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Intake of Radionuclides with the Diet in Uranium Mining Areas

Carvalho FP[a]*, Oliveira JM[a], Malta M[a]

[a] Instituto Superior Técnico/Laboratório de Protecção e Segurança Radiológica, Universidade de Lisboa, Estrada Nacional 10, km 139, Bobadela 2695-066, Portugal

Abstract

To assess the ingestion of radionuclides from uranium mining and milling tailings and other sources related to the legacy of uranium mining, several meals from villages in the region were analyzed. For comparison, it was analyzed a meal prepared with non-local products. Local homemade meals contained much higher radioactivity than the comparison meal. The highest concentrations were consistently those of ^{226}Ra followed by uranium and ^{210}Po. Ingestion of local foods by the most exposed population group (critical group) might exceed the recommended dose limit for members of the public, i.e., being higher than 1 mSv/year above the natural radioactive background.

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Keywords: Uranium mines; uranium waste; radioactivity; agriculture; radionuclide ingestion; radiation dose

1. Introduction

The environmental radiological monitoring of old uranium mine regions is performed annually by the IST/LPSR in order to fulfill European Union Member State obligations derived from the EURATOM Treaty, in particular Articles 35 and 36[1]. The aim of such monitoring program is to keep updated knowledge on environmental radioactivity, to identify undue exposure of population members to ionizing radiation from uranium mining and milling waste, and to identify dispersion and transfer pathways of radionuclides from those wastes in order to abate

* Corresponding author. Tel.: 351 219 946 332; fax: 351 219 550 117.
E-mail address: carvalho@ctn.ist.utl.pt

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radiation exposure of Population. Radioactivity exposure pathways of humans include external radiation, inhalation of radon and dust, and ingestion of radionuclides with the diet. In uranium mining and milling areas the existence of spoil heaps and mine drainage into water streams, may easily lead to accumulation of uranium series radionuclides in vegetables and other agriculture products, thus entering the food chain[2-6]. Although vast surveys of the areas around old uranium mines have been performed for monitoring the dispersal of radionuclides in the environment, the most realistic assessment of the radionuclide intake is the direct measurement of radioactivity in meals cooked by local families. In a preliminary study using this method we collected meals in villages near uranium mine sites and analyzed them for radionuclides. Radionuclide activity concentrations determined were used to compute the radiation dose through ingestion for adults and children.

2. Materials and Methods

The collaboration of several families regularly consuming vegetables and legumes from their kitchen gardens was requested. These were located in the village of Cunha-Baixa, close to the old uranium mine with the same name, and in the village of Caldas de Felgueira, on the banks of Ribeira da Pantanha stream flowing through the Urgeiriça mining and milling area[7]. To the families were given plastic boxes with tight closure for collection of homemade meals. These consisted of vegetable soups, a popular diet item for daily consumption, made with vegetable products from their kitchen gardens. A total of 8 meal samples were obtained, 6 from Cunha Baixa and 2 from Caldas de Felgueira. For comparison, a meal was purchased in a restaurant in the region where food was prepared with products purchased in the supermarket and originated in non-local production. This reference meal was composed of a cabbage soup and a dish of rice, salad and pork meat.

Meals were kept frozen until freeze drying in the laboratory. The resulting dry powder was thoroughly homogenized and sample aliquots were taken for analysis of uranium and thorium isotopes, ^{226}Ra, ^{210}Pb and ^{210}Po. The chemical separation of radioelements and their determination by alpha spectrometry, as well as the analytical quality assurance procedures followed are described in detail elsewhere[8-11].

Activity concentrations of radionuclides are given in Bq/kg (fresh weight). The committed effective radiation doses for adult and children were computed using the concentrations determined in the meals and the activity to dose conversion factors recommended by the International Commission on radiological Protection (ICRP) and adopted by the EU and the IAEA[12,13].

3. Results and Discussion

Results of radionuclide analysis in the meals showed larger concentrations of uranium series (^{238}U) radionuclides than thorium series (^{232}Th) radionuclides (Table 1). The average activity concentration of each radionuclide in these meals may be compared with concentrations determined in the reference meal (Table 2). Generally, radionuclide concentrations in the meals from villages were significantly higher than in the reference meal. For example, on average, ^{226}Ra concentrations were 20 times, and ^{210}Po concentrations were 60 times higher than in the reference meal. In the case of Cunha-Baixa this was due to the use of vegetables produced in kitchen gardens near the mines and mines waste heaps in the preparation of family meals. Kitchen gardens at Cunha-Baixa are located in fields along the water stream receiving discharges of mine drainage. Water from this water stream and from irrigation wells nearby is contaminated with acid seepage from past uranium mining activities and contains relatively high concentrations of dissolved radionuclides, such as uranium and radium[14]. The kitchen gardens at Caldas de Felgueira were irrigated with water from the Ribeira da Pantanha stream, which for many years received drainage and wastewater from Urgeiriça mine and milling area[15].

Radionuclides that were consistently high in all meals were ^{226}Ra, followed by uranium isotopes (^{238}U, ^{235}U, ^{234}U) and ^{210}Po. This ranking of radionuclide accumulation in vegetables is the same as determined in analyzed fresh products from the region, i.e., without cooking[8,15]. Radium-226 in the meals from the villages averaged 5258±6356 mBq/kg (fresh weight) which can be compared with the reference soup with 262±22 mBq/kg fresh weight. The main dish of the reference meal with rice and pork meat was even lower, with 38.3±4.6 mBq/kg. It was also noticed that the meals from Caldas da Felgueira were higher in ^{226}Ra activity concentrations than those of Cunha-Baixa (Table 1). The accumulation of radionuclides from mine drainage, and especially the accumulation of ^{226}Ra by wild plants growing on sludge and agriculture crops had been reported before, and it was related to irrigation with contaminated water[16-18].

Table 1. Activity concentrations of radionuclides (mBq/kg fresh weight) in family meals from villages near uranium mines.

Sample	Origin	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Soup #4	Cunha Baixa	223±7	10.2±1.1	212±7	53.8±6.6	6764±649	172±12	145±9	15.8±3.5
Soup #5	Cunha Baixa	340±11	15.7±1.8	330±11	290±16	4787±308	280±18	174±9	14.2±2.0
Soup #6	Cunha Baixa	266±10	12.4±1.6	285±10	140±22	2367±151	153±8	144±11	23.9±9.8
Soup #7	Cunha Baixa	189±7	8.5±1.2	186±7	63.3±12.5	5187±757	141±13	1255±25	2.4±0.3
Soup #8	Cunha Baixa	10.6±0.7	0.25±0.07	11.7±0.7	88.5±27.1	1578±188	95.0±8.8	39.2±2.3	0.9±0.4
Soup #9	Cunha Baixa	6.8±0.3	0.16±0.04	6.9±0.3	8.1±0.9	136±6	210±16	4.1±0.2	0.16±0.11
Soup #10	Caldas da Felgueira	1624±48	78.5±6.3	1577±47	428±92	1282±64	185±13	170±9	44.8±18.9
Soup #11	Caldas da Felgueira	310±13	21.9±3.3	289±12	138±10	19966±3353	15.2±1.1	7.9±0.6	8.8±2.4
Maximum	-	1624	78.5	1577	428	19966	280	1255	44.8
Minimum	-	6.8	0.16	6.9	8.1	136	15.2	4.1	0.16
Mean	-	371	18	362	151	5258	156	242	14
Standard deviation	-	521	25	506	140	6356	78	415	15

Table 2. Activity concentrations of radionuclides (mBq/kg fresh weight) in a restaurant meal cooked with non-local products.

Sample	Origin	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{210}Po	^{232}Th
Soup	Caldas da Felgueira	12.3±1.0	0.51±0.16	16.2±1.1	255±202	262±24	20.6±1.7	4.1±0.4	<18
Main dish	Caldas da Felgueira	7.4±0.9	0.32±0.09	14.8±1.4	8.0±1.5	38.3±4.6	39.2±5.5	48.1±8.5	1.2±0.5

Assuming a consumption rate of 1 kg/day of soup made with vegetables from the local production, therefore using a conservative scenario, and applying the activity-to-dose conversion factors, the radiation dose computed for the members of this critical group for radionuclides analyzed, might average for a child (2-7 years old) 1.7 mSv/year and reach a maximum of 4.6 mSv/year. For the adult person (>17 years old) the annual average dose from ingestion would be 0.71 mSv/year with a maximum of 2.1 mSv/year. These estimates of the annual dose rate due to the ingestion of local foods are one order of magnitude higher than for the reference meal (Tables 3 and 4).

The largest contribution to radiation dose from the diet comes from the ^{226}Ra in vegetables (Fig. 1). It was noticed also that ^{210}Po concentration in vegetables from villages was higher than in the reference meal and therefore the relative contribution of this radionuclide to dose was higher as well.

4. Conclusions

The determination of radionuclides, mostly of the uranium family, in homemade soups prepared with vegetables and legumes from local production in villages near old uranium mines, showed that local population has higher radionuclide intake than populations at large and must be considered the most exposed (critical group) amongst the general population. This higher intake comes from the transfer of radionuclides originated in uranium waste to vegetables grown in the family kitchen gardens, followed by consumption. Previous work has shown that dispersion of radionuclides with water used for irrigation is the main pathway for the uptake of radionuclides, such as ^{226}Ra, by plants[16, 17]. The computation of the radiation dose due to radionuclides ingested with the diet indicated that there are

Table 3. Effective radiation dose extrapolated to annual basis (mSv/year) for members of the population computed on the basis of the analyzed meals from uranium mine areas.

Sample	Origin	Absorbed radiation dose from ingestion (mSv/year)	
		Age group: 2-7 years	Age group: > 17 years
Soup#4	Cunha Baixa	1.9	0.81
Soup#5	Cunha Baixa	1.6	0.67
Soup #6	Cunha Baixa	0.93	0.37
Soup #7	Cunha Baixa	3.3	1.1
Soup #8	Cunha Baixa	0.51	0.21
Soup #9	Cunha Baixa	0.21	0.069
Soup #10	Caldas da Felgueira	0.87	0.34
Soup #11	Caldas da Felgueira	4.6	2.1
Maximum	-	4.6	2.1
Minimum	-	0.21	0.069
Mean	-	1.7	0.71
Standard deviation		1.5	0.65

critical groups that may receive through ingestion an annual effective dose exceeding the maximum recommended limit i.e., 1 mSv/year from radionuclides and exposures related to practices.

Table 4. Effective radiation dose extrapolated to annual basis (mSv/year) for members of the public computed on the basis of the comparison meal made with non-local products (restaurant).

Sample	Origin	Absorbed radiation dose from ingestion (mSv/year)	
		Age group: 2-7 years	Age group: > 17 years
Soup	Restaurant	0.09	0.036
Main dish	Restaurant	0.12	0.04

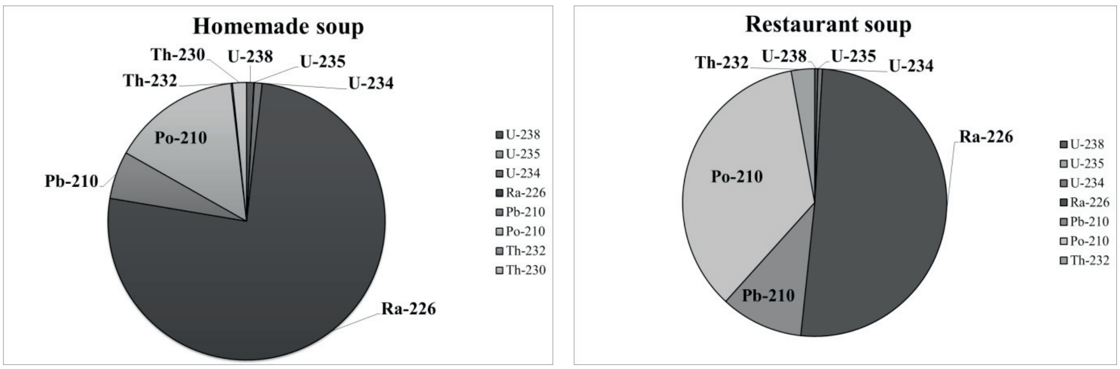

Fig. 1. Relative contribution of radionuclides to radioactivity determined in soups prepared with vegetables from different origins.

This preliminary study was based on a small number of meals collected in villages of the uranium region, and a larger investigation on local diets and encompassing more villages is now underway. Notwithstanding, these findings advice focused attention on mine drainage and irrigation water, including specific measures to control the quality of irrigation water in order to abate radionuclide accumulation in agriculture products and to reduce human exposure through ingestion of agriculture products.

References

1. EURATOM Treaty at http://eur-lex.europa.eu/en/treaties/dat/12006A/12006A.htm (accessed 9-3-2014).
2. Carvalho FP. Past uranium mining in Portugal: legacy, environmental remediation and radioactivity monitoring. In: IAEA. *The Uranium Mining Remediation Exchange Group (UMREG). Selected Papers 1995–2007*. Vienna, STI/PUB/1524; 2011. p. 145-155.
3. Carvalho FP, Oliveira JM, Libânio A, Lopes I, Ferrador G, Madruga JM. Radioactivity in Public Water Supplies in the Uranium Mining Regions in Portugal. In: AIEA. *Proceed. of an International Workshop on Environmental Contamination from Uranium Production Facilities and Remediation Measures,* held in Lisbon 11-13 Feb 2004. Vienna; 2005. p. 41-51.
4. Carvalho FP, Madruga JM, Reis MC, Alves JG, Oliveira JM, Gouveia J, Silva L. Radioactive survey in former uranium mining areas in Portugal. In: AIEA. *Proceed. of an International Workshop on Environmental Contamination from Uranium Production Facilities and Remediation Measures,* held in Lisbon 11-13 Feb 2004. Vienna; 2005. p. 29-40.
5. Carvalho FP, Madruga MJ, Reis MC, Alves JG, Oliveira JM, Gouveia J, Silva L. Radioactivity in the environment around past radium and uranium mining sites of Portugal. *J Environ Radioact* 2007; **96**: 39-46.
6. Carvalho FP, Oliveira JM, Lopes I, Batista A. Radio nuclides from past uranium mining in rivers of Portugal. *J Environ Radioact* 2007; **98**: 298-314.
7. Carvalho FP, Oliveira JM, Malta M. Radioactivity in Iberian Rivers with Uranium Mining Activities in their Catchment Areas. *Procedia Earth and Planetary Science* 2014 (submitted in this volume).
8. Carvalho FP, Oliveira JM. Alpha emitters from uranium mining in the environment. *J Radioanal Nucl Chem* 2007; **274**: 167-174.
9. Oliveira JM, Carvalho FP. A Sequential Extraction Procedure for Determination of Uranium, Thorium, Radium, Lead and Polonium Radionuclides by Alpha Spectrometry in Environmental Samples. In: Proceedings of the 15th Radiochemical Conference. *Czechoslovak Journal of Physics* 2006; **56** (Suppl. D): 545-555.
10. Carvalho FP, Oliveira JM. Performance of alpha spectrometry in the analysis of uranium isotopes in environmental and nuclear materials. *J Radioanal Nucl Chem* 2009; **281**: 591-596.
11. Pham MK, Benmansour M, Carvalho FP, Chamizo E, Degering D, Engeler C. Certified Reference Material IAEA-446 for radionuclides in Baltic Sea seaweed. *Appl. Radiat. Isot.* 2013 (Available online 20 November 2013).
12. ICRP. Age-dependent Doses to Members of the Public from Intake of Radionuclides, Part 5, Compilation of Ingestion and Inhalation Dose Coefficients. ICRP Publication Nº 72, Ann. ICRP 26, Elsevier Science, Oxford, UK; 1996.
13. COUNCIL DIRECTIVE 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation.
14. Carvalho FP, Oliveira JM, Faria I. Alpha Emitting Radionuclides in Drainage from Quinta do Bispo and Cunha Baixa Uranium Mines (Portugal) and Associated Radiotoxicological Risk. *Bull Environ Contam Toxicol* 2009; **83**: 668-673.
15. Carvalho FP, Oliveira JM, Malta M. Analyses of radionuclides in soil, water and agriculture products near the Urgeiriça uranium mine in Portugal. *J Radioanal Nucl Chem* 2009; **281**: 479-484.
16. Carvalho FP, Oliveira JM, Neves MO, Abreu MM, Vicente EM. Soil to plant (*Solanum tuberosum L.*) radionuclide transfer in the vicinity of an old uranium mine. *Geochem: Explor Environ, Anal* 2009; **9**: 275-278.
17. Carvalho FP, Oliveira JM, Malta M. Radionuclides in plants growing on sludge and water from uranium mine water treatment. *Ecol Eng* 2010; **37**:1058-106.
18. Carvalho FP. Environmental Health Risk from Past Uranium Mining and Milling Activities. In: C.A. Brebbia, editors. *Environmental Health Risk IV*. UK; 2007. p 107-114.