Open access e-Journal
Earth Science India eISSN: 0974 – 8350
14 (3), 2021, 132- 143
http://www.earthscienceindia.info/

Study of Radioactive Aerosols and Ambient Gamma Radiation Levels at a Rural Site of South India

Charan Kumar K and Kamsali Nagaraja*
Atmospheric and Space Science Research Laboratory, Department of Physics,
Bangalore University, Jnanabharathi Campus, Bengaluru–560056 India
*Email:kamsalinagaraj@gmail.com

ABSTRACT

Simultaneous measurements of radioactive aerosols (radon progenies), gamma radiation dose and meteorological parameters were carried out at National Atmospheric Research Laboratory, Gadanki, India, during 2012. Radon progenies had Pearson correlation coefficients of 0.52, -0.55, and 0.72, respectively, with relative humidity, temperature, and pressure. The correlation coefficients of ambient gamma dosage with relative humidity, temperature, and pressure are, respectively, -0.02, -0.15, and 0.22. For the NARL environment, radioactive aerosols exhibit a 0.24 correlation with gamma dosage. About 85% of Radon progeny data lie below 20 mWL, and for ambient gamma, about 99% of data lie between 135 to 245 nSv/hour. The mean radon progenies were found to be 4.24$\pm$ 2.41 (SD) mWL, and the mean ambient gamma dose levels was found to be 178.7$\pm$15.1 (SD) nSv/hour at NARL, which is well below the limits prescribed by UNSCEAR-2008.

Keywords: Radioactive aerosols, ambient gamma dose, Gadanki, meteorological parameters

INTRODUCTION

Radon (^{222}Rn) is an odourless, colourless, and tasteless radioactive gas. The natural radioactive decay of Uranium, which is found in all rocks and soils, produces ^{222}Rn. The ^{222}Rn is a naturally occurring radioactive gas found in high concentrations in enclosed spaces like homes and offices. The ^{222}Rn gas released into the air from the Earth, where it decays and creates more radioactive particles (WHO, 2011). The ^{222}Rn accumulation in indoor environments and its effects on human health is well reported for the Indian environment (Sathish *et al.,* 2011; Arora *et al.,* 2019). The potential short- and long-term implications of anthropogenic activities on air mass transport and lower atmospheric dynamics have frightened atmospheric physicists worldwide, prompting them to seek new and inventive techniques to comprehend the effects. Because of its unique nature of radioactivity and chemical inertness, researchers in atmospheric physics are gaining interest in studying the outdoor dynamics of ^{222}Rn and its progenies in the lower atmosphere (Kikaj *et al.,* 2019). ^{222}Rn and its isotopes exposure to humans and the accumulation of its progeny within the body is unavoidable. Researchers have attempted to use the basic character of ^{222}Rn and radioactive aerosols to investigate a variety of atmospheric physics research challenges. Radon has a half-life of 3.823 days, comparable to certain air pollutants' residence time and involved in different atmospheric processes (Chambers *et al.,* 2016).

Please cite this article as*: Charan Kumar K and Nagaraja Kamsali (2021) Study of radioactive aerosols and ambient gamma radiation levels at a rural site of south India. Earth Science India, v. 14 (3), pp. 132-143 https://doi.org/10.31870/ESI.14.3.2021.8*

^{222}Rn decays to ^{218}Po with the emission of an alpha particle. The emitted α-particles ionize the atmospheric constituents, and this energy is the primary reason behind the lower atmosphere's weak electrical nature (Nagaraja *et al.,* 2006). Variations in ^{222}Rn impact the atmospheric potential gradient, atmospheric air conductivity, and Air-Earth current values near the Earth's surface. Unlike ^{222}Rn, Polonium has a particle nature and attaches itself to the dust particles and aerosols or suspended particles in the lower atmosphere. Inhalation of such radioactive aerosols is far more dangerous than inhalation of ^{222}Rn, leading to severe health problems such as lung cancer (Kelly *et al.,* 2020). ^{222}Rn immediate decay products have comparatively less half-life until the decay chain reaches stable lead (^{210}Pb). The Radioactive products present in the atmosphere, gaseous and fine particles in nature, transform into radioactive aerosols through coagulation and gas-particle conversion. The behaviour and interaction of these radioactive aerosols with the environment as atmospheric tracers has been reported (Papastefanou *et al.,* 2009). It is essential to study ^{222}Rn alone and its daughters with special emphasis on alpha-emitting radionuclides, and studies on the ^{222}Rn progeny and ambient gamma dose levels are reported for India's different environments (Al Azmi *et al.,* 2019; victor *et al.,* 2019). The simultaneous measurements of radioactive aerosols, ambient gamma dose and meteorological parameters were carried out at National Atmospheric Research Laboratory (NARL, 13.457 °N 79.175 °E), Gadanki, India and results are presented.

SITE DESCRIPTION

The simultaneous measurements of ^{222}Rn progenies, ambient gamma levels and selected meteorological parameters were carried out at NARL, an institute dedicated to studying atmospheric dynamics and its variations. Institute is equipped with MST radar facility, Ray Leigh Doppler Lidar, Ionosonde, Doppler Sodar. The area is surrounded by a complex with dense vegetation and rocky hills. At NARL, both the southwest and northeast monsoons have an impact. The general direction of the wind changes from season to season. The wind is most southerly and southeasterly in April. It is westerly from May to September, and from October to November, it is northeasterly (Kumar *et al.,* 2016; Renuka *et al.,* 2020).

EXPERIMENTAL METHODOLOGY

The measurements of ^{222}Rn decay products (in PAEC) was carried out using Genitron made Alpha Progeny Meter (AlphaPM) in synchronization with AlphaGUARD PQ 2000 PRO (AG). AlphaPM is a new radon progeny meter consisting of an alpha sensitive microchip for progeny concentration measurement. The detection principle is a quasi-continuous sampling of filter paper on which aerosol transported particles are sucked. Alpha particle detecting chip is a CAM300AM semiconductor detector. Once connected to AG, all the data is transferred to data expert software used for administration, evaluating, and output of data acquired. Potential alpha energy concentration is in units of mWL (WL-Working Level), MeV/L, and J/m^3. One working level (WL) is defined as the concentration of short-lived decay products of radon in equilibrium with 3,700 Bq/m^3 in the air (Kumar *et al.,* 2016). The continuous ambient gamma dose measurements were carried out using an integrated Geiger-Muller (GM) tube inside the AG. AG is a professional ^{222}Rn monitoring head based on pulse ionization chamber principle and DSP technology. The sophisticated instrumentation and accurate continuous measurements make it a reference device in calibrating several other passive devices. The GM-tube inside AG is wholly protected, and the measuring range is from 20nSv/hour to 10mSv/hour (Sv-Sievert) with a resolution of 1nSv/hour. AG also consists of temperature, atmospheric pressure and relative humidity sensors, making it an ideal instrument to monitor Radon and ambient gamma dose with the maintenance-free operation (Al Azmi, 2013).

The correctness of the ^{222}Rn concentration values measured with AlphaGuard monitors is assured through multiple references of the instrument calibration to recognized calibration standards. The AlphaPM was calibrated with Am-241, an alpha-emitting source, by the manufacturer. The calibration factor is determined by the efficacy of the detector, the airflow, and the degree of filter absorption (AlphaGuard Manual, 2006). Both AlphaPM and

AG were installed inside the Stevenson screen to avoid the direct effect of solar radiation, rainfall, and wind at the height of 1m from the Earth's surface.

RESULTS AND DISCUSSION

The dynamics of the atmosphere near the ground are affected by the climate. Short-term fluctuations in all meteorological parameters are common and are primarily generated by turbulence in the atmosphere. Solar radiation influences all meteorological factors, either directly or indirectly, resulting in typical daily or yearly trends (Guan *et al.,* 2007; Rao *et al.,* 2013; Rao and Reddy, 2018). Observations were undertaken to analyze fluctuations in ^{222}Rn progenies, ambient gamma dose rate, and temperature to see if incoming solar radiation affects the radioactive aerosols and meteorological variables. Observations are for typical fair-weather days during 19-20, March, 2012 and are shown in Fig. 1.

The temperature gradient from the Earth and lower atmosphere is the fundamental cause of fluctuations in ^{222}Rn activity, its progeny, temperature, and relative humidity. The atmosphere is mainly steady during the night, and convective activity begins to build as the sun rises. It peaks in the afternoon, transporting atmospheric air to higher altitudes due to vertical mixing, and is primarily caused by incoming solar radiation and outgoing longwave radiation from the Earth. But no pronounced diurnal trend is observed in ambient gamma dose levels at 1 m above Earth's surface. After sunrise, a gradual decrease in ^{222}Rn progenies was observed till 2000 hours during 19-20, March, 2012. During this time, the temperature was at its maximum in the late afternoon, between 15:00 and 17:00 hours of IST. The behaviour of ^{222}Rn progenies in the south Indian environment has been thoroughly documented by Nagaraja *et al.* (2003) and Ashok *et al.* (2008). It is caused mostly by the concentration of atmospheric elements near the Earth's surface due to the establishment of a temperature inversion before sunrise. Due to thermal energy, the temperature inversion breaks up after sunrise, and atmospheric constituents and radioactive aerosols travel to higher altitudes, leaving a lower concentration near the surface. The decrease in radioactive aerosol concentration was observed till 20:00 hours on 19-03-2012. After 20:00 hours, an increasing ^{222}Rn progeny activity trend was observed until the sunrise event on 20-03-2012. The accumulation of atmospheric constituents near the surface builds up at night and reaches the maximum in the early morning hours (Chambers *et al.,* 2016). The Pearson correlation coefficient between ambient temperature and radioactive aerosol activity was determined to be -0.61.

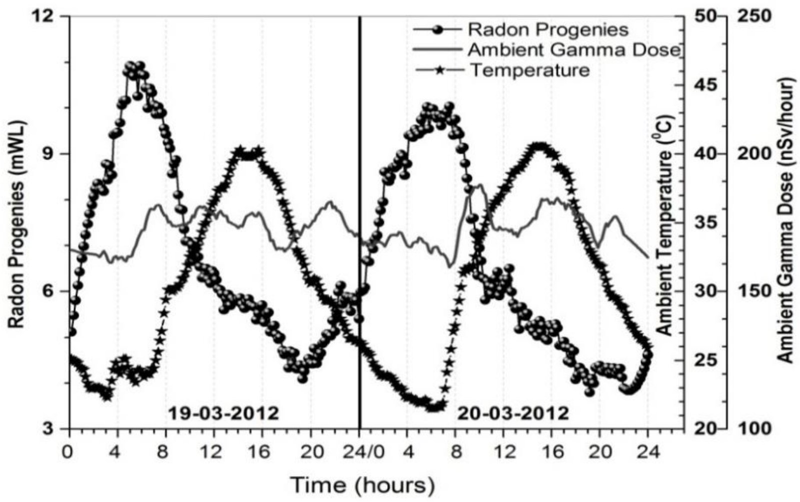

Fig. 1: ^{222}Rn progenies, ambient gamma dose and temperature during 19-20, March, 2020.

The activity concentration of radioactive aerosols shows a positive correlation with relative humidity for the environment of NARL during 19-20, March, 2020, as shown in Fig. 2.

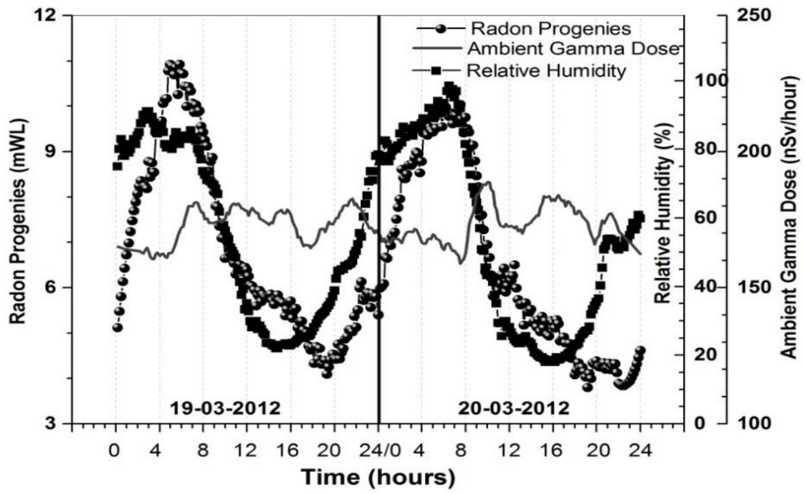

Fig. 2: ^{222}Rn progenies, ambient gamma dose and relative humidity during 19-20, March, 2020.

But no pronounced correlation was found between ambient gamma levels and relative humidity. A positive Pearson's correlation coefficient of +0.78 was found between ^{222}Rn progenies activity and relative humidity (Ashok *et al.,* 2008). Similar diurnal variations in radioactive aerosols were observed for most fair-weather days during the study period. To understand the significance of the effect of meteorological parameters, data points of radon progenies, ambient temperature, relative humidity, air pressure and ambient gamma level collected during fair weather days was analyzed. The scatter plot between ^{222}Rn progenies and ambient temperature yielded a Pearson's correlation coefficient (r) of +0.55 (Ashok *et al.,* 2008; Shetty *et al.,* 2017), and among all the possible fits; the most suitable fitting was linear as shown in Figs. 3-5.

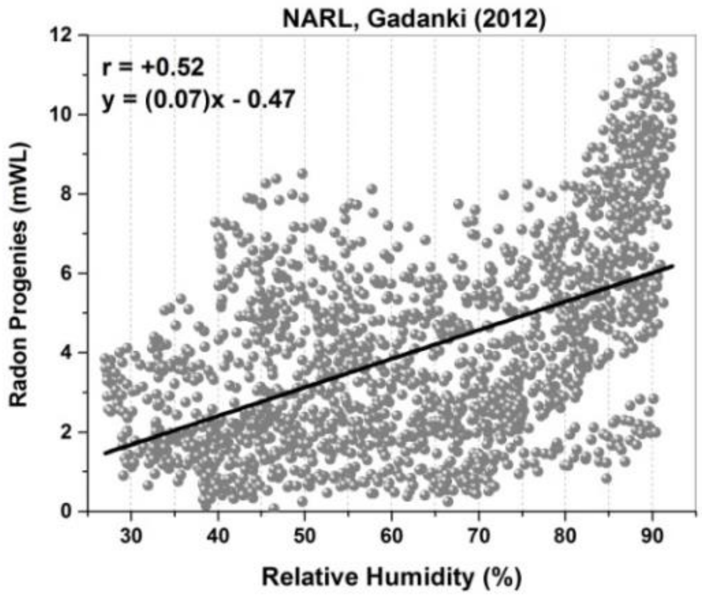

Fig. 3: Scatter plot between ^{222}Rn progenies and relative humidity.

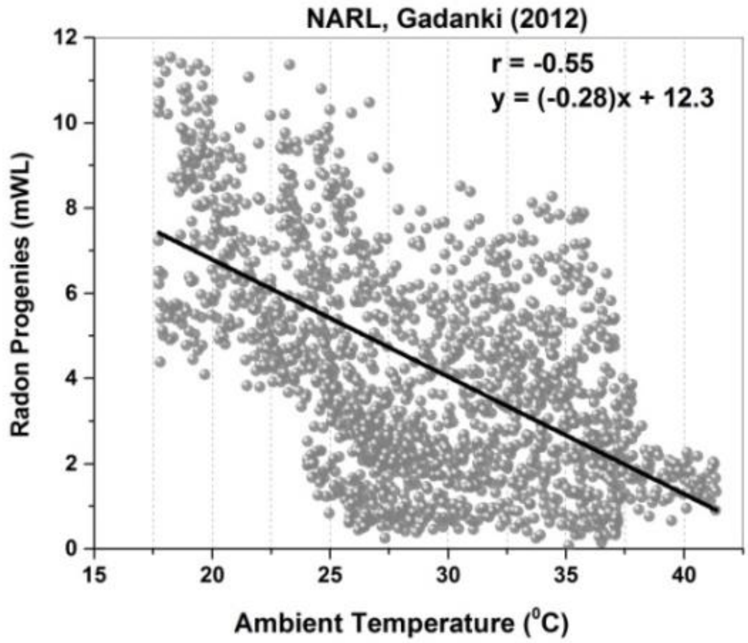

Fig. 4: Scatter plot between ^{222}Rn progenies and temperature.

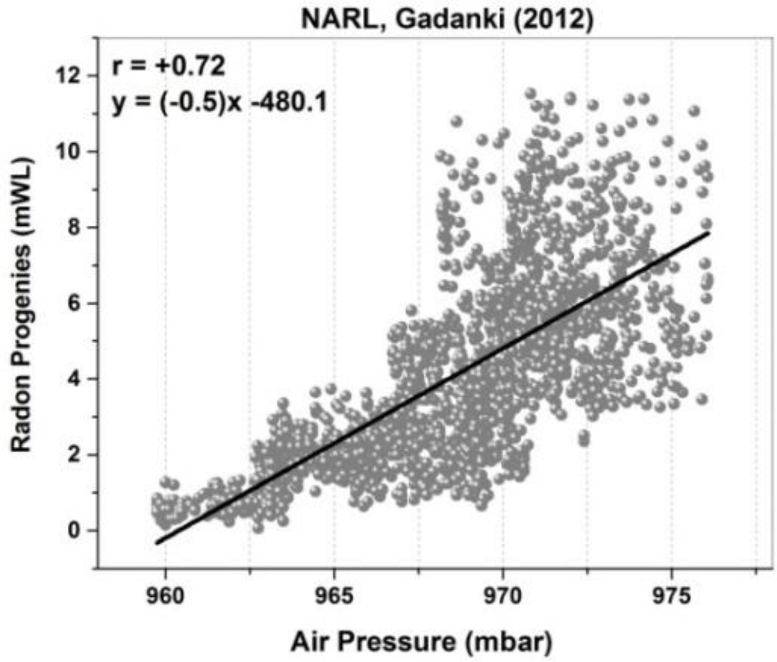

Fig. 5: Scatter plot between ^{222}Rn progenies and air pressure.

Hence, a moderate linear correlation exists between ^{222}Rn progenies and temperature for almost all the fair-weather data points. Similarly, the scatter plot between ^{222}Rn progenies and relative humidity resulted in an r-value of +0.52, as shown in Fig. 3, which is slightly less than that of ambient temperature with linear fitting as the best possible fit. Relatively, a higher Pearson's r of +0.72 was found between ^{222}Rn progeny levels and air pressure, as shown in Fig 4. Hence, it is evident that radioactive aerosols significantly depend on the meteorological factors characterized by solar radiation. Within a fair-weather day, the effect of meteorological parameters on the ambient gamma dose is weak. To explore the possible effect, if any, the relationship between ambient gamma dose and selected meteorological parameters was carried out using scatter plots. It was curious to observe that, over NARL for all the fair weather days, the effect of selected meteorological parameters, *i.e.* ambient temperature, relative humidity, air pressure on ambient gamma dose levels, is weak with a Pearson correlation coefficient of -0.15, -0.02 and +0.22, respectively, as shown in Figs. 6-8.

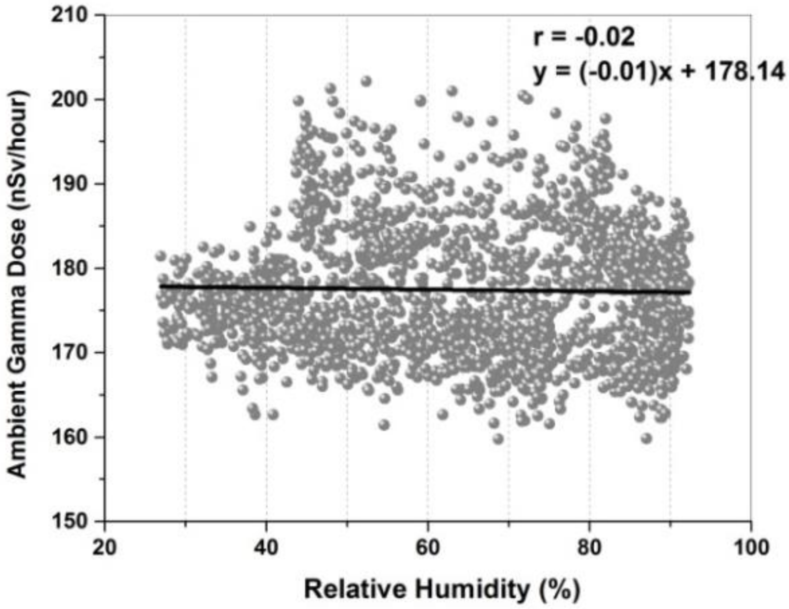

Fig.6: Scatter plot between ambient gamma dose and relative humidity

It justifies that the significant contribution towards ambient gamma dose near the surface is from constantly emitting radiations from the soil surface, and during fair weather days, the variations in gamma dose are constant over time (Shetty *et al.,* 2017). The possible relationship between ambient gamma dose levels and ^{222}Rn progenies activity (radioactive aerosol activity) was investigated, and the scatter plot is shown in Fig. 9.

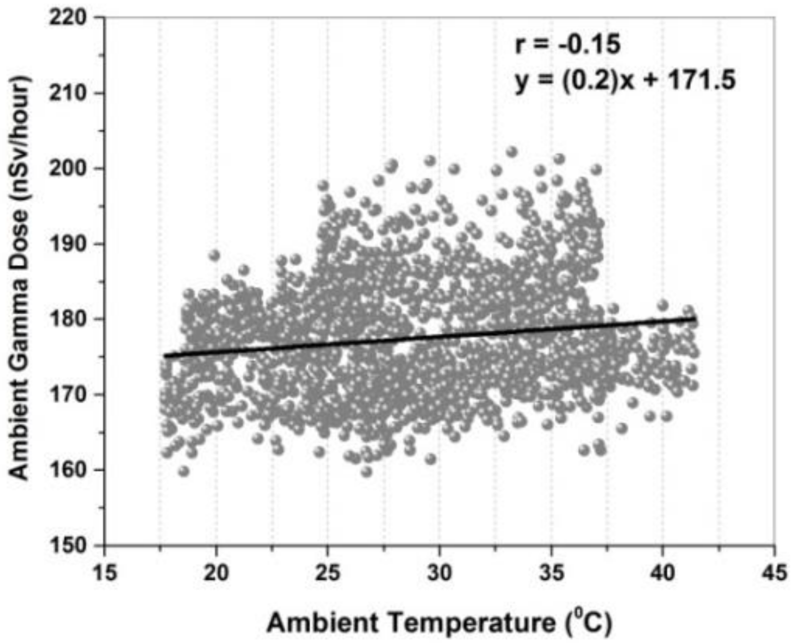

Fig. 7: Scatter plot between ambient gamma dose and temperature.

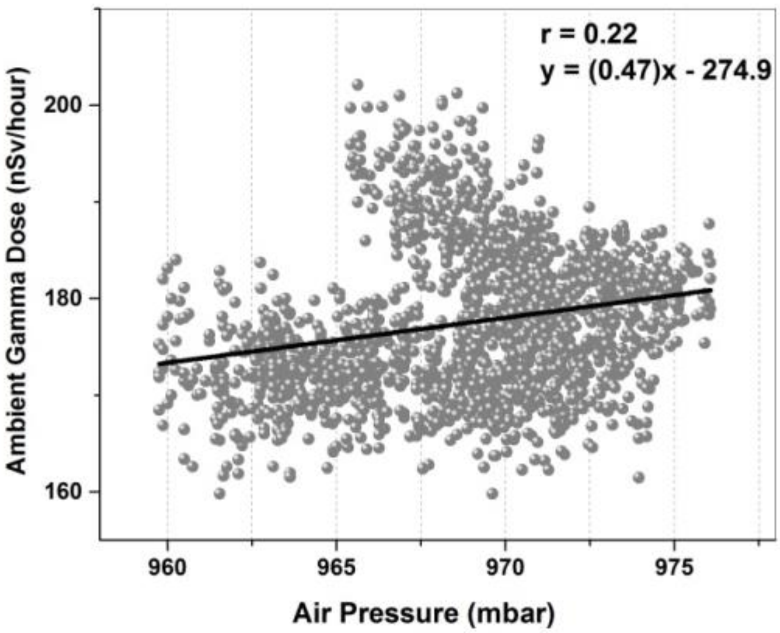

Fig. 8: Scatter plot between ambient gamma dose and air pressure.

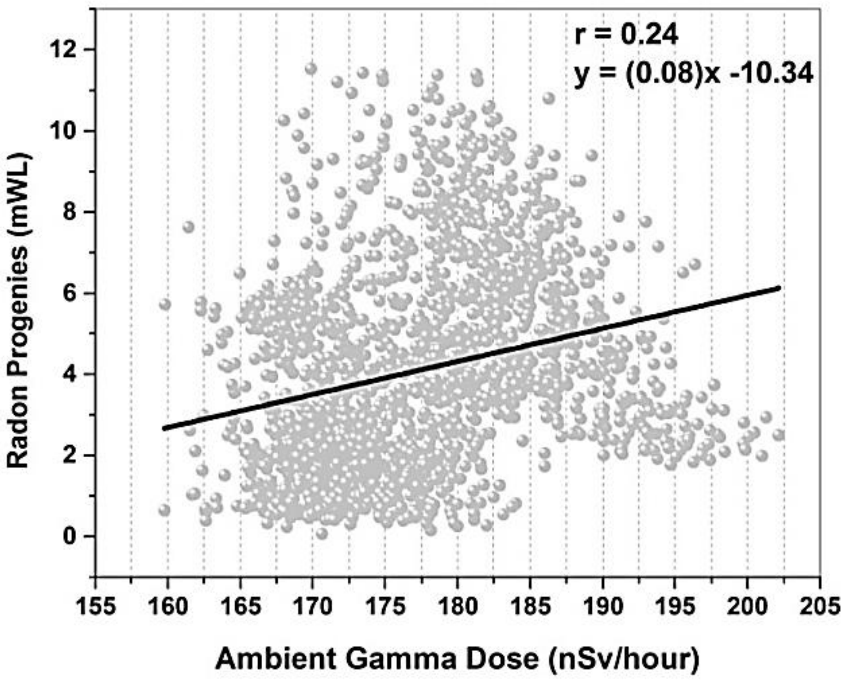

Fig. **9:** Scatter plot between ambient gamma dose and activity of radioactive aerosols.

The results show a weak linear relationship between ^{222}Rn progenies activity and ambient gamma dose levels with Pearson's coefficient of +0.24. The higher Pearson's coefficients for ^{222}Rn progenies with meteorological parameters compared to ambient gamma dose with meteorological parameters indicate that the meteorological parameters mainly control the concentration of radioactive aerosols near Earth's surface on fair weather days. In contrast, ambient gamma dose levels during fair weather days mainly depend on the orography of the study region. To understand the extent to which the activity of ^{222}Rn progenies and ambient gamma dose levels vary, around 25000-30000 fair weather data points were used for the frequency distribution analysis. It was found that about 85% of the activity of ^{222}Rn progeny data points lies below 20mWL. Interestingly, the best possible fit for the frequency distribution was exponential decay with adj r^2 of +0.979, as shown in Fig.10.

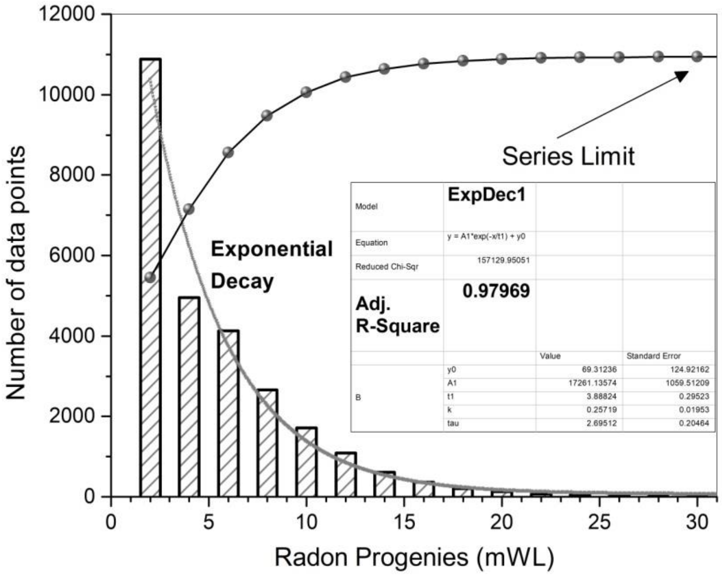

Fig. 10: Frequency distribution of activity of radioactive aerosols at NARL.

Fig. 11 depicts the frequency distribution of ambient gamma dose levels. The best possible fitting of the distribution was Gaussian, with an adj r^2 of 0.999, and 99 percent of data lie between 135 and 245 nSv/hour, indicating narrow background gamma radiation levels over NARL, with a mean value of 178.7±15.1 (SD) nSv/hour. It confirms that the ambient gamma dose level is below 2.4 mSv /year, the permissible limit prescribed by UNSCEAR 2008.

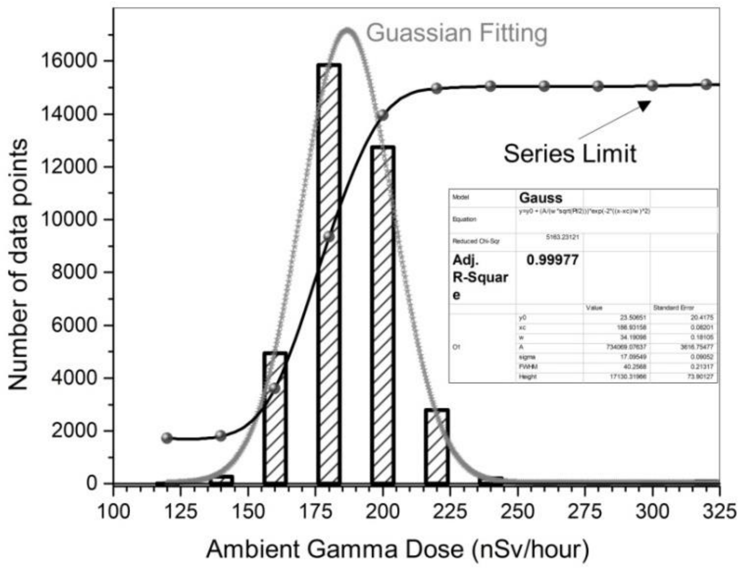

Fig. 11: Frequency distribution of ambient gamma dose levels at NARL.

The time series inspection of the activity of radioactive aerosols shows that over NARL during fair weather days, a distinct diurnal trend in activity was observed. The Fast Fourier Transform (FFT) technique was utilized for frequency domain analysis of ^{222}Rn progeny data to see the periodicity in time series. Because the energy in physics is proportional to the amplitude squared, the frequency domain is the square of FFT's magnitude. FFT is the most fundamental of the Fourier spectral techniques, and it necessitates evenly sampling the data stream for 10 minutes. Frequency domain information is in the amplitude format with N as data size is given by:

$$Amplitude = \frac{Magnitude[FFT(A)]}{N}$$

$$= \sqrt{\frac{[real[FFT(A)]]^2 + [imaginary[FFT(A)]]^2}{N}}$$

Hence, FFT was used to see the possible signatures of hidden memory in the time series of radioactive aerosols and found that, apart from 24 hours' usual diurnal trend, peaks at around 12 hours and 6 hours was also found, as shown in Fig. 12. The observed peaks for ^{222}Rn progenies in FFT may be due to variations in ^{222}Rn activity and attributed to the daily evolution of ^{222}Rn progeny activity, governed by the nocturnal accumulation and mixing of atmospheric boundary layer constituents (Galmarini, 2006). The power as MSA is represented as,

$$\mathrm{P} = \sqrt{\mathrm{mWL/(counts\ per\ day)}^{1/2}}\ .$$

It was reported by several researchers that, during precipitation, the radioactive aerosols in the atmosphere are washed out by the raindrops leading to reduced activity near the surface. But due to the employment of passive techniques, it is challenging to monitor radioactive aerosols during precipitation continuously. Over NARL, continuous monitoring of radioactive aerosols' activity, selected meteorological parameters and rainfall was carried out during 01-09, December 2012, shown in Fig.13 and 14.

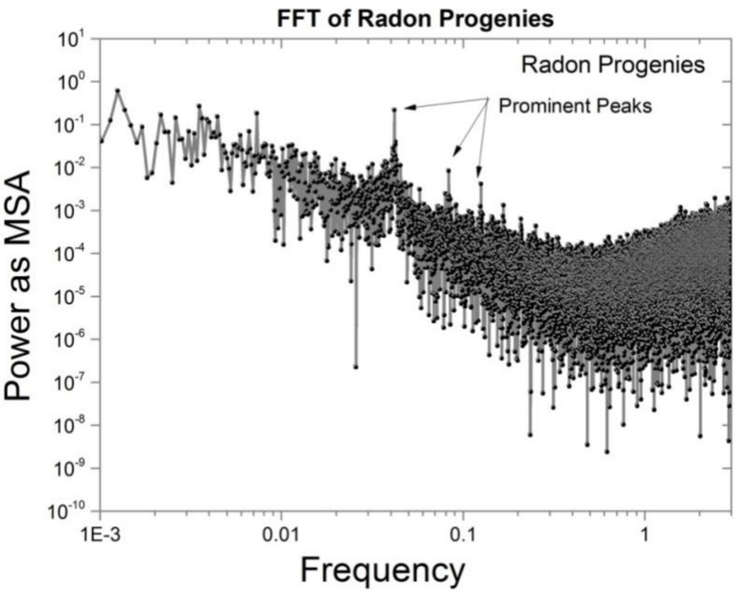

Fig. 12: FFT spectrum of activity of radioactive aerosols.

As reported, before precipitation, a well-defined diurnal trend was observed in the activity of radioactive aerosols. Once the precipitation started, a significant reduction in activity was observed with higher relative humidity levels and intensified precipitation. The reduction in the activity of radioactive aerosols was of the order of 15-20 mWL, and during precipitation, activity concentration values were below 5 mWL. After the precipitation event, it took several days for ^{222}Rn progeny activity to regain its usual diurnal trend. It may be due to a significant reduction in ^{222}Rn exhalation from the soil during and after precipitation (Nagaraja *et al.,* 2003).

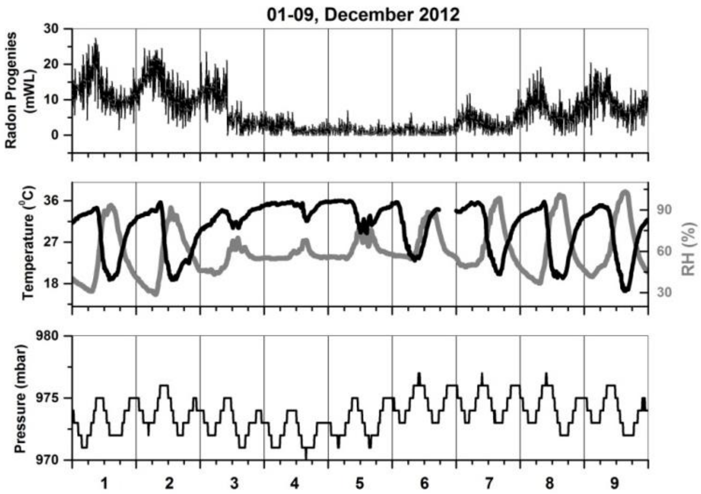

Fig. 13: Variation of ^{222}Rn progeny, temperature, relative humidity and pressure.

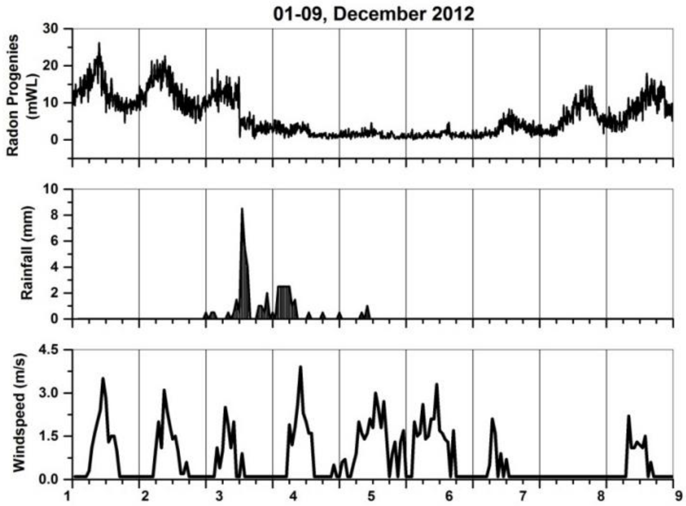

Fig. 14: Daily variation of radioactive aerosols with rainfall and wind speed during 01-09, Dec. 2012.

Table-1: Statistics of all the measured parameters at NARL.

Month (2012)	^{222}Rn **Progeny ± Standard Deviation (mWL)**	**Gamma Dose ± Standard Deviation (nSv/hour)**	**Temperature (^{0}C)**	**Relative Humidity (%)**	**Air Pressure (mbar)**
January	7.45 ± 4.06	177.30 ± 17.09	24.44	68.38	971.85
February	4.80 ± 2.57	170.85 ± 14.34	26.14	62.10	970.70
March	3.63 ± 2.25	178.36 ± 14.75	29.77	57.48	969.11
April	1.57 ± 1.05	172.66 ± 14.85	32.04	59.46	968.09
May	2.11 ± 1.22	172.60 ± 14.36	33.05	52.15	964.65
June	2.03 ± 1.47	172.22 ± 14.45	31.01	58.66	964.57
July	0.76 ± 0.71	171.96 ± 14.22	30.74	56.57	961.90
August	3.01 ± 1.99	198.50 ± 17.25	29.70	64.71	967.57
September	6.58 ± 3.53	186.58 ± 15.57	30.15	67.23	968.96
October	5.66 ± 2.93	181.81 ± 14.61	27.94	74.36	971.18
November	7.02 ± 3.48	181.49 ± 14.70	26.14	71.12	972.44
December	6.29 ± 3.62	180.37 ± 14.52	24.30	76.01	974.03
Average	4.24 ± 2.41	178.72 ± 15.05	28.78	64.02	968.75

The detailed statistics of all the measured parameters are presented in Table-1. It was observed that over NARL, a well-defined monthly variability was observed for ^{222}Rn progeny activity during 2012, and this may be attributed to the significant effect of meteorological parameters on the activity of radioactive aerosols (Nagaraja *et al.,* 2003). But for change in ambient gamma dose levels has not produced any pronounced monthly variability and is attributed to the constant background radiation levels.

CONCLUSIONS

The analysis of radioactive aerosols (^{222}Rn progeny activity), ambient gamma dose and selected meteorological parameters was carried out at NARL, Gadanki, India. All the measured quantities except ambient gamma dose show diurnal and monthly variability. But, no significant relationship was found between ambient gamma dose levels and meteorological parameters. The frequency distribution of ^{222}Rn progenies has shown exponential decay, and Gaussian fitting was most suitable for ambient gamma dose levels. The FFT analysis has revealed the possible memory hidden in the time series of ^{222}Rn progenies. A significant effect of precipitation on the ^{222}Rn progeny activity was found at NARL and may be attributed to the washout of radioactive aerosols from the atmospheric air by the rain droplets. The mean ^{222}Rn progenies activity at NARL was found to be 4.24$\pm$2.41 (SD) mWL, and the mean ambient gamma dose levels at NARL was found to be 178.7$\pm$15.1 (SD) nSv/hour, which is well below the limits prescribed by UNSCEAR.

Acknowledgements: Authors are thankful to Indian Space Research Organization for the financial assistance through the RESPOND project (Grant Number: B 19012/72/2011-II dated 23-12-2011) and Director, National Atmospheric Research Laboratory for the permission to carry out experiments.

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(Received: 04.06.2021; Accepted: 12.07.2021)