Remediation of NORM/TENORM Contaminated Sites in Brazil

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Abstract

Today, the decision about clean-up of Brazilian sites contaminated with NORM/TENORM residues is addressed on a case-by-case basis, since there is no general guidance for actions soon after the problem has been identified. The Brazilian Environmental Agency established background value prevention and intervention as the first step in implementing remediation actions based on human risk assessment, which are only applied to sites contaminated with chemical elements. These values were based on the Soil Screening Guidelines (SSG), which is a framework for developing risk-based soil screening levels (SSLs) to protect human health. This paper describes the methodology used for developing the Soil Screening Guidelines for radionuclides, The SSG goals have been applied in preliminary remediation in Brazilian sites contaminated by NORM/TENORM. Background values of ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb were determined by analyzing different types of soil that are typical of the Southeast region of Brazil. Intervention and action values were estimated using typical exposure scenarios that could represent possible future uses of the soil after the remediation action. They included possible residential, agricultural, and industrial uses. Dose factors and soil-plant transfer coefficients were shown to be the most sensitive parameters of the proposed model.

Keywords: NORM, remediation, soil contamination.

1. Introduction

The Earth's crust contains radionuclides that are a major source of Naturally Occurring Radioactive Materials, NORM. Most of these radionuclides belong to the decay chains that begin with ^{235}U, ^{238}U and ^{232}Th. Some industrial processes that deal with natural resources concentrate this radionuclide to a degree that can be a health risk to people and the surrounding environment if they aren't controlled.
However, in recent years many industrial installations have increased the proportion of this NORM in end products, concentrates, or waste. These facilities are defined as a place in which the raw material that contains radionuclides of the natural uranium and/or thorium series are processed or manufactured, including waste piles and waste storage areas (CNEN, 2005).
In Brazilian territory, which covers 8.5 million km2, the types of installations that process ore containing significant amounts of uranium and/or thorium can be classified into the following groups (PONTEDEIRO, 2006):

- Nb/Ta carbonatite and pegmatite deposits (columbite/tantalite);
- Deposits of Sn, Nb/Ta, and Zr in intrusive granitic bodies;
- Deposits of Zr, Ti, and monazite sand;
- Phosphate deposits;
- Deposits containing copper in the Carajás mining province.

Nb/Ta carbonatite and pegmatite deposits (columbite/tantalite)

In Brazil, at least four carbonatite structures from the cretaceous age are known. They contain niobium deposits. Two of these deposits are being mined and are responsible for 70 % of world niobium production. The minerals belong to the pyrochlore group. The carbonatites underwent intense weathering which facilitates open pit mining.
One of the deposits, located in the state of Minas Gerais, has reserves of around 460 million tons of ore with an average yield of 2.5% to 3.0% Nb_2O_5 and levels of up to 0.008% U_3O_8 and 0.13% ThO_2, which are concentrated during niobium processing. In the other mine, located in the state of Goias, the reserves are around 10.2 million tons with an average yield of 0.9% Nb_2O_5. The pyrochlore ore contains up to 0.03% U_3O_8 and 0.05% ThO_2. Some areas of these carbonatites have the highest levels of natural radioactivity in Brazilian territory (PONTEDEIRO, 2006).

Deposits of Sn, Nb/Ta, and Zr in intrusive granitic bodies

The main sources of columbite/tantalite ore are in the pegmatite province located in the states of Bahia and Minas Gerais and in the northeast region (in the states of Rio Grande do Norte and Paraíba). This ore is generally extracted by prospectors and exported after it's been concentrated. Yields of radioactive elements in the ore reach 1% U_3O_8 and 1.8% ThO_2, but they are normally much lower (HEIBRON et al., 2001)

Deposits of Sn, Nb/Ta, and Zr in intrusive granitic bodies

Located in the northern part of the Amazonian Craton in the state of Amazonas, these deposits are related to two granitic intrusions from the middle Proterozoic (± 1,700 Mya). In these areas, the ore has average yields of 0.176% Sn, 0.808% ZrO_2, 0.223% Nb_2O_5, 0.29% Ta_2O_5 and reserves of around 10,000 t of uranium and thorium, with 0.03% uranium and 0.13% thorium.

Deposits of Zr, Ti and monazite sand

In the coastal region of the north and northeast of Brazil, from the state of Rio de Janeiro to the Amazon river valley, parallel to the coast there are continent-spanning sediments from the tertiary age, grouped together with the name the Barreiras Formation. These sediments are made up of sand, red clay, and banded iron concretions. They come from erosion of gneissic and granitic rocks in the Brazilian Shield and contain minerals such as zirconite, ilmenite, rutile, and monazite. The zirconite has average yields of 0.03% U_3O_8 and 0.016% ThO_2 (PONTEDEIRO, 2006). In some areas, there are "black sand stains" with naturally high radioactivity. Guarapari beach, in the state of Espirito Santo, is known to be one of the places on the planet with the highest levels of natural radioactivity.

Phosphate Deposits

The majority of the phosphate deposits being processed in Brazil are related to carbonatites located in areas around cratons, bordering the great sedimentary basins, mainly in the area of

the Parana basin in the south and southeast regions in Brazil. They are almost all from the cretaceous era, are circular, and are relatively small, from 6.5 to 65 km^2.
Some of the phosphate deposits are in the same niobium pyrochlore carbonatite deposits mentioned above. This phosphate ore has yields of up to 14% P_2O_5, averages of up to 0.0185% U_3O_8 and 0.0480% ThO_2, reaching ^{226}Ra concentrations around 3000 Bq kg^{-1}.
In Brazil, phosphate deposits in sedimentary rock aren't very common (SAUEIA et al., 2013). Metaphophsorites with low levels of metamorphism from the upper proterozoic were enormous deposits, but with low yields of uranium and thorium. There is a significant exception, the Itataia deposit in the state of Ceara from the lower proterozoic, with yields of up to 26% P_2O_5 and 0.19% U_3O_8. In this deposit, the uranium is a co-product of phosphate and the industrial/mineral complex can be considered to be a nuclear facility.

Carajas Mineral Province Copper Deposits

The Carajas mineral province is located in the state of Pará in the north of Brazil. It has one of the largest iron ore reserves in the world, as well as significant deposits of manganese, nickel, and copper/gold. The mineral ore is calcopyrite with some anomalies: copper, between 0.5% and 11%; gold, between 0.5 and 15 ppm; and uranium, between 28 and 380 ppm. Near Salobo, concentrations can reach 500 ppm U_3O_8 (TALLARICO et al., 2000).

Mining and processing this ore, whether it be tin, phosphate, niobium, monazite, or rare earths, generates large amounts of NORM residues. Obviously, improper disposal of this material can cause situations that result in contaminated areas. These areas can lead to problems including damage to human health because of unneeded exposure, compromised water resources, restrictions to land use, and damage to public and private land resulting in reducing the value of the property, as well as damaging the environment.
A contaminated area is defined as an area, lot, locale, installation, building, or natural area where there is significant human traffic such as a park, that contains levels or concentrations of any waste substance at levels that are a risk to human health, the environment, or any property, in which the material has been deposited, accumulated, stored, buried, or infiltrated in a planned, accidental, or even a natural way.
The origin of contaminated areas is mainly related to past lack of knowledge about safe procedures in dealing with hazardous substances. The procedures didn't exist and so weren't obeyed, and accidents or leaks took place throughout the production process, transport, and storage of raw material, processed material, and waste. This is most significantly in the case of NORM installations, because often neither the investors nor the general public knew of the generation or inadequate disposal of waste that contained uranium, thorium, and their decay products.
Soil and ground water are the priority when management of contaminated areas is considered. As well as being property that is extremely important to protect, they are the main ways that contamination propagates to other resources. Specifically, the practice of using open-air stockpiles containing radionuclides must take the soil, water, and air into account as critical pathways that increase the dosage estimate. The chemical availability of radioactive isotopes is affected by the concentration and characteristics of chemical species present in the soil impregnated in the final environmental destination (US EPA, 2000).
Contaminated areas are managed in order to minimize the risk that the populace and the environment are subjected to by means of a set of measures that assure that there is knowledge about these areas as well as their impact. This provides the information needed for the decision to be taken as far as appropriate means of intervention.
The main objective of this study takes this into consideration. It is to introduce and discuss a methodology for evaluation and remediation of NORM contaminated areas. In this case, the

calculation of the dose was chosen as the parameter that intervention and prevention numbers would be based on, since radiation protection guidelines that are presently in use in Brazil are explicitly based this parameter.

2. Materials and Method

The management method for contaminated areas that is proposed in this study is based on a strategy of sequential steps in which information obtained in each step is the basis for carrying out the next. The objective of this is to optimize technical and economic resources. For this reason, two processes have been defined as the basis of management of contaminated areas: the identification process and the remediation process.

The primary objective of the process of identifying contaminated areas is defining their location, as laid out in four steps (CETESB, 2005):

- Defining the study area;
- Identification of the main contaminated areas;
- Preliminary evaluation;
- Confirmatory investigation.

Because of the information for each of the study areas, they are classified as one of three types when the identification process is carried out: potentially contaminated areas (PAs); suspected contamination areas (SAs); or contaminated areas (CAs).

The main objective of the recovery process for contaminated areas is adoption of corrective measures in these areas that allow them to be recovered for a use that is compatible with the goals established that are to be met after the intervention. The first step of the rehabilitation process of a contaminated area is made up of detailed investigative studies and risk evaluation. The detailed investigation is the management step for contaminated areas in which the characteristics of the source of contamination and the affected environment must be evaluated in order to obtain data to carry out the risk evaluation and define the rehabilitation project for the contaminated area. Risk evaluation will aid in determining the need for remediation in function of the present and proposed use of the area. It will be the basis for the establishment of acceptable remediation levels for the usage conditions and occupation of the soil in the area and vicinity and the best remediation techniques for each case.

Contamination control and management of contaminated areas under investigation use Guideline Values as a prevention instrument. Guideline Values determine concentrations of chemical substances that provide guidelines about soil quality conditions. In practice, three guideline values are defined for soil and ground water (CETESB, 2005). They are:

Soil Quality Reference Value (SQRV) is the concentration of a specific substance in the soil or ground water. It defines a soil as clean or ground water quality as natural.

Prevention Value (PV) is the concentration of a specific substance above which there can be harmful alterations to soil quality. This value indicates soil quality that is able to sustain its main functions, protected from ecological receptors.

Intervention Value (IV) is the concentration of a specific substance in the soil above which there are potential direct or indirect risks to human health, considering a generic exposure scenario. For the soil, scenarios have been developed for Agricultural, Residential, and Industrial exposure.

Determination of Soil Quality Reference Values (SQRV) for natural radionuclides.

Today in Brazil, only the state of Minas Gerais has estimated Soil Quality Reference Values for naturally occurring radionuclides of ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra and ^{210}Pb. The state has some unusual lithographic characteristics and unusual characteristics of soil genesis and morphology. These characteristics have a preponderant influence on the geochemistry of trace elements and radionuclides. Around 70% of the soil in the state is highly weathered Oxisols, Ultisols and Alfisols. Weathering of some soil in the state is limited by the originating material, such as Quartzipsamments. Approximately 17% of the remaining soil belongs to the Cambisol class, which often has latosol mineralogy. Clay that is barely reactive predominates in this soil, such as kaolinite associated with iron and aluminum oxides and hydroxides (PEIXOTO, 2013).

To carry out the field work, different parts of Minas Gerais were travelled on the major highways, roads, and accesses. Geological and lithological formation of the soil was used as a control parameter in order to minimize and optimize the number of samples used to determine natural radionuclide concentration values. 120 soil samples were collected in total. First, campaigns were carried out to classify and survey the main soil classes. Collection was carried out randomly within each map unit, observing the predominance of preserved or minimally impacted vegetation, for example native pasture. Later, compound samples were taken at some points in order to evaluate if they were representative and validate the simple sample collection.

Soil samples were taken at depths between 0 and 20 cm, which is equivalent to horizon A for most soil. After the samples had been dried, they were sieved through a 2.0 mm nylon mesh screen. This process produced fine air-dried soil (FADS). After they had been sieved, the samples were placed in a plastic recipient and sent for analysis for activity concentration of ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb.

Uranium and Thorium concentration were determined by spectrophotometric method using Arsenazo III as a reagent (SAVIN, 1961). Gamma spectrometry was used to measure activity concentrations for ^{226}Ra, ^{228}Ra and ^{210}Pb that are present in the soil. Samples were dried and ground and stored in sealed plastic flasks. In order to measure the activity of ^{226}Ra and ^{228}Ra, the samples were analyzed for thirty days after being sealed. This is long enough to ensure radioactive equilibrium between ^{226}Ra its byproducts ^{214}Pb and ^{214}Bi. In general, this takes 40 days. Equilibrium must be achieved because the direct energy of the gamma rays from ^{226}Ra (E = 186.1 keV) is very near that of ^{235}U (E = 185.7 keV). ^{210}Pb was determined by gamma spectrometry (SIQUEIRA, 2009). A low-energy system was utilized that was appropriate for gamma emission less than 100 keV. This is the case of ^{210}Pb, which has a characteristic photopeak of 46.5 keV. Control samples containing radionuclides with known activities were analyzed along with the samples in the study. IAEA/Soil7 reference soil from IAEA was used for this purpose.

Determination of Prevention and Intervention Values

Prevention and intervention values were determined as specified by Peres (2007). In order to do this, data referring to intervention values and suggested action in Regulatory Position 3.01/007, Intervention and Action Levels for Chronic Exposure (CNEN, 2011), were used. This regulation states that, "*generic intervention levels are reference levels established by CNEN a priori which should be considered in the planning phase and utilized in justification and optimization processes for intervention situations. An annual existing dosage of 10 mSv should be used as a generic reference value for interaction action in situations of chronic exposure to members of the public. The estimate or measurement of an existing dose reference value above 10 mSv yr^{-1} always requires evaluation to decide whether to implement*

protective measures or remediation. The decision to apply a specific protective or remediation measure must take into account the current situation. It must be the result of an evaluation in which the measure is justified.
The estimate or measure of an existing dose level that is lower than the generic reference value doesn't normally justify interventions for chronic exposure scenarios. However, there can be situations in which intervention to reduce one or more components that make up the total existing dose are justified. This can be as a result of an optimization process or when protective actions to reduce those components are very simple and justified. CNEN doesn't establish an upper level for intervention that defines when a protective measure or remediation is mandatory, but always considers intervention when the existing dose is above 50 mSv yr^{-1}".
For this reason, the intervention value in this study corresponds to concentration in the soil of a given natural radionuclide that will result in an effective dose of 50 mSv yr^{-1} to an individual in the public. Similarly, the prevention value was defined in function of a dose of 10 mSv yr^{-1} to a member of the public. The choice of these values is in agreement with nuclear legislation active in Brazil.
Prevention and intervention values were determined for ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb radionuclides, which are important for NORM facilities. A statistical distribution of the values obtained was considered. A value of 95 % of dosage distribution was used as the basis for defining recommended intervention and prevention values. As established by Peres (2007), the doses were calculated for two population groups: adults; and children of 10 years of age. Contamination of the soil was considered to be finite and homogenous. Three distinct exposure scenarios were considered:
Agricultural: This is related to green belts and rural areas where there is economic activity related to agriculture, forestry, and pastures. In this scenario, a rural worker resides in a hypothetical area, remaining 24 hours a day in an exposed area for his entire life. Included in this scenario are: large farms, small farms or ranches, houses in rural areas mainly used for recreation, rural residences, fishing areas, and protected springs.
Residential: This is related to living in residential areas in urban environments. Individuals spend most of their time in the area, with exposure throughout their lives. Ingestion of contaminated soil, water, and plants is lower than in the agricultural scenario. This scenario includes all kinds of residences, areas where there is some cultivation of fruit and vegetables, residential building with multiple floors, and establishments such as recreation centers, schools, daycare centers, hospitals, clinics, parks, and green urban areas, bed and breakfasts, hotels, and rest homes.
Industrial: This is related to areas where industrial and commercial activities predominate where people remain for working hours. In this scenario, the adult is exposed for part of the day (during working hours), with children present in a restricted period such as parties and special events. Exposure pathways considered include: external exposure to radiation; inhalation of particles; ingestion of soil; ingestion of ground water; and ingestion of foodstuffs (agricultural products, meat, and milk).
Models utilized to estimate intervention and prevention values were suggested in Safety Series 19 (IAEA, 2001). In addition, soil usage and occupation factors adopted by CETESB were used as a basis for calculations, such as time in the contaminated area for each scenario, inhalation rate, and food and water ingestion rate (CETESB, 2005). Dose coefficients recommended by CNEN Regulatory Position 3.01/011:2011 were utilized for members of the public.

3. Results and Discussion

Soil Quality Reference Values (SQRV)

SQRV for the radionuclides in this study in the soil in Minas Gerais were based on statistical analysis of the data applied to the two sets of concentration results. In order to do this, the third quartile was used to represent 75 % of the soil radionuclide concentration values. Final results are shown on Table 1.

Table 1. Soil Quality Reference Values (SQRV) in soil in the state of Minas Gerais.

Radionuclide	SQRV (Bq kg^{-1})	Range (Bq kg^{-1})	Average (Bq kg^{-1})	Mean (Bq kg^{-1})
^{238}U	101.6	7.8 – 334.3	84.8	73.0
^{232}Th	75.7	8.1 – 184.0	58.1	49.0
^{226}Ra	66.3	6.6 – 229.6	48.9	37.2
^{228}Ra	89.9	7.5 – 198.3	61.0	54.0
^{210}Pb	83.7	15.6 – 188.9	63.0	55.2

As can be seen, concentrations of ^{238}U present in the soil were higher than concentrations of ^{232}Th. This fact can be explained by the intense weathering process that most of the soil in the state of Minas Gerais has undergone, as well as the actual chemical and geochemical characteristics of the two elements. In oxidizing surface environments, in general Th^{+4} remains unaltered and immobile, remaining close to the rock of origin, while U^{+4} oxidizes to U^{+6}, forming uranyl ion $(UO_2)^{2+}$, which confers a large amount of geochemical mobility. The highest activity concentration values for the radionuclides studied were found in regions of the state of Minas Gerais where there are radioactive anomalies, such as: the pegmatite and alkaline magmatic provinces (PEIXOTO, 2013).

Intervention and Prevention Values

Table 2 shows the intervention and prevention values derived for each of the radionuclide of interest in this study. As can be seen, Intervention and Prevention values are much higher than the Soil Quality Reference Values for each of the radionuclide being studied even in the most restricted scenario, agricultural land use.
Application of these values contributes to a preliminary evaluation of areas suspected of being contaminated. An area is classified as a Contaminated Area under Investigation when it can be shown that the radionuclides studied here are at concentrations in the soil higher than the Intervention Values, showing the need for action to protect those at risk. Procedures for management of contaminated areas should be followed.
Tables 3 shows some examples of soil activity concentration results of soil samples collected inside NORM industries, as well as samples collected in the surrounding environment of these facilities.

Table 2. Prevention and Intervention Values for each scenario (Bq kg^{-1})

Radionuclide	Intervention Value (Bq kg^{-1}) (for an effective dose of 50 mSv y^{-1})			Prevention or Action Value (Bq kg^{-1}) (for an effective dose of 10 mSv y^{-1})		
	Agricultural	Residential	Industrial	Agricultural	Residential	Industrial
^{238}U	130000	360000	470000	26000	72000	94000
^{232}Th	190000	190000	65000	38000	38000	13000
^{226}Ra	4500	9000	60000	900	1800	12000
^{228}Ra	750	1700	31000	150	340	6200
^{210}Pb	8500	19000	170000	1700	3800	34000

Table 3. Soil analysis results of activity concentration of soil samples collected inside and surrounding some Brazilian NORM industries.

Company	Local of sampling	^{238}U Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{228}Ra Bq kg^{-1}	^{210}Pb Bq kg^{-1}
A	Areas located inside the facility	6000 ± 300	9100 ± 500	4700 ± 200	7800 ± 200	4900 ± 300
B	Areas located inside the facility	80 ± 20	160 ± 0,02	535 ± 30	143 ± 12	418 ± 23
C	Surrounding environment (rural zone)	130 ± 20	150 ± 20	60 ± 9	460 ± 30	720 ± 50
D	Surrounding environment (rural zone)	70 ± 20	120 ± 20	120 ± 20	100 ± 10	80 ± 20
E	Surrounding environment (rural zone))	90 ± 20	130 ± 20	115 ± 7	128 ± 10	135 ± 10

Note that activity concentration values vary significantly from one company to another, especially for results for soil collected inside the facilities. This is generally due to waste disposal procedures that have been adopted. In many cases, the waste is still disposed of in the open air in the places where it is generated. Evidently, various efforts have been made to manage the waste disposal in these industries. Depending on the activity concentration values, the waste can be sent to landfills, which generally requires a series of studies and negotiations because of the presence of radionuclides. Another option that has been chosen is construction of repositories within the facility itself which are utilized to temporarily store the waste. This solution is generally costly for the investor since in addition to building the repository, the practice involves carrying out a safety analysis study that requires surveys of many parameters in order to estimate the risk considering future scenarios of human exposure to radiation.

4. CONCLUSIONS

The formulae and assumptions about exposure used to develop the screening levels in this work are based on the Intervention and Prevention or Action Levels for Chronic Exposure established by CNEN (CNEN, 2005). These levels are then compared to on-site soil radionuclide levels. Generally, areas of a site which fall below the screening levels may be eliminated from further assessment. Areas above the screening levels generally warrant further evaluation of the potential risks that may be posed by radionuclides to determine the need for responsive action. The guidelines recommend use of a screening tool in which assumptions made in developing the tool are consistent with conditions found to determine if further study of specific portions of a site is warranted. The levels should not be interpreted to represent cleanup standards for a site.

Application of the methodology proposed in this study during the preliminary evaluation phase of an area that is suspected of contamination in NORM facilities in Brazil has been shown to be efficient, fast, and have a low cost. It allows: 1) confirmation of presence of contaminants (in this case radionuclides of the natural series of ^{238}U and ^{232}Th); 2) determination of the extension of the contamination (when possible) and 3) establishment of the concentration range (magnitude) of the contamination.

In addition to this information, the methodology allows the definition of important points in the remediation process, such as mapping the greatest concentration points (hot spots), definition of location of future probes and monitoring wells, *a posteriori* definition of a detailed characterization of the affected area, and also orientation about future monitoring and recuperation plans. In this case, the actions are only carried out if the values found through screening the analyses of samples greatly exceed the limit values for specific contaminants being studied. Without this, the values obtained must be compared with the natural background levels.

Studies developed in Brazil have shown that while a lot of progress has been made in the area of radiation protection and remediation actions, it is still very important to discuss things related to NORM waste management. It must be highlighted that responsibility for the environment can be considered a competitive advantage for companies. The image of a company that values a healthy environment leads to better acceptance for stockholders, consumers, suppliers, and public agencies. It is also relevant today in the financial analysis of the company and in requirements of Brazilian and international law. Today, environmental damage, whatever its source, is often used as an item in evaluating mergers, acquisitions, and privatization of companies. Reducing environmental damage definitely leads to financial benefits.

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