

Geo-spatial analysis of radon in spring and well water using kriging interpolation method

Abdul Razzaq Khan, Muhammad Rafique, Saeed Ur Rahman, Muhammad Basharat, Chand Shahzadi and Ishtiaq Ahmed

ABSTRACT

Radon activity concentration was measured in 101 springs and well water samples collected from Muzaffarabad city and its outskirts. Sixty springs and 41 well water samples were analyzed, using RAD7, an electronic radon detector manufactured by DurrIDGE Company Inc., for the estimation of radon borne activities and their relevant effects. Results obtained show that for spring water, the water borne radon activity varied from 0.246 ± 0.348 to 34.36 ± 5.54 Bq L⁻¹ with an average value of 10.16 ± 2.42 Bq L⁻¹. For well water, the water borne radon activity varied from 0.86 ± 0.10 to 16.12 ± 0.22 Bq L⁻¹ with an average value of 4.21 ± 0.13 Bq L⁻¹. Concentration of radon borne activities were subsequently used for determination of inhalation and ingestion doses. The inhalation and ingestion doses for spring water samples varied from 0.0062 ± 0.0087 to 0.865 ± 0.14 mSv y⁻¹ and 0.052 ± 0.073 to 7.22 ± 1.16 mSv y⁻¹, respectively. The inhalation and ingestion doses for well water varied from 0.022 ± 0.0025 to 0.41 ± 0.0054 mSv y⁻¹ and 0.18 ± 0.021 to 3.38 ± 0.045 mSv y⁻¹, respectively. 33.33% of spring and 7.32% of well water samples were found with values above the recommendation levels of the United States Environmental Protection Agency (~11.1 Bq L⁻¹). As a single measurement cannot serve as a best estimate of the unsampled areas in the region, Kriging interpolation method, a geo-statistical method, was used to get an estimate of spatial distribution of water borne radon in the area of study. Kriging mapping shows that higher radon concentrations are found in areas with lithology consisting of sandstones, siltstones, shales and claystones.

Key words | inhalation and ingestion doses, Kriging interpolation method, RAD7, radon activity concentration, spatial distribution

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INTRODUCTION

Radon-222 (²²²Rn), naturally occurring radioactive gas, is produced as a result of the emission of alpha particle (⁴He) from Radium-226 (²²⁶Ra). Since ²²⁶Ra is found ubiquitously as trace amount in soil, water, minerals forming rocks in the lithosphere, this makes its progeny (²²²Rn) available everywhere in the environment. As ²²²Rn is obtained from the decay of ²²⁶Ra, so higher levels of ²²⁶Ra in bedrocks are expected to produce higher concentrations of ²²²Rn in water. Depending upon flow rate, groundwater can carry

²²⁶Ra generated ²²²Rn over large distances. Flow rate of water is controlled by parameters like rock porosity and permeability. Igneous rocks, such as granites, have high values of permeability. Water with elevated levels of radon shows detectable levels of alpha emitting nuclides, Lead-210 (²¹⁰Pb) and Polonium-210 (²¹⁰Po) (Musa 2003). Radon in water has been identified as public health concern (EPA 1994, 1999). Usually, surface water contains very small amounts of dissolved radon due to rapid volatilization of

the gas into the atmosphere (NCRWACR 2011). Typically, radon concentrations in surface waters are less than $4,000 \text{ Bqm}^{-3}$ (4 Bq/L). On the other hand, water from wells can have high radon concentrations ($>4,000 \text{ Bqm}^{-3}$ or 4 Bq/L) (Hopke *et al.* 2000).

Alpha particles, emitted from the decay of radioactive radon gas and its progenies, are associated with the biological effects under the low radon exposure conditions within closed domestic environments. On passing through a cell nucleus, alpha particle damages deoxyribonucleic acid (DNA) (WHO 2009; Nisar *et al.* 2017) and results in removals and rearrangements of chromosomal regions leading to genetic instabilities implicated in tumor progression. Due to low solubility of radon in water, making it prone to de-gassing from water in contact with air, water releases radon into the indoor air and contributes to the total air borne radon concentrations. Hopke *et al.* (2000), reported that health risk posed by radon released from water, even at typical ground water concentrations, is estimated to be larger than the risk posed by other water contaminants such as disinfection byproducts.

DNA structure, function, and replication can easily be affected by the radiation emitted from inhaled radon and its decay products. These deleterious effects are usually confined to the respiratory tract. Exposure, then, to elevated levels of radon and its decay products over time will increase a person's risk of developing lung cancer during his or her lifetime.

Ingestion of radon in water may also pose a direct health risk through irradiation of sensitive cells in the gastrointestinal tract and other organs once it is absorbed into the bloodstream (Mills 1990; Crawford-Brown 1991). Thus, radon in drinking water could potentially produce adverse health effects in addition to lung cancer (Hopke *et al.* 2000).

For the public, water borne radon is a double source of exposure, via taking water and breathing of radon gas released from water (Cross *et al.* 1985; Nasir *et al.* 2015). Lung cancer (due to breathing) and stomach cancer (due to ingestion) are two important effects linked with the exposure of radon (Mills 1990). Ingestion of radon causes irradiation of sensitive cells in the gastrointestinal tract and other organs once it is absorbed into the bloodstream (Crawford-Brown 1989, 1991).

Interest in the study of Rn in water is increased due to the health hazards of radon in water. In the USA, almost 1–7% of lung cancer sufferers have endorsed the indoor radon exposure resulting from groundwater (Cothorn *et al.* 1986).

Worldwide survey has revealed the mean concentration of radon in groundwater to be about 183 Bq/L (NCRP 1984). For tap water, suggested limit of radon concentration is, however, not recommended but the projected limit is 150 Bq/L . The Environmental Protection Agency (EPA) has proposed a maximum contaminant level of radon in public drinking water supplies to be 11.1 Bq L^{-1} (300 pCi L^{-1}). World Health Organization (WHO) guidelines permit a maximum ^{222}Rn concentration of 100 Bq L^{-1} in drinking water for public water supplies.

This current study provides the results of radon measurements in the wells and spring water collected from different locations of Muzaffarabad, Pakistan, and remote areas of the Muzaffarabad district. Three-dimensional (3D) radon contour maps on a regional scale with a correspondingly higher spatial resolution are drawn. Spring water naturally runs out of the earth surface continuously, whereas well water is drawn using a pump from a drilled well in the ground. Well water, a dug well, is accumulated with time and can have low radon due to water–air interfacing over time. For public interest, 3D contour maps for radon in spring and well water on a regional scale with a correspondingly higher spatial resolution are drawn. As measurements were taken at single specific location and could not be representative of unsampled area, we have used geostatistical method, Kriging interpolation method, to get estimate of spatial distribution of radon concentration in spring and well water. Present research is addressing the question to what extent variation in water borne radon activity (WBRnA) can be found within the context of a geographical information system (GIS), and intended at the compilation of water borne radon prognosis maps. This map could serve as a best guide to local establishments in the future urban planning process, and in particular these 3D GIS based contour maps will serve as baseline information for the compilation of radon risk maps. To represent the spatial distributions of the radon concentration in the spring and well water samples of the study area, the kriging interpolation method was used (Sarma 2009).

STUDY AREA

The study area Muzaffarabad, the state capital of Azad Jammu and Kashmir, is located at the confluence of the Neelum and Jhelum Rivers. Azad Jammu and Kashmir, also called as Pakistan Administered Kashmir, lies between longitude of 73°–75° and latitude of 33°–36° (*Disaster Risk Management Plan, Azad Jammu & Kashmir 2008; Shafique et al. 2012*). The area is divided by three rivers: Jhelum, Neelum, and Kunhar. It includes district Muzaffarabad and small part of the district Mansehra KPK. Muzaffarabad lies at a height of 737 m (2,418 ft) and covers an area of 1,642 km². Muzaffarabad City is located at the junction of river Neelum and river Jhelum, which both originate from the Indian-held Kashmir.

METHODS

Sampling

Since population density of Muzaffarabad city is not uniformly distributed over the territory, random sampling technique was adopted and drinking water samples were collected from the sites that were frequently used by the inhabitants of the area. People consume drinking water – without further treatment – directly from wells and springs. In the present study, 60 springs and 41 well water samples were collected in 40 mL glass vials. While taking a sample, the vial was kept vertically erect below the falling spring water at its actual origin, such that it is filled slowly without producing water bubbles. After the vial is filled up to its edges, teflon cap was placed on it at once and made tight gently. Presence of the bubbles in the vial was checked, in order to assess loss of radon, by holding the vial inverted. Date, time and temperature was noted for each sample. Geographical parameters of the sampling sites were determined using Magellan GPS tracker. GIS based [Figure 1\(a\)](#) and [1\(b\)](#) represents the distribution of sampling sites in the Muzaffarabad district. A record of sample collection time and its radiometric analysis was properly maintained to apply the decay correction factor (DCF) where needed.

Experimental procedure

Measurement of the radon content in the collected sample from spring water was carried out in a solid state nuclear track detection laboratory (SSNTD) of the Department of Physics. For this purpose, RAD 7 set up was used in accordance with the EPA Protocol Test (average taken over at the results of two days).

RAD 7 is solid state detector that can detect alpha particles only. The interior detection cell of the RAD 7 is 0.7-liter electrical conductor hemisphere. At the center of this hemisphere, a semiconductor, ion implanted, planar detector is present. A potential difference of 2,000–2,500 V is created between the detector and the hemisphere cell; that produces an intense field throughout the volume of the chamber. The positive charges are attracted and collected on the detector due to this electric field. With RAD H₂O accessories, the device can be used for the analysis of different water samples. RAD H₂O offers different protocols defined as Wat40 and Wat250.

In the present study, Wat40 protocol in the grab mode was used (RAD7 manual). The analysis technique was a closed loop arrangement with three main components: (i) the RAD 7 device on the left, (ii) a water vial with aerator on the left, (iii) a desiccant tube fitted in the retort stand as shown in [Figure 2](#).

The air was recirculated and extracted the radon from water continuously until the equilibrium state was reached in the first 5 min, after which more radon could not be extracted. For a 40 mL vial sample, we have used Wat40 protocol in the grab mode to get maximum efficiency of 99%.

When radon-222 decays within the chamber, a positively charged Po-218 nucleus is formed as the daughter product, which is pushed by the electric field on to the detector where it is being attached. Po-218 has a half-life of 3.05 min and decays by emitting an alpha particle, which has 50% probability of entering the detector and producing the electric pulse. Succeeding decays of the Po-218 nucleus produce alpha particles of different energies and beta particles, which are not detected. Different isotopes have different alpha energies and produce different strength signals in the detector and are recorded in different windows. The height of the pulse is proportional to the energy of the incoming particle.

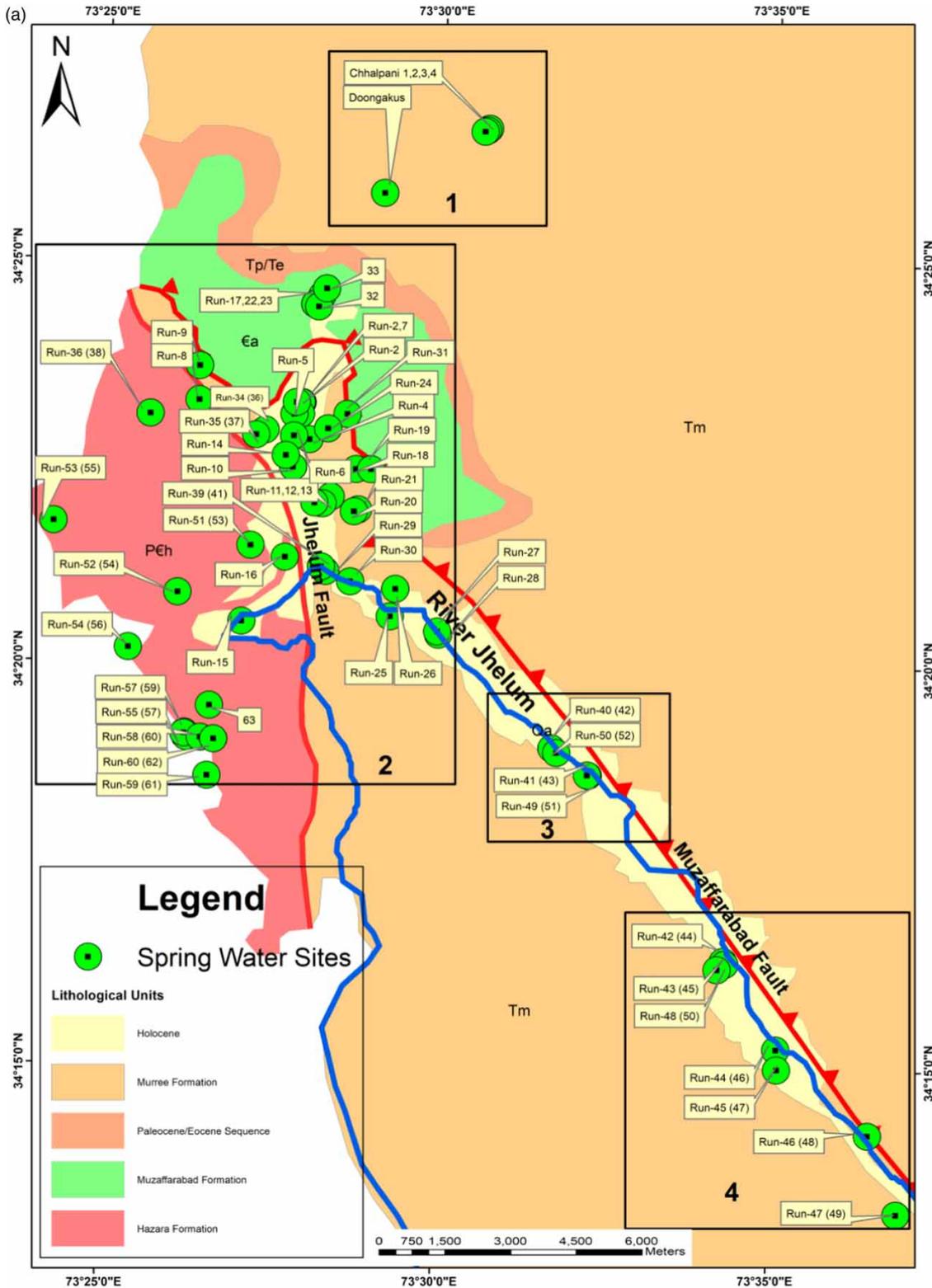


Figure 1 | (a) Geological map of the study area, showing spring water sampling sites, digitized after Calkins et al. (1975) and Geological Survey of Pakistan (Iqbal et al. 2004). (b) Geological map of the study area, showing well water sampling sites, digitized after Calkins et al. (1975) and Geological Survey of Pakistan (Iqbal et al. 2004). (Continued.)

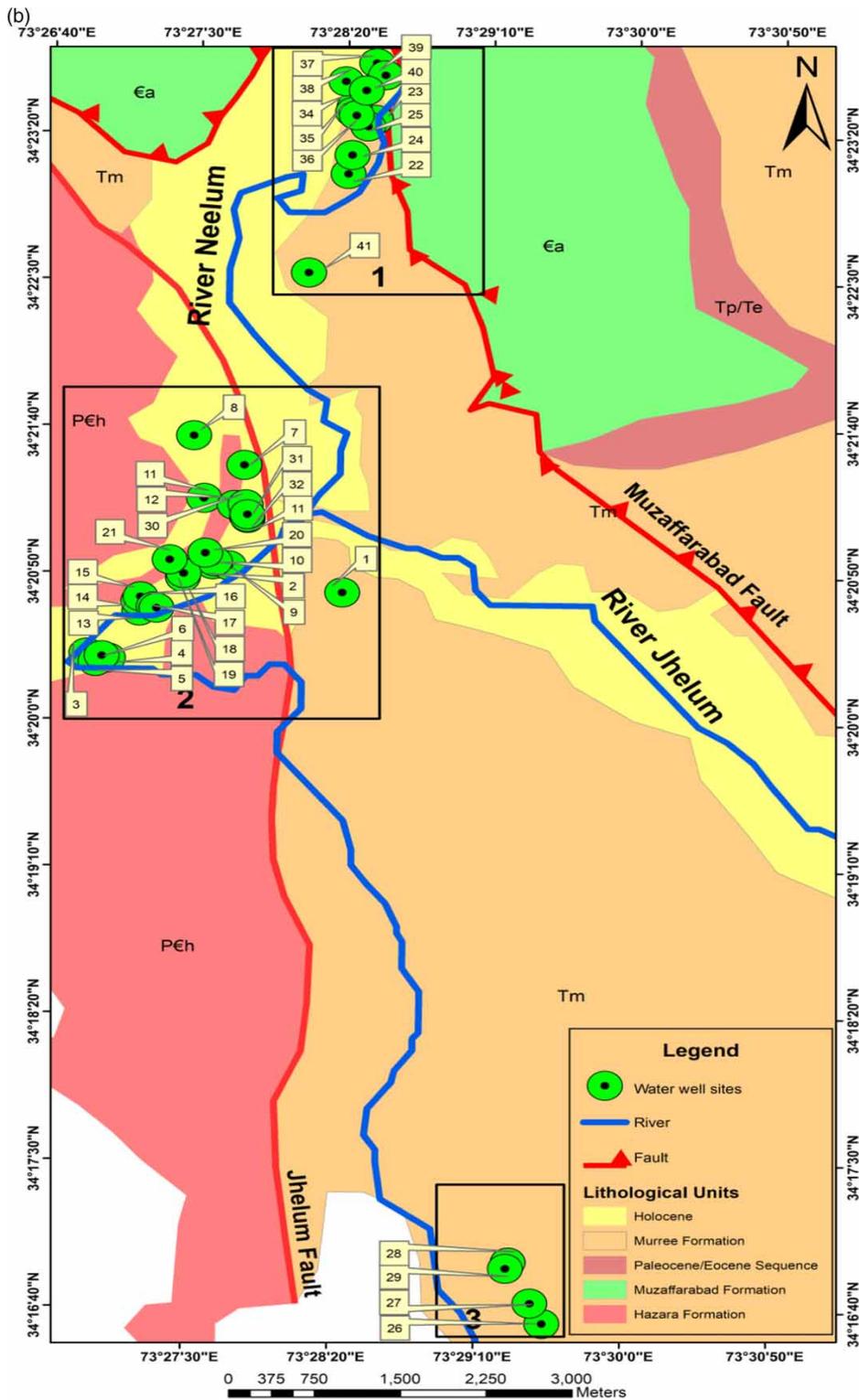


Figure 1 | Continued.

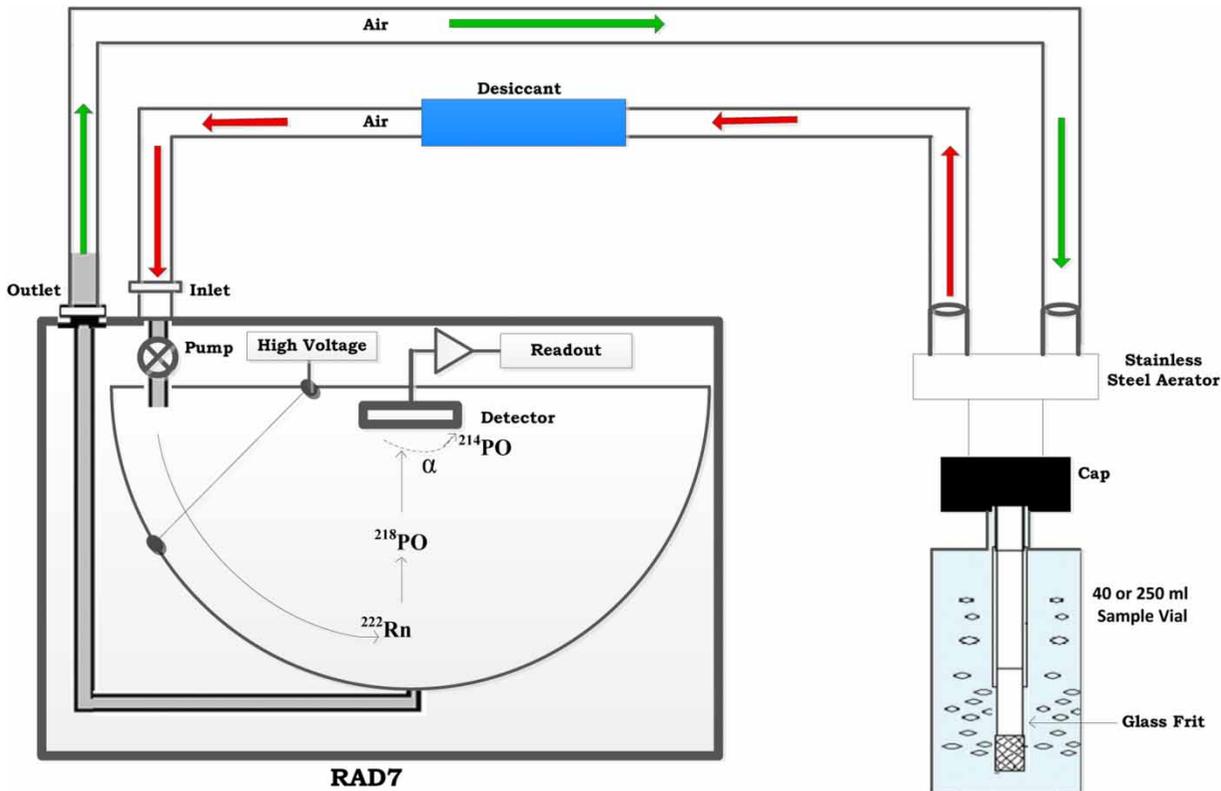


Figure 2 | Schematic diagram for the measurement of radon in water using RAD7 and accessories.

In this way, the mean radon content in the sample is determined using Po-218 activity. In addition to the Po-218, the subsequent radon daughters, Po-214 and Po-210 also emit alpha particles; however, RAD 7 does not detects them.

For the analysis of the next sample, the device is purged for 10 minutes or more so that the relative humidity is less than 6%. With RAD 7, one can measure radon concentration in water over a range from below 10 pCi/L to above 400,000 pCi/L. The sensitivity of the device is 0.8 counts per hour per Bq/m³. From each site, two samples were taken to avoid resampling in case of possible errors in the analysis. Almost all the samples were analyzed in the shortest possible time in consideration of short half-life radon. Most of the samples were analyzed within 24 h of sampling. Yet DCF was applied for 10 h delay and above to correct the sample activity using formula:

$$A_0 = A \times DCF \quad (1a)$$

where A_0 is the original activity of the sample, A is the activity at sample analysis time, DCF is the decay correction

factor. DCF was calculated by using the formula:

$$\text{The decay correction factor (DCF)} = \exp(T/132.4) \quad (1b)$$

where T is the decay time in hours.

Evaluation of the dissolved radon in the water

The dissolved radon in the water is ingested while drinking and inhaled when released from water in to the indoor air. Therefore, the annual effective doses (AEDs) for ingestion and inhalation were evaluated according to the parameters given in the UNSCEAR (2006) report.

$$\text{For ingestion: } E_{WIg} (mSv a^{-1}) = C_{Rnw} \times C_w \times EDC \quad (2)$$

where the parameter E_{WIg} is the effective dose for ingestion, C_{Rnw} is the radon concentration in water (kBq m⁻³), C_w is the weighted estimate of water consumption (60 L a⁻¹)

and EDC is the effective dose coefficient for ingestion 3.5 nSv Bq^{-1} .

For inhalation:

$$E_{Wih} (\text{mSv a}^{-1}) = C_{RnW} \times R_{aW} \times F \times O \times DCF \quad (3)$$

where the parameter E_{Wih} is the effective dose for inhalation, R_{aW} is the ratio of radon in air to radon in tap water (10^{-4}) and the other terms have been explained above.

Geo-spatial analysis

As sample collections were carried out from specific locations and measurements were not representative of the entire region, therefore, to get information about unsampled locations we have made utilization of geostatistical methods (Armstrong 1998). To represent the spatial distributions of the natural radon content present in the water samples of the study area, the Kriging interpolation method was used. The Kriging method, after D. Krige (Krige 1966), was further developed by G. Matheron (Matheron 1970) is regarded as the best linear unbiased estimator (Sarma 2009). The ordinary Kriging formula is given as (Dindaroglu 2014):

$$Z(S_0) = \sum_{i=1}^N \lambda_i Z(S_i) \quad (4)$$

where $Z(S_i)$ is the estimated value at the i th location (i th), λ_i is the unknown weight for the measured value at the i th location (i th) and S_0 is the measurement location.

RESULTS AND DISCUSSION

As discussed earlier, 60 springs and 41 well water samples were analyzed for the estimation of radon borne activities and their relevant effects. Activity values, for both types of samples, are presented in Figures 3 and 4.

For spring water, the water borne radon activity vary from 0.246 ± 0.348 – $34.36 \pm 5.54 \text{ Bq L}^{-1}$ with an average value of $10.16 \pm 2.42 \text{ Bq L}^{-1}$. The minimum value was found in the sample S-43, collected from Bandway Steel Mill, whereas the maximum value was found in the sample; S-7, collected from Madina Plaza Muzaffarabad.

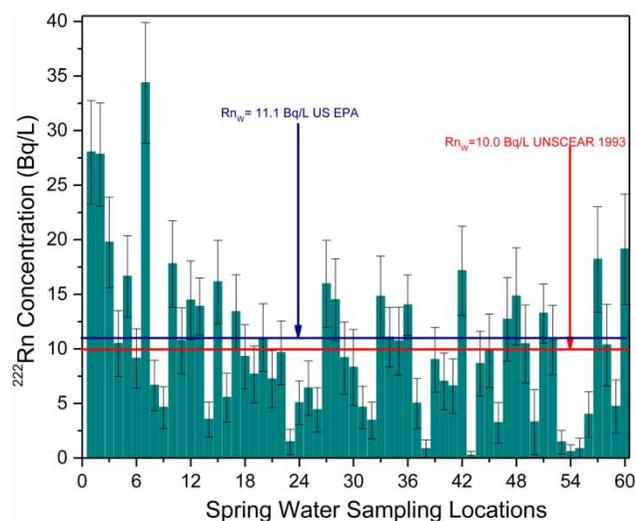


Figure 3 | Radon concentration in spring water samples collected from Muzaffarabad district and environs, Pakistan.

Concentration of radon borne activities were subsequently used for determination of inhalation and ingestion doses. For spring water, the inhalation and ingestion doses varied from 0.0062 ± 0.0087 to $0.865 \pm 0.14 \text{ mSv y}^{-1}$ and 0.052 ± 0.073 to $7.22 \pm 1.16 \text{ mSv y}^{-1}$, respectively. These doses were determined using Equations (2) and (3), respectively.

The effect of values for pH of spring water on water borne radon activities were also analyzed. Maximum value of pH was found to be 8.3 (at S-53 location) and minimum value 6.9 (at S-60 location). Maximum radon concentration 34.36 ± 5.54 was found in sample S-7 with pH 7.46 and

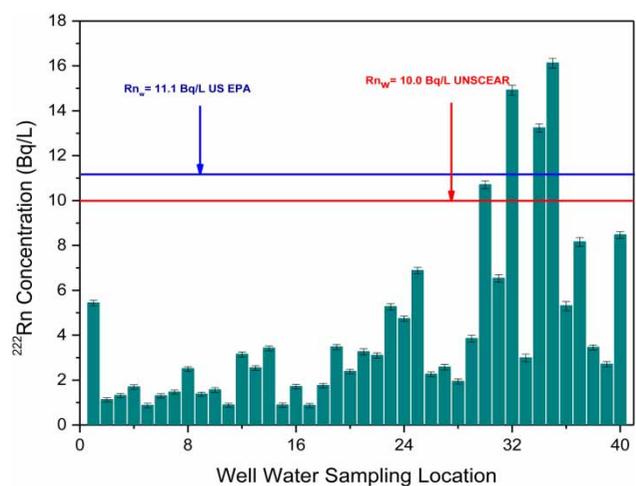


Figure 4 | Radon concentration in Well water samples collected from Muzaffarabad district and environs, Pakistan.

minimum radon concentration $0.246 \pm 0.348 \text{ Bq L}^{-1}$ was found in sample S-43 with pH value 7.82. Relative humidity varied from 8.5 (at S-50 location) to 26.5 (at S-23 location). Average temperature of spring water samples at the time of analyses varied from 14 to 31.6°C . Sampling heights varied from 2,192 to 4,362 feet.

Water borne radon activities (WBRnA) for well water varied from 0.86 ± 0.10 to $16.12 \pm 0.22 \text{ Bq L}^{-1}$ with an average value of $4.21 \pm 0.13 \text{ Bq L}^{-1}$. The minimum value was found in the sample W-05, whereas the maximum value was found in the sample W-35. Concentrations of WBRnA were subsequently used for determination of ingestion and inhalation doses using Equations (2) and (3). The inhalation and ingestion doses varied from 0.022 ± 0.0025 to $0.41 \pm 0.0054 \text{ mSv y}^{-1}$ and 0.18 ± 0.021 to $3.38 \pm 0.045 \text{ mSv y}^{-1}$, respectively. The pH values of all the well water samples were measured and the maximum value was found to be 7.82 (at W-37 location) and minimum value 7.13 (at W-37 location). Commonly accepted pH values range from 6.5 to 8.5. So, pH of all spring and well water samples are within the acceptable range.

Relative humidity varied from 7.75 (at W-37 location) to 15 (at W-2 location). Average temperature of well water samples at the time of analyses varied from 25.12 to 30.4°C . Height of sample collection ranged from 2,246 to 2,628 feet.

Relative frequency graph of measured concentrations of radon in spring and well drinking water samples was constructed. Figures 5 and 6 show spring and well water frequency distribution graphs. Lognormal distribution was fitted on observed data of radon concentrations in both types of water samples. For spring water, the coefficient of determination (adjusted R^2) was obtained as 0.748 by lognormal distribution fit. Adjusted R^2 usually gives an idea of how many radon concentration data points fall within the line of regression equation. Adjusted R^2 tells the percentage of variation in independent variable that will affect the dependent variable. Lognormal distribution seems appropriate fitting for the spring water data. In the case of the well water sample, the coefficient of determination (adjusted R^2) was obtained as 0.986 by lognormal distribution fit. Figure 6 shows radon concentration data with a long tail to the right suggesting that a frequency distribution curve skewed to the right (i.e. had positive skewness). The

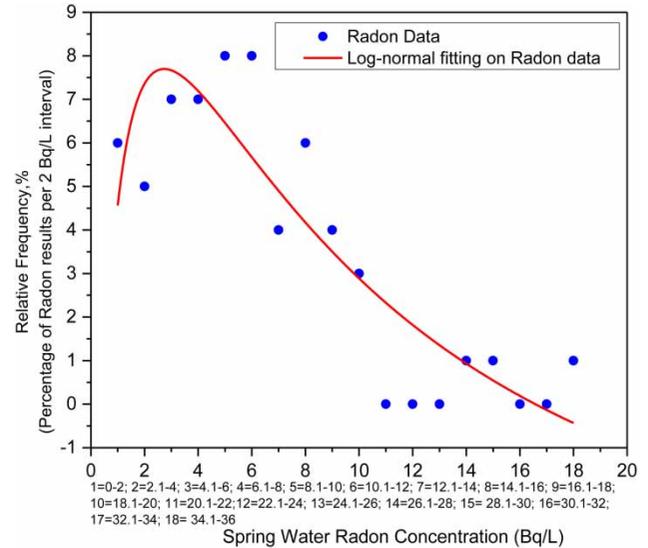


Figure 5 | Lognormal distribution fitting on spring water radon data in Muzaffarabad district.

lognormal distribution of radon measurements observed for well water samples in the current study is consistent with USEPA studies and analyses findings, which have shown that environmental radioactivity data tend to follow a lognormal distribution (www.dep.state.fl.us/water/wf/dw). The reason for lognormal trend of radon in water data may be attributed to heterogeneous distribution of uranium in rocks across which water travels before accumulating inside the well.

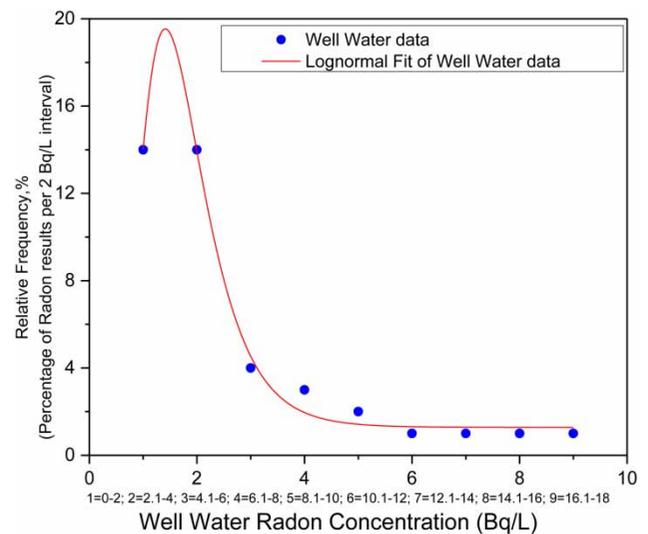


Figure 6 | Lognormal distribution fitting on well water radon concentration in Muzaffarabad district.

For well and spring waters, maximum values of 16.12 ± 0.22 and 34.36 ± 5.54 Bq/L were reported at pH values of 7.44 and 7.46, respectively. Since radon is a direct daughter product of radium and radium dissolved in water with low pH, radon showed highest concentration at intermediate pH levels.

Total AED for spring water samples varied from 0.052 to 7.30 mSv y^{-1} with mean value of $2.16 \pm 0.008 \text{ mSv y}^{-1}$, whereas, for well water, it ranged from 0.184 to 3.43 mSv y^{-1} with a mean value of $0.89 \pm 0.027 \text{ mSv y}^{-1}$. These dose values are well above the safe limit (0.1 mSv y^{-1}) of AED recommended by World Health Organization (WHO 2011).

Mean value of excess lifetime cancer risk (ELCR) for spring and well water samples was computed to be 7.56×10^{-3} and 3.13×10^{-3} , respectively, which, in both cases, is much higher than the upper bound of 0.1×10^{-3} for drinking water, as proposed by the USEPA (USEPA 2012). 98% of the spring water and all of the well water samples have ELCR greater than world standard value of 0.29×10^{-3} (Ononugbo

& Avwiri 2016). Similarly, 90% of spring water and 65% of the well water samples have ELCR above worldwide average of 1.45×10^{-3} (Etuk et al. 2017). 3D contour maps for radon in spring and well water are shown in Figures 7 and 8.

Maximum concentration of water borne radon activities, 34.36 ± 5.54 , 28.01 ± 4.74 and 27.81 ± 4.73 Bq/L were found on locations SW7, SW1 and SW2, respectively. These locations are a lithological part of Murree formation. Lithology of these sampling location mainly consists of sandstones, siltstones with shales and claystones. The variation of the radon concentration in groundwater is mainly controlled by lithology of aquifer, and the depth and the contact of water with the underground rocks containing radioactive elements (Misdaq & Elharti 1997).

Elevated WBRnA from spring and well waters may be due to water interaction during its movement, with bedrocks having elevated concentration of uranium, or possibly through cracks with surfaces of minerals containing higher

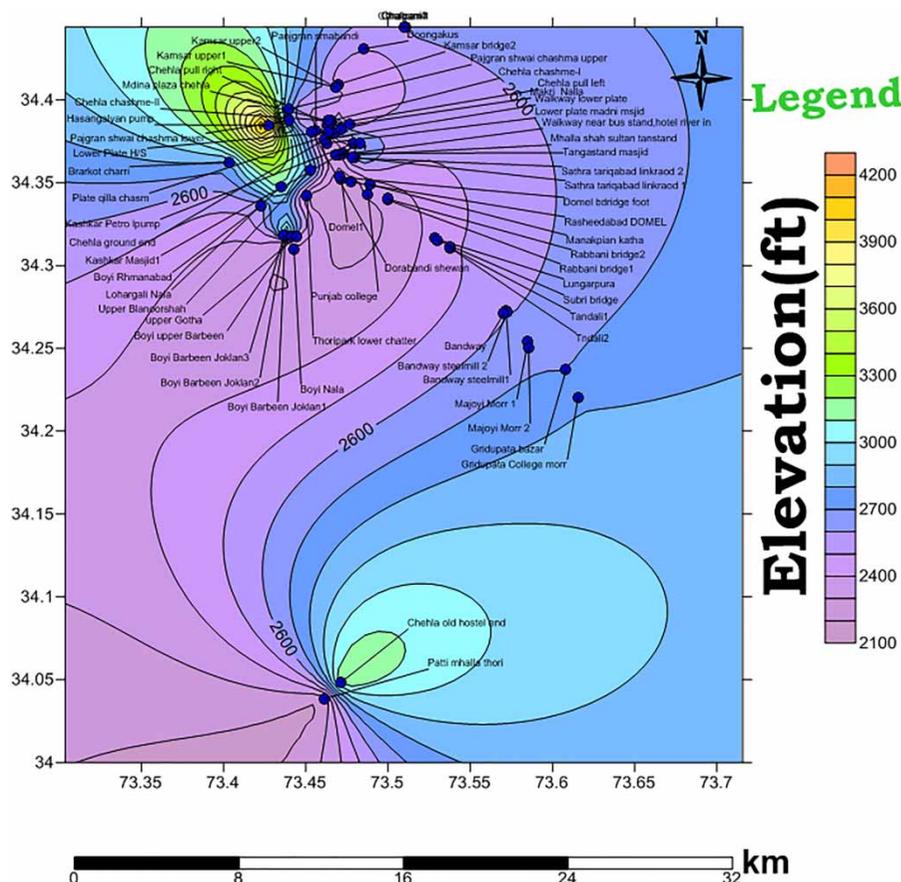


Figure 7 | Three-dimensional contour map of ^{222}Rn concentration in spring water samples from Muzaffarabad city and outskirts, Pakistan.

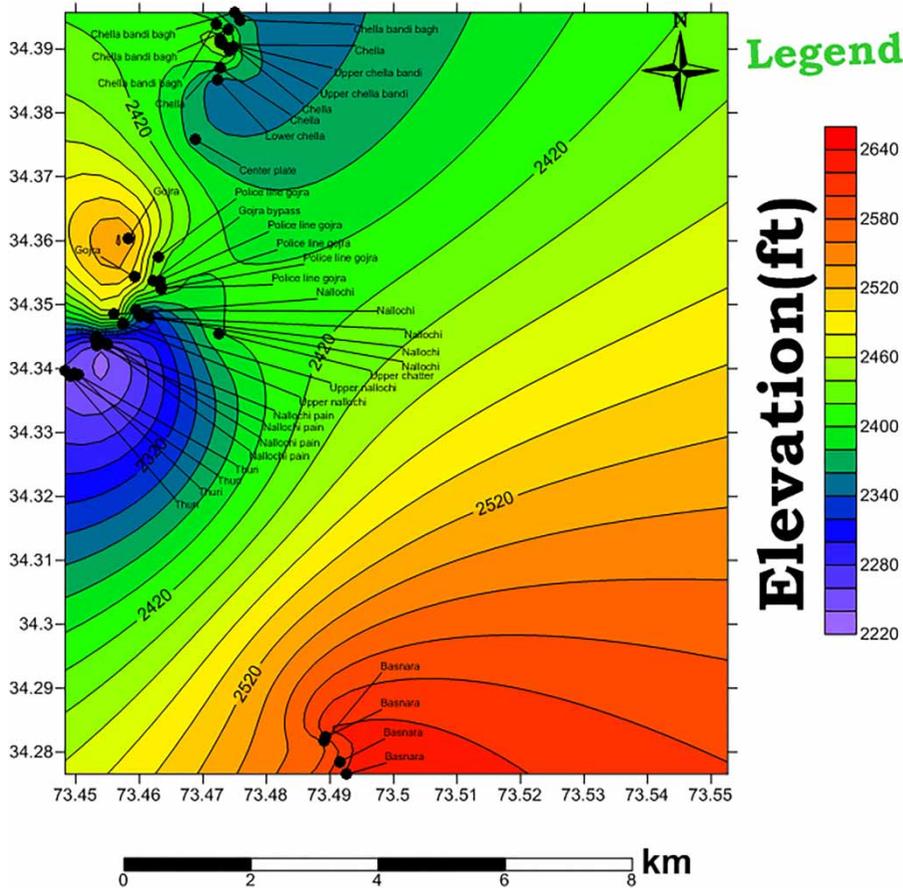


Figure 8 | Three-dimensional contour map of ^{222}Rn concentration in well water samples from Muzaffarabad city and outskirts, Pakistan.

concentrations of ^{226}Ra . Higher levels in spring waters may be the reason, as reported by Akerblom & Lindgren (1997) that elevated WBRn levels are manifested in intramontaneous water flowing through areas with uranium-bearing rock types (e.g. uranium-rich granites, pegmatites and vulcanites). Extensive further studies are needed to be carried out to assess uranium/radium contents of underlying bedrocks with the help of ground radiometry and to establish a relationship with WBRnA.

COMPARISON OF RESULTS WITH LITERATURE DATA

Comparison of results (Table 1) obtained from the current study (0.246 ± 0.348 – 34.36 ± 5.54 Bq/L with average value of 10.16 ± 2.42 Bq/L for spring water, 0.86 ± 0.10 to

16.12 ± 0.22 Bq/L with average value of 4.21 ± 0.13 Bq/L for the well water), with the data available in literature shows that radon concentration in water samples fall far below the water borne activity action contamination level recommended by World Health Organization (100 Bq L^{-1}) publication (WHO 2008) of the drinking water guidelines.

However, 33.33% of spring water samples were found with values above the recommendation levels of the United States Environmental Protection Agency, USEPA ($\sim 11.1 \text{ Bq L}^{-1}$), on the other hand only 7.32% of well water samples were found with radon concentration above USEPA recommendations.

The mean radon concentration of $10.16 \pm 2.42 \text{ Bq L}^{-1}$ in the drinking spring water samples from the studied area is slightly higher than the world average of 10.0 Bq L^{-1} (UNSCEAR 2000).

Table 1 | Comparison of current study results for ^{222}Rn concentration in spring and well water with data available in literature

Water type	^{222}Rn activity concentration (Bq L ⁻¹)	Region of the study	Geology of the area	Reference	
Spring water	1.4–105	South Catalonia, Spain	Volcanic (granite) and sedimentary rocks (e.g. limestone, sandstone)	Fonollosa <i>et al.</i> (2016)	
	2.11–120	Balaton Highland, South Transdanubia and The South Great Plain, Hungary	Sedimentary rocks	Somlai <i>et al.</i> (2007)	
	1.4–43.7	Lithuania	Crystalline and sediment rocks	Ladygiene <i>et al.</i> (1999)	
	1,595	Momin Prohod, Bulgaria		Pressyanov <i>et al.</i> (2007)	
	1.5–181	Padua, Euganean Thermal District, Italy	Volcanic rocks	Cantaluppi <i>et al.</i> (2014)	
	1,029	Galicia, Spain	Granitic and slate rocks	Llerena <i>et al.</i> (2013)	
	0.39 ± 0.19 to 1.17 ± 0.21	Amasya, Turkey		Oner <i>et al.</i> (2009)	
	0.246 ± 0.348 to 34.36 ± 5.54 Bq L ⁻¹ with an average value of 10.16 ± 2.42 Bq L ⁻¹ .	Muzaffarabad, Pakistan	Mainly sedimentary rocks	Current study	
	Well water	1.46 to 53.64	Bursa, Turkey	Geology is basically composed of granitic formations. Most rock formations, such as limestone, clays, lavas, calcareous shale, sandstones and conglomerates are seen in the region.	Akar <i>et al.</i> (2012)
		4–63,560	Stockholm County, Sweden	Various, crystalline bedrock	Skeppstrom & Olofsson (2006)
47–1,600		Visé, Belgium	Granitic bedrock	Bourgoignie <i>et al.</i> (1982)	
77,000		Finland		Salonen (1988)	
1.6–215		Curitiba metropolitan area, Brazil	The substrate of the metropolitan area of Curitiba, Brazil, contains rocks that are mostly metamorphic. In Brazil, there is a great variety of igneous intrusive rocks, including granites and granitoids	Corrêa <i>et al.</i> (2014)	
For Spring season; 2.29 ± 0.17 to 27.25 ± 1.07. For summer season: 1.44 ± 0.18 to 27.45 ± 1.25.		Konya, Turkey	Mostly botanical soil, clayey gravel, gravel clay, clay, gravel, conglomerate	Erdogan <i>et al.</i> (2013)	
0.86 ± 0.10 to 16.12 ± 0.22 Bq L ⁻¹ with an average value of 4.21 ± 0.13 Bq L ⁻¹ .		Muzaffarabad, Pakistan	Mainly sedimentary rocks	Current study	

For spring water (Table 1), current study reported values are considerably smaller than those reported for Spain (in regions of South Catalonia and Galicia), Hungary, Lithuania, Bulgaria, Italy and higher than from the values reported for Amasya, Turkey. For well water (Table 1), WBRnA for current study are smaller from those reported for Turkey (Bursa), Sweden (Stockholm County), Belgium (Visé), Finland, Brazil (Curitiba metropolitan area) and greater than Turkey (Konya).

CONCLUSION

The radon activity concentrations were measured in 101 spring and well water samples collected from Muzaffarabad city and its outskirts. Results are summarized as follows:

1. For spring water, the water borne radon activity vary from 0.246 ± 0.348 to 34.36 ± 5.54 Bq L⁻¹ with an average value of 10.16 ± 2.42 Bq L⁻¹.
2. For well water, the water borne radon activity varied from 0.86 ± 0.10 to 16.12 ± 0.22 Bq L⁻¹ with an average value of 4.21 ± 0.13 Bq L⁻¹.
3. The inhalation and ingestion doses for spring water samples varied from 0.0062 ± 0.0087 to 0.865 ± 0.14 mSv y⁻¹ and 0.052 ± 0.073 to 7.22 ± 1.16 mSv y⁻¹, respectively.
4. The inhalation and ingestion doses for well water varied from 0.022 ± 0.0025 to 0.41 ± 0.0054 mSv y⁻¹ and 0.18 ± 0.021 to 3.38 ± 0.045 mSv y⁻¹, respectively.
5. 33.33% of spring and 7.32% of well water samples were found with values above the recommendation levels of USEPA (~11.1 Bq L⁻¹).
6. Total AED due to WBRn was found to be 2.16 ± 0.008 mSv y⁻¹ and 0.89 ± 0.027 mSv y⁻¹, for spring and well water samples, respectively. These dose values are well above the safe limit (0.1 mSv y⁻¹) of AED recommended by World Health Organization (WHO 2011).
7. Mean value of ELCR for spring and Well water samples, was computed to be 7.56×10^{-3} and 3.13×10^{-3} , respectively, which are much higher than the upper bound of 0.1×10^{-3} for drinking water, as proposed by the USEPA (USEPA 2012).
8. It is inferred that 98% of the spring water and all of the well water samples have ELCR greater than world

standard value of 0.29×10^{-3} , whereas the USEPA, 90% and 65% of the respective samples have ELCR above worldwide average of 1.45×10^{-3} .

Nevertheless, the concentration levels of ²²²Rn in spring water were appreciably higher than Well water samples. This may be due to the reason that Well water continuously get accumulated over the passage of time and at the time of analysis Well water have undergone multiple radioactive decays. On the other hand, spring water was collected just as it oozed out from rock. The source of radon in Well and spring waters may be soil and rocks.

The current study suggests further investigation should be carried out to detect presence of other radionuclide's and heavy metals in drinking water samples. For provision of safe drinking water, physical, chemical and biological parameters should be analyzed. It is suggested that to reduce radon levels in drinking water aeration systems can play an effective role. Through aeration or agitation process radon escapes from water. It is time for government to implement regulations regarding levels of radon in water and air at national level. Current study results provide baseline data for future studies aimed at assessment of possible radon based water contamination in the region.

REFERENCES

- Akar, T. U., Gurler, O., Akkaya, G., Kilic, N., Yalcin, S., Kaynak, G. & Gundogdu, O. 2012 *Evaluation of radon concentration in well and tap waters in Bursa, Turkey*. *Radiation Protection Dosimetry* **150** (2), 207–212.
- Akerblom, G. & Lindgren, J. 1997 *Mapping of Groundwater Radon Potential* (IAEA-TECDOC-980). International Atomic Energy Agency (IAEA), Vienna, Austria.
- Armstrong, M. 1998 *Basic Linear Geostatistics*. Springer Verlag, Berlin, Germany.
- Bourgoignie, R. R., Lejeune, P., Poffijn, A., Sefaert, O. & Uyttenhove, J. 1982 On the Rn-222 and Ra-226 concentrations in water from the Pletrou source (Vise). *Ann. Belg. Ver. Stralingsbescherm* **7** (1), 5–16.
- Calkins, J. A., Offield, T. W., Abdullah, S. K. N. & Ali, S. T. 1975 *Geology of the Southern Himalaya in Hazara, Pakistan and Adjacent Areas*. US Geological Survey, 716-C, C1-29, Reston, VA, USA.
- Cantaluppi, C., Fasson, A., Ceccotto, F., Cianchi, A. & Degetto, S. 2014 *Radionuclides concentration in water and mud of Euganean thermal district*. *Environmental Research* **8**, 237–248.

- Corrêa, J. N., Paschuk, S. A., Kappke, J., Allan, F. N. P., Alana, C. F., Hugo, R. & Valeriy, D. 2014 Measurements of ^{222}Rn activity in well water of the Curitiba metropolitan area (Brazil). *Radiation Physics and Chemistry* **104**, 108–111.
- Cothorn, C. R., Lappenbusch, W. L. & Michel, J. 1986 Drinking water contribution to natural background. *Health Physics* **50**, 33–39.
- Crawford-Brown, D. J. 1989 The biokinetics and dosimetry of radon 222 in the human body following ingestion of ground water. *Environ. Geochem. Health* **11**, 10–17.
- Crawford-Brown, D. J. 1991 *Risk and Uncertainty Analysis for Radon in Drinking Water*. Final Report. American Water Works Association. Risk Analysis **11**, 135–1435.
- Cross, F. T., Harley, N. H. & Hoffmann, W. 1985 Health effect and risk from ^{222}Rn in drinking water. *Health Physics* **48** (5), 649–670.
- Dindaroglu, T. 2014 The use of the GIS Kriging technique to determine the spatial changes of natural radionuclide concentrations in soil and forest cover. *Journal of Environmental Health Science and Engineering* **12**, 130.
- Disaster Risk Management Plan, Azad Jammu & Kashmir 2008 *Report by State Disaster Management Authority Muzaffarabad*, 116 pp. <http://www.ndma.gov.pk/plans/Provincial%20DRM%20Plan%20AJK.pdf>.
- Environmental Protection Agency (EPA) 1994 *Report to the Congress on Radon in Drinking Water, Multimedia Risk and Cost Assessment of Radon*. EPA 811-R-94-001; US Government Printing Office, Washington, DC, USA.
- Environmental Protection Agency (EPA) 1999 Health risk reduction and cost analysis for radon in drinking water. *Federal Register* **64** (38), 9559.
- Erdogan, M., Eren, N., Demirel, S. & Zedef, V. 2013 Determination of radon concentration levels in well water in Konya, Turkey. *Radiation Protection Dosimetry* **156** (4), 489–494.
- Etuk, S., Antia, A. & Agbasi, O. 2017 Assessment and evaluation of excess lifetime cancer risk for occupants of university of Uyo permanent campus. *Nigeria International Journal of Physical Research* **5** (1), 28–35.
- Fonollosa, E., Peñalver, A., Borrull, F. & Aguilar, C. 2016 Radon in spring waters in the south of Catalonia. *Journal of Environmental Radioactivity* **151**, 275–281.
- Hopke, P. K., Borak, T. B., Doull, J., Cleaver, J. E., Eckerman, K. F., Gundersen, L. C. S., Harley, N. H., Hess, C. T., Kinner, N. E., Kopecky, K. J., Mckone, T. E., Sextro, R. G. & Simon, L. 2000 Health risks due to radon in drinking water. *Environmental Science & Technology* **34** (6), 921–926.
- Iqbal, S., Nasir, S. & Hussain, A. 2004 *Geological Map of the Nauseri Area, District Muzaffarabad, AJK*. Geological Survey of Pakistan, Islamabad, Geological Map Series, **6** (14).
- Krige, D. G. 1966 Two-dimensional weighted moving average trend surface for ore – evaluation. *Journal of the South African Institute of Mining Metallurgy* **66**, 13–38.
- Ladygiene, R., Mastauskas, A., Morkunas, G. & Gasiunas, K. 1999 Determination of ^{222}Rn concentrations in Lithuanian spa waters by liquid scintillation counting. *Czechoslovakian Journal of Physics* **49**, 473–478.
- Llerena, J. J., Cortina, D., Duran, I. & Sorribas, R. 2013 Impact of the geological substrate on the radiological content of Galician waters. *Environmental Radioactivity* **116**, 48–53.
- Matheron, G. 1970 The Theory of Regionalized Variables and Its Applications. *Ecole Nationale Supérieure des Mine* **5**, 212.
- Mills, W. A. 1990 Risk assessment and control management of radon in drinking water. In: *Radon, Radium and Uranium in Water* (C. R. Cothorn & P. Rebers, eds). Lewis Publishers, Chelsea, MI, USA, pp. 27–37.
- Misdaq, M. A. & Elharti, A. 1997 Study of the influence of the lithological and hydrogeological parameters of aquifers on the radon emanation from underground waters using solid state nuclear track detectors. *Journal of Radioanalytical and Nuclear Chemistry* **218** (2), 209–214.
- Musa, I. S. M. 2003 *Radon in Natural Waters, Analytical Methods; Correlations to Environmental Parameters; Radiation Dose Estimation; and GIS Applications*. Doctoral Thesis, Division of Radiation Physics, Department of Medicine and Care, Faculty of Health Sciences, Linköping University, S-581 85 Linköping, Sweden.
- Nasir, T., Matiullah Rafique, M. & Tahseen, R. 2015 Measurement of waterborne radon in the drinking water of Dera Ismail Khan city using active and passive techniques. *Nuclear Technology & Radiation Protection* **30** (2), 139–144.
- National Council on Radiation Protection and Measurements (NCRP) 1984 *Exposure From the Uranium Series with Emphasis on Radon and its Daughters*. Report No. 77, Bethesda, MD, USA.
- Nisar, A., Jalil ur, R., Rafique, M. & Tabassum, N. 2017 Age-dependent annual effective dose estimations of ^{226}Ra , ^{232}Th , ^{40}K and ^{222}Rn from drinking water in Baling, Malaysia. *Water Science & Technology: Water Supply* **18** (1), 32–39.
- NCRWACR 2011 *North Carolina Radon-in-Water Advisory Committee Report*.
- Oner, F., Yalim H, A., Akkurt, A. & Orbay, M. 2009 The measurements of radon concentrations in drinking water and the Yeşilirmak River water in the area of Amasya in Turkey. *Radiation Protection Dosimetry* **133** (4), 223–226.
- Ononugbo, C. P. & Awwiri, G. O. 2016 Evaluation of effective dose and excess lifetime cancer risk from indoor and outdoor gamma dose rate of university of Port Harcourt Teaching Hospital, Rivers State. *Scientia Africana* **15** (1), 33–40.
- Pressyanov, D., Dimitrova, I., Georgiev, S., Hristova, E. & Mitev, K. 2007 Measurement of radon-222 in water by absorption in Makrofol. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* **574**, 202–204.
- Salonen, L. 1988 Natural radionuclides in groundwaters in Finland. *Radiation Protection Dosimetry* **24**, 163–166. <https://doi.org/10.1093/oxfordjournals.rpd.a080263>.
- Sarma, D. D. 2009 *Geostatistics with Applications in Earth Sciences*, 2nd edn. Springer, Dordrecht, The Netherlands.

- Shafique, B., Rahman, S., Rafique, M. & Rahman, S. U. 2012 [Monitoring of 222rn/220rn concentrations at the work places of Muzaffarabad, Azad Kashmir](#). *International Journal of Physical Sciences* 7 (41), 5577–5584.
- Skeppstrom, K. & Olofsson, B. 2006 [A prediction method for radon in groundwater using GIS and multivariate statistics](#). *Science of the Total Environment* 367, 666–680.
- Somlai, K., Tokonami, S., Ishikawa, T., Vancsura, P., Gaspar, M., Jobbagy, V., Somlai, J. & Kovacs, T. 2007 [Rn-222 concentrations of water in the Balaton Highland and in the southern part of Hungary, and the assessment of the resulting dose](#). *Radiation Measurements* 42, 491–495.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2000 *Sources, Effects of Ionizing Radiation*. United Nations, New York, NY, USA, pp. 453–487.
- United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 2006 *Annex E*, United Nations, New York, NY, USA.
- USEPA 2012 *Edition of the Drinking Water Standards and Health Advisories*. United States Environmental Protection Agency, Washington, DC, USA.
- WHO 2008 *World Health Organization Guidelines for Drinking Water Quality. Incorporating First and Second Addenda*, 3rd edn. WHO Press, World Health Organization, Geneva, Switzerland.
- WHO 2009 *WHO Handbook on Indoor Radon: A Public Health Perspective*. World Health Organization, Geneva, Switzerland.
- WHO 2011 *Guidelines for Drinking-Water Quality*, 4th edn. World Health Organization, Geneva, Switzerland. www.dep.state.fl.us/water/wf/dw.

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