Radon contamination, risk evaluation, and their spatial distribution in groundwater of three selected northern districts

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ABSTRACT

Radon (^{222}Rn) has a ubiquitous nature in the environment and can pose serious health threats to living beings. Groundwater is the primary source of drinking and domestic uses for more than 3 million people in the area. The current study was conducted for investigation of groundwater for the ^{222}Rn concentration and associated health risks. For that purpose, the groundwater was examined for ^{222}Rn concentration using the RAD7 detector (Durridge Company, USA). Average concentrations of ^{222}Rn were highest at 10.8±3.6 Bq/L in the Lower Dir district and lowest at 6.39±0.98 Bq/L in the Upper Dir. The average concentration of ^{222}Rn was found below the threshold limit set by the US Environmental Protection Agency (USEPA) and the World Health Organization (WHO). However, the concentration in 30% of sampling sites in the Lower Dir showed ^{222}Rn concentrations higher than a recommended level of 11.1 Bq/L. This study evaluated the potential non-cancer risk and estimated lifetime cancer risk (ELCR) of various age groups via ^{222}Rn consumption in groundwater. The results showed ´s total annual mean exposure doses (EwTotal) values for infant of 43.7±10.2, 25.2±3.15, 27.0±1.68 μSv/a, children 33.5±7.80, 19.3±2.42, 20.1±1.29 μSv/a), and adults (32.3±7.52, 18.6±2.33, 20.0±1.24 μSv/a) via groundwater intake in Lower Dir, Upper Dir, and Chitral districts, respectively. Infants showed higher vulnerability to health issues due to ^{222}Rn

contamination in groundwater. ^{222}Rn concentrations showed a weak correlation with various physicochemical parameters in the groundwater.

Keywords: Annual mean exposure doses; Chitral district; estimated lifetime cancer risk; Lower & Upper Dir districts; non-cancer risk; Pakistan

1. Introduction

Radon (^{222}Rn) is a ubiquitous radioactive gas and a significant contributor to human exposure to ionizing radiation (Benà et al., 2024). ^{222}Rn is a key product of the radioactive decay of the uranium (U-238) chain produced in the earth´s crust (Wu et al., 2024). The ^{222}Rn isotope having a half-life of 3.83 days is potentially firm in nature among its other isotopes (Fan et al., 2023). ^{222}Rn naturally is occurs in rocks and soils emits α- particles and is soluble in water under pressure. Consequently, water interaction with uranium-rich soils contributes to a higher quantity of ^{222}Rn in different water sources (Grządziel et al., 2021; Muhammad and Haq, 2023). The level of ^{222}Rn concentration in groundwater is attributed to types of hosting rocks, porosity-permeability, active faults occurrence, and the nature of groundwater aquifers (Chen et al., 2024; Muhammad et al., 2024; Ullah et al., 2022; Zhang et al., 2024; Zhao et al., 2024). In addition, the ^{222}Rn concentration is also elevated in groundwater due to the presence of radium in the rocks and minerals such as phosphate-rich soils around it (Shu'aibu et al., 2021). Exposure to ^{222}Rn in a confined environment poses serious health issues (Bossew et al., 2020).

Radon has been recognized as the main source of natural radiation exposure to humans. The toxicity arises from human exposure to ^{222}Rn and its progeny, which emits alpha particles. These alpha particles are sources of high energy that directly damage DNA, producing oxidative stress, and cause chronic inflammation in lung tissues. Chronic exposure to even at low concentration of ^{222}Rn can result in genetic mutation and lung cancer (Seo et al., 2019; Sutradhar et al., 2023). ^{222}Rn availability in water has attributed to approximately 89% of lung cancer via inhalation and around 11% of stomach cancer due to its direct intake contaminated in water (Suresh et al., 2020). It can damage the respiratory tracts, and digestive organs and can cause leukemia in children (McLaughlin et al., 1993). According to USEPA, the maximum contaminant level (MCL) of ^{222}Rn concentration in drinking water is permissible at 11.1 Bq/L. The guidelines of the World Health Organization and the European Union (EU) recommend that its countries should keep a concentration of ^{222}Rn in water less than 100 Bq/L.

Recently, ^{222}Rn contamination and associated health risks in water have been reported in several studies from China (Zhao et al., 2021), India (Jamir et al., 2023), and Qatar (Manawi et al., 2023). The concentration of ^{222}Rn and associated health risks has been rarely reported in Pakistan (Ijaz et al., 2023; Muhammad et al., 2020). Data were evaluated using various model (Yao et al., 2024; Zhou and Liu, 2022; Zhou et al., 2023; 2022) and geospatial distribution (Gong et al., 2024). However, these studies were mostly limited to the eastern and southern regions of Pakistan. Higher ^{222}Rn concentrations in hot spring water have been reported from the Garam Chasma, Chitral, and suggested detail analyses of groundwater (Ullah et al., 2022). Therefore, the present study was aimed to examine ^{222}Rn concentrations in groundwater of Lower Dir, Upper Dir, and Chitral districts, northern Pakistan. In addition, ^{222}Rn concentrations were evaluated for non-cancer and excess lifetime cancer risk (ELCR). ^{222}Rn concentrations, non-cancer, and ELCR values were plotted on the geospatial distribution maps.

2. Materials and methods

2.1. Study area

The sampling areas for this study included Chitral, Lower Dir, and Upper Dir districts, located in the Khyber Pakhtunkhwa province of Pakistan (Fig. 1). Chitral is situated in the northernmost part of Pakistan along the Kunar River, also known as the Chitral River, which ultimately drains towards the Kabul River. Chitral shares its boundaries with the Ghizer district of Gilgit-Baltistan to the north, Swat to the southeast, and Dir to the west. Surrounded by the elevated mountain ranges of Karakoram and Hindukush, Chitral's elevation ranges from 4500 to 7600 m. The groundwater in the Chitral basin, sourced from rainfall, springs, glaciers, and snow melting, serves as the primary drinking water source for the local population (Baig et al., 2010). The Dir districts are situated in northern Pakistan, with elevations ranging from 1200 to 2800 m above sea level. The area receives an annual average rainfall of approximately 1469 mm, with 254 mm during winter and summer, respectively. Inhabitants utilize tube wells, hand pumps, and dug wells water for various purposes, including drinking. The districts host the Panjkora River, which originates from Kohistan in Upper Dir and eventually joins the Swat River (Hasan et al., 2015; Ullah et al., 2014b).

2.2. Geology

Geological settings are considered a dominant factor in the distribution of groundwater. Soil and rock composition, type, permeability, and porosity characterize infiltration and recharge of groundwater. The area is mostly covered by different types of metamorphic and igneous rocks. Metamorphic and igneous rocks have low permeability which makes hurdle for water infiltration to underlying aquifers. Similarly, clayey soil has played a big role in the stoppage of infiltration in the area. However, the area is also dominated by sedimentary rocks and high structural terrain which are usually characterized by high porosity and permeability. This allow high infiltration of water and also contribute to the escape of different materials from underlying bedrocks to the surface of water. The area is geologically diverse with the presence of multiple types of rocks and different kinds of soils (Sarwar et al., 2021; Sullivan, 1992; Ullah et al., 2014a).

2.3. Sampling procedure

A total of 46 samples were collected from groundwater comprising Lower Dir (n=23), Upper Dir (n=14), and Chitral (n=09), randomly selected within the study area. The exact coordinates of the sample locations were determined using a global positioning system (GPS, Garmin eTrex 10, New Taipei City, Taiwan). Standard protocols were followed during sample collection to prevent potential contamination. Pre-cleaned, specifically designed glass bottles (0.25 L) were used for the collection and analysis of ^{222}Rn. These bottles were washed with 15% nitric acid (HNO_3) before sample collection and then attached to the RAD7 detector equipment (Durridge company, Billerica, Massachusetts, USA, 2016) for the analysis of ^{222}Rn and calibration according to the method as adapted from the (Haroon and Muhammad, 2022).

2.4. Water analyses

The RAD7 offers various options for the analysis of ^{222}Rn in different natural mediums including water. This study followed the standard procedure for determining ^{222}Rn in water samples based on existing research works (Duggal et al., 2013; Yakut et al., 2013). The internal humidity of the RAD7 was maintained at (<9%) for each sample to ensure highly accurate measurements of ^{222}Rn. The RAD7 was set up with a 30-min measurement time for each water sample using the Wat 250 protocol, and an additional 30 min were allocated to lower the internal humidity of the RAD7 to a suitable level of less than 9% for the subsequent examination of the next sample (Haroon and Muhammad, 2022). The recommended calibration method for RAD7 was consistently employed at intervals to ensure the reliability and quality control of water sample measurements. Basic water quality parameters, such as pH, temperature, turbidity, oxidation-reduction potential (ORP), electrical conductivity (EC), and salinity were determined using a multiparameter analyzer (CONSORT 6030, Turnhout, Belgium).

2.5. Annual mean exposure doses

The non-cancer risks associated with ^{222}Rn consumption to human health can be divided into two pathways such as ingestion and inhalation. The annual mean exposure doses via the ingestion (EwIng) pathway are primarily determined as the water quantity consumed by an individual at a specific duration. Furthermore, water usage for bathing and other domestic purposes can help in escaping ^{222}Rn to indoor air which consequently increases individual exposure by inhalation (EwInh) pathway. Due ^{222}Rn ubiquitous presence in the environment makes it a potential threat to individual health (Isinkaye et al., 2023; Vitz, 1991). The EwIng and EwInh values were determined by the equations adapted from (UNSCEAR, 2000, 2008):

$$\mathrm{EwInh} = C_{\mathrm{RnW}} \times F \times O \times R \times \mathrm{DCF} \quad (1)$$

$$\mathrm{EwIng} = \mathrm{CRnW} \times \mathrm{Cw} \times \mathrm{EDC} \quad (2)$$

C_{RnW}: concentration of ^{222}Rn in each water sample (Bq/L), F: equilibrium factor between ^{222}Rn and its progenies (0.4), O: average annual indoor occupancy time per individual (7000 h/y), DCF: dose conversion factor for ^{222}Rn exposure (9 nSv/Bq/h/m^3), Cw: average annual water consumption for infants, children, and adults is 0.6, 0.8, and 1.3 L/day, respectively, R: ratio of ^{222}Rn in the air to that in water (10^{-4}). EDC: effective dose coefficient for ingestion (3.5 nSv/Bq). The total annual mean exposure doses from ^{222}Rn in water (EwTotal) of humans can be calculated by the sum of the annual mean exposure doses for inhalation and ingestion as follows (Muhammad et al., 2024):

$$\mathrm{EwTotal} = \mathrm{EwIng} + \mathrm{EwInh} \quad (3)$$

Radon ELCR was computed using the adapted equation (Muhammad and Haq, 2023):

$$\mathrm{ELCR} = H \times DL \times RF \quad (4)$$

where H: is the mean effective dose, DL: the average duration of life (70 years), and RF: The fatal cancer risk per Sievert ($5.5E10^{-2}$/Sv).

2.6. Statistical analyses

The ^{222}Rn data obtained from RAD7 was first analyzed for descriptive statistics with the help of MS Excel (2022) and then presented the data graphically by plotting it in a sigma plot software (ver.12.5, Systate Inc.). Spatial distribution maps of ^{222}Rn concentration and ELCR data were prepared with the help of interpolation (Kriging) techniques of Arc GIS (software ver.10.8.2).

3. Results and discussion

3.1. Radon concentration in water

The mean concentrations of ^{222}Rn in groundwater of Lower Dir, Upper Dir, and Chitral districts were found 10.8±3.6, 6.39±0.98 and 6.67±1.03 Bq/L, respectively (Fig. 2). The highest ^{222}Rn concentration of 22.6 Bq/L was noted in the sample collected from Ballambut in the Lower Dir district (Fig. 2). The mean concentrations of ^{222}Rn in the studied district were found below the recommended level of 11.1 Bq/L (USEPA, 1999) in studied groundwater. The concentration of ^{222}Rn in 100% of groundwater samples in the Upper Dir and Chitral districts was found within the threshold limit for safe drinking water purposes. However, 34% of groundwater samples collected from the Lower Dir surpassed the recommended level but within the WHO drinking water guidelines (100 Bq/L). Local geology, lithology, and geochemical properties of the Lower Dir could contribute to a higher concentration of ^{222}Rn in groundwater. This region is a tectonically active zone with high susceptibility to erosion, landslide, and mass movement making pathways for increasing ^{222}Rn levels in different environmental media including groundwater. The elevated concentration of ^{222}Rn was attributed to permeable and porous pathways resulting from geological activities including landslide movement, erosion, and bedrock fractures as reported by Ullah et al. (2022) and Muhammad et al. (2024). These types of geological activities help to increase the concentration of ^{222}Rn in water aquifer and the atmosphere from underlying bedrocks which consequently deteriorated water quality. Furthermore, bedrock types of aquifers play a significant role in the presence of excess ^{222}Rn. It is reported that granite rock aquifers possess a much higher concentration of ^{222}Rn than aquifers with sedimentary bedrocks (Duenas et al., 1998; Khan et al., 2022; Ullah et al., 2022).

The average concentration of ^{222}Rn in groundwater of the Lower Dir district was observed higher than that of 6.04±1.85 Bq/L in Kotli, AJK (Muhammad et al., 2024) and 1.82±0.04 Bq/L in Nagaland, India reported (Jamir et al., 2023) as shown in the Table 1. However, the average concentrations of ^{222}Rn reported by this study were noted below the mean concentration of 31.5±4.9 Bq/L observed in the water of the Zarand area, Iran (Mehnati et al., 2022), as shown in Table 1. The excessive concentration of ^{222}Rn in drinking water sources has potential health concerns for the dependent human populations. Human populations if exposed to the highly contaminated water with ^{222}Rn through ingestion and inhalation can face serious health issues such as cancer and non-cancer health detrimental issues.

3.2. Annual mean exposure doses

The annual mean exposure doses for non-cancer health risks of different age groups including infants, children, and adults were calculated for the consumption of groundwater contaminated with ^{222}Rn in the Lower Dir, Upper Dir, and Chitral districts. The values of EwIng for infants were 16.5±3.83, 9.80±1.19, 10.2±0.64 μSv/a, children 6.30±1.47, 3.73±0.45, 3.90±0.24 μSv/a, and adults 5.12±1.19, 2.95±0.37, 3.16±0.20 μSv/a via the groundwater consumption of Lower Dir, Upper Dir, and Chitral districts, respectively. Furthermore, the EwInh values were 27.2±6.33, 15.7±1.96, and 16.8±1.05 μSv/a in the groundwater of Lower Dir, Upper Dir, and

Chitral districts, respectively. Additionally, EwTotal values for infants were 43.7±10.2, 25.2±3.15, 27.0±1.68 μSv/a, children 33.5±7.80, 19.3±2.42, 20.1±1.29 μSv/a, and adults 32.3±7.52, 18.6±2.33, 20.0±1.24 μSv/a in the groundwater of Lower Dir, Upper Dir, and Chitral districts, respectively as shown in (Fig. 3abc). Among age groups, the infants were most vulnerable to the non-cancer health risk posed by ^{222}Rn excessive concentration in groundwater. The concentration of ^{222}Rn in the groundwater of the Lower Dir district posed a higher risk to the dependent community via both ingestion and inhalation. Exposure risk through inhalation was found more severe for infants compared to the ingestion route. The inhalation route posed a serious threat to different age groups for all the study areas compared to ingestion. The groundwater of the Lower Dir district has a significant potential for detrimental impacts on infants' health via ingestion and inhalation than other districts. Higher risk for infants, children, and adults were compared to previous studies (Ismail et al., 2021; Muhammad et al., 2020). However, the EwTotal values were noted as less than the previously conducted study on hot springs in Gilgit-Baltistan by Ullah et al. (2022) as shown in Table 2. According to WHO guidelines of drinking water for safe annual mean exposure dose limit, the water in the area was found below the recommended limits of 100 μSv/a for the human.

This study calculated the ELCR for adults of the population exposed to the ^{222}Rn via groundwater of Lower Dir, Upper Dir, and Chitral districts (Fig. 4ab). The US EPA set a maximum threshold of 1E10-4 for ELCR for adults, beyond this limit it has serious consequences on human health. Results revealed that ELCR values for the lower Dir, Upper Dir, and Chitral districts were less than the US EPA threshold except for a few instances in the Lower Dir district. This means that the groundwater of the area was 100% safe in the Upper Dir and Chitral districts in terms of ^{222}Rn contaminations. However, 30% of groundwater samples showed higher values that needs attention from the policymakers.

3.3. Water physical parameters and correlations

Basic water parameters such as Temperature, pH, EC, Turbidity, ORP, and TDS are important indicators of water quality and its status for different services to humans. This study examined various physical parameters of water to know the status of its quality and correlated to ^{222}Rn concentration in drinking water sources (Fig. 5). Basic water parameters determined were pH (7-7.6), temperature (18.5-30.1^{o}C), EC (303-1415 µS/m), salinity (0.15-0.71 µ/L), ORP (220-292 mV), and turbidity (0.76-2.8 NTU) for the water of Lower Dir and were found in the range for pH (6.4-7.8), temperature (14.4-26^{o}C), EC (49-477 µS/m), salinity (0.03-0.25 µ/L), ORP (253-306 mV), and turbidity (0.6-8.4 NTU) for the water of Upper Dir. The results showed that 15.3% of turbidity values surpassed the threshold limit of 5 NTU for water quality. The maximum variation was detected for EC in the water of Lower Dir and all the other parameters show a normal trend with gentle variation for the water of Lower and Upper Dir districts. Similarly, for the water of Chitral, basic water parameters values were 6.7-7.6, 17.1-23^{o}C, 105-1860 µS/m, 0.05-0.92 µ/L, 245-313 mV, and 0.94-4 NTU for pH, temperature, EC, salinity, ORP and turbidity, respectively. The variation in the minimum and maximum values for EC were pronounced in the water of Chitral and other parameters were found in normal trend. Statistical correlations were determined between the physical basic parameters of water and ^{222}Rn concentration which showed no strong correlation for all the parameters, except for ORP. This finding was in contrast to the previous study of (Muhammad and Haq, 2023). Where the dominant factor on the solubility of ^{222}Rn in the water was temperature and revealed strong positive correlation.

4. Conclusion

This study concluded that the average ^{222}Rn concentration in the groundwater of Lower Dir, Upper Dir, and Chitral districts was found within the threshold limits set by USEPA and WHO.

However, 30% of the Lower Dir district water samples were found with higher levels than recommended values, which can pose detrimental health impacts to the exposed human community. Results revealed higher values of EwIng, EwInh, and EwTotal via groundwater consumption to the infants than other age groups. Results showed that the inhalation pathway was the dominant way to affect the health of dependent populations of different age groups. The studied physiochemical parameters showed a weak correlation with ^{222}Rn concentration in the groundwater. The maximum average ELCR values were also noted for the Lower Dir district. Therefore, this study recommended the aeration or heating of groundwater especially for drinking purposes.

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Author Contribution S. Muhammad *Supervision, performed data analyses, drafted and edited the manuscript.* **R. Ullah & S. Amin** *data curation, data analyses, and edited the manuscript.* **A. Ahmad,** *Funding, technical support, performed data analyses and edited this manuscript.*

Data Availability *The data supporting this study's findings are available from the corresponding author upon reasonable request.*

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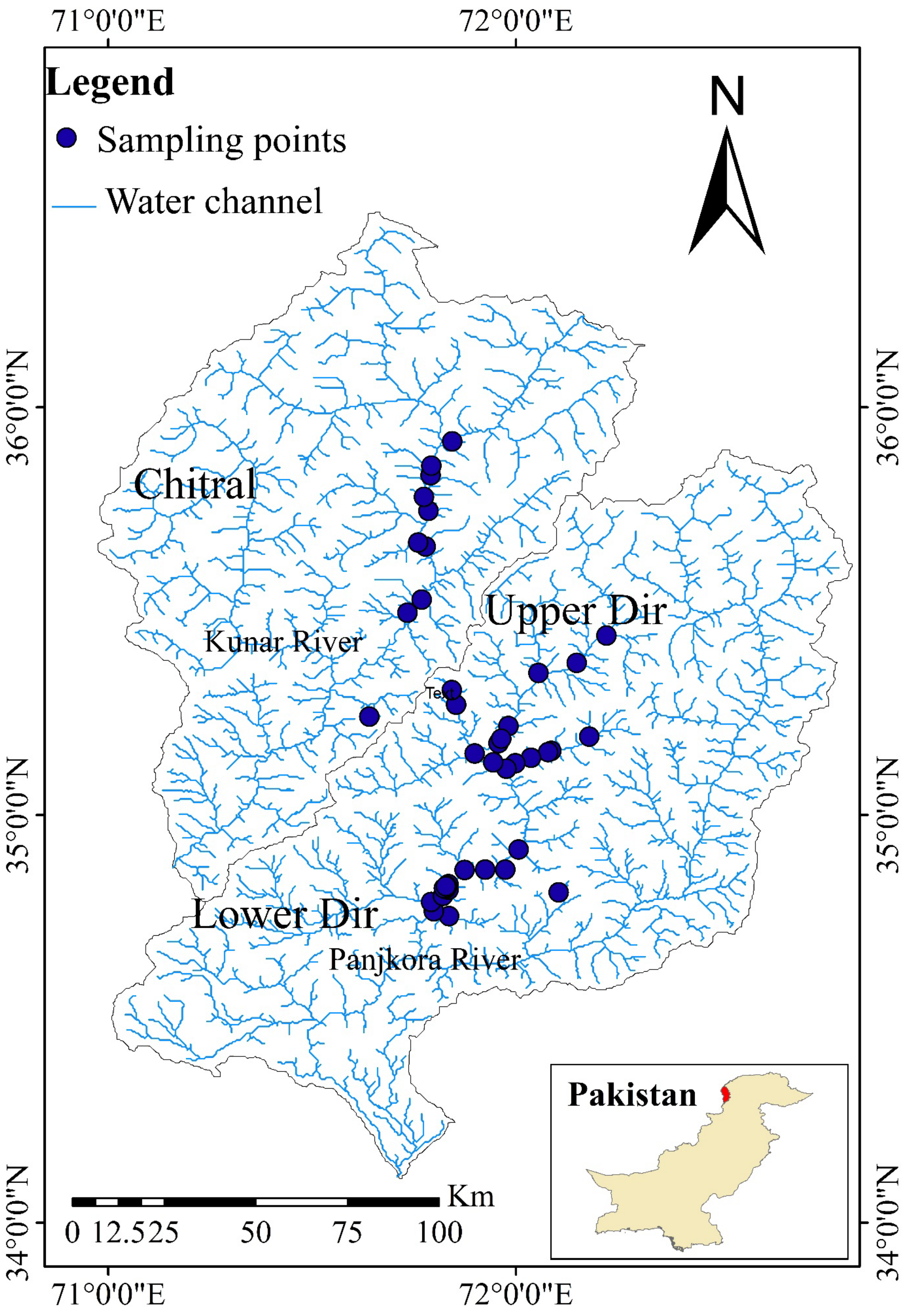
Table 1. Comparison of ^{222}Rn concentration (Bq/L) from the study area with studies conducted worldwide

Location	Country	^{222}Rn concentration	Study reference
Central Vietnam	Vietnam	107±0.91	(Duong et al., 2024)
Nagaland	India	1.82±0.04	(Jamir et al., 2023)
Zarand	Iran	31.5±4.912	(Rahimi et al., 2022)
Bauchi State	Nigeria	38.3±0.0	(Shu'aibu et al., 2021)
Bannu	Pakistan	28±13	(Shakoor et al., 2022)
Bibala	Angola	42±50	(Kessongo et al., 2020)
Dongpo	China	10.5±0.008	(Tan et al., 2019)
University Campus, Giresun	Turkey	27.28±0.98	(Büyükuslu et al., 2018)
Kotli, AJK	Pakistan	6.04±1.85	(Muhammad et al., 2024)
Ho Chi Minh City	Vietnam	0.11±0.01	(Le et al., 2015)
Karak	Pakistan	9.4±0.4	(Khattak et al., 2014)
Novi Sad	Serbia	1.59±1.05	(Todorovic et al., 2012)
Sudety Mountain area	Poland	1703±55	(KozŁowska et al., 2010)
Murree & Islamabad	Pakistan	4.38±0.44	(Ali et al., 2010)
Lower Dir	Pakistan	10.8±3.6	This Study
Upper Dir	Pakistan	6.39±0.98	This Study
Chitral	Pakistan	6.67±1.03	This Study

Table 2. Comparison of annual mean exposure doses of ^{222}Rn (μSv/y) in drinking water between Lower Dir, Upper Dir, and Chitral, Pakistan (current study) and the rest of the world studies

Location	Country	Annual effective doses (mSv/a)		Total	Previous studies
		Inhalation	Ingestion		
Akoko area	Nigeria	160.8	190.5	151.3	(Isinkaye et al., 2023)
Yalova Basin	Turkey	8.06	0.75	8.81	(Tabar and Yakut, 2014)
West Bank	Palestine	4.7	1.60	6.60	(Thabayneh, 2015)
Coonoor	India	0.0033	0.0069	0.0102	(Selvasekarapandian et al., 2002)
Mirpur, AJK	Pakistan	0.045	0.0038	0.049	(Muhammad et al., 2020)
Ashanti region	Ghana	29.72	41.6	71.32	(Asare-Donkor et al., 2018)
Peninsular	Malaysia	0.06	4.45	4.51	(Ismail et al., 2021)
Lower Dir	Pakistan	27.18	5.12	32.30	Current study
Upper Dir	Pakistan	15.66	2.95	18.61	Current Study
Chitral	Pakistan	16.81	3.16	19.97	Current Study



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461 **Fig. 1.** Sampling points of water in the Lower Dir, Upper Dir, and Chitral, northern Pakistan.

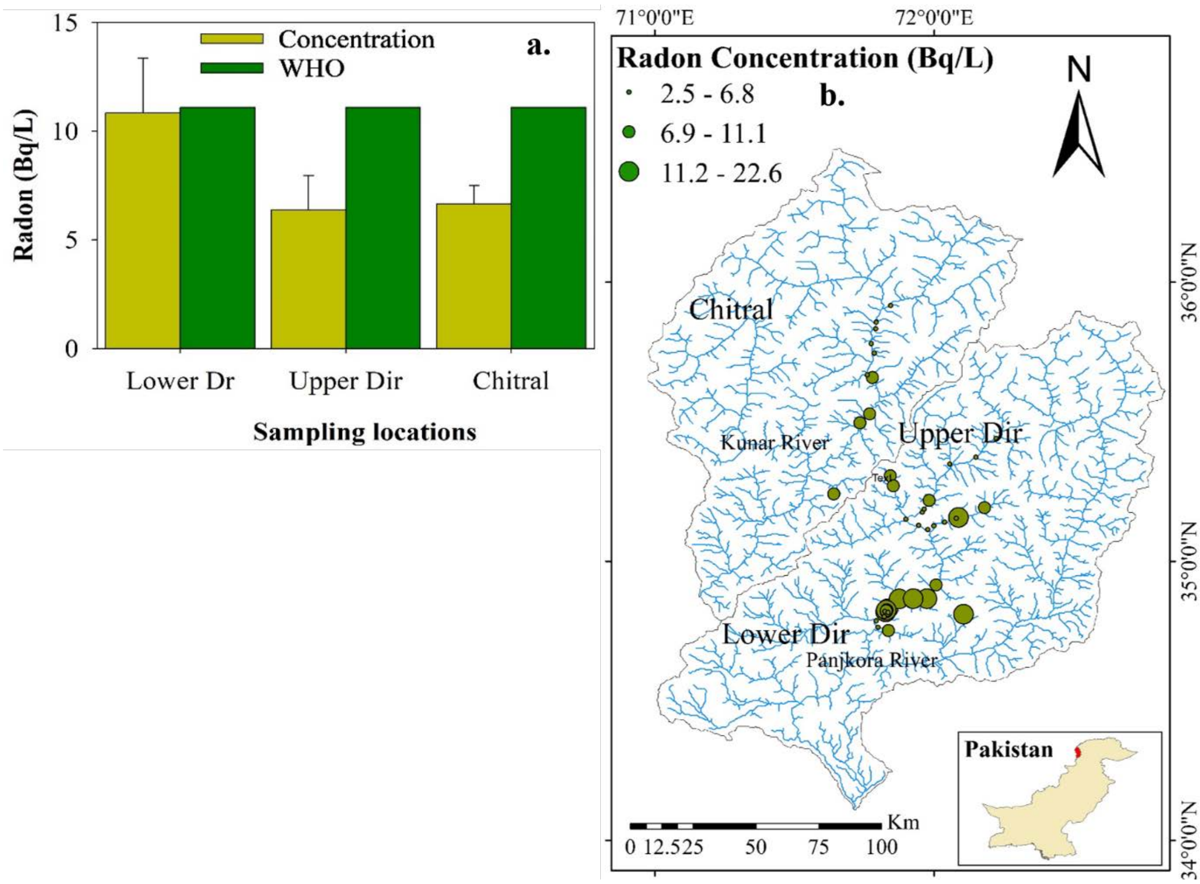

Fig. 2. Radon concentration (Bq/L) in sampling locations of Lower Dir, Upper Dir, and Chitral, northern Pakistan. **a.** Bar diagram of average concentration and b. spatial distribution of each sampling point.

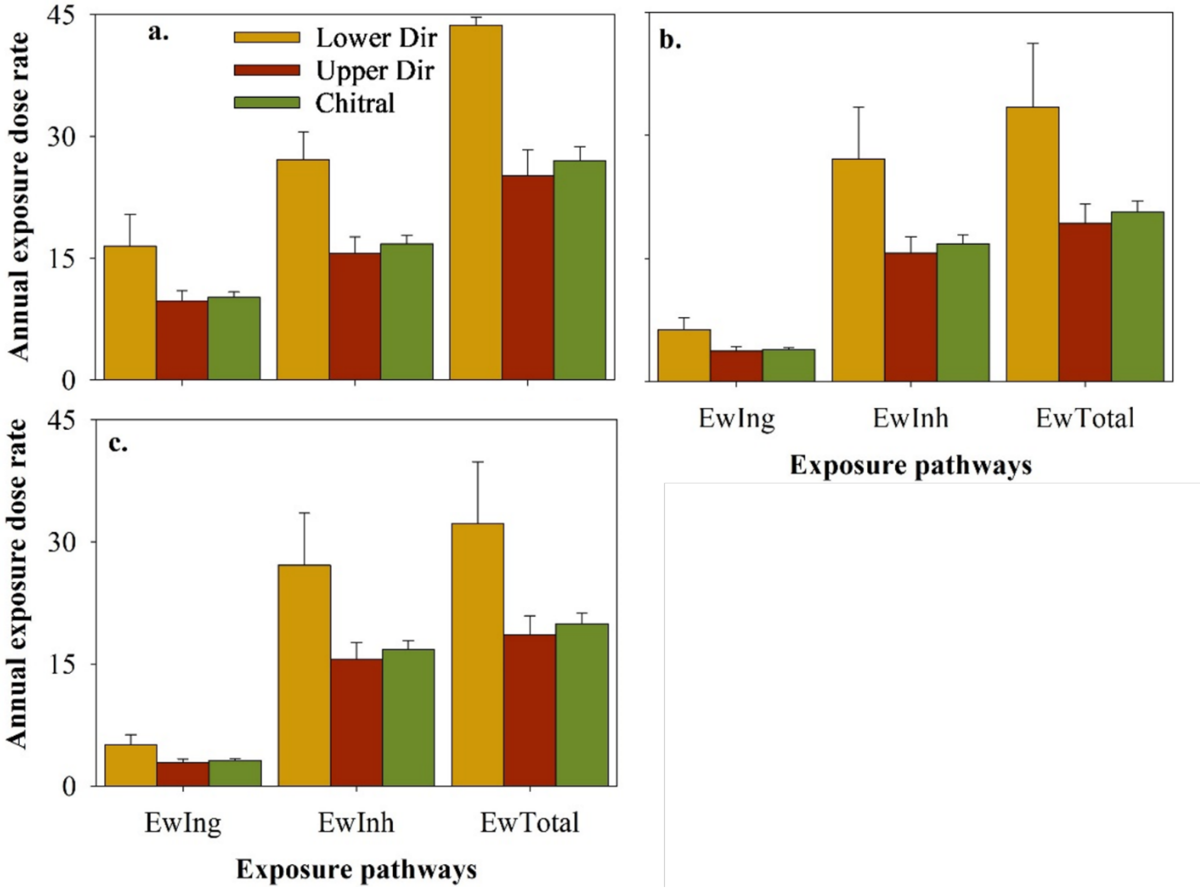

Fig. 3. Annual mean exposure dose rates (μSv/a): EwIng is the mean effective dose of ^{222}Rn ingestion, EwInh is the mean effective dose of ^{222}Rn inhalation, and EwTotal is the total mean effective dose of ^{222}Rn in Lower Dir, Upper Dir, and Chitral, northern Pakistan; **a.** infants, **b.** children, and **c.** adults.

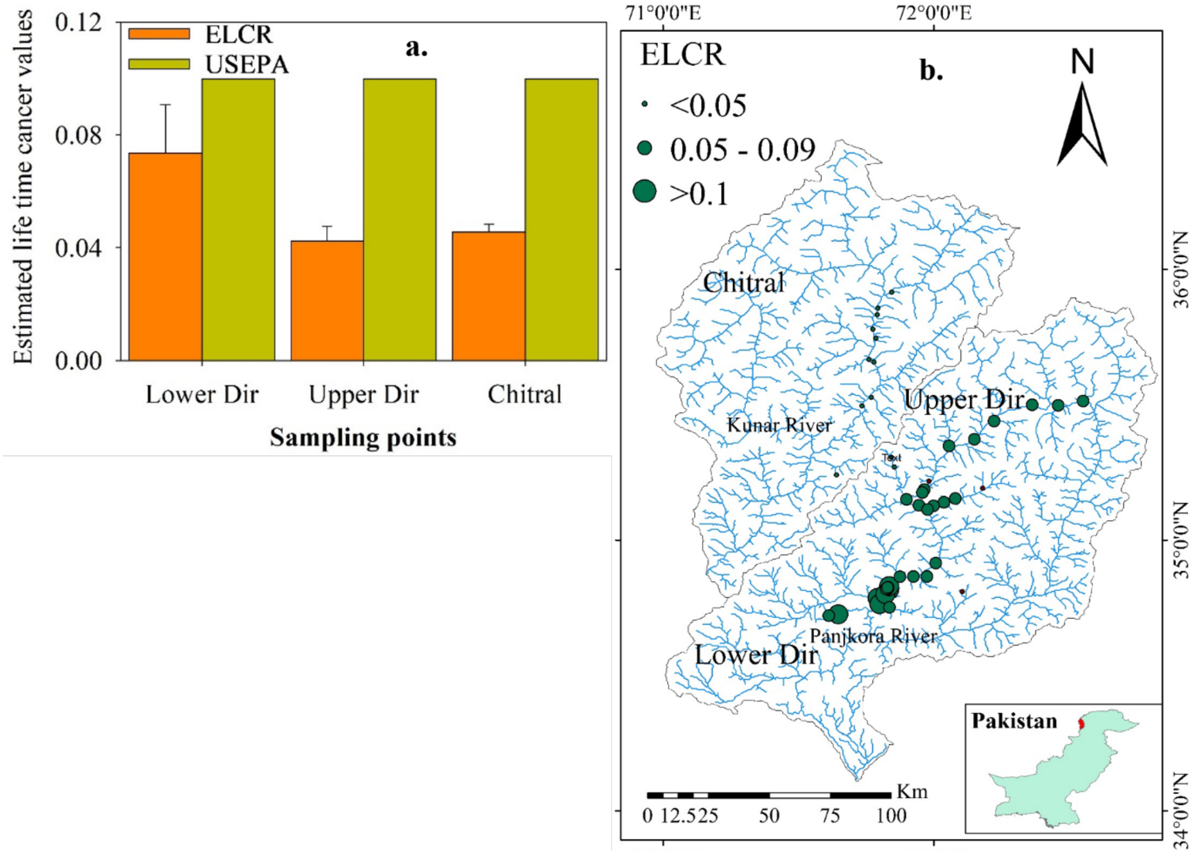

Fig. 4. ELCR through water consumption of Lower Dir, Upper Dir, and Chitral, northern Pakistan. **a.** Bar diagram of average ELCR (10^{-3}), **b.** spatial distribution of ELCR (10^{-3}) for each sampling point.

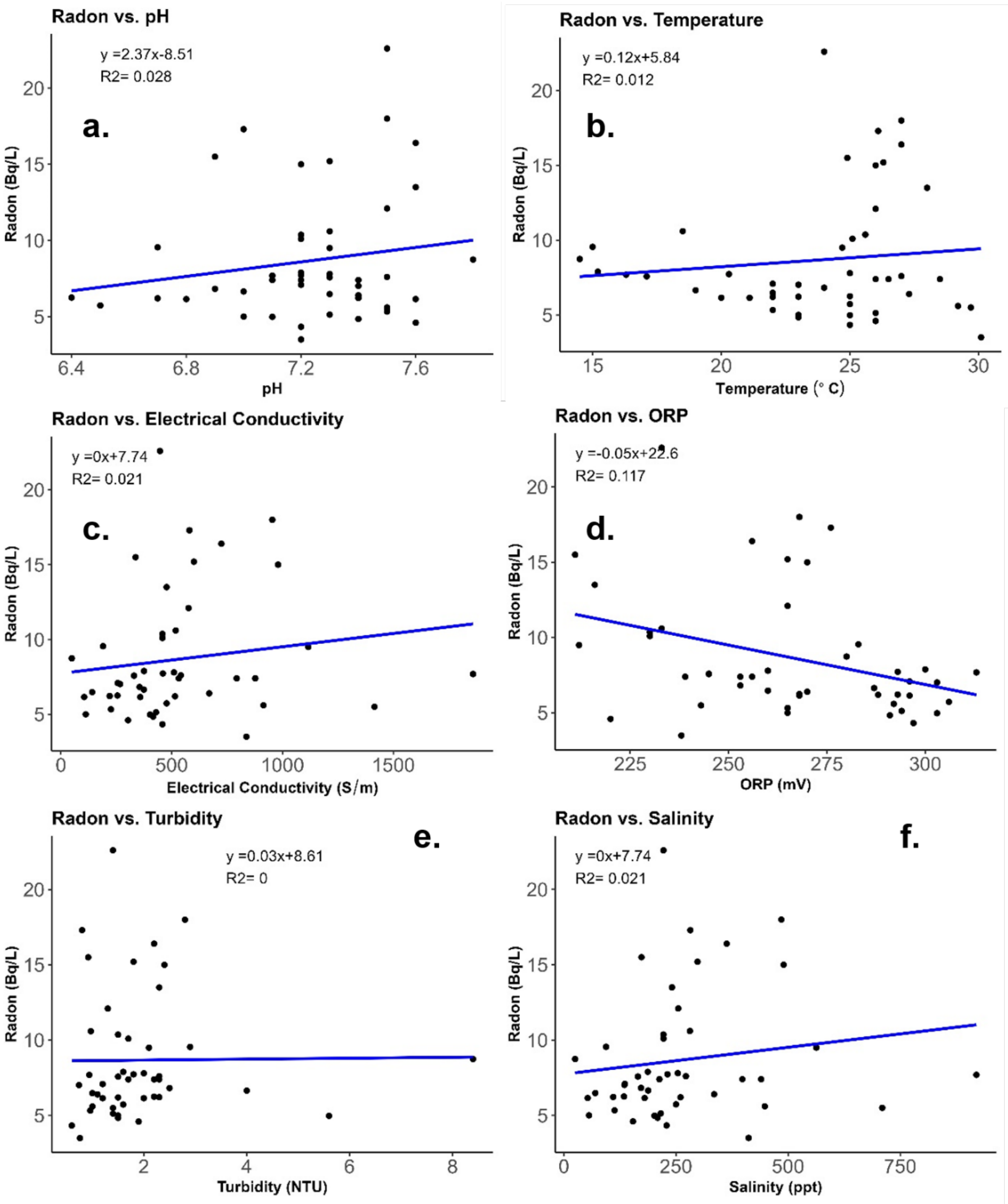

Fig. 5. Correlation of radon concentration with that of other water physical parameters, **a.** pH, **b.** Temperature, **c.** EC, **d.** ORP, **e.** Turbidity, and **f.** Salinity.