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Abstract

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Keywords

Radon (222Rn), inhalation dose, health risk, underground uranium mines

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Contribution of mine water and uranium ore rocks to the ²²²Rn-induced radiation dose received by the mine workers in a low-ore grade underground uranium mine, India

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Abstract

More than 50% of the radiation dose received by underground mine workers is mainly due to the inhalation of radon (²²²Rn) gas and its decay products in an underground mine working space. Monitoring and controlling of ²²²Rn exhalation in the underground mine working play a vital role in minimizing the radiation risk hazards to the mine workers. This study discusses the contribution of mine water and uranium ore to ²²²Rn activity concentration in mine air and its health risk assessment. The annual effective radiation dose (E_{Rn}) due to inhalation of ²²²Rn for mine workers is estimated at 0.10 mSv/y. Furthermore, the estimated excess lifetime cancer risk (ELCR) and radon-induced lung cancer per million per person (RnLCC) is found to be 0.3×10^{-3} and 0.002×10^{-6} . The estimated results of E_{Rn} and RnLCC due to the inhalation of ²²²Rn are well within the prescribed limits of the international regulatory agencies.

Keywords: radon (²²²Rn), inhalation dose, health risk, underground uranium mines

1. Introduction

adon and its decay products of Naturally Κ Occurring Radioactive Materials (NORMs) like uranium and thorium are the major sources of radiation hazards to underground mine workers. The decay of these radioactive materials causes external as well as internal radiation hazards to the workers due to its prolonged exposure. The external radiation is mainly caused due to gamma exposure (γ) , whereas the internal radiation is due to the inhalation of radon gas, produced as a daughter product in the radioactive decay process in the mine environment [1,2]. There are other radionuclides that contribute to the external radiation, but their effect and epidemiological hazards are less than the internal radiation effects [3]. The global average annual human exposure from the natural sources is 2.4 mSv/y, out of which 1.15 mSv/y is due to radon [2].

Radon has three natural isotopes: ²²²Rn ($T_{1/2} = 3.82$ days), ²²⁰Rn ($T_{1/2} = 55.6$ s) and ²¹⁹Rn ($T_{1/2} = 3.92$ s).

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The half-life of the latter radon isotopes is shorter; hence they are incapable of cumulative addition in the internal radiation in the underground mine working places [3].²²²Rn is the immediate daughter product of radium (²²⁶Ra) of the uranium (²³⁸U) decay series, which is abundantly present on earth. In underground uranium mines, the major sources of ²²²Rn emanation were identified as ore rocks, broken ore rock, backfill mill tailings, and mine water [3-5]. The emanation rate of ²²²Rn from the sources depends on various factors such as ore grade, radium content, aerosol particle size, and water flow rate [4,6-8]. Several epidemiological studies have evidenced that exposure of humans to ²²²Rn and its daughter products caused severe diseases like lung cancer, myeloid leukemia, emphysema, respiratory disorders, and mutation of cells in the sputum [9,10]. To reduce the radiation risk within the stipulated safe limits in underground workplaces, appropriate monitoring and measures are considered [1,11-14]. ICRP [16] recommends lower and upper ²²²Rn

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concentration limits of 500-1500 Bg/m³ for underground workplaces. ICRP [11] recommends a reference level of 1000 Bg/m^3 for radon exposure in mines. ICRP [28] recommends 10 mSv per year due to radon exposure which gives approximately 300 Bg/m^3 . United States Public Health Service (USPHS) reports that an increase in lung cancer frequency is observed for a span of 10 years of exposure to the ²²²Rn concentration varying between 1850-5550 Bg/m³ [15]. Several regulatory bodies have recommended the safe limits of radiation exposure, i.e., the International Atomic Energy Agency (IAEA), the International Commission on Radiological Protection (ICRP), the United States Environmental Protection Agency (USEPA), the World Health Organization (WHO), etc. ICRP recommended a limit for exposure of 100 mSv in a specified block of 5 years, i.e., an average of 20 mSv per annum for the underground working space [16]. Also, the International Agency for Atomic Energy (IAEA) recommended the Equilibrium Equivalent radon Concentration (EER) of 1000 Bq m^{-3} in the working space, and USEPA (1991) stipulates the 222 Rn concentration safe limits as 11 Bq/ dm³ for water.

The present study aims to estimate the ²²²Rn inhalation dose and its health risks due to the sources (ore rocks, broken ore materials, and mine water) present in the underground uranium mines using a Smart Radon Monitor (SRM). Very few studies on the contribution of the different sources to ²²²Rn exposure mines were conducted in underground uranium mines. This study shows that the contribution of ²²²Rn activity concentration due to mine water is higher than the ore rocks present in the underground mine environment. This variation is due to the mineral content and ore grade of mine water and ore rocks present in the mine environment. Further, this study was undertaken to estimate the health risk due to the miners' ²²²Rn inhalation in the underground mines.

2. Materials and methods

2.1. Study area and sample collection

Figure 1 shows the study area of the sample collection, located in southeast Singhbhum of Jharkhand, India. The study area is a low-ore-grade underground uranium mine as the ore grade is less than 1% U₃O₈. Thus, the radiation hazards due to external exposure and long-lived α -emitters are not significant. However, the radiological hazards associated with the mining of uranium ore cannot be neglected, even if the grade of uranium ore is low. The most important radiation hazard in a lowgrade uranium mine is due to inhalation of radon and its short-lived decay products.

A split ventilation system is followed in the mine. The ventilation system is continuously upgraded to provide air quantity commensurate with the operations. Ventilation of the mine is provided by means of three axial-vane-type exhaust fans located on the surface at three different places. The total quantity of air handled by these fans is about 230 m³ s⁻¹. Under the influence of negative pressure ranging from 10 to 170 mmwg created by these fans, intake air is taken into the mine through the shafts and declines. It is used to ventilate all the levels and stopes by means of suitably placed air locks and ventilation doors. The fresh air requirement for each level depends on the radon exhalation from different sources, viz. ore body, waste rock, broken ore stockpiles in the production zones, backfill tailings and mine water. The advancing faces are ventilated with auxiliary fans located in mine airways and the air is delivered to the face through metal/canvas ducts of 50 cm diameter.

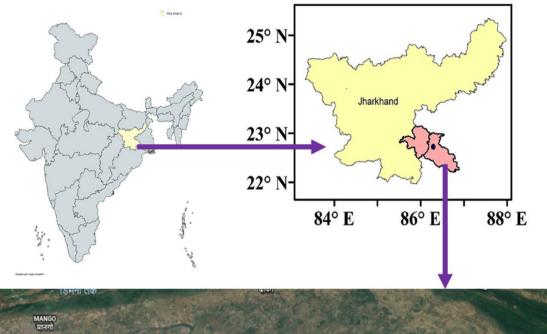
The ore rock and broken rock materials (16) and mine water (21) samples were collected as per the standard procedure [17,18] for the analysis purpose. The mine working depth usually varies from 450 m to 700 m, having two (02) mine entrances and a horizontal cut and fill working method. The ore rock and broken rock samples were collected in airtight plastic bags and the water samples in leak-proof, lowpermeability bottles to reduce the risk of ²²²Rn gas dissolution in the atmosphere. Water samples were collected using 60 cm³ capacity leak-proof bottles.

2.2. Experimental setup

The measurement of radon exhalation from the collected samples was carried out by Smart Radon Monitor (SRM) assembled with an airtight exhalation chamber and bubbler kit with a water bottle, developed by BARC, India [17,18]. The ²²²Rn surface exhalation and mass exhalation rates were measured by placing the uranium ore samples in an airtight ²²²Rn exhalation chamber with a volume of 4.4 dm³ volume connected to a Smart Radon Monitor (SRM) in a closed-loop. The detailed method of measurement of ²²²Rn surface exhalation and mass exhalation rates from the ore body is available in the literature elsewhere [17].

Dissolved radon content in the mine water was measured by connecting the sampling bottles to the bubbler kit and smart radon monitor (SRM) in a closed-loop. This monitor consists of a ZnS (Ag) based Scintillation cell (150 cm³) with a ²²²Rn progeny filter, an inbuilt air suction pump, and a

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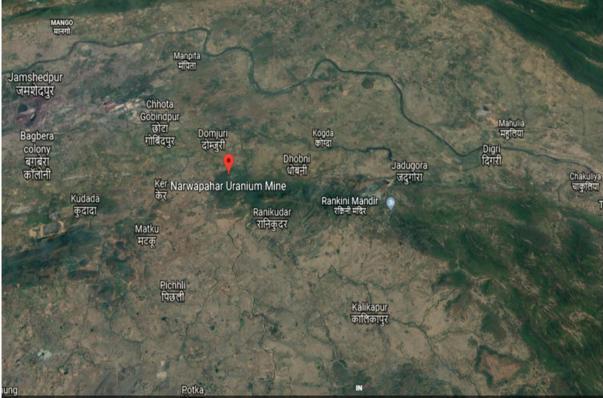


Fig. 1. Map of India showing study area location [17].

photomultiplier (PMT) tube. In this method, the sampling bottle was connected to the bubbler, and care was also taken to avoid bubble formation. The bubbler kit transferred a significant fraction of ²²²Rn dissolved in water to air volume of setup. The detailed method of measurement of dissolved radon content in the mine water is available in the literature elsewhere [18].

3. ²²²Rn activity concentration and dose calculation

In this study, dose estimation due to ²²²Rn gas inhalation is calculated based on radon concentration present in the mine air. The estimation of dose is carried out in two phases, i.e., dose due to uranium ore and dose due to water present in the underground mines.

3.1. ²²²Rn activity concentration estimation due to broken materials in the underground mine working

The analysis of ²²²Rn activity concentration in the underground mine working was based on the exhalation characteristics and airflow quantity [17]. The exhalation characteristics (surface and mass exhalation rate) were discussed elsewhere [17]. The following mathematical expression for the estimation of ²²²Rn activity concentration (C_{Rn}) in the underground mine working is given by Sahu et al. [26]:

$$C_{\rm Rn} = \frac{\left[\left\{\left(J_s\right)_o \times S_o\right\} + \left\{V_b \times \rho \times \left(J_m\right)_o\right\}\right]}{q} \tag{1}$$

where, $(J_s)_o$ is the ²²²Rn surface exhalation rate from the uranium ore (Bq/s per m²), S_o is the exposed area of the orebody (m²), V_b is the volume of broken material (m³), ρ is the ore density (kg/m³), $(J_m)_o$ is the ²²²Rn mass exhalation rate from the broken material (Bq/s per kg), and q is the airflow rate (m³/s) in the underground mine working place.

²²²Rn mass exhalation rate J_m was estimated based on build-up ²²²Rn concentration inside the accumulation chamber of SRM and the quantity M of the investigated sample in terms of kilograms. Mathematically, it is expressed as [26]:

$$J_m = \frac{(C_t - C_0 e^{-\lambda_{em} t_m}) \times V_e \times \lambda_{em}}{(1 - e^{-\lambda_{em} t_m}) \times M}$$
(2)

where C_t is the ²²²Rn concentration at the measurement time (Bq/m³), C_0 is the background ²²²Rn concentration (Bq/m³), λ_{em} is the ²²²Rn decay constant (7.52 × 10⁻⁰³ h⁻¹), V_e is effective size of the concentration chamber (m³), and t_m is the measurement time (h).

²²²Rn surface exhalation rate J_s was estimated based on ²²²Rn concentration accumulated in the accumulation chamber of SRM and the surface area A of the investigated sample in terms of square meters. The ²²²Rn surface exhalation rate of the sample was predicted using the following relation [26]:

$$J_s = \frac{(C_t - C_0 e^{-\lambda_{es} t_s}) \times V_e \times \lambda_{es}}{(1 - e^{-\lambda_{es} t_s}) \times A}$$
(3)

Where λ_{es} is ²²²Rn decay constant (2.09 × 10⁻⁰⁶ s⁻¹), and t_s is measurement time (s).

3.2. Dose estimation due to uranium ore

Inhalation dose estimation due to the ore rock and broken rock materials in the underground mines was estimated based on Equilibrium Equivalent Concentration (EEC) of radon gas and dose coefficient of 10 mSv per WLM (Working Level per Month) [12]. The following relationship was used to determine the contribution of uranium ore to radon dose E_{Rn} [27]:

$$E_{\rm Rn} = \frac{T_e \times C_{\rm Rn} \times F \times 10}{170 \times 3750} \tag{4}$$

where, T_e is the exposure time per year, and C_{Rn} is the total ²²²Rn activity concentration (Bq/m³). For forced-air ventilated mines, the equilibrium factor (*F*) is 0.2 [12]. However, the equilibrium factor can vary from 0.01 to 0.8 depending on the efficiency of the ventilation system.

3.3. Dose estimation due to mine water

Radon gas (²²²Rn) dissolved in the water affects human through its consumption as drinking (ingestion) and release of dissolved ²²²Rn in the environment (inhalation). In this study, effective dose due to ingestion is not considered, since consumption of mine water is prohibited as per the statutory laws in the mine. The annual dose due to inhalation of ²²²Rn E_{In} released from the mine water is calculated using the following expression [1]:

$$E_{In} = C_{RnW} \times Rn_{aW} \times T_{om} \times D_{cf} \times F$$
(5)

where, C_{RnW} is the dissolved ²²²Rn concentration of mine water (Bq/dm³), Rn_{*aW*} (10⁻⁴) is the ratio of ²²²Rn concentration in air and water [1], T_{om} (2000 h/y) is the average occupancy time for a miner [11], D_{cf} (15.7 nSv $[Bq m^{-3}]^{-1}$) is the dose conversion factor [12], and F (0.2) is the equilibrium factor between ²²²Rn and its daughter products for underground mines ventilated by forcing-fan [12]. The dissolved ²²²Rn concentration of mine water was measured using a Smart Radon Monitor (SRM) connected to a bubbler kit. The water sample was collected in a leak-tight bottle (volume ~60 cm³) of low permeability material provided in the radon in the water kit. While sampling into the bottle, caution must be taken to avoid the formation of bubbles/agitation in the liquid. Measurements were performed within 4-5 h after the sampling. The method for measuring dissolved ²²²Rn concentration was provided elsewhere [18].

In the present study, it is assumed that there were 250 working days per year and 8-h working shifts in the underground mine. Thus, a miner is exposed to a constant value of WL for the duration of about 2000 h per year. The occupational exposure time period of 2000 h year⁻¹ for mine workers includes

the time spent by the miners at different places starting from the surface facilities to the mine entrance, underground travel ways, and finally, at actual work sites such as stopes and development drives. The relaxation period in the change room and time spent at the surface job are also included in this overall occupancy period [21].

The total effective radiation dose E_T due to the inhalation of ²²²Rn gas released from different sources present in the mine atmosphere is estimated by

$$E_T = E_{\rm Rn} + E_{\rm In} \tag{6}$$

3.4. Health risk due to ²²²Rn inhalation

The health risks due to the prolonged exposure of ²²²Rn in the underground mine workings are expressed in terms of excess lifetime cancer risk (ELCR) and ²²²Rn-induced lung cancer cases per year per million persons (RnLCC). The equations for ELCR and RnLCC are given below [18,19]:

$$ELCR = E_T \times W_u \times f_{cr} \times 10^{-3} \tag{7}$$

$$RnLCC = E_T \times LCRF \times 10^{-3}$$
(8)

where, W_y is the total working periods by a miner in the mine (47 years), f_{cr} is the fatal cancer risk factor (0.055 per mSv) reported by ICRP-103 [16], LCRF is the lung cancer induction risk factor $(18 \times 10^{-6} \text{ mSv}^{-1})$ reported by Sherafat et al. [20]. In India, the total working periods of a miner in the mine have been assumed to be 47 years, considering the joining age and retirement age of the miner to be 18 and 65 years, respectively.

4. Result and discussion

4.1. ²²²Rn concentration contributed by uranium ore and mine water

The statistical analysis of the ²²²Rn concentration contributed by uranium ore and mine water in a low-ore grade uranium mine is presented in Table 1. The build-up radon concentration contributed by uranium ore was obtained directly from the Smart Radon Monitor connected to the accumulation chamber in which uranium ore samples were placed. The build-up radon concentrations emitted from uranium ore samples varied from 14 to 1300 Bq/m³ with an arithmetic mean of 255 ± 336 Bg/m³. The significant variations in radon concentration may be attributed to the variations of ore grade, radon exhalation characteristics, ²²⁶Ra

Table 1. Statistical parameters of ²²²Rn concentration contributed by uranium ore and mine water.

| Parameters | Uranium ore (Bq/m ³) | Mine water (Bq/dm ³) [18] | |
|---------------------------------|-------------------------------------|--|--|
| No. of samples | 16 | 21 | |
| Range | 14-1300 | 0.4 - 18.2 | |
| Mean | 255 | 7.8 | |
| Standard deviation | 336 | 6.6 | |
| Geometric mean | 124 | 4 | |
| Geometric standard deviation | 3.6 | 4.3 | |

activity concentration, and mineral composition of the uranium ore. The build-up radon concentration values were used to determine the radon mass and surface exhalation rates as provided in Equations (2) and (3).

For calculating ²²²Rn concentration contributed by uranium ore in the mine atmosphere, the total airflow rate, the density, the volume of broken material and the exposed surface area offered for radon exhalation in the different underground uranium mines were taken as 2669 kg/m³, 230 m³/s, 953 m³ and 26,290 m², respectively. It has been reported that the mean 222 Rn surface and mass exhalation rates of uranium ore are 0.17 ± 0.2 Bq/s per m² and 11.8 ±12.4 Bq/s per kg, respectively (Table 2). From Table 2, the maximum contribution of uranium ore to radon concentration in the mine was found to be 228 Bg/m³. The mean radon activity concentration contributed by uranium ore was reported as 30.34 Bq/m³ [17]. The mean radon concentration was about 6% of the lower radon concentration limit and about 2% of the upper radon concentration limit prescribed by ICRP-103 [16]. Further, it was about 3% of the safe radon concentration limit stipulated by ICRP-115 [11]. The mean radon concentration was observed to be well below the reference level of radon concentration prescribed by the regulatory bodies. It revealed that there was adequate air quantity to keep the radon concentration below the prescribed limits in the underground mine.

The dissolved radon concentration of mine water was measured using Smart Radon Monitor (SRM)

Table 2. Statistical analysis of ²²²Rn surface and mass exhalation due to ore rock and broken materials [17].

| Parameter | Range | Mean | Standard deviation | Geometric mean | GSD |
|---|-----------|------|--------------------|-------------------|-----|
| Surface exhalation (Bg/s per m ²) | 0.01-0.74 | 0.17 | 0.2 | 0.09 | 3.3 |
| Mass exhalation (Bq/h per kg) | 0.9–47 | 11.4 | 12.8 | 6.6 | 3.1 |

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connected to bubbler kit. The dissolved concentraton varied from 0.4 to 18.2 Bq/dm³ with an arithmetic mean of 7.8 \pm 6.6 Bq/dm³ [18]. The variation in dissolved radon content depends on the different parameters such as the type of uranium minerals through which water flows, distance from the water sources, and degree of turbulence in the water flow path. The average dissolved radon concentration was about 8% of the threshold limit value of 100 Bg/dm³ recommended by WHO [14]. Based on the dissolved radon content of mine water and the ratio of ²²²Rn concentration in air and water, the mean radon concentration contributed by the mine water to the mine atmosphere was worked out to be about 0.78 Bg/m^3 . It revealed that the contribution of mine water to radon concentration in the mine atmosphere was much lesser than that of uranium ore.

4.2. Contribution of various sources to ²²²Rn dose

Generally, mine water is not used for drinking purposes in underground mine workplaces. Thus, the ²²²Rn inhalation dose is considered in this study. ²²²Rn inhalation dose contributed by the uranium ore was calculated based on the average occupancy time for a miner in a year, the contribution of ore to ²²²Rn activity concentration, the equilibrium factor between ²²²Rn and its daughter products in underground mine ventilated by forcing-fan as provided in Equation (4). The maximum radon dose contributed by the uranium ore was found to be 0.715 mSv/y. The mean ²²²Rn inhalation dose was reported as 0.095 mSv/y [17]. On the other hand, the radon dose contributed by mine water was calculated based on the dissolved ²²²Rn concentration of mine water (Bq/dm³), the ratio of ²²²Rn concentration in air and water, the exposure time per year, the equilibrium factor between ²²²Rn and its daughter products as provided in Equation (5). The maxium radon dose contributed by mine water was found to be 0.011 mSv/y. The contribution of mine water to radon dose was reported as 0.005 mSv/y [18]. Thus, the total maximum and mean ²²²Rn inhalation doses received by the miners contributed by the above two sources were 0.726 and 0.10 mSv/y, respectively. The estimated inhalation dose was far below the threshold limit of 20 mSv/y [16]. However, the World Health Organization reported that there is no real "upper limit" for exposure to radon concentration. So, the best strategy is to keep the exposure As Low As Reasonably Achievable (ALARA). To achieve the ALARA principle, all mining methods and operations must be optimized. The primary focus should be given to controlling the exposures by engineering means, such as providing adequate

Table 3. Inhalation radiation dose levels for nearby studied underground uranium mines, India.

| Mine | Inhalation radiation dose (mSv/y) | Reference |
|----------------------------|--------------------------------------|------------------|
| Jaduguda mines, India | 6.95 | [21] |
| | 2.10 | [22] |
| Bagjata mines, India | 1.76 | [23] |
| Narwapahar mines, India | 5.44-7.33 | [24] |
| | 2.30 | [3] |
| | 0.10 | Present study |

ventilation, adopting a safe equipment layout and work procedures.

Table 3 shows the previous investigation on ²²²Rn inhalation exposure carried out in different underground uranium mines in India. The radon dose obtained in the present study was much lower than the previous values reported by the other investigators. However, since the present study considered only two sources of radon dose, the proper conclusion could not be drawn from Table 3.

4.3. Health risk

Excess Lifetime Cancer Risk (ELCR) was calculated based on total radon dose, the total working years of a miner and fatal cancer risk factor as provided in Equation (7). ²²²Rn-induced lung cancer cases per year per million persons was calculated based on total radon dose and the lung cancer induction factor as provided in Equation (8). The estimated excess lung cancer risk (ELCR) and ²²²Rn induced lung cancer cases per year per million persons (RnLCC) were found to be 0.26×10^{-3} and 0.002×10^{-6} , respectively. The estimated results of RnLCC due to the inhalation of ²²²Rn are well within the prescribed safe limits of 170–230 \times 10^{-6} [25]. The mean RnLCC value in the mine may be higher than this estimated value as the ²²²Rn exhalation in the mines also gets affected by other factors such as backfill materials, ventilation parameters, and geological conditions (hydrology, barometric pressure, fold and fault) [4].

5. Conclusion

In the present study, ²²²Rn activity concentration due to the ore rocks, broken materials, and mine water collected from the underground uranium mine was determined by using Smart Radon Monitor (SRM). This monitor is portable, low-cost, highly sensitive, less time-consuming, and a user-

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friendly instrument useful for active monitoring of ²²²Rn concentration in underground working spaces. This study shows that the estimated ²²²Rn effective dose is 0.5% of the safe limit of the radiation dose prescribed by ICRP. Higher ²²²Rn concentration in mine water is due to the mineral content and geological parameters of the underground working space, i.e., rock properties and radium content. The estimated effective annual dose may be higher due to other parameters such as backfill materials, hydrogeology of the area, mining operations such as drilling and blasting, etc. Thus, it is recommended to further study the contribution of ²²²Rn in the underground working space due to these parameters.

Ethical statement

The authors state that the research was conducted according to ethical standards.

Funding body

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Conflicts of interest

The authors declare no conflict of interest.

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