

Risk Assessment and Management of Uranium Mill Tailings Legacies – 17508

M.L. Dinis^{1,2}, A. Fiúza^{1,2}

¹CERENA-Polo FEUP - Centre for Natural Resources and the Environment

²FEUP - Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465, Porto, Portugal

ABSTRACT

In Portugal, the exploitation of uranium ore was active from 1913 to 2000. The exploitation activities took place at 61 sites. The great majority of these mining sites are located in central-east Portugal, including the Urgeiriça mine that was the most important uranium deposit exploited in the country. The mining activities at this site generated 1.6 million m³ of radioactive wastes disposed in an open air area originating the largest radioactive tailings pile of the country.

The environmental remediation of the Urgeiriça site was one of the main priorities of the national plan for the rehabilitation of former uranium mining sites. Site remediation, that included the mine industrial area with several support infrastructures and a processing plant, was executed between 2006 and 2008 (1st phase) and between 2008 and 2014 (2nd phase). Nevertheless, the environmental rehabilitation of the Portuguese uranium legacies sites is not yet concluded. One of the last largest mining complexes, the Quinta do Bispo open pit, is scheduled to be rehabilitated only in 2020.

This work presents a study to evaluate the environmental impacts from the uranium mill tailings legacies before and after the remediation works, focusing on two different approaches: a deterministic methodology for dose assessment and a probabilistic methodology with the Monte Carlo method to estimate the incremental lifetime risk. Probabilistic distributions of the input parameters were used in the risk calculation approach.

The methodology for dose and risk assessments may be applied to other sites that are still under intervention and the results may be used to prioritize the remediation stages.

INTRODUCTION

Over the last decades the legacy of former uranium exploitations and uranium processing plants has been documented in many parts of the world. Uranium mill tailings are an important component of these legacies due to their composition, quantities and continuous transformation. The tailings are the finely ground silt-like material that is left after uranium is extracted from ore that still contains in most cases around 75 % of the original activity, as only the first isotopes of the uranium 238 and 235 decay series are removed by leaching, while all the others isotopes remain un-leached. In addition, the tailings may suffer the oxidation of the sulfides and they also contain almost all the heavy metals associated with the ore, as well as a variety of other anionic pollutants.

In Portugal, the uranium mining ore was active from 1913 to 2000 mainly in the

central region of the country. Several ore bodies were exploited in Viseu and Guarda districts after the first mineral deposit was discovered in 1907. Until the forties of last century the main interest was the production of radium salts. After the World War II uranium became the element of interest being exploited mainly at Urgeiriça (Viseu) and Cunha Baixa (Viseu) besides others 16 mines (Fig. 1). The Urgeiriça mine complex was one of the largest deposits in Europe.



Fig. 1: Location of the two main Portuguese districts of former radioactive ores exploration (Viseu and Guarda).

The exploration activities took place at 61 sites for uranium and radium ores, mostly in small open pits, although the larger ones were underground mines, or a combination of both. The mining methods were determined by the characteristics of the exploited ore bodies: underground, open pit, combined underground/open pit, sometimes using heap or in situ acid leaching (ISL). The most relevant uranium mine complexes occurred at Urgeiriça, Cunha Baixa, Quinta do Bispo, Bica, Castelejo and Pinhal do Souto. In most cases the uranium was extracted and transported to Urgeiriça or other major mining facility for milling and extraction of the uranium. In 1991, the residual in-situ leaching of the Urgeiriça Mine was extinguished but the processing plant was still used for the ore processing of ores from other mines until 2000. Other leaching facilities also operated at Bica (Sabugal, Guarda), Quinta do Bispo and Cunha Baixa (Mangualde, Guarda), Senhora das Fontes (Pinhel, Guarda) and Castelejo (Gouveia, Guarda)[1] but the Urgeiriça “Old Dam” accumulated the tailings from almost all the high grade ores. In addition, during the last two decades of the mining activity, several tonnes of

sludge containing the radium precipitates produced by addition of barium chloride and radionuclides resulting from the neutralization of acid mine waters were placed at the “Old Dam”.

Therefore, the intense exploration of radioactive ores during almost one century generated 13 million tons of wastes that were left in the mining sites [2]. These wastes comprised mostly mine tailings, low-grade ores and sludge enriched in metals and radionuclides, contributing to the contamination of soil, water and air not only in these sites but also in their vicinity, causing an additional radiological exposure to local populations and ecosystems. However, not all the sites presented the same risk. The situation was more serious in specific mining locations like the Urgeiriça site. Mining activities at Quinta do Bispo, Cunha Baixa and Bica (Sabugal) also generated tailings, because the marginal ore was locally processed by heap leaching.

In particular, the mining and milling activity at Urgeiriça complex led to the accumulation of large amounts of solid wastes mainly at three locations: i) a tailings disposal with an estimated volume of 1.39 million m³ of sludge produced in the milling facility occupying an area of 13.3 ha (“Old Dam”); ii) a waste rock pile containing some low-grade ore (91 000 m³ on an area of 1.5 ha) near the main shaft and a few tons of a high-grade uranium ore (not milled) that existed near the plant.

The Urgeiriça tailings dam, “Old Dam”, was the main concern of the national plan for the rehabilitation of former uranium mining sites due to both its volume and radioactive content. The 1 460 000 m³ of materials deposited are very heterogeneous: size grain, chemical composition and radiometry. Also, this site is surrounded by small farms and country houses, with most of the population living in the village of Canas de Senhorim, a settlement with about 5000 inhabitants. Others sites that were also considered priorities for the environmental remediation were the mines of Cunha Baixa, Quinta do Bispo and Bica [3].

The Cunha Baixa uranium mine is surrounded by pine trees forest and agricultural fields. It is located in the municipality of Mangualde (Viseu) with an area of 16 km² and 884 inhabitants.

This mine included facilities for both underground and open pit extraction. At the end-of-life the stored marginal ore was processed by heap leaching performed inside the open pit. The leachates percolated through the pit bottom and were collected and pumped from the first level of the underground mine, pre-concentrated by ion-exchange and sent to Urgeiriça plant as charged resins for further treatment. Approximately 0.5 Mtons of ore were extracted from this mine with a production of 901 tons of U₃O₈ [4] and 76 tons of uranium was produced by heap leaching with the treatment of 400 000 tons of low-grade ore from other mines in the region. This mine site generates acidic waters (pH = 3) with high concentration of ²³⁸U and ²²⁶Ra as well as other metals. In the past, the barren solution from ion exchange was treated for pH neutralization and radium precipitation, but the resulting sludge was not confined.

The Quinta do Bispo uranium mine was located at approximately 2 km from the Cunha Baixa mine. This mine was one of the largest open pits in Portugal with a depth of about 80 metres and a surface area of 158 000 m². About 460 000 tons of

ore were extracted from this mine. Marginal ore was leached in pads, pre-concentrated by ion-exchange and the charged resins were transported to Urgeiriça for further treatment [3]. The use of waste rock from the mine was allowed for the construction of house foundations until 1980.

The Bica underground mine operated from 1912 to 1944 and from 1951 to 1999. The ore was processed in a very small treatment plant. When the exploitable reserves were exhausted the remaining ore was submitted to in-situ leaching. As a consequence, there existed tailings and sludge as a matter of concern for radiological exposure.

Since 1996 the Portuguese government had to deal with the decommissioning of the mines, mills and other facilities and the rehabilitation of the mining sites [5]. Nevertheless, only in 2001 (Decree Law 198/A), the Portuguese government assumed that the rehabilitation of these areas left by old mining activities was a question of public interest [1], [6]. In 2002, the uranium mines sites were classified as priority areas for intervention [1], [6], [7].

In particular, for the remediation of the Urgeiriça site, the reclamation program was announced in 2005 and the rehabilitation of the “Old Dam” was considered a key element of the overall environmental remediation program [3], [5]. These tailings were a source of external radiation and, furthermore, a source of radon originating higher radiation levels comparing to the background values. Radioactivity measurements done at the “Old Dam” showed high concentrations for radionuclides from the ^{238}U decay series. Due to precipitation, water, wind and soil erosion radionuclides could be continuously transported and redistributed in this region as well as in its vicinity. Therefore, people living in nearby areas may have been exposed to additional levels of radiation.

The environmental remediation of the Urgeiriça site included the confinement of the radioactive wastes of the “Old Dam” in addition to the 140 000 m³ of materials from another waste dump and from another contaminated area in the neighbourhood, that were deposited in the “Old Dam” during the remediation stage [3].

This work presents a study to evaluate the environmental impacts from the uranium mill tailings legacies before and after the remediation works focusing on two different approaches. One approach is based on a deterministic methodology for dose assessment and the second approach is based on a probabilistic methodology applying the Monte Carlo method to estimate the resulting risk. Probabilistic distributions of the input parameters were used in the risk calculation approach generated with Matlab® code.

METHODOLOGY

Survey Area – Assessment of External Radiation and Radon Concentration

Previous assessment of the “Old Dam” tailings pile comprised radon measurements, surface radiometry and external radiation dosimetry in order to assess the dispersion of radioactive materials and establish a pattern that could be used for comparative purposes after the remediation. The remediation project included, among others targets, radiological emanations and radon fluxes. Reducing radon

concentration to acceptable or legal values was one of the main issues for the environmental remediation of the “Old Dam” tailings pile [8].

Radon concentrations were measured at 1 m above the ground with passive detectors LR115. About 52 samplings points were chosen according to background radiometer variation, between 140 and 10 000 cps (measurements carried out with a portable gamma ray scintillometer Saphymo SPP2) [1]. Surface radiometry and external radiation doses registered 15 000 cps and 7.5 $\mu\text{Gy/h}$, respectively, in some zones of the pile [1]. For radon concentrations the values ranged from 195 to 1205 Bq/m^3 with an average value of 557 Bq/m^3 and a standard deviation of 309 Bq/m^3 [8].

The materials of the tailings pile were very heterogeneous not only in what concerns to radionuclides concentration [9], [10], [11], [12] but also for materials composition, particle size and moisture. Gamma radiation measurements carried out along drill holes performed in the tailings and at the surface, showed a significant compositional and particle size heterogeneity of the materials disposed [8].

The remediation program for this site included the placement of a multi-layer cover for the “Old Dam” and the remediation of the mine industrial area, where the remains of the former milling and processing plant were located (the tailings, the unprocessed ore naturally leached, and the waste rock deposit). The wastes arising from the industrial area were transferred to the tailings pile “Old Dam”.

The structure of the tailings disposal had to be geo-technically stabilized (field works started in 2005), confined in-situ by a peripheral concrete support structure, equipped with surface and deep drainage systems, and sealed off with a multi-layer cover consisting of geological and synthetic materials along all the surface of the tailings deposit.

The multilayer cover is composed of a compacted layer of geological materials at the bottom followed by a clay layer of 0.60 m, a 2 mm high-density polyethylene (HDPE) liner in conjunction with a geo-textile membrane with 10 mm, a drainage layer of 0.30 m of gravel, then a 0.50 m of sand layer and finally a layer of topsoil with 0.50 m [13]. After remediation, the tailings were fenced off to prohibit public access. The rehabilitation of the “Old Dam” took place between 2006 and 2008 [8], [13], [14].

Radiometry and external radiation doses measured after complete remediation decreased to 300 cps and 0.35 $\mu\text{G/h}$ [15]. This value is the same order of magnitude of the regional background 0.4-0.5 $\mu\text{G/h}$ for this area [15].

Radon values in air measured on late 2008, after the placement of the cover, averaged between 55 and 65 Bq/m^3 [15]. Radon concentrations recently measured at 20 cm depth of the cover ranged from 33 and 65 Bq/m^3 [16] and therefore the cover is efficiently retaining the radon.

Effective dose assessment

The exposure scenario adopted in this study considers both internal and external exposure for estimating the dose and the associated risk incurred from the exposure at the Urgeiriça uranium mining site. Collected data from previous assessments were used as an input for dose calculations.

The inhalation dose was calculated using a deterministic methodology where all input parameters were defined as a single fixed value [8], according to Eq. 1 [17]:

$$H_{\text{int}} = C_{\text{Rn}} \times F \times E \times (\text{DCF}) \quad (\text{Eq. 1})$$

where H_{int} is the annual effective dose due to radon inhalation (mSv/y), C_{Rn} (Bq/m^3) is the average radon concentration measured with passive detectors LR115, at 1 m from the ground, F is the equilibrium factor between radon and its progeny, E (hours/year) is the occupancy factor and DCF (mSv/y per Bq/m^3) is the dose coefficient factor (effective dose received by adults per unit of ^{222}Rn activity per unit of air volume).

For radon concentration measured at 1 m from the ground an average value of $557 \text{ Bq}/\text{m}^3$ was used [8].

Radon progeny has a very important contribute to internal exposure (alfa – radiation). Many measurements of radon progeny have been reported which suggest that a rounded value for the equilibrium factor of 0.6 may be appropriate for the outdoor environment. Nevertheless, the UNSCEAR Report (2000) also noted that there is a wide range of values from individual measurements which is understandable given the many environmental factors, including exhalation rates and atmospheric stability that influence the activity ratios. A typical worldwide F factor of 0.4 is recommended by UNSCEAR [17]. For the exposure frequency parameter (E) it was considered a critical receptor represented by farmers from the small farms in the vicinity of the site (farmer residing onsite), for whom the time not spent in their houses is likely spent outdoors. The exposure scenario supposes that the receptors spend 12 h/day during all year inside the house and 8 h/day, 7 days per week, 50 weeks per year, outdoor working in open-air farm activities (accounting for the receptor being away from the site for 2 weeks per year) [18], [19].

The dose coefficient factor (DCF) or radon equilibrium equivalent concentration (EEC) represents the conversion of potential alpha energy exposure ($\text{Bq h}/\text{m}^3$) to effective dose equivalent (nSv). This conversion factor is the concentration of radon that, in equilibrium with each one of the daughters, would have the same potential alpha-energy per unit volume as the actual mixture [8]. A value of $9 \text{ nSv}/\text{h}$ per Bq/m^3 ($9 \times 10^{-6} \text{ mSv}/\text{h}$ per Bq/m^3) was used [20].

In these conditions, the internal component of dose assessment is not absolute but estimated with a considerable degree of uncertainty due to the assumptions and the used single value for each one of the input parameters.

The external dose assessment (H_{Ext} , mSv/y) is obtained directly from the gamma dose rate (D_γ , $\mu\text{G}/\text{h}$) measured in the previous assessment and monitoring programs at 1 m from the ground [12], [21]. A maximum value of $7.5 \mu\text{G}/\text{h}$ was adopted from these studies.

The dose given in $\mu\text{G}/\text{h}$ is converted in mSv/y through the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv Gy^{-1}) [22] and corrected for the considered exposure scenario (8h/d, 365 d/y).

$$H_{\text{Ext}} = D_{\gamma} \times 0.7 \times 10^{-3} \times E \quad (\text{Eq. 2})$$

The total dose (total effective dose) over a year, for the considered scenario, is the dose from inhaled radon (mSv/y) added to the dose from external radiation (mSv/y).

Risk Assessment

Traditional approaches, such as the deterministic ones, often report the risks as “central tendency” (mean or median), “high end” (e.g., 90th percentile or above) or “maximum anticipated exposure” but a probabilistic risk assessment approach can be used to describe more completely the uncertainty surrounding such estimates and identify the key contributors to the variability or uncertainty in predicted exposures or risk estimates. This information can then be used to compare the risks related to different management options, or to invest in research with the greatest impact on risk estimate uncertainty [23].

The U. S. Environmental Protection Agency evaluates the risk due to radiation exposure as the carcinogenic slope factor, representing the lifetime excess total cancer risk per unit intake or exposure (excess lifetime cancer risk - ELCR). The cancer slope factor represents the slope of the dose-response curve, at very low concentrations, thus quantifying the cancer inducing potential; the unit is the inverse of a dose. The product of the cancer slope factor by the dose received estimates the ELCR for a member of the critical group due to a specific exposure. And in this way, the ELCR represents the probability of cancer inducing by this particular exposure in excess relatively to the background risk, also known as the incremental lifetime cancer risk (ILCR). Therefore, the radiological risk assessment is an estimate of the probability of a fatal cancer over the lifetime of an exposed individual while a radiological dose assessment calculates the amount of radiation energy that might be absorbed by a potentially exposed individual as a result of a specific exposure.

The acceptable risk is generally defined as 10^{-6} for the general public and 10^{-5} for workers. This means that an additional one case of cancer is accepted for populations of 1 million or 100 000, respectively. A risk level of 1 in a million or 1 in one hundred thousand also implies a likelihood that up to one person out of one million (or 100 000) equally exposed people would develop cancer if exposed continuously (24 hours per day) to a specific radiation dose over 70 years (an assumed average lifetime). This value is in excess to the normal background number of cancer originated by multiple and indeterminate causes, respectively 200 000 or 20 000 [24].

In this study, a probabilistic approach was used to assess the incremental lifetime cancer risk incurred from radon inhalation (R_{Rn}) and from external exposure to gamma radiation (R_{Ext}) in the considered scenario.

First, the annual risk resulting from radon inhalation was obtained by combining the average indoor radon measurements (C_{Rn} , Bq/m³) with the inhalation rate at the exposure location (BR, m³/d), the exposure frequency (E, d/year) and the radon cancer slope factor (CSF, Risk/Bq) as given by Eq. 2. A single input fixed value of 4.86×10^{-10} (Risk/Bq) was adopted for the cancer slope factor [24]. Probabilistic

distributions were used for the other input parameters [8].

$$R_{Rn} = C_{Rn} \times F \times BR \times E \times (CSF) \quad (\text{Eq. 3})$$

The long-term variation of outdoor radon equilibrium factor adjust to a log-normal distribution [25]. A value of 0.51 ± 0.12 for the mean and the standard deviation, respectively, were used in this simulation for the input parameters of the log-normal distribution [26].

The values measured for radon concentration were considered to adjust also to a log-normal distribution with a mean and standard deviation of 557 and 309 Bq/m³, respectively, for the input distribution parameters [8].

For the daily inhalation rate, it was adopted a log-normal distribution, normalized to the average body weight, with a mean and a standard deviation of 16.45 and 4.69 m³/d, respectively [27].

The outdoor exposure frequency was assumed to follow a triangular distribution with 180 d/year (minimum), 365 d/year (maximum), and 345 d/year (most probable) as the input parameters of the distribution. The likeliest value was established using the rationale that two weeks spent away from the site was a plausible likeliest value. The minimum value assumes that a person spends 50 % of his or her time at the site and the rest away from it. The maximum value for EF assumes that the person spends all of his or her time at the site [28], [29], [30]. The annual cancer risk induced by the external exposure to gamma radiation (R_{Ext}) was estimated, based on the calculated gamma dose for the external exposure (H_{Ext}), according to Eq. 4:

$$R_{Ext} = H_{Ext} \times (EF) \times (RF) \times (SF) \quad (\text{Eq. 4})$$

where R_{Ext} is the annual cancer risk for external exposure due to gamma dose (the health risk from a given radiation dose), H_{Ext} is the calculated dose (input as mSv/365 d) [12], EF is the outdoor exposure frequency (probabilistic distribution as d/year), RF is the fatal cancer risk per Sievert (mSv⁻¹), SF is the gamma shielding factor outdoor (fraction of outdoor gamma that is shielded; 1.0 = no shielding). For this input, SF, a triangular probabilistic distribution was adjusted with 0 (minimum), 1 (maximum) and 0.7 (most probable) [31], [32], [33], [34]. For fatal cancer risk per Sievert or risk factor, (RF), ICRP 60 considers 0.04 for workers and 0.05 for the public accounting for stochastic effects [35].

The obtained value for the annual cancer risk is then multiplied by the exposure lifetime of 70 years to give an estimate of the probability of a fatal cancer over the lifetime of an exposed individual, or a 30-year exposure period of an individual farmer residing onsite, to calculate the incremental lifetime risk (ECLR) which represents the probability of cancer inducing by this particular exposure in excess relatively to the background risk.

The Monte Carlo method was then applied to generate the input distributions, used in the risk calculation approach as well as to generate the distribution for the resulting risk given as an output (developed in Matlab© code).

RESULTS AND DISCUSSION

The inhalation dose estimated from the deterministic approach resulted in 5.61 mSv/y while for external gamma radiation the resulting dose is 15.33 mSv/y. The total annual effective dose is 20.94 mSv/y. An estimated dose can be assumed as low priority if a conservative estimate is low enough: below 10 % of the recommended limit of 1 mSv/y for an individual of the public [35], [36]. In this specific case, a high priority should be assigned.

The obtained results indicate that the external exposure to gamma radiation is the major exposure pathway. However, we must be aware that a maximum value was used as an input of the external gamma dose rate and this should be taking into account when interpreting these results. Moreover, the inhalation dose is underestimated as dust inhalation was not included due to the lack of accurate data [8]. Other studies carried out at this site [37] describe an exposure scenario composed by external radiation, inhalation of dust, radon or radon products, or accidental ingestion of soil with an effective dose estimated in 39 mSv/y. In the same study it is reported that radon and progeny inhalation is the major exposure pathway, accounting for almost 87 % of the total dose and the external gamma radiation is the minor exposure pathway, accounting for approximately 13 %. Dust inhalation or accidental ingestion of soil have a minor importance and can be neglected [37].

The probabilistic distributions of the input parameters for risk estimates (cancer risk) due to radon inhalation and due to external gamma radiation exposure are presented in Fig. 2.

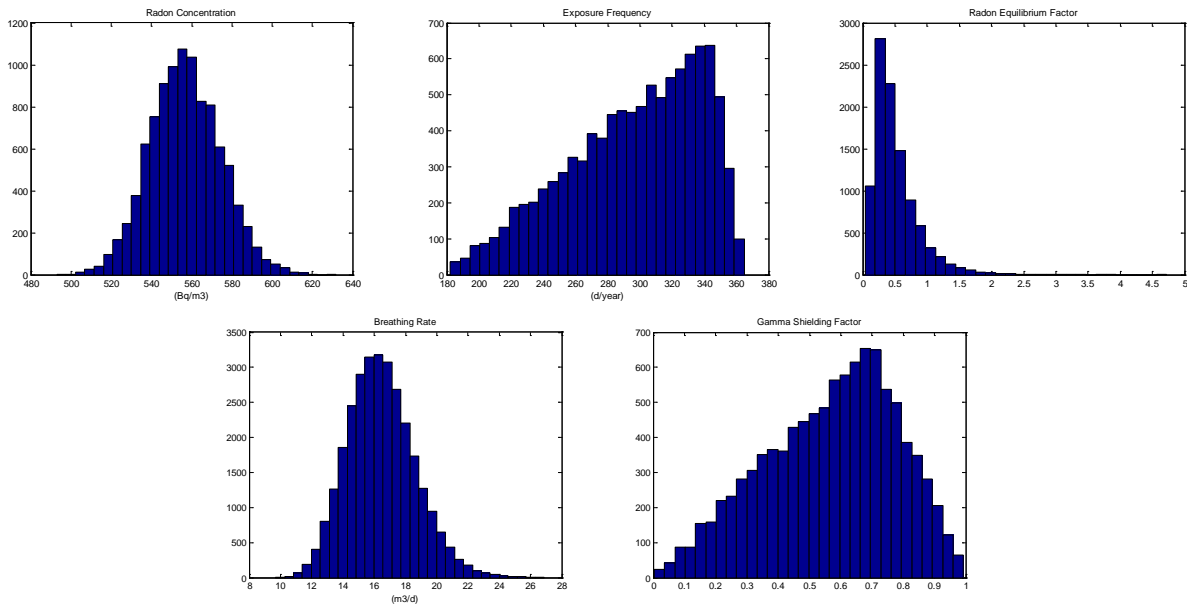


Fig. 2: Probabilistic distributions of the input parameters for risk calculations.

The probabilistic distributions of the resulting annual cancer risk and the cumulative probability functions are presented in Fig. 3 and in Fig. 4 for radon inhalation and external exposure pathways.

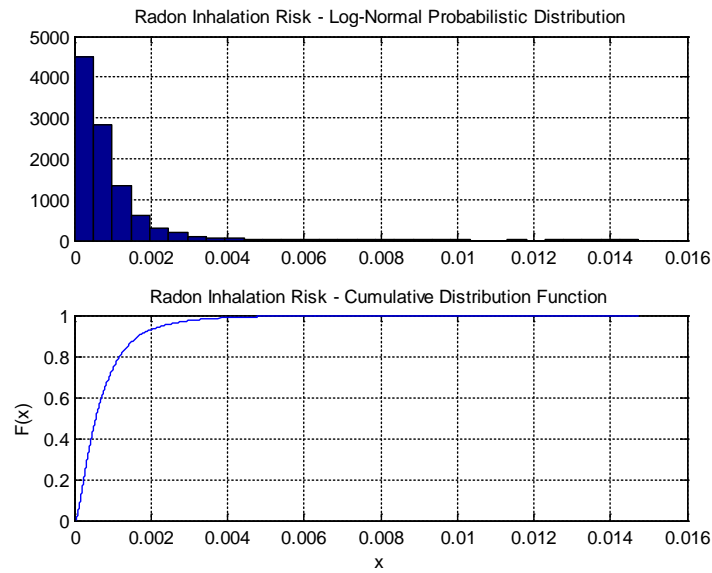


Fig. 3: Radon inhalation risk – log-normal probabilistic distribution and cumulative probability function.

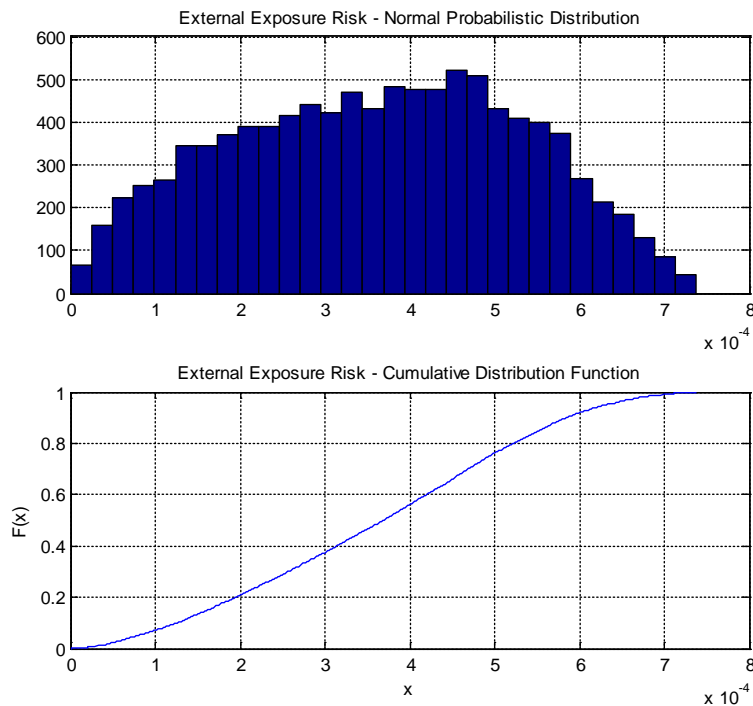


Fig. 4: External gamma radiation exposure risk – normal probabilistic distribution and cumulative probability functions.

The probability plot shows that 95 % of this area is characterized by an annual inhalation cancer risk below $\cong 0.003$ (Fig. 3) and an annual cancer risk for external radiation below $\cong 6.5 \times 10^{-4}$ (Fig. 4).

A summary of the risk assessment for the considered exposure scenario is presented in Table 1 and Table 2, for radon inhalation and for external exposure, respectively. The presented results include: i) the annual cancer risk; ii) the cancer risk incurred by a 30-years exposure period and iii) the incremental lifetime cancer risk (70 years) [29]. Data on mean, median and 95th percentile are presented for radon inhalation and gamma radiation exposure pathways.

TABLE I: Cancer risk from radon inhalation (farmer scenario)

| Radon inhalation exposure | Mean | Median | 95th perc. |
|----------------------------------|-----------------------|-----------------------|------------------------------|
| Annual cancer risk | 8.21×10^{-4} | 5.67×10^{-4} | 2.40×10^{-3} |
| 30-year exposure cancer risk | 2.46×10^{-2} | 1.70×10^{-2} | 7.20×10^{-2} |
| Incremental lifetime cancer risk | 5.75×10^{-2} | 3.97×10^{-2} | 1.68×10^{-1} |

TABLE II: Cancer risk from external gamma radiation exposure (farmer scenario)

| External gamma radiation exposure | Mean | Median | 95th perc. |
|------------------------------------------|-----------------------|-----------------------|------------------------------|
| Annual cancer risk | 2.96×10^{-4} | 3.03×10^{-4} | 5.16×10^{-4} |
| 30-year exposure cancer risk | 8.89×10^{-3} | 9.08×10^{-3} | 1.55×10^{-2} |
| Incremental lifetime cancer risk | 2.07×10^{-2} | 2.12×10^{-2} | 3.61×10^{-2} |

The resulting annual cancer risk for radon inhalation pathway is 8.21×10^{-4} (50th percentile = 5.67×10^{-4}) and the 95th percentile is 2.40×10^{-3} . The annual cancer risk for a 30-year exposure period, which represents the probability of an individual to develop a fatal cancer due to radon inhalation over the average 30 years spent at the site, is 2.46×10^{-2} . This corresponds to a lifetime cancer risk for an average life expectancy of 70 years of 5.75×10^{-2} .

The cancer risk for external exposure (gamma radiation) is 2.96×10^{-4} (50th percentile = 3.03×10^{-4}) and the 95th percentile is 5.16×10^{-4} . The annual risk for a 30-year exposure period, is 8.89×10^{-3} . This corresponds to a lifetime cancer risk for an average life expectancy of 70 years of 2.07×10^{-2} .

As expected, and for this exposure scenario, radon inhalation pathway poses a higher cancer risk than external radiation exposure and therefore this is the main exposure pathway.

An estimated lifetime risk of lethal cancer of 5.75×10^{-2} for radon inhalation and of 2.07×10^{-2} for external gamma radiation exposure means that if an individual is born and lives in the critical group location during his entire lifetime (70 years), the probability of dying prematurely from radiation-induced cancer might be increased in 6 % and in 2 % due to radon inhalation (which will be even more accentuated when considering the inhalation of radon progeny daughters) and external gamma radiation exposure, respectively.

With the remediation of the Urgeiriça site and, in particular, by sealing off the tailings with a multi-layer cover, the external gamma radiation was reduced to an average value of $0.35 \mu\text{Gy/h}$ upon all the surface of the pile and the radon concentration averaged 60 Bq/m^3 after the placement of the cover. The calculated annual cancer risk and the incremental lifetime cancer risk for radon inhalation, in this new situation, is 9.12×10^{-5} and 6.38×10^{-3} , respectively. For external exposure the new values are 1.68×10^{-5} and 1.17×10^{-3} , respectively.

With the remediated areas, there has been a reduction by a factor of 2 in the absorbed gamma dose rates and in radon inhalation doses, following the remediation works, and 1 order of magnitude for the incremental lifetime cancer risk.

CONCLUSIONS

This work presents a study to evaluate the environmental impacts from the uranium mill tailing legacies before and after the remediation works focusing on two different approaches. One approach is based on a deterministic methodology for dose assessment and the second approach is based on a probabilistic methodology applying the Monte Carlo method to estimate the resulting cancer risk (annual, 30-years exposure period and incremental lifetime). Probabilistic distributions for almost input parameters were used in the risk calculation approach.

The dose and risk calculations were applied only to radon inhalation and gamma radiation external exposure but it can be extended to any other pathway as well as to other exposure scenarios, as long as the needed data are available. As an example, the main concern at Cunha Baixa mine site was groundwater contamination and in this way, water ingestion from wells should be the main target as well as contaminated soils near the mine site, used for agricultural purposes by the local's inhabitants. Moreover, the conceptual model and the input parameters of the exposure scenario should be verified periodically on the basis of available site characterization data. Other pathways also imply different radionuclides of concern. The deterministic approach may identify the exposure pathways that should have a low/high priority for further study and data acquisition.

The environmental remediation works at the Portuguese uranium legacy sites are not finished yet. One of the last largest mine complexes, the Quinta do Bispo open pit, is planned to be intervened only in 2020.

From the 61 radioactive sites identified at the end of 2015, 34 have already been remediated; in 6 mining areas there were on-going efforts to conclude the remediation works; and there were 21 uranium mining areas with remediation works to be carried out until 2022 [38]. In many of the remediated sites water treatment and effluents treatment based on active solutions are ongoing and it seems that these solutions will continue in a long-term without interruption. In addition, the monitoring and maintenance of these sites must assure that these wastes remain covered. Therefore, the assessment should be verified periodically along with some monitoring.

The methodology presented for dose and risk assessments may be applied to the other sites that are still to be intervened. The results may be used in the hierarchy process of the remediation stages as well integrated in the planning for long term stewardship during the remediation phases.

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