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Assessment of human exposure to dissolved radon in groundwater around the uranium industry of Jaduguda, Jharkhand, India

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Measurement of dissolved radon and ^{226}Ra in groundwater was carried out in 30 different locations around the uranium mining and ore processing area of Jaduguda, Jharkhand, India. Activity concentration of ^{226}Ra was found to be very low, whereas dissolved radon was observed to be slightly elevated due to geological features of the area consisting of uranium mineralization. No definite relation was observed between radon and ^{226}Ra in groundwater. The annual dose due to ingestion of radon containing water estimated at 60% locations was less than 100 μSv. The maximum ingestion dose was found to be 300 μSv to adult members of the public. The average dose due to the ingestion of radon in water was 94 μSv, which is less than the stipulated limit of 1000 μSv prescribed by regulatory body.

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RADIATION is a natural part of the environment and all people receive exposure from naturally occurring radioactivity in soil, water, air and food. The largest fraction (>50%) of natural radiation exposure we receive is attributed to the radioactive gas radon[1,2]. This is emitted from uranium, a naturally occurring mineral present in soil and rocks. The nuclides of the uranium series which can be harmful to health because of their presence in drinking water are ^{226}Ra and ^{222}Rn (ref. 3). Uranium-bearing ore mineral continuously releases radon into groundwater. It can travel from the point of formation to an area of no or low uranium content showing a high value of waterborne radon. Consequently, groundwater has potentially much higher concentrations of radon than surface water. The average concentration of radon is usually less than 0.4 Bq l^{-1} in public water supplies derived from surface waters and about 20 Bq l^{-1} from groundwater sources[4].

When water containing radon is ingested, a radiation dose is received by the digestive system of the body. Though part of the dissolved radon is released when we exhale a greater portion is absorbed in the intestinal wall. The decay of radon and its short-lived daughters continues to give dose. The greatest risk of intake of water containing high dissolved radon is stomach and colon cancer[5]. USEPA has proposed a limit of 11,000 Bq m^{-3} of dissolved radon in drinking water. WHO has suggested that public water supply should not exceed 100 Bq l^{-1} of dissolved radon. Countries like Sweden, Finland, USA and the Czech Republic have proposed separate limits for dissolved radon in different sources of water supply. The regulatory authority in India has proposed no limit for dissolved radon in drinking water supply. In view of this, a study has been carried out in the uranium mineralized area of Jaduguda, Jharkhand to estimate waterborne radon in groundwater and ingestion dose among the public.

The Proterozoic Jaduguda U (–Cu–Fe) deposit in the Singhbhum shear zone, eastern India hosts the oldest and most productive uranium mine in the country[6]. Jaduguda (long. 22°30′, lat. 86°20′) (Figure 1) is well known for its heterogeneous high mineral deposits. The geological futures of the area have been well documented[7]. Many copper, nickel and uranium mining activities are continuing since the past several decades[8]. The region receives >1000 mm of rainfall annually with a maximum temperature in summer being >45°C and minimum during winter <7°C.

Dissolved radon measurement in community groundwater samples was carried out using AquaKit supplied with AlphaGuard[9], a widely used primary device for the measurement of radon. It is suitable for continuous monitoring of radon concentration[10] in the range of $2–2 \times 10^6$ Bq m^{-3}. The AquaKit is an attachment consisting of two glass vessles, a battery-operated pump and related piping (Figure 2). One glass vessel holds the sample, whereas the second one is a security vessel that remains empty and prevents the entry of sample into AlphaGuard in case of any malfunction. The regular attachment of AlphaGuard from the air inlet probe is removed and a different socket is joined to couple with the AquKit. The sample container is taken to the spot and 100 ml volume of water is transferred carefully with minimum disturbance. The sample jar and safety jar are connected with the alpha pump and moisture trapper[9,11]. AlphaGuard is kept in the flow mode. The pump is operated at 0.5 lpm for 30 min. The water sample starts bubbling which emanates dissolved radon and allows it to flow to the ionization chamber region of AlphaGuard. The radon concentration measured is converted to dissolved radon as follows

$$C_{\text{water}} = C_{\text{air}} \left[\frac{(V_{\text{system}} - V_{\text{sample}})}{V_{\text{sample}}} + K \right] - Co,$$

where C_{water} is the dissolved radon in water (Bq l^{-1}), C_{air} the radon concentration in the measuring set-up or AlphaGuard reading (Bq m^{-3}), Co is background of alpha guard (Bq m^{-3}), V_{system} is interior volume (ml) of measuring set-up, including ionization chamber of AlphaGuard, alpha pump, sampling vessels and tubing and K is the radon distribution coefficient (0.26).

Radium in water sample was estimated by the emanation method[12]. ^{226}Ra was estimated by allowing the build-up of its daughter ^{222}Rn for a known period[13,14]. The in-built radon was collected in a scintillation cell and counted after equilibrium (between radon and its progeny) was attained[13–15]. The radon already present in the solution was removed using an evacuation pump. After ensuring radon-free aliquot, the solution was retained for two weeks or more depending on the expected level of radium in the sample. During this period, ^{226}Ra through alpha decay led to the formation of its progeny ^{222}Rn. The in-built radon was collected in a previously evacuated scintillation cell. The scintillation cell was left for minimum 200 min for ^{222}Rn and its progeny to attain equilibrium. The scintillation cell was coupled to a photo multiplier tube. Alpha counts were recorded for a desired period in order to get counts above 95% confidence level. The ^{226}Ra activity is calculated using the equation

$$^{226}\text{Ra(Bq)} = \frac{1.883 \times 10^{-3} \times C \times 0.037}{E \times (1 - e^{-\lambda\theta}) \times (1 - e^{-\lambda T}) \times e^{-\lambda t}},$$

where C is the net counts obtained after subtraction of the background, E the efficiency of the cell (75%), t the counting delay (min), T the counting duration (min), θ the build-up period (min) and λ is the decay constant of ^{222}Rn (1.258×10^{-4} min^{-1}).

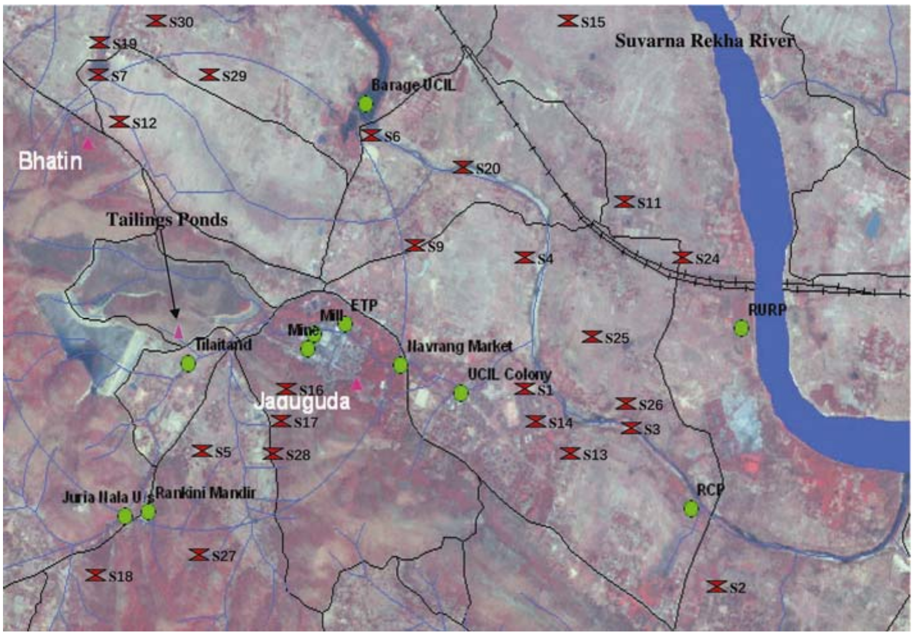

Figure 1. Map of study area (source: Geology Department, Uranium Corporation of India Ltd.). ⧗ Sampling Points, ▲ Uranium Mine/Mill/Tailings Ponds, ● Prominet Industries/places ……▶ surface water flow direction.

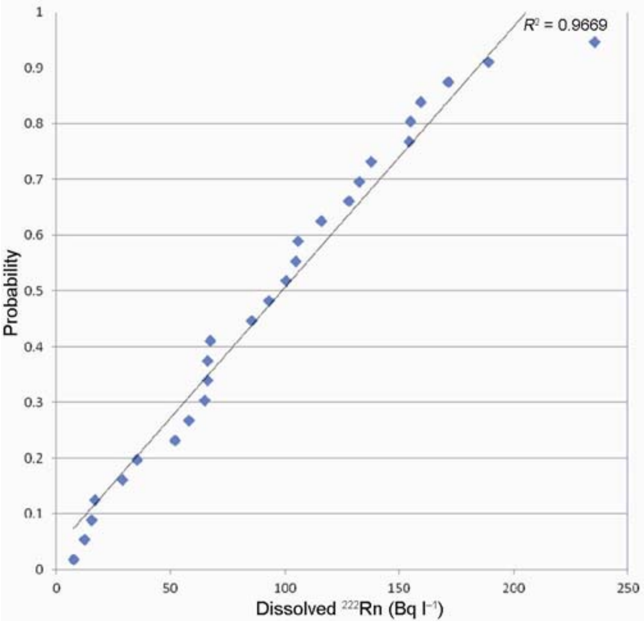

Figure 2. Probability plot of dissolved radon in groundwater.

The radium content of the original sample was obtained from the above equation by further applying the correction for sampling parameters.

Measurement of dissolved radon and radium in groundwater samples was carried out in 30 different locations (Table 1) around uranium mining and ore processing facility at Jaduguda. Table 2 presents the summary statistics. It can be observed from Table 2 that the arithmetic mean and median are greater than the standard deviation. Also, kurtosis is less than 3, so the distribution may follow a normal trend. Again the data are highly skewed with a skewness of 1.5, which can be attributed to the presence of outliers either due to variation in geological formation or due to large-scale variation between different seasons. The probability plot of the data (Figure 3) incorporating the outlier values indicates that the distribution is neither normal nor lognormal. For normal probability plot the R^2 value is 0.79, whereas for lognormal probability plot the R^2 value is 0.90. The lognormal distribution of the dataset is also not favoured by large difference in the geometric mean and median concentration of 102.7 and 84 Bq l^{-1} respectively. Barring the three outliers the probability plot reasonably represents the normal distribution (Figure 3) with skewness of 0.4 and approximately identical mean and median (94.7 and 93 Bq l^{-1} respectively). The outlayer locations are at Sarkdih and Kendadih are in the proximity of uranium mineralization and elevated concentration is anticipated. Groundwater in contact with rocks deformed by shear may contain higher concentrations of ^{222}Rn than groundwater from adjacent unsheared rocks[16,17]. In Singhbhum shear zone wide variation in dissolved radon concentration is observed ranging from 7.5 to 389 Bq l^{-1}. The measured dissolved radon concentration is far lower than in the case of similar studies (Table 3) carried out in the United States, Poland, Greece, Venezuela and Sweden and elevated compared to those for Taiwan, Lebanon and the Himalayan region of India[18–26]. Studies carried out in the Virginia, USA showed that 84% of groundwater samples exceeded the proposed limit for dissolved radon with a maximum concentration of 1184 Bq l$^{\cdot}$ Radon-222 activity in water may be interpreted as recoil flux from mineral surfaces[17], or as a combination of recoil and diffusion of Rn from microfractures resulting from a higher effective surface area[20–22] for Rn relative to Ra. The United States Geological Survey has done extensive studies on dissolved radon distribution in the US and found a significant positive correlation between dissolved radon in groundwater and uranium distribution in the rock strata[16,17,27–29]. The present investigation is confined to distribution of dissolved radon in groundwater; rock strata analyses was not carried out, although an attempt was made to observe the correlation between dissolved ^{222}Rn and dissolved ^{226}Ra in groundwater. The correlation was insignificant ($R^2 = 0.089$), which is presented in a scatter diagram (Figure 4). This is in agreement with studies which suggest that dissolved radon concentration in groundwater is unsupported by soluble ^{226}Ra in aquifer, but may supported when ^{226}Ra is concentrated in aquifer material[30,31].

In the present study seasonal and temporal variation of dissolved radon concentration was not considered. It has been found that the groundwater temperature in this

Table 1. Dissolved radon and radium in groundwater and ingestion dose

Location in Jaduguda, Jharkhand	Sample ID	^{226}Ra (mBq l^{-1})	Radon (Bq l^{-1})
Community Centre, UCIL Colony	S1	15	154.2
Rakha Copper Market	S2	3.5	64.8
Sitadanga Village Temple	S3	3.5	137.7
Ichra Village	S4	6	189.1
Chatikocha Village	S5	3.5	7.5
Mechua Village	S6	5	132.6
Baregutu Village	S7	5	105.6
Kendadih Village	S8	40	385.5
Govt. School, More	S9	12	15.4
Degree Ashram Village	S10	13	100.5
Old Rakha Mine Station	S11	5	235.4
Sadakdih Village	S12	33	372.1
Matigora, Hanuman Temple	S13	3.5	12.3
A&B Type, UCIL Colony	S14	12	85.6
Kuldih Village	S15	3.5	171.6
Tilaitand, Near Bittle Shop	S16	3.5	67.4
Tilaitand end, Main Road	S17	8	35.3
Kalikapur Village	S18	3.5	104.8
Jharia, road side	S19	3.5	159.3
Kalimandir, Station Road	S20	16	389.6
Moubhandar, Near HCL Plant	S21	31	116
Surda crossing	S22	3.5	155
Purnapani Village	S23	3.5	66
Old Rakhamines Village	S24	3.5	128
Itabhata Village	S25	5	29
Sitadanga Village Church	S26	3.5	52
Rankini Temple	S27	80	93
Tilatand Village	S28	18	17
Bhatin Village	S29	4	66
Jharia Village	S30	7	58

Table 2. Summary of Dissolved radon (Bq l^{-1}) in groundwater

Mean	123.5
Standard error	19.0
Median	102.7
Mode	66
Standard deviation	104.3
Sample variance	10,886.3
Geometric mean	84.0
GSD	2.7
Kurtosis	1.9
Skewness	1.5
Minimum	7.5
Maximum	389.6
n	30

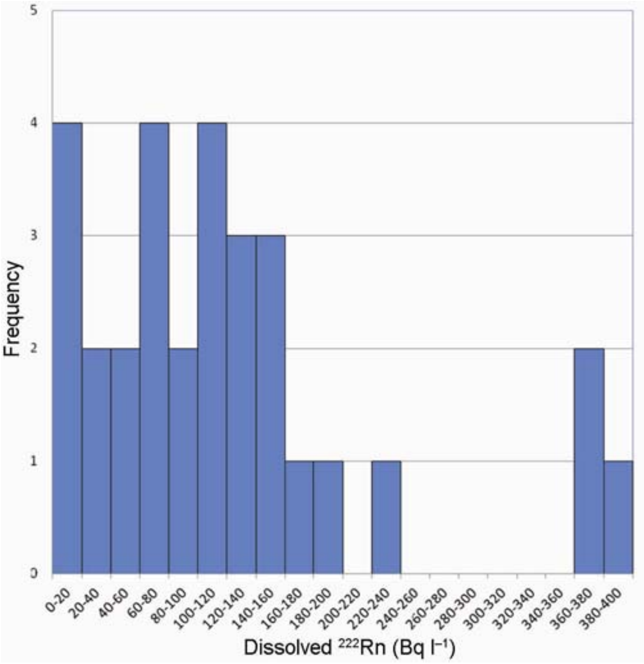

Figure 3. Frequency distribution plot of dissolved radon in groundwater.

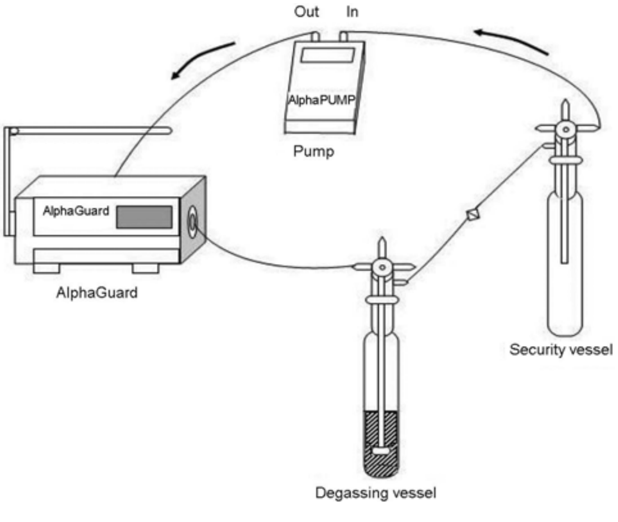

Figure 4. Measurement of dissolved radon by AquaKit.

Table 3. Dissolved radon in groundwater of different region world wide

Country	Dissolved radon (Bq l^{-1})
Virginia, USA[16]	1184
Poland[21]	1703
Migdonia Valley, Greece[22]	160
El Castano, Venezuela[23]	576
Sweden[24]	15,000–30,000
Ain Ibil, Lebanon[25]	49
Taiwan[27]	40
Himalayan region, India[22]	69
Bangalore region, India[36]	283
Jaduguda (present study)	123

region remains almost steady over time and major fluctuation in dissolved radon concentration is not anticipated in different seasons from the same source. However, the impact of seasonal variation on dissolved radon level in shallow aquifer or shallow well water cannot be ruled out. The elevated concentration of dissolved radon in the groundwater of the studied area may be attributed to uranium mineralization. Moreover, radon is easily soluble in water and not easily absorbed onto the mineral surface; so its concentration is normally higher than its precursor. The wide variation of ^{222}Rn in different locations could be explained by physical properties of the aquifer material and varying concentration of uranium in solid phase[26]. Earlier studies have suggested that average radon in water consistent with relative abundance of uranium in host rocks and aquifer lithology can be a useful tool for predicting groundwater radon concentration[30].

Annual ingestion dose among the public can be calculated using the formula

$$D_{\text{eff}}\ (\mu\text{Sv y}^{-1}) = A_C W_D V,$$

where D_{eff} is the annual effective dose (Sivert Sv)), A_C the activity concentration of ^{222}Rn (Bq l^{-1}) and V is the annual water intake. W_D is the effective dose equivalent per unit water activity concentration of the radionuclide and for ^{222}Rn't is 0.35×10^{-8} Sv/Bq (refs 2 and 32). The World Health Organization (WHO) and the US Environmental Protection Agency (EPA) have estimated that people consume 2 litre of water per day[33]. Various studies have proposed that a part of the dissolved radon escapes during normal water usage[34,35]. It has also been estimated that the average amount of water from which ^{222}Rn is not removed in direct usage is 0.6 litre per day (ref. 2). Thus an annual consumption of 222.6 litre of water is considered while evaluating the ingestion dose. The annual dose estimated at 60% locations is less than 100 μSv. The maximum dose was found to be 300 μSv, which is less than 1100 μSv ingestion dose reported in Polish groundwater[21] and 2280.2 μSv y^{-1} (ingestion and inhalation) reported in Bangalore region, South India[36]. The average ingestion dose in the study was around 60 μSv which is 6% of prescribed dose limit 1000 μSv by Atomic Energy Regulatory Board

Dissolved radon has no prescribed limit in drinking water in many countries including India. USEPA recommends 11.1 Bq l^{-1} in drinking water and the European Union consists 100 Bq l^{-1} as the reference level for dissolved radon. It has been observed that more than 50% of the samples exceed the European reference level which can be attributed to the local geological features of the area, including widespread uranium mineralization. However, in more than 60% samples the annual radiation dose to the public is less than 100 μSv or accounts for only 10% of the prescribed limit of 1000 μSv. Public exposure and health risk from ingesting radon in drinking water are considered negligible.

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Development of hydrophobic platinum-doped carbon aerogel catalyst for hydrogen–deuterium exchange process at high pressure

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The use of catalysed exchange of deuterium (D) between hydrogen (H) gas and liquid water using the bithermal hydrogen water (BHW) process is a promising and environment-friendly approach for the production of heavy water. However, the use of this approach is limited by the lack of a suitable catalyst that has good activity at high operating pressures required for practical applications. We report the development of hydrophobic platinum-doped carbon aerogel (PtCA) catalyst which shows good catalytic activity for H/D isotope exchange reactions at operating pressures up to 20 bar.

Keywords: Carbon dioxide activation, hydrogen isotope separation, hydrophobic catalyst, platinum-doped carbon aerogel.

CATALYSED exchange of deuterium (D) between hydrogen (H) gas and liquid water (liquid phase catalytic exchange (LPCE) reaction) using bi-thermal hydrogen water (BHW) process is receiving considerable attention for heavy water production[1]. This is because it is more energy-efficient and environment-friendly compared to the existing 'girder sulphide' and 'ammonia–hydrogen' processes. The catalysed exchange involves two steps – first, deuterium exchange between hydrogen gas and water vapour at the catalytic site and subsequently equilibration of deuterated water vapour with liquid water at gas–liquid interface, which results in the transfer of deuterium from gas to liquid phase. Success of the process requires simultaneous presence of H_2 gas and water vapour at the catalytic site and easy accessibility of liquid water to water vapour involved in isotopic exchange reaction. However, because the solubility of H_2 gas in water is poor, if the catalytic site is covered with liquid water, the exchange of deuterium between hydrogen gas and water vapour is highly impeded[2]. Therefore, a primary requirement for the LPCE process is that the catalyst should allow access of gaseous reactants to the catalytic

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