Agriculture and soil-to-plant radionuclide transfer in the surroundings of an old uranium mine

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INTRODUCTION

Uranium mining and uranium ore treatment were carried out at the mine of Cunha Baixa, near the city of Viseu in the North of Portugal, from 1967 till 1991. Uranium ore was initially extracted by underground works and in the latest years by the open pit method. The mining and milling wastes were deposited on the surface, near the old mine, and remain uncovered. Sulphuric acid was used to leach uranium from the rock by the heap leaching method on the surface and by underground in situ leaching. After closure of the mine, and up to today, acid mine waters have been continuously treated to neutralise the acid and to remove dissolved radium. Sludge from acid mine water treatment have been dumped into the open pit. Radionuclides in several of these materials were analyzed before (Carvalho et al., 1996).
As the mine is located very close to the village of Cunha Baixa and inhabitants grow vegetables and fruits for their own supply and as livestock feed, radioactivity was determined in soils, irrigation water, and horticulture products as part of a radioactivity assessment of this area.

MATERIALS AND METHODS

Sampling was carried out in May and July 2007 in the area of Cunha Baixa and Quinta do Bispo mines, including the agriculture zone located between these two mines and drained by Castelo Stream, a small river tributary to the Mondego River (Figure 1).
Water samples were collected in irrigation wells and in the Castelo Stream. Samples of surface soils and horticulture products were collected as available, pre treated at the village, and transported to the laboratory for radioactivity analyses
Water physical chemical parameters were measured *in situ* with a portable multiparameter probe Horiba U-22. Water for radionuclide analyses were filtered through 0.45 μm pore size large diameter membrane filters. Filtered water was acidified to pH<2 and filters saved for drying, determination of suspended load and analyses of radionuclides in the particulate fraction. Soils (20 cm upper layer) were collected with a stainless steel spoon, from 4 points in the same garden, combined, and stored in a plastic box for transportation. In the laboratory, soils were dried in the oven to constant weight, sieved and the fine fraction with grain size less than 63 μm retained for radionuclide analysis. Horticulture products were transported in ice chests and prepared as for consumption, *i.e*, lettuce leaves washed, fruits and cucumbers pealed off. Edible portions were saved, freeze dried, homogenized to powder and aliquots used for radionuclide analyses.

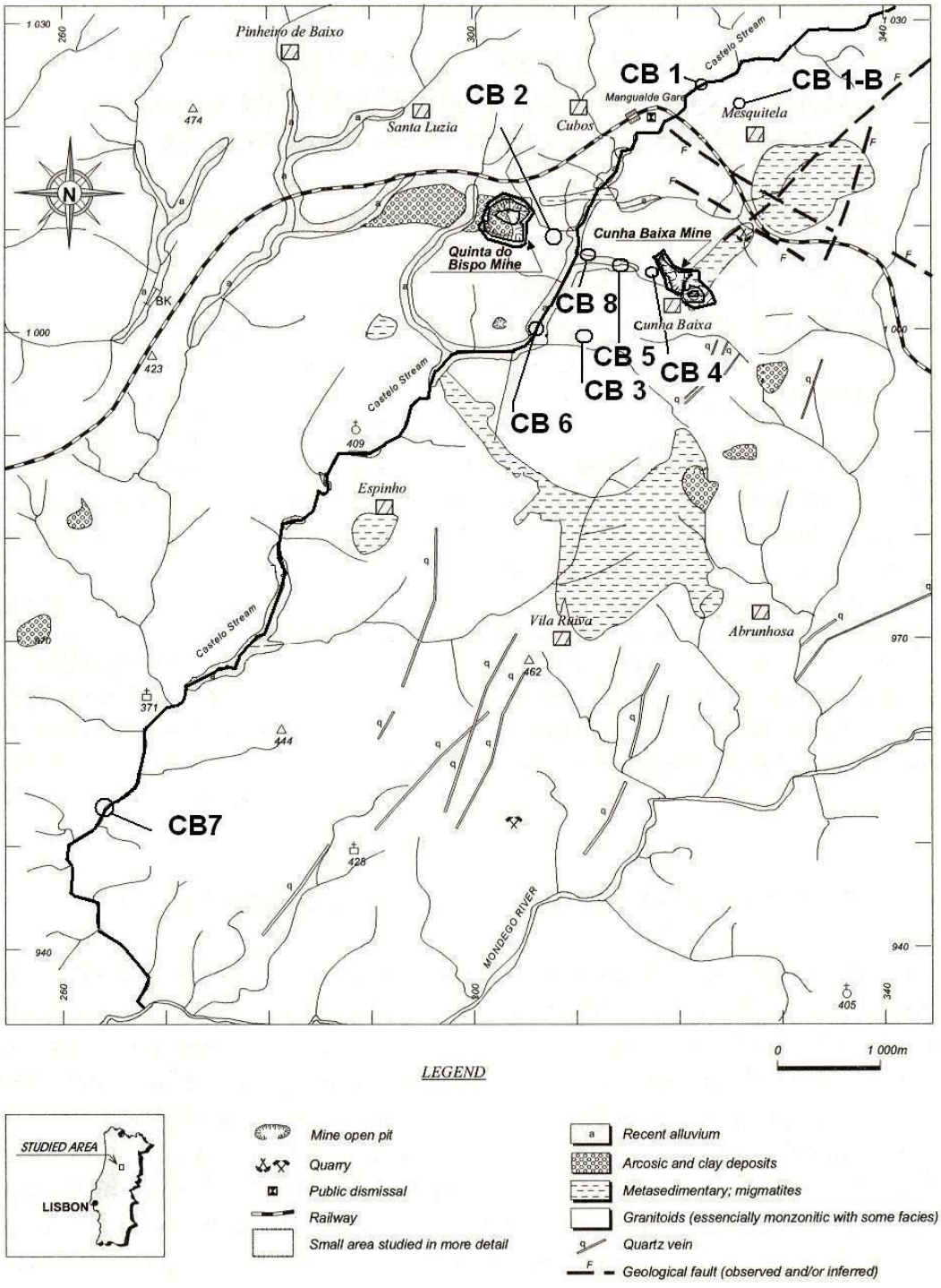

Figure 1: *Location of the old uranium mines of Cunha Baixa and Quinta do Bispo, with sampling stations*.

All radionuclide analyses were performed using internal isotopic tracers and radionuclides separated and purified by radiochemical methods prior to electroplating onto stainless steel and silver discs, as described elsewhere (Oliveira and Carvalho, 2006). Alpha sources were

measured in vacuum chambers using large surface ion implanted detectors with 450 mm^2 active surface (OCTECTEPlus, ORTEC EG&G). Analytical quality control was performed by analyses of certified reference materials and participation in international intercomparison exercises organized by the IAEA and the EU (Oliveira and Carvalho, 2006).

RESULTS AND DISCUSSION

Table 1 shows the results for the main soluble radinuclides in water from Castelo Stream, *i.e.*, CB1, CB6, CB7. Station CB1, located upstream the uranium mine areas and mine waste discharges was selected as a reference site (Figure 1). Results for downstream stations showed no significant enhancement of radionuclide concentrations in this river. These samples were collected during a period of no mine water discharges (treated or untreated) into the stream and, thus, results contrast with situations of high radionuclide concentrations measured before (Carvalho et al., 2006).

Table 1. *Radionuclide concentrations in the soluble phase (Bq m^{-3} ± 1SD) of river water and water from wells in the area of Cunha Baixa.*

Sample	^{238}U		^{235}U		^{234}U		^{230}Th		^{226}Ra		^{210}Po		^{232}Th	
CB1 river	22.0	± 0.6	0.93	±0.08	21.9	±0.6	4.0	±0.4	23.6	±1.6	8.0	±0.4	0.20	±0.08
CB6 river	32.4	± 1.1	1.6	±0.2	32.3	±1.1	2.6	±0.2	22.6	±3.1	22.9	±0.9	0.34	±0.07
CB7 river	56.5	±1.7	3	±0.2	58.4	±1.8	2.4	±0.2	26.7	±5.7	8.8	±0.4	0.27	±0.05
CB1-B well	173	± 4	7.6	±0.4	167	±4	2.7	±0.2	124	±8	27.5	±0.7	0.24	±0.05
CB2 well	405	± 16	18.6	±1.2	391	±16	2.2	±0.2	83.5	±9.6	15.2	±0.7	0.41	±0.08
CB3 well	23.7	± 1.0	0.9	±0.1	24.0	±1.0	2.3	±0.2	40.0	±2.6	15.1	±0.7	0.40	±0.10
CB4 well	1701	± 77	81.8	±4.5	1581	±72	3.1	±0.2	171	±10	34.0	±1.3	0.39	±0.08
CB5 well	4176	± 280	212	±16	3892	±261	3.6	±0.3	184	±40	50.6	±6.6	0.34	±0.08
CB8 well	37.5	± 1.2	1.8	±0.2	38.9	±1.2	0.5	±0.1	85.6	±15	5.6	±0.2	0.17	±0.07

CB2 water is from a well under the influence of mine water discharge from Quinta do Bispo mine, and radionuclide levels were about 2.3 times higher for uranium, but 0.7 times only for radium, in comparison with CB1-B reference levels. Other wells at the left river bank, near Cunha Baixa mine (*i.e.,* CB3, CB4, CB5 and CB8) are impacted in variable degree by acid mine water from Cunha Baixa mine. The enhancement of radioactivity in water from wells in this area may be noticed particularly in results for CB4 and CB5 waters. For example, ^{238}U in CB5 water is 24 times higher than in water from the reference well. All these wells are intensively used for irrigation of horticulture plots and orchards.

Together with soils and water from wells, several horticulture products and fruits were analyzed. Table 2 shows the results for lettuce, one of the most common products grown in these plots. Station CB1-B was the reference sample (baseline) to compare with samples from the horticulture zone near the mine area. Products irrigated with water containing enhanced levels of radioactivity, displayed enhanced concentrations of uranium, ^{230}Th, ^{226}Ra and ^{210}Po when compared with CB1-B. Though radionuclide concentrations in soil may vary also in the region, the main source of radionuclides to lettuce seems to be the irrigation water. Concentrations measured in fruits and potatoes, especially when the peel was removed before analysis, were lower than those measured in lettuce.

Radionuclides accumulated in these vegetables might be transferred to humans and to cattle through consumption of these horticulture products.

***Table 2**. Radionuclide concentrations in lettuce leaves (Bq kg^{-1} dry weight ± 1SD) from the area of Cunha Baixa, May 2007. Average dry:wet weight ratio: 0.036.*

Station	^{238}U		^{234}U		^{230}Th		^{226}Ra		^{210}Po		^{232}Th	
CB1-B	0.52	±0.02	0.53	±0.02	0.49	±0.02	1.7	±0.1	2.10	±0.08	0.092	±0.007
CB2	4.2	±0.1	4.0	±0.1	3.7	±0.1	5.7	±0.4	10.1	±0.4	1.26	±0.05
CB3	0.54	±0.02	0.56	0.03	0.39	±0.03	3.0	±0.4	4.7	±0.3	0.28	±0.02
CB4	154	±4	152	±4	86	±4	100	±6	101	±4	2.0	±0.1
CB5	13.1	±0.4	12.5	±0.4	5.7	±0.3	78	±7	8.0	±0.3	0.97	±0.08

CONCLUSIONS

Radionuclides dissolved in the water of Castelo Stream were at background concentration levels due to temporary suspension of acid mine water discharges from Cunha Baixa mine. This indicates that radioactivity levels in surface waters may return rapidly to normal after a suitable treatment of mine water discharges allowing for ecosystems to recover rapidly from acid and radioactive drainage.

In the agriculture area surrounding the old mines of Cunha Baixa and Quinta do Bispo, especially in the flood plain of Castelo Stream, the water from wells used for irrigation contains enhanced concentrations of radionuclides belonging to uranium series. These enhanced concentrations are a consequence of the use of sulphuric acid for *in situ* uranium leaching in the mine, which allowed the acid to reach the aquifer and spread in groundwater. Radionuclides in groundwater of the shallow aquifer, both in suspended particles and in solution, are a function of water acidity (Carvalho et al., 2006). Water from irrigation wells transfer part of these radionuclides to top soil and to plants grown in the area. Fruits (apples) have practically no enhanced radionuclide concentrations. Potatoes grown with these waters have slightly enhanced concentrations, especially of ^{226}Ra, but about 50% were in the potatoes peel and may be removed by peeling (Carvalho et al., 2008). However, leafy vegetables, such as lettuce, showed an important increase in radium and uranium concentrations that is more significant than in other products. A more detailed control of radioactivity in the diet based on locally grown products is recommended in order to assess the radiation dose through ingestion to the population.

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