Risk assessment of ^{226}Ra and ^{222}Rn from the drinking water in the Jalandhar and Kapurthla districts of Punjab

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Abstract

Naturally occurring Radium (^{226}Ra) and Radon (^{222}Rn) concentration in the drinking water samples of 20 villages of Jalandhar and Kapurthla districts of Punjab has been determined for health risk assessment. Activity concentration of ^{226}Ra and ^{222}Rn in the studied water samples varies from 0.08 to 0.13 Bq l^{-1} and 1.42 to 5.26 Bq l^{-1} , with the average concentration of 0.1 Bq l^{-1} and 3.51 Bq l^{-1} respectively. The dose to the internal parts of the body is received from ^{222}Rn through inhalation and ingestion while ^{226}Ra contributes dose mainly through ingestion. The annual average inhalation and ingestion dose from ^{222}Rn in water in the studied area is 0.89 nSv a^{-1} and 0.74 $\mu\text{Sv a}^{-1}$. The mean annual ingestion dose from ^{226}Ra in water is 18.73 $\mu\text{Sv a}^{-1}$. The total mean annual effective dose due to ^{226}Ra and ^{222}Rn in water is 19.46 $\mu\text{Sv a}^{-1}$. As the total annual effective dose due to ^{226}Ra and ^{222}Rn in water samples of the study area is lower than the reference dose level of 10 mSv, the health hazards related to these radionuclides are expected to be negligible.

Keywords ^{226}Ra · ^{222}Rn · Rad7 · Inhalation and ingestion dose

1 Introduction

Presence of high ^{222}Rn concentration in the indoor environment contributes a major proportion to the sources of lung cancer. As we spend most of our time at home, houses become significant location of ^{222}Rn exposure [13]. In United States, after smoking, ^{222}Rn is the second-largest cause of lung cancer and may result in about 21,100 lung cancer deaths every year [9]. The main source of ^{222}Rn in air is the presence of Uranium (^{238}U) and Radium (^{226}Ra) in soil. ^{222}Rn is formed as a result of natural decay of ^{226}Ra . From there it is transported to the soil surface and then to the houses through advection and diffusion. ^{222}Rn present in water also contributes to ^{222}Rn in air through degassing. ^{222}Rn contributes the dose to the lungs and to the stomach through inhalation and ingestion [22]. ^{226}Ra is almost pervasive in soil, rock, air, water, and food. The median concentrations of ^{226}Ra and ^{228}Ra in drinking water

are generally low, but there are regions where higher concentrations are known to occur. The redistribution of ^{226}Ra in the environment occurs due to the mining of coal and uranium ore. But it does not affect much overall. The chemical properties of ^{226}Ra are alike some other alkaline earth elements, peculiarly similar to Ba and Ca. ^{226}Ra exists in only the +2 oxidation state in solution and does not easily complex in water. The chloride, nitrate and bromide salts of ^{226}Ra are soluble in water but the carbonate and sulphate salts are very insoluble [1]. The gastrointestinal absorption of ^{226}Ra in human adults is about 15–21 percent of the total ingested amount of ^{226}Ra from water and food. Normal elderly human that ingested mock ^{226}Ra dial paint containing ^{224}Ra absorbed approximately 20 percent of ingested ^{226}Ra as an average [15, 16]. 25 to 35 percent of the ingested amount of ^{226}Ra remains in the body for 5–6 days, if 0.05 mg of ^{226}Ra is taken by an adult human male through mouth [27]. The retention of ^{226}Ra in human

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body is dependent on its gastrointestinal absorption (f_1 value), which is 0.2, 0.6 and 0.3 respectively for adults, infants and children. 70% of the ^{226}Ra is excreted from the human body on the first day after the ingestion while the rest of it follows the metabolic path of calcium [5]. The behaviour of ^{226}Ra is analogous to Ca in case of its absorption to blood from the GI tract or lungs. But its movement between the body parts like bones, soft tissue is different from that of Ca. In an oral exposure, around 80 percent of absorbed ^{226}Ra into the intestines from blood [30].

The main objective of the present study is to estimate the ^{222}Rn and ^{226}Ra concentration in the drinking water samples of the studied area for the radiation dose assessment. This study is first time reporting the ^{226}Ra concentration in the drinking water samples of Jalandhar and Kapurthla districts of Punjab and also, an attempt has been made to find their contribution to the inhalation and ingestion dose. There is no previous study about the ^{226}Ra concentration in the drinking water samples but there are many studies conducted to estimate air borne radon in the nearby provinces and in the nearby states [2, 12, 20].

2 Study area

The state of Punjab stretches from $29^{\circ}32'$ to $32^{\circ}32'$ N latitude and $73^{\circ}55'$ to $76^{\circ}50'$ E longitude, occupying a land of 50,362 sq. kms in the north-western part of India. Sandy loam to clayey with an average pH value of 8.15 is found in Doaba region (Jalandhar, Kapurthala, Hoshiarpur and Nawanshahr districts) of Punjab, making alkalinity and salinity problematic for this place. The alluvial soil of Jalandhar and Kapurthala districts are widely described as arid and brown soil or tropical arid brown soil. The major type of soil found in South-western Punjab is desert soil and sierozem soil and in the Eastern Punjab, it is loamy to clayey. Gamma spectrometric studies showed that the average concentration of thorium in this area is higher than the world average value of thorium (45 Bq kg^{-1}) in soil [18].

3 Sampling technique

In this investigation, water samples were collected from the twenty villages of Jalandhar and Kapurthla districts of Punjab. Groundwater is the main source of drinking water in these districts. So, the water samples were collected from the main source for the assessment of the radiation dose. Samples were taken in specific bottles designed for the RAD7 device and provided by the manufacturer. Four

samples were taken from each source of water. Getting a good sample requires care and practice. Sampling technique, or lack of it, is generally the major source of error in measuring the ^{222}Rn content of water. It has been taken in mind the water sample taken is a representative of the water being tested and it has never been in contact with air. It was made sure that the sample has not been through a charcoal filter, or been sitting for days in a hot water tank. Before taking the sample from the source, the water was run for at least fifteen minutes in order to make sure that water is coming directly from deep. A bowl was put up to the faucet so that the water overflowing the bowl prevents the water when leaving the faucet from touching the air and the vial is filled with water at the bottom of the bowl. ^{222}Rn concentration in the water samples was measured immediately after collecting the sample and for ^{226}Ra concentration measurements; these 250 ml vials were sealed and stored for one month prior to the measurement after ^{222}Rn in the vials was removed. After a period of one month, ^{226}Ra is considered to be in equilibrium with ^{222}Rn . At the end of this period, the growing ^{222}Rn will be equal to ^{226}Ra activity, which was measured using an electronic ^{222}Rn detector RAD7 [7].

4 Methodology

^{222}Rn in water was measured using the RAD H2O accessory, which enables us to measure ^{222}Rn in water over a wide concentration range from less than 10 pCi l^{-1} to more than $400,000 \text{ pCi l}^{-1}$. The RAD H2O setup consists of three components, the RAD7, 250 ml water vial with aerator, and the desiccant as shown in the Fig. 1. Before going for a measurement, the RAD7 should not contain any moisture or ^{222}Rn . To achieve this, the RAD7 was purged for some time using the larger drying unit. The small drying tubes were used for the actual measurement as the RAD7 is calibrated with the small drying tubes. ^{222}Rn gas is mildly soluble in water. Being a gas, it tends to leave the water upon contact with air. This is known as aeration. The rate of ^{222}Rn transfer from water to air increases with temperature, agitation, mixing, and surface area. The ^{222}Rn continues to release from until a state of equilibrium is developed between the air and water. In RAD H2O method the water sample is aerated in closed loop configuration. The air recirculates through the water and continuously extracts the ^{222}Rn until a state of equilibrium develops. In RAD H2O configuration this state of equilibrium is reached within about 5 min. The extraction efficiency of ^{222}Rn from the water in this scheme is very high, typically 94% for a 250 ml sample. The exact value of the percentage ^{222}Rn

Fig. 1 Map of study area

extracted from water depends somewhat on ambient temperature, but it is always well above 90%. There is little or no temperature effect on the overall ^{222}Rn measurement in water. The RAD7's internal sample cell is a 0.7 l hemisphere, which is coated with an electrical conductor. At the centre of this hemisphere, a solid-state, ion-implanted, planar silicon alpha detector is present. A high voltage power circuit is used to charges the inside conductor to a potential of 2000–2500 V, with respect to the detector. This creates an electric field throughout the volume of the cell. When a ^{222}Rn nucleus decays inside the cell, it is transformed to ^{218}Po as a positively charged ion. The electric field within the cell propels this positively charged ion to the detector and it sticks there. The short-lived ^{218}Po nucleus decays through alpha emission upon the detector's active surface and produce an electrical signal, which is proportional to the energy of the emitted alpha particle [7].

5 Molecular diffusivity coefficient and Partition coefficient

^{222}Rn is soluble in water but its solubility is dependent on the temperature ($K = f(T)$). The effect of temperature on the partition of ^{222}Rn in water and air was explained on the basis of data reported by Meyer and Schweidler [21]. The temperature dependence of partition coefficient of ^{222}Rn in water to air is well reported in literature. Some studies also show its dependence on salinity and both [35].

Partition coefficient, $K = 0.105 + 0.405e^{(-0.0502T)}$

where T is temperature in $^{\circ}\text{C}$.

Temperature dependence of Molecular diffusivity coefficient (D_o) for ^{222}Rn in water is reported in the literature by Peng in 1974 as a function of temperature [23].

Molecular diffusivity coefficient (D_o) = $10^{-\left(\left(\frac{980}{T+273}\right)+1.59\right)}$

6 Dose estimation

Ingestion dose due to ^{226}Ra in drinking water is calculated by using the following equation

$$Ra_{\text{ing}} = C_{\text{Ra}} \times U_a \times D_f$$

where C_{Ra} is the ^{226}Ra concentration in water in Bq l^{-1} , U_a is the annual water consumption in l a^{-1} i.e. 730 l a^{-1} and D_f is the dose conversion factor in $0.28 \mu\text{Sv Bq}^{-1}$.

^{222}Rn in water is a source of Stomach and Lung dose through ingestion and through inhalation as it is released to indoor air from water. The annual effective doses for ingestion and inhalation were calculated according to parameters introduced by UNSCEAR report [32]. Inhalation dose, $Rn_{\text{inh}} = C_{\text{Rn}} \times \text{Equilibrium factor} \times \text{Dose conversion factor} \times \text{Indoor occupancy} \times 10^{-4}$ where C_{Rn} is ^{222}Rn concentration in water; Average indoor occupancy time per person is about 7000 h a^{-1} ; Equilibrium factor between ^{222}Rn and its progeny is equal to 0.4; Dose conversion factor for ^{222}Rn exposure is $9 \text{ nSv}/(\text{Bq h m}^{-3})$.

$$\text{Ingestion dose, } Rn_{\text{ing}} = C_{\text{Rn}} \times U \times D$$

where U is the annual intake of water i.e. 60 l a^{-1} and D is the The effective dose coefficient from ingestion equals $3.5 \text{ nSv}/(\text{Bq l}^{-1})$ [3, 4]

7 Results

The values of partition and molecular diffusion coefficient for ^{222}Rn in the studied water samples show a variation in the range from 0.25 to 0.28 and 1.06 to 1.18 respectively. This manuscript presents the ^{226}Ra and ^{222}Rn concentration in the drinking water samples of the some villages of Jalandhar and Kapurthla district of Punjab. The concentration of the ^{226}Ra and ^{222}Rn in the surveyed water samples varies from $< 0.074 \text{ Bq l}^{-1}$ to 0.13 Bq l^{-1} and from 1.42 Bq l^{-1} to 5.26 Bq l^{-1} . The annual effective dose (AED) due to ^{226}Ra in water varies from BDL to $27.01 \mu\text{Sv a}^{-1}$ with a mean value of $18.73 \mu\text{Sv a}^{-1}$. The inhalation and ingestion dose from ^{222}Rn in water varies from 0.35 nSv a^{-1} to

1.33 nSv a^{-1} and from $0.3 \mu\text{Sv a}^{-1}$ to $1.10 \mu\text{Sv a}^{-1}$. The total mean AED dose from both ^{226}Ra and ^{222}Rn in water varies from $0.3 \mu\text{Sv a}^{-1}$ to $28.01 \mu\text{Sv a}^{-1}$ with mean value of $19.46 \mu\text{Sv a}^{-1}$ (Table 1).

8 Discussions

^{222}Rn and ^{226}Ra activity concentrations have been measured in drinking water samples of Jalandhar and Kapurthla districts of Punjab and have been compared with the ^{222}Rn and ^{226}Ra concentrations of different water samples of the other countries of the world. Different values of the reference level of the ^{222}Rn in water are given by the different international agencies. The maximum contamination level of ^{222}Rn in water is 100 Bq l^{-1} according to WHO [29], 40 Bq l^{-1} according to UNSCEAR [31] and 11 Bq l^{-1} according to US-EPA [8]. ^{222}Rn concentration in all the studied samples is less than the reference level defined by WHO, UNSCEAR and US-EPA. The US-EPA [24] has given the safe limit of 0.74 Bq l^{-1} for ^{226}Ra level in water and the ^{226}Ra concentration in the studied water samples is less than this action level. 0.1 mSv is the dose level for alpha and beta activities in water that may be inimical to human health. If we consider the ^{222}Rn – ^{226}Ra dose contribution as a gross alpha appraisal for the present study, the values of the doses obtained are below the WHO safe level. These values of doses are also less than the world average values of 1.2 mSv a^{-1} and 0.29 mSv a^{-1} for inhalation and ingestion dose from ^{222}Rn [33] (Tables 2, 3).

9 Conclusions

In the present investigation, an active radon monitor has been used to calculate the ^{226}Ra concentration. It has been back calculated from the radon concentration. In is intended to estimate ^{226}Ra concentration using ICPMS directly from the collected water samples. The comparison of these two techniques will give the idea about the uncertainties of estimating ^{226}Ra using an active radon monitor. A positive and significant co-relation is observed between the ^{226}Ra and ^{222}Rn concentration in water (Fig. 2). So, the

Table 1 ^{226}Ra , ^{222}Rn concentration and their dose contributions

S. no	Location	Temperature (°C)	K	D _o	^{226}Ra concentration (Bq l ⁻¹)	^{222}Rn concentration (Bq l ⁻¹)	Ra _{ing} (μSv a ⁻¹)	Rn _{ing} (μSv a ⁻¹)	Rn _{inh} (nSv a ⁻¹)	Total AED (μSv a ⁻¹)
1	Lohian Khas	17.4	0.27	1.08	0.079	2.73	16.18	0.57	0.69	16.76
2	Sultanpur Lodhi	17.9	0.27	1.10	0.110	5.24	22.49	1.10	1.32	23.59
3	Kapurthala	16.7	0.28	1.06	0.123	3.74	25.23	0.79	0.94	26.01
4	NIT Jalandhar	19.1	0.26	1.13	0.128	4.12	26.11	0.87	1.04	26.97
5	Phagwara	17.8	0.27	1.10	BDL	1.42	0.00	0.30	0.36	0.30
6	Busawal	17.5	0.27	1.09	0.076	2.51	15.53	0.53	0.63	16.06
7	Nakodar	18.7	0.26	1.12	0.132	4.72	27.01	0.99	1.19	28.00
8	Malsian	18.2	0.27	1.11	0.079	2.73	16.15	0.57	0.69	16.72
9	Gidderpindi	17.5	0.27	1.09	0.080	2.77	16.42	0.58	0.70	17.00
10	Kartarpur	18.5	0.27	1.12	0.089	2.77	18.12	0.58	0.70	18.70
11	Husain Pur	17.9	0.27	1.10	0.118	4.38	24.17	0.92	1.10	25.09
12	Makho	18.1	0.27	1.11	0.076	3.14	15.53	0.66	0.79	16.19
13	Pajjan	17.5	0.27	1.09	0.091	4.13	18.57	0.87	1.04	19.44
14	Dhilwan	18.9	0.26	1.13	0.121	5.26	24.73	1.10	1.33	25.83
15	Fattu Dhingra	20.6	0.25	1.18	0.089	3.29	18.16	0.69	0.83	18.85
16	Bara pind	18.5	0.27	1.12	0.076	2.46	15.59	0.52	0.62	16.10
17	Goraya	19.4	0.26	1.14	0.086	3.25	17.58	0.68	0.82	18.26
18	Jalandhar city	16.4	0.28	1.06	0.100	3.46	20.51	0.73	0.87	21.24
19	Lambra	16.8	0.28	1.07	0.090	4.27	18.33	0.90	1.08	19.23
20	Nihaluwal	17.6	0.27	1.09	0.089	3.85	18.10	0.81	0.97	18.91

Table 2 ^{226}Ra concentration in water samples of some countries

Location	^{226}Ra concentration	Source of water	References
Serbia	0.1–14.8 Bq l ⁻¹	Well water	[6]
India	12.6–79.2 Bq l ⁻¹	Groundwater	[37]
Austria	3.7–211 mBq l ⁻¹	Mineral water	[34]
Italy	0.4–7.2 mBq l ⁻¹	Tap water	[25]
Spain	< 0.2–142 mBq l ⁻¹	Well, Spas and Spring	[14]
Punjab, India	< 0.074–0.13 Bq l ⁻¹	Drinking water	Present study

Fig. 2 Schematic diagram of RAD H2O Accessory

^{226}Ra in water also contributes to the ^{222}Rn content of that water. It is also clear from the study that the ^{222}Rn in water contributes dose mainly by ingestion. The ingestion dose from the ingested ^{226}Ra from the water is more as compared to the ingestion dose from the ^{222}Rn in water (Fig. 3). Total AED from ^{226}Ra in water is more than the total AED from ^{222}Rn in water. Total AED for the residents of the study area is lower than the recommended dose level. So, the studied area is safe as far as the exposure from ^{226}Ra and ^{222}Rn in water is concerned. The results of this study provide the baseline data about the natural radioactivity in the environmental medium. This data will help the policy makers to determine the contamination limit of this radionuclide (Fig. 4).

Fig. 3 Ingestion dose from ^{226}Ra and ^{222}Rn in water**Table 3** ^{222}Rn concentration in water samples of some countries

Location	^{222}Rn concentration	Source of water	References
Austria	12 Bq l ⁻¹	Drinking water	[10]
Punjab, India	0.4–16.7 Bq l ⁻¹	Groundwater	[19]
Milano, Italy	3.9–6.9 Bq l ⁻¹	Tap water	[25]
Brazil	0.9–36 Bq l ⁻¹	Groundwater	[17]
China	8–12 kBq m ⁻³	Tap water	[36]
UK	0.2–30.4 Bq l ⁻¹	Domestic water	[11]
Iran	MDA–31.9 Bq l ⁻¹	Water source, Spring river	[26]
Turkey	1.5–53.6 Bq l ⁻¹	Well water	[28]
Punjab, India	1.4–5.26 Bq l ⁻¹	Drinking water	Present study

Fig. 4 Co-relation between ^{226}Ra and ^{222}Rn in water

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Compliance with ethical standards

Conflict of interest The authors declare no conflicts of interest.

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