Indian J. Phys. **83** (7), 1001-1010 (2009)

Seasonal variation on radon emission from soil and water

Yogesh Prasad[1], Ganesh Prasad[1], G S Gusain[1], V M Choubey[2] and R C Ramola[1]*

[1]Department of Physics, H N B Garhwal University, Badshahi Thaul Campus, Tehri Garhwal-249 199, Uttarakhand, India

[2]Wadia Institute of Himalayan Geology, Dehradun-248 001, Uttarakhand, India

E-mail : rcramola@bsnl.in

Abstract : Radon is being measured continuously in spring water and soil-gas at Badshahi Thaul Campus, Tehri Garhwal in Himalayan region by using radon Emanometer since December 2002. An effort was made to correlate the variance of radon concentrations in spring water and soil-gas with meteorological parameters at the same location. The main meteorological parameters that affect the radon emanation from host material is surrounding temperature, barometric pressure, wind velocity, rain fall and water level of the spring. The correlation coefficient between radon concentration in spring water and different atmospheric parameters was computed. The correlation coefficient between radon concentration in spring water and the maximum atmospheric temperature was 0.3, while it was 0.4 for minimum atmospheric temperature at the monitoring site. The correlation coefficient for radon concentration in spring water with minimum and maximum relative humidity was 0.4. Spring water radon concentration was found positively correlated (0.6) with water discharge rate of the spring. A weak correlation (0.09) was observed between the radon concentration in spring water and rain fall during the measurement period. As temperature of near surface soil increases, the radon emanation coefficient from the soil surface also increases. The possible effects due to global warming and other climatic changes on environment radiation level were also discussed in detail.

Keywords : Radon, meteorology, transfer efficiency.

PACS Nos. : 89.60.-k, 23.60.+e

1. Introduction

Radon tends to migrate from its source mainly upward, this rate of migration is affected by many factors, such as distribution of uranium in the soil and bed rock, soil porosity and humidity, micro-cracks of bed rock, rainfall, air temperature, barometric pressure, surface wind and so on [1,2]. One of the most important physical characteristics of soil pertinent to indoor radon is its permeability *i.e.* how readily air or fluid may flow through it

* Corresponding Author

[3]. The emission coefficient of radon from different material also affects the concentration of radon in the indoor air. If the soil above the bedrock has an enhanced content of ^{238}U, the soil may be the major source of the radon in the environment. With the importance of the source of the radon gas there are certain additional parameters connected to the soil-gas and ground water radon which affecting the true level of the radon in the surrounding environment [4–8]. Static parameters of the soil *i.e.* grain size; porosity and soil type are constant with respect to time span and season. The water content has a large impact on the emanation coefficient and on the soil transport parameters for radon gas and therefore affect the radon concentration in the soil [9]. The National Research Council (NRC) estimates that 160 Americans die annually from lung cancer caused by radon in drinking water [10]. A variety of factors shape the risk exposure people face, the level of radon dissolved in drinking water, the volume of water used, the transfer of the radon into the indoor atmosphere, the breathing rate, the rate of radioactive decay and occupancy factor [11].

In this study we report the data from our laboratory generated from daily measurements of radon from spring and soil-gas over the last four years. The effect of other environmental parameters was also observed in the radon emission from spring and soil located at same location. The recorded data in different seasons were also correlated with recommended international limit given by various scientific agencies.

2. Experimental method

In soil radon emanometry, the auger holes, each 60 cm in depth and 5.5 cm in diameter, were left covered for 24 hours, so that the amount of radon and thoron became stable. The soil-gas probe was fixed in the auger hole and formed an air-tight compartment [12]. The rubber pump, soil-gas probe and alpha detector (Lucas Cell) were connected in closed circuit. Air was circulated through a ZnS coated chamber for 15 min until the radon formed a uniform mixture with the air. The alpha particles produce scintillation on the sample chamber which are sensed by a photomultiplier tube and converted to the electronic meter-reading. The resulting display of the counts is converted into Bq/m^3 by using the calibration factor 1 count/min = 66.3 Bq/m^3 determined by [13].

A 750 ml water sample was taken in a 1-litre radon-tight reagent bottle connected in a closed circuit with a ZnS(Ag) coated detection chamber through a hand-operated rubber pump and a glass bulb containing $CaCl_2$ to absorb the moisture. Air was then circulated in the closed circuit for 15 min until the radon formed a uniform mixture with the air and the resulting alpha activity was recorded. The calibration factor 1 count/min = 0.0663 Bq/*l* was used to convert the recorded alpha counts in Bq/*l* [13].

2.1. Indoor radon concentration resulting from water use :

The indoor radon concentration due to the spring water is calculated by using the mass conservation formula [14]. The distance of house from the water spring is about 50 m.

$$C_a = \frac{C_w We}{V \lambda_0}$$

where C_w = radon concentration in water entering the residence (Bq/m^3)

W = water use rate per resident (m^3/person h)

e = the use weighted average transfer efficiency of radon from water to air (dimensionless)

V = the volume per resident of the dwelling (m^3/person)

λ_v = air-exchange rate of the residence (h^{-1}), assumed to be much greater than the decay constant for radon (0.0076 h^{-1}).

The equation must be expressed in terms of a transfer factor, *f*

$$C_a = f.C_w$$

$$f = \frac{We}{V \lambda_0}$$

There are two distinct approaches for determining $f.C_w$ is measured daily by using radon emanometry method. Water use rate in the particular house is calculated by taking average water used by the three people in twenty four hours; it is calculated 1.66 × 10^{-3} h^{-1} in winter and autumn season and 1.87 × 10^{-3} m^{-3} person^{-1} h^{-1} in summer and rainy season. Volume of the room is 5 m × 3.5 m × 4 m (70 m^{-3}). Geometric mean value of air exchange rate is considered same as [14] analysed from different set of data and different ventilation conditions (0.68 h^{-1}). The use weighted average transfer efficiency are considered between minimum and maximum value (limit between, 0.3–1.0) *i.e.* 0.6 (say).

3. Results and discussion

The radon anomalies in spring water and soil-gas are associated with the meteorological parameters recorded at measuring site. The radon spectrum of recorded data from the spring water and soil-gas gives spike like anomalies with changing meteorological data and water level of the spring. The variations in average radon concentrations in soil-gas and spring water at Badshahi Thaul, Tehri Garhwal, for different seasons are given in Table 1. The main meteorological parameters that affect the radon emanation from host material is surrounding temperature, barometric pressure, wind velocity, rain fall and water level of the spring. The recorded values of air temperature, relative humidity, rain-fall and water discharge rate from a spring at study area are given in Table 2.

The radon concentration in spring water along with the meteorological parameters was measured since December 2002. The monthly average radon value in spring water during

Table 1. Monthly average radon value recorded in spring water and soil-gas with seasonal average and standard deviation.

Months	Radon concentration in spring water (Bq/m^3)	Standard deviation	Average for one season (Bq/m^3)	Soil-gas radon concentration (Bq/m^3)	Standard deviation	Average for one season (Bq/m^3)
Dec-02	1137	191		N.A	N.A	N.A
Jan-03	1422	660		N.A	N.A	N.A
Feb-03	2300	560	2125	N.A	N.A	N.A
Mar-03	2744	454		N.A	N.A	N.A
April-03	3311	571		N.A	N.A	N.A
May-03	3551	482	4352	N.A	N.A	N.A
Jun-03	5915	1790		N.A	N.A	N.A
Jul-03	7703	661		N.A	N.A	N.A
Aug-03	10009	1601	9202	N.A	N.A	N.A
Sep-03	10124	1979		N.A	N.A	N.A
Oct-03	5167	1002		N.A	N.A	N.A
Nov-03	5171	898	4645	N.A	N.A	N.A
Dec-03	3851	824		N.A	N.A	N.A
Jan-04	3174	1201		N.A	N.A	N.A
Feb-04	1930	1029	2573	N.A	N.A	N.A
Mar-04	2210	324		1445	1918	
April-04	2619	594		2338	1403	
May-04	2599	818	2639	1945	4099	1982
Jun-04	2717	537		1718	1753	
July-04	3046	862		557	313	
Aug-04	3069	788	4405	7035	3751	3532
Sep-04	6199	1802		1072	913	
Oct-04	6119	1316		8708	8449	
Nov-04	4188	953	4281	10336	5621	8248
Dec-04	2405	805		6925	913	
Jan-05	2125	490		3819	2430	
Feb-05	2327	1065	2399	4461	3853	3177
Mar-05	2676	758		1859	843	
April-05	2931	1026		3956	3567	
May-05	2778	918	3011	4221	2579	3990
Jun-05	3490	672		3728	2075	
Jul-05	4984	1467		2246	2362	
Aug-05	7963	2267	6337	5912	4057	5968
Sep-05	5838	1086		8584	6084	
Oct-05	7803	3277		8348	5030	
Nov-05	5126	1182	5921	7996	3664	8085
Dec-05	3675	888		7656	3869	
Jan-06	2264	1656		3889	4186	
Feb-06	1453	544	1869	1409	814	4446
Mar-06	1710	585		6253	3911	
April-06	2148	794		3438	2755	
May-06	2551	529	2240	1917	916	3148

the year 2003, was recorded minimum (1422 Bq/m^3) in January and the maximum (10124 Bq/m^3) in September. The average minimum temperature, maximum temperature, rain fall and water discharge rate from the spring for January 2003 were recorded 1.6 °C, 12.5 °C, 2.1 mm and 3.5 *l*/min, respectively (Table 2). For September 2003, the average minimum temperature, maximum temperature, rain fall and water discharge rate from the spring were recorded 14.1 °C, 22.0 °C, 2.7 mm and 8.6 l/min, respectively. The radon emission and water discharge rate are largely influenced by the environmental temperature and rain fall in the area. Enrichment of radon in spring water has been observed after the rain fall possibly with increasing of more catchments area additional radon is supplied from the surrounding rocks. The variation of the radon concentration in spring water with the water discharge rate of the spring during the year 2003 is shown in Figure 1. The water discharge of the spring increases between 24 to 48 hours after the rain fall. It was also observed that with mild rain fall the radon concentration increases by depletion of radon in water was noticed after heavy rain fall. Possibly heavy rain fall dilutes the radon concentration in per litre volume. The average seasonal variations of radon concentration in the spring water were also calculated during 2003 and are given in the Table 1. The lowest radon value in the spring water was recorded in winter season (2125 Bq/m^3) and highest in rainy season (9202 Bq/m^3).

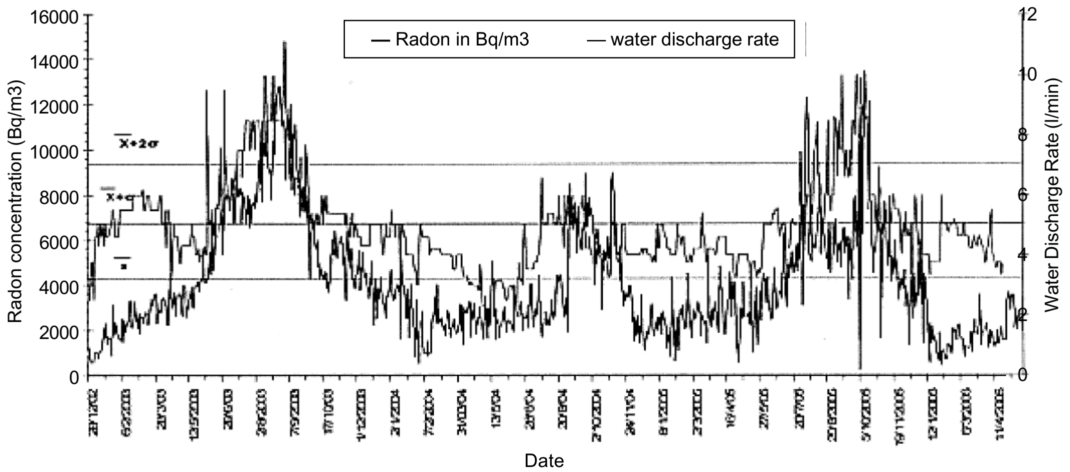

Figure 1. Variation in radon concentration and water discharge rate from the spring.

The monthly average variation of radon for the year 2004 was recorded minimum in February (1930 Bq/m^3) and maximum in the September (6199 Bq/m^3). The average seasonal variation of radon for the year 2004 was recorded minimum (2573 Bq/m^3) in the winter season and maximum (4281 Bq/m^3) in autumn. The shift in monthly and seasonal variations for the year 2004 in comparison to the year 2003 is due to the variation in meteorological parameters as shown in Table 2. The rain fall during February 2003 (10.4 mm) was higher than the rain fall in February 2004 (0.1 mm). Consequentially the water level of the spring

Table 2. Monthly average values of meteorological parameters with monthly average of water discharge rate from spring.

Months	Average temperature (ºC)		Average relative humidity (%)		Average rain fall (mm)	Average water discharge rate (l/min)
	Min	Max	Min	Max		
Dec-02	1.3	14.2	50.1	71.2	0.5	3.3
Jan-03	1.6	12.5	52.7	70.3	2.1	3.5
Feb-03	1.4	11.2	64.7	82.1	10.4	4.8
Mar-03	5.0	16.8	55.6	76.6	1.2	5.6
April-03	9.6	23.1	38.9	57.5	1.8	5.6
May-03	11.1	26.1	33.9	52.9	0.8	4.5
Jun-03	14.9	26.0	61.7	77.0	1.8	4.2
July-03	15.3	23.6	81.0	95.7	13.4	5.7
Aug-03	15.2	23.3	83.4	96.3	6.6	7.8
Sep-03	14.1	22.0	88.6	95.8	2.7	8.6
Oct-03	8.2	22.2	51.3	68.7	0.0	6.0
Nov-03	4.0	17.8	50.3	75.2	0.0	5.4
Dec-03	1.2	12.7	61.5	85.0	0.9	4.9
Jan-04	0.4	11.0	65.6	86.7	3.9	4.7
Feb-04	4.1	14.1	57.3	86.8	0.1	4.1
Mar-04	10.2	22.0	30.0	60.3	0.0	4.4
April-04	12.1	24.3	42.9	64.3	1.6	3.9
May-04	14.6	26.0	41.0	62.3	1.5	3.0
Jun-04	15.5	24.2	68.9	84.3	4.0	2.6
Jul-04	16.5	22.9	82.2	93.9	12.5	2.8
Aug-04	16.2	22.5	84.0	97.8	8.5	4.1
Sep-04	14.7	22.7	80.8	96.7	5.4	5.2
Oct-04	8.8	18.7	69.3	86.7	3.5	4.8
Nov-04	5.5	16.4	65.5	93.8	0.0	4.0
Dec-04	3.3	14.4	60.5	81.3	0.1	4.4
Jan-05	0.9	10.7	70.5	89.2	1.2	4.1
Feb-05	2.0	10.8	70.1	87.2	4.2	4.3
Mar-05	6.4	17.6	60.3	72.5	2.4	4.1
April-05	9.7	23.4	34.4	52.9	0.5	4.2
May-05	11.6	26.6	36.0	63.0	0.8	4.1
Jun-05	14.7	29.3	33.5	59.3	0.4	3.6
July-05	15.8	22.2	86.7	97.5	13.2	5.0
Aug-05	15.4	22.8	84.0	96.0	3.4	5.1
Sep-05	13.7	22.1	82.1	94.2	7.0	5.7
Oct-05	9.6	20.8	69.6	90.8	0.05	8.0
Nov-05	5.4	18.5	48.9	72.0	0.0	6.5
Dec-05	3.2	16.3	41.3	70.6	0.0	5.3
Jan-06	N.A	N.A	N.A	N.A	N.A	4.8
Feb-06	N.A	N.A	N.A	N.A	N.A	3.8
Mar-06	N.A	N.A	N.A	N.A	N.A	4.8
April-06	N.A	N.A	N.A	N.A	N.A	4.4
May-06	N.A	N.A	N.A	N.A	N.A	3.5

during 2003 (4.8 litre/min) was higher than the water level of the spring during 2004 (4.1 litre/min). This is also the main cause of shifting the minimum monthly average radon concentration in February 2004, although it was recorded minimum in the month of January during 2003.

The monthly average radon concentration during 2005 was recorded minimum (2125 Bq/m^3) in January and maximum (7963 Bq/m^3) in August. The seasonal average radon concentration in the spring water was recorded 2399 Bq/m^3 and 6337 Bq/m^3 in winter and rainy season, respectively. These variations are similar to the same recorded in previous two years. In 2006, the monthly average radon concentration in spring water was recorded minimum (1453 Bq/m^3) in February and maximum (2551 Bq/m^3) in May. The similar behaviour of radon concentration in spring water with seasonal changes during last four year shows that the environmental parameters play an important role in emanation of radon from the spring water.

The correlation coefficient was also computed between radon concentration in spring water and different atmospheric parameters. The correlation coefficient between radon concentration is spring water and the maximum atmospheric temperature was computed 0.3, while it was 0.4 for minimum atmospheric temperature at the monitoring site. The correlation coefficient for radon concentration in spring water with minimum and maximum relative humidity was calculated 0.4. Spring water radon concentration was found positively correlated (0.6) with water discharge rate of the spring. A weak correlation (0.09) was observed between the radon concentration in spring water and rain fall during the measurement period. Radon in groundwater, especially in deep or confined aquifers, is less perturbed by metrological variation [15].

The monthly and seasonal variation in soil-gas radon concentrations are shown in Table 1. The soil-gas radon concentration was recorded continuously at monitoring site from March 2004 to May 2006. During this period, soil-gas radon concentration was found to vary from 557 Bq/m^3 to 10336 Bq/m^3. In the year 2004, average soil-gas radon concentration was recorded minimum (1982 Bq/m^3) in summer season and maximum (8248 Bq/m^3) in autumn. However, this variation can not be taken as representative as there was no data for winter season. In 2005, the average soil-gas radon concentration was recorded minimum (3177 Bq/m^3) in winter season and maximum (8085 Bq/m^3) in autumn season. The data is again incomplete for 2006 with average soil-gas radon concentration 4446 Bq/m^3 in winter season and 3148 Bq/m^3 in summer season. The lowest and highest monthly average soil-gas radon concentrations during the year 2004 were recorded 557 Bq/m^3 and 10336 Bq/m^3 in July and November, respectively. The lowest monthly average soil-gas radon concentration (1859 Bq/m^3) during 2005 was recorded in March and highest monthly average soil-gas radon concentration (8584 Bq/m^3) was recorded in September. The minimum monthly average soil-gas radon concentration (1409 Bq/m^3) for 2006 was recorded in February and maximum average soil-gas radon concentration

(6253 Bq/m^3) was recorded in March. The diverse variation in soil-gas radon concentration shows that the radon emission from soil is highly perturbed by the meteorological conditions of the monitoring site.

The correlation coefficient of soil-gas radon concentration with meteorological parameters was computed. The measurements were taken in the air tight auger hole of 60 cm depth to mitigate the meteorological effect on radon accumulation in augur hole. Only moderate to heavy rain fall, humidity, atmospheric temperature and barometric pressure could affect the accumulation conditions in the closed compartment. Soil-gas radon concentration shows negative correlation (–0.3) and (–0.2) with atmospheric maximum and minimum temperature, respectively. As temperature of near surface soil increases, the radon emanation coefficient from the soil surface also increases, which results in the decrease in soil-gas radon concentration in auger hole. King also found the inverse relationship between radon emanation and air temperature in the San Andreas Fault region [16]. Soil-gas radon shows weak positive correlation (0.2) with radon concentration in spring water due to the different radon migration mechanism in soil-gas radon and spring water. Soil-gas radon concentration shows a weak positive correlation (0.1) with minimum and maximum relative humidity. No correlation (0.01) was observed between soil-gas radon concentration and rain fall in the study area.

Table 3. The indoor radon concentration due to the spring water.

	W	S	R	A
Indoor radon concentration (Bq/m^3)	65	63	69	81
Average radon concentration in water (Bq/m^3)	2125	4352	9202	4645
Contribution of water radon to indoor air (C_a) (Bq/m^3)	0.04	0.10	0.21	0.09

W - Winter, S - Summer, R - Rainy, A - Autumn

The water radon levels during this period did not exceed the then benchmark maximum contaminant level of 11.1 Bq/l [17]. The contribution of water radon in indoor air of the nearest house from the spring has been determined by the long term average of water use rate, water radon concentration and indoor radon concentration in particular house volume. The estimate the fraction of indoor radon concentration from water use rate other parameters are considered within the limit established by Nazaroff *et al* (1988a). Ground water supply does not contribute significantly to indoor radon concentrations in different season (Table 3). From this model the water radon concentration contributes 0.1% to 0.3% in indoor radon concentration in the house situated at particular location. There are some uncertainties in use–weighted transfer coefficient because any residence uses water in a mix of applications, some with high transfer efficiency and other with low value. The random air exchange rate of houses also has no definite pattern in different season, which is also an uncertain parameter in calculating the indoor radon fraction due to ground water uses. The distance of house from the ground water source also affect the radon release in available volume.

4. Conclusions

The study of seasonal variation of radon in soil-gas and ground water is very effective for the purpose of risk calculation and contribution of radon sources in indoor radon concentration at specific site. With static parameters of the soil *i.e.* grain size; porosity and soil type are constant with respect to time span and season, the meteorological parameters play an important role in radon emission from different medium. Observed radon levels are not higher than the contaminant level set by US Environmental Protection Agency, therefore the use of water at particular site is safe for general public. The method of calculating radon fraction in indoor air is general and could be equally applied to other places where the groundwater radon concentrations are particularly high. The groundwater and soil-gas radon concentration is expected to reflect not only the permeability and emission coefficient of medium but also structural properties of bed rock.

Acknowledgments

Authors are thankful to Department of Science and Technology, New Delhi, Council of Scientific and Industrial Research, New Delhi and Board of Research in Nuclear Sciences, Department of Atomic Energy, Government of India for providing financial support in the form of project.

References

[1] J Planinic, V Radolic and Z Lazanin *Appl. Radiat. Isot.* **55** 267 (2001)

[2] M Inceoz, O Baykara, E Aksoy and M Dogru *Radiation Measurements* **41** 349 (2006)

[3] W W Nazaroff, B A Moed and R G Sextro *Soil as a source of indoor Radon Generation, Migration and Entry* (New York : A Wiley-Interscience Publication) 57 (1988)

[4] M Singh, R C Ramola, N P Singh, S Singh and H S Virk *The Study of Radon Diffusion in Air and Soil.* Proc. 5th National Seminar on Solid State Nuclear Track Detectors Calcutta 134 (1987)

[5] I Tell, I Bensryd, L Rylander, G Jonsson and E Daniel *Appl. Geochem.* **9** 647 (1994)

[6] Li Font *Radon Generation, Entry and Accumulation Indoors* PhD Thesis University Autonoma de Barcelona, Spain **ISBN** 84-490-1265-1 (1997)

[7] R C Ramola, R B S Rawat, M S Kandari and V M Choubey *Radiat. Prot. Dos.* **74** 103-105 (1997)

[8] V M Choubey, K S Bisht, N K Saini and R C Ramola *J. Appl. Radiat. Isotopes.* **51** 587 (1999)

[9] E Stranden, A K Kolstad and B Lind *Health Phys.* **47** 480 (1984)

[10] National Research Council *Risk Assessment of Radon in Drinking Water* (Washington, DC : National Academy Press) p18 (1999)

[11] D F Vitaliano *Costs and Benefits of Mitigating Radon in Drinking Water, Public Works Management and Policy* **7** 291 (2003)

[12] P C Ghosh and N S Bhalla *Ind. J. Earth Scie.* **8** 1 (1981)

[13] V M Choubey, T V Ramachandran, M S Negi and R C Ramola *Indian J. Environmental Protection* 329 (2000)

[14] W W Nazaroff, S M Doyle, A V Nero (Jr.) and R G Sextro *Radon Entry via Portable Water* (New York : A Wiley-Interscience Publication) 131 (1988)

[15] Chi-Yu King *Radon Gas Geochemistry, Groundwater and Earthquakes* Proce. The 7th Tohwa University International Symposium on Radon and Thoron in the Human Environment (eds.) A Katase and M Shimo 115 (1988)

[16] Chi-Yu King *Nature* (London) **271** (5645), 516 (1978)

[17] US Environmental Protection Agency National Primary drinking water regulations; Radon-222; proposed rule (40 CFR Parts 141 and 142) Washington DC: Federal Register (1999)