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DEALING WITH A FORMER RARE EARTH SEPARATION FACILITY SITE (USIN): CASE OF A CONTAMINATED SITE IN SÃO PAULO CITY, BRAZIL

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ABSTRACT

São Paulo city has currently 2,224 contaminated lands which are at different stages of the environmental management process. It is possible to find areas developing environmental analysis and investigations, fields with confirmed risk to human health and without an approved remediation plan, even places during the remediation process and revitalized spaces, which could be an otherwise land use. The USIN facility stopped their operation in 1992 and was initially used to store minerals and residues from the processing of minerals. Furthermore, was equipped to process rare earth compounds using solvent extraction techniques, and also store by-product material consisting of 2% of U_3O_8 and 20% of ThO_2 concentrate. The former facility site consisted of 3 sheds constructed in an area of 60,000 m^2 located in a past urban industrial district, Campo Grande, in São Paulo city. Over the years, Campo Grande district was changing its land use, from industrial use to residential and commercial use, hence the number of residences in the neighborhood of USIN has increased. According to São Paulo Environment Agency (CETESB), USIN's area was classified as an area with confirmed risk. This study approach chemical and radiological contaminants present in the area, also the remediation actions adopted. Despite CETESB classification, results indicated than the area has been remediated, a segment of 18,400 m² it was already cleaned-up, and the remainder currently is in the remediation process

1. INTRODUCTION

Over the years, the anthropic activities have caused degradation and environmental contamination, mainly in cities whose history has been marked by the development of the industry. According to United Stations Environmental Protection Agency (USEPA) contaminated land is a result from a diversity of intended, accidental, or naturally occurring activities and proceedings such as manufacturing, mineral extraction, abandonment of mines, national defense activities, waste disposal, accidental spills, illegal dumping, leaking underground storage tanks, hurricanes, floods, pesticide use, and fertilizer application. These areas can present of varying size and significance in sceneries ranging from abandoned buildings in inner cities to large areas contaminated with toxic materials from past use. Contaminated lands include sites contaminated by inappropriate handling or disposal of toxic and hazardous materials and wastes, also sites where toxic materials may have been deposited as a result of natural disasters or acts of terror [1]. Therefore, contaminated land affects the soil quality and can also implicate the groundwater quality, according to the contaminant behavior.

As for contaminated land, Brazil has a law with criteria and guiding values of soil quality about the presence of chemical substances and guidelines for the process of managing contaminated areas [2]. According to this law contaminated land refers to places where there are the presence of chemical substances above the reference values and due to anthropic activities which limit the use of this environmental resource either for current or future use [2], [3].

Regarding guiding values of soil quality, these are defined according to international standards and considering the exposure scenarios and Human Health Risk Assessment. The exposure scenarios can be rural, residential, and manufacturing, each one has different reference values. There are three sorts of reference values: reference quality, is the concentration of a substance that defines the natural quality of the soil; prevention, is the limit concentration of a substance in the soil, such that soil is capable of sustaining its primary functions; investigation, is the concentration of a given substance in soil or groundwater above which there are potential direct or indirect risks to human health [2].

Establishing the reference values, if an area has substances above these values, the process of managing contaminated areas is initiated, according to the federal law. Under Environment Agency of São Paulo State's (CETESB) report, in 2018 São Paulo states currently has 6,110 contaminated and rehabilitated areas whereas São Paulo city owns 2,224 areas [4].

Lots of historically contaminated sites, particularly those associated with product manufacture, are in urban areas. These sites relied on easy access to highways and railroads. In some cases, the sites have initially been in rural areas, but urban development has subsequently trespassed on or enveloped the sites [5].

Around the world, many sites were contaminated by human-made radionuclides or materials containing naturally occurring radionuclides (NORM). Sites that have not been operated with satisfactory controls or because of past operations without regulatory control, and now pose a problem for governments for a number of reasons, including location, the possible need for remedial action, and difficulties in assessing the possible health and environmental impact of such sites, because of this, requires clean-up operations [6].

Natural radionuclides are ubiquitous in the human environment. All minerals and raw materials contain radionuclides of natural, terrestrial origin. The main radionuclides of interest are those from the ^{238}U and ^{232}Th decay series and ^{40}K. Certain minerals contain uranium and or thorium series radionuclides at significantly elevated activity concentrations. Furthermore, during the extraction of minerals from the Earth's crust and subsequent physical and or chemical processing, the radionuclides may become irregularly distributed between the various materials resulting from the process. Selective mobilization of radionuclides can disrupt the original decay chain equilibrium.

As a result, radionuclide concentrations in materials arising from a process, including process residues, may exceed those in the original mineral or raw material, sometimes by orders of magnitude. NORM is, by definition, a naturally occurring radioactive material. Industrial activities involving NORM, aside from the mining and extraction of uranium, are extraction of rare earth elements; production and use of thorium and its compound; production of niobium and ferroniobium; mining of ores; production of oil and gas. Consequently, all the industrial activities cited generate or have the potential for generating NORM residue. An example is the processing of heavy mineral sand, which is recovered from beaches and dunes and is a principal worldwide source of minerals such as zircon, ilmenite, rutile, and monazite [5].

The most important sources of rare earth elements are monazite (Ce, La, Nd, Th) PO_4 and bastnaesite. The monazite ore concentrate is obtained from suitable sands by a gravimetric and electromagnetic sorting [7]. The Brazilian monazite sand is basically composed of four minerals: ilmenite ($FeTiO_3$); zirconite or zircon ($ZrSiO_4$); rutile (TiO_2) and monazite, a rare-earth orthophosphate containing up to 6% ThO_2 and 0.3% U_3O_8. The most notable deposits of monazite sand in Brazil were found in the Brazilian coastal area, in the north of the state of Rio de Janeiro. The chemical processing of monazite was carried out in Brazil until 1994 to produce rare earth chlorides. The process generated two by-products, both containing most of the thorium and uranium, initially contained in the monazite concentrate [8].

In 1949, started the chemical processing of monazite to produce lanthanide chlorides and tri-sodium phosphate at the Santo Amaro mill (USAM), located in the city of São Paulo, Brazil. The first phase of the process consisted of the extraction, washing, and drying of monazite bearing sands taken from beaches. The next step was a physical separation process to slip the four minerals: ilmenite, rutile, monazite, and zircon. The heavy mineral separation processes performed at a mill located close to the monazite bearing sand deposits. After that, the monazite's concentrate (90–95%) was sent to USAM to be chemically processed. In Figure 1 the basic steps of the USAM monazite chemical processing are shown [9].

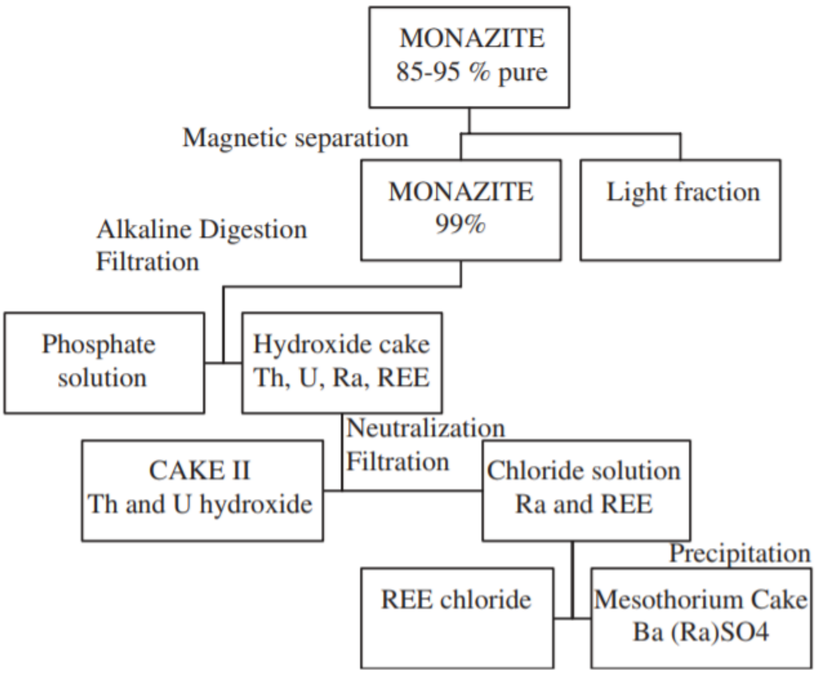

Figure 1: steps of the monazite chemical processing [9]

Thus, as a consequence of the monazite processing, basically three different kinds of wastes were produced: (1) a light mineral fraction, from monazite physical purification; (2) the Cake II (average content 20% of thorium hydroxide and 1% of uranium hydroxides), from the monazite alkaline digestion; and (3) the mesothorium cake ($Ba(Ra)SO_4$). In consequence, disposal sites had to be set to store the monazite's wastes and residues [9]. USAM stopped its activities in 1992, decommissioning started in 1994 and finished in December 1998[10].

Interlagos factory site (USIN) is in an area of 60,000 m^2 located in the Interlagos neighborhood, a former industrial district in the south region of the São Paulo city. The plant was installed in a shed and starts operating in 1989, performing the separation of rare earth elements (REE), produced by USAM, into light and heavy groups by a chemical process. Figure 2 shows an old picture to USIN site.

Figure 2: USIN site[9]

The USIN area was also used by the company to deposit different materials and wastes from the physical treatment of the ore[9], [10]. These residues were classified as silica and used as backfill on USIN site. However, they had a source of contamination in the area. In 1992 a radiometric survey of the area revealed the existence of soil contamination. The investigations led to the conclusion that during the operational phase of the installation, there was leakage of contaminated solvent that reached groundwater. The installation was deactivated, and the equipment transferred. In March 1994, the USIN's sheds after adaptation were classified as Radiative Installation and received the rejects resulting from the decommissioning of the USAM[10].

Concerns related to contaminated areas are an essential theme for sustainable city planning. Therefore, the actions developed to deal with the soil contamination in a radiative installation have high relevance. Hence, the purposes of this research are identifying the chemical and radiological contaminants present in the area, and the remediation actions adopted to diminish the risks for human health.

2. METHODOLOGY

This study is a literature review and qualitative exploratory research, with information obtained from secondary sources. A survey was carried out to find publications and gathering information regarding the industrial activities developed by USIN and what is the current scenario in the area. A request for Environment Agency of São Paulo (CETESB) was made aiming to view administrative file nº228/11, and process nº 33/00882/10, a contaminated area process of Indústrias Nucleares do Brasil (INB) – USIN, in 05/30/2019. The request for process visualization based on Federal Law 10.650, of April 16, 2003[11], which provides public access to data and information in the organs and entities that belong to Sisnama. The process view was authorized in 06/11/2019 and done in 17/06/2019, in Santo Amaro environmental agency.

3. RESULTS AND DISCUSSION

According to the information available on INB's website[12], the USIN (Interlagos Plant) is, located in Jurubatuba neighborhood and installed in an area of 60,000m². It is used to storing residues and wastes of low radioactivity, resulting from the deactivation of the former Santo Amaro Plant (USAM), an amount of 1,757 tons of materials, among which are Cake II, equipment, parts, and tools of the old factory. Figure 3 shows USIN site in 2019.

Figure 3: USIN site 2019[13]

In the past, there were three sheds on the site. Sheds A and B temporarily stocked samples of ores obtained from uranium prospecting. Shed C stored empty metal drums and the Rare Earth separation unit worked in shed A.

A report in the CETESB's administrative file nº228/11 and process nº 33/00882/10, USIN's land was acquired in 1960. Currently, there is only one shed, Shed A, with six internal compartments containing waste and residues from USAM and waste from the USIN land decontamination process.

From 1989 to 1992 the industrial unit operated, which performed the separation of rare earth by extraction with solvents. The production process used rare earth solution, diethyl-hexyl phosphoric acid, hydrochloric acid, and liquid ammonia. In 1995 shed A was reformed and converted into an initial deposit of radioactive waste. Between 1998 and 2002 the land was partially decontaminated and sheds B and C were demolished. During the years of 1995 until 2006 several surveys were carried out to sample groundwater and soil.

In a report delivered in 2009 to CETESB, contamination in groundwater was identified. As a result of a leakage in the effluent separation box in shed A, there is a plume of contamination in groundwater, with the presence of Cl, SO_4, NO_3, U, ^{226}Ra, ^{228}Ra and ^{210}Pb.

As provide for a report delivered by INB in December 2010 to CETESB, the land was backfilled with silica contaminated by heavy minerals, including monazite, waste from the beneficiation process carried out by USAM, as it was a wetland region. The backfill provides soil contamination with As, Ba, ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{210}Pb.

In 2009, the decontamination plan presented by INB was approved by CNEN, through the PR/CNEN Ordinance No. 087. The plan foresees, in addition to decontamination, the removal of wastes from the shed to another location. In June 2010, the city of São Paulo issued decree no. 51,579, declaring a part of the USIN's land of public interest, to open a connecting street between Avenida Miguel Yunes and Avenida Interlagos, which would promote the expropriation of part USIN’s site.

After presenting the investigations carried out to CETESB and receiving authorization from CNEN to decontaminate the area, the remediation process began. For groundwater contamination, water was pumping and treated. The effluent was stored. Concerning the soil contamination, was adopted soil excavation. A survey of the hotspots and the delimitation of the contaminated area was carried out. A part of shed A received the fractions of soil removed that had presented ^{226}Ra and ^{228}Ra activity above 30Bq/g, stored as radioactive waste. Dispersion of contamination beyond the USIN site was not observed. The company worked in the soil decontamination of the areas outside the shed in order to deliver to São Paulo’s city hall.

INB carried out the remediation of a wide strip of land of 40 meters in the USIN facility site. The strip is in the USIN border with its neighboring. The total area remediated amounts to approximately 18,400 m^2. In December 2013, CNEN published Resolution No. 161, releasing for unrestricted use the USIN remedied land strip and in the interest of the São Paulo’s City Hall [14]. The INB also released the results of the remediation carried out in a presentation titled Remediation Project of a Contaminated Site with monazite residues [15][16].

According to the list of contaminated and rehabilitated areas of CETESB of 2017, USIN site is a contaminated area with confirmed risk [17]. Under CETESB's considerations in dispatches contained in proceeding files, it was mandatory to remove the residues in shed A. Then, it would allow leading investigation and remediation of the section under the shed A. So, the clean-up process could consider completed. However, this target would only be achieved with the construction of a final repository for radioactive waste, which is the adequate facility for storage such waste. Although, the final repository construction is the responsibility of CNEN [18]. Nevertheless, until now, there is no repository in the country, and, because of this, the remediation process cannot be completed in USIN site.

In a technical report issued by CETESB on 12/21/11 is highlighted that the plume of contamination in groundwater is localized and does not present dispersion. Furthermore, the chemical, analytical, and radiometric results attest to the stability, and safety of the storage conditions of the waste deposit for the workers, the environment and the population.

The district in which the site is, Campo Grande, is historically industrial. Nonetheless, in the last decade, the neighborhood has changed land use, is currently classified as mixed use [19]. According to CETESB's list of contaminated and rehabilitated areas in the state of São Paulo for the year 2017, the Campo Grande district has 65 contaminated areas [17]. Besides, due to the number of contaminated areas and the presence of several contaminants in groundwater, there are restrictions on the use of groundwater in the Jurubatuba neighborhood, giving to an ordinance published by the Water Department of the state of São Paulo [20], [21].

The INB provided information about USIN site for CETESB until the year 2011, and then direct reporting to IBAMA. According to the proceeding files, CETESB issued its last opinion on 01/31/12, requesting the filing process, since the case passes to IBAMA. In 2013 IBAMA issues a license to INB to operate the Land Decontamination Plan and an Initial Deposit of Nuclear Materials and Radioactive Waste, as previously authorized by CNEN. The license was valid for 05 years, and IBAMA also requested reports related to decontamination performed in the stretch land that had demanded by São Paulo prefecture and had already been authorized by CNEN.

The environmental licensing process for nuclear activities is a federal competence, and for this reason, IBAMA is the responsibility for monitoring the process of managing contaminated areas of USIN. IBAMA has a Federal Technical Register of Potentially Polluting Activities [22], which includes the USIN area. However, since this register refers to the activity developed, it does not present any information about contamination of the area, such as the status within the process of management of contaminated areas.

4. CONCLUSIONS

Nuclear activities are not the only that produces radioactive waste; several activities can generate not just residues but also contaminated sites by radioactive substances. Although USIN site appears in the list of contaminated and rehabilitated areas of CETESB as a contaminated area with confirmed risk, through the developed research, it is possible to recognize that USIN site is an area in a remediation process. However, end-up the remediation process rely on a final repository for radioactive wastes. Consequently, the absence of a final repository for radioactive wastes impairs the adequate environmental management of a contaminated site by radioactive substances, since during and after the remediation processes wastes will be generated, requiring a place to be disposed.

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