Inhalation dose due to radon and its progeny at Pune

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The radon and its progeny concentrations are measured near the surface of the earth at the Indian Institute of Tropical Meteorology, Pune, India. In-situ measurements of the meteorological parameters such as temperature and relative humidity are also made simultaneously. The concentrations show maxima in the early morning hours when the turbulence mixing is the minimum; whereas in the afternoon the turbulence mixing is the maximum and concentrations exhibit minima. The diurnal and seasonal variations in the concentrations of radon and its progeny are found to exhibit correlation with the relative humidity, and anti-correlation with the temperature. The annual effective dose for Pune environment is found to be 0.55 mSv which is within the permissible limit.

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1 Introduction

The exposure of human beings to ionizing radiation from natural sources is a continuing and inescapable feature of life on earth. For most individuals, this exposure exceeds that from all man-made sources combined. There are two main contributors to natural radiation exposures: high-energy cosmic ray particles incident on the earth’s atmosphere and radioactive nuclides that originated in the earth’s crust and are present everywhere in the environment, including the human body itself.

Radon gas and its progeny are naturally occurring radionuclides formed within the decay series of $^{238}\text{U}$ present in all natural sources of the atmosphere. Inhaled radon progeny is the major source of background radiation exposures that account for more than 50% of the radiation dose to the general population. The principle decay modes and half-lives of $^{222}\text{Rn}$ and its short-lived daughters in order are $^{222}\text{Rn}$-$\alpha$, 3.82 days; $^{218}\text{Po}$ (RaA)-$\alpha$, 3.05 min; $^{214}\text{Pb}$ (RaB)-$\beta$, 26.8 min; $^{214}\text{Bi}$ (RaC)-$\beta$, 19.7 min; and $^{214}\text{Po}$ (RaC')-$\alpha$, 164 $\mu$s. Both polonium isotopes are alpha emitters. The $^{222}\text{Rn}$ relatively short-half life (56.6 s) and does not have much time to travel from its production site to the immediate environment of human beings. The relative concentrations of the various radionuclides are strongly affected by dynamical processes, including the attachment of the decay products to aerosol particles.

The $^{222}\text{Rn}$ emanated from the earth’s surface distribute in the atmosphere depending on the geological and meteorological parameters. As temperature inversions occur in atmosphere, $^{222}\text{Rn}$ concentrations will vary accordingly and show diurnal and seasonal variations. Extensive studies on $^{222}\text{Rn}$ concentration and its behaviour in atmosphere along with meteorological parameters such as temperature, humidity, wind velocity, wind direction, atmospheric pressure etc., have been carried out and reported by large number of researchers in India and all over the world. The diurnal/seasonal variations of radon and its progeny concentrations, their dependence on the meteorological parameters are presented for a continental location Pune (18 N, 74 E), India. Also the annual effective dose due to radon and its progeny concentration is estimated.

2 Experimental Details

Many techniques have been established for measuring the concentrations of radon and its progeny in air. These techniques are mainly based on the detection of emissions from radioactive decay of radon and its daughter products. Most of the methods are based on detection of alpha particles; some on detection of gamma emission while a few utilize beta decays. In case of progeny, either the individual decay products concentration or Potential Alpha Energy Concentration (PAEC) is measured. Due to low...
concentrations of environmental radon and its decay products, the precision and accuracy and detecting efficiency of the techniques are the important issues.

2.1 Radon concentration

The concentration of $^{222}$Rn can be measured by various methods. The most commonly used instruments/methods to measure the atmospheric radon concentration are the ionization chambers$^{18}$, Scintillation cells$^{19}$, Two-filter method$^{20}$ and Electrostatic collection method$^{21}$. However, these instruments and methods are not suitable in atmospheric air to measure the $^{222}$Rn concentrations, which are generally very low.

In order to study the diurnal variation of radon and its progeny concentration in atmosphere, sampling duration should be very small and the activity should be determined immediately. In such cases, active devices are helpful. In the present work, in order to determine the diurnal behaviour of $^{222}$Rn and its progeny concentration, active device technique is employed. The most simple, convenient and easy to handle in the field measurements is the Low Level Radon Detection System$^{22}$ (LLRDS).

Radon decays to $^{218}$Po atoms, which on formation are positively charged$^{23}$. They continue to carry positive charges for a small duration even after formation until neutralized by free electrons or until it plates out on condensation nuclei or nearby surfaces. The life span of these free or unattached atoms may vary from few seconds to several minutes, depending on the airborne concentration of condensation nuclei and availability of surfaces nearby to plate on. This property of the born atoms of radon decay products is made use of in the LLRDS.

The LLRDS consists of sample collection chamber of 24 cm diameter and 11.5 cm height and a volume of 5 litres, and is provided with an air inlet and outlet. In this method, the LLRDS is evacuated and air is allowed inside the chamber for about 3–4 min so that the air pressure inside the chamber becomes equal to the atmospheric pressure. After collection of air sample in the chamber, at least 10 min delay is allowed for complete decay of thoron, which may be present in the chamber. A negative potential of ~800 V is applied generally for about 90 min for the saturation of radon daughter atoms on the collection plate. At the end of this period, the plate is removed and counted for alpha activity. The concentration of radon is calculated using the following expression$^{22}$:

$$^{222}Rn = \frac{1000 \times C}{E \times V \times F \times Z} \text{ (Bq m}^{-3})$$

where,

- $C$ is the gross counts;
- $E$ the alpha counting efficiency;
- $V$ is the volume of the LLRDS chamber (litres);
- $F$ the collection efficiency of the $^{218}$Po atoms on the plate and is empirically related to the relative humidity by, $F = 0.9 \times [1 - \exp(0.039 \times H - 4.118)]$; $H$ the relative humidity (%); and $Z$ is the correction factor for build-up of radon progeny on the disc and decay during exposure and counting period.

2.2 Radon daughters concentration

Several methods are available to measure the radon daughters viz. two filter method$^{20}$, Tsivoglou method$^{24}$, Thomas method$^{25}$, Kusnetz’s method$^{26}$ (using active devices) and twin cup dosimeter method employing SSNTD technique$^{27}$ (passive devices). In the present work, radon progeny concentrations in atmosphere have been estimated by the modified Tsivoglou method$^{28}$. The most familiar of the modifications of the original Tsivoglou method was done by Thomas and LeClare$^{20}$. The method consists of collecting a sample of the airborne progeny of radon at a height of one meter from the earth surface, on to a glass micro fiber filter paper (Whatman 2000, 2.5 cm diameter) at known rate of deposition for a specific duration. Typically, the collection rate may vary from a few litres per minute to several tens of litres per minute depending on such factors as required sensitivity, levels of expected airborne activity, equipment and pump available. The sampling time generally is about 15-30 min. These two parameters are not critical except that the sampling for unusually long time (>90 min) is not productive since half-life of the longest lived radon progeny is only 26 min. In 90 min the activity deposited on the filter paper is about 90% of the saturation activity. After collection the filter paper is alpha counted during three intervals. The three counts thus obtained are equated to a set of three simultaneous equations derived theoretically. Solution of these three equations yields the concentration of the airborne radon progeny. A number of modifications of this procedure has been suggested and used in practice$^{4}$. The modifications are mainly in the counting procedures and computation methodology. The expressions for the individual radon progeny as well
as the PAEC or Working Level (WL) can be derived in terms of the sampling, counter and counting parameters viz. $V$, $E$, $C_1$, $C_2$ and $C_3$. The sets of expressions change depending on the sampling and counting intervals. For example, the following expressions are corresponding to a sampling time of 30 min and counting intervals of 2 to 5 min, 6 to 20 min and 21 to 30 min. The counting intervals are so chosen here to correspond to the counting intervals suggested by Thomas and LeClare\textsuperscript{20} as optimum.

$$ RaA = \frac{4.249019\times(C_1) - 2.062417\times(C_2) + 1.949949\times(C_3)}{VE} \quad (\text{Bq m}^{-3}) $$

$$ RaB = \frac{-0.355129\times(C_1) + 0.006232\times(C_2) + 0.240618\times(C_3)}{VE} \quad (\text{Bq m}^{-3}) $$

$$ RaC = \frac{-0.215175\times(C_1) + 0.371319\times(C_2) - 0.502945\times(C_3)}{VE} \quad (\text{Bq m}^{-3}) $$

$$ WL = \frac{0.048445\times(C_1) - 0.019335\times(C_2) + 0.037053\times(C_3)}{VE} \quad (\text{mWL}) $$

where $C_1$, $C_2$ and $C_3$ are the gross counts during the three counting intervals; $E$ the efficiency of alpha counting system; $V$ is the sampling rate in liters per minute (LPM).

**2.3 Estimation of inhalation dose due to radon and its progeny**

The inhalation dose due to radon and its progeny is calculated by using the conversion coefficient $9$ nSv (Bq h m$^{-3}$)$^{-1}$, equilibrium factor $0.6$ and outdoor occupancy factor\textsuperscript{20} $1760$ h. The dose coefficient for radon dissolved in blood is calculated using conversion coefficient $0.17$ nSv (Bq h m$^{-3}$)$^{-1}$ following the inhalation intake. Finally an estimation of the effective dose (m Sv y$^{-1}$) is estimated by using the formula\textsuperscript{29}.

$$ \text{Dose} = [0.17 + 9 \times 0.6] \times 1760 \times 10^{-6} $$

**3 Results and Discussion**

Radium present in the earth crust is the source of atmospheric radon. When radium decays with the emission of alpha particle, the recoil range of radon atom at the instant of formation is about 20-70 nm in mineral, 60 μm in air and 0.1 μm in water. Thus, the emanation of radon atoms from soil particles can cause the radon to recoil (a) within the particle itself or (b) from one particle to other or (c) into the interstitial pores of the soil\textsuperscript{11}. Those recoils that occur through process (a) and (b) will be trapped in the soil particles, while those that occur through the process (c) will escape into the air. The transfer of radon atom from mineral grains to the soil capillaries depends on the water content in the interstitial pores. Some of the recoiled atoms come to rest in the moisture content in the soil pores. Soil moisture plays an important role in the emanation of radon and its diffusion in soil. The soil moisture in interstitial pores directly affects radon emanation by capturing the radon recoiled from the soil matrix. These capture increases the radon atoms to remain in the pore space. Once radon enters the pore space, its partitions between the gas and liquid phases depend on the volume of water in pore space and temperature. The solubility of radon in water decreases with increasing temperature. Both partitioning and increased emanation causes the concentration of radon in air filled pores to be higher under moisture condition than in dry condition. The atmospheric concentrations of $^{222}\text{Rn}$ and its progeny at the ground level are governed by its exhalation rate and atmospheric diffusion depending on meteorological parameters\textsuperscript{11}.

**3.1 Radon concentration in the atmosphere**

Concentrations of radon in the outdoor environment are affected not only by the magnitude of the exhalation rates in the general area but also by atmospheric mixing phenomena. Solar heating during the daytime tends to induce some turbulence, so that radon is more readily transported upwards and away from the ground. During night atmosphere is relatively calm with low winds and little convective motion. Radon exhaled from the soil accumulates near the ground leading to gradual increase in the concentrations.

A typical diurnal variation of the radon concentration on 11 February 1998 for Pune is presented graphically in Fig. 1. The radon concentration in air varies between $4.1-55.2$ Bq m$^{-3}$ with a mean value of $17.6$ Bq m$^{-3}$ showing significant diurnal variation almost by a factor of $13-14$. The
radon concentration in air is found to be maximum during the early morning hours (03:00 to 05:00 hr of Indian Standard Time) and starts to decrease after sunrise, and reaches minimum value during afternoon hours (10:00 to 18:00 hr) and thereafter it increases (Fig. 1). Figure 2 represents the diurnal variation temperature and relative humidity on the same day. The concentration of radon varies proportionately with the relative humidity. This is due to the fact that as temperature increases, the relative humidity decreases resulting in the decrease of moisture content in the atmosphere. This causes increased vertical mixing and raising of aerosols to the higher altitudes which result in lower concentrations at ground level. When the temperature decreases and relative humidity increases, as a result the vertical mixing and raising of aerosols to the higher altitude reduces. As a consequence, the aerosols to which radon and its progeny are attached will be present at higher concentrations at ground level air. This results in the increase of radon concentration in the ground level air.

Diurnal variations in radon concentration are universally ascribed to variations in atmospheric stability. Early morning atmospheric temperature inversions lead to an extremely stable atmosphere. This restricts the vertical turbulent mixing that leads to relatively higher radon concentrations near the earth’s surface. After sunrise, solar radiations warm the lower atmosphere, breaking up the inversion leading to a substantial decline in concentration. Concentrations remain low until late afternoon when radiant cooling of the surface leads to increase in atmospheric stability and a corresponding increase in radon concentration. Several researchers have observed and reported diurnal variations of radon of similar trend. It is seen from the observations, Figs (1 and 2), that the concentration of radon shows a positive correlation with the relative humidity. However, there is a negative correlation coefficient between radon and ambient temperature indicating these two are incoherent. Magalhaes et al. also observed a negative correlation of radon with temperature. The relative humidity is usually associated with periods when wind speed is low and atmosphere is stable. The low winds and atmospheric stability are probably the most important factors resulting in increased concentration of radon.

### 3.2 Radon progeny concentration in the atmosphere

In order to assess the dose due to radon and its progeny, several researchers carried out investigations by measuring the concentrations of radon only. However, it should be noted that, the dose is mainly caused by the short-lived progeny of radon—$^{218}$Po, $^{214}$Pb and $^{214}$Bi. Concerning the radiation dose estimates, the decay product of radon in air is ordinarily not given individual activity concentration, but rather by Potential Alpha Energy Concentration (PAEC). The PAEC of an atom of the decay products is the sum of alpha energies emitted during the decay of the atom up to the $^{210}$Pb. The PAEC of any mixture of activity concentrations of short-lived radon progeny is the sum of potential alpha energies of these atoms present per unit volume of air. For determination of PAEC in air, the activity concentrations of all individual radon decay products have to be known. Therefore, it is important to study the distribution of individual progeny products and equilibrium with their parent radionuclide radon.

A typical diurnal variation of radon progeny in the atmosphere on 11 February 1998 for the environment of Pune is presented graphically in Fig. 3. The atmospheric concentrations of $^{218}$Po, $^{214}$Pb, $^{214}$Bi and PAEC varies from 1.1–36.9 Bqm$^{-3}$, 1.0–2.9 Bqm$^{-3}$, 0.2–3.2 Bqm$^{-3}$ and 0.1–1.3 mWL showing significant diurnal variations of the order of 34, 3, 16 and 13, respectively (Fig. 3). Similar variations of radon progeny concentrations were observed by Jacobi and Andre and reported the variations of the order of 10–100. It is clear from the mean values of radon progeny concentrations presented in Fig. 3 that, the concentration of $^{218}$Po is higher when compared to other two progeny. This is due to the fact that, the $^{218}$Po, being positively charged and immediate progeny of $^{222}$Rn attaches to the atmospheric aerosols and hence contributes significantly to its atmospheric concentration. It is evident from the Fig. 3 that the concentration of radon progeny starts decreasing after 06:00 hr. The concentration decreases and shows minimum during the afternoon between
10:00–18:00 hr, and reaches its maximum during the time period of 04:00–07:00 hr. This trend of variation is consistent with the findings of Porstendorfer\textsuperscript{30} for the environment of Germany. They found that, the large variations of radon progenies in atmosphere during a single day are caused by changes in the eddy diffusivity in the boundary layer. The observed trend of diurnal variation is also in consistence with the trends observed in the case of variation of radon exhalation rates and concentration in atmospheric air, which have been discussed earlier.

The diurnal variation in the radon progeny concentrations can also be attributed to the same reasons as already discussed in previous section. Because of the temperature inversion during early morning hours the radon gas and the aerosols are present in higher concentrations at ground level air, resulting in the higher concentration of radon progeny in air samples near the ground. As the ambient temperature increases, the convective current of the atmospheric air transports the radon gas and aerosols to the upper atmosphere. This results in the decrease of activity concentration of radon and its progeny at the ground level during the period 10:00–18:00 hr. Similar trend of diurnal variations have also been observed and reported by Shenber\textsuperscript{39} for Tripoli, Rangarajan \textit{et al}.\textsuperscript{4} for the environment of Bombay, Wilkening\textsuperscript{31} for USA and a number of researchers for other environs.

### 3.3 Seasonal variation of radon and its progeny

The seasonal variations in the concentrations of radon and its progeny are presented in Fig. 4. It can be observed that the concentrations were maximum during winter and minimum in summer/rainy seasons. The concentration of radon varies between 5.3–24.0 Bq m\(^{-3}\) for summer, 5.5–15.1 Bq m\(^{-3}\) for rainy and 4.7–80.4 Bq m\(^{-3}\) for winter season, respectively. Similarly, the progeny concentration also varies between 0.8–4.7 Bq m\(^{-3}\) for summer, 1.3–7.2 Bq m\(^{-3}\) for rainy and 2.0–11.2 Bq m\(^{-3}\) for winter season, respectively. The activity is higher in winter than in summer/rainy season\textsuperscript{6,36,37,40} since during winter season the radioactive gases are trapped near the surface because of temperature inversions. In summer, the higher rate of vertical mixing and dispersion lifts the aerosols to higher altitudes resulting in a decrease in the concentration near the ground level air. During monsoon, the density of aerosols in the upper atmosphere is low as compared to the earth’s surface through wet precipitation. Consequently there is a reduction in concentration of radon and its progeny concentration in monsoon. Wilkening\textsuperscript{31} reported similar trend of variations for the environment of USA. Magalhaes \textit{et al}.\textsuperscript{37} have observed a two order of magnitude of variability, with a maximum of 50 Bq m\(^{-3}\) in winter and a minimum of 0.5 Bq m\(^{-3}\) in the summer months. In addition, radon exhalation rate also decreases during monsoon as soil pores get filled by water and hence, resulting in lower concentration of radon progeny in air.

### 3.4 Radiation dose due to outdoor radon

For the estimation of the annual effective dose due to outdoor radon an occupancy factor of 0.2 is used\textsuperscript{1,29}. Fig. 5 shows the seasonal variation of the concentration of radon progeny PAEC.
concentrations of radon, its progeny and the inhalation dose. The average concentrations of radon and its progeny are 11 and 1.8 Bq m\(^{-3}\) for summer, 9.40 and 3.5 Bq m\(^{-3}\) for rainy, and 28.5 and 5.7 Bq m\(^{-3}\) for winter season, respectively. Similarly the dose due to radon and its progeny concentration for summer, rainy and winter seasons are 0.11, 0.09 and 0.28 mSv y\(^{-1}\), respectively. The average annual effective dose from the inhalation of radon for a person who lives in the environment of Pune is found to be 0.55 mSv which is within the permissible limit. In the past, Debaje et al.\(^{34}\) had also estimated and found the annual dose of 0.58 mSv for the environment of Pune.

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