Assessment of physicochemical and radon-attributable radiological parameters of drinking water samples of Pithoragarh district, Uttarakhand

Prakhar Singh  
Uttarakhand Science Education and Research Centre

OP Nautiyal  
Uttarakhand Science Education and Research Centre

Manish Joshi  
Bhabha Atomic Research Centre

Ankur Kumar  
Gurukula Kangri University: Gurukula Kangri vishwavidyalaya  https://orcid.org/0000-0001-8628-5702

Taufiq Ahamad  
Uttarakhand Science Education and Research Centre

Kuldeep Singh  
Government Post Graduate College New Tehri

Research Article

Keywords: Annual effective dose, Groundwater, Radon activity, Spatial distribution, TDS

DOI: https://doi.org/10.21203/rs.3.rs-499817/v1

License: This work is licensed under a Creative Commons Attribution 4.0 International License. 
Read Full License
Assessment of physicochemical and radon-attributable radiological parameters of drinking water samples of Pithoragarh district, Uttarakhand

Prakhar Singh¹, O. P. Nautiyal¹, Manish Joshi², Ankur Kumar*³, Taufiq Ahamad⁴, Kuldeep Singh⁵

¹Uttarakhand Science Education and Research Centre, Dehradun - 248006, India
²Radiological Physics and Advisory Division, Bhabha Atomic Research Centre, Mumbai-400094, India
³Department of Physics, Gurukula Kangri Vishwavidyalaya Haridwar – 249404, India
⁴Department of Physics, Shri Guru Ram Rai PG College, Dehradun-248001, India
⁵Department of Physics, Govt. PG College, Tehri Garhwal, New Tehri-249001, India

*E-mail address of the corresponding author: physicsankur@gmail.com

Abstract

This study evaluates the quality of drinking water samples (sample size=52) taken from various locations of Pithoragarh district, Uttarakhand. The parameters include physiochemical properties viz. total dissolved solids (TDS in mg/L), electrical conductivity/salinity (µS/cm), pH and radiological dose attributable to radon in water (µSv/y). TDS values for the tested samples varied within the range of 18-434 mg/L with average value of 148 mg/L. Electrical conductivity and pH for these samples was measured as 36-868 µS/cm (average: 296 µS/cm) and 6.8-8.2 (average: 7.2), respectively. Radon activity concentration for these water samples was measured using scintillation-based radon monitor, immediately after sampling at the location site. Radon activity concentration was measured as 0.6-81.9 Bq/L with an average value of 17.8 Bq/L. The
paper also estimates the annual effective ingestion dose ($\mu$Sv/y), annual effective inhalation dose ($\mu$Sv/y) and total effective dose ($\mu$Sv/y) attributable to radon in drinking water samples. Spatial patterns for the observed variations have also been interpreted for the dataset obtained over the terrestrial region.

**Keywords**

Annual effective dose; Groundwater; Radon activity; Spatial distribution; TDS.

**Introduction**

Water quality is one of the important parameters determining the health risks to humans. Measurement of various characteristics of water, associated interpretations and hazard estimations have been performed worldwide at different places of relevance. It is a fact that numerous contaminants pollute water sources and this impacts human health risks. The source of contaminants could be natural or anthropogenic and the level of degradation of water quality depends on their source-term. Radioactive contamination is also a serious issue around the globe. While the mineralogy acts as the source term for physicochemical parameters, natural radio-nuclides present in the earth crust acts as the main source for radiological impurity of ground and surface water [1]. Total dissolved solids (TDS), salinity/electrical conductivity and pH are three vital physicochemical parameters which vary for different water sources and also get affected due to human activities. Similar to the air matrix, radon/thoron gases and its decay products primarily released from the natural radio-active precursors (decay chains) determines the radioactivity concentration profile of water. $^{222}$Rn and $^{220}$Rn are two isotopes of radon with different half-life ($^{222}$Rn has a half-life of 3.82 days and $^{220}$Rn has a half-life of 55.5 seconds) [2]. Due to its gaseous nature, it travels through cracks and pores between the grain in the soil and easily escape into the ground and surface water. Previous studies in other parts of Uttarakhand Himalaya indicate the presence of a significant amount of natural radioactivity in the air, soil, and water [6-12]. Pithoragarh is a high-altitude terrestrial region and groundwater is the main source of water to be used for domestic as well as industrial purposes. Most of the population in the region depends on natural spring sources for drinking water [13]. $^{222}$Rn emits alpha particles and its short-lived progenies ($^{218}$Po, $^{214}$Po, $^{214}$Pb and $^{214}$Bi) interact with atmospheric particles. Previous studies in Kumaun Himalayas reported radiological dose levels due to $^{222}$Rn activity concentration. [15-18]. Such measurements, their usage in the estimations of radiological doses
and the interpretations are highlighted as an important data-base by standard international commissions. Therefore, measurement of radioactivity concentration and the estimation of radiological doses due to the ingestion of Rn\(^{222}\) has been done by several researchers in past as well [19-21].

This study focuses on the measurement of physicochemical properties and Rn\(^{222}\) concentration in drinking water samples of Pithoragarh district, Uttarakhand, India. Rn\(^{222}\) concentration was measured using a portable Rn\(^{222}\) monitor on the spot of sampling right after the sample is collected. The physicochemical properties (TDS, Electrical Conductivity, and pH) were measured in the laboratory. Statistical analysis of the obtained data is discussed in detail in this work. QGIS (Quantum GIS) 3.4 Madeira LTS (Long Term Support) is used for the spatial or geological analysis of the data over the study region.

**Geology of Study Area**

Pithoragarh is the easternmost district of Uttarakhand state of India. It confines higher Himalayan mountain ranges, passes, valleys, alpine meadows, forests, waterfalls, rivers, glaciers, springs, and snow-capped peaks. The geographical area of Pithoragarh is 7,110 square Km. The district is located in the Kumaun division of Uttarakhand. The northern part of the district is attached to the Tibetan plateau and Nepal is on its eastern border that is separated by the Kali river also known as Sharda river. The elevation from sea level ranges between 500 to 6400 m. Variation of temperature is considerable from area to area depending upon the altitude. The lower Himalaya is confined by the Main Boundary Thrust (MBT) in the southern part and Main Central Thrust in the Northern part. It consists of the late Proterozoic to early Cambrian sediments intruded by some granites and volcanic rocks [22]. The rock types in this land are quartzites, siltstone, carbonates, Phyllite, schist with subordinate impure marbles, metamorphic rocks, and orthogneisses [23]. Almost 50 % of the area is occupied by snowy Himalayan mountain ranges in the north forming several glaciers in the region. Therefore, at lower reaches, numerous natural springs are available for domestic and irrigation purposes. The district receives a good amount of rainfall, which infiltrates into the ground through the soil matrix and recharges the groundwater level. During the post-monsoon period, the water level reduces gradually affecting running water inventory for the pre-monsoon phase. The radon exhalation and water discharge rate are largely influenced by the meteorological parameters like pressure,
temperature, relative humidity etc. Radon level in water was found to be higher in monsoon and post-monsoon season in comparison to pre-monsoon season in a study conducted in Garhwal region [27]. The concentration of radon is linked to water quality in terms of radiological considerations. This study was performed for the post-monsoon season only and the results are interpreted in terms of water quality characterization using the results obtained from the measurements made in this season. Whereas physicochemical properties represent the parameters mainly for post-monsoon season, annual inferences are also made for radiological dose estimations. As the radon concentration values remains in mid-level (maximum in pre-monsoon and minimum in monsoon period) range, these can be assumed as representing yearly averaged values for calculations.

Figure 1: Sampling locations on the map of the study area (Pithoragarh district of Uttarakhand state of India).

Experimental Techniques and Methodology

Sample collection
52 water samples of drinking water were collected from different villages of the study area from various sources like natural springs (49) and hand pumps (3) in the month of December. Radon concentration profile follows a seasonal pattern due to the change in the water level as the function of rainfall. Radon concentration is expected to be at maximum level in the summer season while it reduces in monsoon period due to the increase in water level. The water samples that have been taken from the locations represent water which is used by the local population for domestic purpose as well as for drinking. Water samples were collected in polypropylene double capped sampling bottles. The bottles were washed thoroughly prior to sampling to reduce external contamination. Before pouring the sample into the bottles, a container is used to collect the water. The polypropylene sample bottle was then immersed into the container. One end of a polypropylene pipe is immersed into the opening of water source. After ensuring that no air bubble is forming inside the pipe, the other end of the pipe is purged into the sampling bottle, replacing the water in the sample bottle. To collect water sample from the hand-pump, the pipe was plunged inside the pumping cylinder of hand-pump up to the water level and hand-pump lever was operated to pump out water. After ensuring that there are not any bubbles forming inside the pipe, the other end of pipe then plunged inside the sample bottle that was immersed inside the water in large container. The sample bottle was capped inside the container to prevent the atmospheric contaminants. This sampling methodology was used to reduce the chance of instant degassing of radon while pumping the water out from the handpump, and to reduce external contamination from the atmosphere. After sampling, these water samples were analyzed for Rn\textsuperscript{222} on the spot with a portable Rn\textsuperscript{222} monitor. After the Rn\textsuperscript{222} measurement, water samples were taken to the laboratory for physiochemical analysis.

**Measurement of Rn\textsuperscript{222} concentration**

Water samples were analysed for Rn\textsuperscript{222} concentration with Smart RnDuo using a bubbler kit. Smart RnDuo is an online active Rn\textsuperscript{222} monitor that is developed by Bhabha Atomic Research centre, Mumbai. The working of Smart RnDuo is based on the alpha scintillation method. An Ag:ZnS coated cell (Lucas cell) is connected to a photomultiplier tube that records scintillation counts when an alpha particle hits the wall of the lucas cell [28]. The basic set-up for the measurement of radon concentration in water involves 50 ml glass sample bottle attached to a bubbler. The bubbler is connected to the radon monitor using polypropylene pipes and a particle filter. The air pump is used to circulate the air within the sample through the Lucas cell to obtain
escaping Rn$^{222}$ from the sample inside the Lucas cell. For 15 minutes of the measurement cycle, a pump is set to on for 5 minutes and for the rest 10 minutes, the alpha count continues. After completing one cycle, average Rn$^{222}$ activity concentration is displayed on the monitor. Figure 2 shows the schematic diagram for the setup for the measurement of Rn$^{222}$ activity concentration using the bubbler method [29]. This instrument gives Rn$^{222}$ concentration in the air volume having unit Bq/m$^3$. To calculate Rn$^{222}$-activity concentration in water volume (liquid) one can use the following equation 1 for the setup [30].

$$C_{\text{liq}} = C_{\text{air}} \left[ K + \frac{V_{\text{air}}}{V_{\text{liq}}} \right] \quad (1)$$

Where $C_{\text{air}}$ is the Rn$^{222}$ activity concentration of air given by the Rn$^{222}$ monitor, $K$ is the partition coefficient (i.e. $K=0.25$ for water and air), $V_{\text{air}}$ and $V_{\text{liq}}$ is the volume of air present in the setup and volume of liquid sample present in the sampling bottle, respectively.

**Figure 2**: Schematic diagram of measurement of Rn$^{222}$ activity concentration using bubbler kit attached to portable Smart RnDuo active radon monitor.

**Measurement of Physiochemical parameters**

The measured physiological parameters for the water samples i.e., Total Dissolved Solids (TDS), pH, and electrical conductivity due to salinity were measured using standard analytical methods available at Nuclear Research Laboratory, USERC, Dehradun. The Oakton CON 550 Benchtop Conductivity/TDS meter kit was used for the measurement of TDS and Electrical Conductivity of the water samples. The instrument measure electrical conductivity/TDS though a conducting probe, and has conductivity measuring range of 0-200 mS/cm with an accuracy of ±1.0% full-scale ±1 digit, and TDS measuring range of 0-100g/L with a resolution of 0.1/1mg/L. TDS is the
measurement of the total weight of solids dissolved in the water sample volume that is expressed in parts per million (mg/L) or milligrams per unit volume. pH shows whether the sample is alkaline or acidic. It is a negative logarithm of hydrogen ion concentration. For pH measurement, GeNai pH digital meter – 117800GB is used in the laboratory. It has pH range of 1-14 with 0.5±1 digit accuracy pH. [31-33].

**Estimation of radiological dose**

The annual effective dose due to ingestion and inhalation of Rn$^{222}$ has been calculated using the parameters established in the reference [34,35].

The annual effective dose via ingestion can be calculated using the following equation2:

$$E_{w,lg} (\mu Sv, y^{-1}) = C_{Rn} \times C_w \times (EDC)$$  \hspace{1cm} (2)

Where, $E_{w,lg}$ is the annual effective ingestion dose, $C_{Rn}$ is the Rn$^{222}$ activity concentration in water (Bq/L), $C_w$ is the estimation of water consumption for the average population (600 L/y) and $EDC$ is the ingestion effective dose conversion coefficient i.e. 3.5 nSv/Bq [36]. Annual effective inhalation dose can be calculated by the equation 3:

$$E_{w,ih} (\mu Sv, y^{-1}) = C_{Rn} \times R_{a,w} \times F \times O \times (DCF)$$  \hspace{1cm} (3)

Where, $E_{w,ih}$ is the annual effective inhalation dose, $C_{Rn}$ is the Rn$^{222}$ activity concentration in water (Bq/L), $R_{a,w}$ is the ratio of Rn$^{222}$ in air and tap water ($10^{-4}$)[36], $F$ is the equilibrium factor between Rn$^{222}$ and its progenies (0.4)[37], $O$ is average annual indoor occupancy time per person (7000 h/y) and $DCF$ is the dose conversion factor due to Rn$^{222}$ exposure i.e. 9 nSv/(h.(Bq/m$^3$))[36].

Age-dependent annual effective ingestion dose was calculated using equation 4 as follows [36,38]:

$$AED_{ing} (\mu Sv, y^{-1}) = A \times w_{ing} \times DCF_{ing}$$  \hspace{1cm} (4)
Where, $AED_{ing}$ is the age-dependent annual effective ingestion dose($\mu$Sv/y), $A$ is the activity concentration of Rn$^{222}$ in water (Bq/L), $w_{ing}$ is average annual water intake by the local population (L.y$^{-1}$) and $DCF_{ing}$ is the age-dependent dose conversion factor (nSv/Bq) [39,40].

**Result and discussion**

The statistical parameters (i.e. minima, maxima, mean, standard deviation, skewness, and kurtosis) calculated for Rn$^{222}$ activity concentration (Bq/L), TDS (mg/L), pH, Electrical conductivity ($\mu$S/cm), age-dependent annual effective ingestion dose ($\mu$Sv/y), Annual effective ingestion dose ($\mu$Sv/y), Annual effective inhalation dose ($\mu$Sv/y) and Total annual effective dose ($\mu$Sv/y) in Table 1.

**Radiological exposure due to Rn$^{222}$ activity concentration in water**

The Rn$^{222}$ activity concentrations in the water samples are found to be in the range of 0.6 to 81.9 Bq/L with a mean value of 17.8 Bq/L. These values are within the safe limit prescribed by WHO (2008) and EU (2001a), that is 100 Bq/L [41-42].

The frequency plot regarding the data obtained for Rn$^{222}$ activity concentration in collected water samples is shown in Figure 3(a). The frequency plot indicates that more than half (54%) of samples have Rn$^{222}$ activity level below 10 Bq/L and in 56% samples radon level is below 11.1 Bq/L (recommended level by USEPA (1991)) [43]. All of samples have Rn$^{222}$ levels within the safety limit recommended by WHO (2008). To obtain the best fit distribution among Lognormal, Exponential, Gamma and Weibull, K-S (modified) test and A-D test has been performed on radon dataset, and goodness-of-fit statistical parameters are shown in Table 2. Results shown in table 2 and fig. 3 suggest that except exponential distribution, all other distributions (Lognormal, Gamma and Weibull) captured the trend of the variation of the data. [44, 45]. Figure 3(b) demonstrate the probability plot generated for the radon dataset. Lognormal, Gamma and Weibull dataset lie over the fitted reference line, indicating good fit for the data. However, in the Exponential probability plot, data points outreach the reference fitted line which indicates rejection of Exponential distribution over the radon dataset.
Figure 3: (a) Frequency plot and (b) probability plot of $^{222}\text{Rn}$ activity concentration in water samples taken from Pithoragarh, district of Uttarakhand.
Physicochemical parameters

Total dissolved solids (mg/L) values of the samples occur in the range between 18 and 434 mg/L with an average value of 148 mg/L. From frequency plot (Fig 4a), it can be seen that; the TDS concentration was below 50 mg/L in 37% samples and in 97% samples its value was below 300 mg/L. Also shown in Table 1, this dataset is not uniform with -0.2 kurtosis and lightly-tailed or non-uniformly random. Goodness-of-fit statistics using K-S (modified) and A-D test has been performed over TDS dataset, and calculated parameters are shown in the Table 2. The goodness-of-fit statistics suggest that Lognormal, Gamma and Exponential distribution are not followed by the TDS dataset. However, Weibull distribution is accepted as best fit among assumed Distributions.

Figure 4(b) demonstrate the probability plot of the TDS dataset is rejected for Lognormal, Gamma and Exponential distribution. However, it is observed that the K-S (modified) test signifies that data-points are within the reference fitted line which confirms that Weibull distribution is best fit for the dataset (Table 1), meanwhile Lognormal, Gamma and Exponential distributions are rejected for the TDS dataset.

The pH values of collected samples range from 6.8 to 8.2 with a mean value of 7.2. Thus, the samples are slightly alkaline but safe for drinking and other domestic purposes. Skewness and kurtosis values show asymmetry of the dataset distribution which indicates a lack of external parameters to affect the pH of the water samples. Electrical conductivity varies from 36 µS/cm to 868 µS/cm, with a mean value of 296 µS/cm.
Figure 4: (a) Frequency plot and (b) probability plot of TDS present in the water samples taken from Pithoragarh, district of Uttarakhand.
Radiological Dose assessment due to Rn$^{222}$ exposure

The age-dependent annual effective ingestion dose (AEI$_g$D) estimated due to Rn$^{222}$ exposure to the local population is shown in Table 1. For infants (age below 1 year), the annual ingestion dose ($\mu$Sv/y) varies from 2.7 to 376.6 $\mu$Sv/y with an average value of 81.7 $\mu$Sv/y. These values estimated for the children aged between 1 and 3 years vary within the range of 0.9-125 $\mu$Sv/y with an average value of 27.2 $\mu$Sv/y. For children aged 4 to 8 years, the value of AEI$_g$D ranges between 1.0 and 144.9 $\mu$Sv/y with an average value of 31.4 $\mu$Sv/y. For toddlers (male) aged 9-13 years, the average value of AEI$_g$D is 21.8 $\mu$Sv/y within the range of 0.7-100.3 $\mu$Sv/y. For teenagers (male) who come in the age group of 14 to 18 years, the range of AEI$_g$D occurs between 1.2 and 171.9 $\mu$Sv/y with the mean value of 37.3 $\mu$Sv/y. For the adults, the AEI$_g$D ranges from 1.5 to 209 $\mu$Sv/y with an average value of 45.4 $\mu$Sv/y. For Toddlers (female) aged between 9 to 13, the AEI$_g$D values obtained within the range of 0.7-100.3 $\mu$Sv/y with an average value of 21.8 $\mu$Sv/y. The AEI$_g$D value for teenagers (female) ranges between 1.2 and 171.9 $\mu$Sv/y with the mean value 37.3 $\mu$Sv/y. For Adult females, the obtained values vary within the range of 1.5-209.2 $\mu$Sv/y with an average value of 45.4 $\mu$Sv/y. The AEI$_g$D values for infants and children are higher than for teenagers and adults because of the high value of dose conversion factors for infants and children. High dose conversion factors for infants and children indicate that this age group is at relatively higher risk due to similar exposure of Rn$^{222}$ compared to other age groups. For the study area, these dose values are well within the safe limits prescribed by various agencies [41]. Therefore, water samplers analyzed for this region are safe for drinking for all age groups. Figure 5 shows the violin plot for annual effective ingestion dose for the various age groups. The violin-like graph shows the kernel densities of the dataset of age-dependent annual ingestion dose. The white dots between the densities are the median value. It can be seen that for infants the median as well as the span of the dataset is highest.
The annual effective Ingestion dose (µSv/y) due to Rn$^{222}$ exposure through drinking water, annual effective inhalation dose (µSv/y) due to inhalation of Rn$^{222}$ escapes from the water and total annual effective dose (µSv/y) are also represented in Table 2. The annual effective ingestion dose ranges between 1.5 and 209.2 µSv/y with the mean value 45.4 µSv/y. The annual effective inhalation dose occurs in the investigation within the range of 1.8-258.2 µSv/y and the average value is 56.0 µSv/y. The total annual effective dose due to Rn$^{222}$ exposure ranges between 3.3 and 467.4 µSv/y with a median value of 44.5 µSv/y. Figure 6 illustrates the violin plot of annual effective ingestion dose (µSv/y), annual effective inhalation dose (µSv/y) and total annual effective dose (µSv/y), respectively. These values are under the recommended limits (WHO, 2004) for drinking water. The white dots in between quartiles indicate the mean value of the data and the red line demonstrates the median value. The mean value is in the second quartile for all the doses which usually happens when most of the data occur at low values. The total dose increases when the Rn$^{222}$ concentration increases. It is also demonstrated in the study that although children drink less water than adults yet they receive more doses due to high dose.
conversion factor proposed for infants. Similar conclusions are also made by other studies in past [35,39]. No significant difference is found between AED for males and females.

Figure 6: Violin plot for annual inhalation dose, annual ingestion dose and total annual effective dose.

Spatial Analysis

This study is a field survey; hence, it is imperative to analyze the dataset over terrestrial land, with available data points. QGIS 3.4 LTS software is used for spatial analysis of Rn$$^{222}$$ activity concentration (Bq/L) and TDS (mg/L). It is to be noted that the sampling of water in the study was carried out on point locations of the map. The latitude and longitude were also taken by GARMIN-GPS during the measurement of the location of sampling. The pointwise data is not enough to illustrate areal distribution. Therefore, the inverse distance weighting (IDW) interpolation method is used to calculate the approximate values on the neighbouring locations where the data was not taken during fieldwork. It is an adaptive interpolation method that distributes the data values over a spatial area. The simplest weighting function that is used to
interpolate data over the area is the inverse of the distance between two points with measured
data on the location of sampling. This is a good method when need to apply for fewer location
points otherwise kriging should be used for interpolation [46]. However, the accuracy of the
predicted values for regions other than the sample nodes increases with the increase in sample
size. The study represents the spatial distribution based on chosen data nodes only for indicative
purposes.

Figure 7 illustrate spatial distribution of TDS (mg/L) and Rn$^{222}$ activity concentration (Bq/L). It
is observed that the most of the samples show low values over the region for TDS (mg/L) as well
as Rn$^{222}$ activity concentration (Bq/L). The water samples taken from the location of Usail,
Satgad, Bisabajer and Jajardeva show comparatively higher values of TDS than other locations
of the study area. The samples taken from the southern region have a TDS value of more than
160 mg/L. Figure 7(b) demonstrates the predicted Rn$^{222}$ activity concentration values in water
samples for the study region. It is observed that the northern area shows high values of
Rn$^{222}$ activity concentration in water samples although TDS is comparatively lower than 200
mg/L in the water samples taken from those nearby locations. The northern area of Pithoragarh is
high altitude Terrain. More studies are needed to investigate higher Rn$^{222}$ concentration values
observed for high altitude regions. The observance of higher Rn$^{222}$ activity concentration in low
altitude regions (e.g. sampling node close to Dharchula) indicate the possibility of transportation
of minerals by natural spring water, aquifers and rivers.
Conclusion

In this study, 52 samples of drinking water were collected from Pithoragarh district of Uttarakhand, India. The samples were investigated for Rn$^{222}$ activity concentration (Bq/L) level, TDS concentration (mg/L), Electrical Conductivity ($\mu$S/cm) and pH. Age-dependent radiological doses to local population due to radon ingestion and inhalation due to water intake were also calculated and are compared with reference parameters recommended by ICRP (2002). All of the water samples have lower Rn$^{222}$ activity concentration level than the reference limit recommended by European Commission (2001) and WHO (2008). The Rn$^{222}$ activity concentration dataset follows Lognormal, Weibull and Gamma frequency distribution when tested with K-S (modified) and A-D tests for goodness-of-fit. Most of the samples have TDS below 250mg/L which indicates that the samples are of good quality (mineral-wise). For the case of frequency distribution of TDS, Weibull distribution was found to be best fitted as per the tests for goodness-of-fit. On average, the tested water samples were found to be slightly alkaline. Higher age-dependent annual effective ingestion dose values for the infants reflect the impact of higher dose conversion factors. No significant difference was found between annual effective ingestion doses for males and females. The estimated dose values were found to be lower than the recommended levels, wherever applicable. The spatial distribution analysis demonstrated that the water samples from the southern region have higher TDS (mg/L) values and the Northern region have higher Rn$^{222}$ activity concentration (Bq/L) levels. This study concludes that the springs and handpumps are the source of safe drinking water in Pithoragarh district of Uttarakhand, India.

Acknowledgment

The authors acknowledge the Board of Research in Nuclear Science (BRNS), Department of Atomic Energy (DAE) Project Ref. 36(4)/14/2016-BRNS, Mumbai, India, for extending their laboratory facilities for conducting the experimental work and funding for this research work.

Declaration of Conflict of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
Table 1: Statistical parameters of the measurements.

| Statistical Parameters | Radon Activity Concentration (Bq/L) | TDS (mg/L) | EC (µS/cm) | pH | Age-dependent Annual Effective Ingestion Dose (µSv/y) | Annual Effective Inhalation Dose (µSv/y) | Annual Effective Ingestion Dose (µSv/y) | Total Annual Effective Dose (µSv/y) |
|------------------------|-------------------------------------|------------|------------|----|----------------------------------------------------|------------------------------------------|------------------------------------------|--------------------------------------|
| **Minima**             | 0.6                                 | 18.0       | 36.0       | 6.8| 2.7 0.9 1.0 0.7 1.2 1.5                           | 0.7 1.2 1.5                              | 1.8 1.5 3.3                             |                                      |
| **Maxima**             | 81.9                                | 434.0      | 868.0      | 8.2| 376.6 125.6 144.9 100.3 171.9 209.2             | 100.3 171.9 209.2                        | 258.2 209.2 467.4                       |                                      |
| **Mean**               | 17.8                                | 148.0      | 296.0      | 7.2| 81.7 27.2 31.4 21.8 37.3 45.4                   | 21.8 37.3 45.4                           | 56.0 45.4 101.4                         |                                      |
| **Standard Deviation** | 21.4                                | 107.0      | 214.0      | 0.3| 98.4 32.8 37.9 26.2 44.9 54.7                   | 26.2 44.9 54.7                           | 67.5 54.7 122.1                         |                                      |
| **Skewness**           | 1.5                                 | 0.5        | 0.5        | 0.2| 1.5 1.5 1.5 1.5 1.5 1.5                        | 1.5 1.5 1.5                             | 1.5 1.5 1.5                            |                                      |
| **Kurtosis**           | 1.7                                 | -0.2       | -0.2       | -1.1| 1.7 1.7 1.7 1.7 1.7 1.7                       | 1.7 1.7 1.7                             | 1.7 1.7 1.7                            |                                      |
Table 2: Goodness of Fit statistics for Rn$^{222}$ activity and TDS dataset in water samples.

| Distribution | Goodness of Fit tests | Statistics | P-value | Decision at level (5%) |
|--------------|-----------------------|------------|---------|------------------------|
| Radon        | K-S modified test     | 0.075      | >0.15   | Accept Lognormal        |
|              | A-D test              | 0.477      | 0.225   |                        |
| Weibull      | K-S modified test     | 0.099      | >0.1    | Accept Weibull          |
|              | A-D test              | 0.456      | >=0.25  | Accept Weibull          |
| Exponential  | K-S modified test     | 0.184      | 0.022   | Reject Exponential      |
|              | A-D test              | 2.072      | 0.007   | Reject Exponential      |
| Gamma        | K-S modified test     | 0.117      | 0.235   | Accept Gamma            |
|              | A-D test              | 0.566      | 0.180   | Accept Gamma            |
| TDS          | K-S modified test     | 0.215      | <=0.01  | Reject Lognormal        |
|              | A-D test              | 2.494      | 2.04E-06| Reject Lognormal        |
| Weibull      | K-S modified test     | 0.173      | >0.1    | Accept Weibull          |
|              | A-D test              | 2.039      | <0.01   | Reject Weibull          |
| Exponential  | K-S modified test     | 0.193      | 0.014   | Reject Exponential      |
|              | A-D test              | 1.873      | 0.012   | Reject Exponential      |
| Gamma        | K-S modified test     | 0.174      | 0.006   | Reject Gamma            |
|              | A-D test              | 2.053      | <0.005  | Reject Gamma            |
References

[1] Cosma, C., Moldovan, M., Dicu, T., Kovacs, T. Radon in water from Transylvania (Romania). RADIATION MEASUREMENTS 43 pp. 1423-1428., 6 p. (2008)

[2] Bajwa, B. S., Kumar, S., Singh, S., Sahoo, S. K., & Tripathi, R. M. (2017). Uranium and other heavy toxic elements distribution in the drinking water samples of SW-Punjab, India. Journal of Radiation Research and Applied Sciences, 10(1), 13-19.

[3] Veiga, R., Sanches, N., Anjos, R. M., Macario, K., Bastos, J., Iguaitemy, M., ... &Umisedo, N. K. (2006). Measurement of natural radioactivity in Brazilian beach sands. Radiation measurements, 41(2), 189-196.

[4] El-Arabi, A. M. (2007). 226Ra, 232Th and 40K concentrations in igneous rocks from eastern desert, Egypt and its radiological implications. Radiation Measurements, 42(1), 94-100.

[5] Duggal, V., Sharma, S., & Mehra, R. (2017). Radon levels in drinking water of Fatehabad district of Haryana, India. Applied Radiation and Isotopes, 123, 36-40.

[6] Kumar, A., Singh, P., Agarwal, T., Joshi, M., Semwal, P., Singh, K., ... &Ramola, R. C. (2020). Statistical inferences from measured data on concentrations of naturally occurring radon, thoron, and decay products in Kumaun Himalayan belt. Environmental Science and Pollution Research, 27(32), 40229-40243.

[7] Semwal, P., Agarwal, T. K., Singh, K., Joshi, M., Gusain, G. S., Sahoo, B. K., &Ramola, R. C. (2019). Indoor inhalation dose assessment for thoron-rich regions of Indian Himalayan belt. Environmental Science and Pollution Research, 26(5), 4855-4866.

[8] Ramola, R. C., Prasad, M., Kandari, T., Pant, P., Bossew, P., Mishra, R., &Tokonami, S. (2016). Dose estimation derived from the exposure to radon, thoron and their progeny in the indoor environment. Scientific reports, 6(1), 1-16.

[9] Semwal, P., Singh, K., Agarwal, T. K., Joshi, M., Pant, P., Kandari, T., &Ramola, R. C. (2018). Measurement of 222 Rn and 220 Rn exhalation rate from soil samples of Kumaun Hills, India. Acta Geophysica, 66(5), 1203-1211.

[10] Ramola, R.C., Choubey, V.M., Negi, M.S., Prasad, Y., Prasad, G. Radon occurrence in soil-gas and groundwater around an active landslide. Radiation Measurements, 2008, 43(1), pp. 98–101
[11] Choubey, V.M., Bartarya, S.K., Ramola, R.C. Radon in Himalayan springs: A geohydrological control. Environmental Geology, 2000, 39(6), pp. 523–530.

[12] Ramola, R.C., Rawat, R.B.S., Kandari, M.S., Choubey, V.M. Measurement of radon in drinking water and indoor air. Radiation Protection Dosimetry, 1997, 74(1-2), pp. 103–105.

[13] Choubey, V. M., S. K. Bartarya, and R. C. Ramola. "Geological Controls on Radon in Soil and water of Pithoragarh region, Kumaun Himalaya, India." In Radon investigations in the Czech Republic XI and the 8th international workshop on the Geological Aspects of Radon Risk Mapping.—72–78, Czech Geol. Surv. Prague. 2006.

[14] Jobbagy, V., Kavasi, N., Somlai, J., Dombovari, P., Kardos, R., Kovacs, T. Radioanalytical investigations of uranium concentrations in natural spring, mineral, spa and drinking waters in Hungary. JOURNAL OF RADIOANALYTICAL AND NUCLEAR CHEMISTRY 286 : 2 pp. 417-422. , 6 p. (2010)

[15] Ramola, R.C., Choubey, V.M., Prasad, Y., Prasad, G., Bartarya, S.K. Variation in radon concentration and terrestrial gamma radiation dose rates in relation to the lithology in southern part of Kumaon Himalaya, India. Radiation Measurements, 2006, 41(6), pp. 714–720

[16] Ramola, R.C., Choubey, V.M., Prasad, G., Gusain, G.S., Tosheva, Z., Kies, A. Radionuclide analysis in the soil of Kumaun Himalaya, India, using gamma ray spectrometry. Current Science, 2011, 100(6), pp. 906–914

[17] Bourai, A.A., Gusain, G.S., Rautela, B.S., Joshi, V., Prasad, G. and Ramola, R.C., 2012. Variations in radon concentration in groundwater of Kumaon Himalaya, India. Radiation protection dosimetry, 152(1-3), pp.55-57.

[18] Kumar, A., Singh, P., Semwal, P., Singh, K., Prasad, M. and Ramola, R.C., 2021. Study of primordial radionuclides and radon/thoron exhalation rates in Bageshwar region of Kumaun Himalaya, India. Journal of Radioanalytical and Nuclear Chemistry, pp.1-7.

[19] Duggal, V., Sharma, S., Srivastava, A. K., & Mehra, R. (2018). Measurement of radon concentration in drinking water in Bhiwani district of Haryana. Journal of the Geological Society of India, 91(6), 700-703.
[20] Sahoo B.K., Mayya Y.S, Sapra B.K., Gaware J.J. Banerjee K.S. and Kushwaha, H.S., 2010 Radon exhalation studies in an Indian Uranium tailings pile, Radiation Measurements, 45, pp. 237-241.

[21] Ajay, K., Manpreet, K., Rohit, M., Sumit, S., Rosaline, M., Singh, K. P., & Bajwa, B. S. (2016). Quantification and assessment of health risk due to ingestion of uranium in groundwater of Jammu district, Jammu & Kashmir, India. Journal of Radioanalytical and Nuclear Chemistry, 310(2), 793-804.

[22] Valdiya, K.S. 1980. Geology of Kumaun Lesser Himalaya. WIHG Publ. Dehra Dun. 291 pp.

[23] Valdiya, K. S. (1988). Tectonics and evolution of the central sector of the Himalaya. Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences, 326(1589), 151-175.

[24] Chakraborty, C., Mandal, N., & Ghosh, S. K. (2003). Kinematics of the Gondwana basins of peninsular India. Tectonophysics, 377(3-4), 299-324.

[25] Misra, R. C., & Bhattacharya, A. R. (1976). The Central Crystalline Zone of northern Kumaun Himalaya: Its lithostratigraphy, structure and tectonics with special reference to plate tectonics. Him. Geol, 6, 133-154.

[26] Bhargava, O. N. (1980). Pre-Tertiary orogenies in the Himalaya: a review of various evidences. GeologischeRundschau, 69(3), 811-823.

[27] Prasad, Y., Prasad, G., Gusain, G.S., Choubey, V.M. and Ramola, R.C., 2009. Seasonal variation on radon emission from soil and water. Indian Journal of Physics, 83(7), pp.1001-1010.

[28] Gaware, J. J., Sahoo, B. K., Sapra, B. K., & Mayya, Y. S. (2011). Indigenous development and networking of online radon monitors in the underground uranium mine. Radiation Protection and Environment, 34(1), 37.

[29] Singla, A. K., Kansal, S., & Mehra, R. (2021). Quantification of radon contamination in drinking water of Rajasthan, India. Journal of Radioanalytical and Nuclear Chemistry, 327(3), 1149-1157.

[30] Kumar, M., Kaushal, A., Sahoo, B. K., Sarin, A., Mehra, R., Jakhu, R., ... & Sharma, N. (2019). Measurement of uranium and radon concentration in drinking water samples and assessment of ingestion dose to local population in Jalandhar district of Punjab, India. Indoor and Built Environment, 28(5), 611-618.
[31] Mueller, J. D., & Rehder, G. (2018). Metrology of pH measurements in brackish waters—
part 2: experimental characterization of purified meta-cresol purple for spectrophotometric
pH measurements. *Frontiers in Marine Science, 5*, 177.

[32] Qadir, R. W., Asaad, N., Qadir, K. W., Ahmad, S. T., & Abdullah, H. Y. (2020).
Relationship between radon concentration and physicochemical parameters in
groundwater of Erbil city, Iraq. *Journal of Radiation Research and Applied Sciences, 1-9.*

[33] Jeyaraj, M., Indhuleka, A., & Arunpaul, C. (2019). Investigation of Water Quality Index
of River Noyyal and Its Connected Ponds Coimbatore Tamil Nadu India. *Oriental Journal
of Chemistry, 35*(3), 1125.

[34] Mittal, S., Rani, A., & Mehra, R. (2016). Radon levels in drinking water and soil samples
of Jodhpur and Nagaur districts of Rajasthan, India. *Applied Radiation and Isotopes, 113*,
53-59.

[35] Ravikumar, P., & Somashekar, R. K. (2014). Determination of the radiation dose due to
radon ingestion and inhalation. *International Journal of Environmental Science and
Technology, 11*(2), 493-508.

[36] United Nations Scientific Committee on the Effects of Atomic Radiation. (2000b).
Sources, effects and risks of ionizing radiation, report to the general assembly with
scientific annexes, United Nations, New York. 1, 126–127.

[37] UNSCEAR (1993) United Nations Scientific Committee on the effects of atomic
radiation—sources and effects of ionizing radiation, 1993 report to the General Assembly,
with scientific annexes. United Nation Sales Publication E.94.IX.2. United Nations, New
York.

[38] International Commission on Radiological Protection. (2002). Guide for the practical
application of the ICRP human respiratory tract model. ICRP supporting guidance 3.
Oxford: Pergamon.

[39] Kaur, M., Tripathi, P., Choudary, I., Mehra, R., & Kumar, A. (2017). Assessment of
annual effective dose due to inhalation and ingestion of radon in water samples from some
regions of Punjab, India. *International Journal of Pure and Applied Physics, 13*(2), 193–
200.

[40] Kumar, A., Arora, T., Singh, P., Singh, K., Singh, D., Pathak, P. P., & Ramola, R. C.
(2021). Quantification of radiological dose and chemical toxicity due to radon and
uranium in drinking water in Bageshwar region of Indian Himalaya. *Groundwater for Sustainable Development*, **12**, 100491.

[41] WHO (2008). Guidelines for drinking water quality. World Health Organization, Geneva.

[42] European Commission. (2001). Commission recommendations of 20th December 2001 on the protection of the public against exposure to radon in drinking water. 2001, 2001/982/ Euratom, L344/85.

[43] USEPA (1991) United States Environmental Protection Agency-National primary drinking water regulations for radionuclides: notice of proposed rule-making. Fed Reg 56:33050–33127.

[44] Leon Harter, H., Khamis, H.J. and Lamb, R.E., 1984. Modified Kolmogorov-Smirnov tests of goodness of fit. Communications in Statistics-Simulation and Computation, **13**(3), pp.293-323.

[45] Stephens, M. A. (1977). Goodness of Fit with Special Reference to Tests for Exponentiality, Technical Report No. 262, Department of Statistics, Stanford University, Stanford, CA.

[46] Lu, G. Y., & Wong, D. W. (2008). An adaptive inverse-distance weighting spatial interpolation technique. *Computers & geosciences*, **34**(9), 1044-1055.