Activity of radon (^{222}Rn) in the lower atmospheric surface layer of a typical rural site in south India

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Analysis of one year measurements of *in situ* radon (^{222}Rn) and its progenies along with surface air temperature, relative humidity and pressure near to the Earth's surface has been carried out for the first time at the National Atmospheric Research Laboratory (NARL, 13.5°N and 79.2°E) located in a rural site in Gadanki, south India. The dataset was analysed to understand the behaviour of radon in relation to the surface air temperature and relative humidity at a rural site. It was observed that over a period of the 24 hours in a day, the activity of radon and its progenies reaches a peak in the morning hours followed by a remarkable decrease in the afternoon hours. Relatively, a higher concentration of radon was observed at NARL during fair weather days, and this can be attributed to the presence of rocky hills and dense vegetation surrounding the site. The high negative correlation between surface air temperature and activity of radon ($R = -0.70$, on an annual scale) suggests that dynamical removal of radon due to increased vertical mixing is one of the most important controlling processes of the radon accumulation in the atmospheric surface layer. The annual averaged activity of radon was found to be 12.01±0.66 Bq m^{-3} and 4.25±0.18 Bq m^{-3} for its progenies, in the study period.

1. Introduction

Radon emission from the ground has been extensively studied over the last decades from several viewpoints. Radon and its progeny deliver the highest radiation dose to human beings among all natural radioactive sources (UNSCEAR 1993, 2000); it represents a very interesting behaviour of natural trace gas for a number of atmospheric and geophysical research applications like pollutant dispersion model, boundary layer studies, relation with ground-level ozone (O_3), NO_x and particulate matter of several size fractions (Kataoka *et al.* 2001; Sesana *et al.* 2003; Desideri *et al.* 2006, 2007; Zoran *et al.* 2014; Chambers *et al.* 2015). The occurrence of anomalous changes in radon concentrations prior to earthquakes has been studied (Walia *et al.* 2006; Ghosh *et al.* 2009; Zoran *et al.* 2012). Radon is a radioactive noble gas emitted mainly by the soil. The origin of radon in the earth's crust stems directly from uranium and its decay products, which are distributed in minute quantities in the ground within a few meters below the surface. Radon is gaseous in nature and transported by diffusion, advection through the pore space until they decay or released into the atmosphere (Israelsson *et al.* 1973; Clements and Wilkening 1974; Pressyanov *et al.* 1995). The principal decay modes and half-lives of radon and its short-lived daughters in order are ^{222}Rn-α – 3.82 days; ^{218}Po-α – 3.05 min; ^{214}Pb-β – 26.8 min; ^{214}Bi-β – 19.7 min; and ^{214}Po-α – 2×10^{-4} s, among which the alpha

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emitters are the main contributors for the ionisation of air.

It is clear from literature that no studies on the behaviour of the rare natural tracer radon (^{222}Rn) at National Atmospheric Research Laboratory (NARL), Gadanki, south India are reported. Hence, a one-year time series (January–December 2012) of radon and its progenies were measured at NARL and analysed. The measurement of radon, its progeny, temperature (T), relative humidity (RH) and pressure (P) were carried out for every 10-min and averaged for required period at a fixed location. In this paper, the following research issues are addressed:

- Analysis of the one-year time series, statistics and investigation of the monthly variability of the activity of radon and its progenies.
- Dependence of radon concentration on T, RH and P.
- Effect of precipitation on radon concentration and its recovery after rainfall.
- Estimation of ion pair production rate due to radon and its progeny.

2. Site description

Continuous observations were carried out at NARL, Gadanki (13.5°N, 79.2°E), a rural tropical warm location in peninsular India, about 2 km away from the main residential areas with no major industrial activities. The locative map and topography of NARL are shown in figure 1. The laboratory is surrounded by many rocky hills, terrains, dense vegetation and it experiences both summer (south-west) and winter (north-east) monsoons. Overall wind direction is southerly and south-easterly during April, westerly from May to September and north-easterly during October and November (Naja and Lal 2002; Renuka *et al.* 2014).

3. Experimental methodology

Several techniques have been established for measuring the concentration of radon in air, which are mainly based on the detection of emissions from radon or its radioactive decay products (Nagaraja *et al.* 2003; Sesana *et al.* 2003; Desideri *et al.*

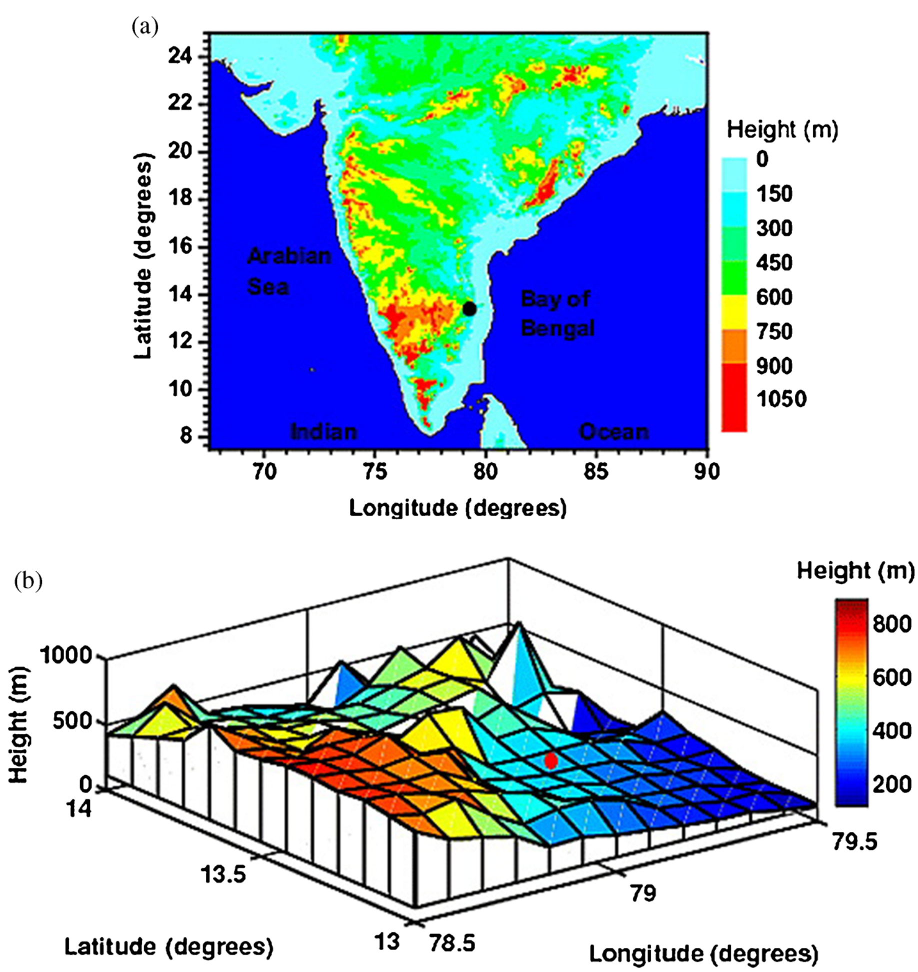

Figure 1. The locative map and topography of NARL.

2007; Calin and Calin 2011; Chambers *et al.* 2015; Kobayashi *et al.* 2015). But, due to low concentrations of environmental radon, the precision, accuracy and detection efficiency of the techniques are of important issues. AlphaGUARD PQ-2000PRO, a compact, portable measuring system, was used for the continuous monitoring of the radon concentration along with T, RH and P, while Alpha PM was used for radon progeny measurements. The alpha guard uses the principle of pulse ionisation chamber and measures radon by 3D alpha spectroscopy technique with DSP technology. It consists of the cylindrical ionisation chamber of active volume 0.56 L and is operated at a potential of +750 V. The center electrode is connected to the signal input of the highly sensitive preamplifier unit and then further fed to digital processing. Its measuring range for radon is 2–20,00,000 Bq m^{-3} and the sensitivity analysis of 4.5 CPM/100 Bq m^{-3}. Main climatic parameters recorded simultaneously are temperature, relative humidity and pressure. The instrument is coupled to get synchronised results and then fed to a memory module for data acquisition (Griffiths *et al.* 2013; Zoran *et al.* 2014; Zimnoch *et al.* 2014).

4. Results and discussions

4.1 *Diurnal variations*

Diurnal variations of the activity of radon and its progeny along with the temperature and relative humidity for a typical fair weather day on 23rd February 2012 are shown in figures 2 and 3, respectively. The radon concentration varied from 2 to 65 Bq m^{-3} showing a significant diurnal variation, almost by a factor of 33, whereas values of radon progeny varied between 2 and 25 Bq m^{-3}. The concentrations show maxima during the early morning hours, generally between 0600 and 0830 h of Indian Standard Time (IST) and decreases after sunrise, attaining minima during the afternoon, 1400–1600 h of IST at NARL (Nagaraja *et al.* 2003; Prasad *et al.* 2005; Desideri *et al.* 2006).

It is observed that the concentration of radon and its progeny follow the trend of the relative humidity, in general. The *in situ* diurnal cycle of radon is triggered by soil emission and atmospheric dynamics (primarily small-scale vertical mixing), so that when the atmosphere is stable (mostly at night), the radon accumulates at the surface.

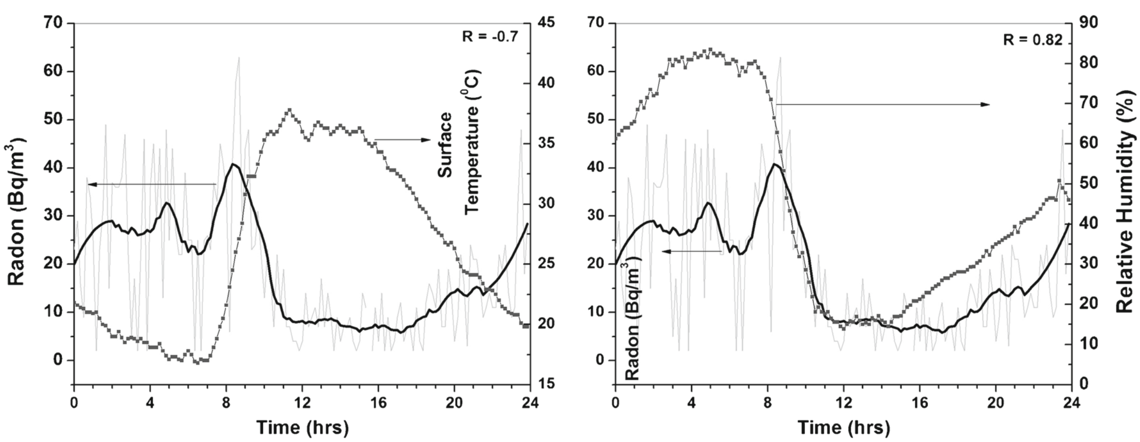

Figure 2. Diurnal variations of activity of radon along with T and RH.

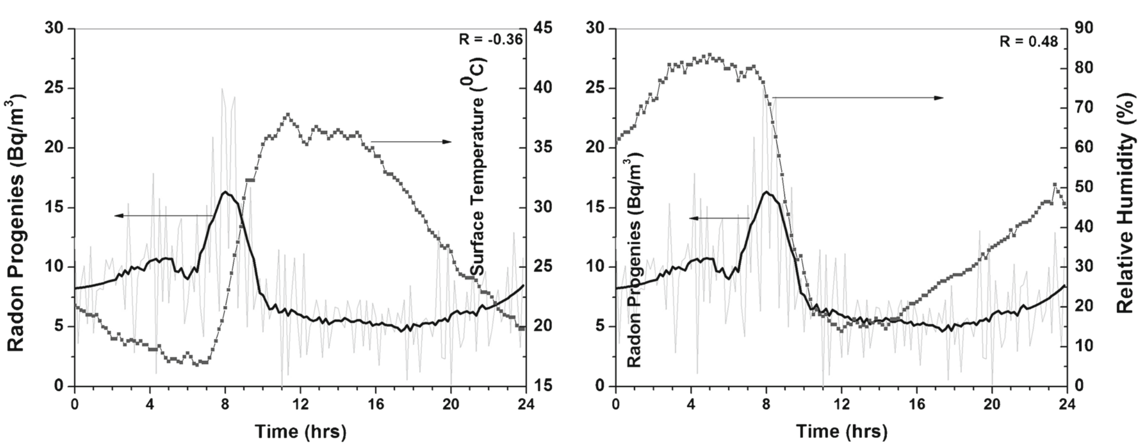

Figure 3. Diurnal variations of activity of radon progenies along with T and RH.

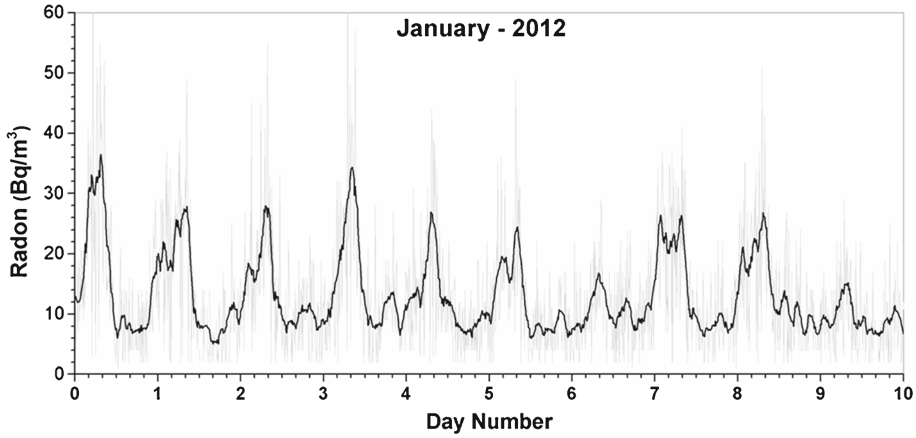

Figure 4. Pattern of nocturnal radon accumulation and subsequent daytime decrease (black lines are hourly averaged).

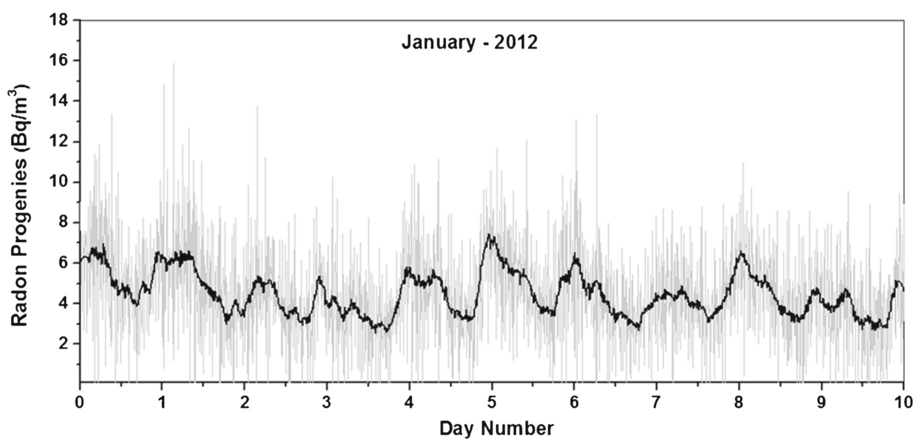

Figure 5. Pattern of nocturnal radon progeny accumulation and subsequent daytime decrease.

After sunrise, when the turbulent vertical mixing starts, the temperature increases, the relative humidity decreases – resulting in the decrease of moisture content in the atmosphere (Wilkening 1990) and the surface concentration of radon rapidly decreases, even though it is still emitted from the ground. When the temperature decreases and relative humidity increases, the vertical mixing and raising of aerosols to the higher altitude reduces. As a consequence, the radon gas and the aerosol to which radon progenies is attached will be present at higher concentrations during the night and in the early morning hours at ground level. This results in the increase of radon concentrations near the surface of the earth (Wilkening 1990; Porstendorfer 1994; Lebedyte *et al.* 2002).

4.2 *Nocturnal stability and mixing processes*

The regular pattern of nocturnal radon and its progenies accumulation and subsequent daytime decrease for continuous 10 days, in January 2012, are presented in figures 4 and 5. A higher value of the activity of radon (50–60 Bq m^{-3}) was observed in this series and this may be due to rocky hills and dense vegetation around the study region (Jayaratne *et al.* 2011). The pattern immediately indicates conditions of high nocturnal stability and temperature inversions in early morning hours and good daytime mixing conditions, especially in the afternoon hours. This type of diurnal trend is frequently observed in the summer and more rarely in winter (Nagaraja *et al.* 2003; Sesana *et al.* 2003). At NARL, winter patterns differ from summer ones by a smaller difference between daily minimum and maximum concentrations in winter than in summer and a longer accumulation phase, and the accumulation phase typically comes to an end at around 7:00–8:00 in the morning in the summer months, whereas it goes on until about 09:00 in winter months.

4.3 *Estimation of ion pair production rate due to alpha particles*

The decay of radon ^{222}Rn and its progenies ^{218}Po and ^{214}Po are followed by emission of alpha particles of energy 5.49, 6 and 7.69 MeV, respectively. This alpha energy ionises the atmospheric air and free small ions are formed. These ions play a very important role in atmospheric electricity, ion–aerosol interaction and its modelling plays a vital role in cloud physics, earthquake prediction, air pollution studies, etc. (Srinivas *et al.* 2001; Nagaraja *et al.* 2006, 2009; Freund *et al.* 2009; Matthews *et al.* 2012; Pawar 2013; Mizuno and Takashima 2013). The total alpha energy released by the radon progenies together is extracted from the Alpha Progeny Meter and the total energy released ε (eV cm^{-3} s^{-1}) due to both radon and its progeny concentration is arrived using the standard approach (Hoppel and Frick 1986). For a nitrogen and oxygen rich environment, the energy required to ionise a single molecule is 32 eV. Hence, ion pair production rate is calculated using equation (1):

$$q = \frac{\varepsilon}{32}\ \mathrm{eV \cdot cm^{-3} \cdot s^{-1}} \qquad (1)$$

A typical variation of ion-pair production rate at NARL, estimated by the activity of radon and its progenies is shown in figure 6; it varied between 1 and 18 ion-pairs cm^{-3} s^{-1} and in the inset, a box plot shows the variability of radon, its progenies and ion pairs. It is observed that ion pairs are produced the maximum in early morning hours, lowest during afternoon hours and it follows the trend of radon and its progenies (Kolarz *et al.* 2009; Nagaraja *et al.* 2009).

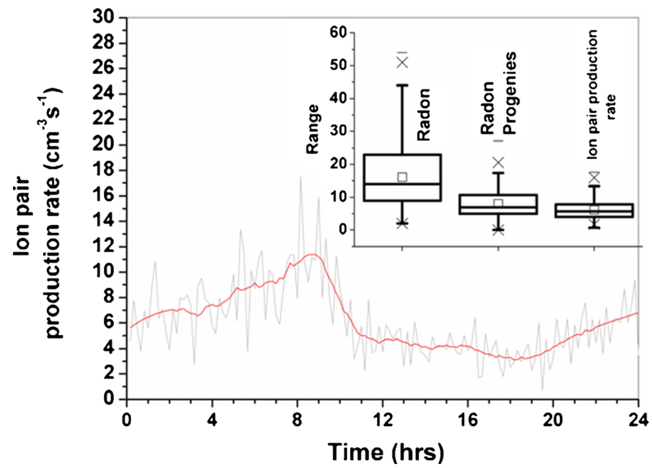

Figure 6. Variation of ion-pair production rate at NARL.

4.4 *Effect of precipitation on radon concentration*

A significant effect of precipitation on radon activity above the surface was observed at NARL during 1–9 December 2012, as shown in figure 7. During the precipitation, water content fills up the pore spaces in the soil. Primarily, the radon originates from the soil through pore spaces; precipitation significantly weakens the diffusion of radon and the reduction in exhalation. Due to this process, the radon concentration is decreased after rainfall. A negative correlation of –0.7 was observed between daily averaged activity of radon and daily rainfall. The radon will regain its diurnal trend, once the regular nocturnal stability, temperature inversions and good daytime mixing conditions are resumed (Fujinami 1996; Seftelis *et al.* 2007; Di Carlo *et al.* 2009).

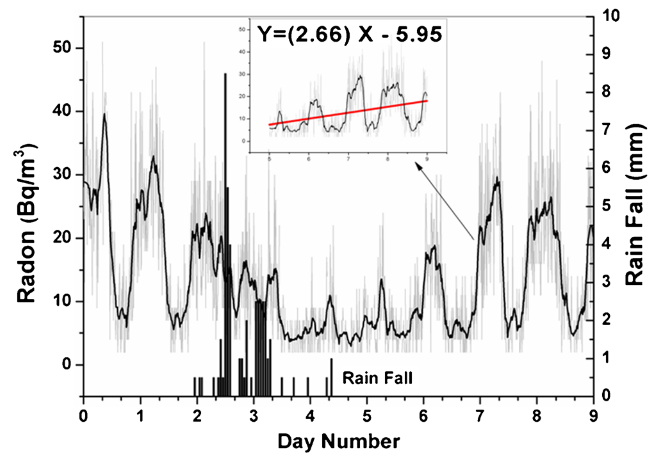

Figure 7. Effect precipitation on radon concentration.

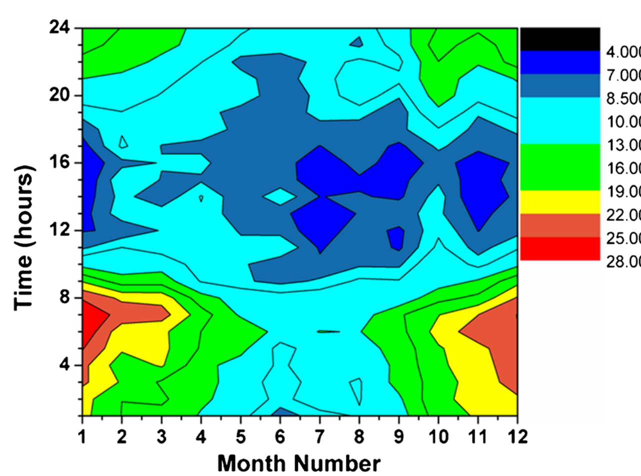

Figure 8. Contour plot of hourly averaged radon for 12 months.

4.5 *Annual variation of radon and its progenies*

The contour plot of hourly-averaged radon for 12 months period is shown in figure 8. From the figure, the average concentration of radon is higher in the winter period (early morning hours) than in the summer period and lower activity was observed in monsoon period (May, June, July) (Nagaraja *et al.* 2003; Prasad *et al.* 2005). The annual statistics of radon, its progenies and meteorological parameters for the study period is shown in table 1.

The whiskers diagram showing the variability of radon for all months is shown in figure 9. The last box shows the average for the entire study period with a maximum of 18 Bq m^{-3}, minimum of 6 Bq m^{-3} and an average of 12.01±0.66 Bq m^{-3}.

Table 1. *Statistics of radon, its progenies and selected meteorological parameters.*

Month	Radon (Bq/m^3)	Radon progenies (Bq/m^3)	Surface air temperature (°C)	Relative humidity (%)	Pressure (mbar)
Jan-12	14.1±0.8	7.5±0.1	24.4	68.4	971.9
Feb-12	13.1±0.7	4.8±0.1	26.1	62.1	970.7
Mar-12	11.3±0.6	3.6±0.2	29.8	57.5	969.1
Apr-12	9.5±0.5	1.6±0.1	32.0	59.5	968.1
May-12	9.0±0.5	2.1±0.1	33.1	52.2	964.7
Jun-12	8.9±0.5	2.0±0.1	31.0	58.7	964.6
Jul-12	9.8±0.5	0.8±0.1	30.7	56.6	961.9
Aug-12	11.0±0.6	3.0±0.2	29.7	64.7	967.6
Sep-12	13.8±0.7	6.6±0.3	30.2	67.2	969.0
Oct-12	13.1±0.8	5.7±0.2	27.9	74.4	971.2
Nov-12	15.4±0.9	7.0±0.3	26.1	71.1	972.4
Dec-12	15.1±0.9	6.3±0.3	24.3	76.0	974.0
Average	12.01±0.66	4.25±0.18	28.8	64.0	968.8

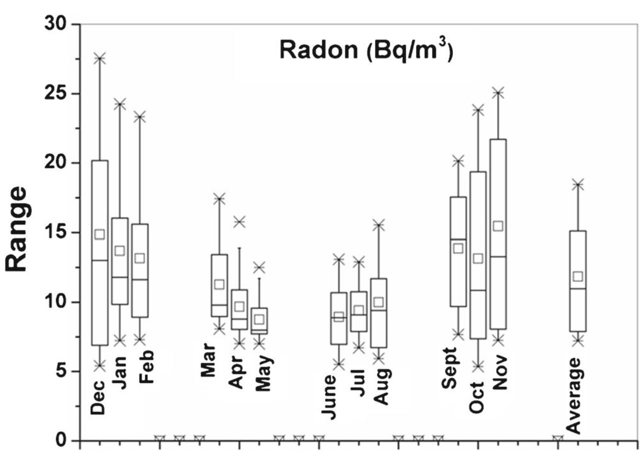

Figure 9. Whiskers diagram showing the monthly variability of radon.

5. Conclusion

A one-year measurement of surface radon and its progeny activity have been analysed along with surface air temperature, relative humidity and pressure. The study was carried out in order to see the impact of temperature and relative humidity on the dynamical mixing and dilution of atmospheric surface radon and on its emission from the ground surface. It has been found that the pronounced correlation coefficient of radon and its progeny activity concentration with temperature and relative humidity confirm the hypothesis that radon is a good indicator of the small-scale stability of the atmospheric surface layer. The instantaneous concentration of radon activity in the atmospheric surface layer is controlled by a coupling of soil emission and convective mixing. Within a fair weather day, the radon concentration has shown high values of the order of 50–60 $Bq\,m^{-3}$, which may be attributed to the rocky hills and dense vegetation area around the measuring site. Precipitation is considered to be able to affect the surface radon evolution at NARL site (negative correlation). The monthly average of radon activity for the study period was found to be 12.01±0.66 Bq m^{-3}, and for radon progeny activity it was found to be 4.25±0.18 $Bq\,m^{-3}$. The overall analysis suggests that longer data records are required in order to evaluate typical behaviour, mean concentrations and trends of the outdoor radon and short-lived radon progeny for a wide range of applications such as natural tracer, atmospheric electricity and pollutant dispersion models (dynamics of radon with trace gases like ozone, NO_x etc.).

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