EVALUATING INTERVENTION OPTIONS FOR A CONTAMINATED LAND – THE USIN SITE

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1 ABSTRACT

Wastes from a monazite processing plant has contaminated a site located in an industrial district of São Paulo, the most densely populated Brazilian City. In its 60,000 square meters of area, contaminated zones were found encompassing an area of about 6,500 square meters. In some places, contamination was found below the superficial layer of the soil, being the radionuclide vertical distribution not uniform. The ^{228}Ra soil activity concentration reached values up to 33,000 Bq/kg while those for ^{226}Ra reached values up to 6,700 Bq/kg. Based on pathway analysis model and considering both the current land use and a hypothetical residential scenario, the residual contamination levels of radionuclides in soil have been derived. Considering the unrestricted use of the site, the preliminary assessment pointed out a volume of around 400 m^3 of soil to be taken out of the site. This paper discusses alternatives to site remediation taking into account the present radiation protection framework, specially concerning the principles of the system of radiological protection for intervention.

2 INTRODUCTION

Monazite basically consists of an orthophosphate of rare earths containing thorium and uranium. The fifth monazite deposit in the world is located along the Brazilian coast. According to documentation, the monazite exploration in Brazil began in 1884, when monazite-bearing sand was transported to Europe in clandestine ships as ballast for ship. In 1904 a French-Brazilian company started exporting monazite concentrate. By this time, the monazite interest increased due to the use of thorium nitrate by gas lantern mantle industries, to the use of ^{228}Ra (mesothorium) in luminous watch dials and to the use of ^{228}Ra and thorium in therapeutic applications. After that period, the new applications of lanthanide elements turned monazite into a much more important commodity than it was in pre-war years [1]. In 1949 an industrial unit for chemical processing of monazite, aiming production of lanthanide chlorides and tri-sodium phosphate, started operation at Santo Amaro mill, located in São Paulo city, the largest Brazilian city.

As a consequence of the monazite processing, three basically different kinds of wastes were produced:

a) From monazite physical purification a so-called light mineral fraction, app. 4.0 10^4 Bq/kg of ^{228}Ra and 6.3 10^4 of ^{226}Ra ;

b) From the monazite alkaline digestion the mesothorium cake ((Ba(Ra)SO_4)); and,
c) The Cake II (average content 20% of thorium hydroxide and 1% of uranium hydroxides).

Through the years a big amount of those radioactive wastes have been generated, being presently stored in shallow ground silos, rubber drums in temporary storage buildings and buried rubber drums in trenches scattered in different sites of the country.

As the radiological protection rules had not been implemented in Brazil until the 60s, the monazite waste storage and transport were performed without adequate care. So metal barrel containing wastes were laid in open air and spilled wastes ending in soil contamination. Also, the light mineral fraction has been used as landfill in some sites.

The Interlagos mill site is located in an industrial district of São Paulo city, encompassing an area of about 60,000m^2, and belongs to the state company responsible for monazite processing in Brazil. In one of the three buildings of the USIN site, a lanthanide separation plant was located during a short period of time; this processing operation aimed to split the light from the heavy lanthanides. The mill was closed in 1990 and shortly afterward the public minister requested to the Brazilian Nuclear Energy National Commission (CNEN) a radiological survey of the site. The public minister request was in attendance on the public concern about the careless waste stored at the site.

This paper aims to evaluate the site cleanup taking into account the radiation protection framework at present, especially that concerning the intervention principles.

3 METHODOLOGY

Pathway analysis model is a generic method used as a tool to help the decision-aiding process concerning site restoration (2,3). The model uses mathematics expression in potential scenarios of site occupation for dose assessment. The model allows the identification of the main exposure pathway, and critical radionuclide, and permits the derivation of cleanup levels (DCLs), in such way that the total doses due to all potential exposure pathways do not exceed the dose limit criteria established for the situation. For the calculation of the total dose, as a first step, the soil concentration for each radionuclide is set as 1 $Bq.g^{-1}$. Then, a straight relationship between dose and radionuclide concentration in soil is established and clean up level can be derived for each exposure scenario.

3.1 Dose limits-Framework of radiation protection

The basic principles of the radiological protection system for practices are justification, optimization and the limitation of individual dose. All activities involving radiation sources have to be justified, meaning that it should produce more benefit than harm to individuals or to society. The best radiological protection option shall be selected for any source or practice, aiming to deliver the dose as low as reasonably achievable, taking into account social and economic circumstances; and, the sum of all individual additional annual doses that are attributable to all relevant practices should respect a dose limit. In relation to dose limitation, the Brazilian Standard Guide (BSG) recommends a dose limit of 1 mSv/y. In the scope of optimization this Guide refers only to nuclear activities and considers that a value of no more than 0.3 mSv in a year should be appropriate as a constraint for a single source, once the annual dose for a member of the public should be less than 1 mSv due to all practices (4) Practices are activities involving radiation sources with the aim of obtaining some benefit, despite the fact that can cause some additional dose to people, being a matter of a planned choice. On the other hand, during the last years, the radiation protection staff has been dealing with a different activity, which concerns radioactive residues from past activities or accidents. Intervention is a process that is intended to decrease the existing exposure situations both from radioactive residues produced by past-unregulated activities and accidental events, being a matter of health concern (5). The International Commission on Radiation Protection advises to deal separately with practices and intervention.

The principles of the System of Radiological Protection for intervention are the justification of intervention; the proposed intervention should do more good than harm, and optimization of the protective actions; the form, scale and duration of intervention should be optimized so as to maximize the net benefit. There is no recommendation of dose limit, although the IAEA recommends action levels for emergency situations and for chronic exposure, in particular for indoors radon inhalation pathway.

According to ICRP 82 (5), the use of a dose limit in existing exposure situations "might involve measures that would be out of all proportion to the benefit obtained and would then conflict with the justification principle". However the ICRP-82 recommends a level of existent dose of 10 mSv/y, below which intervention is optional but not likely to be justified and above which may be necessary. Intervention is not addressed in the BSG, presently under review.

An intervention at USIN site aiming to reduce the public dose is easily justified, because it would be also concerned with the decrease of public anxiety. Also, the reintegration of the site to the city, considering the economical status of the affected area and its surroundings, is a good reason for cleanup studies. Regarding to the dose level, as there is not any Brazilian Standard Guide concerning intervention, in a first approach, several criteria have been considered to derive intervention level and to assess the area to be decontaminated, such as 0.1 mSv/y as suggested for long-term residual dose, 0.3 mSv/y, as the dose constraint for a practice, 1 mSv/y, as the dose limit to members of the public due to practices, and 10 mSv/y, as the presently ICRP recommended intervention level.

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3.2 Scenario of land use

Adjacent properties at USIN site are primarily devoted to industrial activities, but the neighborhood has been changing nowadays and some service offices have been working at that district. Nevertheless, taking into account the long half-life of the radionuclides concerned, as well as the fast social change that usually occurs in Brazil, one may consider that some time ahead the district could also be used for housing purposes. Then, two exposure scenarios were considered:

Scenario a) considering the current use of the land, an industry located at the site would be a probable scenario. It was then considered a worker staying 2190 h/a at the site, being 1918 h/a working inside the plant and 272 h/a outside, in recreational activities. The thickness of the plant floor is 20 cm and it is situated over the contaminated areas.

Scenario b) the most conservative scenario is the occupation of the site by a family intruding the site. In this case, a child less than 10 years old remains 5840 hours/a (16 h/day) inside home and 1489 h/a (4 h/day) outside in the surroundings of the house, in recreational activities. An adult remains inside the house in home activities 7008 h/a (19 h/d) and 701 h/a (2 h/d) outside, in the surroundings. The house is a wood house (40% shielding) and has been built over the most contaminated area. Once the local groundwater is muddy water and the city is supplied by tap water, ingestion of water has not been considered for this scenario. Also, the ingestion of vegetables from the site was not considered, as it is not a realistic assumption for such large cities in Brazil. The dose due to radon inhalation was not considered, taking into account the ventilation rate in such house and the actual international recommendation for intervention level specific to this chronic exposure situation. Then, the main exposure pathways would be external exposure and dust inhalation and ingestion. The main parameters used for dose calculation are shown on Table 1. Inhalation and ingestion default are from Greenhalgh et alli (6), the other parameters are from Kennedy and Peloquin (3).

Table 1. Main parameter values used in the dose assessment.

Soil ingestion rate	Outdoor: 36.5 g/y
Dust loading for exposure period	Outdoor: 1.10^{-4} $g.m^{-3}$
	Indoor: 5.10^{-3} $g.m^{-3}$
Volumetric breathing rate	Adult: 8030 m^3/a
	Child: 5500 m^3/a
External dose rate factor (FDC)	Adult: 0.7 Sv/Gy
	Child: 0.8 Sv/Gy
Kerma rate in air (contamination uniformly distributed in the top 30 cm of soil)	U chain(^{226}Ra):adult: 2.73 10^{-7} $Gy.h^{-1}.Bq^{-1}.cm^3$ Child:0.077 Sv y^{-1} Bq $^{-1}$ kg
	Th Chain: adult: 3.77 10^{-7} $Gy.h^{-1}.Bq^{-1}.cm^3$ [3] Child: 0.106 Sv y^{-1} Bq $^{-1}$ kg
Soil density	1.3 $g.cm^{-3}$ (local data)

4 RESULTS AND DISCUSSION

Survey at USIN site.

Results of a preliminary radiological survey at the site confirmed the public concerns and showed surface and deep soil contamination from different materials including Cake II and Mesothorium cake. Basically the contamination was distributed in three areas (table 2 and figure 1):
Area 1: inside building C (9 m^2);
Area 2: surrounding building B (4800 m^2); and,
Area 3: surrounding the old football field (1750 m^2).
The range of ^{228}Ra and ^{226}Ra concentrations at these areas is showed on Table 2. It must be observed that the contamination is not homogeneously distributed at the areas, and some points of higher contamination levels can be found. The background level for this part of the town has been estimated as 59 Bq/kg of ^{228}Ra and 44 Bq/kg of ^{226}Ra.

Table 2. Range of ^{228}Ra and ^{226}Ra concentrations at the three contaminated areas found at USIN site

Area	^{228}Ra Bq/kg	^{226}Ra Bq/kg	$^{228}Ra/^{226}Ra$ Range	mean
Inside building C	153-16,500	50-1,150	12-91	54
Surrounding building B	387-33,100	65-6,710	0.9-12.4	3.8
Near soccer field	271-2,380	62-1,860	0.9-1.3	0.9

For the dose assessments, the concentrations of Th and U in soil, characterized by three different activity ratios $^{228}Ra/^{226}Ra$, were taken into account (table 2). For the two exposure scenarios the external exposure is the main radiation pathway, being responsible for 80 % of the dose; 60 % of this dose is due to radionuclides from the Th chain in the soil. As secular equilibrium in the decay chains was determined in a large part of the gamma spectrometry analyzed samples, it was decided ^{228}Ra to be the radionuclide clean up director, since ^{228}Ra can be easily measured by gamma spectrometry in laboratory and in field radiometric measurement. Besides, the limitation of one radionuclide will certainly result in the limitation of the others.
Considering the background level at site for both scenarios, the residual concentration levels for radionuclides in soil were derived, and are showed on Table 3.
Comparing the soil concentrations at site and the derived levels, it can be observed that except for the industry scenario (dose criteria of 10 mSv/y), all alternatives necessarily imply on the need for some cleanup at the site.

Table 3. Derived concentration levels (DCL) for the different contaminated areas, scenarios and dose criteria, taken into account the different ratio $^{228}Ra/^{226}Ra$ for each area.

Scenario	Dose criteria mSv/y	DCLs (^{228}Ra (Bq/kg))		
		Inside Building C	Surrounding Building B	Near to soccer field
Family intrusion	0.3	243	220	176
	1	672	596	449
	10	6193	5426	3959
Industry	0.3	1851	1593	1169
	1	6032	5173	3759
	10	59789	51199	37059

Considering that the site releasing for restricted use could increase the already existent anxiety in the population, decrease the economic value of the site and its surroundings and include government actions in order to keep control on the site to meet the required restriction, the chosen option was the unrestricted use. In this case the family intrusion scenario seems to be the most restrictive one and was adopted to derive intervention levels. As the derived values obtained for each dose limit in the three areas are similar, the rounded average was assumed as intervention level for each dose criteria. The area to be decontaminated according to these criteria can be seen in Figure 1. A line related to a 5 mSv/y criteria was also included, for illustration. As expected, the area to be decontaminated increases with the decrease of the dose criteria but the difference between a 10 mSv/y and a 5 mSv/y is small, showing the effect of “hot spots” on the over all contamination. Also the areas related to the lower dose criteria, 0.1, 0.3 and 1 mSv/y are not very different, as they are mainly related to the clean up of the same contaminating material.

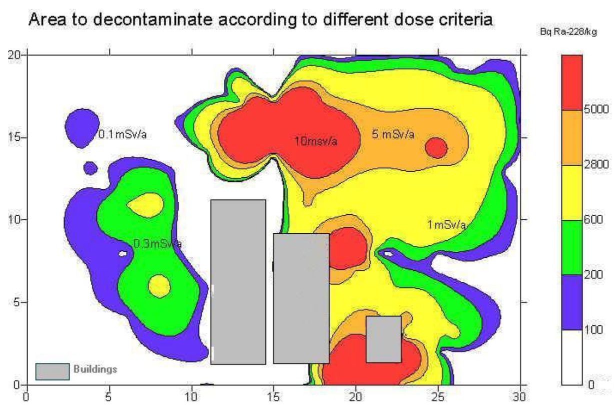

Figure 1: Areas to be decontaminated according to the different dose limits.

For unrestricted use of the site, the preliminary assessment pointed out a volume of around 400 m^3 of soil to be taken out of the site for the dose limit of 0.3 mSv annual. The soil volume to be taken out for dose limit of 1 mSv/y would be around 300 m^3. Due to the heterogeneous distribution of the contamination and the high concentration in some soil spots at the areas, for the dose limit of 10 mSv/y the volume will be smaller, around 50 m^3. However, for a real evaluation of soil volume to be taken out, a radiological survey on a smaller and specific grid on surface and in depth has to be performed.

This preliminary assessment point out to a optimized value of additional annual dose of about 0.3 mSv/y, once the form, scale and duration of clean up will be not very different from that for 1 mSv/y. It must be emphasized that unprocessed ore has been used as landfill to parts of the area and the activity concentration of this natural raw material is on the order of magnitude of that related to the 1 mSv/y criteria. The use of very low dose criteria, 0.1 mSv/y, would mean the clean up of amounts of material that may be considered as excluded. As so, the use of lower criteria would not be justified.

Although the use of the dose level of 10 mSv/y would lead to a smaller waste volume, the use of higher dose level values for USIN site intervention would increase the public anxiety concerning the contamination on the site, due to the lack of a Brazilian Standard Guide (BSG) for intervention and the BSG recommendation that a dose limit of 1 mSv/y should be applied to members of the public.

The generation of low activity wastes results in a new problem and costs. According to BSG, wastes with concentration less than 74.00 Bq/g can be stored in municipal landfills. So, most of the soil taken out of the site could be sent to one of the four sanitary landfills close to the site in São Paulo city.

5 CONCLUSION

An intervention would be the recommended approach regarding the USIN site although the system of radiation protection for intervention is not yet included into Brazilian standards and recommendations and its use would lead to misunderstanding and public concern about the level of dose post-remediation. The residential scenario should be preferred over the industrial one, since it addresses the unrestricted release of the site. Taking into account the public concern and anxiety, the site location and size, it is suggested that an additional annual dose to a member of the public should be no higher than 1 mSv/y. Due to the observed distribution of contamination at the areas, this preliminary assessment points out to the possibility of optimize the post remediation additional dose to close to 0.3 mSv/y. However, this optimization procedure would require a more detailed radiological survey at the site.

6 REFERENCES

1. Paschoa A.S., Radiation Protection in Australia, Special Issue, Vol.11 (1993), No.4, 170-173.

2. Till,J.E., Moore, R.E., Health Physics, 55 (1988) No.3, 541-48.

3. 3. Kennedy,W.E. Jr., Peloquin., Richland, Pacific Northwest Laboratory, (1990). NUREG/CR--5512.

i. 4. Diretrizes Básicas de Radioproteção. Rio de Janeiro, CNEN
(1988),Norma CNEN-NE-3.01.

5. International Commission on Radiological Protection, (1999) Publication 82.

6. Greenhalgh J.R., Fell T.P. and Adams N., Chilton (1985), National Radiological Protection Board, (NRPB-R162), 1985.