URANIUM LEGACY SITES IN PORTUGAL: ENVIRONMENTAL RADIOACTIVITY AND MITIGATION OF RADIOLOGICAL IMPACT

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Abstract

Uranium legacy sites in Portugal include milling tailings, mining waste, old infrastructures, and acid mine drainage with elevated radioactivity concentrations that in some areas could represent a radiation hazard to public health. A national programme was approved for remediation of abandoned and degraded mine areas. Remediation work was carried out in several mine sites already, including Urgeiriça and Cunha Baixa. Radiation exposure and environmental contamination at several sites is described, contrasting the situations before and after site remediation and identifying radiation protection enhancements and socioeconomic gains achieved.

1. INTRODUCTION

Mining of radioactive ores in Portugal was carried out from 1908 to 2001, initially for producing radium and after 1944 for producing uranium. Sixty uranium deposits were exploited and the ore was processed in milling facilities, generally near the mines [1]. After cessation of mining and milling activities the industrial legacy included about 160 Mt of mining waste and 13 Mt of milling waste, most of the last one disposed near the facilities of Urgeiriça mine, in the centre-North of the country. After closure of the mining company Empresa Nacional de Uranio (ENU), concerns of populations and local administrations about the fate and impact of uranium residues on public health, led the Parliament to approve in 2001 a recommendation for assessment of environmental and public health impact in those areas. National Laboratories for Public Health, Geological Survey, and Nuclear Technology Institute (ITN) worked together in assessing the impact of uranium residues (MinUrar Project). Based on the reports produced, recommendations were made for cleanup and environmental remediation of contaminated sites [2, 3]. Although the issue of radioactive and environmental impact of this mining industry has long been debated and the need for environmental remediation perceived, such recommendations contributed to support the approval in 2004 of a remediation plan for abandoned mines initiated thereafter to mitigate radioactive and environmental impacts [4–6]. Herein, environmental radioactivity information gathered over several years through the monitoring carried out by Laboratório de Protecção e Segurança Radiológica (LPSR)/IST (former ITN) is briefly reviewed and the outcome of remediation work in enhancing radiation protection is succinctly summarized.

2. WORK PERFORMED

The evaluation of contaminated and radioactive waste of uranium mines was extensively carried out by LPSR (ex-ITN) from 2000 to 2002, with field work in all old mine and milling sites. Abandoned mines, old facilities, and waste piles were identified, visited, geo referenced, monitored for ambient radiation dose, and samples of waste collected for determination of activity concentration of radionuclides by radioanalytical techniques in the laboratory [7–9].

The risk assessment requested by the Government in 2001 from National Laboratories, enabled an in depth study of selected uranium sites (MinUrar project). Several mine sites were selected to represent the most contaminated areas (those with old milling facilities and milling tailings besides uranium mines), areas with old mines and mining waste but no milling tailings, and reference areas in the same region (i.e., without mines and without mining and milling waste). These sites were investigated for radioactive and stable metal contamination and, furthermore, an epidemiological study was carried out on the resident population applying a careful study design. Results were reported to the Government and Parliament and made available to the public [2, 3].

Decision on environmental remediation and proper decommissioning of abandoned and degraded mine areas was approved by the Government and entrusted to the mining State holding Empresa de desenvolvimento Mineiro (EDM). EDM initiated remediation works in 2005. During 2006–2008 the main site with mining and milling tailings at Urgeiriça, was clean up and waste removed and concentrated in two previously existing dump areas, Barragem Velha and Barragem Nova. Barragem Velha was reshaped and covered with a multilayer cap. A drainage system was built to collect mine water and seepage into an automatic water treatment plant [6]. Remediation of other uranium mine sites is currently in progress or planned. Until 2014 were cleaned and remediated the uranium mine sites of Urgeiriça, Valinhos, Abrutiga, Espinho, Cunha Baixa, Senhora das Fontes, and Bica mines, most of them with milling tailings and acid mine drainage.

As by law LPSR/IST (formerly ITN) is in charge of the environmental radioactivity monitoring and implementation of the EURATOM Treaty article 35, the assessment of radioactivity in regions of remediated sites and not yet remediated sites is regularly carried out and results reported to the Government, to the CEC, and to the public [10]. Sites that had not been assessed in the framework of MinUrar project have gradually been assessed, radioactivity measured, and risks evaluated and reported.

3. RESULTS AND DISCUSSION

A detailed account of environmental radioactivity and radiation issues in all sites could not be reported here. Instead, some selected sites are presented.

The Urgeiriça mine and milling site is located in Viseu county. Mine operation started in 1913, and a chemical treatment plant was added in 1944 for production of uranium concentrates. When facilities were closed in 2001, there was two main milling tailings dump areas, Barragem Nova and Barragem Velha, plus mining waste and low grade ore in other locations inside the mining area facilities. The mine and mill facilities are located in the urban perimeter of Urgeiriça village, but over the years the area was surveyed by ENU company and the access restricted to employees. With the company shut down, waste piles could become areas of public access and contaminated materials could be dispersed. Figure 1 gives an aerial view of the rehabilitated site taken in 2007.

FIG. 1. The milling tailings waste pile Barragem Velha, at Urgeiriça, after placement of the multilayer cap (Aerial view, 2007).

External radiation doses on the top of milling tailings averaged 7.5 μSv/h with a maximum of about 12 μSv/h, and radon exhalation from tailings averaged 3.5 Bq m^{-2} s^{-1} with a maximum of about 7 Bq m^{-2} s^{-1} [10, 11]. External radiation, re-suspension of tailings' radioactive dust, radon emanation, and dispersal of tailings' materials by landslides, surface runoff, and leaching of radionuclides by rainwater were of environmental concern. Furthermore, the work carried out in the framework of MinUrar project identified high radioactivity levels in seepage from tailings piles, mine water, and surface runoff that were of radiological concern. The epidemiological study carried out concluded that population living near the milling tailings had seen their health condition diminished in comparison with the reference groups, and analysis of ^{210}Po in human hair and radiation induced chromosome damage in blood cells indicated higher accumulation of uranium daughters in the body and enhanced radiation exposure than in reference population groups [3].

The cleaning of mining and milling facility areas in Urgeiriça, with disposal of waste from facility areas on top of milling tailings piles followed by the placement of a multilayer cap totalizing 1 m thickness, was carried out from 2006 to 2008 (25 months' work). A drainage system was built and an existing water treatment station improved and automated. The waste dump Barragem Nova, that received contaminated mud and sludge from mine water treatment was also re shaped and covered with shielding layers and soil after 2008.

The outcomes of these works were the reduction of ambient radiation dose on top of Barragem Velha to about 5% of previous dose rate, i.e, down to 0.30 μSv/h (natural background dose rate in the area is 0.2-0.4 μSv/h), the elimination of re suspension of radioactive waste particles into the atmosphere, and the reduction of radon exhalation to near background levels (0.2 Bq m^{-2} s^{-1}).

Radioactivity in the stream Ribeira da Pantanha, receiving process water from milling tailings for decades was, in 2005, at 22.6 ± 0.8 Bq/L of dissolved ^{238}U and 0.247 ± 0.016 Bq/L of dissolved ^{226}Ra, while in 2012 it was of 0.782 ± 0.024 Bq/L and 0.065 ± 0.003 Bq/L for the same radionuclides, respectively [11, 12]. Sediments from the bed of this stream displayed very high concentrations in the past and are gradually diminishing now. Radionuclides in horticulture products grown on the banks of Ribeira da Pantanha using stream water for irrigation were also often higher than in reference areas [11–14]. With the cleanup of the area and contaminated water treatment, radionuclide concentrations are expected to decrease.

The clean-up of the Urgeiriça mine area and confinement of milling tailings and other contaminated waste in Barragem Vellha corresponds to securing a waste volume of 1.6×10^6 m^3. This was performed in 25 months, with a cost of 6 million Euros from public funds. With this remediation work there were gains in radiation protection of the public, environmental clean-up of a stream basin, reduction of the contamination of agriculture and pasture lands in the vicinity of tailings and along the stream, and reduction of contaminated discharges into River Mondego. Furthermore, there were gains also achieved with the decontamination of property (buildings, industrial area, machinery, lands) that can be now released and sold by the mine holding company.

Cunha Baixa mine near the city of Mangualde, is another major uranium mine site located in the village with the same name [15]. Besides the usual impacts of external radiation dose, dust resuspension and radon, in this area contaminated acid mine water from underground mine spread into groundwater and reached the wells of agriculture properties in a field located at altitude lower than the mine. For example, concentrations of dissolved radionuclides in the water of a well in this zone were, in 2007, 4176 ± 280 mBq/L of ^{238}U and 184 ± 40 mBq/L of ^{226}Ra (water pH = 3.88) and, in 2012, concentrations were of 4983 ± 222 mBq/L and 552 ± 33 mBq/L (water pH = 4.12) for the same radionuclides respectively, and showing no improvement. Since mine closure, chemical processing of acid mine water from the underground mine did not help much to reduce radioactivity in the water of those irrigation wells. These contaminated wells were recently sealed and a surface water reservoir was built in terrains of the former mine concession for supplying clean irrigation water to farms in this valley. Views of the water treatment station and a recreational area created by the rehabilitation are shown in Figs. 2 and 3.

The survey of radioactivity in horticulture products from this area was performed several times over the years and radionuclide concentrations were above average natural background radioactivity in comparison areas of the region. Internal radiation dose to members of the public in Cunha Baixa village, consuming horticulture products from their farms, were higher than the EU recommended dose limit for members of the public, 1 mSv/h [12]. This radiation exposure through intake of radionuclides with the diet is expected to decrease with the use of clean water in irrigation. Furthermore, the treatment of Cunha-Baixa contaminated and acid mine water was modified with the introduction of a passive treatment in constructed wetlands and using the chemical treatment as a second resource in mine water treatment.

FIG. 2. Partial view of the old uranium mine site of Cunha Baixa, with the new water treatment station, water reservoir for irrigation, and passive water treatment ponds in first plane (View from the village, 2014).

FIG. 3. Partial view of the small lake and leisure area of the remediated former uranium mine site of Valinhos, near Canas de Senhorim (2014).

Major remediation works were carried out in other mine sites. Many of the former uranium mine sites have no milling tailings and some of them have mining waste that actually contains radioactivity at about background level. Two sites with such features were Valinhos and Espinho mines, in the regions of Urgeiriça and Cunha Baixa, respectively. In both sites did existed flooded open pits and piles of mining waste. Contaminated materials were removed and sites were cleaned, including scrapping of surface ground, and landscape re shaped to adapting for new uses. The Valinhos mine site was transformed in a leisure park, with picnic areas and a fishing pond with facilities for using small boats. Espinho mine site was re-naturalized, keeping the 35 m deep open pit as a water pond used for sports fishing and as water source for firefighting. External radiation and radioactivity levels in environmental materials of these sites are at the level of regional background. For example, in Valinhos area the external radiation dose was determined at < 1µSv/h and radioactivity in the water pond at about background values for natural waters. These environmentally remediated sites, after due clearance by radiation protection authorities, could be used for other societal purposes soon. The challenge is the suitable transfer of site property along with responsibilities on surveillance, maintenance and proper use, to new owners.

Many former uranium sites were not remediated yet. Some of them are of more concern because of radioactive materials and potential for environmental radioactive contamination. For example, some open pit mines such as Mondego Sul, Boco, and Murtorios, contain large volumes of contaminated water and are located near surface water streams, such as the River Mondego, and are of strategic value as sources of drinking water [13, 16]. Adequate action shall be taken in the future, expectedly to ensure suitable water quality and radiation protection to population.

These sites, especially those with large quantities of milling tailings and high radionuclide concentrations, are deemed to be kept isolated from biosphere by multilayer caps placed on tailings and by drainage systems [15]. Such engineered waste management solutions are adequate but will not stand alone and require maintenance in the future and continued radiological monitoring and surveillance. In particular, water, surface air and food chains will continue to require radioactivity monitoring to ensure that radiation dose to members of the public is in compliance with European Union directives and radiation dose limits, as well as international standards [17].

The environmental remediation program for abandoned mine sites, was started in 2005 and the cost so far is of 145 MEuros (of which about 70 MEuros for radioactive mines) for a total of near 30 mine sites remediated or in course of remediation. The remediation program identified 175 degraded and abandoned mine sites and amongst them 61 uranium/radium mine sites [6]. The investment made in remediation of abandoned mine sites (radioactive and non-radioactive) is aimed at protecting public health and environment, and preserving the territory and natural resources for future generations. Remediation works implemented contributed already to a significant abatement of radiation exposure allowing for safer implementation of economic activities, such as agriculture, grazing, and tourism in the surroundings of legacy sites. Environmental remediation contributed also to re build confidence in mining activities and improved life quality in the region.

4. CONCLUSIONS

Research and monitoring of environmental radioactivity at old uranium mine sites and in their surrounding environment allowed identifying existing exposures to ionizing radiation from uranium waste and identifying pathways of radionuclides' transfer in the environment leading to enhanced exposure of population members to ionizing radiation. In the terrestrial environment at these mine sites, besides the direct exposure to external radiation that more easily could be controlled using multilayer caps as a shielding, the main challenge are environmental pathways that may transfer radionuclides from uranium waste to humans. Amongst radionuclides of the uranium family, radium–226 usually is the most water soluble and mobile [18]. Control of water pathways and radium transfer to plants, cattle, and humans may provide control on this major component of radiation exposure of local population members. On the long run, the confinement of residues to isolate them from water cycle and from the food chains may ensure environmental health and radiation safety to population.

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