two large mines Quinta do Bispo and Cunha Baixa have for many years released untreated and treated drainage into the Ribeira do Castelo stream. Agriculture is abundant in this area, especially near Cunha Baixa and in alluvial soils along the Castelo stream. Water from this stream and from irrigation wells dug near the stream is used for irrigation. Most agriculture plots are kitchen gardens, but one large apple orchard does exist at Cunha Baixa village. At the time of this survey no environmental remediation had been initiated in these mines. Station CB1 a similar kitchen garden at the village of Mesquitela and on the banks of the Ribeira do Castelo stream was used as a reference site (Figure 1).

In the third area, in the Sabugal county, four old uranium mines are located in a valley running NNE-SSW from Águas Belas to the historic village of Sortelha. The valley is drained by two streams, Quarta-feira and Valverdinho, which merge into the Ribeira de Inguias stream, and further south this one discharges into the River Zêzere. In the valley and along the streams there is abundant agriculture production which uses water from wells dug on stream banks for irrigation. Most production of agriculture plots are for family consumption, but wide productions of maize and wine do exist as well.

Samples of water from irrigation wells, soil from the agriculture plots, and plants from the available production were collected in the three areas on May 2010. Some agriculture plots were located close to the mines and others at km from the mines, along the streams receiving drainage from mine areas.

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 (lettuce, cabbage, tomatoes, rtc.) 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 retaken for radioanalysis.

Radionuclides were determined in all samples applying radiochemical separation of radionuclides, electrodeposition and radiation measurement by alpha spectrometry. The procedures followed are described in detail elsewhere[1,2,3].

3. Results and Discussion

Results of radionuclide determinations are shown in Tables 1, 2 and 3, for part of the samples analyzed. Due to space constraints only a few results are shown. In the area of Nelas county, radionuclide concentrations determined in all vegetables and especially in lettuce showed a trend with concentrations increasing down the Pantanha stream from near Urgeiriça to a maximum at Caldas de Felgueira (Table 1). While, for example, ^{238}U in lettuces at the reference station U1 was 51.6±2.0 mBq/kg, at Caldas de Felgueira it was 771±22 mBq/kg (wet weight). This increase goes along with the increase in radionuclide concentrations in bottom sediments and water in the stream. Although radioactive discharges into the stream at Urgeiriça have decreased after implementation of waste water treatment, radionuclide concentrations in the stream materials are still measurable, especially in bottom sediments which will keep a record of past effluent discharges for years. Pumping water and mud from the stream into the kitchen gardens has facilitated radionuclide transfer to vegetables[4,5,6].

In the Cunha Baixa area (Table 2) many agriculture plots have irrigation wells with acidic water contaminated with radionuclides from Cunha Baixa mine seepage. Agriculture plots along the water line from Quinta do Bispo mine also showed contamination although this water was used for irrigation. Soils and vegetables from agriculture plots in these areas all showed radionuclide concentrations enhanced in comparison to values determined at the reference station C1 (Table 2). Common garden products, such as lettuces, displayed higher concentrations near the Cunha Baixa mine. However, apple production in the orchard near Cunha Baixa (#C5) which used as irrigation water a deep aquifer free of radioactive contamination, showed no enhanced radioactivity levels when compared with #C2, confirming water as the main source of radionuclides accumulated in plants[6]. Several km downstream along Ribeira do Castelo (C#5), radionuclide concentrations in soils and agriculture products showed concentrations with no contamination from the mines.

In the Águas Belas-Sortelha valley the agriculture soils along the Quarta-feira stream showed enhanced concentrations from dispersion of waste materials from Vale de Arca mine by surface runoff into the stream (Table

3). The same was observed in some soils around the Bica mine. Agriculture production near these mines displayed higher radionuclide concentrations especially of radium. In vegetable products from Bica area ^{226}Ra was near three times higher than in the reference station and the product displaying the highest radium concentrations was the watercress, near Quarta-feira village (QF#2). Previous research in the area around the Bica mine has indicated contamination of the aquifer and advised continued water treatment to neutralize acidity and precipitate radionuclides [7,8,9].

Table 1. Activity concentrations of radionuclides in soils (Bq/kg dry weight), irrigation water (mBq/L) and vegetables (mBq/kg wet weight) from Ribeira da Pantanha basin (Sites: 1, Algeraz; 2 Valinhos; 3,Vale Escuro; 4,Urgeiriça; 7, Caldas de Felgueira).

Sample	Id.	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Reference Soil	#U1	198±6	9.6±0.9	201±6	456±19	395±46	302±12	302±12	178±9
Water	#U1	70.7±2.1	3.3±0.2	69.9±2.1	1.5±0.6	38.4±2.4	88.6±2.8	12.2±0.5	0.22±0.03
Lettuce	#U1	51.6±2.0	2.1±0.3	52.3±2.0	59.0±14.2	189±12	119.5±5.2	67.4±3.6	17.6±6.9
Lettuce	#U3	87.4±4.5	4.6±0.9	90.5±4.6	48.7±2.5	718±114	171±5	183±5	24.3±1.6
Lettuce	#U4	124±4	38.4±0.7	128±4	94.0±14.4	1754±194	156.3±6.6	53.0±2.6	30.4±7.1
Lettuce	#U7	771±22	35.5±2.9	766±22	227±16	2963±419	706±22	197±9	39.6±4.7
Carrots	#U7	834±33	27.7±4.3	849±34	160±10	1830±181	390±41	253±12	20.5±2.8

Table 2. Activity concentrations of radionuclides in soils (Bq/kg dry weight), irrigation water (mBq/L) and vegetables (mBq/kg wet weight) from Ribeira do Castelo basin (Sites: 1, Mesquitela; 2, Gomes; 3, Aida; 4, Figueiredo; 5, Pisão; 6; Adelino; 7 Qta Carvalhais)

Sample	Id.	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Soil	#C1	314±10	15.0±1.6	316±11	560±24	512±52	567±26	567±26	132±8
Irrig water	#C1	44.4±1.1	2.1±0.1	43.7±1.1	0.86±0.06	37.0±2.8	37.1±1.5	4.7±0.2	0.13±0.02
Lettuce	#C1	437±12	21.4±1.6	462±12	253±17	683±35	527±13	122±4	80.0±6.5
Apple	#C2	45.4±1.8	1.8±0.3	47.1±1.9	24.6±1.4	1627±54	157±5	216±9	3.7±0.5
Lettuce	#C3	3231±136	147±10	2988±126	500±22	1205±178	9141±454	3162±150	28.9±1.7
Apple	#C3	31.5±1.5	1.20±0.4	29.4±1.4	30.0±8.1	1305±70	115±5	668±25	5.2±3.5
Lettuce	#C4	1142±31	50.5±3.8	1100±30	381±23	3359±248	1159±44	931±18	49.9±5.7
Apple	#C5	5.2±0.5	0.26±0.11	6.6±0.6	7.2±0.7	287±16	16±2	107±9	3.6±0.5
Lettuce	#C6	193±8	6.6±1.2	197±8	153±10	3309±226	714±24	294±8	36.2±4.1
Apple	#C6	3.2±0.3	0.14±0.06	4.8±0.4	4.6±0.4	555±30	63.7±6.8	63.9±2.6	1.8±0.3
Lettuce	#C7	803±23	34.6±3.2	758±22	624±34	6260±768	1752±67	2479±122	277±18

Table 3. Activity concentrations of radionuclides in soils (Bq/kg dry wt) and vegetables (mBq/kg wet wt) from Águas Belas-Sortelha valley and Ribeira de Inguias basin (Sites: 1, Águas Belas; 2, Vale de Arca; 4, Bica; 5, Quarta-Feira; 6; Caldeirinhas; 7, Caria).

Sample	Id.	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Soil	#QF1	763±24	37.3±3.2	762±24	666±42	795±64	551±19	551±19	139±10
Lettuce	#QF1	194±6	7.9±0.9	205±6	176±9	460±30	400±30	430±10	75±4
Lettuce	#QF2	158±6	6.1±0.9	154±6	1607±115	8300±1435	346±22	262±19	415±52
Water cress	#QF2	2842±62	133±5	2852±62	904±37	7312±832	2428±67	3734±131	135±7
Lettuce	#QF4	369±9	17.7±1.0	366±9	60.6±4.3	614±106	41.2±1.8	97.0±10.0	25.3±2.2

Lettuce	#QF5	77.1±3.4	1.8±0.4	67.6±3.2	43.1±10.5	671±50	185±15	170±6	15.6±6.5
Lettuce	#QF6	108±4	4.3±0.7	114±5	102±6	3578±415	51.7±2.4	17.8±0.8	35.9±2.6
Lettuce	#QF7	112±5	4.8±09	121±5	83.4±5.0	2042±160	406±29	210±6	42.2±3.4

These results confirm previous reports on these areas, including the fact that not all plants are similar accumulators of radionuclides from water and soil, and some may even preclude root absorption and exclude radioelements. Soil composition may also play a role in concentration of radionuclides by plants [6,10,11].

4. Conclusions

In the three areas investigated, enhanced radionuclide concentrations were measured in soils and agriculture products, especially vegetables from plots near old uranium mines and their waste piles. The increment of radionuclide concentrations varied amongst sites, but the previous work suggested that the vehicle of radionuclidetransfer to plants was the use/dispersion of contaminated mine waste and, in particular, mine drainage[6]. Acid and radioactive mine drainage seeped into the surface aquifer and many irrigation wells, supply contaminated water. Furthermore, surface wells dug near the waterlines receiving mine drainage are recharged with contaminated water from the streams which expands contamination through use in irrigation. In contrast to this, agriculture products grown near mine areas but irrigated with water from deep bore holes showed no radioactive contamination. Also the products from agriculture plots located further away from the mines and grown with clean surface water showed normal (background) radionuclide concentrations. These results suggested that non-contaminated agriculture products may be grown if mine water and drainage from the mines is not used as irrigation water. Once all the mine water discharges will be treated, the quality of surface waters will improve and the transfer of radionuclides to vegetables in agriculture plots might be prevented more efficiently. Nevertheless, guidelines establishing radioactivity limits for several types of water, including irrigation water and soils totally lack in national legislation which renders advice and regulatory control of water and soil use very difficult.

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