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Radon exhalation rate in Chhatrapur beach sand samples of high background radiation area and estimation of its radiological implications

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Abstract : Chhatrapur beach placer deposit, situated in a part of the eastern coast of Orissa, is a newly discovered high natural background radiation area (HBRA) in India. The sand samples containing heavy minerals, were collected from Chhatrapur region by the grab sampling method at an interval of ~ 1 Km. Radon exhalation rates were measured by "Sealed Can Technique" using LR-115 type type II in the sand samples containing heavy minerals collected from the beach. Radon activity is found to vary from 1177.1 to 4551.4 Bq m⁻³ whereas the radon exhalation rate varies from 423.2 to 1636.3 mBq m⁻²h⁻¹ with an average value of 763.9 mBq m⁻²h⁻¹. Effective dose equivalent in sand samples estimated from exhalation rate varies from 49.9 to 193.0 μSv y⁻¹ with an average value of 90.1 μSv y⁻¹. From the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K computed radium equivalent is found to vary from 864.0 to 11471.5 Bq kg⁻¹ with an average value of 3729.0 Bq kg⁻¹. External hazard index, H_{ex} range from 2.3 to 31.0 with a mean value of 10.1, which is quite high. This value supports the conclusion based on high mean absorbed gamma dose rate in air due to the naturally occurring radionuclides as 1627.5 nGy h⁻¹. A positive correlation has been found between U concentration and radon exhalation rate in the sand samples. The use of sand as construction material may pose a radiation risk to ambient environment.

Keywords : Sand samples, Radon exhalation rate, SSNTD, uranium concentration

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1. Introduction

The Chhatrapur beach placer deposit is located in Ganjam District, Orissa on the southeastern coast of India. It is known as High Back Ground Radiation Area (HBRA) due to the presence of radiogenic heavy minerals. This is characterized by a number of well

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developed sand dunes, which are parallel to the coast. Heavy mineral concentrations are found as discrete patches within the beach sands, consisting dominantly of quartz sands. Distribution of naturally occurring radionuclides mainly ^{238}U , ^{232}Th and ^{40}K depends on the distribution of rocks from which they originate and on the processes through which they are concentrated. Higher concentrations of radionuclides such as ^{238}U , ^{232}Th and ^{40}K occur in minerals such as monazites and zircons. Around the world though, there are some areas with sizable populations that have high background radiation levels. The highest are found primarily in Brazil, India and China. The higher radiation levels are due to high concentrations of radioactive minerals in sand and soil.

High background radiation areas (HBRAs) are found in the world and are due the local geological controls and geochemical effects and cause enhanced levels of terrestrial radiation [1, 2]. Wide ranging radiation studies have been carried out in the HBRAs in Brazil [3], in China [4], in India [5-7], in Iran [8] and in USA and Canada [9] and in some other countries [2] to estimate risks and effects of longterm radiation exposure. In India there exists some natural HBRA's due to monazite sand bearing placer deposits along its long coast line such as Ullal in Karnataka [10], Kalpakkam in Tamil Nadu [11] and Kerala and the southwestern coast of India [5,6]. Recently Mohanty *et al.* (2004) [12] found higher levels of natural radionuclides (^{238}U , ^{232}Th & ^{40}K) in the newly discovered HBRA on the Chhatrapur beach placer deposit of Orissa.

Radon an α -radioactive inert gas indicates the presence of radium and its ultimate precursor uranium in the ground. As an inert gas and having sufficiently long lifetime (3.8 days) it can move freely through the materials like soil, sand, rock *etc.* Short lived radon progenies have been established as causative agents of lung cancer [1, 13]. Radon exhaling properties of porous materials are of prime importance for the estimation of radiation risk and have been the subject of several investigations [14-16]. The correlation between the uranium concentration and the radon emanation potential of the source material is required for radon risk. In the present study radon exhalation rate measurements from the sand samples of the Chhatrapur beach placer deposit have been carried out for finding its correlation with uranium concentration and the estimation of radiation risk.

2. Experimental method

The "Sealed Can Technique" using the solid-state nuclear track detectors, a simple and efficient method [16-18] was used for the radon exhalation measurements. Collected sand samples from the Chhatrapur Beach Placer Deposit were dried and sieved through a 100-mesh sieve and were dried and sieved through a 100-mesh sieve and were placed in the Cans (diameter 7.0 cm and height 7.5 cm) similar to those used in the calibration experiment [17]. In each Can a LR-115 type-II plastic track detector (2cm \times 2cm) was fixed at the top inside of the Can as shown in Figure 1. The sensitive lower surface of the detector is thus freely exposed to the emergent radon from the material in the Can so that it is capable of recording the alpha particles resulting from the decay of radon in the

Can. Radon and its daughters reach an equilibrium concentration after a week or more and thus the equilibrium activity of emergent radon could be obtained from the time of exposure and the geometry of the Can. After the exposure for 100 days, the detectors from all the Cans were retrieved. The detectors were etched in 2.5N NaOH at 60°C for a period of 90 minutes in a constant temperature water bath for revelation of tracks. Resulting alpha tracks on the exposed face of the detector foils were scanned under an optical microscope at a magnification of 400X. The calibration factor of 0.056 Tr cm⁻²d⁻¹ obtained from an earlier calibration experiment [17], was used to compute the radon activity from the track density.

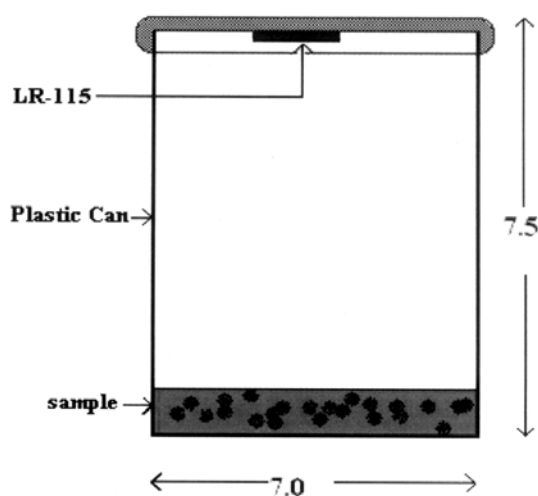


Figure 1. Assembly for the measurement of radon exhalation rate using "Sealed Can Technique".

Following expression given the exhalation rate [16, 18, 19] :

$$E_x = \frac{CV \lambda}{A \left[T + \frac{1}{\lambda} \{e^{-\lambda T} - 1\} \right]} \quad (1)$$

Where E_x is Radon exhalation rate (Bq m⁻² h⁻¹), C is integrated radon exposure (Bq m⁻³ h), V is the effective volume of Can (m³), λ is the decay constant for radon (h⁻¹), T is the exposure time (h) and A is the area covered by the Can (m²).

3. Radiation risk

As sand is frequently used as a building construction material in India, the estimation of radiation risk to the population is quite important and can be computed from the radon exhalation rate as well as from the activity concentration of the radio-nuclides present in the sand samples.

3.1. Effective dose equivalent :

The risk of lung cancer due to radon and its daughters is estimated from the radon exhalation rate in terms of effective dose equivalent. The contribution to indoor radon concentration from sand samples can be obtained from the expression [20] :

$$C_{Rn} = \frac{E_x \times S}{V \times \lambda_v} \quad (2)$$

Where, C_{Rn} , E_x , V and λ_v are radon concentration (Bq m^{-3}), radon exhalation rate ($\text{Bq m}^{-2} \text{h}^{-1}$), room volume (m^3) and air exchange rate (h^{-1}) respectively. In these calculations, the maximum radon concentration from the building material is assessed by assuming the room as a cavity with $S/V = 2.0 \text{ m}^{-1}$ and air exchange rate of 0.5 h^{-1} . Annual exposure to potential alpha energy E_p (effective dose equivalent) is then related to the average radon concentration C_{Rn} by the following expression :

$$E_p [\text{WLM} \cdot \text{y}^{-1}] = \frac{8760 \times n \times F \times C_{Rn}}{170 \times 3700} \quad (3)$$

where, C_{Rn} is in Bq m^{-3} , n the fraction of time spent indoors; 8760, the number of hours per year and 170, the number of hours per year and 170, the number of hours per working month. The values of $n = 0.8$ and $F = 0.42$ were used to calculate E_p . From radon exposure, the effective dose equivalents were estimated by using a conversion factor of $6.3 \text{ mSv (WLM)}^{-1}$ given by ICRP, 1987 [21].

3.2. Radium equivalent activity and gamma dose rate :

To estimate the gamma dose rate activity concentrations of $^{238}\text{U}/^{226}\text{Ra}$, ^{232}Th and ^{40}K for these samples were taken from Mohanty *et al* 2004 [12]. The distribution of $^{238}\text{U}/^{226}\text{Ra}$, ^{232}Th and ^{40}K in sand samples is not uniform. Uniformity with respect to exposure to radiation can be defined [22] in term of Radium Equivalent Activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of $^{238}\text{U}/^{226}\text{Ra}$, ^{232}Th and ^{40}K :

$$\text{Ra}_{\text{eq}} = C_U + 1.43 C_{\text{Th}} + 0.07 C_K \quad (4)$$

It has been assumed here that $370 \text{ Bq kg}^{-1} \text{ } ^{238}\text{U}$ or $259 \text{ Bq kg}^{-1} \text{ } ^{232}\text{Th}$ or $4810 \text{ Bq kg}^{-1} \text{ } ^{40}\text{K}$ produce the same gamma dose rate. To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv Gy^{-1}) and outdoor occupancy factor (0.2) proposed by UNCSEAR (2000) [2] were used. The effective gamma dose rates in units of mSv y^{-1} were calculated by the following relation :

$$\begin{aligned} \text{Effective gamma dose rate (mSv y}^{-1}\text{)} = \\ \text{Dose rate (nGy.h}^{-1}\text{)} \times 8760\text{h} \times 0.2 \times 0.7 \text{ Sv.Gy}^{-1} \times 10^{-6} \end{aligned} \quad (5)$$

Krieger (1981) [23] proposed the following relation for external hazard index, H_{ex} for limiting the radiation dose from building materials in Germany to 1.5 mGy y^{-1} :

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \tag{6}$$

This criterion considers only the external exposure risk due to γ -rays and corresponds to maximum Ra_{eq} of 370 Bq kg^{-1} for the material. These very conservative assumptions were later corrected by Keller and Muth, 1990 [24] and the maximum permission concentrations were increased by a factor of 2 which gives

$$H_{ex} = \frac{C_U}{740 \text{ Bqkg}^{-1}} + \frac{C_{Th}}{520 \text{ Bqkg}^{-1}} + \frac{C_K}{9620 \text{ Bqkg}^{-1}} \leq 1 \tag{7}$$

Table 1. Radon Activity, Radon exhalation rates and effective dose in bulk sand samples.

Sample Name	Radon Activity (Bqm ⁻³)	Radon Exhalation Rate (mBqm ⁻² h ⁻¹)	Effective Dose (μ Sv y^{-1})	Uranium concentration (ppm)
CHS1	1462.9	525.9	62.0	9.7
CHS2	2057.1	739.6	87.2	16.2
CHS3	2128.6	765.3	90.2	20.2
CHS4	2157.1	775.5	91.4	20.2
CHS5	3488.6	1254.2	147.9	28.3
CHS6	2242.9	806.3	95.1	21.8
CHS7	1514.3	544.4	64.2	9.7
CHS8	1177.1	423.2	49.9	4.9
CHS9	1400.0	503.3	59.4	13.8
CHS10	1328.6	477.6	56.3	6.5
CHS11	1571.4	564.9	66.6	12.1
CHS12	1662.9	597.8	70.5	12.1
CHS13	2428.6	873.1	103.0	24.3
CHS14	1214.3	436.6	51.5	4.9
CHS15	1528.6	549.5	64.8	12.1
CHS16	1497.1	538.2	63.5	9.7
CHS17	2074.3	745.7	87.9	16.2
CHS18	2628.6	945.0	111.4	24.3
CHS19	4551.4	1636.3	193.0	48.5
CHS20	4217.1	1576.1	185.9	40.5
Minimum	1177.1	423.2	49.9	4.9
Maximum	4551.4	1636.3	193.0	48.5
Average	2116.6	763.9	90.1	17.8
S.D	934.3	342.8	40.4	11.1
R. Std. %	44.1	44.9	44.8	62.3

4. Results and discussion

The values of radon activity, radon exhalation rate and uranium concentration in the sand samples are presented in Table 1. Radon activity is found to vary from 1177.1 to 4551.4 Bq m⁻³ with an average value of 2116.6 Bq m⁻³ whereas the radon exhalation rate varies from 423.2 to 1636.3 mBq m⁻²h⁻¹ with an average value of 763.9 mBq m⁻² h⁻¹. Uranium concentrations varies from 4.9 to 48.5 ppm with an average value 17.8 ppm. As can be seen from the graph (Figure 2) of there is a positive correlation between the radon exhalation rate and uranium concentration. Effective dose equivalent in the sand samples estimated from exhalation rate varies from 49.9 to 193.0 μSv y⁻¹ with an average value of 90.1 μSv y⁻¹.

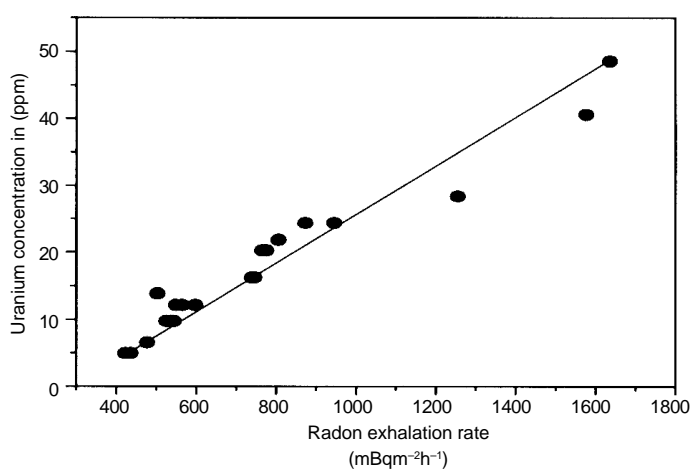


Figure 2. Variation of radon exhalation rate with U-concentration in the sand samples.

From the activity concentration of ²²⁸U, ²³²Th and ⁴⁰K in these sand samples the radium equivalent due to presence of radionuclides is given in Table 2.

Radium equivalent activity was found to vary from 854.0 to 11471.5 Bq kg⁻¹ with an average value of 3729.0 Bq kg⁻¹. Average absorbed gamma dose rate in air due to the naturally occurring radionuclides (²³⁸U, ²³²Th and ⁴⁰K) is 1627.7 nGy h⁻¹ which is much higher than the world average value of 55 nGy h⁻¹ (UNSCEAR, 1988) [25], while effective gamma dose rate varies from 0.5 to 6.1 mSv y⁻¹ with an average value of 2.0 mSv y⁻¹. Values of external hazard index, H_{ex} for the sand samples studied in this work range from 2.3 to 31.0 with a mean value of 10.1. As all the values of H_{ex} are higher than unity, the use of sand from this region as building construction material may give significant radiation dose and pose radiological threat to the population.

5. Conclusion

Radon exhalation rate in the sand samples collected from Chhatrapur beach placer deposit a newly discovered high natural background radiation area (HBRA), in Orissa are measured using the "Sealed Can Technique". Radon activity is found to vary from 1177.1 to 4551.4

Table 2. Effective dose equivalent, Radium equivalent activity, annual effective gamma dose rate and External Hazard index of sand samples.

S. No.	Radium equivalent (Bqkg ⁻¹)	Absorbed dose rate (nGyh ⁻¹)	Annual effective gamma dose rate (mSvy ⁻¹)	External Hazard index H _{ex}
CHS1	1689.3	740.0	0.9	4.6
CHS2	3498.5	1530.0	1.9	9.4
CHS3	3975.0	1730.0	2.1	11.1
CHS4	4047.9	1770.0	2.2	10.9
CHS5	6507.4	2840.0	3.5	17.6
CHS6	4853.0	2120.0	2.6	13.1
CHS7	1708.8	750.0	0.9	4.6
CHS8	1218.0	530.0	0.6	3.3
CHS9	2465.0	1080.0	1.3	6.7
CHS10	1027.0	450.0	0.5	2.8
CHS11	2442.2	1070.0	1.3	6.6
CHS12	2445.0	1060.0	1.3	6.0
CHS13	5166.9	2260.0	2.8	14.0
CHS14	864.0	375.0	0.5	2.3
CHS15	1876.5	830.0	1.0	5.1
CHS16	1145.5	500.0	0.6	3.1
CHS17	3635.5	1580.0	1.9	9.8
CHS18	5451.5	2375.0	2.9	14.7
CHS19	11471.5	5000.0	6.1	31.0
CHS20	9083.5	3960.0	4.9	24.5
Minimum	864.0	375.0	0.6	2.3
Maximum	11471.5	5000.0	6.1	31.0
Average	3729.0	1627.5	2.0	10.1
S.D	2719.3	1185.02	1.5	7.3
R. Std. %	72.9	72.8	72.5	72.7

Bq m⁻³ whereas the Radon exhalation rate varies from 423.2 to 1636.3 μBq m⁻² h⁻¹. Effective dose equivalent varies from 49.9 to 193.0 μSv y⁻¹. From the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in the sand samples radium equivalent, Absorbed gamma dose rate and external hazard index were calculated the values are quite high. All the values of H_{ex} are more than 1, the use of sand from this region as building construction material may pose radiation risk to the population. A positive correlation has been observed between U-concentration and radon exhalation rate.

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