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Radioecological characterization of a uranium mining site located in a semi-arid region in Brazil

Horst M. Fernandes [a,b,*], F. Fernando Lamego Simoes Filho [a],
Valeska Perez [c], Mariza Ramalho Franklin [a],
Luiz Alberto Gomiero [d]

[a] *Instituto de Radioprotecao e Dosimetria, Environmental Impact Assessment,*
Av. Salvador Allende s/n – Recreio, 22780-160,
Rio de Janeiro, Brazil
[b] *University of Central Florida*
[c] *Instituto de Engenharia Nuclear*
[d] *Industrias Nucleares do Brasil*

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Abstract

The work presents the radioecological characterization of the new Brazilian uranium mining and milling site located in a semi-arid region of the country. The process characterization demonstrated that in heap leach plants most of the ^{226}Ra remains in the leached ore. Despite the potential higher availability of radium isotopes in the soils of the studied region the lack of precipitation in that area reduces the leaching/mobilization of the radionuclides. High ^{226}Ra and ^{228}Ra concentrations were found in manioc while ^{210}Pb was significant in pasture. It was suggested that a range from 10^{-3} to 10^{-1} may conveniently encompass most of the transfer factors (TF) values for soil/plant systems (i.e. involving different cultures, different soils and natural radionuclides). Impacts due to aerial transportation of aerosols and radon generated in the mining were proved to be minimal and restricted to an area not greater than 15 km^2.

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[*] Corresponding author. Institute of Radiation Protection and Dosimetry, Environmental Impact Assessment Section, Av. Salvador Allende s/n – Recreio, 22780-160, Rio de Janeiro, Brazil. Tel.: +55 21 34118087; fax: +55 21 24422699.

E-mail address: monkenhorst@yahoo.com.br (H.M. Fernandes).

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Finally, uranium complexation by carbonates was shown to be the main mechanism responding for the elevated radionuclide concentration in groundwater.

Keywords: Uranium mining; Semi-arid region; Natural radionuclides

1. Introduction

It is well reported in the literature that the operation of uranium production centers has the potential to increase exposure of members of the public to natural radiation through different pathways.

Uranium mining involves the removal from the ground of large quantities of ore containing uranium and its decay products. Underground and open-pit mining are the main techniques. It is reported that underground mines accounted for 40% of the world's total uranium production in 1996 and open-pit mines, 39%. Uranium is also mined using in situ leaching, which produced 13% of the world's uranium in 1996; the remaining 8% was recovered as a byproduct of other mineral processing (BRGM, 2001).

Milling operations involve the processing of the ore to extract the uranium in a partially refined form, known as yellowcake. Around the world, 115 uranium mining and milling operations are conducted in several countries. In 1997 about 90% of world uranium production took place in nine countries: Australia, Canada, Kazakhstan, Namibia, Niger, the Russian Federation, South Africa, the United States, and Uzbekistan.

The resulting exposure to ionizing radiation from mining and milling operations will depend upon the type of the plant operation; specifically, the way effluents are released into the environment and also on social-economical habits of the exposed population. UNSCEAR (2000) recognizes that there are few new data on releases of radionuclides into the environment from mining and milling operations and most of the attention has been focussed during the past decades to the emissions of radon into the atmosphere. However, little attention has been paid on releases into superficial water and underground water bodies.

It is also believed (see UNSCEAR, 2000) that for mining operations in arid areas, liquid effluents are minimal, and radionuclide releases via this pathway are estimated to be of little consequence.

Based on available data, the radon emission rates were estimated in UNSCEAR (1993) to be 10 Bq s^{-1} m^{-2} of tailings during the operational phase of the mill and 3 Bq s^{-1} m^{-2} from abandoned but stabilized tailings (assumed period of unchanged release of 10,000 years). Liquid emissions into groundwater and superficial water bodies may lead to high exposure levels to members of the public (Fernandes et al., 1996). In addition to tailings dam, waste rock piles can constitute an important source of pollutants to the environment, especially if acid drainage generation takes place (Amaral et al., 1988; Fernandes et al., 1998; Fernandes and Franklin, 2001).

In Brazil, the first uranium production center (the Poços de Caldas Project) ceased operations in 1997 and is about to be decommissioned. The uranium production now takes place at the production center of Caetite, located in a semi-arid region for which no environmental information has been made available in the literature so far. Thus, the objective of the present work is to present a radioecological characterization of the neighboring environment of the new Brazilian uranium mining and milling site and discuss the potential radiological environmental impacts associated with that particular uranium project. This paper also intends to call the attention of operators to the importance of carrying on detailed characterization of site-specific socio-economical aspects and operational conditions to improve their environmental performance.

2. Description of the study site and operation characteristics

2.1. Environmental and socio-economical characterization

The Uranium Production Center of Caetité (URA) is located in the center-southwestern region of Bahia state (13°56′36″ S and 42°15′32″W) close to the municipality of Caetité which has 45,000 inhabitants (half of whom live in rural areas) (see Fig. 1).

The climate is tropical, warm and humid with annual average precipitation rate of 800 mm (almost 70% of this amount is recorded in the period between December and March). The average temperature is 22 °C and the predominant wind direction is southwest. The region is characterized as semi-arid. Although perennial rivers do not exist in the area, during the rainy seasons these rivers present high flow rates.

The area under the influence of the mining and milling site comprises a circumference with a radii of 20 km centered in the plant. It is estimated that approximately 1000 families live within this area. Groundwater is the main water supply for the local population; however some of the wells present saline waters often with high uranium concentrations. Agricultural activities comprise the production of manioc, corn, sugar cane and black beans. Table 1 summarizes the agricultural production in the area of direct influence of the uranium plant. In addition to this production, cattle are also raised along with pork and poultry.

2.2. Site and plant characterization

The geological content of the ore is 3000 ppm and the average thickness of the ore body is approximately 7 m. The uraniferous mineralization is associated with albitites ($NaAlSi_8O_8$). Thirty-five anomalies were identified in an area of approximately 1200 km^2 (Gomiero et al.,

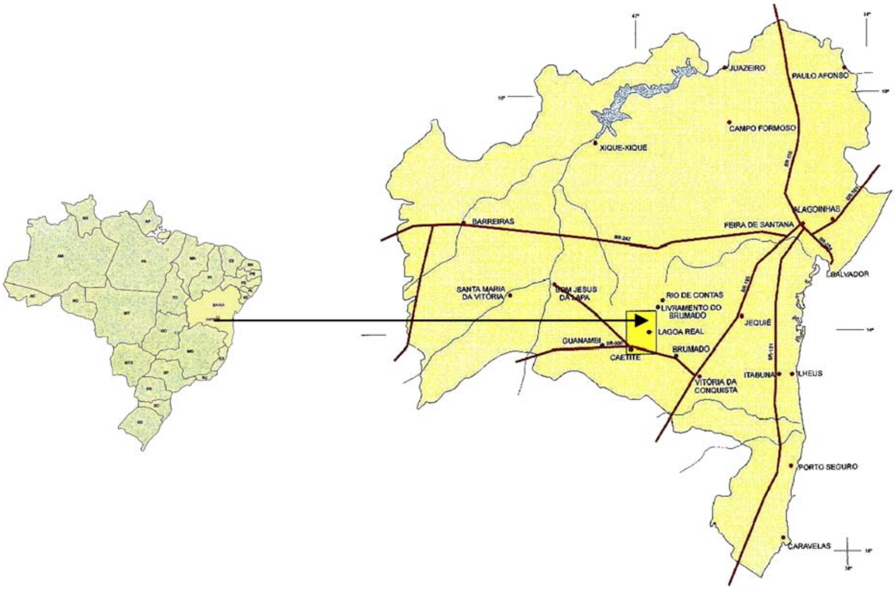

Fig. 1. Localization of the study area.

Table 1
Survey of agricultural production at farms located in the area under potential influence of the uranium plant

Products	Production (t $year^{-1}$)
Black beans	98.9
Corn	327.6
Manioc powder	455.2
Sugar Cane	124.8
Mango	74.5×10^3 (unities)
Banana	37.7×10^3 (racemes)

Source: INB (1999).

2004). The average composition of the rocks is presented in Table 2. Albitite (70%) is the predominant mineral in the rock composition, followed by pyroxene (10–20%), garnet (2–5%) and magnetite (1–3%).

The extraction of uranium from the ore is achieved by means of a heap leach process. The process is represented in Fig. 2. After the ore crushing and the addition of sulfuric acid in the run-of-mine, in the proportion of 1.5 $kg_{H_2SO_4}\ t_{ore}^{-1}$, piles containing between 25,000 and 35,000 tons of ore are formed. Each pile is submitted to three washing cycles:

(1) acid leaching step: with a 25 g l^{-1} H_2SO_4 solution, in the ratio of 0.6 $m^3\ t_{ore}^{-1}$. The solution is dispensed by means of perforated tubes at a rate of 30 L $h^{-1}\ m^{-2}$.
(2) washing: with another H_2SO_4 solution (5 g L^{-1}) in the ratio of 0.3 $m^3\ t_{ore}^{-1}$, at the same percolation rate.
(3) washing: with raw water, in the ratio of 0.3 $m^3\ t_{ore}^{-1}$, keeping the same percolation rate.

The resulting solutions are captured in ponds where they are mixed in such a way that a feeding solution 2 g l^{-1} of uranium is achieved. Uranium is extracted from the feed solution by means of an organic solvent that consists of 7% of a tertiary amine, 3% of tridecanol and 90% of kerosene. Uranium is re-extracted with NaCl being subsequently precipitated by the addition of ammonium hydroxide solution. The liquid effluent generated in the milling plant consists of an acid suspension with high concentrations of other elements that are dissolved along with uranium, such as Al, Fe, Ca, Mg and Si. This acid effluent is neutralized with CaO to pH 8.0. The slurry is pumped to ponds, provided with sub-aerial drains that allow for the sedimentation of the solids in suspension. The overlying solution is re-circulated to the process.

Table 2
Ore composition in the Caetité mining site

Element	Concentration (%)	Element	Concentration (%)
SiO_2	43.1 ± 1.3	BaO	0.16 ± 0.05
CaO	6.8 ± 1.1	ZrO_2	0.04 ± 0.01
Al_2O_3	13.8 ± 0.8	ZnO	0.011 ± 0.002
Fe_2O_3	5.4 ± 0.8	SnO_2	0.0056 ± 0.06
TiO_2	2.4 ± 0.4	Cr_2O_3	0.0056 ± 0.0005
U_3O_8	0.35 ± 0.009	MnO	0.094 ± 0.001
MgO	2.4 ± 0.4	Nb_2O_5	0.1 ± 0.02
V_2O_5	0.1 ± 0.02		

Source: INB (1999).

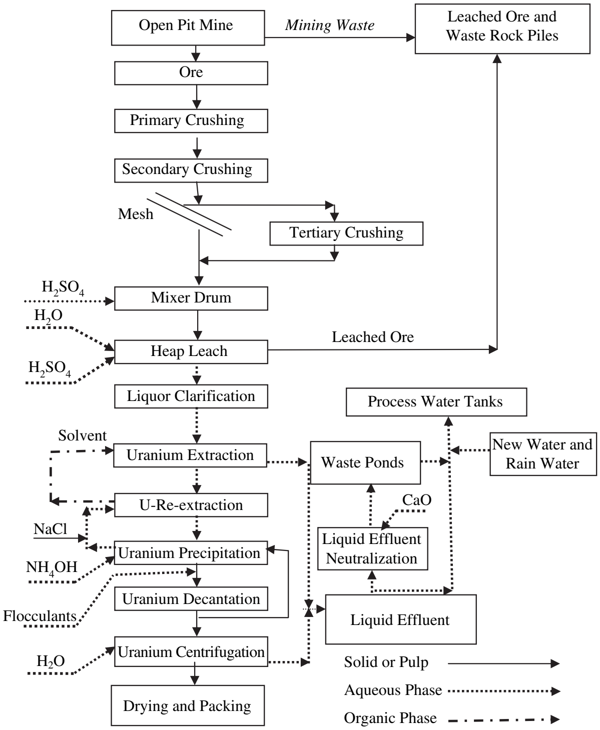

Fig. 2. Operational flowchart of the Caetité uranium production plant.

The resulting wastes of the overall mining and milling processes can be divided into wastes from the mining operations (waste rock and drainage waters) and wastes from the industrial process (leached ore and effluents from the milling plant).

From the results presented in Table 3 it can be seen that while uranium is removed to a large extent from the ore in the sulfuric leaching (85%), Ra-isotopes are not. The main implication of these findings is that the leached ore will constitute an important source of Ra in the long term.

3. Materials and methods

Soil, aerosol and vegetable samples were collected in five different sampling stations. Radon detectors (track etch) were placed for the measurement of environmental radon at these same locations.

Table 3
Radiological characterization of the materials coming from the leaching circuit

Material	^{238}U	^{226}Ra	^{228}Ra
Ore (Bq kg^{-1})	53080 ± 11975	33280 ± 9549	83.0 ± 21.0
Leached ore (Bq $kg^{-1})_{seco}$	7582 ± 3290	34520 ± 9512	75.00 ± 12
Leaching solution (Bq l^{-1})	157400 ± 30770	23.0 ± 4.0	6.0 ± 0.5
1st washing water (Bq l^{-1})	54840 ± 13162	16 ± 4.5	2.3 ± 0.6
2nd washing water (Bq l^{-1})	526 ± 176	10.0 ± 3.0	<0.5

Source: INB (1999).

Additionally, radon and aerosol sampling stations were fixed in the mining and milling area (stations INST 001 and INST 008).

Fig. 3 shows the map with the environmental aerosol and radon sampling stations. The dominant wind directions are (ESE) (52%), (SE) (26%) and (E) (8%). Because of this the preferential impact sectors due to aerial emissions from the mining and milling plant are (WNW), (W) and (NW). As a result it can be seen that stations AERO 001, AERO 008, AERO 010 and AERO 013 are located within the preferential impact sectors. On the other hand, station AERO 42 is intended to represent a background area in respect of the aerial emissions from the mining and milling site.

In each sampling point soil samples were taken in three different depths, i.e. 0–5, 5–10 and 10–20 cm to check for possible vertical variations. Twenty-four samples, 2 kg each were obtained by the end of the sampling campaign. In the laboratory samples were homogenized. For the aerosol sampling high volume samplers (HIVOL-AGV-PTS type) were used. Samples were collected every three consecutive months yielding a total period of 240 h (about 24,000 m^3) of sampling. Vegetable samples were washed and peeled, when necessary, dried in air and weighed for determination of the fresh mass. After that, they were oven dried at 80 °C for approximately 16 h. The samples were then roasted for 24 h in an oven at 450 °C. The ashes were then digested with a mixture of strong acids (HCl, $HClO_4$ and HF).

Water samples from the aquifers were filtered with 0.45 μm Millipore filters and acidified with HNO_3 prior to the analysis.

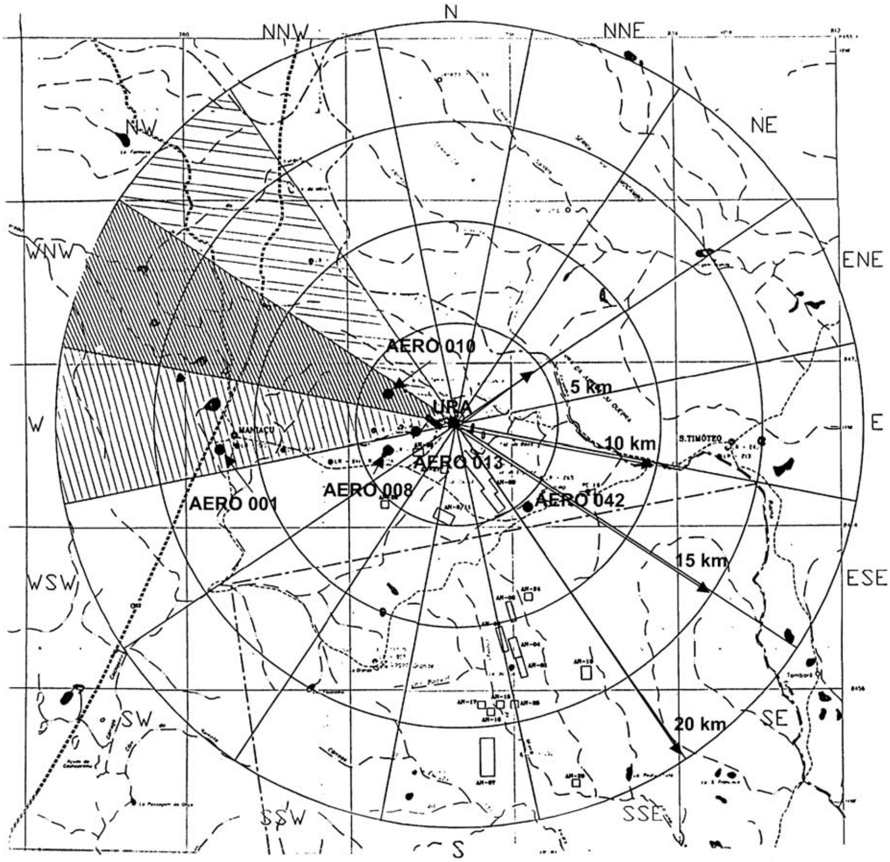

Fig. 3. Location of the sampling stations for soil, aerosols, radon and groundwaters.

Radium and lead determinations were performed by total alpha and beta counting according to Godoy et al. (1994). Radium isotopes were co-precipitated as $Ba(Ra,Pb)SO_4$, by adding H_2SO_4 and $BaCl_2$ to the sample solution. The mixed sulfates were washed and dissolved with nitrilotriacetic acid (NTA). Barium (radium) sulfate was re-precipitated by adding acetic acid until pH 4.5–5.0, whereas lead remained in the aqueous phase. The aqueous and solid phases were separated and the $Ba(Ra)SO_4$ was purified by dissolution in an ethylene diamine tetra-acetic acid (EDTA) solution at pH 10. Then the sulfate mixture was re-precipitated by acetic acid (pH 4.5–5.0). The precipitate was filtrated and, after setting aside for 1 month for in growth of ^{222}Rn and its short-lived progeny, ^{226}Ra was determined by gross alpha counting. ^{228}Ra was determined by beta counting, whereas the alpha particles of ^{226}Ra were stopped by a filter paper. Corrections were made if there was any contribution from the daughter products of ^{226}Ra. Lead was precipitated from the NTA solution as PbS. After elimination of the sulfur by nitric acid addition, lead was precipitated as $PbCrO_4$ from the solution at pH 4.5–5.0. About 2 weeks later, ^{210}Pb was determined through its daughter product ^{210}Bi by beta counting. Alpha and beta activities were counted in a low-background anti-coincidence proportional detector (Bertold, model LB770-1). The chemical yield of the method was approximately 85% for radium and lead and the relative standard deviation for individual activities was about 10%. The minimum detectable activities, with the counting time of 100 min for alpha and 400 min for beta, 95% confidence level, were: ^{226}Ra, 5 mBq; ^{228}Ra, 90 mBq; and ^{210}Pb, 16 mBq.

To determine ^{238}U and ^{232}Th, the samples were dissolved with 65% nitric acid (Merck Suprapur). After 72 h of digestion, the samples were then diluted (1:20) with ultra pure water with a specific resistivity of 18 MΩ cm obtained by filtering double-distilled water through a Milli-Q (Millipore). Indium and thallium were added as internal standards to all samples, blank samples and standard solutions, in such a way as to produce a final solution containing 20 ng ml^{-1} of each one. The analyses were performed by inductively coupled plasma–mass spectrometry (ICP–MS) on a Perkin–Elmer SCIEX ELAN 6000 instrument equipped with the standard spray chamber and cross-flow nebulizer, applying the quantitative method and using 1, 5, 10, 15 and 20 ng ml^{-1} solutions of the Perkin–Elmer standard solutions. Determination of the element concentrations was performed with the major isotope ^{232}Th and ^{238}U. Major and trace metals in groundwater were also measured by inductively coupled plasma–mass spectrometry (ICP–MS).

The procedure for the determination of radon was the same as that described in Binns et al. (1998).

4. Results and discussion

4.1. Radionuclides in soil

The alkaline complex of Poços de Caldas is a volcanic round structure of approximately 35 km in diameter, in which the radionuclides are found homogeneously distributed. These soils derive from the volcanic magma spreading that covered the whole region and uplifted the tableland 400–500 m high over a pre-exiting substrate (granite). In the Caetité region, there was no volcanic spreading and ^{238}U mineralization took place in rock fractures located in the prevailing rock bodies due to the process of albitization, as a consequence of the magma intrusion derived from tectonic shield processes.

The activity concentrations of the analyzed radionuclides are shown in Table 4. No significant differences in activity concentrations of soil samples collected in the different depths (soil layer) were observed. Because of this, we chose to report the results as average values of the concentrations recorded in the three layers. It is remarkable the higher ^{228}Ra concentrations in comparison to ^{226}Ra concentrations, being this finding in good agreement with data published by Linsalata et al. (1991).

It can be assumed that ^{238}U and ^{226}Ra are in equilibrium (average ^{226}Ra/^{238}U ratio equal to 1.05 ± 0.17), however the average ^{210}Pb/^{238}U ratio was 2.99 ± 1.03 showing that ^{210}Pb concentrations in soil are consistently higher than those of the parent radionuclide; this finding is

Table 4
Radionuclide activity concentrations in soil

Sampling station	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
A1					
TC	180	143	156	–	99.4
RAC	28	18	3.0	10	23
A2					
TC	46.5	43.4	158	–	87.6
RAC	9	5.0	7.0	9.0	22
B1					
TC	41.9	41.1	154	84.8	62.5
RAC	4	5.0	4.0	4.0	11
B10					
TC	70.5	53.2	203	92.9	98.0
RAV	7	8.0	3.0	5.0	13
C					
TC	68.4	87.0	140	74	75.6
RAC	10	9.0	4.0	48	22
D					
TC	53.0	50.0	204	99.0	89.7
RAC	5	6.0	3.0	2.0	20
E1					
TC	41.6	48.3	149	–	82.0
RAC	4	6.0	3.0	3.0	14
E2					
TC	48.3	47.0	175	–	59.6
RAV	5	5.0	3.0	3.0	12
AVG					
TC	68.7 ± 45.0	62.6 ± 32.6	173 ± 42.4	87.7 ± 10.8	80.8 ± 19.6
RAV	9.0 ± 8.0	8.0 ± 4.0	4.0 ± 1.0	8.0 ± 15	17 ± 5.0
Pocos de Caldas	184 ± 107	148 ± 103	135 ± 101	359 ± 85.6	284 ± 97.6

TC, total concentration; RAC, readily available concentration; in Bq kg^{-1}. Each result represents the average of the concentration in the three different soil layers (0–5; 5–10; 10–20). Each concentration in one soil layer is a result of duplicate analysis. Results of Poços de Caldas from Amaral et al. (1992). Locations A1 and A2 relative to AERO 008; B1 to AERO 010; C to AERO 013; D to AERO 42.

caused by the dry and wet deposition of ^{210}Pb onto the surface of the soils. ^{228}Ra and ^{232}Th are in close equilibrium with an average ^{226}Ra/^{232}Th ratio of 0.92 ± 0.14.

In comparison to data of the Poços de Caldas mining site (see Amaral et al., 1992) it can be seen that the results from Caetité area are consistently lower, especially for ^{232}Th and ^{228}Ra which can be explained by the geological evolution of both systems as explained above. Interesting to observe that the ^{210}Pb/^{238}U, ^{226}Ra/^{238}U, ^{228}Ra/^{232}Th ratios of 0.73, 0.80 and 0.79, respectively, are lower than those observed for Caetité soils. It can be postulated that a more efficient removal process of ^{210}Pb and Ra isotopes from Poços de Caldas soils is taking place in comparison to Caetité.

The relative mobilization of radionuclides in the soil will depend, among other factors, on the associations of these radionuclides with the different geochemical phases of the soil. Sequential extractions are generally used to discuss the distribution of chemical elements in the different geochemical phases of soils and sediments. Variations in the experimental conditions for sequential extraction techniques are numerous (e.g. choice of reagents, reagent concentrations, reaction temperatures, and duration of reaction periods) and controversial, but at least the procedure can shed some light on this discussion.

Cardoso (2004) assessed the distribution of radionuclides in soils of the Caetité region using a sequential extraction methodology that divided the soil into five fractions: (1) slightly acid (supposed to characterize the readily available metals; 2 M $CH_3COOH + 2$ M CH_3COONa at pH 4.7); (2) reducible phase (in which the metals are supposed to be associated with Mn-oxides; 0.1 M $NH_2OH{-}HCl$); (3) oxidizable (H_2O_2 30%); (4) alkaline (in which metals are associated with Fe-oxides; 0.1 M NaOH, pH 12); and (5) residual phase (taken as the summation of the previous phases). The author concluded that the contribution of the residual fraction in the bulk content of radionuclides in soil decreases according to the flowing order ^{232}Th and ^{210}Pb (~90%), ^{238}U (~75%), ^{226}Ra (~67%) and ^{228}Ra (~36%). If these results are used as a reference to explain the dynamics of radionuclides in Poços de Caldas soils, it can be suggested that relatively higher potential mobility of radium isotopes in those soils may explain the lower ratios of ^{226}Ra/^{238}U and ^{228}Ra/^{232}Th as these radionuclides would be mobilized from the soils due to the higher precipitation rates observed in the Poços de Caldas region. However, the same mechanism cannot be used to explain the behavior of ^{210}Pb. This radionuclide is hardly ever leached significantly from a soil column, rather it is well known for its particle-reactive nature, which is exactly what makes it a good tracer for erosion and sedimentation studies. The high percentage of ^{210}Pb in the non-mobile phases of Caetité soils supports this statement. So, if there is a removal of ^{210}Pb from the Poços de Caldas soils, then this indicates that the particles to which the ^{210}Pb is bound have been removed, likely by erosion, what is also in good agreement with the higher precipitation rates observed at the Poços de Caldas region.

Blanco et al. (2005) presented results of natural radionuclides in soil and plants of a mineralized uranium area in southwestern Spain. In the non-affected area of the former uranium mining activities, the average activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{228}Ra were 178 ± 58; 303 ± 45; 28 ± 39 and 39 ± 12, respectively. These figures lead to ^{226}Ra/^{238}U and ^{228}Ra/^{232}Th ratios of 1.77 and 1.37, respectively. Blanco et al. (2005) reported ^{226}Ra/^{238}U activity ratio in soil to be equal to 1.20 for the same area.

It has been suggested by Ivanovich and Harmond (1992) that the disequilibrium between Ra and U isotopes may be due to four factors: (1) precipitation/dissolution reactions; (2) alpha recoil; (3) diffusion; and (4) Szilard-Chalmers effect. Dowdall and O'Dea (2002) report that ^{238}U loss in organic soils in Donegal, Ireland, may be primarily due to the oxidation of organic material within the soil. They support this hypothesis with evidence that ^{238}U in this fraction tends to increase downward in a soil profile as conditions become increasingly reducing. At the lower, more reducing depths of the soil profile, ^{238}U and ^{226}Ra were essentially in equilibrium.

4.2. Radionuclides in vegetation

According to Chen et al. (2005) there are considerable differences in the uptake and translocation of long-lived radionuclides among different plant species. Up until now, several investigations on mobilization of natural radionuclides (such as ^{238}U and ^{226}Ra) in different compartments (soil, plant, and water), as well as the transfer between them, have been performed at different uranium mining sites around the world. However, there is not much information about the transport of ^{238}U, ^{226}Ra and ^{232}Th from soil to plant in temperate to tropical climate ecosystems.

Table 5 depicts the radionuclide concentrations in vegetables of the study area. The most outstanding figures relate to radium isotope concentrations in manioc and that of ^{210}Pb in pasture. Higher ^{228}Ra concentration in manioc in comparison to ^{226}Ra, of about one order of magnitude, have also been reported by Santos et al. (2002), although these authors reported activity concentrations in fresh weight without providing the fresh/dry weight ratio used in the work. The relatively higher uptake of ^{228}Ra in comparison to ^{226}Ra by manioc may be explained by the much more labile behavior of the former in soils of the study area as demonstrated above. The elevated concentrations of ^{210}Pb in pasture may also be explained by the fall-out and are in good agreement with the high concentration of the radionuclide in soils.

Table 6 shows the calculated transfer factors (TF; concentration vegetable/concentration in soil) for the different species and radionuclides based on the total soil concentration (TC) and on the readily available concentration (RAC), obtained from Cardoso (2004).

It is observed that 50% of the values are of the order of 10^{-3}; 25% of 10^{-2}; 20% of 10^{-4} and 5% of 10^{-1}, with the highest observed TF ^{228}Ra and ^{226}Ra in manioc, being 3.8×10^{-1} and 9.07×10^{-2}, respectively The highest ^{228}Ra concentration in manioc is not only a function of the higher radionuclide total concentrations in soil. In fact, the average readily available ^{228}Ra content in soil is 21%. This value is higher than that observed for ^{226}Ra, i.e. 12.8%. Therefore, in addition to the higher ^{228}Ra concentrations in total soil, a significant part of this amount exists in a more labile state when compared with other radionuclides.

Despite the high ^{210}Pb concentrations in soil, the radionuclide is not transferred to the plants to a high extent. According to Cardoso (2004) the average content of the radionuclide in the residual fraction of the Caetité soils is equal to 89%, which is almost identical to the value reported for ^{232}Th. The elevated concentrations of lead deposited by fallout in pasture, however, may play a relevant role in the transfer of the isotope to cattle, possibly contributing to the increase of its concentration in milk.

Based on the average TF obtained for each radionuclide amongst the different studied vegetables, a sequence of absorption by the plant may be proposed: $^{228}\text{Ra} > {}^{226}\text{Ra} \sim {}^{210}\text{Pb} > {}^{238}\text{U} \sim {}^{232}\text{Th}$. However, if the extreme values observed for manioc are removed, the TF value for all the radionuclides in the studied vegetables could be represented by a single average value of 6.0×10^{-3}.

Markose et al. (1993) studied the migration characteristics of ^{226}Ra from soil and uranium mill tailings to edible and wild plants. The TF showed wide variation (10^{-3} to 10^{-1}) in different species but were approximately in the same range as those observed in the present work. Controlled experiments carried out by growing edible plants on soil and uranium tailings also indicated large variation in TF when calculated on the basis of total radium, while reasonably consistent TFs (1.6×10^{-2} to 4.2×10^{-2}) were obtained when calculated on the basis of

Table 5
Radionuclide concentrations (Bq $\text{kg}^{-1}_{\text{(dry)}}$) in vegetables of the study area

Product	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Corn	0.023 ± 0.017	0.13 ± 0.11	0.12 ± 0.049	0.051 ± 0.031	0.22 ± 0.16
Manioc	0.11 ± 0.07	5.7 ± 4.17	2.7 ± 1.9	0.028 ± 0.047	31.0 ± 22.0
Black beans	0.19 ± 0.19	0.45 ± 0.36	0.17 ± 0.062	0.73 ± 1.23	0.60 ± 0.46
Pasture	0.53 ± 0.18	0.38 ± 0.26	4.98 ± 0.76	0.45 ± 0.32	0.83 ± 0.62

Table 6
Calculated soil–plant transfer factors

Product	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra
Corn	3.36×10^{-4}	2.03×10^{-3}	7.06×10^{-4}	8.97×10^{-4}	2.75×10^{-3}
Manioc	1.54×10^{-3}	9.07×10^{-2}	1.57×10^{-2}	4.90×10^{-3}	3.84×10^{-1}
Black bean	2.75×10^{-3}	7.19×10^{-3}	9.80×10^{-4}	1.27×10^{-2}	7.36×10^{-3}
Pasture	2.68×10^{-3}	6.03×10^{-3}	2.88×10^{-2}	7.84×10^{-3}	1.02×10^{-2}
Average	1.82×10^{-3}	2.65×10^{-2}	1.16×10^{-2}	6.59×10^{-3}	1.01×10^{-1}

available radium in soil/tailings. Vera Tome et al. (2003) reported transfer factors for ^{238}U, ^{232}Th and ^{226}Ra in the range of 10^{-2} to 10^{-1} which is also compatible with data from the present work. Bettencourt et al. (1988) presented a study of the uptake of ^{226}Ra by vegetables in zones contaminated by wastes from an abandoned radium salts factory and from uranium mines and in uncontaminated regions of purely natural radioactivity. They reported that ^{226}Ra TF values for cabbage leaves varied from 7.0×10^{-3} to 1.1×10^{-1} and for herbage from 9.0×10^{-2} to 5×10^{-1}. Finally, it should be pointed out that IUR (1994) reported TF values in the range of 10^{-3} to 10^{-1} for both ^{238}U and ^{232}Th.

The interpretation of the presented results, along with data published in the literature, suggest that a range from 10^{-3} to 10^{-1} may conveniently encompass most of the TF values for soil/plant systems (involving different cultures, different soils and natural radionuclides). If this is true, the whole worldwide available data pool can be used to provide, at least for the purpose of dose assessments, the general trend for the uptake of radionuclide from the soil by different plants. This would be useful to avoid to some extent, expensive and time consuming experimental studies. However, when discussing the uptake of radionuclides by plants from the soil, the challenge is to move away from the classical transfer factor approach and sequential extractions, which tend to lump together a variety of controlling variables, towards a more mechanicistic description of the plant uptake.

4.3. Radionuclides in air

4.3.1. Aerosol

In the analysis of radionuclide concentrations in aerosol data from sampling stations inside the installation are compared with those located outside (i.e. in the environment). The sampling points are located in the platform of the milling area (INST 001) intended to characterize the contributions from the leached ore piles and from the milling operations and at the settling ponds area (INST 008), supposed to give information about the contributions of the ore crushing operations. The results are shown in Table 7.

The $^{238}U/^{226}Ra$ ratios observed in the samples from both locations give good indication of the origin of the material in the sampling filters, i.e., in sampling location INST 001 the $^{238}U/^{226}Ra$ ratio is equal to 3.70, while in station INST 008 it is close to unity, 1.05. The $^{238}U/^{226}Ra$ ratio in the ore is 1.7 which is closer to these radionuclides ratio in aerosols of sampling station INST 008 and half of the value observed in station INST 001. As a conclusion, emissions from the milling plant, where uranium is concentrated, are reflected in the aerosol collected in INST 001 while in INST 008 the influence of the ore is more evident.

Table 8 shows the radionuclide concentrations in aerosols collected in the environmental stations.

Table 7
Radionuclide concentrations in aerosols samples collected inside the uranium mining and milling area

Sampling point	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra	Weight (g)
INST 001 ($\times 10^{-5}$Bq m^{-3})	68 ± 42	19 ± 9.8	44 ± 13	4.92 ± 2.0	7.40 ± 4.7	1.10 ± 0.23
(Bq kg^{-1}) × 10^{3}	15 ± 7.4	4.4 ± 1.8	9.7 ± 3.0	1.08 ± 0.56	1.57 ± 0.97	
INST 008 ($\times 10^{-5}$Bq m^{-3})	94 ± 49	90 ± 52	94 ± 35	4.92 ± 4.0	43 ± 50	1.06 ± 0.16
(Bq kg^{-1}) × 10^{4}	1.90 ± 0.9	2.1 ± 1.7	2.1 ± 1.2	0.1 ± 0.06	1.0 ± 0.12	

Sampling point locations: INST 001, milling plant platform; INST 008, close to the ponds with sub-aerial drains.

^{210}Pb activity concentrations in aerosol are the highest amongst the investigated radionuclides and are fairly constant throughout the sampling points. This finding confirms the hypothesis that ^{222}Rn decay and the subsequent fall-out of ^{210}Pb onto soil and pasture surfaces are responsible altogether for the high concentrations of this radionuclide in those matrixes. ^{238}U activity concentrations are higher in sampling stations AERO 010 and AERO 013 that are located in the preferential impact sector regarding aerial emissions from the mining and milling site, and located within a 5 km circle of radii centered at the plant. Average ^{238}U concentrations in stations AERO 001, AERO 042 and AERO 008 is $3.41 \pm 0.3 \times 10^{-5}$ Bq m^{-3}, while the radionuclide concentration in stations AERO 010 and AERO 013 are respectively 11.7 and 8.1×10^{-5} Bq m^{-3}. These figures suggest that mining and milling operations are probably increasing aerosol concentrations in the environment. It is interesting to observe that the highest of ^{226}Ra concentrations are in stations AERO 010, AERO 013 and AERO 008 compared to those observed in stations AERO 001 and AERO 042. The most probable reason for this finding is the transport of ^{226}Ra from the leached ore piles to the environment, emphasizing the contribution the role of the plant as source of these contaminants to the environment through the atmospheric pathway. The ^{232}Th/^{228}Ra ratios in stations 001 and 042 are respectively 0.72 and 0.76, which come to be identical values while in stations AERO 010 and AERO 013 (under the direct influence of plant emissions), the ratio is equal to 0.28 and 0.55. Again, stations AERO 001 and AERO 042 show a consistent identity among themselves. The evidence above points to the fact that aerosol samples collected in stations AERO 042 and AERO 001 have a different source than other stations, but are free from contributions of the industrial plant. If we take into consideration the sectors represented by the monitoring stations in Fig. 2 and assume the total area of a circle centered in the installation, it could be suggested that the aerial emissions would potentially impact an area not bigger than about 15 km^2.

Table 8
Radionuclide concentrations ($\times 10^{-5}$ Bq m^{-3}) in aerosols samples collected from sampling stations located outside the mining plant

Sampling points	^{238}U	^{232}Th	^{226}Ra	^{228}Ra	^{210}Pb	Particulate (g)
AERO 001 (WSW) 10–15 km	3.53	0.68	0.45	0.94	22	1.03
AERO 008 (WSW) 0–5 km	3.04	1.23	1.11	1.47	27	1.86
AERO 010 (WNW) 0–5 km	11.7	0.75	5.49	2.61	27	1.08
AERO 013 (W) 0–5 km	8.10	3.65	3.28	6.56	27	3.00
AERO 042 (ESE) 5–10 km	3.67	0.48	0.28	0.63	22	0.62

4.3.2. Radon

The average values related to sampling stations inside of the facility are shown in Table 9.

The results indicate that the leaching area presents the highest values amongst the sampling station, while radon concentrations in the other sampling stations are virtually the same. It is to say the ore pile constitutes the most relevant single radon source in the mining and milling site.

Table 10 presents radon concentration data in the environment. With the exception of the value recorded in station AERO 001 (located at the preferential impact sector) the gas concentrations in the other stations are lower than those observed in the mine area, being the lowest value observed in station 042 which is located at the opposite direction to the preferential impact sector in relation to the mining and milling area emissions. That may, in principle, suggest some contribution of the mining operations to these sampling points. However, radon concentration in air is a result from the emissions of the soil (local production) that will depend on the lithologies and their uranium content, which is the most likely primary parameter that controls the radon concentration in the outdoor environment. However, it is not sufficient to explain the large variability of the gas on a single lithogeochemical unit (Ielsh et al., 2001). In a uranium area, the transported amount from the source areas may, in principle, be thought of as another important contributor. On the other hand, the concentration shows a diurnal pattern with maximum value occurring in the early morning hours and minimum in the afternoon; the early morning peaks are due to the establishment of temperature inversions overnight. Finally, lower Rn exhalation rates from the ground surface are expected in the wet seasons due to the higher soil moisture, which introduces a seasonal variation in radon concentration. Due to the measurement technique used in this work, all the above-mentioned processes are lumped in the reported values. This allows for a better comparison with results from other studies, which used the same analytical approach.

Binns et al. (1998) report an average ^{222}Rn concentration from different environmental sampling points, a total of 118 measurements, equal to 68 ± 6 Bq m^{-3}. All these measurements were made before the beginning of mining and milling operations. As a result this value can be taken as characterizing a pre-operational situation of the site. Martin et al. (2004) report variations of one order of magnitude (5–45 Bq m^{-3}) in the same sampling station from wet to dry season in the Djarr Djarr locality, in the Northern Territory in Australia. The authors reported that the average concentration over a yearly cycle, during a time interval of 4 years (1998–2001) was 9.3 Bq m^{-3}. It was also reported by those authors that Rn concentrations in the remediate area of Nabarlek can reach 300 Bq m^{-3} with down gradients of one order of magnitude. The contribution of the Ranger mining site to adjacent communities was estimated to vary in the range of 1–8 Bq m^{-3}. The conclusion we can come up with is that non-mining

Table 9
Radon concentration inside the uranium mining and milling area

Sampling station	Description	Bq m^{-3}
Rn 001 ($n = 8$)	Milling plant	90 ± 71
Rn 003 ($n = 8$)	Waste-rock piles	102 ± 50
Rn 004 ($n = 20$)	Leached ore piles	627 ± 238
Rn 008 ($n = 8$)	Ponds with sub-aerial drains	118 ± 61
Rn 009 ($n = 8$)	250 m away from the leaching platform	104 ± 82
Rn 010 ($n = 8$)	Open pit, ore body 1	116 ± 60
Rn 011 ($n = 8$)	Open pit, ore body 3	135 ± 71

Table 10
Radon concentrations in sampling stations located outside the mining plant

Sampling station	Sector	Bq m^{-3}
AERO 001 ($n = 4$)	W	109
AERO 008 ($n = 4$)	WSW	57
AERO 010 ($n = 4$)	WNW	77
AERO 013 ($n = 4$)	WNW	73
AERO 042 ($n = 4$)	SE	44

related factors can result in large differences in Rn behavior in regions adjacent to mining areas. In this study, station AERO 001, located at 1 km from the mine site within the preferential impact sector, would be eventually the only area that could be suffering the impacts from the mining and milling operation radon emissions.

4.4. Radionuclide in water bodies

Due to the fact that the watercourses are not perennial at the study region and also because of the fact that liquid effluent emissions are not supposed to take place at the plant, our discussion will focus solely on the analysis of the meaning of radionuclide concentrations in groundwater.

The mine operator installed several wells throughout the region. Some of them are used to supply water for the local inhabitants; others are used as water supply to the industrial operations. Because of that, some of the wells are located in areas that can be affected by the mining and milling operations and others are not supposed to suffer any impact from the industrial operations.

In order to examine the eventual contributions of the mining and milling operations in groundwater, results from four monitoring wells that belong to the operator monitoring program were selected to be investigated. Three of them are located in the mining and milling area, they are: ASUB 001 and ASUB 279 (both of them potentially influenced by mining operations and close to the mine pit) and ASUB 276 (close to the tailings pond and also receiving the potential contribution from the leached ore/waste rock piles). The well ASUB 211 is out of the influence of mining and milling operations and was selected to serve as a reference point (sort of local background). The activity concentrations of ^{238}U, ^{226}Ra and ^{210}Pb are presented in Table 11 which also presents results of ^{238}U, ^{226}Ra and ^{210}Pb in Brazilian groundwater samples for comparison.

The elevated ^{238}U concentrations in waters from ASUB 001 and 279 could suggest that the aquifers could be receiving the contributions of drainage waters accumulated in the mine pit (concentrations are reported in Table 12) or be a consequence of the mutual contact of groundwater and rocks of the aquifers. Regarding ^{226}Ra and ^{210}Pb, concentrations they are similar to what is observed in the reference station and no contributions from the pollutant sources can be clearly assigned based on these radionuclide concentrations. The relatively high values of ^{210}Pb are probably due to the presence of dissolved ^{222}Rn in water.

If the ^{238}U/^{226}Ra ratios of the two sampling points, representing the main ore bodies of the mine pit, are taken into consideration, it will be seen that in the mine waters they are very similar to each other, i.e. 33.3 and 27.7, despite the difference in absolute concentrations of both radionuclides. This is an indication that both radioisotopes are being leached to the same extent in all the mining area. In groundwaters, the observed ratios were 17.4, 192 and 2.3 for stations

Table 11
Activity concentrations (Bq l^{-1}) of ^{238}U, ^{226}Ra and ^{210}Pb in groundwater

Well	^{238}U	^{226}Ra	^{210}Pb
ASUB001 ($n = 12$)	7.30	0.42	0.20
ASUB279 ($n = 12$)	12.3	0.064	0.13
ASUB276 ($n = 12$)	0.09	0.039	0.086
ASUB211 ($n = 3$)	0.15	0.75	0.10
Results from Brazilian groundwater[a]	0.015 $<1.2 \times 10^{-4}$–0.093 $n = 210$	0.014 0.001–3.79 $n = 358$	0.040 0.009–0.98 $n = 210$

[a] Source: Godoy and Godoy (2006).

ASUB 001, ASUB 279 and ASUB 276, respectively. On the other hand, ASUB 211 station, which is located in an area out of the potential influence of the pollutant sources, presented an inverse tendency, i.e. a $^{238}U/^{226}Ra$ ratio of 0.2.

Added et al. (2005) reported $^{238}U/^{226}Ra$ concentrations in aquifers of a semi-arid region in Tunis as high as 37.8. However, in deep aquifers the ratio dropped to 0.24 as a consequence of the development of reducing conditions. Under these circumstances, the $^{238}U/^{226}Ra$ ratio tends to decrease as a consequence of the lower solubility of U(IV) in reducing environments. Hakam et al. (2001) measured the $^{238}U/^{226}Ra$ ratio in 15 wells in Morocco. The observed values varied in the range of 0.8–14 with an average value of 2.12. As a result, it can be suggested that, with the exception of the ratio observed in station ASUB 279, the others observed lie in the range reported in the literature.

The use of $^{234}U/^{238}U$ ratios which is extensively applied as a geochemical tool to investigate movement and flow relationships in hydrological units (Dabous and Osmond, 2001), would give relevant information regarding hydrogeological aspects of the studied wells and would be a very important tool in the appreciation of the most probable events in charge of ^{238}U elevated concentrations in the studied groundwaters, however, in the absence of the $^{238}U/^{234}U$ ratio data, the hypothesis of groundwater contamination or elevated concentrations being caused solely by a process of water–rock interaction needed to be approached by a different way.

In oxidized surface and groundwaters uranium is transported as highly soluble uranyl ion (UO_2^{2+}) and its complexes, the most important of which are the carbonate complexes (Langmuir, 1997; Dabous and Osmond, 2001). The carbonate complexes are extremely important because they greatly increase the solubility of uranium minerals, facilitate U(IV) oxidation, and also limit the extent of uranium adsorption onto particles in oxidized waters, thus increasing its mobility. Carbonate ions may result from the weathering of the Al-silicates according to the following reaction:

$$2KAlSi_3O_8 + 2CO_2 + 11H_2O \rightarrow Al_2Si_2O_5(OH)_4 + 2K^+ + 2HCO_3^-$$

Table 12
Average radionuclide activity concentrations (Bq l^{-1}) in waters accumulated in the mine pit

Sampling station	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra	pH
Area body 01 ($n = 16$)	4.95 ± 5.5 or 0.41 ppm	0.15 ± 0.16	0.05 ± 0.03	0.04 ± 0.08	0.05 ± 0.10	8.27 ± 0.66
Area body 03 ($n = 22$)	57 ± 6.4 or 4.70 ppm	2.03 ± 1.29	0.22 ± 0.16	0.21 ± 0.25	0.23 ± 0.25	7.84 ± 0.40

Source: INB (2003).

in which K^+ may be replaced by other alkaline and alkaline earth metals. In the case of Na, its concentration is a consequence of the plagioclase dissolution, remembering that albite ($NaAlSi_3O_8$) is a major component of the rocks of the study area.

In order to explore the potential role of uranium complexation by carbonates and discuss the probable process in charge of the elevated uranium concentrations in groundwater, physical-chemical characterization of the waters from ASUB 001, 279 and 211 (reference point) were taken. These values are presented in Table 13.

The most outstanding differences concern the concentrations of bicarbonate and pH, with higher values in stations ASUB 001 and 279; and chloride, with higher concentration in station ASUB 211.

The higher Cl^- concentration in ASUB 211 is unlikely to be derived from rock weathering, probably resulting from its accumulation by evaporation, which is the dominant component of the local water balance. On the other hand, the HCO_3^- ions may result from the weathering of the Al-silicates as explained above.

Uranium speciation in samples of well ASUB 001 and ASUB 211 was undertaken. The computational code MINETQ A2 (Allison et al., 1991) was used. In the case of waters from ASUB 001, uranium speciation is as follows: 7.4% as UO_2CO_3(aq), 85.3% as $UO_2(CO_3)_2^{2-}$ and 7.2% as de $UO_2(CO_3)_3^{4-}$. In ASUB 211, the speciation gives the following results: 36.7% as UO_2CO_3 (aq) and 45.2% as $UO_2(HPO_4)_2$. The results allow us to accept that the complexation with carbonates is playing a very important role in the distribution of uranium in waters where the radionuclide concentrations are more elevated.

As a result, the higher concentrations of Cl^- in ASUB 211 and the higher concentrations of HCO3$^-$ in ASUB 001 and 279 allow us to propose a more effective connection of the aquifer represented by ASUB 211 with superficial waters in comparison to what would take place in ASUB 001 and 279. That is to say that the recharge events shall be more effective in ASUB 211 than in ASUB 001 and 279, the latter being more isolated from superficial waters. If this hypothesis were accepted, the dissolution of ^{238}U from the rock would account for the higher concentrations of the radionuclide in groundwater, and the contamination of these waters by the drainage accumulated in the open pit would not be favored.

Table 13
Chemical characterization of underground waters

Chemical species (mg l^{-1})	ASUB001	ASUB279	ASUB211
Na	100	107	176
K	7.67	7.67	6.33
Mg	10.8	7.33	12.9
Ca	81	52	27
Ba	0.32	0.09	6.19
Mn	0.77	0.19	0.67
Fe	1.13	1.79	0.53
SiO_2	57	62	98
PO_4	0.04	0.03	0.05
SO_4	2.4	11	20
F^-	2.13	1.77	0.37
Cl^-	107	173	425
HCO_3^-	288	194	9.90
Al^{3+}	–	–	0.36
pH	6.6	6.75	4.80

5. Conclusions

It was demonstrated that in heap leach plants most of the ^{226}Ra fractionates from the other radionuclides and remains in the leached ore that is subsequently disposed of in piles. On the other hand, in a leach-in-tank operation the radionuclide goes along with the other pollutants into the tailings. This characteristic of the heap leach processes has an important bearing in the overall potential environmental impacts.

Despite the potential higher availability of radium isotopes in the soils of Caetité region the lack of precipitation in that area reduces the leaching/mobilization of the radionuclides, which is reflected in the higher observed ^{226}Ra/^{238}U and ^{228}Ra/^{232}Th ratios in comparison to Poços de Caldas.

The highest radionuclide concentrations amongst the investigated vegetables were those of radium isotopes in manioc and of ^{210}Pb in pasture. The relatively higher uptake of ^{228}Ra in comparison to ^{226}Ra by manioc may be explained by the much more labile characteristic of the former in soils of the study area. The elevated concentrations of ^{210}Pb is explained by fall-out and are in good agreement with the high concentration of the radionuclide in soils.

The interpretation of the presented results along with data published in the literature suggest that a range from 10^{-3} to 10^{-1} may conveniently encompass most of the transfer factors (TF) values for soil/plant systems (i.e. involving different cultures, different soils and natural radionuclides).

Impacts due to aerial transportation of aerosols and radon generated in mining were proved to be minimum and restricted to an area not greater than 15 km^2.

Uranium speciation in groundwater samples demonstrated that complexation with carbonates is playing a very important role in the dissolution of uranium in those waters. This mechanism is the probable reason for the elevated observed radionuclide concentrations, and the weathering of Al-silicates being the most relevant source of bi-carbonate to these waters. The higher Cl^- concentration in a well located outside the mining area probably originated from its accumulation by evaporation in the soils of the study area. That is to say that the recharge events shall be more effective in ASUB 211 than in ASUB 001 and 279, being the latest more isolated from superficial waters.

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