

TECHNICAL PAPER

Evaluation and study of radon occupational exposure levels in dams: a case study of the Gallery of Medau Zirimilis Dam (Sardinia, Italy)

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Key Points:

- Long-term exposure to radon in underground workplaces can increase lung cancer risk.
- The objective of recent legislation related to occupational exposure to radon is to protect workers' health and safety.
- This study provides technical guidelines for indoor radon measurements in tunnels and dam galleries.

Abstract

Protection of people from radon exposure in workplaces is an important factor in decreasing lung cancer risk. International authorities have set 'reference levels' to control enhanced radon exposure. Radiometric investigations, including in-situ radon measurements and laboratory gamma-spectroscopy, were carried out to monitor the radon exposure and define the radon transfer model in the workplaces of Medau-Zirimilis Dam in Sardinia. It was found that a shale formation is the main source of radon accumulated in the gallery of this dam. The water passing through the bedrock transports a considerable amount of sediments containing uranium-bearing minerals. Outgassing of radon from water flowing in the drainage canal and its accumulation due to poor ventilation could be the main reasons for the enhanced indoor radon level. The methodology introduced here can be adopted in underground workplaces like mines and tunnels to understand the natural radiation hazard and the effectiveness of the possible mitigation strategies.

Keywords: radon exposure; workplace; radiometric investigations; geological structure; radon transfer model

s uranium (²³⁸U) decays naturally over time, it releases radiation and forms a new radioactive element called ²²⁶Ra, which decays to form deadly radon (²²²Rn). Radon is a colourless, odourless and tasteless radioactive gas that is the most significant single fraction of radiation exposure from natural sources (1, 2). Radon can be found everywhere, including in the air, soil/ rocks and groundwater (3, 4). Long-term exposure to radon and its radioactive decay products (i.e. ²¹⁸Po, ²¹⁴Po, ²¹⁴Pb and ²¹⁴Bi) in dwellings, public buildings and workplaces (mainly by inhalation and digestion) may cause DNA alterations and increase lung cancer risk (5–7).

In the case of workplaces, exposure to high radon levels is more common in underground working areas (8), such as basements, mines, tunnels, caves, subway stations and waterworks (9, 10). The primary sources of high radon concentration are the gas flux from the soil containing contents of uranium-bearing minerals. Radon exhalation from construction material and radon dissolved in groundwater are the possible secondary sources (7, 11-13). In addition, in underground water work areas, radon can release from water to the air at high flow rates, and therefore, it can cause a significant dose contribution (10, 14, 15).

The radon concentration in the water depends on many factors, such as the pathway of the aquifers, the mineralogical composition of the aquifer and the permeability of the soil. Different aquifer rock types could have varying uranium contents and therefore also varying radium (radon's parent element) concentrations, which influences the radon concentration released as gas and dissolved in the groundwater (10, 16–18). To protect the public from the adverse health effects of radon, the European Commission recommended the Basic Safety Standard (Council Directive 2013/59/ Euratom), in which the reference level of 300 Bq.m⁻³ has been set up for radon concentration in dwellings and workplaces. The current Italian legislation on the protection from ionising radiation, especially Decree No. 230 of 17/03/1995 as amended by Decree No. 241 of 26/05/2000, also states that in workplaces with the presence of personnel for a total of 2,000 h per year, the average concentration of radon should not exceed 500 Bq.m⁻³.

Study objectives

This study aims to investigate the source of indoor radon and the mechanism of radon transfer from the source to the indoor environment in the gallery of Medau Zirimils Dam.

Previously, a study was sponsored by the Ente Acque della Sardegna (ENAS) water authority of Sardinia and has been entrusted by the spin-off company of the University of Cagliari (E-laboRAD). That survey was part of a project that began in May 2019 and included the verification of radon concentration within 27 days for the dams present in the territory of Sardinia, in which passive dosimeters with CR-39 detectors are used to measure the annual average radon exposure.

Records of elevated indoor radon concentrations were observed in this dam during the preliminary analysis of radon levels. This highlighted a condition that required further investigations (follow-up measurements) using an active radon monitor. Therefore, a set of in-situ and laboratory tests were considered to identify the source of radon and recognise how radon is transferred from the source to the indoor space of the Medau Zirimilis Dam. The measurement results were analysed, and the rate of radon production from solid sediments was estimated. Finally, possible remediation solutions for the protection of the life and health of the employees were introduced.

Materials and methods

Site description

The Medau Zirimilis Dam is an embankment dam, which is located on the Casteddu River southwest of Sardinia. The dam was constructed for irrigation purposes and to supply drinking water. The materials for the construction of the dam were taken from the pediments and terrace levels. The dam was completed in 1991 with a capacity of 19 Hm³, a longitude of 480 m and a maximum height of 151 m above sea level, reaching 44 m from the lower part of the reservoir (19).

As shown in Fig. 1, the dam is located at the transition area where the Paleozoic basement is related to the Hercynian orogenic evolution outcrops. The Paleozoic basement consists of an originally sedimentary succession (Lower Cambrian to Early Carboniferous) that was deformed during the Carboniferous period under the low metamorphic to anchizonal setting (20, 21). The Arburese Unit outcropping at the dam site consists of the Arenarie di San Vito Formation (Middle Cambrian to Lower



Fig. 1. Simplified map of the main geological features and the location of Medau Zirimilis Dam (10).

Ordovician), which is several hundred metres thick and is composed of decametric to metric alternations between micaceous metasandstones, quartzites and metasiltstones. Clastic, poorly cemented sediments, from gravels to sands and silts, of the Upper Pleistocene to the Holocene overlay the metamorphic basement (21).

The structural framework is mainly linked to the Hercynian Orogeny and Pliocene tectonic events. The Hercynian tectonics favoured the thrust of the Arburese Unit over the Upper Ordovician to Lower Carboniferous succession and its deformation in large-scale folding with axes of folds E–W and N–S oriented. The Pliocene tectonics shallowly modified the structural setting through normal fault systems E–W and N–S to NNW–SSE oriented (20–22). In the case of hydrogeological properties of the Paleozoic basement, it is known that the metamorphic rocks form a low-permeability aquifer where groundwater circulation occurs in the fissured zones close to the surface (on average, the first tens of metres). In the most heavily fractured zones (i.e. near the faults), the groundwater flow is more active and can occur at a greater depth (21, 23).

Methodology

The preliminary inspections done in the previous study showed elevated radon levels in the dam gallery.

Specifically, the outgassing and dispersion of radon dissolved in water (or, more accurately, radon derived from sediments) flowing through drainage systems was most likely to cause high indoor radon concentrations. In an attempt to verify this hypothesis, sediment samples from three different parts of the drainage canal of the gallery (see Fig. 2) together with a water sample were collected. In addition, the indoor radon concentration was monitored in the centre of the gallery, located at the red point in Fig. 2c, using two continuous radon monitors (CRMs) (MR1 plus and Radex MR-107). The collected sediments were first mixed, then oven-dried for 24 h at 110°C, sieved (mesh size finer than 200 microns) and eventually introduced to radon exhalation rate testing. The natural radionuclide concentrations of the homogenised sediment sample were also measured using the gamma-ray spectrometry technique. Furthermore, the concentration of radon in the water sample taken from the drainage system of the dam was determined.

Based on the results of these tests, the steady-state radon concentration, radon emanation factor and radon production rate of the sediment sample were calculated and compared with levels of radon dissolved in the water sample.





Fig. 2. (a) The gallery of Medau Zirimilis Dam, (b) the drainage canal of the dam gallery containing considerable contents of sediments and (c) the section plan of the dam and the radon measurement point is also specified.

Radon production rate of sediments

The experimental set-up to measure the radon exhalation rate from the surface of the sediment sample makes use of a Radex MR-107, which is an active radon monitor that was placed inside an accumulation chamber consisting of a pressure-resistant vacuum glass chamber. The soil sample was first weighted and placed inside a plastic container with a known volume. Afterward, as shown in Fig. S-2 (supplementary file), whilst the sample was placed inside the system, the chamber was efficiently closed to keep the air tightness.

The activity concentration of radon inside the chamber was measured hourly for about 120 h. The radon growth model released from the surface of the sediment sample can be modelled using the two-dimensional diffusion theory (24), according to Equation 1:

$$C(t) = C_0 e^{(-t\lambda_c)} + C_m (1 - e^{(-t\lambda_c)})$$
⁽¹⁾

Where (25)

 λ_{e} = effective radon decay constant (h⁻¹), 'accounting' for the radon decay constant, the leak rate of the system and the so-called 'back diffusion'

 C_0 and C_m = the radon concentration (Bq.m⁻³) in the closed chamber at time = 0 and its maximum value, respectively.

The values of λ_e and Cm for each sample were extrapolated using non-linear least-squares fitting of the experimental data with Equation 1 (26, 27), and then the radon emanation coefficient (E) was calculated using the following equation (28):

$$E = \frac{C_m V_{eff}}{A_{Ra} W}$$
(2)

Where

W = the weight of the sample (kg)

 A_{Ra} = the 226Ra mass activity (Bq.kg⁻¹) of the test sample

 V_{eff} = the effective volume of the sampling device (m³)

From the abovementioned parameters and considering the γ_d as the density of the disturbed sample, the radon production rate P_{Rn} (Bq.m⁻³.h⁻¹) was also estimated using the following equation (28, 29):

$$P_{Rn} = A_{Ra} E \gamma_d \lambda_e \tag{3}$$

Determination of radionuclide concentrations in sediment samples

The radionuclide (i.e. 226 Ra, 232 Th and 40 K) activity concentrations were measured using an Ortec NaI (Tl) detector (3 × 3 inches) placed in the laboratory of nuclear physics of the University of Cagliari (Fig. S-2,

4 (page number not for citation purpose) Supplementary file). The homogenised sediment sample (with a weight of approximately 32g) was filled in a plastic beaker, sealed carefully and left undisturbed for about 4 weeks to reach equilibrium in the ²³⁸U chain before measurement. IAEA reference sources of RGU1 and RGTh1 together with a high pure potassium nitrate salt were used for calibration of the system. Energy calibration of the detector was carried out using disc sources of ⁶⁰Co and ¹³⁷Cs. The spectrum was processed by the MAESTRO multichannel analyser emulation software. The effect of the background was also filtered whilst doing the calculations for the estimation of the activity concentration of radionuclides

Radon in water measurement

To measure the concentration of radon in water, the MR1 plus radon monitor was connected to the H_2O -Kit accessory, which was provided by the manufacturer (see Fig. S-3, Supplementary file). A glass vessel of 300 mL of mixed active water sampled from three locations within the drainage canal of the dam gallery was set up in a closed-air loop with the MR1 plus. By operation of the instrument's pump, the sample was aerated via a bubbler, and the radon that was in the water throughout the loop was distributed. Then, the radon gas was introduced into the Lucas cell. A porous membrane filter was used to stop water spray from reaching the Lucas.

Results and discussion

At the time of writing this paper, no ventilation method was adopted in the dam gallery. The results of the radon survey in this paper and related comments regarding the radon transfer scenario expressed herein are based on the available geological information, the conditions recorded during site investigation work and the results of tests made in the field and laboratory. However, there may be conditions existing at the site, which have not been disclosed by the investigation available and which have not been taken into account in the paper. Results obtained in this paper provide no other representation whether expressed or implied, in relation to the real-time levels of epidemiological risk of natural radiation in the Medau Zirimilis Dam.

The indoor radon level in the gallery of Medau Zirimilis dam was monitored for 72 h using two continuous radon monitors. Due to the loss of the electric power supply during the test operation, the MR1 plus was turned off. Because of this, only the values recorded by Radex MR-107 are presented here (see Fig. 3). According to this figure, although there are fluctuations in the reordered indoor radon concentrations, the radon activity in the dam gallery tends to increase over time due to gas accumulation in a closed space (dam gallery). It is worth adding that the enclosed space condition was maintained



Fig. 3. Results of indoor radon concentration monitoring inside the dam gallery and the trend line of radon activity growth (red line).

during the testing to allow the build-up of radon and simulate actual conditions. The fluctuations seen in the measured radon values are mainly because of disequilibrium between the radon decay products. This can be because of the large dimensions of the dam gallery and therefore longer testing time, which would be required. However, the trend line chart in Fig. 3 shows that radon grows up and tends to reach a flat line, but as mentioned earlier, longer measurement period would have been required to see a rather full flat line in the radon activity growth chart. It usually takes 3–7 days to reach a relative equilibrium in a closed space. In general, the results of indoor radon monitoring show a remarkably high value of the mean indoor concentration of radon of about 683 ± 516 Bq.m⁻³ with minimum and maximum values of 30 and 1,998 Bq.m-3, respectively (see Table 1). To identify the source of indoor radon, sediment samples together with a water sample were collected. The sediments were introduced to the gamma-ray spectrometry and radon exhalation rate tests. The results of the measurements are shown in Figs 4 and 5. A significantly high 226 Ra concentration (226 ± 7 Bq.kg⁻¹) was found in the sediment sample. However, the concentration of ²³²Th (37 \pm 5 Bq.kg⁻¹) and ⁴⁰K (210 \pm 198 Bq.kg⁻¹) was relatively low. These concentrations may be justified by the behaviour of these radioisotopes in the environment. In fact, when the water passes through the bedrocks hosting uranium-bearing minerals (the shales from the Genna Muxerra formation can be an example for this study - see also Fig. 1), ²²⁶Ra from the host rocks can be easily dissolved in water due to its relative solubility and transported through the aquifer. Whilst ²³²Th is very stable, it does not dissolve in a solution (30) and therefore cannot be transported by water. The low concentration of ⁴⁰K can also be related to the low concentration of this element in the host rock.

To measure the radon production rate, the technique of radon accumulation inside a closed chamber was

Table 1. Summary of radon in the air and radon in water test results

Measurement type	Testing time (h)	Statistic of radon concentrations (Bq.m ⁻³)			
		Mean ± SD	Median	Min	Max
Radon in the air	72	683 ± 516	554	30	1,998
Radon in the water	146	4,760 ± 3,879	3,968	203	17,268



Fig. 4. Monitored radon concentrations released from sediment sample and the fitted radon growth model, based on the results of radon exhalation rate testing.

employed. Using the non-linear least-square fitting of Equation 1 on the measured data, the steady-state radon concentration was predicted to be $3,652 \pm 160$ Bq.m⁻³, and then utilising Equations 2 and 3, the radon emanation factor and the radon production rate of the sediment sample were calculated (Table 2). A considerably high value of radon production rate was estimated (1,380 ± 239 Bq.m⁻³.h⁻¹). This can be explained by the presence of high ²²⁶Ra content and small particle size (less than 200 microns) of the sediments that can cause higher emanation of radon and subsequently higher gas production rates.



Fig. 5. Gamma spectrum of the sediment sample obtained by the NaI (Tl) detector.

Table 2. Natural radionuclide activity concentrations and estimated values of radon emanation factor and radon production rates of the solid sediment

Radionuclide activity concentrations				
$Ra \pm \sigma (Bq.kg^{-1})$	Th $\pm \sigma$ (Bq.kg ⁻¹)	$K \pm \sigma$ (Bq.kg ⁻¹)		
226 ± 7	37 ± 5	210 ± 198		
R	adon exhalation rate testing			
Extrapolated steady-state activity ± standard error (Bq.m ⁻³)	Radon emanation factor	Radon production rate (Bq.m ⁻³ .h ⁻¹)		
3,652 ± 160	3.2 ±0.17	1,380 ± 239		



Fig. 6. Monitored radon in water concentrations, and the water sampled from the drainage system.

The radon concentration of the water sample containing 300 mL of mixed active water was measured for about 140 h according to the Lucas cell method. As presented in Table 1, the mean concentration over the measurement period is about 4.96 kBq.m⁻³. This concentration ranges between 0.2 and 17.2 kBq.m⁻³. Figure 6 shows the recorded values of radon in water concentrations in which high variations can be seen. The reason for this is the presence of the non-equilibrium state between radon and its progeny. In fact, the measurement of radon in water is very complicated. Technically, the setup for radon in water measurement aerates the water to allow radon released into the air, and then radon in the air is measured. Continuous operation of the pump to aerate the water does disturb the equilibrium state between radon and its progeny, and this is why we see high fluctuations. Therefore, probably internal factors like the pumping rate and the parameters related to the aeration from the water surface are responsible for the variations.

In surface waters such as lakes and rivers, the radon concentration levels are generally very low values in the range of 5 to 10 Bq.m⁻³ since radon can readily escape from the water surface and subsequently diluted in the free air (31). However, some lakes with high levels of uranium or other radioactive minerals in the surrounding rocks and soil may have higher levels of radon. In the case of outgassing of radon in underground spaces (e.g. Waterworks), the radon released from water accumulates in indoor spaces and contributes to high concentrations. It is believed that the same phenomenon occurs in the gallery of Medau Zirimilis Dam. The radon originating from uranium-bearing minerals of the bedrock is transported through the aquifer and penetrates the gallery through the cracks and joints in the structure. Then, whilst the water is being collected by the drainage system of the dam, a part of this radon escapes to the indoor air due to the aeration process. Due to the lack of proper ventilation, the released radon can accumulate inside the gallery in concentrations that may exceed the reference levels.

Conclusions

This study aimed to first evaluate the indoor radon exposure levels of the staff working in the gallery of Medau Zirimilis dam. Elevated values of indoor radon were found in initial investigations. Therefore, further measurements were considered in this study in an attempt to identify the source of radon and explore how it enters the gallery. Geogenic variables such as the geochemical mineralogical properties of bedrock are the primary parameters that affect the concentration of radon in groundwater. The presence of fractures and fissured zones close to the surface provides pathways for radon transportation through the aqueous media. In the cases where water found a way to the surface, the radon can be exhaled into the air from the water's surface. Consequently, because of poor ventilation, radon can accumulate in indoor spaces and therefore can potentially increase lung cancer risk for workers. The possible ways to decrease radon concentration mainly include limitation of radon entries and/or increase of the ventilation rates. Implementation of fans and even taking benefits of natural ventilation by providing more air entries (removing the glasses in windows and replacing them with perforated meshes) can help to decrease the indoor radon concentration effectively. Besides, by identifying cracks, holes and joints of the gallery's structure and restricting water entry routes to the gallery (e.g. by means of cement paste), it would be possible to decrease the amount of water flowing in the discharge channel of the dam gallery and limiting radon release from water to the air. This would also increase the efficiency of the ventilation systems as there would be less radon in the air that needs to be mitigated.

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Open Research

The indoor radon concentration data measured by a third party using CR-39 detectors are not available to the public. All the radon survey data utilised here are created for this research only, and the digitised format of data is available upon sending an email request to the corresponding author.

Conflict of interest and funding

The author declare no conflict of interest.

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