Analysis of Radium and Radon in the Environmental Samples and some physico-chemical properties of drinking water samples belonging to some areas of Rajasthan and Delhi, India

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ABSTRACT

Indoor radon measurements in some dwellings of Rajasthan and Delhi areas have been carried out using LR-115 type II Cellulose Nitrate films in the bare mode. The Radon values vary from 22.30 Bq/m$^3$ to 219.3 Bq/m$^3$ in the Delhi region and 55.76 Bq/m$^3$ to 107.80 Bq/m$^3$ in the Rajasthan areas, which is well within the recommended action level. The Radon concentration in water samples of some locations of Rajasthan and Delhi have also been measured by using RAD 7. The radon values vary from 0.581 Bq/l to 1.54 Bq/l in the Delhi region and from 0.0342 Bq/l to 3.45 Bq/l in the Rajasthan region. The Radium concentration in soil samples have been found to vary from 1.74 Bq/Kg to 5.03 Bq/Kg in the Delhi region and from 1.97 Bq/Kg to 2.91 Bq/Kg in the Rajasthan region. The values are found to be within the safe limits. Some of the physico-chemical properties such as pH, conductivity, hardness and total dissolved solids (TDS) have been reported in water samples. The results reveal that these areas are safe from the health hazard point of view as the radioactivity is concerned. A weak correlation has been observed between radon and radium.

Keywords: RAD 7, LR-115, Radon, Radium

INTRODUCTION

The exposure due to inhalation of radon and its daughters present in the environment is highest of the natural radionuclides to which human beings are exposed. It is an established fact that the enhanced levels of indoor radon in dwellings can cause health hazards and may cause serious diseases like lung cancer in human beings [1-3]. Therefore measurement of $^{222}$Rn concentration in the environment is of special interest to mankind. $^{222}$Rn is a noble radioactive gas produced by decay of $^{226}$Ra, which is a member of $^{238}$U series. By diffusion through soil, $^{222}$Rn enters into the atmosphere. The concentration of radon and its decay products show large fluctuations depending upon the building materials, underground soils, ventilation conditions and wind speed etc. [4-6].

Uranium, the heaviest radioactive toxic element is found in almost all types of soils, rocks, sands and water. Henshaw et al. (1990) has claimed that indoor radon exposure is associated with the risk of leukaemia and certain other cancers, such as malenoma and cancers of the kidney and prostate. If uranium rich material lies close to the surface of earth there can be high radon exposure hazards [8-10].

Radium is a solid radioactive element under ordinary conditions of temperature and pressure. It decays to radon emitting α-particles followed by γ-radiations. It is the concentration of radium which governs how many radon atoms are formed. How many of the produced radon atoms leave i.e. emanate from, the mineral grain or matter and enter the pore spaces depends on ; where the radium atoms are situated in the grain, the texture and size of the grain,
the permeability of the grains, temperature and pressure [11-13]. The measurements of radon thus necessitate the need for uranium and radium estimation in the parent source for public health risk measurements.

Various physio-chemical water quality parameters like pH, conductance, hardness and total dissolved solids in drinking water samples were also measured. It has no direct adverse effects on health. However a lower value below 4 will produce sour taste and higher value above 8.5 an alkaline taste [14]. Conductance is the measure of the ability of an aqueous solution to carry an electric current. The elevated dissolved solid that conductance indicates can cause mineral taste in drinking water. TDS is the term used to describe the inorganic salts and small amount of organic matter present in solution in water.

Uranium, radium and radon exhalation studies have been carried out in Hamirpur, Kullu and Una districts of Himachal Pradesh [15,16]. However in the present investigations, the survey has been carried out first time for the measurements of radium and radon exhalation rate from some soil samples of some areas of Rajasthan and Delhi. The radon concentration in dwellings and in the water samples has also been reported from the same areas. The aim of the work is to estimate the radium and radon content for health risk assessments in the study area. An attempt has also been made to find a correlation between the radium and other water quality parameters.

MATERIALS AND METHODS

To understand the migration and exhalation of radon in soils of different areas of Rajasthan and Delhi, India. The soil samples were collected from different villages belonging to these areas.

2.1.1 Measurements of radon exhalation rate and radium concentration

The ‘can technique’ [17] has been used for the measurement of radon exhalation rates in soil samples as shown in Fig.A.1. The samples from different villages were finely powdered and sieved through a 200 mesh sieve. The fine powder (250 g) of soil sample from each village was placed and sealed in different bottles for 30 days so as to attain the equilibrium. After one month, LR-115 type 2 plastic track detectors were fixed on the top inside of these glass bottles (acting as emanation chambers). The bottles were then sealed and left as such for 90 days so that the detectors can record α-particles resulting from the decay of radon. The exposed detectors were etched in 2.5N NaOH solution at 60°C for two hours using a constant temperature bath. The tracks were counted using an optical microscope at 400X magnification.

The radon exhalation rate in terms of area is calculated from the equation [17,18]

\[
E_A = \frac{CV}{A \left[ T + \frac{1}{\lambda} (e^{\lambda T} - 1) \right]} \tag{A.1}
\]

where \(E_A\) is the radon exhalation rate expressed in Bqm\(^2\)h\(^{-1}\), \(V\) is the effective volume of the bottle in m\(^3\), \(T\) is the exposure time in hour (h), \(\lambda\) is the decay constant for radon (h\(^{-1}\)) and \(A\) is the area of the bottle (m\(^2\)). The radon exhalation rate in terms of mass is calculated from the expression:

\[
E_M = \frac{CV}{M \left[ T + \frac{1}{\lambda} (e^{\lambda T} - 1) \right]} \tag{A.2}
\]

Here \(E_M\) is the radon exhalation rate in terms of mass (Bq kg\(^{-1}\)h\(^{-1}\)) and \(M\) is the mass of rock sample (250 gm).

The ‘can technique proposed by 19Alter and Price (1972) and later developed by 20Somogyi (1990) has been used to calculate the radium concentration in rock samples. The radium concentration in rock samples was calculated using the relation:

\[
C_{\text{Radium}} = \frac{hA}{K T e M} \tag{A.3}
\]

where \(C_{\text{Radium}}\) is the effective radium content of rock sample (Bq kg\(^{-1}\)), \(M\) is the mass of rock sample (250g), \(A\) is the area of cross-section of bottle (7.55x10\(^{-3}\) m\(^2\)), \(h\) is the distance between the detector and the top of the rock sample (0.153 m), \(K\) is the sensitivity factor, which is equal to 0.0245 tracks cm\(^{-2}\)d\(^{-1}\) per Bq m\(^{-3}\) [21] and \(T_e\) is the effective exposure time which is related with the actual exposure time \(T\) and decay constant \(\lambda\) for \(^{222}\)Rn with the relation:
2.1.2 Radon concentration measurements in water

For radon in water measurement, the RAD-H$_2$O closed loop aeration method has been employed; The RAD7 continually measures radon and thoron concentration, showing both on a spectrum printout, and also functions as a sniffer with audible count signal to locate radon entry points. The unit features the fastest response and recovery time of any system on the market, and is able to measure radon concentrations at the 200 Bq/m$^3$ action level in less than 1 hour with 10% standard deviation. The virtual absence of intrinsic background (0.2 Bq/m$^3$) gives the RAD7 an extremely low detection threshold, easily measuring below 4 Bq/m$^3$. The instrument is microcomputer controlled, featuring step-by-step instructions for ease of use. The instrument is a complete, portable stand-alone system with a built-in air pump, supplied in a rugged carrying case, total weight 5 kg. Additional accessories allow measurement of radon in soil and water (continuous and sample measurement); the RAD AQUA continuous water measurement accessory allows measurement of water radon to extremely low concentrations, whereby the air volume and water volume are constant and independent of the flow rate. The air circulates through the water and continuously extracts the radon until a state of equilibrium develops. The RAD-H$_2$O system reaches this state of equilibrium within about 5 minutes, after which no more radon can be extracted from the water. The extraction efficiency, or percentage of radon removed from the water to the air loop, is very high, typically 99% for a 40 mL sample and 94% for a 250 mL sample. The RAD H$_2$O gives results after 30 minutes analysis with a sensitivity that matches or exceeds that of liquid scintillation methods.

\[
T_e = \frac{1}{\lambda} \left(1 - e^{-\lambda T}\right)
\]  

(A.4)

Fig. A.1: The apparatus used to study the radium and radon exhalation rate of soil samples.
2.1.3 Measurements of radon Concentration In air
In the present investigations the indoor $^{222}\text{Rn}$ concentration has been studied in dwellings of some areas of Rajasthan and Delhi. The houses were chosen randomly. The track etch detector technique has been used to measure the level of indoor radon concentration in the dwellings. The LR-115 type 2 (Pelliculable) plastic track detectors having a size of about 1.5 cm x 1.5 cm fixed on micro glass slides were suspended at the centre of the room in the bare mode for a period of three months to assess the variations of indoor radon concentration levels. All the measured dwellings have a single floor level (ground floor). The exposed detectors were etched in 2.5 N NaOH solution for 90 minutes in a constant temperature bath ($60^\circ\text{C}$). After etching the detectors were thoroughly washed and scanned manually for track density measurements using Carl Zeiss binocular optical microscope at a magnification of 400X. The track density so obtained was converted into the units of Bq/m$^3$ using the calibration factor (0.020 $\pm$ 0.002 tracks cm$^{-2}$ d$^{-1}$/Bq m$^{-3}$) which satisfies the conditions prevailing in the Indian dwellings. The average background track density for the unexposed films of LR-115 type 2 detector was found to be 35 tracks cm$^{-2}$ and this value was subtracted from the observed values.

2.1.4 pH, TDS and Conductivity Measurement
For the measurement of pH, Conductivity and TDS a samples of water was collected from natural ground water source in some areas of Rajasthan and Delhi. Water Quality Kit is used to monitor pH, Conductivity and TDS in groundwater. It is manufactured by Naina Solaris Limited, New Delhi. It is calibrated with standard solution before dispatch so it does not required daily calibration. It is calibrated only when the instrument is continuously off for more than 150 hours.

<table>
<thead>
<tr>
<th>S. No</th>
<th>Location</th>
<th>Radium conc. C$_{\text{Ra}}$ (Bq kg$^{-1}$)</th>
<th>Radon exhalation rate E$_{\text{d}}$(mBqkg$^{-1}$h$^{-1}$)</th>
<th>E$_{\text{a}}$(mBqm$^{-2}$h$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Smaypur</td>
<td>1.74</td>
<td>2.15</td>
<td>7.12</td>
</tr>
<tr>
<td>2</td>
<td>Badli</td>
<td>5.03</td>
<td>6.21</td>
<td>20.5</td>
</tr>
<tr>
<td>3</td>
<td>Bhalasva</td>
<td>4.49</td>
<td>5.54</td>
<td>18.3</td>
</tr>
<tr>
<td>4</td>
<td>Yamuna-vihar</td>
<td>1.93</td>
<td>2.39</td>
<td>7.91</td>
</tr>
<tr>
<td>5</td>
<td>Sikar</td>
<td>2.63</td>
<td>3.25</td>
<td>10.7</td>
</tr>
<tr>
<td>6</td>
<td>Lakshmangarh</td>
<td>1.97</td>
<td>2.43</td>
<td>8.07</td>
</tr>
<tr>
<td>7</td>
<td>Bisau</td>
<td>2.79</td>
<td>3.44</td>
<td>11.4</td>
</tr>
<tr>
<td>8</td>
<td>Ramgarh</td>
<td>2.71</td>
<td>3.34</td>
<td>11.1</td>
</tr>
<tr>
<td>9</td>
<td>Ratangarh</td>
<td>2.90</td>
<td>3.58</td>
<td>11.8</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Location</th>
<th>Radon conc.(Bq/m$^3$)</th>
<th>Radon conc.(Bq/L)</th>
</tr>
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<tbody>
<tr>
<td>1</td>
<td>Pitampura</td>
<td>1540</td>
<td>1.54</td>
</tr>
<tr>
<td>2</td>
<td>Balbirnagar</td>
<td>991</td>
<td>0.99</td>
</tr>
<tr>
<td>3</td>
<td>Loni boarder</td>
<td>581</td>
<td>0.58</td>
</tr>
<tr>
<td>4</td>
<td>Peepli village</td>
<td>1470</td>
<td>1.47</td>
</tr>
<tr>
<td>5</td>
<td>Yamuna vihar</td>
<td>205</td>
<td>0.21</td>
</tr>
<tr>
<td>6</td>
<td>Smaypur</td>
<td>171</td>
<td>0.17</td>
</tr>
<tr>
<td>7</td>
<td>Bisau</td>
<td>3450</td>
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</tr>
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<td>Sikar</td>
<td>478</td>
<td>0.45</td>
</tr>
<tr>
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<td>Ramgarh</td>
<td>1430</td>
<td>1.43</td>
</tr>
<tr>
<td>10</td>
<td>Lakshmangarh</td>
<td>8830</td>
<td>8.83</td>
</tr>
<tr>
<td>11</td>
<td>Ratangarh</td>
<td>34.2</td>
<td>0.03</td>
</tr>
</tbody>
</table>

**RESULTS AND DISCUSSION**

The values of radium and radon exhalation rate in soil samples from 09 villages of some areas of Rajasthan and Delhi, India are given in table A.1. It is evident from the table that the radon exhalation rate in soil samples varies from 2.15 mBqkg$^{-1}$h$^{-1}$ to 6.21 mBqkg$^{-1}$h$^{-1}$ in Delhi region and 2.43 mBqkg$^{-1}$h$^{-1}$ to 3.58 mBqkg$^{-1}$h$^{-1}$ in the Rajasthan region. The radium concentration varies from 1.74 Bq kg$^{-1}$ to 5.04 Bq kg$^{-1}$ in Delhi region and 1.98 Bq kg$^{-1}$ to 2.90 Bq kg$^{-1}$ in the Rajasthan area.

In the present investigations the observed values of radium and radon exhalation rate are lower in soil to those determined in the soil of Una district by Kumar et al. (2001) and also lower than those reported in the soil of Hamirpur and Kullu districts of Himachal Pradesh [16a,23]. The values of radon exhalation rate and radium concentration in rocks are quite low as compared with those reported in the soil of Hamirpur district [16]. These
values of radon concentration are also less than the permissible value of 370 Bq kg$^{-1}$ [24] and much lower than those reported by $^{224}$Nageshvara Rao et al. (1996) and $^{226}$Mittal et al. (1998) for Rajasthan area. This may be due to the fact that our sample collection is from the surface whereas the others have reported work in the mining area of this region. The values of radium and radon exhalation are also below the recommended safe limit and are not significant from the exploration point of view. Thus, the results reveal that the area is safe for as the health hazard effects of radium and radon exhalation are concerned. Transport of radon / radium through the soils/rocks largely depends on the geology of the area, which include lithology, compression, porosity and structural / tectonics features like thrust, faults, joints and fractures.

The radon concentration levels recorded in the dwellings of some areas of Rajasthan and Delhi are given in table A.2. The radon concentration varies from 22.30 Bq/m$^3$ to 219.31 Bq/m$^3$ in Delhi area and 55.76 Bq/m$^3$ to 107.79 Bq/m$^3$ in the Rajasthan area. The maximum value 219.31 Bq/m$^3$ is found in Pitampura, Delhi as compared to Rajasthan where the maximum value is 107.79 Bq/m$^3$ in Sikar district of Rajasthan. The higher value of radon in Delhi area may be due to the fact that Delhi lies on the fault line.

Table A.3 shows the value of radon concentration in water samples belonging to the areas of Rajasthan and Delhi. The values varies from 0.39 Bq/l to 1.54 Bq/l in the Delhi region and in Rajasthan area the value varies from 0.03 Bq/l to 3.45 Bq/l. The maximum value is found in the Biasau of the Rajasthan area. This may be due to the leaching of radioactive rich rocks in the area. According to USEPA, The safe limit of radon concentration in water samples is 11 Bq/l. The values are lower than the recommended limit in all the regions. Hence these areas are safe for the drinking purposes.

Table A.4 shows the values of pH, conductivity, TDS and salinity in water samples from the study areas. The pH of water samples from Delhi areas ranges between 7.29 to 8.11 and from Rajasthan areas ranging between 7.02 to 7.25. The safe limit for pH as recommended by $^{224}$WHO (1971) is 7.0-8.5. All the water samples except one ( Pitampura from New Delhi, pH value 8.11) have a pH value within the safe limit. The Samples analyzed have conductivity values ranging from 0.397 to 3.92mS/cm (Table 4). Three samples have been found to contain conductance values more than the recommended levels [27]. However, it has no health significance. High values of conductance in some water samples confirm their natural mineralization. The value of TDS in drinking water samples is found to vary from 0.399 to 1.966 ppt (Table A.4).
CONCLUSION

From the above studies it is concluded that:
1) The indoor radon concentrations in the dwellings and radium in the soil samples lies below the recommended limit so the studied areas is safe as far as the health hazards effects are concerned.
2) The radon concentrations in drinking water samples also lies below the recommended limit by various agencies but the value of Lakshmangarh area of Rajasthan is comparitibly higher than the nearby areas.
3) The values of drinking water quality parameters viz. pH, conductivity and TDS below the recommended level except few samples of conductivity which has shown values more than the recommended levels.

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