



# Diurnal variations of $^{218}\text{Po}$ , $^{214}\text{Pb}$ , and $^{214}\text{Po}$ and their effect on atmospheric electrical conductivity in the lower atmosphere at Mysore city, Karnataka State, India



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## ABSTRACT

The short-lived radon daughters ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ ) are natural tracers in the troposphere, in particular near the ground surface. They are electrically charged particles and are chemically reactive. As soon as they are formed they get attached to the aerosol particles of the atmosphere. The behavior of radon daughters is similar to that of aerosols with respect to their growth, transport and removal processes in the atmosphere. The electrical conductivity of the atmosphere is mainly due to the presence of highly mobile ions. Galactic cosmic rays are the main source of ionization in the planetary boundary layer; however, near the surface of the earth, ions are produced mainly by decays of natural radioactive gases emanating from the soil surface and by radiations emitted directly from the surface. Hence the electrical conductivity of air near the surface of the earth is mainly due to radiations emitted by  $^{222}\text{Rn}$ ,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ , and depends on aerosol concentrations and meteorological parameters. In the present work the diurnal and seasonal variations of radon and its progeny concentrations are studied using Low Level Radon Detection System and Airflow Meter respectively. Atmospheric electrical conductivity of both positive and negative polarities is measured using a Gerden Condenser. All the measurements were carried out simultaneously at one location in Mysore city ( $12^\circ\text{N}$ ,  $76^\circ\text{E}$ ), India. The diurnal variation of atmospheric electrical conductivity was found to be similar to that of ion pair production rate estimated from radon and its progeny concentrations with a maximum in the early morning hours and minimum during day time. The annual average concentrations of  $^{222}\text{Rn}$ ,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Po}$  at the study location were found to be 21.46, 10.88, 1.78 and 1.80  $\text{Bq m}^{-3}$  respectively. The annual average values of positive and negative atmospheric electrical conductivity were found to be 18.1 and 16.6  $\text{f S m}^{-1}$  respectively. The radon and its progeny concentrations are higher in winter than in summer and rainy season.

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## 1. Introduction

Radon is a radioactive gas, formed in the decay chain of naturally occurring primordial radionuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{235}\text{U}$  present in the earth's crust.  $^{222}\text{Rn}$  formed from the  $^{238}\text{U}$  series has a half-life of 3.823 days;  $^{220}\text{Rn}$  also called as thoron, is formed from the  $^{232}\text{Th}$  series has a half-life of 55.6 s.  $^{219}\text{Rn}$  also called as actinon is emanating from  $^{235}\text{U}$  series has a half-life of 3.96 s. The relative abundances of the isotopes in natural uranium by weight are  $^{238}\text{U}$  – 99.28% and  $^{235}\text{U}$  – 0.71%. On the other hand,  $^{232}\text{Th}$  is even more abundant than  $^{238}\text{U}$  in the earth's crust. When radon is formed, it

may diffuse from the rocks and soils to enter the atmosphere. The amounts of  $^{222}\text{Rn}$ ,  $^{220}\text{Rn}$  and  $^{219}\text{Rn}$  in the atmosphere depend primarily on their half life and on the concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{235}\text{U}$  in the earth crust. The fraction that emanates to the atmosphere depends on the type of rock or soil matrix, water content, atmospheric pressure and other geological and climatic factors.

The short-lived daughters of  $^{222}\text{Rn}$  ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ ) are natural tracers in the atmosphere, in particular at the boundary layer near the ground (Baskaran, 2011). The distribution of  $^{222}\text{Rn}$  and its daughters have been widely used as tracers to understand and quantify several atmospheric processes such as: i) source tracking and transport time scales of air masses; ii) stability and vertical movement of air masses; iii) removal rate constants and residence times of aerosols; iv) chemical activities of analog species; v) deposition rates and washout ratios of aerosol particles; vi)

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sources of continental dust in an air mass; and vii) processes of attachment of metal ions to atmospheric aerosols (Baskaran, 2011; Turekian and Graustein, 2003).

The concentration of  $^{222}\text{Rn}$  daughters in the atmosphere is governed by their rate of production, rate of decay and removal by scavenging (Papastefanou, 2009). They are electrically charged particles and are chemically reactive. As soon as they are formed they attach to the aerosols of the atmosphere (Porstendorfer et al., 1991). The behavior of  $^{222}\text{Rn}$  daughters is similar to that of the behavior of the aerosols with respect to growth, transport and removal processes in the atmosphere.

The electrical conductivity of the atmospheric air is mainly due to the presence of highly mobile small ions according to the relation:

$$\sigma = \sigma^+ + \sigma^- = e \left( \sum_i n_i^+ k_i^+ + \sum_i n_i^- k_i^- \right)$$

where  $\sigma^+$  and  $\sigma^-$  are the positive and negative conductivities respectively,  $n^+$  and  $n^-$  are the positive and negative small ions,  $k^+$  and  $k^-$  are the corresponding their mobility and  $e$  is the fundamental electric charge.

Galactic cosmic rays are high-energy particles coming from outside the solar system and are the main source of ionization in the planetary boundary layer; however, near the surface of the earth, ions are produced mainly by decays of natural radioactive gases emanating from the soil surface and by radiations emitted directly from the surface. Cosmic rays cause about 20% of the ionization at the ground level. The rate of ionization at any location on the surface of earth due to cosmic rays is almost constant with time, and varies between 1 and 2 ion-pairs  $\text{cm}^{-3} \text{s}^{-1}$  (Chalmer, 1967; Israelson and Tamm, 2001). Therefore any temporal variations observed in the rate of ionization at lower layers of the atmosphere is almost entirely due to the variations in the concentrations of natural radioactive substances (Ragini et al., 2009). The ionization due to terrestrial radioactive sources normally decreases rapidly with altitude, and at about 1 km its contribution to the total ionization is less than that from cosmic rays.

Historically speaking the discovery of the presence radioactivity in the atmosphere by Elster and Geitel helped in understanding the presence of atmospheric ions. Apart from electrical gradient above the surface of Earth, air conductivity near the surface of Earth is the most frequently measured quantity in atmospheric electricity. Air conductivity is strongly influenced by aerosol pollution; in aerosol polluted air conductivity is considerably reduced by ion-aerosol attachment (Dhanorkar and Kamra, 1997).

The objective of the present work is to estimate the ion pair production rate due to the alpha particles emitted by radon and its daughter products and relate it with the measured atmospheric electrical conductivity. The study of radioactivity and atmospheric electrical conductivity are important in understanding the atmospheric electrical phenomenon. Mysore City ( $12^\circ\text{N}$  and  $76^\circ\text{E}$ ), Karnataka State, India is a low latitude station and has importance in the study of global electric circuit. Measurement of atmospheric conductivity helps in understanding the level of aerosol pollution in the air. Of late air in the city of Mysore is getting polluted due to enormous increase in automobile traffic since the city has become an important tourist destination and also due to establishment of large number of medium scale industries like chemical, textile, electronics, automobile and distillery around the city. Therefore an attempt has been made to understand the air pollution level in Mysore with the measurement of atmospheric conductivity in addition to the estimation of the concentration of radioactive

elements that are responsible for creating atmospheric ions. A study like this requires investigating radon, its progeny concentrations, electrical conductivity along with meteorological parameters on different time scales in particular close to the surface of the Earth. Although a few measurements have been done at the experimental location, an extensive and detailed investigation of this kind has been done for the first time.

## 2. Materials and methods

In order to study the diurnal variation of radon and its progeny concentration in the atmosphere, sampling duration should be very small and the activity should be determined immediately. In the present work, the  $^{222}\text{Rn}$  concentration is measured using Low Level Radon Detection System (LLRDS) and  $^{222}\text{Rn}$  progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Po}$ ) concentration is measured using Airflow Meter. Atmospheric electrical conductivity is measured using Gerdien condenser. All the measurements were carried out simultaneously at 1 m above the ground surface in an open field near the Department of Studies in Physics, University of Mysore, Mysore, Karnataka State, India. The measurement was carried out continuously at every 2 h intervals on fair-weather days during 2012–2013. Meteorological parameters such as, atmospheric temperature, pressure, relative humidity, wind speed, wind direction and rainfall are also recorded.

### 2.1. Low Level Radon Detection System

The schematic diagram of LLRDS is shown in Fig. 1. It consists of a cylindrical sample collection chamber of 24 cm diameter, 11.5 cm height with a volume of 5 L, and is provided with an air inlet and outlet. In this method, the LLRDS is evacuated and air from the experimental site is allowed to enter the chamber by pressing the swage connector 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 swage connector blocks the entry of outside air to the chamber and also avoids leakage of air from the chamber. Then at least 10 min delay is allowed for complete decay of thoron, which may be present in the chamber.  $^{222}\text{Rn}$  decays to  $^{218}\text{Po}$  atoms, which on formation are positively charged. They continue to carry positive charge for a small duration even after formation until they are neutralized by free electrons or nearby surfaces or until it settles on the charged plate. The

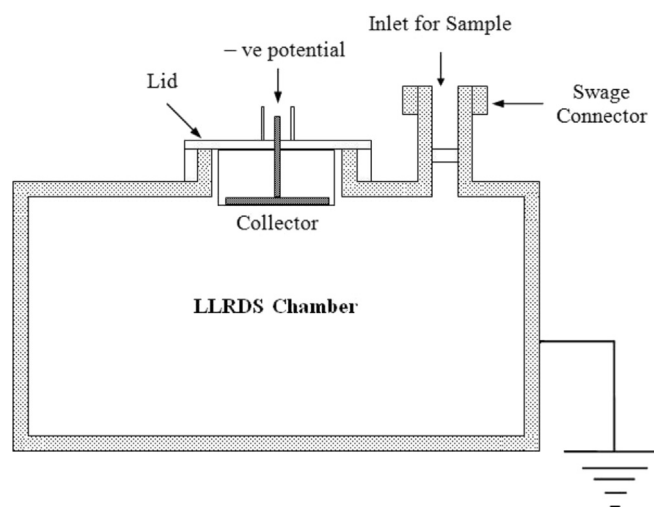


Fig. 1. Schematic diagram of the Low Level Radon Detection System.

life span of these free or unattached atoms may vary from a few seconds to several minutes, depending on the airborne concentration of condensation nuclei and availability of surfaces nearby to plate on (Srivastava et al., 1984). A negative potential of  $\sim 800$  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, assumed to be due to  $^{218}\text{Po}$ . The concentration of radon is calculated using the following expression (Srivastava et al., 1984; Chandrashekhara et al., 2006):

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

where,

- $C$  – is the gross counts
- $E$  – is the alpha counting efficiency
- $V$  – is the volume of the LLRDS chamber (L)
- $F$  – is the collection efficiency of the  $^{218}\text{Po}$  atoms on the plate and is empirically related to the relative humidity through,  $F = 0.9 * [1 - \exp(0.039 * H - 4.118)]$
- $H$  – is the relative humidity (%)
- $Z$  – is the correction factor for build up of radon progeny on the disc and decay during exposure and counting period. This is estimated to be 5000 (Srivastava et al., 1984).

## 2.2. Air sampling for radon progeny measurements

An airflow meter kept at a height of 1 m above the ground surface was used to measure the radon progeny concentration. Airflow meter consists of a 15 cm long and 1 cm diameter tube made up of Perspex. The method consists of collecting a sample of the airborne radon progeny on a glass fiber filter paper. A vacuum pump is connected to one end of the tube and the air is sucked typically for a period of 30 min. The collection rate may vary from a few liters per minute to several tens of liters per minute depending on such factors as required sensitivity, levels of expected airborne activity, equipment and pump available. In the present study the flow rate was about 40 L per minute. These parameters are not crucial except that the sampling for a very long time ( $>90$  min) is not fruitful since half life of the longest lived radon progeny is only 26 min. The filter paper was then alpha-counted at any specific delay time. Total activity on the filter paper is measured at three different counting intervals of 2–5, 6–20 and 21–30 min. Activities of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Po}$  are calculated using the equations (Kusnetz, 1956; Raghavayya, 1998):

$$C_{218\text{Po}} = \frac{4.249019(C_1) - 2.062417(C_2) + 1.949949(C_3)}{VE} \quad (\text{Bq m}^{-3})$$

$$C_{214\text{Pb}} = \frac{-0.355129(C_1) + 0.006232(C_2) + 0.240618(C_3)}{VE} \quad (\text{Bq m}^{-3})$$

$$C_{214\text{Po}} = \frac{-0.215175(C_1) + 0.371319(C_2) - 0.502945(C_3)}{VE} \quad (\text{Bq m}^{-3})$$

where

- $C_1, C_2$  and  $C_3$  are the gross counts during the three counting intervals
- $E$  is the efficiency of alpha counting system (26%)
- $V$  is the sampling rate in liter per minute

## 2.3. Estimation of ion pair production rate

The total energy released  $\epsilon$  ( $\text{eV cm}^{-3} \text{s}^{-1}$ ) due to both radon and its progeny concentration is computed using the expression (Hoppel and Frick, 1986):

$$\epsilon = (5.49 \times C_{222\text{Rn}} + 6.00 \times C_{218\text{Po}} + 0.85 \times C_{214\text{Pb}} + 7.69 \times C_{214\text{Po}}) \times 10^6$$

from which the ion-pair production rate  $Q = \epsilon / 32$  ion pairs  $\text{cm}^{-3} \text{s}^{-1}$  is calculated.

## 2.4. Gerdien condenser

Atmospheric electrical conductivity was measured using a Gerdien condenser (Dhanorkar et al., 1989). The schematic diagram of Gerdien condenser is shown in Fig. 2. It consists of cylindrical tube of 10 cm diameter and 41 cm length and an inner co-axial electrode of 1 cm diameter and 20 cm length. A dc potential of 35 V is applied to the outer electrode and inner electrode is used as a collector. The positive potential applied to the outer electrode will repel the positive ions towards the inner electrode generating an electrical pulse. The electrical signal received from the inner electrode was amplified and collected using data logger card and recorded on a computer. Two sets of Gerdien condensers were used for the simultaneous measurement of atmospheric electrical conductivity due to positive and negative ions. The conductivity is given by

$$\sigma = \frac{i\epsilon_0}{CV}$$

where  $C$  is the capacitance of the Gerdien condenser and  $V$  is the voltage applied across the electrodes. The critical mobility of ions in the instrument was greater than  $10^{-4} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$  and was capable of resolving the values of conductivity as small as  $0.3 \text{ f S m}^{-1}$  (Dhanorkar et al., 1989). To protect from the external electrical fields, the Gerdien condenser was insulated and kept inside second cylinder which was electrically grounded. The measurement was done continuously on all fair-weather days for a period of 2 years. We made 60 samples per second and then it was averaged for 30 s and average values of positive and negative conductivity are recorded in a computer. For any desired length of time the data is averaged and analyzed with the help of an external program.

## 3. Results and discussion

### 3.1. Diurnal variations

The diurnal variations of radon, its progeny ( $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ , and  $^{214}\text{Po}$ ) concentrations, ionization rate due to radon and its progeny

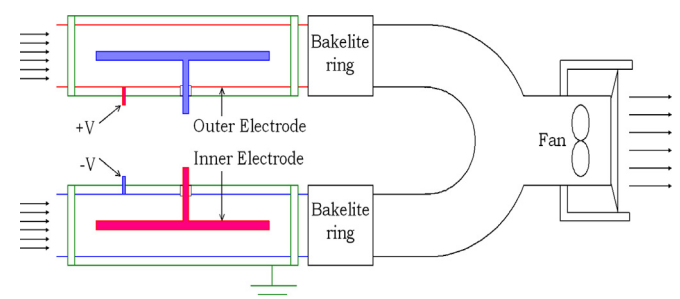


Fig. 2. Schematic diagram of Gerdien Condenser.

and atmospheric electrical conductivity for a typical day during December 2012 are shown in Table 1. The annual average radon concentration varied from 9.65 to 37.70 Bq m<sup>-3</sup> with a median of 19.73 Bq m<sup>-3</sup> showing significant diurnal variation by a factor of 4. The concentrations were maximum during the early morning hours (Table 1), between 04 and 06 h of Indian Standard Time (IST) at Mysore. It decreased after sunrise, attaining a minimum between 10 and 15 h. The individual concentrations of radon progeny were measured since each will contribute separately in the ionization of the atmosphere. Ionization rate estimated from radon and its progeny concentrations at the study location varies from 2.81 to 10.92 ion-pairs cm<sup>-3</sup> s<sup>-1</sup> (Hoppel and Frick, 1986).

During nighttime the atmosphere is relatively calm with low winds and virtually no convective motion. Thus the radon exhaled from the soil accumulates near the ground leading to gradual increase in the concentrations, which inturn increases the ionization rate and atmospheric electrical conductivities.

After sunrise due to convective motion radon and its progeny concentrations move upwards leading to decrease in concentrations near ground surface (Porstendorfer et al., 1991; Shenber, 1992). This will decrease the ion production rate near earth's surface during day time. The electrical conductivity of the atmosphere is determined by the number and mobility of the charge carriers present. During day time the UV radiation generates ultra fine aerosol particles (Kikas et al., 1996). These particles will also reduce small air ions in the atmosphere by ion-aerosol recombination process. This process also results in depression of conductivity during day time.

### 3.2. Seasonal variations

The variation of radon, its daughter products ion pair production rate and atmospheric electricity during winter, summer and rainy seasons are shown in Fig. 3. The concentration of radon and its progeny are maximum in winter followed by rainy and summer seasons. The maximum and minimum levels attained are almost the same in all the seasons. Ion pair production rate varied from 2.9 to 13.5 ion-pairs cm<sup>-3</sup> s<sup>-1</sup> in winter, and during summer and rainy season it varied from 2.4 to 7.8 and 3.1 to 11.4 ion-pairs cm<sup>-3</sup> s<sup>-1</sup> respectively. The temperature inversion during winter leads to the accumulation of more radon near the ground surface thus increasing ionization rate. During summer the gases will move upward carrying radon with it and thereby reducing ionization near the surface which inturn reduces conductivity (Sannappa

et al., 1999; Nagaraja et al., 2003). The annual average concentrations of <sup>222</sup>Rn, <sup>218</sup>Po, <sup>214</sup>Pb, and <sup>214</sup>Po are 21.46, 10.88, 1.78 and 1.80 Bq m<sup>-3</sup> respectively.

The average value of atmospheric electrical conductivity varied from 16 to 77 f S m<sup>-1</sup> in winter, and during summer and rainy season it varied from 13 to 64 f S m<sup>-1</sup> and 13 to 44 f S m<sup>-1</sup> respectively. In fair-weather condition, atmospheric electrical conductivity near the ground surface is governed by the small ions having the sizes of less than 0.1 μm and indicates the level of air pollution in that region. The total conductivity varies from 2 f S m<sup>-1</sup> for highly polluted region to 20 f S m<sup>-1</sup> relatively clean regions (Cobb and Wells, 1970; Sheftel et al., 1994). The annual average value of atmospheric electrical conductivity at our study location was found to be 34.8 f S m<sup>-1</sup>. This will indicate that the study location is relatively clean from air pollution.

During all the seasons, conductivity showed similar diurnal variation with higher values during night and in the early morning and lower values after sunrise. In summer and rainy season the conductivity was lower compared to that in winter. This is due to higher concentrations of radon and its progeny resulting in higher ionization rates in winter than in summer and rainy seasons. The ion pair production rate due to radon and its progeny concentration in rainy season was higher than summer season; whereas the observed atmospheric electrical conductivity values are slightly higher in summer season than in rainy season. During rainy season even though ionization takes place over the ground level, the ions are washed away due to rain resulting in decrease in the conductivity values. The atmospheric electrical conductivity shows good correlation with ion pair production rate with a correlation of 0.95 ( $n = 12$ ). This indicates that the measured value of atmospheric electrical conductivity data can be used as to know the level of radioactive elements in the earth crust and atmosphere.

### 4. Conclusion

The diurnal and seasonal variations of radon, its progeny, ion pair production rate and atmospheric electrical conductivity are studied at one location in Mysore city, Karnataka State, India. The average concentrations of <sup>222</sup>Rn, <sup>218</sup>Po, <sup>214</sup>Pb, and <sup>214</sup>Po in Mysore city were found to be 21.46, 10.88, 1.78 and 1.80 Bq m<sup>-3</sup> respectively. The annual average value of atmospheric electrical conductivity was found to be 34.8 f S m<sup>-1</sup>. This will indicate that the study location is relatively clean from air pollution. The atmospheric electrical conductivity show diurnal variation with maximum in

**Table 1**

Annual average diurnal variation of radon, its progeny concentration, ion production rate and atmospheric electrical conductivity.

| Time (hours) | Concentrations (Bq m <sup>-3</sup> ) |                   |                   |                   | Ionization rate<br>(no cm <sup>-3</sup> s <sup>-1</sup> ) | Conductivity (f S m <sup>-1</sup> ) |          |       |
|--------------|--------------------------------------|-------------------|-------------------|-------------------|---|-------------------------------------|----------|-------|
|              | <sup>222</sup> Rn                    | <sup>218</sup> Po | <sup>214</sup> Pb | <sup>214</sup> Po |   | Positive                            | Negative | Total |
| 0            | 18.46                                | 11.98             | 1.56              | 1.80              | 5.89  | 21.68                               | 20.12    | 41.80 |
| 2            | 23.41                                | 13.84             | 2.49              | 1.61              | 7.06  | 24.40                               | 23.17    | 47.57 |
| 4            | 35.35                                | 19.66             | 3.35              | 2.22              | 10.38   | 30.20                               | 28.87    | 59.07 |
| 6            | 37.70                                | 19.58             | 3.35              | 2.86              | 10.92   | 31.44                               | 30.19    | 61.63 |
| 8            | 20.66                                | 9.60              | 2.62              | 0.89              | 5.63  | 16.72                               | 15.18    | 31.90 |
| 10           | 14.13                                | 6.75              | 1.64              | 1.71              | 4.14  | 12.66                               | 11.21    | 23.87 |
| 12           | 9.65                                 | 3.56              | 0.57              | 1.97              | 2.81  | 10.12                               | 8.95     | 19.07 |
| 14           | 12.07                                | 5.66              | 0.89              | 1.83              | 3.60  | 7.26                                | 6.61     | 13.87 |
| 16           | 17.83                                | 7.82              | 1.06              | 1.66              | 4.95  | 10.96                               | 9.78     | 20.73 |
| 18           | 18.79                                | 8.20              | 1.39              | 1.94              | 5.26  | 15.53                               | 12.84    | 28.37 |
| 20           | 25.69                                | 11.77             | 1.31              | 1.40              | 6.98  | 16.53                               | 14.70    | 31.23 |
| 22           | 23.77                                | 12.16             | 1.14              | 1.73              | 6.80  | 19.86                               | 18.11    | 37.97 |
| Minimum      | 9.65                                 | 3.56              | 0.57              | 0.89              | 2.81  | 7.26                                | 6.61     | 13.87 |
| Maximum      | 37.70                                | 19.66             | 3.35              | 2.86              | 10.92   | 31.44                               | 30.19    | 61.63 |
| Median       | 19.73                                | 10.69             | 1.48              | 1.77              | 5.76  | 16.62                               | 14.94    | 31.57 |
| Average      | 21.46                                | 10.88             | 1.78              | 1.80              | 6.20  | 18.11                               | 16.64    | 34.76 |

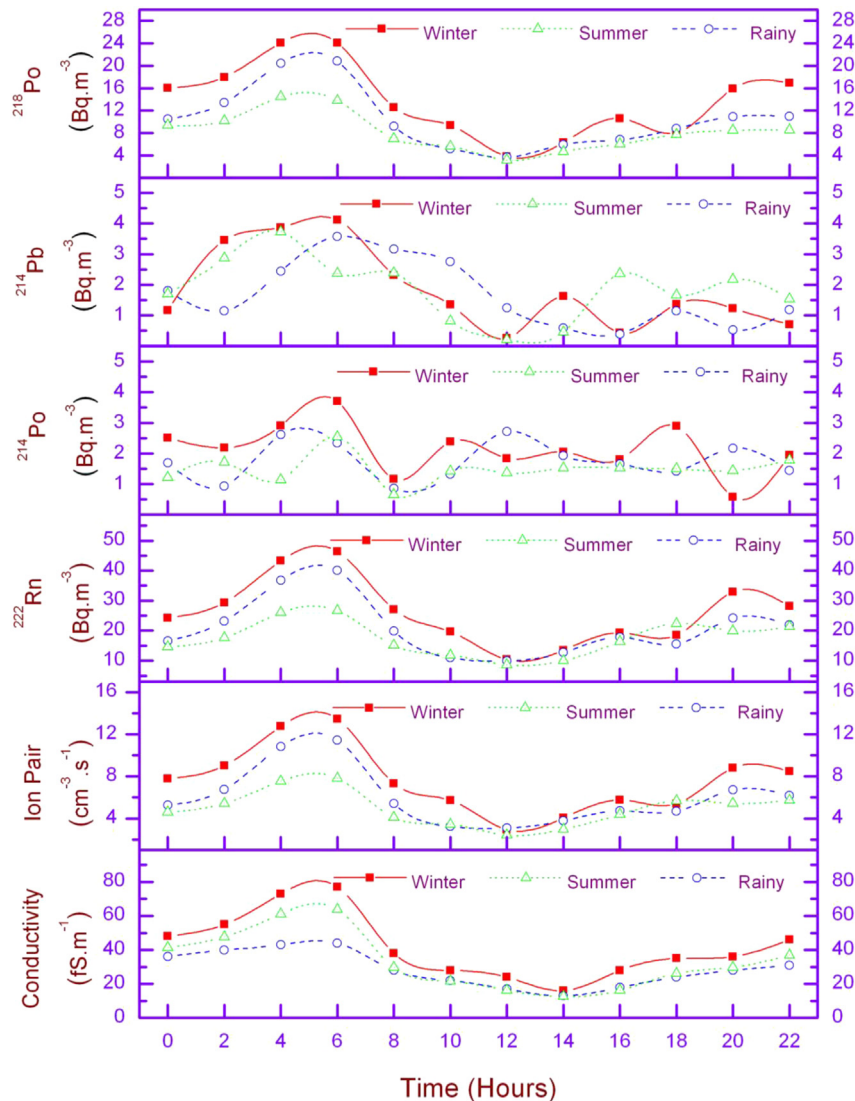


Fig. 3. Average diurnal variation of radon and its progeny concentration, ion production rate and atmospheric electrical conductivity in different seasons (2012–13).

the early morning hours and a minimum during day time which is similar to the diurnal variation of radon, its progeny concentrations and estimated ion production rate. During rainy season even though ionization takes place over the ground level, the ions are washed away due to rain resulting in decrease in the conductivity values. The atmospheric electrical conductivity shows good correlation with ion pair production rate with a correlation of 0.94 ( $n = 12$ ). The atmospheric electrical conductivity data can be used to predict the concentration of radon gas and aerosol pollution level in the atmosphere.

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