

<https://doi.org/10.48047/AFJBS.6.15.2024.6664-6679>



African Journal of Biological Sciences

Journal homepage: <http://www.afjbs.com>



Research Paper

Open Access

Measurement of Radon Evaluation in Soil Samples Collected Over Groundwater and Non-groundwater Sites from Pune City Area.

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Volume 6, Issue 15, Sep 2024

Received: 15 July 2024

Accepted: 25 Aug 2024

Published: 05 Sep 2024

doi: [10.48047/AFJBS.6.15.2024.6664-6679](https://doi.org/10.48047/AFJBS.6.15.2024.6664-6679)

Abstract:

Soil is the uppermost layer of the earth surface playing crucial role in supporting life by providing essential component. It consists of organic and Inorganic matter including natural radionuclides like Uranium (^{238}U), Thorium (^{232}Th), Potassium (^{40}K). Radon(^{220}Rn) and thoron(^{222}Rn) is a decay product of radium and Thorium. These gases escape from soil matrix and enter homes through emanation and exhalation. The half life span of the radon atom is 3.824 days, can diffuse within the indoor environment and travel a few meters away. The Study encompasses 11 Ground water (GW) and 11 Non- Ground water (NGW) sites in Pune, Maharashtra, India. The soil samples were analysed with HPGe Detector and Smart RnDuO Detector for the Radon and thoron measurement. The average global concentrations of ^{226}Ra , ^{232}Th , and ^{40}K are 32 Bqkg^{-1} , 45 Bqkg^{-1} , and 412 Bqkg^{-1} , respectively. The results obtained for the radon concentration, mass and surface exhalation rate in the present study are within the range of the worldwide average value and remain higher for GW than NGW. The study suggests that the radionuclide activity concentrations in the studied regions are within safe limits and do not pose significant risks to public health.

Key Words: Radon, Thoron, Radioactive, Natural Radioactivity.

1.1. Introduction:

Soil, as the upper layer of the Earth's crust, plays a crucial role in supporting life by providing essential components. It also contributes significantly to the natural background radiation dose received by human beings (UNSCEAR, 2008; Karunakara et al., 2014;

Yasshodara et al., 2013; Murugesan et al., 2016; Daulta et al. 2019; Belyaeva et al., 2021; Filgueira et al., 2020). The formation of soil involves various physical and chemical processes such as rock deformation, weathering, decomposition, organic matter addition, and water movement. It consists of organic and inorganic matter, including natural radionuclides like Uranium (^{238}U), Thorium (^{232}Th), Potassium (^{40}K), and their decay products, which are inherent constituents of soil. The distribution and radiological effects of these primordial radionuclides are of utmost importance for human health (Amanjeet, et al., 2017; Lamarsh, 1983; Sing et al., 2017; Sannappa et al. 2003; Ridvan et al.,2011; Ramola et al., 2008; Tzortzis et al., 2004; Chiozzi et al., 2002; Arafa, 2004).

Radon (^{222}Rn) and thoron (^{220}Rn) gases are decay products of radium (^{226}Ra) and thorium (^{232}Th) (Nazaroff, et al., 1988; Lara, et al., 2015; UNSCAR, 2000). The primary source of indoor radon is the underlying bedrock and soil, as uranium and thorium are present in all types of soils and rocks on the Earth's surface. These gases escape from the soil matrix and enter our homes through a two-stage process: emanation and exhalation. During emanation, radon and thoron atoms are released from solid mineral grains into air-filled pores. In the exhalation process, they are transported to the atmosphere through convection and diffusion. The exhalation rate serves as an indicator of natural radioactivity levels, representing the emission of radon per unit area per unit time. Numerous factors influence the exhalation rate, including the concentration of parent radium content, porosity of soil or rock materials, atmospheric temperature, pressure differentials, geological and meteorological factors, ventilation rate in dwellings, and more.

Radon, with its relatively longer half-life (3.824 days), can diffuse within the indoor environment and travel a few meters away. Thus, the primary contribution to indoor activity typically comes from the top few meters of the subsurface soil. On the other hand, thoron, with its shorter half-life (55.6 seconds), mainly contributes to the indoor environment from the top layer of the subsurface soil. The exhalation of these gases from soil and building materials constitutes the main sources of indoor exposure (Ramola, 2008; Che et al.,2010; Aswal, 2016; Bourai,2016; Singh,2016).

The aim of this research is to assess the concentrations of ^{222}Rn , ^{220}Rn , in soil samples collected from groundwater (GW) and non-groundwater (NGW) sites in the Pune region, Maharashtra. A comparative study on the assessment of radon concentration in soil samples collected from GW and NGW sites in the Pune region, Maharashtra, India is essential for

several reasons. Radon is a radioactive gas that emanates from the ground and can pose health risks when it accumulates in indoor environments. Understanding the differences in radon concentrations between these two types of sites can provide insights into the potential risk associated with groundwater sources and help in implementing suitable mitigation measures. This study can also shed light on the geology of the region, helping to identify areas with higher radon levels, thereby aiding in informed urban planning and public health initiatives.

2. Materials and methods

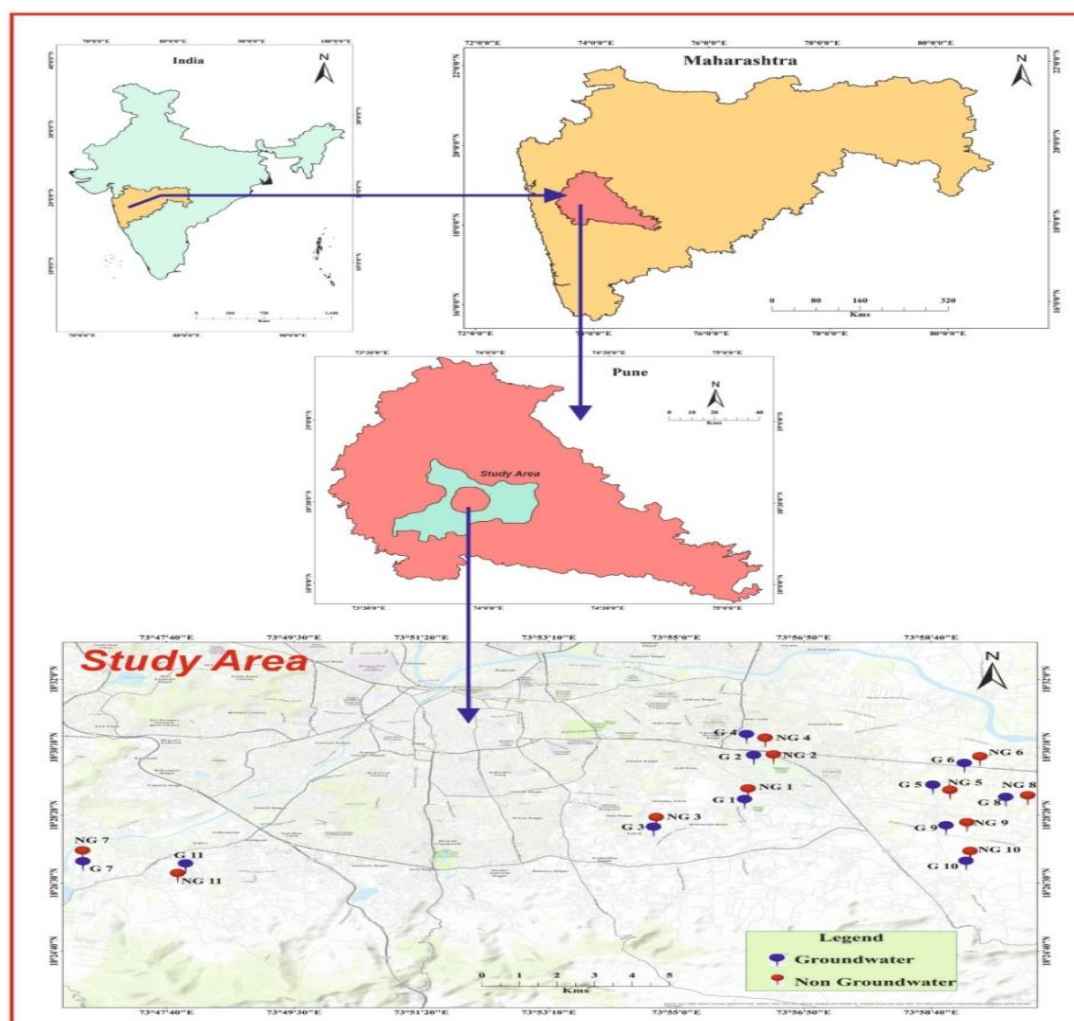
2.1 Study area and identification of GW locations

The geology of the Pune region in Maharashtra, India, is a fascinating blend of ancient geological events and diverse rock formations. This region is part of the Deccan Plateau, and its geological history stretches back millions of years. The dominant geological feature is the Deccan Traps, a massive volcanic plateau formed during the Late Cretaceous period, around 65 to 70 million years ago. These basalts, which can be up to 2,000 meters thick, cover large portions of the region. The Deccan Traps are known for their rugged topography and distinctive basalt formations, featuring volcanic plateaus, rolling hills, and unique geological features (Basavaiah et al., 2018). In addition to the Deccan Traps, the Pune region is also influenced by the Western Ghats, which run along its western boundary. These ancient mountains were formed through tectonic processes and erosion over millions of years, creating a stunning landscape with steep slopes and lush biodiversity. The presence of groundwater within the hard rock terrains of the Deccan Traps is noteworthy. Aquifer horizons exist in vesicular, fractured, and weathered basalts, and the region's geology has led to the formation of natural zeolites containing water molecules within the rock structure (Bhakare, 2010; Querol et al., 2002).

Dharmadhikari's (2019) study delineated the locations of GW veins and NGW areas. Soil samples were collected from these sites, and an HPGe gamma detector were used to measured activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in the soils. The study employed dowsing, an ancient technique known for its 90% accuracy rate, to pinpoint GW veins, using L-rod dowsing specifically. To corroborate GW vein presence, vertical electrical sounding (VES) stations were established at all locations, (Dharmadhikari et al. 2011; 2019). Resistivity measurements utilized a computerized DC resistivity meter SSRMPAT with $\pm 5\%$

allowable tolerance, following the Schlumberger electrode configuration, as outlined by Sharma and Baranwal (2005) and Mahmoud Zadeh et al. (2012).

Figure I : Study Area of Pune Region



2.2 Sample collection, processing, and analysis of samples

A comprehensive scientific study was conducted, involving the collection of a total of 22 surface soil samples from the Pune region. These samples were specifically taken from two distinct locations: 11 samples from areas classified as having GW, and the remaining 11

samples from NGW locations. To ensure accurate analysis, the collected soil samples underwent a standardized procedure, as outlined by the Environmental Measurement Laboratory (EML) in 1983 and the International Atomic Energy Agency (IAEA) in 1989. Following these established guidelines, the samples were carefully processed. Upon completion of the processing stage, the samples were placed in 300 ml polypropylene containers. To allow the radioactive isotope ^{226}Ra to reach equilibrium with its decay products, the containers were sealed and stored for a period of 30 days. Throughout this period, utmost care was taken to prevent the escape of the radioactive gas ^{222}Rn from the containers.

2.2.1 Analysis of samples

The activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in the samples were determined by the gamma spectrometry employing a 48% relative efficiency p-type low background HPGe detector with an energy resolution of 2.0 keV at 1.33 MeV (BEGE-5030, Canberra Industries Inc., USA). The spectrum was acquired and analyzed using a 32K multichannel analyzer (Multiport Lynx MCA, CANBERRA) and GENIE-2000 software. The detector efficiency calibration was performed using the IAEA quality assurance reference materials: RG U, RG Th, RG K, and Soil-6 procured from IAEA. The details of the gamma spectrometer and efficiency calibration were presented in the previous publication (Karunakara et al., 2013; Mohan et al., 2018, 2019; 2019, Sudeep et al., 2023). The samples were counted long enough to reduce the counting/statistical error. The ^{226}Ra activity was evaluated from the weighted mean of the activities of three photo peaks of ^{214}Bi (609.4, 1120.4, and 1764.7 keV). In the case of ^{232}Th , two photo peaks of ^{208}Tl (583.1 and 2614.5 keV) were used. Fig 2 depicts the different gamma line of various energies (photopeaks). The activity of ^{40}K was derived from the 1460.8 keV gamma line of this isotope. The minimum detection levels (MDL) for the above detecting system were 0.3 Bq kg^{-1} , 0.5 Bq kg^{-1} , and 1.3 Bq kg^{-1} , respectively for ^{226}Ra , ^{232}Th , and ^{40}K for a counting time of 60,000 s.

2.3. Estimation of dose rate

2.3.1 Estimation of activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in soil

Activity concentrations of ^{226}Ra , ^{232}Th , and ^{40}K in soil were calculated using the formula (1) (Knoll, 1998). The radionuclide peaks are shown in Fig 2 for GW vein and NGW.

$$\text{Activity (Bq/kg)} = [(g/t_1) - (b/t_2) \pm SD] \times 100/\eta \times 100/a \times 1000/\text{wt} \text{ --- (1)}$$

Where,

g =Gross count (sample +background) in t_1 second

b = Background count of counter in t_2 second

SD = Standard deviation

η =Percent efficiency of HPGe system for a particular energy peak

a =Abundance factor of radionuclide for a particular energy peak

wt =dry weight of soil sample taken for analysis

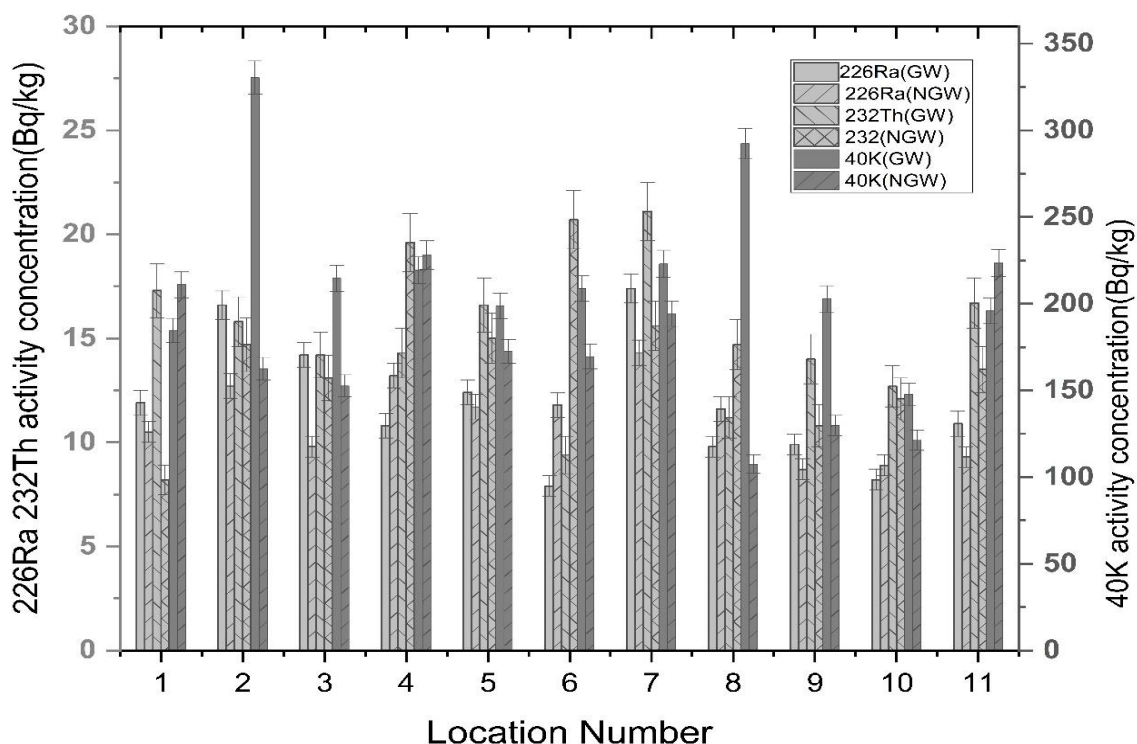
2.3.2 Radium equivalent activity $Ra_{(eq)}$

Radium equivalent activity $Ra_{(eq)}$ is the activity levels of ^{226}Ra , ^{232}Th , and ^{40}K which considers the radiological hazards associated with them. The distribution of radionuclides in the soil is not uniform. Therefore, the consistency of human exposure to radiation from ^{226}Ra , ^{232}Th , and ^{40}K is defined as radium-equivalent activity and it is calculated as follows (Beretka and Mathew,1985; UNSCEAR,2000)

$$Ra_{eq} = R_{Ra} + 1.43 \times R_{Th} + 0.077 \times R_K \text{ --- (2)}$$

where Ra_{eq} (Bq kg^{-1}), R_{Ra} , R_{Th} and R_K are the activity (Bq kg^{-1}) of ^{226}Ra , ^{232}Th and ^{40}K

Figure II: ^{226}Ra , ^{232}Th and ^{40}K activity concentration (Bq/kg) on GW and NGW locations



2.4. Measurement of ^{222}Rn and ^{220}Rn Concentration:

2.4.1. ^{222}Rn Mass Exhalation rate

The active device AQTEK SMART RnDuo is used to assess radon content in soil for current work. The SMART RnDuo is a commercially available portable instrument that detects ^{222}Rn , ^{220}Rn , and alpha rays in the air, ground, and water. The basic principle of the SMART RnDuo instrument is to detect alpha particles using ZnS: Ag scintillation. The acquisition policy and measurement processes are described in further detail elsewhere (Gaware *et al.* 2011). Study materials samples (~1000 g) soil samples has been collected over GW and Non-GW zones for ^{222}Rn , ^{220}Rn concentration. A mortar and pestle or a vibrating ball machine are used to grind the sample into a fine powder. To test radon, a fine powder of an object is placed in the exhalation chamber. The Smart RnDuo is coupled to a ~680gm sample capacity closed accumulation chamber (Figure 3A). The chamber is made up of stainless-steel cylinders with dimensions of 8cm in height and 10 cm in diameter that are connected at the top to a Lucas cell and photomultiplier tube. To avoid ^{222}Rn interruptions, the SMART Duo was used in diffusion mode at ^{222}Rn (if available in samples). After 1 hour, the concentration of ^{222}Rn in the compartment was measured and continued until it was saturated.

The ^{222}Rn concentration data with time elapsed was plotted. Radon growth with time in Bq/m^3 within the accumulator chamber is given

$$C_{(t)} = \frac{J_m M}{V \lambda} (1 - e^{-\lambda t}) + C_0 e^{-\lambda t} \text{ --- (4)}$$

Where $C_{(t)}$ is ^{222}Rn concentration (Bqm^{-3}) at time t Bqm^{-3}

$J_m = ^{222}\text{Rn}$ mass exhalation rate (Bq / Kg / h)

$\lambda = \text{effective decay const.} (h^{-1})$

$C_0 = ^{222}\text{Rn}$ concentration (Bqm^{-3}) present in the chamber volume at $t=0$

$M = \text{mass the dry sample (Kg)}$

$V = \text{Volume of accumulator chamber and scintillation cell volume (volume of detector + porous volume of sample + residual air volume of the mass exhalation chamber) (m}^{-3}\text{)}$

The porous volume (V_p) can be estimated using the following equation.

$$V_p = V_s - \frac{M}{\rho_g} \text{ --- (5)}$$

V_s is the sample volume in the mass exhalation chamber, ρ_g is the specific gravity of the sample which can be taken as 2.7 gm/cc for a clay-type soil sample.

^{222}Rn mass exhalation rate can be calculated by the formula:

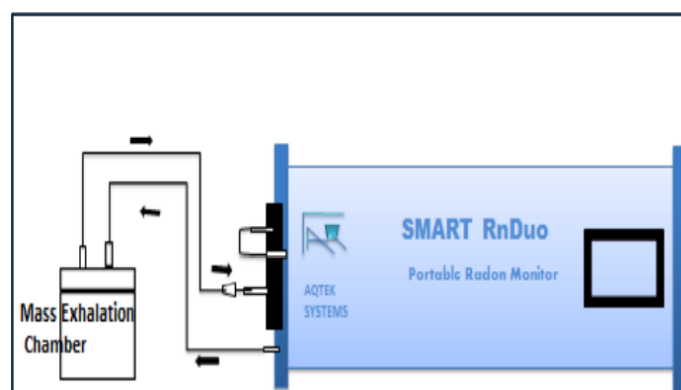
$$J_m = \frac{BV}{M} \text{ --- (6)}$$

Where $J_m = \text{Radon } (^{222}\text{Rn}) \text{ mass exhalation rate}$

$B = \text{Slop of the linear fit graph of radon concentration (Bqm}^{-3}\text{) Vs Time (hr)}$

$V = \text{Effective volume, } M = \text{Total mass of the dry sample.}$

Figure III. A. Set-up for measurement of the radon mass exhalation rate Using Smart RnDuo



2.4.2. ^{220}Rn surface exhalation rate

In case of ^{220}Rn monitoring by SMART RnDuo, a program-based sampling is carried out using a flow-mode sample connected to a pump inlet of the monitor. Flow mode is sampling type for the measurement of the ^{220}Rn surface exhalation rate. As the half-life of ^{220}Rn is 55 sec., so ^{220}Rn decay before reaching the scintillation detector from soil chamber. To get all ^{220}Rn into scintillation detector, pump is used with flow mode in this case. The exhalation chamber is connected to scintillation ^{220}Rn monitor through the airflow pump forming a close circuit loop as shown in figure 3B. The measurement cycle is about 15 min. In a 15 min cycle, sampling pump is kept on for an initial 5 min, which gives a measure of ^{220}Rn and background, followed by decay of 5 min, which ensure near-complete decay of ^{220}Rn and then, the last 5 min counting gives the measure of background counts for that cycle. At equilibrium, the ^{220}Rn surface exhalation rate is given by the equation.

$$J_s = \frac{C_{eq} V \lambda}{A} \text{-----(4)}$$

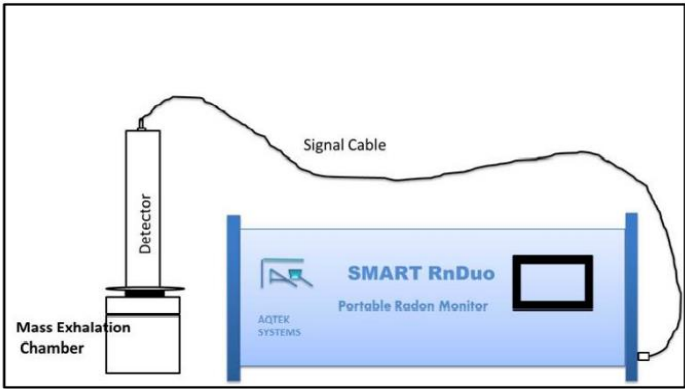
Where C_{eq} is equilibrium ^{220}Rn concentration (Bqm^{-3})

λ is the ^{220}Rn decay constant (0.0126s^{-1})

V is the sum of residual volume of mass exhalation chamber and the internal volume of RnDuo detector and tubing volumes (m^3)

A is the surface area (m^2) of the sample emitting ^{220}Rn .

Figure III. B. Set-up for measurement of the thoron surface in soil Using Smart RnDuo



3.0. Result and Discussion

Figure 2 presents the activity concentration results of radionuclides ²²⁶Ra, ²³²Th, ⁴⁰K, and Ra_(eq) in soil samples. The average global concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K are 32 Bqkg⁻¹, 45 Bqkg⁻¹, and 412 Bqkg⁻¹, respectively.

The overall activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K vary in the ranges of 7.9-17.4 Bqkg⁻¹ (mean: 11.8 Bqkg⁻¹), 9.4-21.1 Bqkg⁻¹ (mean: 14.8 Bqkg⁻¹), and 147.8-330.4 Bqkg⁻¹ (mean: 229.8 Bqkg⁻¹), respectively over the GW region.

Similarly, the overall activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K vary in the ranges of 8.7-14.3 Bqkg⁻¹ (mean: 11.1 Bqkg⁻¹), 8.2-20.7 Bqkg⁻¹ (mean: 14.3 Bqkg⁻¹), and 107.5-228.1 Bqkg⁻¹ (mean: 170.2 Bqkg⁻¹), respectively.

Table 1: Analytical result of the concentration, mass exhalation and Surface exhalation of ²²²Rn, and ²²⁰Rn of soil samples

Sample	²²² Rn (Radon)		²²⁰ Rn (Thoron)		Ra(eq) Activity (Bq kg ⁻¹)
	²²² Rn Conc. (Bq/m ³)	Mass Exhalation	²²⁰ Rn Conc. (Bq/m ³)	Surface exhalation(mBq/kg/sec)	

Code			JM(mBq/kg/hr)							
	GW	NGW	GW	NGW	GW	NGW	GW	NGW	GW	NGW
S1	526.9±24.2	276.8±22.5	13.4	4.68	920.5±165.0	755±138.2	37.2±6.6	30.5±5.5	50.8±3.0	23.7±1.5
S2	543.7±25.2	304.5±21.3	18.6	9.19	1382.1±192.8	1178.3±174.7	55.9±7.8	47.7±7	64.6±3.1	34.8±2.5
S3	533.0±22.5	227.4±19.7	16.5	6.78	1097.5±161.2	962.6±153.5	44.4±6.5	38.9±6.2	51.0±2.7	29.6±2.1
S4	460.7±25.7	225.7±24.6	11.7	7.29	1493.3±187.0	1165.8±162.3	60.4±7.5	47.2±6.5	48.1±2.9	42.8±2.6
S5	529.7±24.5	269.4±24	14.2	7.91	1085.5±161.2	920.8±153.5	43.9±6.5	37.2±6.2	51.4±3.0	34.3±2.3
S6	368.0±22.5	200.5±22.1	9.0	3.27	1768.3±192.8	1291±174.7	71.6±7.8	52.2±7.0	37.4±2.3	42.5±2.6
S7	720.3±26.0	372.4±24.8	20.1	8.58	3381±146.0	861±123	136.9±5.9	34.8±4.9	64.7±3.2	37.9±2.3
S8	413.9±22.2	208.2±22.1	11.2	5.18	1039.8±158.2	657.1±133.5	42.1±6.4	26.6±5.4	48.3±2.6	33.3±2.3
S9	423.1±23.2	212.2±20.1	11.5	5.23	1057.6±207.2	643.3±131	42.8±8.3	26.0±5.3	45.5±2.8	25.0±1.9
S10	401.3±24.2	250.0±22.1	10.9	4.5	1492.8±207.2	653.3±137.2	60.4±8.3	26.4±5.5	37.7±2.4	27.0±1.9
S11	522.5±24.1	254.5±23.9	13.8	8.36	1412.3±173.2	989.3±158.5	57.1±7.0	40.0±6.4	49.8±2.8	30.1±2.1
Mean	494.8±24.1	254.7±22.5	13.7	6.4	1466.5±177.4	916.1±149	59.3±7.1	37.1±6	49.9±2.8	32.8±2.2
Range	368.0±22.2- 720.3±26.0	200.5±19.7- 372.4 ±24.8	9.0- 20.1	3.27- 9.19	920±146- 3381±207.2	643.3±123- 1291±174.7	37.2±5.9- 136.9±8.3	26.0±4.9- 52.2±7	37.4-64.7	23.7- 42.8
Median	522.5±24.2	250±22.1	13.4	6.78	1382±173.2	920.8±139	55.9±7.0	37.2±6.2	49.85±2.8	33.3±2.3

Table 1 presents the radon concentration results in soil samples. The overall radon concentrations of ^{222}Rn and ^{220}Rn vary in the ranges of 368.0±22.2-720.3±26 Bq/m³ (mean: 494.8±24.1 Bq/m³), 920±146-3381±207.2 Bq/m³ (mean: 1466.5±177.4 Bq/m³), respectively. The mass and surface exhalation of ^{222}Rn and ^{220}Rn ranges from 9.0-20.1 mBq/kg/hr with a mean value of 13.7 mBq/kg/hr and 37.2-136.9 mBq/kg/sec with mean value 59.3 mBq/kg/sec respectively over the GW region. The activity concentration of $\text{Ra}_{(\text{eq})}$ ranges from 37.4 to 64.7 Bqkg⁻¹ with a mean value of 49.97 Bqkg⁻¹ over the GW region.

The overall radon concentrations of ^{222}Rn and ^{220}Rn vary in the ranges of 200.5±19.7-372.4 ±24.8 Bq/m³ (mean: 254.7±22.5Bq/m³), 643.3±123-1291±174.7Bq/m³ (mean: 916.1±149 Bq/m³), respectively. The mass and surface exhalation of ^{222}Rn and ^{220}Rn ranges from 3.27-9.19mBq/kg/hr with a mean value of 6.4 mBq/kg/hr and 26.0-52mBq/kg/sec with mean value 37.1 mBq/kg/sec respectively over the NGW region. The activity concentration of $\text{Ra}_{(\text{eq})}$ ranges from 23.7 to 42.8 Bqkg⁻¹ with a mean value of 32.8 Bqkg⁻¹ over the NGW region.

The results obtained for the radon concentration, mass and surface exhalation rate in the present study are within the range of the worldwide average value (Table 3) and remain higher for GW than NGW.

4.0. Conclusion

The study presented in figure1 provides important information about the activity concentration of radionuclides ^{226}Ra , ^{232}Th , and ^{40}K in soil samples from two regions, GW and NGW. The activity concentrations of these radionuclides vary within specific ranges for both regions, with mean values falling close to the average global concentrations reported by the OECD.

It is noteworthy that the results for radon concentration, mass, surface exhalation rates and $\text{Ra}(\text{eq})$ activity concentration fall within the range of worldwide average values. With consistently higher values observed in the GW region compared to the NGW region. These findings emphasize the importance of monitoring and understanding the spatial distribution of radionuclide and radon concentrations, as they have implications for environmental and health assessments in these regions.

Overall, the study suggests that the radionuclide activity concentrations in the studied regions are within safe limits and do not pose significant risks to public health. However, continued monitoring of radionuclide levels in soil and the environment remains crucial to ensure the safety of the population and prevent potential stochastic effects associated with high radium equilibrium index and radon concentration.

Acknowledgement:

The authors thankful to” Mahatma Jyotiba Phule Research and Training Institute (MAHAJYOTI), Nagpur” for their financial assistance

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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