Spatiotemporal distribution of dissolved radon in uranium impacted aquifers of southwest Punjab

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Abstract

Salinity, water logging, high nitrate, fluoride and dissolved uranium concentration in drinking water of southwest Punjab has impacted the local population leading to health issues and additional burden on economy. Though it was known that both U and its daughter products especially Rn contribute to radiological dose to the population through drinking water, there were no correlation studies carried out between these radioactive elements in U impacted regions of southwest Punjab. In this study, an initiative has been taken to assess the doses due to dissolved radon in drinking water. In addition, the U–Rn couple is evaluated in detail along with other hydrochemical parameters. The radon concentration ranges from 360–1700 Bq/m^3 for Faridkot and 140–1400 Bq/m^3 for Muktsar for both seasons and the related average total dose due to radon for both season of Faridkot and Muktsar are 9.79 μS/year and 7.74 μS/year respectively. The total dissolved uranium is in range of 16–350 μg/L for Faridkot and 14–106 μg/L for Muktsar for both seasons. An inverse correlation was observed between Rn and U, which could be attributed to diverse geochemistry of U and Rn in groundwater.

Keywords Radon toxicity · Seasonality · Rn-TDS correlations · U contamination · Rn-U correlations

Introduction

Naturally present radionuclides are not of concern if present within the permissible limits, however concentrations above permissible limits pose serious health issues to mankind. The main processes governing the distribution of radioactive elements in groundwater are dissolution of minerals, weathering reactions under natural conditions as well as due to man-made activities such as contribution of fertilizers, flyash or wastes from nuclear industry. Radon is an odorless, colorless and tasteless inert gas that is generated through decay of U-Th elements present in the bedrocks.

Uranium-238 decays to radon-222 releasing four alpha and 2 beta particles. Among the radon isotopes, Rn-222 is the major contributor for health hazards owing to its relatively long half-life (3.8 days) compared to other isotopes i.e. ^{219}Rn $t_{1/2}$: 3.96 s and ^{220}Rn $t_{1/2}$: 55.6 s which decay much before accumulating in groundwater and migrating to farther distance. The occurrence of high concentration of dissolved radon in drinking water can pose a potential health risk to human due to exhalation of radon and its decay products from water into the household air and also due to direct ingestion of radon through drinking water [1]. It was reported that most of the dose results from inhalation rather than ingestion that is true in general, if we are considering inhalation of the radon emanating from the indoor building materials as well. However, if we are considering only water as the source of radon available for inhalation then the dose due to ingestion is more than inhalation. The major diseases associated with radon are lung and stomach cancer [2–4]. The solubility of radon in water is low, 0.01 mol/kg/bar at 293 K [5]. It degases as soon as it comes in contact with the air [6]. Mean concentration of radon in groundwater worldwide is 183 Bq/L [7]. USEPA suggested limit for radon in drinking water as 148 Bq/L [8]. Many studies were carried

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out in the world as well as in India on dissolved radon in drinking water. The dissolved radon and uranium concentrations in Juban district, Yemen ranged from 1.0 ± 0.2 Bq/L to 896 ± 0.8 Bq/L and 0.33 ± 0.01 μg/L to 24.6 ± 0.6 μg/lL respectively. The study indicated that higher concentration of U and ^{222}Rn were found to be from the basement aquifer, while the lowest concentration of both radionuclides were from the sandstone aquifer and high concentrations of Rn were found along the fault zones. Strong relationship between concentrations of ^{222}Rn with that of U and salinity of water were also observed [9]. In Northeastern Hungary radon concentrations of 100 Bq/L to 350 Bq/L was reported by Kasztovszky et al. [10]. The reported values in central Tamil Nadu are in the range of 0.10–211.60 Bq/L and the main reasons for high concentration of radon and U were redox conditions and fracture systems [11]. Another study from Mandya region in Karnataka showed high radon concentration in groundwater mainly around the granitic rock exposures with values in range of 6.44 ± 0.20 to 44.83 ± 0.54 Bq/L [12]. Groundwater in Fatehabad region of Haryana showed radon concentrations in the range of 1.4 ± 0.6 Bq/L to 22.6 ± 2.4 Bq/L where 86% of the collected samples were found within the permissible limits [13]. Kumar et al. in 2016 reported dissolved radon ranging from 5.4 to 20.1 Bq/L in Jammu and Kashmir [14]. Radon concentrations in soil and water of Malwa belt of Punjab was found to vary from 1.9 to 16.4 kBq/m^3 and 5.01 kBq/m^3 to 11.6 kBq/m^3 respectively [15].

High concentration of uranium in drinking water has also shown negative impacts on human health. It is nephrotoxic causing kidney damage [16]. Since chemical toxicity of uranium is more compared to radiological toxicity [17], WHO has prescribed a guideline value of 30 μg/L uranium in drinking water [18]. Southwest part of Punjab is already reported with high U in groundwater [19–21] and hence measurement of Rn, which is a decay product of U also assumes significance due to its implications on public health. In this paper, we report the dissolved radon values and their variation in space and time, estimate radiation dose due to dissolved radon, discuss general water quality aspects and evaluate the correlation of Rn with U and other hydrochemical parameters.

Study area description

The study area is bounded by four districts of Punjab in east, west and north (Bathinda, Fazilka, Moga and Ferozpur) while in south is the state of Rajasthan. The sediments of the alluvial plains belong to Recent to Sub-Recent age and are classified into two unit i.e. newer alluvium and older alluvium. These alluvial deposits were formed by the sediments transported from adjoining areas comprising Siwaliks, granites and other metamorphic rocks [22]. The groundwater is the main resource for domestic and irrigational purposes in the study area. Intense irrigation practices have led to depletion of groundwater near to one meter per year in major parts of the Punjab state [23]. The study area covers two districts of Punjab state, viz., Muktsar district and Faridkot district (Fig. 1).

Muktsar district lies between the latitudes 29°54′20″ and 30°40′20″N, longitudes of 74°15′ and 74°16′E. It is divided into 4 blocks namely Sri Muktsar Sahib, Lambi, Malout and Gidderbaha. It falls on main Indus basin and Satluj sub-basin with an area of about 2630 km^2, with no river flowing through it. It is mainly fed by canal network of Sirhind feeder. The slope of the district is north-east to south-west with an average elevation of 200 m amsl [24]. It has a dry sub-humid type climate with annual average rainfall of 431 mm. The period of 2004–2010 observed an average annual value of 267 mm [25], which was less compared to period of 2012–2016 (375 mm) [26, 27]. 79% of its rainfall occurs during the south-west monsoon and 21% from western disturbance and thunderstorm. The alluvial plain occupuies major part of the district.

Faridkot district lies between the latitudes 29°54′00″ and 34°35′00″N, longitudes 74°15′ and 75°25′E on Satluj basin with an area of about 1419 km^2, with no river flowing through it. It is divided into 2 blocks namely Faridkot and Kotkapura. It is mainly fed by Sirhind canal (~228 km long) with an average elevation of 204 m amsl [28]. It has a semi-arid hot climate with annual average rainfall of 449 mm. The period of 2004–2010 observed average annual value of 275 mm [25] which was less compared to period of 2012–2016 (458 mm) [27]. 78% of its rainfall is from the south-west monsoon and 22% from western disturbance and thunderstorm. 89% of the district area is under cultivation and is mainly irrigated by canal water since the groundwater is highly saline.

Methodology

Samples (twenty nos.) were collected from dug wells, tube wells and hand pumps from the study area (Fig. 1). The depth from which samples were collected from Faridkot district are in the range of 5–91 m bgl and for Muktsar district it is in the range of 8–21 m bgl. To get representative sample the wells were flushed until a constant temperature was obtained and then water sample were collected. In the field physio-chemical parameters i.e. electrical conductivity (EC), total dissolved solids (TDS), pH, temperature were measured using potable water quality meter (Hanna make). Samples for total uranium measurement were collected in
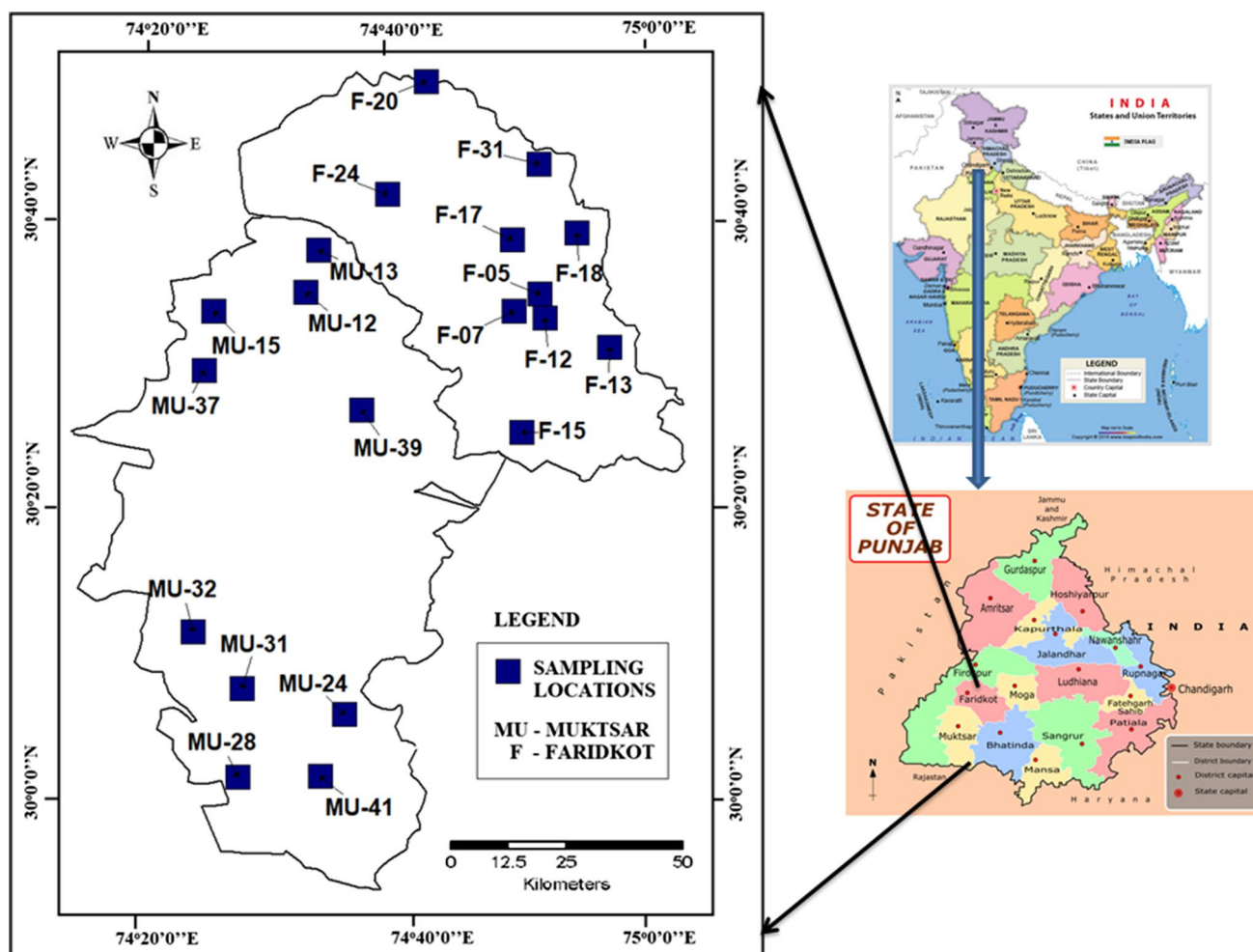
Fig. 1 Study area and sampling locations

acid-leached bottles (20 ml Tarson make). Before collecting the sample for measurement, bottles were thoroughly rinsed using sample water.

Measurement of uranium was carried out by fluorimeter (UA1, Quantalase), which measures fluorescence of uranyl phosphate complex that is formed by addition of sodium pyrophosphate to the sample [29]. The phosphate complex is preferred because of its stablity and enhanced fluorescence. The standard addition method was followed for the measurement of dissolved uranium. This method helps in nullifying the effect of chemical quenching by the interfering ions like iron. The sample was counted repeatedly four times and the average value was taken as the representative value. The error reported is the standard deviation (1 σ) of the four readings. The typical detection limit is 0.2 µg/L. The detailed procedure for measurement and validation is described in Rishi et al. [21]. A suitable dilution was done in the case of sample containing high uranium content.

Radon samples were collected through a pipe with one end inside the pump while the other end was placed at the bottom of the glass bottle. The glass bottles were dipped in bucket filled with water and water was allowed to overflow through the bottle for 3 to 4 volume exchanges to avoid loss of gas through degassing. Radon was measured using indigenous Smart Radon Monitor (SRM) on site. The principal of SRM is based on scintillation caused by radiation in ZnS:Ag crystal which is then detected by PMT. The gas is bubbled out of water sample by purging the bottle and with the help of pump it is carried into scintillation cell where radiation is detected. Before taking the instrument to the field the instrument was calibrated using a standard source and the calibration factor was added to the system for auto correction of the measured values in the field. Each sample was counted in six cycles and the average value of the counts was multiplied with the calibration factor to get the representative dissolved radon value in Bq/m^3. The error reported is the standard deviation (1 σ) of the six readings. The sensitivity of the

instrument is 1.2 CPH/Bq/m^3 with low detector volume of about 150 cm^3 [30]. Instrument sensitivity is free from the influence of environmental parameters like humidity and trace gas concentration. Minimum detection limit of the instrument is 14 Bq/m^3 at 95% confidence for 1 h counting. The location map and contour map were prepared using Map info 8 professional and the correlation plots were made using ORIGIN 6.1 plotting software.

Results

The physico-chemical and environmental isotope data is presented in Table 1

From the Table 1 it can be observed that most of the samples are saline in nature with TDS values more than 500 mg/L (Table 1) except F-18 with TDS value of 407 mg/L (Figs. 2 and 3). Most of the samples are above the permissible limit for drinking purpose as per BIS and WHO guidelines [31, 32]. The pH of the samples ranges from 6.6 to 8.2 and is within the WHO and BIS specified permissible range of 6.5–8.5. Two samples from Faridkot district (F-13 and F-18) are alkaline in nature with pH value of 8.7 and 9.3 respectively, which can be due to the influence of domestic wastes near that area (Figs. 2 and 3). The temperature of samples

are in range of 25–32 °C with no significant seasonal variation observed. The deeper groundwater was found to be warmer compared to shallow groundwater.

Rn dose calculation

The dose from radon was calculated for both ingestion and inhalation. The formula for dose due to ingestion is as follows [33]:

$$E = K \times C_{Rn} \times K_m \times t \tag{1}$$

where E is effective dose from ingestion, K is ingestion dose conversion factor of ^{222}Rn, C_{Rn} is measured concentration of radon, K_m is water consumption and t is duration of consumption of water. For inhalation, the parameters used are, (a) ratio of radon in air to radon in tap water supply, (b) average indoor occupancy time per person, (c) equilibrium factor between radon and its progeny and (d) dose conversion factor for radon exposure.

The total dose due to radon ranges from 3.6 to 16.58 μSv/year for Faridkot with an average value of 9.79 μSv/year while it ranges from 1.41 to 14.04 μSv/year for Muktsar with an average value of 7.74 μSv/year. The average dose of radon due to ingestion is 7.28 μSv/year for Faridkot and 5.75 μSv/year for Muktsar, which is more than that from

Table 1 Physiochemical and isotope data of the measured samples

I.D.	Type	Depth (m bgl)	Temp. (°C)	pH	TDS mg/L	Uranium μg/L	Radon Bq/m^3	Error Bq/m^3	Season
Faridkot									
F-05	BW	31	28	7.3	1155.08	66.25	1688.33	64.39	Postmonsoon
F-12	TW	91	28.96	7.86	1066.64	41.6	950	41.29	
F-15	TW	31	28.22	7.87	2816.68	124.87	1379.67	61.78	
F-17	TW	18	26.49	8.2	2180.85	19.2	1220	39.86	
F-20	HP	11	27.4	7.1	935.32	15.84	1132.33	57.77	
F-24	HP	5	27.08	6.6	2680	83.7	1363.33	65.63	
F-07	BW	61	28.5	8.01	1584.55	259.75	367	30.15	Premonsoon
F-13	BW	49	27.57	8.7	1308.51	353.45	839	46.05	
F-18	BW	46	25.8	9.3	407.36	278.01	476.33	36.88	
F-31	HP	34	25.99	7.62	1090.76	221.16	555	55.75	
Muktsar									
MU-12	HP	14	25.75	7.24	574.19	29.74	949	52.58	Postmonsoon
MU-15	BW	14	26.8	7.09	2795.91	36.39	700	32.95	
MU-28	HP	12	27.56	7.3	2308.82	54.32	1045	42.92	
MU-31	HP	14	26	6.7	3569.76	36.45	1429.67	64.51	
MU-32	HP	15	27.61	6.9	1116.22	14.01	1258.67	60.63	
MU-13	BW	8	26.95	7.7	4745.61	105.37	774	51.57	Premonsoon
MU-24	HP	21	28.4	8.09	3904.09	90	649	40.64	
MU-37	BW	12	28.79	7.8	1791.58	84.43	217.67	21.49	
MU-39	BW	12	26.4	7.12	1593.93	47.11	714.33	42.76	
MU-41	BW	15	31.65	7.77	1508.84	67.78	143.33	23.23	

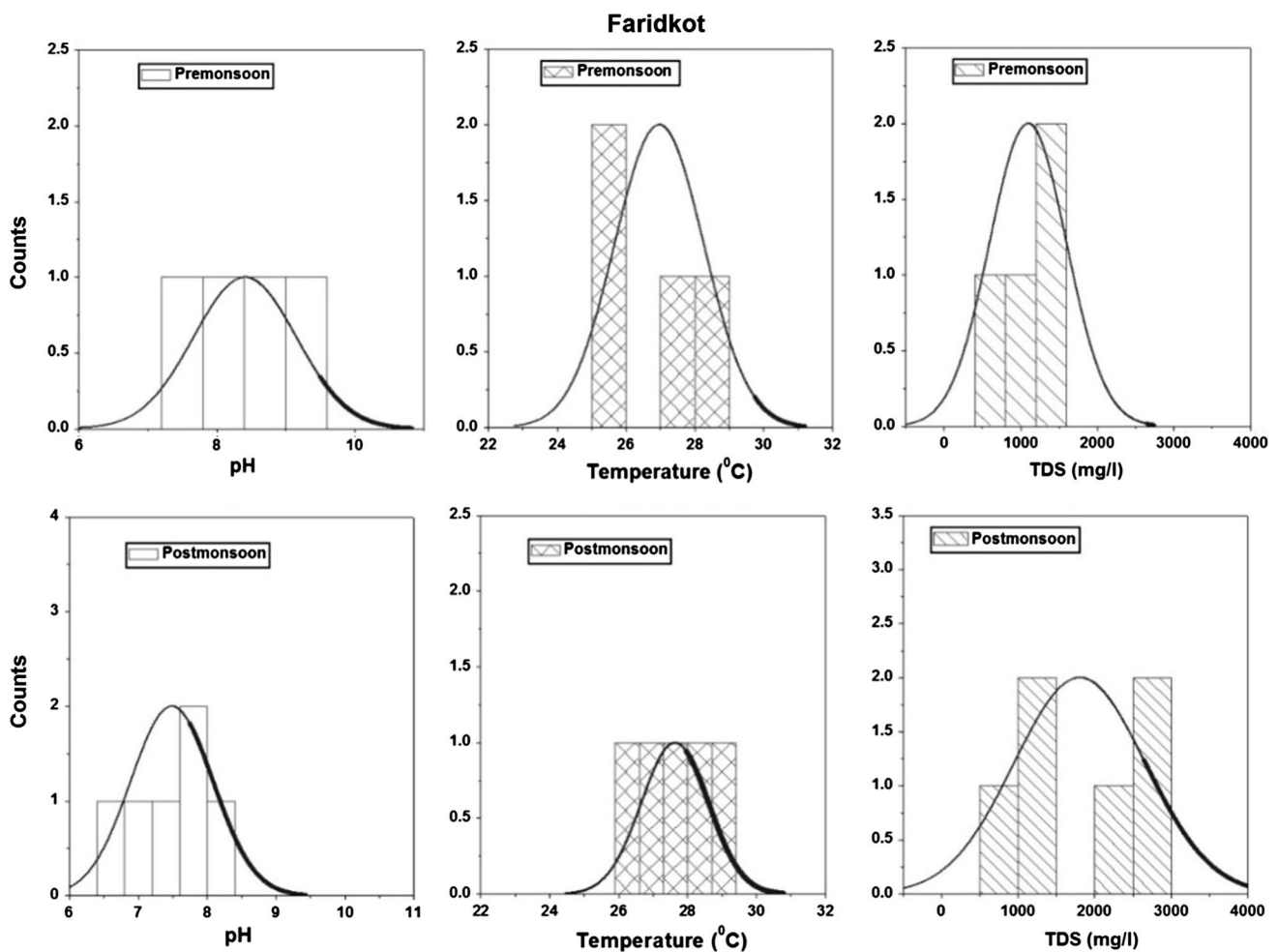**Fig. 2** Histograms of Temperature, pH, TDS for both season of Faridkot district

average inhalation dose of 2.51 μSv/year and 1.99 μSv/year for Faridkot and Muktsar district respectively (Table 2).

The averaged doses for premonsoon season due to ingestion are 4.08 μSv/year and 3.65 μSv/year and due to inhalation are 1.41 μSv/year and 1.26 μSv/year for Faridkot and Muktsar districts respectively. While for postmonsoon season, averaged doses due to ingestion are 9.41 μSv/year and 7.86 μSv/year and due to inhalation are 3.25 μSv/year and 2.71 μSv/year for Faridkot and Muktsar respectively. The seasonal fluctuation can be clearly seen from the above calculated values i.e. doses are higher from both inputs i.e. ingestion as well as inhalation for postmonsoon season. This is due to release of soil-pore trapped radon into the infiltrating water after monsoon period.

Discussion

Rn and U spatial variations

The radon concentrations for Faridkot district range from 367 to 839 Bq/m^3 for premonsoon season and 950–1688 Bq/m^3 for postmonsoon season. The radon values in Muktsar district range from 143 to 774 Bq/m^3 for premonsoon and 700–1430 Bq/m^3 for postmonsoon respectively. The dissolved Rn obtained from the groundwater of this study area was found to be less compared to the groundwater collected from Juban district, Yemen (1.0 ± 0.2 Bq/L to 896 ± 0.8 Bq/L) [10], Northeastern Hungary (100 Bq/L to 350 Bq/L) [11] and Tamil Nadu (0.10 to 211.60 Bq/L) [12]. The concentrations were comparable to Fatehabad, Haryana [14] and Jammu and Kashmir [15]. An increase in the Rn concentration was observed in groundwater during postmonsoon season which could be due to dissolution of radon from vadose zone with infiltrating rainwater.
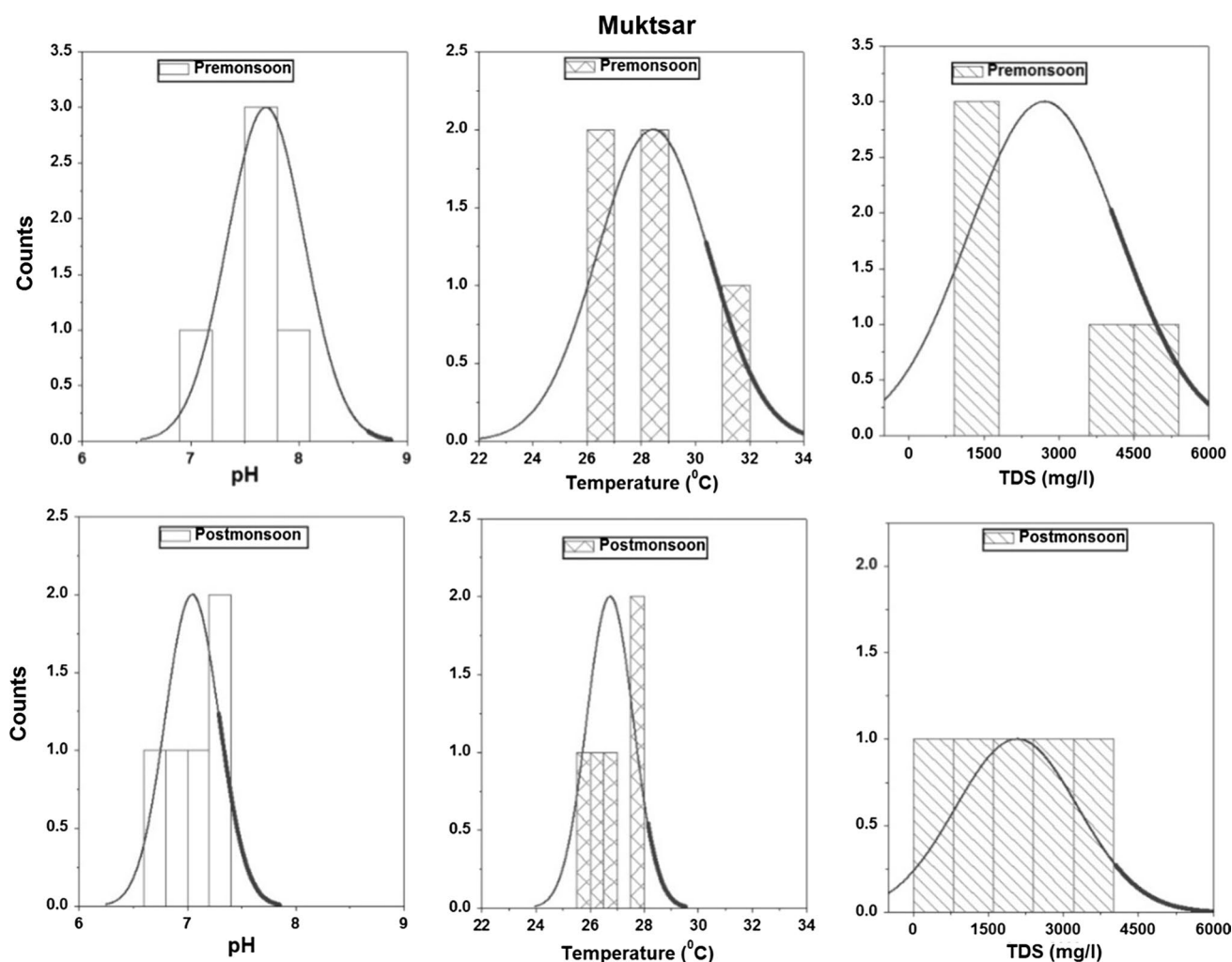
Fig. 3 Histograms of temperature, pH, TDS for both season of Muktsar district

The concentrations of uranium in groundwater of Faridkot district (Fig. 4) are in range of 221–354 μg/L for premonsoon season while it is in range of 16–125 μg/L for post monsoon season. In groundwater of Muktsar district (Fig. 5) the U concentrations are in the range of 47–105 μg/L for premonsoon and 47–54 μg/L for postmonsoon. Both districts showed higher U concentration in premonsoon period compared to postmonsoon period. This trend is found to be opposite of Rn concentration. This reduction in U values in postmonsoon indicates the dilution of U in groundwater by freshly added rainwater.

The lithological source for uranium was found to be local sediments that are derived from rock weathering in a study. Singh et al. reported that soil uranium concentrations are in the range of 1.03–9.82 ppm [34]. The radon exhalation rate

in these collected samples has been found to vary from 1.03 to 8.83 mBq/kg/h [34]. Radon being a decay product in U series, it gets emanated from the sediments and dissolves in groundwater. Authors also found a positive correlation between the radon exhalation rate and the uranium concentration of soil samples [34].

Since the data points are not sufficient to evaluate the seasonal variation of Rn and U, both seasons data was plotted together for preparing contour diagrams (Fig. 6). An inverse relationship was observed between uranium and radon concentrations in the study area i.e. areas with high U showed low Rn and vice versa.

Table 2 Annual effective doses due to presence of Radon

I.D.	Annual effective dose due to ingestion	Annual effective dose due to inhalation	Total dose
	μSv/year	μSv/year	μSv/year
F-05	12.32	4.25	16.58
F-12	6.94	2.39	9.33
F-15	10.07	3.48	13.55
F-17	8.91	3.07	11.98
F-20	8.27	2.85	11.12
F-24	9.95	3.44	13.39
F-07	2.68	0.92	3.60
F-13	6.12	2.11	8.24
F-18	3.48	1.20	4.68
F-31	4.05	1.40	5.45
MU-12	6.93	2.39	9.32
MU-15	5.11	1.76	6.87
MU-28	7.63	2.63	10.26
MU-31	10.44	3.60	14.04
MU-32	9.19	3.17	12.36
MU-13	5.65	1.95	7.60
MU-24	4.74	1.64	6.37
MU-37	1.59	0.55	2.14
MU-39	5.21	1.80	7.01
MU-41	1.05	0.36	1.41

Vertical distribution of U, Rn and physiochemical parameters

Depth profile of temperature shows an increase in temperature with depth while TDS was found to decrease with increasing well depths. pH of the groundwater doesn't not show any definite pattern with depth (Fig. 7).

Due to limited number of samples collected from the study area, the depth profile was plotted after combining all the data for both seasons of Muktsar and Faridkot districts. Figure 8 shows scatter in values of Rn and U for both seasons where dissolved radon show increase in concentration in postmonsoon season compared to premonsoon for both the districts. This is justified as vadose zone contains trapped radon gas which is decay product of parent nuclide (uranium). This radon from vadose zone emanates into the infiltrating water during the postmonsoon season and hence groundwater during postmonsoon season shows increased values for radon concentration. On the contrary, dissolved uranium decreases in postmonsoon season due to dilution caused by infiltrating rainwater.
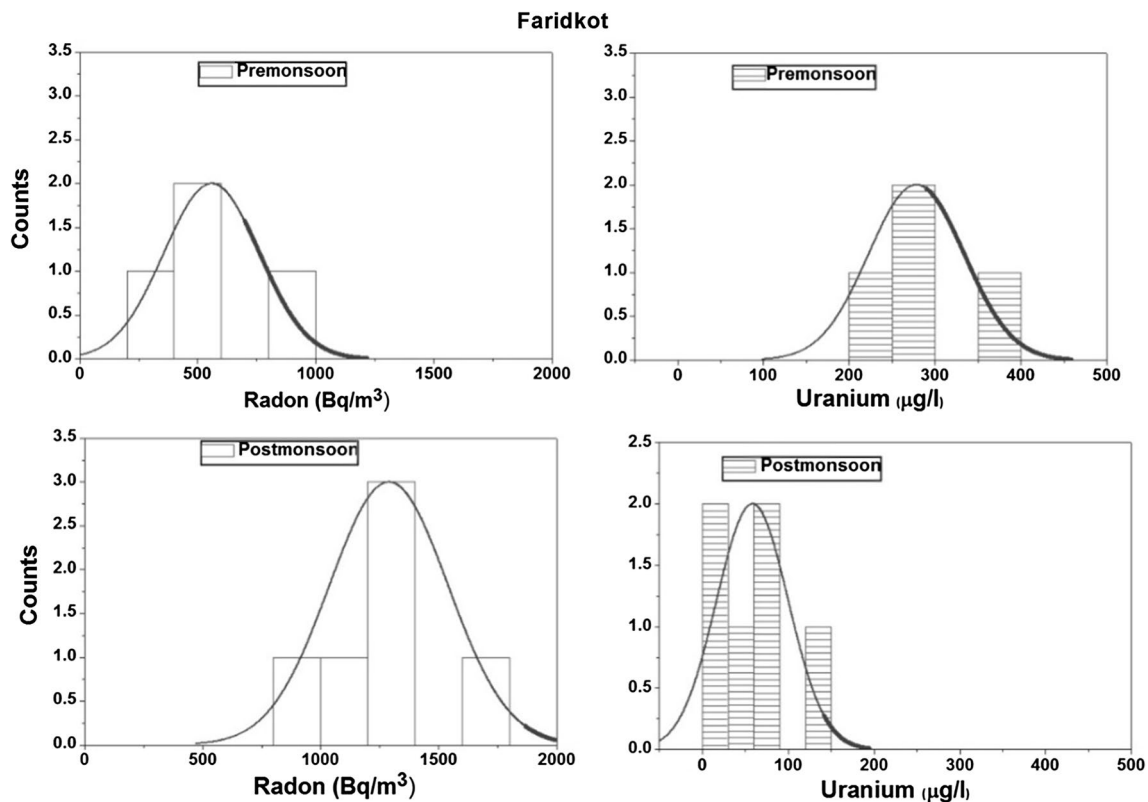
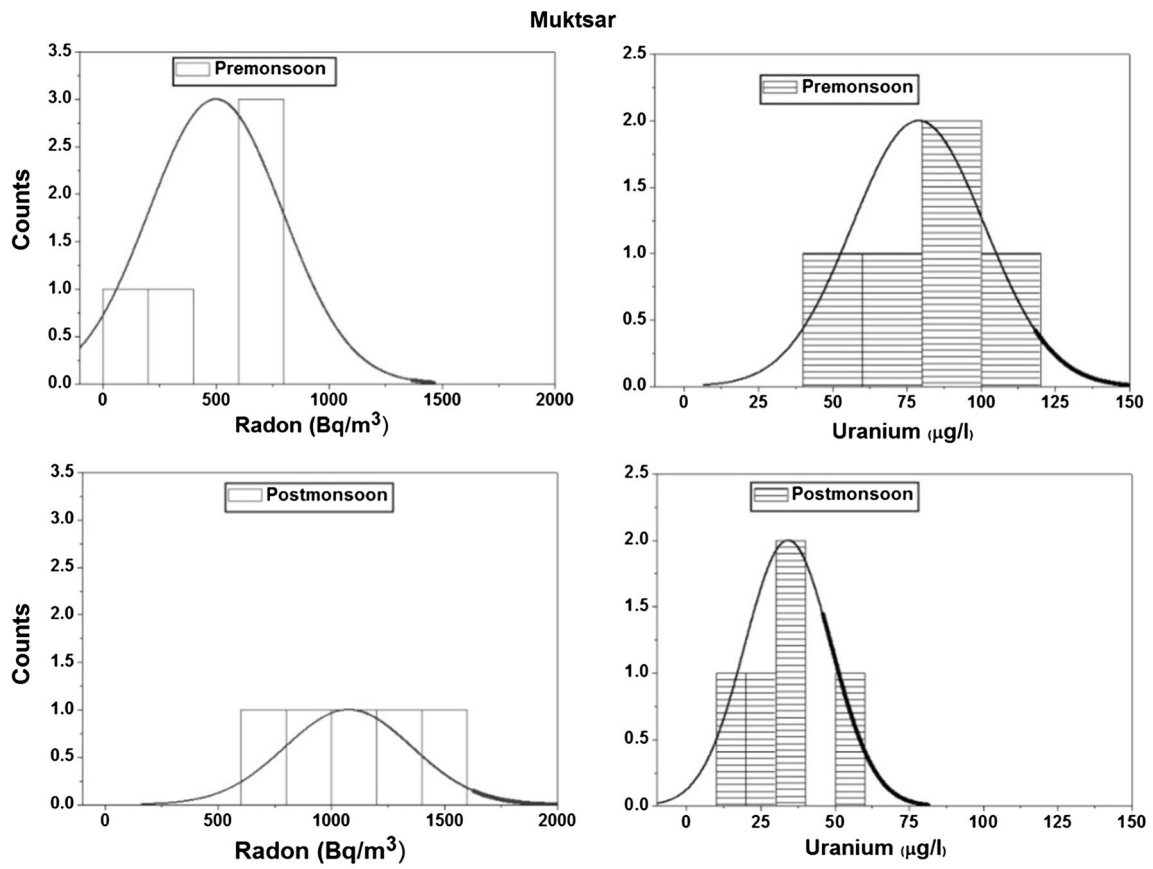**Fig. 4** Histograms of U and Rn for both season of Faridkot

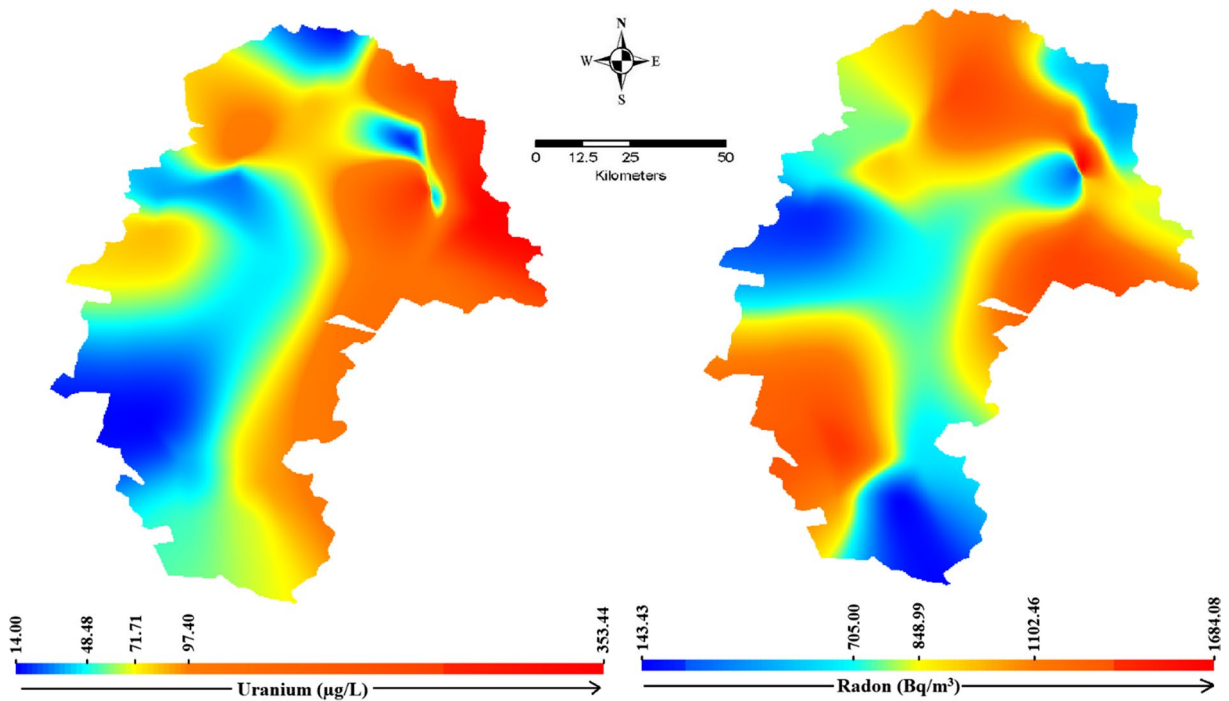**Fig. 5** Histograms of U and Rn for both season of Muktsar

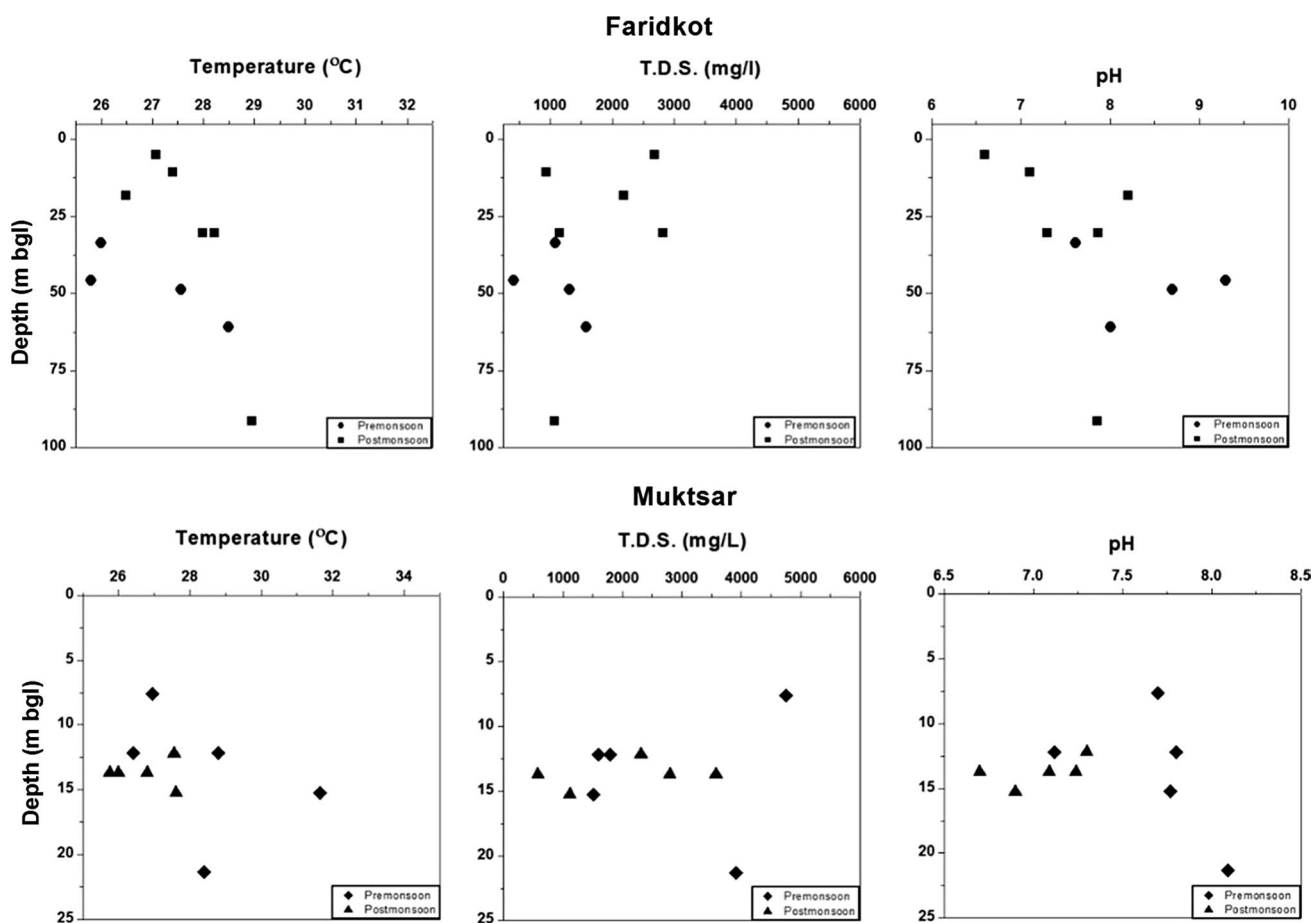**Fig. 6** Contour diagrams for Rn and U for Faridkot and Muktsar districts (both seasons combined)

Fig. 7 Depth profile for temperature, TDS and pH for Faridkot and Muktsar districts (both seasons combined)

Rn correlations with U and physical parameters

Correlation study helps in determining the role of hydrochemical parameters on the distribution of Rn concentration in the study area. These correlation studies can help in predicting the release mechanism and migration behavior of Rn to farther distances. Radon showed negative correlation with all the physico-chemical parameters under study (Fig. 9). The solubility of gases in solvent decreases if temperature increases and the main reason is dissolution of gas in solvent is an exothermic process ($\Delta H_{soln} < 0$). And if heat is supplied to an exothermic process it tends to shift its equilibrium in reverse direction. Conversely, adding heat to the solution provides thermal energy that overcomes the attractive forces between the gas and the solvent molecules, thereby decreasing the solubility of the gas.

Hence negative correlation between Rn and temp can be understood. Similar trends are observed in Muktsar district in both pre and postmonsoon seasons while in Faridkot district no particular trend was observed which could be due to limited number of samples. But a slight decreasing trend can be noticed if pre and postmonsoon samples are considered separately. Negative correlation between Rn and TDS may be due to precipitation of Ra (precursor of Rn) as carbonate (CO_3^{2-}), bicarbonate (HCO_3^{-}), chloride (Cl^{-}) forms. According to studies carried out by Langmuir et al. [35, 36] radium is readily removed from system by adsorption on clay and silicates present in rock material and it co precipitates with insoluble sulphates.

Correlation of Rn with uranium showed negative correlation with correlation coefficient of -0.39 (Fig. 10). Uranium decays to give Th-230 and then Ra-226 and this
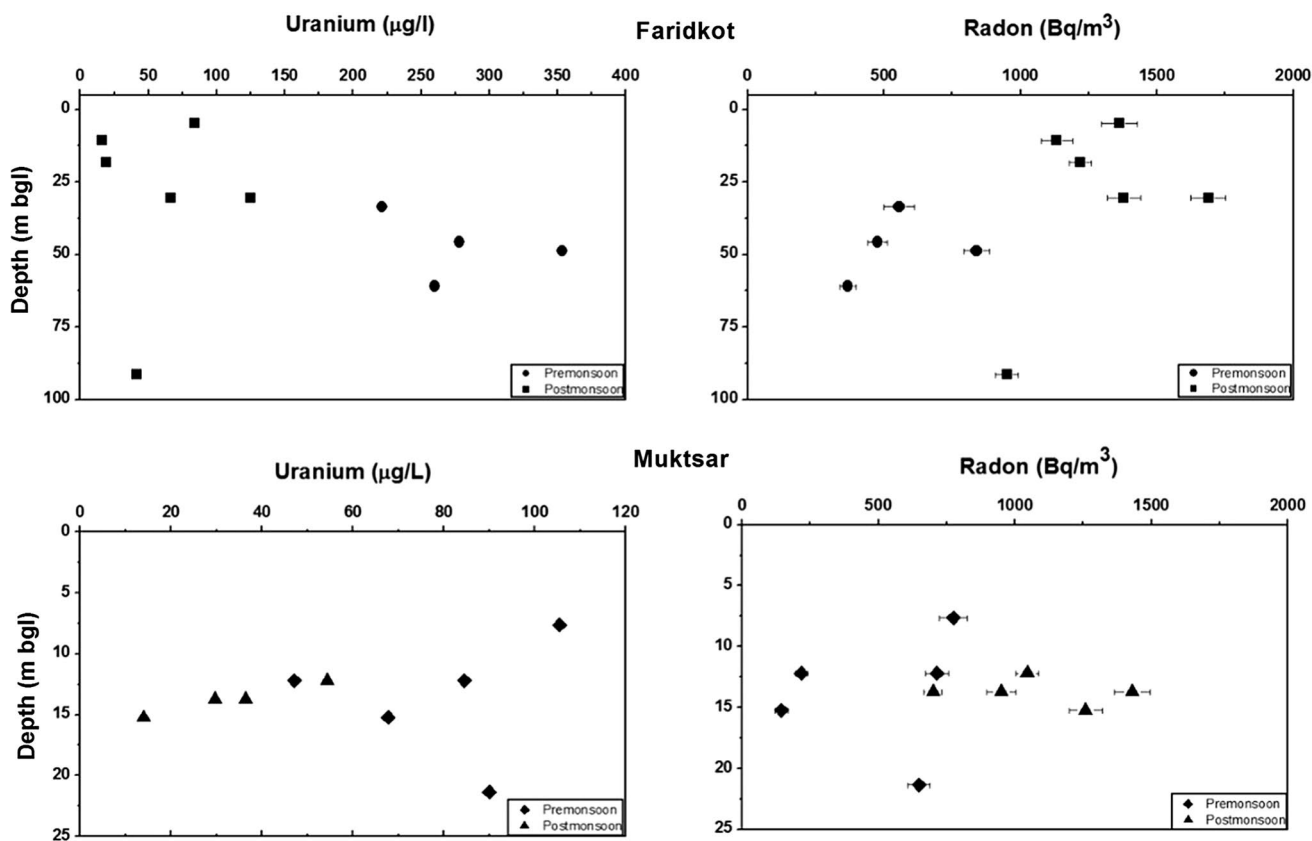
Fig. 8 Depth profile of U and Rn for both seasons of Faridkot and Muktsar districts

newly formed radium moves along the flow path and gets removed from the system by absorption, precipitation or cation exchange [37]. Hence, site of high radium concentration can be different from site of its generation due to its parent decay i.e.Th-230. When radium decays to give its daughter nuclide i.e. radon it is propelled out from the site of generation and an alpha gets recoiled in opposite direction. The diffusion length of Rn-222 depends upon density and composition of matrix. It is studied that the diffusion length of radon is short and is of the order of 0.02–0.07 mm [38]. Depending on the position of radium grain the gas may go deep in rock matrix or can go in the flowing groundwater [39]. The degree of emanation to groundwater depends on aquifer conditions like fractures in bed rock and its connection with groundwater, permeability of bedrock etc. Hence the site of high uranium concentration is different from site having high radon concentrations; thus a negative correlation between these two elements can be expected.

Summary

The groundwater in uranium impacted districts viz., Faridkot and Muktsar of southwest Punjab was studied for its dissolved radon concentration, radiological dose and correlation with U and physico-chemical parameters. The vertical distribution of temperature shows a slight increase while TDS shows decrease with depth and pH value shows scatter for Faridkot district. In the case of Muktsar district, a scatter is observed for all the three parameters. Results from the study showed negative correlation between Rn and TDS, which may be due to removal of precursor radionuclide i.e. radium from the system as it precipitates forming carbonate and bicarbonate complex leading to lesser concentration of radon when TDS increases. It is well known that with increasing temperature the dissolution of gases decreases and similar trends were noticed in the current study. A negative correlation was observed between radon and uranium and hence disequilibrium condition exists, which may be due to adsorption or cation exchange of radium. The radon concentrations seem to increase in postmonsoon season which may be due to dissolution of radon present in pores with the
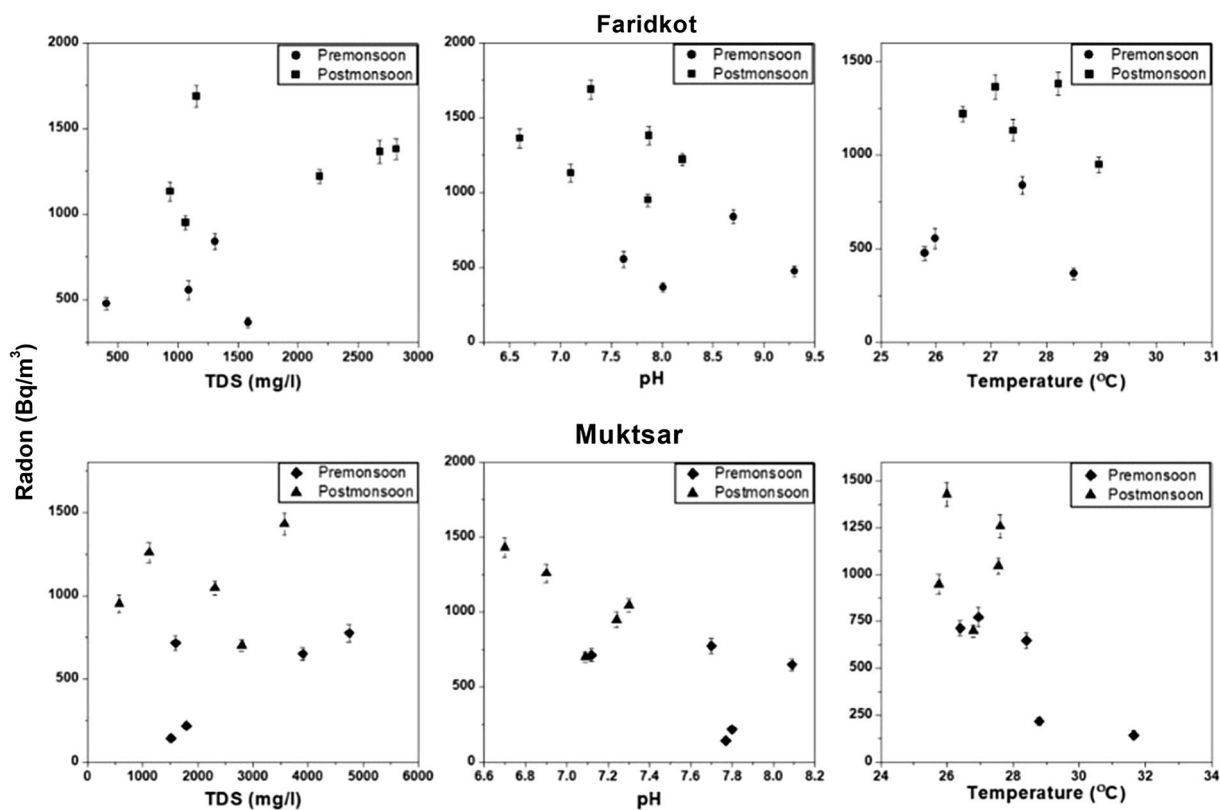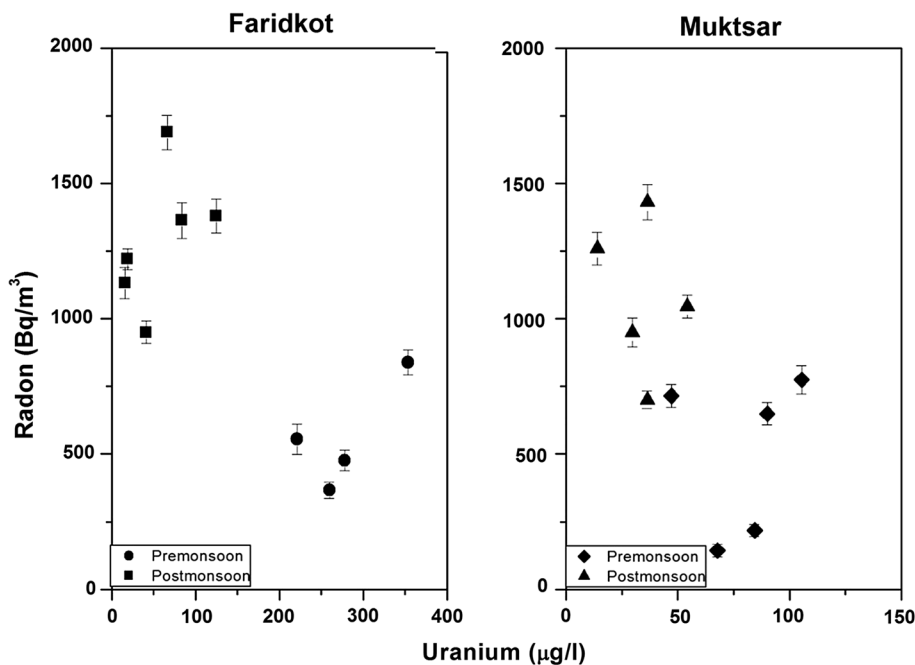
Fig. 9 Correlation of Radon with temperature, TDS for both seasons of Faridkot and Muktsar districts

Fig. 10 Correlation plots for Rn and U for both seasons of Faridkot and Muktsar districts

infiltrating water while a decreasing trend is observed for uranium which is due to dilution by the recharging water.

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