Journal of Environmental Management 88 (2008) 914–925

Journal of
Environmental
Management

www.elsevier.com/locate/jenvman

Critical analysis of the waste management performance of two uranium production units in Brazil—part II: Caetite production center

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Received 22 May 2006; received in revised form 3 April 2007; accepted 27 April 2007
Available online 9 July 2007

Abstract

This paper discusses the environmental waste management of the Heap-Leach Uranium Production Facility of Caetité located in a semi-arid region in Brazil. A comparison is made with the first uranium production site of the country located in Poços de Caldas. It is demonstrated that differences in the operational process along with different environmental conditions can lead to different impacts. In the present case groundwater is the potential most sensitive environmental medium despite the well-established consensus in the literature that radon and aerosol emissions may turn-out to be the most relevant environmental aspects of an installation located at this type of region. Most of the ^{226}Ra content in the ore remains in the leached ore that is deposited with the waste rock. A lack in appropriate prediction of the hydrological balance has been causing unanticipated emissions of liquid effluents into the environment. Chemical treatment of this effluent may be needed. Contamination of groundwater in the short term by the waste ponds is not to be expected but it can be a relevant issue in the long term. As a consequence, careful closure schemes will need to be put in place. Finally, the overall costs with remediation in the Caetité production center are lower than those observed at the Poços de Caldas mining site.

Keywords: Uranium mining; Waste management; Heap-Leach plant; Groundwater contamination

1. Introduction

In the previous paper of this study the waste management strategies adopted by the uranium production center of Poços de Caldas were examined.

The objective of this paper is to examine the potential impacts of the wastes generated at the production center of Caetité, emphasizing the extent to which the composition of the ore, the operational process and the environmental conditions contribute altogether to the potential impact scenarios. A comparison with the production center of Poç os de Caldas is made with the aim of demonstrating that, regarding environmental impacts, mining sites must be treated in a case-by-case basis. It is stressed the difficulties one may find in drawing general considerations about the potential environmental impacts of uranium mining and milling projects.

One of the main contributions of this study stems from the fact that it is recognized that there are few new data on releases of radionuclides into the environment from uranium mining and milling operations and most of the attention has been driven during the past decades to the emissions of radon into the atmosphere, having little attention been paid on releases into superficial and underground water bodies (UNSCEAR, 2000).

It is also reported in UNSCEAR (2000) that for mining operations in semi-arid and arid areas, liquid effluent releases are minimal, and radionuclide inputs via this pathway are estimated to be of little consequence. As it will be demonstrated in this paper, this belief is not necessarily true, and inaccurate information may lead different international agencies and regulatory authorities to propose inaccurate recommendations and regulatory requirements.

DOI of original article: 10.1016/j.jenvman.2007.01.045

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doi:10.1016/j.jenvman.2007.04.022

2. Site description

The Caetité plant is located in the central-southwestern region of Bahia state (latitude 13°56′36″S and longitude 42°15′32″W). The annual average precipitation rate is 927 mm. The region is characterized as a semi-arid one. The geological uranium content of the ore is 3000 ppm and the average thickness of the ore body is about 7 m. The uranium mineral is associated with albites ($NaAlSi_8O_8$). Thirty-five anomalies were identified in an area of about 1200 km^2 (Gomiero et al., 2004). The average elemental composition of the rocks is presented in Table 1. Albitite (70%) is the predominant mineral, followed by pyroxene (10–20%), garnet (2–5%) and magnetite (1–3%). Pyrite is not present in the host rock.

The extraction of uranium from the ore is achieved by means of a Heap-Leach process. The process is represented in Fig. 1. the ore crushing and the addition of a sulfuric acid solution, in the proportion of $15\,kg_{H_2SO_4}\,t^{-1}$ore, piles containing between 25,000 and 35,000 t of ore are formed. Each pile is submitted to a three washing step cycle:

First washing step: a H_2SO_4 solution of $25\,g\,L^{-1}$, in the ratio of $0.6\,m^3\,t_{ore}^{-1}$, is irrigated over the piled ore by means of perforated tubes, at a rate of $30\,L\,h^{-1}\,m^{-2}$.

Second washing step: with a $5\,g\,L^{-1}$ H_2SO_4 solution, in the ratio of $0.3\,m^3\,t_{ore}^{-1}$, at the same percolation rate.

Third washing step: with raw water, in the ratio of $0.3\,m^3 t_{ore}^{-1}$ at the same percolation rate.

The resulting solutions are collected in ponds where they are mixed in such a way that a final solution $2\,g\,L^{-1}$ of uranium, as U_3O_8, is achieved. Uranium is extracted from this feed solution by means of an organic solvent that consists of 7% of tertiary amine, 3% tridecanol and 90% kerosene. Uranium is stripped with a NaCl solution from which it is subsequently precipitated by the addition of ammonium hydroxide solution The liquid effluent generated in the milling plant consists of an acid solution with high concentrations of other elements that have been dissolved along with uranium, as for example Al, Fe, Ca, Mg and Si. This effluent is treated with CaO to a pH of 8.0. The slurry is pumped to high density poly-ethylene (HDPE) lined ponds provided with drains that allow for the sedimentation of the solids in suspension. The supernatant is re-circulated to the process. This process constitutes a closed circuit one with no liquid effluent being released into the environment under normal operation.

Table 1
Average rock composition in the Caetité mining site

Oxide	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).

3. Waste characterization

Wastes can be divided into two categories: (1) wastes from the mining operations that include waste rock and drainage waters and (2) wastes from the industrial process, consisting of the leached ore and effluents from the milling plant.

3.1. Wastes from mining operations

3.1.1. Solid wastes

Fig. 2 presents the amounts of waste-rock material generated by the mining operations. A comparison is made with the predicted amounts. The average total generated amount is 2.7 times higher than the predicted one. It already represents almost one-third of the total amount expected for this phase of the project, i.e., 12×10^6 t in 16 years. This difference may have an important impact in the long-term disposal of the waste-rock material and was caused by a lack of accurate definition of the ore zones in the mining area. This reinforces the idea that a good and precise mining plan is of vital importance to avoid unwanted problems related the waste-rock management in the future.

3.1.2. Drainages

Annual rainfall rates in the semi-arid tropics usually amounts to between 500 and 900 mm. The potential evapotranspiration on the other hand is usually very high, approximately 1500–2000 mm. It is a highly seasonal environment with dry and wet seasons. The rainfall intensities are often high and erratic. If these ratios are taken in the long term a deficit in the water balance will be registered. However, a rainfall surplus develops during shorter period of time, i.e., monthly, weekly or daily. Sandström (1995) presents calculations, based on 3-month intervals, which show a surplus of 4 mm while calculations based on daily intervals shows one of 200 mm. Due to the soil characteristics, the runoff is very high and infiltration through the soil is not efficient. Under these conditions the drainage management system of a particular installation may not be able to hold the amount of water contributed to the mining and milling area and unwarranted discharges into the environment may take place.

In the Caetité mining project it was assumed that the catchment area of the mine pit was 0.44 km^2. The total volume of all the reservoirs that form the drainage management system of the site, as shown in Fig. 3, is 110,000 m^3. During heavy rainfall events the mine pit catches part of the drained waters and floods, leading to the interruption of mining activities. Despite the hydrological deficit in the long run, the operations cannot remain

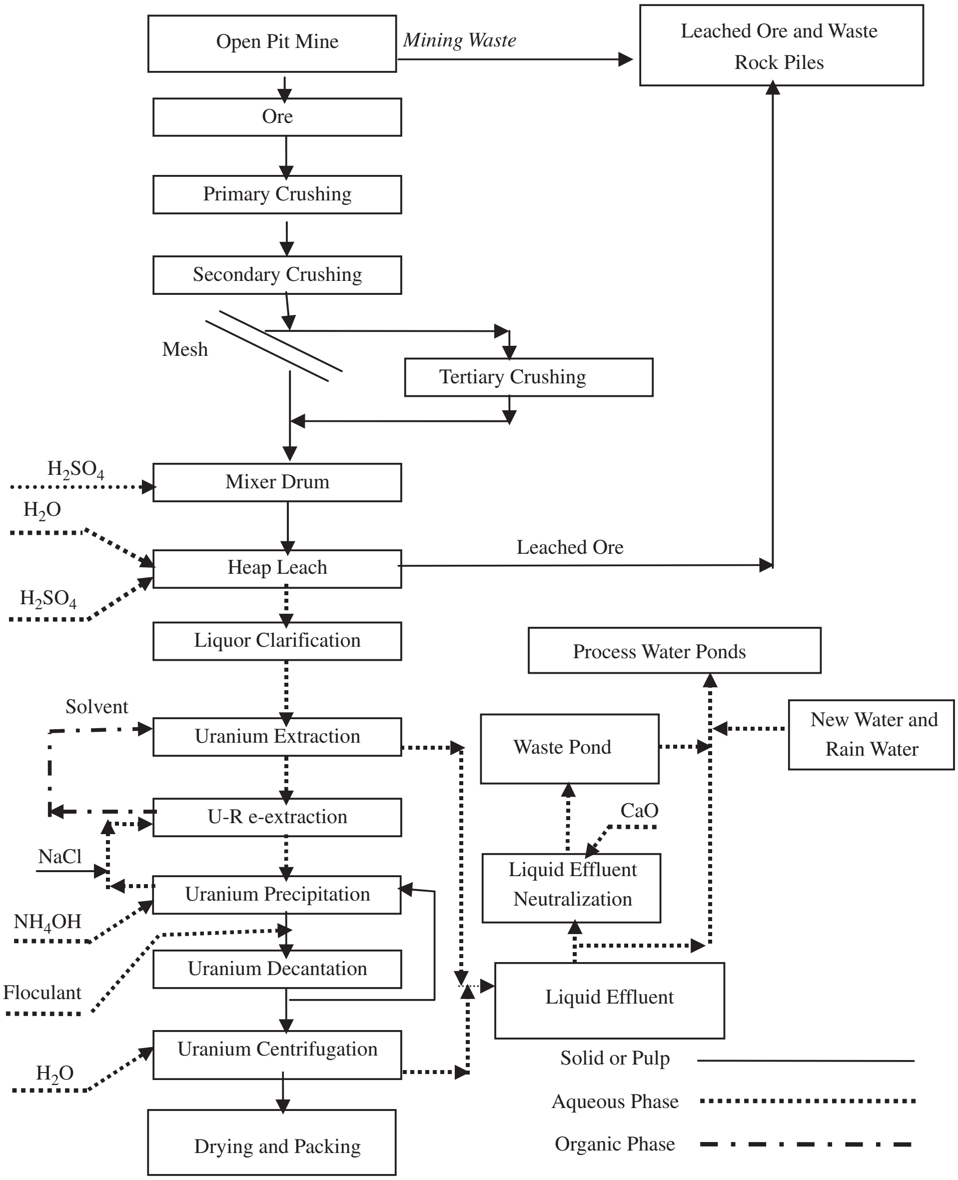

Fig. 1. Operational flowchart of the Caetité process.

paralyzed waiting for water evaporation. As a consequence, this amount of water has to be pumped of from the mine pit and stored in the available holding ponds as no liquid effluents are allowed to be released into the environment.

Table 2 shows the rainfall records from 2000 to 2004. It can be seen that approximately 70% of the annual rainfall are observed between November and March, i.e., amounting 560 mm. Taking runoff rates in the range of 0.2–0.4 it can be roughly estimated that the amount of water contributed to the mine pit would fall in the range between 50,000 and 100,000 m^3. When the waste ponds in operation (see Fig. 3) are completely full with liquids and solids, 77,000 m^3 must be subtracted from the overall available volume, leaving only 33,000 m^3 available for water retention. This amount is not enough to accommodate the water that needs to be pumped from the mine pit. As a result there is an urgent need for additional storage ponds to accommodate this excess of water or the introduction of a different drainage management system, as it will be discussed latter in this paper. The operation of a second waste pond cell can alleviate the present deficit; however, when this pond becomes full of wastes again the problem will return. The main mistake here was not to consider the time waters accumulated in the mine pit would need to evaporate and that by pumping these waters to holding ponds would make the available volume of these ponds be exhausted and whenever other rainfall event of high intensity takes place, unauthorized discharges into the environment would inevitably take place.

Regarding water quality, it can be seen from the results presented in Table 3 that uranium preferentially dissolves in relation to the other radionuclides. Different from what happens in Poços de Caldas mining site, where the

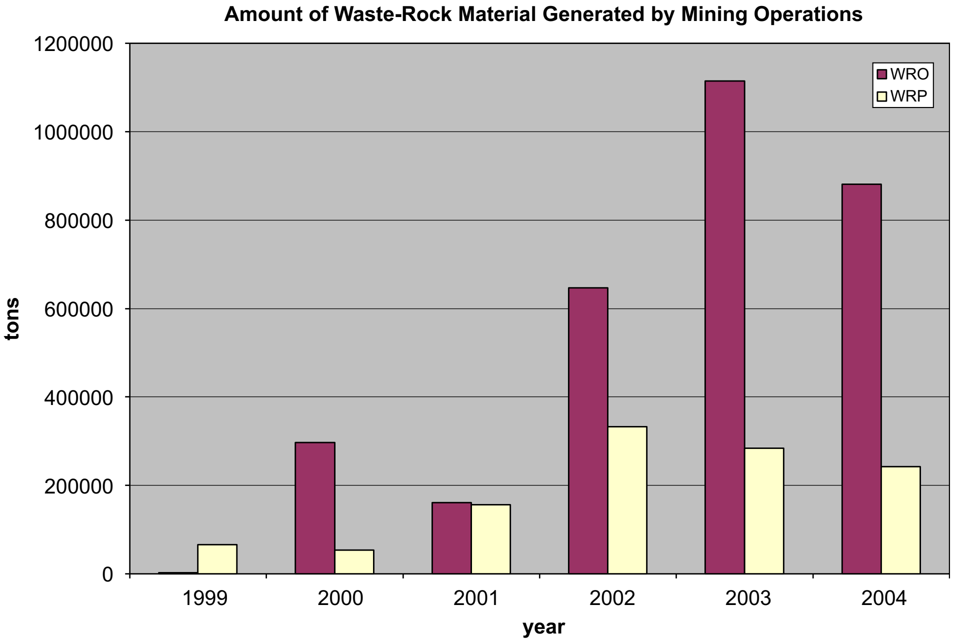

Fig. 2. Amounts of waste-rock material generated by mining operations (WRO, amount of waste-rock material observed after mining operations; WRP, amount of waste-rock material predicted before operations).

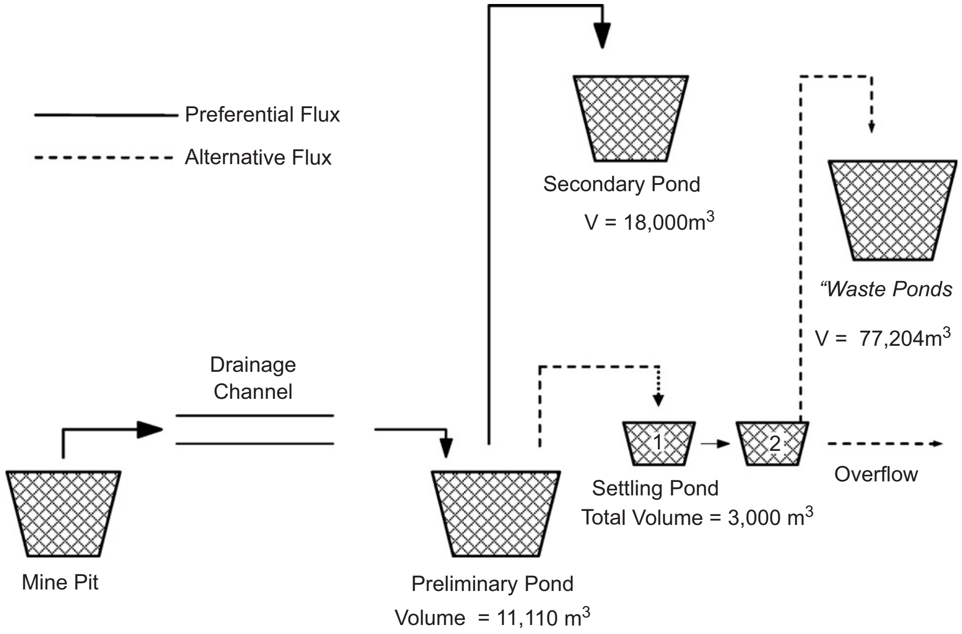

Fig. 3. Drainage management scheme.

oxidation of pyrite leads to high uranium concentrations (in the range between 71 and 315 $\mathrm{Bq\,L^{-1}}$) and low pH values (2.9–3.7) in mine and waste-rock drainages (Fernandes et al., 1998), the high uranium contents in the waters accumulated in the Caetité mine pit may be explained by the formation of carbonate complexes, associated with the relatively high pH values (Fernandes et al., 2006). These complexes largely replace the U (VI)-hydroxyl complexes above pH 6–7. The carbonate complexes are extremely important because they greatly increase solubility of uranium minerals, facilitate U (IV) oxidation, and also limit the extent of uranium adsorption in oxidized waters, thus increasing uranium mobility (Langmuir, 1997).

3.2. *Wastes from ore processing*

The leached ore is the primary waste generated in the uranium production. Table 4 presents the radiological characterization of the solid and liquid streams in the leach heap circuit.

One can see that while uranium is removed to a large extent from the ore in the sulfuric acid leaching solution (85%), Ra-isotopes are not. The removal of ^{228}Ra is relatively higher than that of ^{226}Ra. These results imply that the leached ore will contain most of the radium isotope amounts present in the ore before the leaching step and because of that will constitute the most important single potential source of these radionuclides to the environment. However, and despite the relative lower concentrations of these radionuclides in the liquors of the three washes, the waste ponds will also constitute a potential source of these radionuclides to the groundwater beneath them. It is very important to observe that the fate of radium isotopes in the Heap-Leach process is different from what is observed in the leach-in-tank one. In the later, radium isotopes follow the other potential pollutants in the ore and will end up in the mill tailings along with them. In the Heap-Leach process radium isotopes does not enter the milling circuit along with the other potential contaminants. These isotopes remain mostly in the leached ore. The pH values after the third wash are still low—1.84—implying that the leached ore still contains a residual acidity.

After the extraction of uranium from the leaching solution, a raffinate is formed. This acidic solution is neutralized to a pH equal to 8.0 and the pollutants present in the raffinate will end up in the waste ponds as slurry according to what is shown in Fig. 1. The composition of the raffinate before and after the neutralization is presented in Table 5. The composition of the solid phase formed after the neutralization is also presented. The concentration of ^{226}Ra in solution even after pH elevation cannot be considered as negligible from the environmental point of view. Data in Table 6 were input into the geochemical computational code MINTEQA2 (Allison et al., 1991) to examine the solid phases that would be formed from the raffinate in two different pH values, i.e., 1.5 and 8.0. The results indicate that when pH is 1.5 precipitation of barite ($BaSO_4$) strengite ($FePO_4$-$2H_2O$), hematite (Fe_2O_3), gypsum ($CaSO_4$) and $AlOHSO_4$ would be taking place. In pH 8.0 there would be the formation of diaspore (AlOH) and hydroapatite ($Ca_5(PO_4)_3OH$). Co-precipitation of ^{226}Ra and ^{228}Ra as $Ba(Ra)SO_4$ and $Ca(Ra)SO_4$ will be probably taking place and these phases will control the release of these radionuclides in the long term.

4. Identification and analysis of the potential environmental impacts associated with the mining wastes at the Caetite site

Due to the fact that liquid effluent emissions do not take place during normal operations, ground waters would be the potential most affected medium regarding the impacts of mining and milling wastes during normal operations. Atmospheric impacts due to radon emissions were assessed as being of low relevance (Carmo 2005).

The leached ore is encapsulated within the waste rock from mining operations to form a single unit. By the occasion of these cells deactivation soil will be placed over them, and vegetation will be developed. In order to estimate the potential impacts caused by the radium content in this system, the following processes need to be taken into account: (i) water infiltration through the

Table 2
Precipitation at the Caetité site between 2000 and 2004

Month	Rainfall (mm)					
	2000	2001	2002	2003	2004	Average
January	86.85	108.20	156.90	163.06	332.27	169.46
February	158	16.76	137.88	4.57	245.36	112.51
March	94.46	175.27	16.76	16.76	173.74	95.40
April	79.22	38.10	13.71	73.15	32.00	47.24
May	86.56	67.06	16.74	12.19	4.57	37.42
June	94.48	35.52	22.82	3.05	6.10	32.39
July	16.75	22.85	0.00	13.72	6.10	11.88
August	65.53	13.71	10.65	19.81	0.00	21.94
September	63.99	38.09	117.30	9.14	0.00	45.70
October	77.71	105.10	3.05	7.62	57.92	50.28
November	185.88	65.49	39.61	120.38	246.85	131.64
December	362.60	73.07	248.40	96.00	76.20	171.25
Total	1372.11	759.22	783.82	539.47	1181.11	927.11

Table 4
Radiological characterization of the materials coming from the leaching circuit

Material	^{238}U	^{226}Ra	^{228}Ra
Ore (Bq kg^{-1})	53,080 ± 11,975	33,280 ± 9549	83.0 ± 21.0
Leached ore (Bq kg^{-1}) dry	7582 ± 3290	34,520 ± 9512	75.00 ± 12
First washing solution (Bq L^{-1})	157,400 ± 30.770	23.0 ± 4.0	6.0 ± 0.5
Second washing water (Bq L^{-1})	54,840 ± 13,162	16 ± 4.5	2.3 ± 0.6

Table 3
Mean radionuclide concentration values in pit waters (Bq L^{-1})

Point	^{238}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra	pH
Area 01 ($n = 16$)	4.95 ± 5.5	0.15 ± 0.16	0.05 ± 0.03	0.04 ± 0.08	0.05 ± 0.10	8.27 ± 0.66
Area 03 ($n = 22$)	57 ± 64	2.03 ± 1.29	0.22 ± 0.16	0.21 ± .25	0.23 ± 0.25	7.84 ± 0.40

Obs: monthly sampling along year 2003.

Table 5
Chemical composition of the raffinate

Chemical species	Raffinate (mg L^{-1})	After pH increase to 8.0	
		Liquid phase	Solid phase (%)
SiO_2	1.120		1.95
Cao	987	823	Not available
Al_2O_3	3640	1.0	6.35
Fe_2O_3	4200	<0.03	7.33
TiO_2	480	<0.03	0.84
MgO	2020	659	2.37
BaO	1.0	<0.08	–
ZrO_2	15	<2	–
ZnO	16	0.06	–
Cr_2O_3	<10	0.1	–
MnO	159	<0.5	0.28
V_2O_5	167	0.5	–
U_3O_8	3.0	0.016	52 (ppm)
P_2O_5	1240	–	2.16
SO_4	31,300	1400	73.8
Radionuclides	Activity concentration		
	Bq L^{-1}		Bq g^{-1}
^{238}U	31	<0.2	0.54
^{230}Th	18	<0.02	315
^{226}Ra	25	3.0	0.38
^{210}Pb	65	0.02	1.13
pH	1.0 and 1.5	8.0	
Density	1.01 g/cm^{-3}	1.0 g cm^{-3}	

Source: INB (1999).

Table 6
Input data for the simulation of leached ^{226}Ra concentration from the leached ore pile in Caetité Production Center

Parameter	Description	Adopted value	Comment
L	Length of the source volume	100 (m)	Assuming basic pile geometry
W	Width of the source volume	50 (m)	
T	Thickness of the source volume	3.5 (m)	
P_r	Precipitation rate	0.927 (m yr^{-1})	
R	Runoff rate	0.32 (m yr^{-1})	Estimates taking into account a run-off factor of 0.4
E	Evapotranspiration	1.8 (m yr^{-1})	
θ_c	Volumetric moisture Content	0.1 ($m^3\,m^{-3}$)	
K_{dc}	^{226}Ra distribution coefficient	3.6×10^6 (mL g^{-1})	K_d was taken as the ratio between ^{226}Ra content in the second washing waters and in the solid material
ρ_c	Bulk density of the material	1.5 (g cm^{-3})	

soil cover and percolation through the waste rock, (ii) dissolution of gypsum and consequent Ra release, (iii) radionuclide transport through the pile, (iv) contaminant release at the toe of the pile, (v) contamination of superficial and groundwater. As mentioned before, the release of radium will depend upon the dissolution of gypsum which follows first-order kinetics.

For the sake of simplicity, the potential releases of this source were simulated by means of the construction of a worst-case scenario also known as a screening exercise. In this case the concentration of radium in the drainage of the pile was assessed in a situation in which the leached ore would be totally exposed to rain water. The source volume routine of the computational code GWSCREEN (Rood, 1994) was used to simulate Ra release from the leached ore. The governing equation of this part of the model is

$$dQ/dT = -(\lambda_L + \lambda_d + \lambda_0)Q. \tag{1}$$

Its solution is

$$Q(t) = Q_0 e^{-(\lambda_L + \lambda_d + \lambda_0)}, \tag{2}$$

where Q_0 is the initial mass or activity in the source volume (mg or µg) and t is the time (years).

The initial mass or activity in the source volume (Q_0) is given by

$$Q_0 = C_0 * L * W * T, \tag{3}$$

where L, W, T are the length, width, and thickness of the source volume (m) and C_0 is the initial concentration (Bq or mg m^{-3}) in the solid and liquid phases.

The leach rate constant is given by

$$\lambda_L = \frac{P}{\theta_c R_{dc} T}, \tag{4}$$

where P is the net water percolation rate (m yr^{-1}), θ_c is volumetric moisture content in the source volume (m^3 m^{-3}), R_{dc} is retardation factor in the source volume, $T = T_{1/2}$ is contaminant half life (yr^{-1}).

The net water percolation rate (P) is the amount of water that enters the soil per unit time that is not lost to evapotranspiration or runoff. Under steady-state condition (no net storage), this can be approximated by

$$P = P_r - R - E, \tag{5}$$

where P_r is the precipitation (m yr^{-1}), R is runoff (m yr^{-1}), E is evapotranspiration (m yr^{-1}).

The value P/θ_c represents the pore velocity in the contaminated zone and is based on the unit gradient model, which assumes that the hydraulic conductivity is equal to the net percolation rate.

The retardation factor in the source volume (R_{dc}) is given by Eq. (6):

$$R_{dc} = 1 + \frac{K_{dc}\rho_c}{\theta_c}, \tag{6}$$

where K_{dc} is the sorption coefficient at source (mL g^{-1}) and ρ_c is the bulk density (g cm^{-3}).

The contaminant flux out of the base of the source volume is calculated as

$$q_c(t) = \lambda_L Q(t), \tag{7}$$

where $q_c(t)$ is the contaminant flux out the bottom of the source volume (Bq yr^{-1}).

Calculations were undertaken for an individual pile of 25.000 t of leached ore.

In this kind of approach the choice of the K_d values is of critical significance as it will govern the ^{226}Ra concentrations in the leachate. Regarding the K_d of Ra in the source term it was decided to adopt a value derived by the ratio of the measured radium content in the ore to the measured content in the second wash water. The variation of the moisture content in the wastes will not affect significantly the radionuclide concentration in the leachate but will play an important role in the determination of the radionuclide fluxes from the system. Moisture content will depend, among other things, on the infiltration rate. It was estimated that ^{226}Ra fluxes would vary in the range of 4.76×10^6–3.81×10^7 Bq a^{-1} corresponding to leach rates of 5.52×10^{-6}–4.41×10^{-5} a^{-1}, respectively.

The other sources of pollutants are the waste ponds in which the wastes from the uranium extraction process are deposited. The GWSCREEN (Rood, 1994) model was used to assess these impacts into groundwater. Simulations were run considering the absence of an upper cover and bottom geosynthetic liners and/or clay layer. That would represent a worst-case scenario. In fact, the actual construction scheme of these cells consists of a compacted clay layer 20 cm thick in the bottom of the ponds overlaid by a geosynthetic membrane—HDPE. Consequently, if the impacts are negligible under the worst-case scenario they will not be relevant in the real situation either. Simulations were run for ^{226}Ra and ^{210}Pb as its progeny assuming that these radionuclides are critical contaminants amongst the overall species contained in the waste material.

Simulations were done in two steps. The first one considered the waste pond receiving a continuous flux of waste (solid + liquid materials) during 10 years. In fact this is a conservative assumption as the operational lifetime of these cells is designed to be of 4 years. The simulation of this part of the cell operation was done with the aid of the pond model routine of the GWSCREEN. In the second phase of the simulations, i.e., after the effluent input cessation the waste cell was modeled as buried source and the respective routine of the GWSCREEN model was adopted. The K_d in the unsaturated zone and in the aquifer were conservatively fixed to 10 L mg^{-1} as to maximize the potential of any impact in groundwater and follows the values adopted in Rood (1994). The input data of both simulations are presented in Table 7 and the results showed in Fig. 4. The main differences between the input data refer to the percolation rates (Darcy velocity). In the pond model routine a value equal to 58.7 m yr^{-1} was used while in the buried source routine a value equal to 0.927 m yr^{-1} (see Table 2) was taken into consideration. This value corresponds to the average local precipitation rate at the site. In doing so, the leach rate in the pond model routine presents a value equal to 1.20×10^{-1} yr^{-1} and for the buried source routine a value equal to 1.60×10^{-3} yr^{-1} is obtained. These values are order of magnitude higher than those obtained for the leached ore deposited along with the waste-rock material. As a result, these sources (waste ponds) are to be of greater concern regarding ^{226}Ra releases into the environment. The transit times in the unsaturated zone are practically equivalent, 96 years in the pond model routine and 108 years in the buried source one. This means that no contamination will be observed in the aquifer before 100 years (the worst-case scenario taken into account, i.e., the presence of the 20 cm clay layer and the geomembrane not taken into account). It can be seen from Fig. 4 that if the pond was allowed to work with the percolation rate of 58.7 m yr^{-1} a peak concentrations of 384 and 377 Bq L^{-1} would be observed for ^{226}Ra and ^{210}Pb, respectively, in 107 years. Peak concentrations for the same

Table 7
Input data for the simulation of groundwater concentration under the influence of the tailings holding ponds of Caetité Production Center

Variable	Pond model routine	Buried source routine
Number of radioactive progeny	1	1
Length of source parallel to groundwater flow (m)	129	129
Width of source perpendicular to groundwater flow (m)	129	129
Thickness of source (m)	3.0	6.0
Percolation rate (Darcy velocity $m\,yr^{-1}$)	58.7	0.927
Volumetric water content in source	0.86	0.86
Volumetric water content in unsaturated zone	0.15	0.15
Bulk density at source ($g\,cm^{-3}$)	0.33	0.33
Sorption coefficient at source ($mL\,g^{-1}$)[a]	500	500
Bulk density in unsaturated zone ($g\,cm^{-3}$)	1.72	1.72
Unsaturated zone thickness (m)	5.0	5.0
Sorption coefficient in unsaturated zone ($mL\,g^{I}$)	10	10
Initial activity (Ci)	0.0078	0.0078
Molecular weight ($g\,mole^{-1}$)	226	226
Solubility limit ($mg\,L^{-1}$)	1.0	1.0
Half lives of contaminant and progeny (y)	1620 and 21	1620 and 21
Bulk density of aquifer ($g\,cm^{-3}$)	2.75	2.75
Porosity of aquifer	0.005	0.005
Sorption coefficient(s) in aquifer ($mL\,g^{-1}$)	0.0 and 0.0	0.0 and 0.0
Dispersivity X direction (m)	10	10
Dispersivity Y direction (m)	1	1
Pore velocity ($m\,yr^{-1}$)	584	584
Well screen thickness (m)	1.0	1.0
Distance to receptor along X-axis (m)	70	70
Distance to receptor along Y-axis (m)	0.0	0.0
Activity release rate to pond ($Ci\,yr^{-1}$)	0.003	Not applicable
Operation time of pond (year)	10	Not applicable
Liquid effluent flow rate ($m^3\,yr^{-1}$)	15,000	Not applicable
Evaporation rate constant from pond (yr^{-1})	0.0	Not applicable
Effective width of pond at aquifer (m)	129	129

[a] Sorption coefficient at source indicates the partition of radionuclide between the solid and liquid phases of the waste material.

radionuclides would be, respectively, 1.17 and 1.16 $Bq\,L^{-1}$ achieved in 109 years by the buried source routine. The total dose of ^{226}Ra and ^{210}Pb for the peak concentrations would be equal to 1.84 $mSv\,yr^{-1}$ due to the ingestion of groundwater at a rate of 2 $L\,day^{-1}$ by an intruder that would have dug a well in the vicinity of the pond. Obviously, this dose rate is higher than the 1.0 $mSv\,yr^{-1}$ allowed by the Brazilian legislation and has to be avoided.

As a result of these simulations, it can be concluded that no impact into the aquifer is expected to take place during the operational phase of the waste cells, especially if one takes into account that the clay layer and the geomembrane would reduce the infiltration of contaminated waters into the underlying soils. However, in the long term the potential contamination of the aquifer beneath the waste pond cannot be discarded. This will make it necessary the adoption of careful closure measurements of these cells to reduce, as much as possible the infiltration of rainwater. This can be achieved by the application of a geomembrane and another clay layer over the wastes. The compaction of the wastes to be achieved by the accumulated liquid drainage by means of the sub-aerial drains will also contribute to reduce the hydraulic conductivities of the wastes.

The contribution of the drainage accumulated in the mine pit into the groundwater is also to be assessed. Results from three monitoring wells were investigated. Two of them are located in the mining and milling area and close to the mine pit. They are: GW 001 and GW 279. The third one, GW 211, is out of the influence of the mining and milling operations and was selected to serve as a reference (background) point. The activity concentrations of ^{238}U, ^{226}Ra and ^{210}Pb in ground waters are presented in Table 8.

^{226}Ra and ^{210}Pb concentrations in groundwater are compatible to those observed in the reference station (GW 211). As a result, no contribution from the drainages accumulated in the mine pit can be assigned as influencing the groundwater. On the other hand, the elevated ^{238}U concentrations in waters from GW 001 and GW 279, higher than the concentration in GW 211, could suggest that the aquifer may be receiving the contributions of drainage waters accumulated in the mine pit.

A detailed hydrogeological study, concerning the potential impacts of mine waters into groundwater is underway. In this paper this issue will be treated based on the investigation of geochemical evidences that can explain the higher concentrations of uranium in groundwater in the

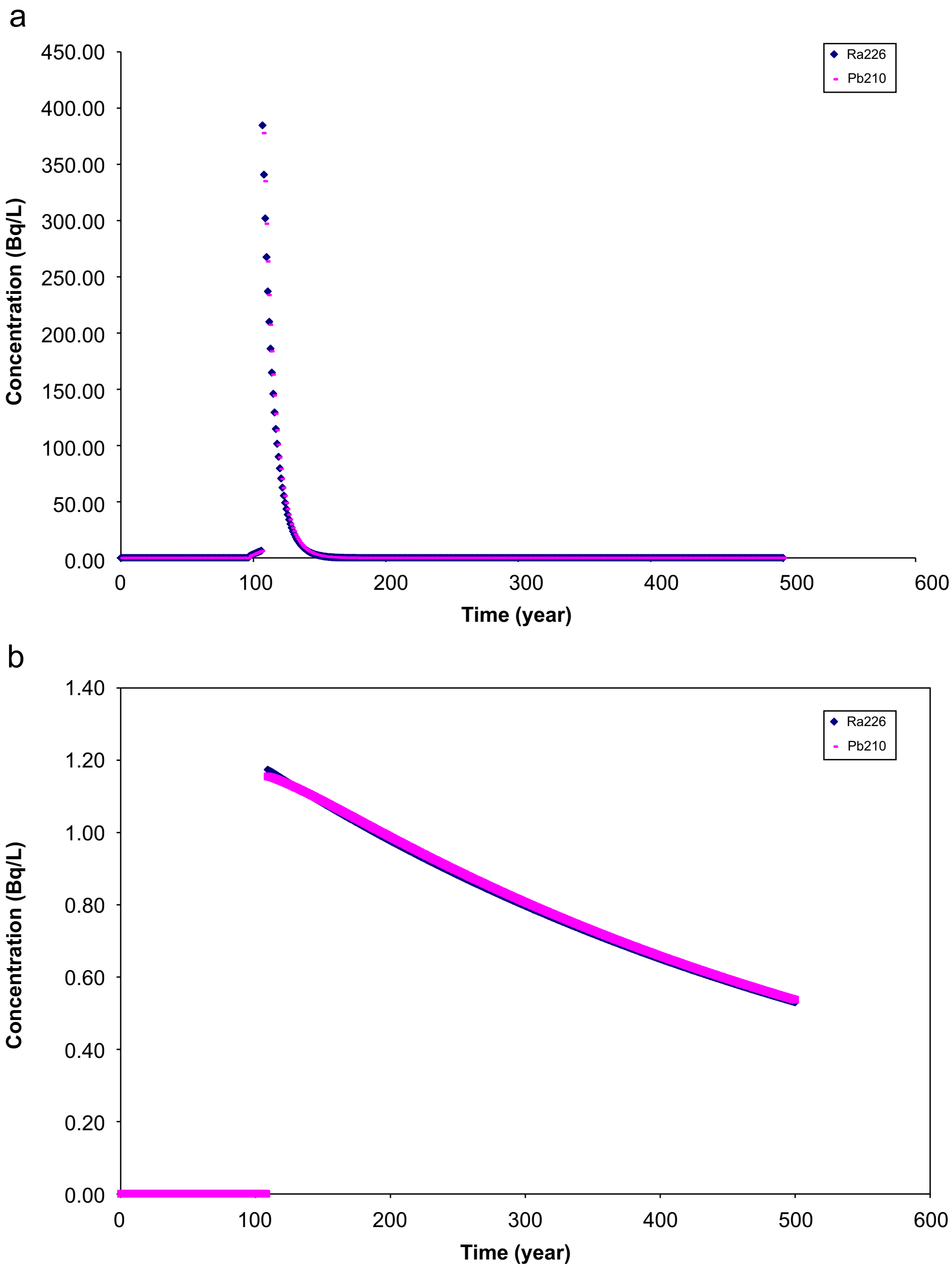

Fig. 4. (a) Simulated evolution of ^{226}Ra and ^{210}Pb concentrations in the aquifer beneath the waste pond using the pond model routine of the GWSCREEN model. (b) Simulated evolution of ^{226}Ra and ^{210}Pb concentrations in the aquifer beneath the waste pond using the buried source routine of the GWSCREEN model.

Table 8
Activity concentrations of ^{238}U, ^{226}Ra and ^{210}Pb in groundwater

Sampling station	^{238}U (Bq L^{-1})	^{226}Ra (Bq L^{-1})	^{210}Pb (Bq L^{-1})
GW 001 ($n = 12$)	7.30	0.42	0.20
GW 279 ($n = 12$)	12.3	0.064	0.13
GW 211 ($n = 3$)	0.15	0.75	0.10

area under the influence of the mine pit. In oxidized surface waters as well as in groundwater 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). Carbonate ions, like HCO_3^-, may result from the weathering of the Al-silicates according to the following reaction:

$$2KAlSi_3O_8 + 2CO_2 + 11H_2O \rightarrow Al_2Si_2O_5(OH)_4 + 2K + 2HCO_3^- \quad (8)$$

in which K^+ may be replaced by other alkaline and alkaline earth metals.

In order to investigate the potential role of carbonate complexation of uranium and discuss the probable process in charge of the elevated uranium concentrations in groundwater physical–chemical characterization of the waters from GW 001, GW 279 and GW 211 (reference point) were taken. These values are presented in Table 9.

The most outstanding differences between the composition of the studied waters have to do with the concentrations of bicarbonate and pH—with higher values in stations GW 001 and 279—and chloride, with higher concentration in station GW 211.

The predominance of HCO_3^- and Na^+ ions is usual for fractured aquifers in geological settings formed by metamorphic rocks with granite or gneissic composition, i.e., the significant Na concentrations relies on the fact that the metal is a component of albite ($NaAlSi_3O_8$) which is present in the rocks of the study region. Groundwater salinization is mainly related to sources of sodium and chloride concentrated in the vadose zone. In this context, the groundwater coming from wells placed in shallower fractures should have higher salinity than others coming from deeper ones. On the other hand, the HCO_3^- ions may result from the weathering of the Al-silicates as explained above.

Table 9
Chemical characterization of underground waters

Chemical species (mg L^{-1})	GW 001	GW 279	GW 211
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

Uranium speciation in samples of well GW 001 and GW 211 was undertaken. The computational code MINETQ A2 (Allison et al., 1991) was used. In the case of waters from GW 001 uranium speciation is as follow: 7.4% as UO_2CO_3(aq), 85.3% as $UO_2(CO_3)_2^{2-}$ and 7.2% as de $UO_2(CO_3)_3^{4-}$; while in GW 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 propose that the complexation with carbonates may be playing a very important role in the distribution of uranium in waters.

One needs to observe the huge difference between HCO_3^- in waters from wells 001 and 211, 288 and 9.90, respectively. The reason for the difference may rely on the pH values found in both waters. In the first the pH value is 6.6 and drops to 4.8 in the second. As a result dissolution of bicarbonates may be taking place and it is reflected in the uranium speciation results as showed above.

It seems then that elevated levels of ^{238}U in groundwaters are mainly a consequence of the complexation of the metal by carbonates and not by any sort of the contamination of these waters by the drainage accumulated in the open pit would not be favored.

Table 10 provides a comparison between different aspects of both mining centers (Pocos de Caldas and Caetite Production Centers).

Table 10
Comparison between mining characteristics of the two production centers

System	Poços de Caldas	Caetité
Waste rock (t)	105×10^6	3.1×10^6 (in 4 years) 12×10^6 (estimated in 16 years)
Ore grade (ppm)	800–1200	2300–3000
Tailings (t)	2.05×10^6	7.2×10^3–12×10^3 (per year) 1.15×10^5–1.92×10^5 (estimated for 16 years)
Tailings area (km^2)	1.8	0.47
U_3O_8—production (t)	1200	1000 (in 4 years)
Ore (t)	1.76×10^6	6.0 × 105 (in 4 years)
Environmental/climatological characteristics	Rainfall—1800 mm yr^{-1}	Rainfall—800 mm yr^{-1}
	Well developed agriculture activities	Absence of expressive agriculture
	River waters (in which effluents are released) are used for irrigation	Rivers are not perennial (no effluent release is permitted)
	No major use of groundwater	Groundwater is used for water consumption (confined fissural aquifers, low recharge rates)
Potential environmental impacts	Superficial water bodies (operational and post-operational phase)	Effluent releases not allowed in the operational phase
	Groundwater contamination not expected by radionuclides	Potential contamination of superficial water bodies in the post-operational phase
	Studies need to be developed concerning potential contamination due to the mine pit	Groundwater suspected of being affected by drainage disposed in the open pit and auxiliary holding ponds

Regarding the Pocos de Caldas Project, the relatively high rainfall rates (1800 mm yr^{-1}) associated with the occurrence of pyrite in the host rocks, giving rise to acid drainage and turns the continuous liquid emissions on a major environmental issue. In that particular case, release limits need to take into account the potential affected population exposure scenario (which includes the intensive use of river waters for irrigation and cattle watering) in order to keep the radiological doses to members of the general public as low as reasonably achievable. This exposure scenario is completely different from the one prevailing at the Caetite site. There, agricultural activities are very restricted and destined mainly for the subsistence of the local population. Due to the fact that the rivers are not perenial, no irrigation takes place. Water is supplied mainly from aquifers, which constitute the most sensitive environmental compartment regarding the contamination by radioactive and non-radioactive pollutants. As a result, the potential impacts associated with the Caetite unit are lower than those that can be assigned to the Pocos de Caldas site and this fact is translated into the costs regarding waste and environmental management.

The long-term duration of acid drainage generation at the Pocos Caldas site makes the adoption of permanent solutions necessary to replace the present collect and treat strategy. Regarding acid drainage, uranium is of major concern amongst the radionuclides associated with the waste-rocks/ore, because it is preferentially mobilized by the acid solutions percolating the dumps. As it was demonstrated in Part I of this study, remediation of the waste-rock piles needs to focus on the reduction of oxygen inputs to the dump. In addition to the waste-rock piles, the tailings dam is another relevant source of pollution in the present and in the long term. Differently from the Caetite plant, tailings from Pocos de Caldas contain most of the potential contaminants present in the ore. The presence of residual pyrite in the milled ore favors the occurrence of acid drainage in that material. In addition to these impacts, the emissions of radon from the tailings may be of concern in a possible case of area occupation by intruders. As a result, remediation of this area needs to involve both the reduction of oxygen diffusion into the tailings, to avoid the occurrence of acid drainage, along with the reduction of radon exhalation from the tailings.

Differently from the Pocos de Caldas site, the problem of acid drainage is not present in the waste-rock piles of the Caetite mine due to the absence of pyrite in the rocks. As a consequence of the co-disposal of the leached ore with the waste-rock material, these systems will contain virtually most of the total amount of radium in the ore and this radionuclide, differently from Pocos de Caldas, will be the critical one controlling future potential impacts from these sources. The lowest rainfall rates observed in the Caetite regions alleviate the problem of discharge management in the long run. However, and due to the intense showers that take place, especially during summer time, mine pit flooding and occasional unauthorized discharges into the environment are issues that have to be fixed. The construction of a water treatment plant may be an adequate solution for both of these problems. Since the prevention of oxygen infiltration into the waste-rock/leached rock system is not a relevant issue to be avoided the closure of these systems will be driven solely by the reduction of water infiltration into them in order to reduce the potential mobilization of the radium isotopes.

The far higher amounts of rock-materials moved in Poços de Caldas mining activities in relation to Caetité is a result of two components: the higher depths that the mineralized zone was located, and secondly the lower grade of the ore. Despite the fact that uranium production in Brazil is designed to supply solely the internal market, the operation of production centers, processing low-grade ores, may not be warranted if environmental costs are considered.

Costs with remediation activities at Poços de Caldas project amounted to \$731,315 between 1997 and 2001. In addition to this \$2.6 million have been spent with the neutralization of acid drainage between 1984 and 2002. This corresponds to 52% of the annual expenditures with effluent treatment of the whole operations of the Caetité production center. It is expected that the costs with the complete remediation of one of the waste-rock piles of the Poços de Caldas site will fall in the range of \$1.5–\$4.0 million depending on the type of remediation scheme to be put in place. Costs with the decommissioning of waste-rock and leached ore piles are on the order of \$25,000 per ha in Caetité project, which correspond to an annual average cost with the remediation of these systems of about \$125,000. If these costs are projected to the lifetime of this phase of the project, i.e., 16 years, the overall costs with the remediation of waste rock and leached ore piles will amount \$2.0 million. This figure is equivalent to the lower cost estimate of the remediation of only one of the waste-rock piles of the Poços de Caldas mining site (not considering the expenditures already put in place).

5. Conclusions

It was demonstrated in this paper that the type of impacts arising from the operation of a mining and milling facility will depend not only on the type of project (open pit/underground galleries), process (acid/alkaline leaching—Leach in Tank/Heap Leach), and ore grade but also, and perhaps most significantly, on climatological/environmental conditions. Contrarily to what is generally reported about environmental impacts of uranium plants located in arid/semi-arid regions, the aquatic pathway proved that it can be a more sensitive medium than the atmospheric one. As a consequence detailed investigation has to be carried out including a good understanding about the operational process, its possible influences on the mobilization and distribution of the potential contaminants, the main mechanisms in charge of the transport of contaminants in the environment. All these must be interpreted in an integrated way. Regulators, especially from developing country may rely on international recommendations that are based mainly in the experience collected from work published in the scientific literature and that arise from developed countries. Also of relevance is the fact that radiation safety and environmental impact assessment may be dealt with separately and appreciated by different organizations. By doing so the interactions between the relevant processes in the mobilization and transport of radionuclides from the source to the receptors may be lost. This work clearly demonstrates the importance of an integrated approach in which radiological and non-radiological aspects are addressed simultaneously.

It can be proposed that the Heap-Leach process adopted in Caetité is much more environmental friendly than the Leach-in-Tank one adopted at Poços de Caldas site. Finally, any conclusion regarding the potential impacts associated with wastes generated by uranium mining and milling projects shall be made on a case-by-case basis.

Regarding the Caetité site, the main issues to be considered in the overall waste management scheme of the plant have to do with:

(1) improvement of the modeling of the long-term release of Ra-isotopes from the leached/waste-rock piles;
(2) development of hydrogeological studies in the mine pit area in order to gain a better understanding on the potential contamination of groundwater by drainage accumulated in the mine pit;
(3) development of improved mining operations in order to reduce the generation of waste-rock material;
(4) continuous improvement of measures to avoid groundwater contamination by holding ponds (both in the milling circuit as well as in the waste deposit area);
(5) installation of drainage water treatment plant (e.g., ion-exchange resin, precipitation, etc.) in order to avoid unwarranted releases in the environment (during heavy rainfall events) and interruption of mining operations due to the mining flooding.

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