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The National Radioactivity Monitoring Program for the Regions of Uranium Mines and Uranium Legacy Sites in Portugal

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Abstract

Following closure of the uranium mining company, the areas of former radium and uranium mines were assessed for environmental radioactivity, stable metals and public health impact. Concentrations of radionuclides were found highly enhanced in milling tailings, in mine drainage, and in some surface water streams. An environmental remediation plan was advised and it is ongoing. Furthermore, field surveys are annually carried out and results reported to the Government, the European Union, and made available to the public. This monitoring program enabled assessing the radiological risk, and every year contributes to substantial improvement of radiological safety of population and environment.

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Keywords: uranium sites; milling tailings; radioactivity; environmental remediation; radiation dose

1. Introduction

In Portugal, between 1908 and 2001, 60 deposits of radioactive ore were extracted for the production of radium and uranium. On 2001, with the closure of Empresa Nacional do Urânio (ENU-SA), the assets of this company were transferred to the Empresa de Desenvolvimento Mineiro (EDM), State holding company under the Ministry of Economy for the mine sector. The environmental remediation works of uranium legacy sites were entrusted to EXMIN,

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a society initially set up by EDM for the remediation, under the supervision of the State Follow up Commission (CAC) with representatives of Ministries of Economy, Environment, Science & Technology, and Health set up by the Decree-Law 198A/2001. Later, after approval of the general remediation plan and several site specific remediation plans by the CAC, the EXMIN ceased functions and the remediation activities were directly ascribed to EDM[1,2]. The Nuclear and Technological Institute (ITN), now IST/CTN, was charged to implement the monitoring of environmental radioactivity in regions impacted by extraction of radioactive ores during the phases of exploitation, closure, and requalification, such as established in the Decree Law 165/2002 article 14. To this purpose in 2006 was set up a regular monitoring programme of environmental radioactivity in the areas of old uranium mines and milling sites which started implementation in 2007. This programme carries out extensive environmental surveys for determination of radionuclide concentrations, especially those of the uranium series, and ambient radiation doses. Results of this national monitoring programme are annually reported to the Government, released to the public, and transmitted to the European Union as required by the EURATOM Treaty, articles 35 and 36.

Regular monitoring of these regions is needed to ensure the updated assessment of environmental radioactivity and to generate scientifically sound information for radiological protection of the population and environment, including protection of the quality of natural resources, such as, water, soils, and agriculture and animal products.

2. Past research and monitoring activities in uranium regions

The current and regular national monitoring of uranium regions, implemented since 2007, was not the beginning of the environmental and radioprotection work carried out in this field. The Department of Radiological Protection and Safety (DPSR) had already carried out during the previous 20 years periodic monitoring of environmental radioactivity and published institute reports on the impact of active uranium mining. That work, although thoroughly done, suffered limitations inherent to the spectrometry equipment and knowledge available at that time about key radionuclides, such as ^{210}Po and ^{222}Rn. Notwithstanding, it was an important contribute to keep institutional control on mining and milling waste, to improve waste management, and to figure out the radioactive impact on the environment [3]

With the closure of ENU-SA and cessation of all mining and milling activities in 2001, a new period would start. During decades radioactive waste was controlled and mining concession areas were of restricted access and surveyed by the Junta de Energia Nuclear (JEN) and later by the Empresa Nacional de Uranio (ENU). With closure, the abandon could lead to a major disruption in waste management and in surveillance of properties and facilities and, thus, to a major change in radiation protection measures. There was a need to follow this change and prevent loss of institutional control on radioactive waste. The need for assessment of sites and for an eventual remediation plan in order to meet radiation protection goals was put forward [4](Fig. 1).

The assessment of radioactivity levels in the uranium mining areas was resumed by the time of discontinuing the uranium production activities through a research project aimed at identifying the uranium mining and milling waste in those regions and the potential radiological exposure risks. This project was supported by an IAEA Technical Cooperation project (POR/9/014, 2001-2003) and allowed for revisiting and referencing all the uranium legacy sites, measuring ambient radiation doses, and analyzing the composition of wastes left on site by past mining and milling activities. A clear picture about characteristics of sites, types of contamination, and need for remediation was obtained and consolidated. This project concluded with an International Workshop jointly organized by ITN/DPSR and the IAEA, in Sacavém, 11-13 February 2004, on the "Environmental Contamination from Uranium Production Facilities and Remediation Measures" (Proceedings published by the IAEA in 2005) [3].

This assessment was followed by the research project entitled "Study of Effects of Uranium Mining Residues on the Public Health" (MinUrar Project), funded by the Ministry of Health as a result of a recommendation approved by the Parliament (Recommendation to the Government Nº34/2001). The MinUrar project (2003-2006) was jointly implemented by the Nuclear and Technological Institute (ITN/DPSR), the National Health Institute and the Geological Survey and carried out an in-depth study of the uranium residues impact and its effects on the environment and public health.

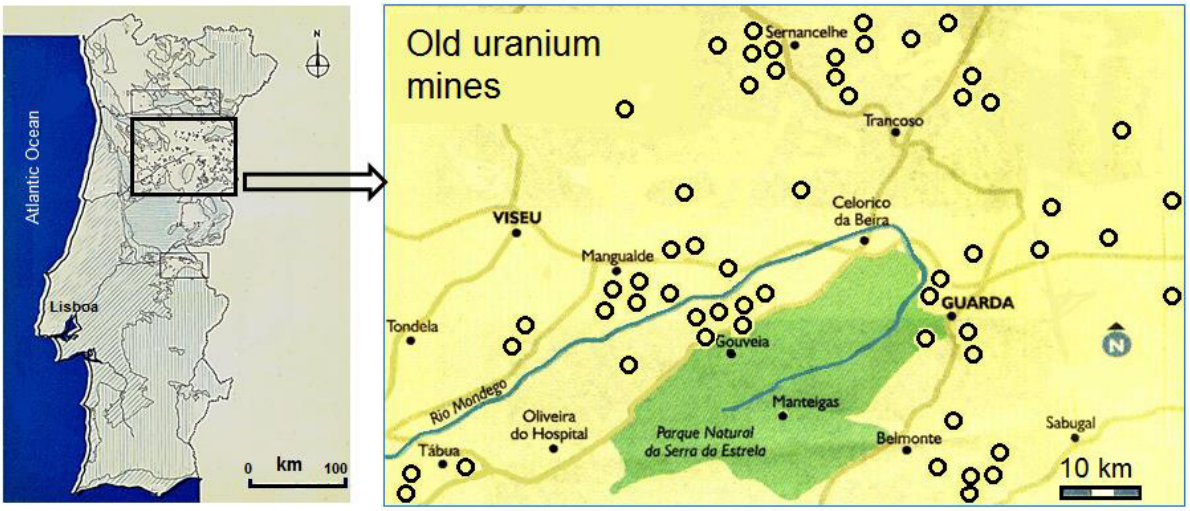

Fig. 1. Uranium mining areas in Portugal, periodically surveyed for environmental radioactivity. Open circles indicate old uranium mines.

3. Results of the MinUrar project

The MinUrar project was implemented through three main axes: environmental radioactivity, heavy metals, and impact on public health[5]. Investigation on environmental radioactivity levels showed that mining and milling tailings at Canas de Senhorim/Urgeiriça contained high concentrations of uranium family radionuclides (Table 1). For instance, the concentrations of ^{226}Ra in mill tailings reached 25 kBq kg^{-1} whereas in sludge from ponds used to treat acid mine water, ^{238}U concentrations reached about 42 kBq kg^{-1}. The areas receiving surface runoff and drainage from milling tailings displayed enhanced concentrations of those radionuclides also, and concentrations in the most contaminated soils were up to 200 times higher than concentrations in agriculture soils of the region. The same trend was observed with the ambient radiation doses that reached values of 41 mSv a^{-1} on uranium tailings at Urgeiriça and decreased to 2.5 mSv a^{-1} on agriculture fields. Clearly, the uranium milling tailings were a source of radiation originating external radiation dose rates largely exceeding the regional radiation background and the additional dose limit of 1 mSv/year to members of the public (Directive 96/29/Euratom). Furthermore, due to the very high radium (^{226}Ra) concentrations in the waste, tailings were a powerful source of radon (^{222}Rn) and radioactive dust. Atmospheric processes and surface runoff following heavy rains, would gradually contribute further to disperse the radionuclides from the milling tailings in the surrounding environment. Radionuclide concentrations determined in agriculture products, indicated higher values in these areas. Radioactivity in drinking water was also elevated in some investigated counties and especially in water from private supplies (wells) in some areas near Canas de Senhorim, and Cunha Baixa. Suspended dust particles in surface air at Canas de Senhorim carried also higher radionuclide concentrations than air dust in other counties. Therefore, if the area of milling tailings was left without control and with no remediation, this could originate an exposure to radionuclides and radiation doses exceeding the maximum permissible limits for the safety of the population and compromise the quality of environmental resources of this region[5].

The dispersion of heavy metals and other inorganic contaminants in the area of Canas de Senhorim was detected mainly in the hydrographical basin of streams receiving drainage from the mining facilities and milling tailings and, thus, it was concluded that uranium ore processing had left its fingerprint in the environment through enhanced concentrations of co-occurring stable metals, such as lead and zinc[5].

Table 1. Radionuclide concentrations (Bq kg^{-1} ±1SD) in uranium waste at Urgeiriça mine and mill site after closure of ENU-SA Company.

	^{238}U	^{235}U	^{234}U	^{230}Th	^{226}Ra	^{210}Po	^{232}Th
Milling tailings, Barragem Velha	2530±94	118±12	2876±105	10337±598	24717±2039	20354±681	412±32
Sludge ponds, Barragem Nova	41598±1228	1959±67	40182±1187	13390±613	1690±150	1176±43	386±22
Spoil heap, Santa Barbara	6108±173	276±14	6175±175	8052±282	3608±133	3501±112	112±7
Waste heap by the ore mill	38316±1154	1717±67	38247±1152	30115±113	15569±707	30824±1147	426±21
Soil, Aldeia de Aguieira	211±7	10±1	201±7	128±6	140±8	227±8	117±6

A soil from the region is shown for comparison.

Regarding the effects from exposure to waste from past uranium mining activities on human health, the results indicated the existence of a negative impact on populations' health living near the uranium mining and milling areas, although a direct cause-effect relationship could not be established[5].

By the end of MinUrar project results supported recommendations made in the final report which included the implementation of an environmental remediation plan, the periodic environmental radioactivity monitoring in the areas impacted by the uranium residues, as well as the follow up of remediation works, and radiological surveillance of the environment in the post-remediation[5].

4. Outline of a remediation programme

The need for environmental remediation of former uranium sites was foreseen and proposed for enhanced radiation protection of the public[3,6]. Following the sound recommendations made by MinUrar project in 2005, the implementation of a remediation plan for the abandoned mines, including radioactive and non-radioactive mines was approved and entrusted by the Ministry of Economy to EDM. This plan achieved the rearrangement and confinement of radioactive waste in Urgeiriça in the period 2005-2007. Other mine sites, such as Valinhos and Espinho, were cleaned up and requalified and, remediation of Cunha Baixa site is ongoing and others, such as Quinta do Bispo mine site may follow[7,8].

5. Regular monitoring of uranium regions

This radioactivity monitoring programme of the former uranium mining and milling areas currently takes into account the main five mining areas in the center of the country, where most of the mining and milling waste is deposited and to where the waste from small mines is planned to be relocated and stored. This monitoring plan takes into account the potential transport of radionuclides from mining and milling waste into ground water and rivers crossing the region and possible transfer pathways of radionuclides along the food chain to man.

Samples collected yearly in the region include soils, vegetables, well water used for irrigation, drinking water from public supplies, atmospheric dust, and radon in surface air. The sampling areas encompass the main mines and milling tailings areas and rivers receiving discharges and surface runoff. Occasional samples of milk, poultry and cooked meals are purchased in the area for radio analytical measurements. The focus of environmental monitoring has been maintained in the main mine areas, i.e., Urgeiriça, Quinta do Bispo, Cunha-Baixa, Bica, Castelejo, Mortórios, with occasional visits to and sampling in other areas. A thorough monitoring has been made of the main rivers receiving mine water discharges, especially the River Mondego. The main radionuclides of the uranium series, namely the alpha emitters uranium isotopes, ^{230}Th, ^{226}Ra, ^{210}Po and ^{210}Pb (beta emitter) are systematically determined in the samples through radiochemical analyses and alpha spectrometry measurements. A periodic analytical quality control programme is implemented through participation in international interlaboratory comparison exercises organized by the IAEA and EU, and routine analyses of certified reference materials. The results consistently are of good quality and show accurate and precise determination of radionuclides[2,4,9].

The results of the monitoring are included in the Annual Report on the environmental radioactivity surveillance, delivered to the authorities and released to the public by the ITN (now IST/CTN). These reports show extensive tables with radionuclide concentrations in all type of samples totalizing around 400 radionuclide measurements per year[9].

Results from several years of monitoring enabled identifying hot spots of radioactive waste, areas with wells showing too high radionuclide concentrations for use in irrigation and consumption, and kitchen gardens with vegetable products not suitable for human consumption. Appropriate recommendations have been made to the population concerned. In general, the population is not exposed to high values of external radiation doses and, with few exceptions, is supplied nowadays with water suitable for human consumption.

6. The need to plan the post remediation phase

A few sites of uranium mining legacy were remediated so far, including the Urgeiriça site where the uranium mill was operated and most of the milling waste deposited. The milling waste pile Barragem Velha was confined with a multi-layer cover and the remediated site was fenced in 2007. This area is now of restricted access and requires maintenance and continuous radiation monitoring that is carried out by EDM. Other areas were cleaned up at Urgeiriça including mill facilities, warehouses, terrains and roads. A public park was build up on the terrains of the former ENU. The mine drainage and seepage from the waste pile are recovered and treated in an automated water treatment plant at Barragem Nova[8]. Mine sites as Valinhos and Espinho were cleaned up and re-engineered to make them suitable for new uses and are intended to be released from institutional control. Other areas, such as Cunha Baixa, are under remediation and Quinta do Bispo is planned for remediation soon. The issue still open to finding a suitable solution is the future use and control of these areas. There is a need for custody of remediated sites and permanent radiological surveillance, at least until confirmation that remediation works were sufficient to reduce radon concentration in the air, offer efficient shield against gamma radiation, and that are stable solutions providing long term confinement for solid and liquid effluents. The regular national monitoring programme implemented by the IST/LPSR ensures every year, as required by national legislation and by the EURATOM Treaty, that mine sites, soils, rivers and atmosphere are controlled for radioactivity, and that water, agriculture and animal products (meat, milk, cheese) from the region are suitable for human consumption and, thus, for trade and export. Due to the long lived radionuclides present in the uranium waste, waste confinement and management need to be effective in keeping them apart from the biosphere for long time (centuries). In this regard, international recommendations for enhanced radiation safety and regulatory control have been agreed and provide suitable guidance applicable to uranium legacy[10].

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