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Paper

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Assessment of radiation dose due to ^{210}Po in water and food samples of Chamarajanagar district, Karnataka, India

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Abstract

Groundwater is in direct contact with the soil and rocks that dissolve many compounds and minerals including uranium and its daughter products. ^{210}Po is one of the decay products of ^{238}U series that cause internal radiation dose in humans when consumed in the form of water and food, including sea food. Therefore, activities of ^{210}Po have been studied in ground and surface water, and in food samples that are commonly used in Chamarajanagar region of Karnataka, India. The average ^{210}Po concentration in bore well water samples and surface water samples are 3.21 and 1.85 mBq L^{-1}, respectively. In raw rice and wheat, the average values of ^{210}Po are 96 and 41 mBq kg^{-1}, respectively. In millets and pulses, the average activity of ^{210}Po is 157 and 79 mBq kg^{-1}, respectively. Among food items, the highest activity of 1.3 kBq kg^{-1} is observed in marine crabs and the lowest activity of 2.6 mBq kg^{-1} is found in milk samples. The average ingestion dose due to ^{210}Po in ground and surface water are 2.8 and 1.62 μSv y^{-1}, respectively. The ingestion dose due to various food samples to the population is also calculated. Total ingestion dose due to ^{210}Po to pure vegetarian population and general population are 38.09 and 590.80 μSv y^{-1}, respectively. The concentration of ^{210}Po in water samples and food samples of this region are in a comparable range with the world and Indian average values and lies well below the recommended guideline level.

Introduction

Radiations from the primordial radionuclides are eventually found in soil, water and in the atmosphere. Radionuclides in the ^{238}U decay series are important sources of internal dose in humans ingested via inhalation and ingestion. ^{210}Po with a half-life of 138.4 days decays to ^{206}Pb by emitting an alpha particle (5.3 MeV) and delivers a significantly higher dose via ingestion(1). Most of the ^{210}Po found in our environment occur naturally and distributed through two main processes: (i) the dissolution of ^{226}Ra and ^{222}Rn in water sources and (ii) the release of ^{222}Rn from the Earth's crust(2). ^{210}Po is the last unstable isotope in the ^{238}U decay series and can be found in the environment wherever ^{238}U or its daughter products such as ^{226}Ra, ^{222}Rn or ^{210}Pb are present.

^{222}Rn is the major source of ^{210}Po in the environment which diffuses from rocks and soils to the atmosphere where it finally decays to ^{210}Pb, ^{210}Bi and then to ^{210}Po. ^{210}Po attaches itself further electrostatically to aerosol particles and are transported back to Earth's surface to soil, plants and aquatic environments prevalently by dry deposition and aqueous wash out. ^{210}Po and ^{210}Pb are directly taken up by plants either from the soil through the root system or by foliar absorption from the air. They are also produced in plants when ^{226}Ra is absorbed from soil and water(3). It is well known that ^{210}Po is accumulated in various marine organisms at high concentrations and is a major cause of ingestion dose due to radionuclides(1).

As an alpha particle emitting decay product of ^{226}Ra, ^{210}Po is classified as a Group 1 human carcinogen(4). Polonium has a high specific activity of 1.66×10^{14} Bq g^{-1}. Therefore, 1 μg of ^{210}Po emits as many alpha particles per second as 4.5 mg of ^{226}Ra (specific activity = 3.66×10^{10} Bq g^{-1}), 262 mg of ^{238}Pu; or 446 kg of ^{238}U(1). The International Atomic Energy Agency passed a regulatory guideline associated with the need for radiation protection measures for the workers involving minerals. Dose due to ingestion of

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^{210}Po is ~7% of the total natural internal radiation dose to the public[5] and ~18% of the average internal dose of the population is due to ingestion of ^{210}Po along with its precursor ^{210}Pb[6]. Renal excretion of ^{210}Po is slow compared with other elements because it binds strongly to haemoglobin and plasma proteins and is not filtered by kidneys[7]. Its toxicity is determined solely by its chemical properties, which directs its distribution and retention in organs and tissues where the alpha particles do the harm[8]. ^{210}Po entering blood is deposited predominantly in soft tissues, with the greatest concentrations in the reticuloendothelial system, principally the liver, spleen and bone marrow, as well as in the kidneys and skin, particularly hair follicles[9]. The activity concentration of ^{210}Po $\geq$ 0.1 Bq L^{-1} in drinking water poses human health concerns. In consideration of this, an attempt has been made to study the activity of ^{210}Po in drinking water and dietary sources and the dose received by the local public due to ingestion of ^{210}Po through food and drinking water is estimated.

Study area

Chamarajanagar district lies in the southern tip of Karnataka, India between north latitudes 11° 40′ 58" and 12° 6' 32", and east longitudes 76° 24' 14" and 77° 64' 55". The geographical area is 5648 km^2 and has an average elevation of 662 m above sea level. The district has five taluks viz. Chamarajanagar, Gundlupet, Hanur, Kollegal and Yelandur (Figure 1)[10]. The major food sources are rice, ragi, wheat, cereals and pulses. Rarely some people use fresh water and marine food. Lean and poultry products are consumed by few people occasionally. The general mineral available in the district is black granite. Bore wells are the major sources of water supply for domestic and other purposes in the entire district[11, 12]. Groundwater which originates from granitic rocks that leach and dissolve radioisotopes may result in higher doses to the local population. Therefore, the distribution of ^{210}Po in the groundwater and common food sources are studied in the present investigation.

Materials and methods

Water samples of ~20 L, collected from different regions of Chamarajanagar district were analysed for the ^{210}Po activity by radiochemical method. Ground and surface water samples were collected from ~10 villages of each taluk in pre cleaned plastic cans and the pH of the water was measured at each location.

Water sample was filtered using Whatman 42 filter paper and transferred to a clean tub. Hydrochloric acid was added to maintain the pH of the solution to 2.0. Ferric chloride anhydrous, iron carriers (5 g) were added to the solution and stirred for an hour using a specially designed mechanical stirrer. Ammonia solution (25%) was slowly added until the pH of the solution increased to 9.0 to precipitate iron as iron (III) hydroxide in the solution. The solution was stirred steadily for 6 hours and left undisturbed overnight for settling. The upper layer of this solution was discarded. In the case of raw food samples, the samples were dried in hot air oven at 110 °C and crushed into fine power of size 100 μm. The dry weight was recorded and processed as discussed hereafter. The precipitate/dry sample was dissolved using conc. 12.1 M hydrochloric acid. Hydrogen peroxide solution (30%) was added to remove the organic content present in the solution. Hydrochloric acid was added to this solution, stirred using a magnetic stirrer, and evaporated to near dryness. The total dryness was avoided to prevent loss of ^{210}Po due to volatilization and sorption onto the surface of the glass beaker. The residue was treated with 0.5 M hydrochloric acid and to this solution; ascorbic acid was added to avoid interference of ferric ion deposition on the silver disc[2, 13, 14].

Sample processing

A background counted silver disc is immersed into the solution and stirred for 6 hours at 90 °C for spontaneous deposition of ^{210}Po on it (Figure 2). The silver disc is removed from the solution, rinsed with distilled water and ethanol, dried and counted for alpha activity. The activity concentration of ^{210}Po was calculated using equation (1)[13].

$$C_{Po} = C \times \frac{100}{\varepsilon} \times \frac{100}{E_p} \times \frac{1000}{V} \quad (1)$$

Where, C_{Po} is the activity concentration of ^{210}Po (Bq L^{-1}), C is the background subtracted sample counts (s^{-1}), ϵ is the efficiency of alpha counting system (17.65%), E_p is the efficiency of ^{210}Po deposition on a silver planchet (90%) and V is the volume of the water taken for processing (L) and in the case of food samples, dry weight of the samples was considered (kg).

Ingestion dose due to ^{210}Po in water through drinking pathway

The effective dose due to activity of ^{210}Po in the ingested water per annum was calculated using equation (2)[15].

$$D_w = C_{Po} \times W_w \times F_{Po} \quad (2)$$

Where, D_w is the ingestion dose due to ^{210}Po in water (mSv y^{-1}), C_{Po} is the activity concentration of

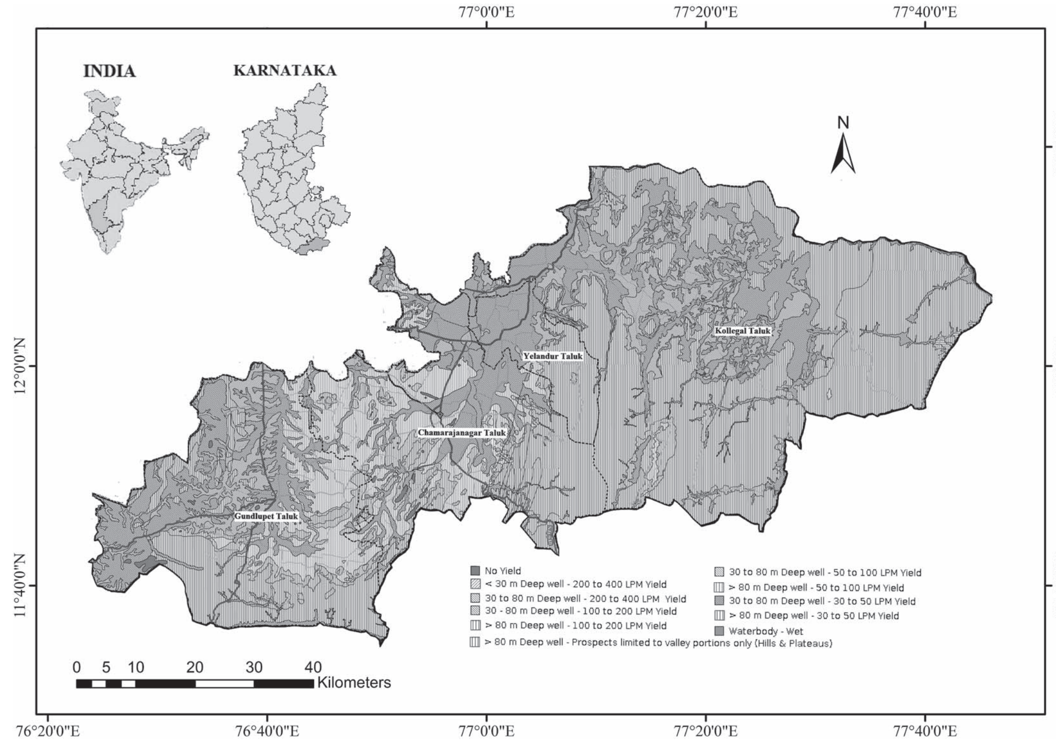

Figure 1. The study area, Chamarajanagar district.

Figure 2. Experimental setup for the spontaneous deposition of 210Po on a silver disc.

^{210}Po (mBq L^{-1}), W is the weighted average of water consumption (730 L y^{-1})[16] and F_{Po} is the dose conversion factor for ^{210}Po (1.2 × 10^{-6} Sv Bq^{-1})[17].

Ingestion dose due to ^{210}Po in food samples

To estimate the dose due to ^{210}Po in food samples through ingestion path way, a demographical survey was conducted for the selected population in all the five taluks of the Chamarajanagar district. The annual intake rate of specific food samples was calculated and used to estimate the annual ingestion dose.

The ingestion dose is calculated using the following equation[18]:

$$D_F = C_{Po} \times W_F \times F_{Po} \qquad (3)$$

Where, D_F is the ingestion dose due to ^{210}Po in food samples (mSv y^{-1}), C_{Po} is the activity concentration of ^{210}Po in food sample (mBq kg^{-1} or mBq L^{-1}), W_F is the average consumption rate of particular food item by the population (kg y^{-1} or L y^{-1}), F_{Po} is the dose conversion factor for ^{210}Po (1.2 × 10^{-6} Sv Bq^{-1})[17].

Results and discussion

^{210}Po activity in water samples and associated effective dose

Activity of ^{210}Po was measured in 108 groundwater samples and 52 surface water samples covering all

Table 1. ^{210}Po concentration in water samples of Chamarajanagar district and associated ingestion dose.

Sl. No	Sampling Taluks	Groundwater			Surface water		
		No. of Samples	Range and average activity of ^{210}Po (mBq L^{-1})	Average dose due to ^{210}Po (μSv y^{-1})	No. of Samples	Range and average activity of ^{210}Po (mBq L^{-1})	Average dose due to ^{210}Po (μSv y^{-1})
1	Chamarajanagar	26	1.61–9.35 (2.65)	2.32	14	0.95–3.23 (1.85)	1.62
2	Gundlupet	21	1.02–12.56 (5.21)	4.57	10	1.62–4.86 (2.64)	2.31
3	Kollegal	24	0.56–4.64 (2.63)	2.30	11	0.36–2.52 (1.55)	1.36
4	Hanur	18	1.14–5.75 (3.90)	3.41	8	0.64–3.90 (1.32)	1.64
5	Yelandur	19	0.32–2.28 (1.67)	1.46	9	0.47–3.00 (1.87)	1.16
	Minimum		0.32	1.46		0.36	1.16
	Maximum		12.56	4.57		4.86	2.31
	Average		3.21	2.81		1.85	1.62
	Standard deviation		1.37	1.20		0.50	0.44
	Median		2.65	2.32		1.85	1.62
	Geometric mean		2.98	2.61		1.80	1.57

the taluks of Chamarajanagar district and the data analysed for each taluk is presented in Table 1. The activity of ^{210}Po in groundwater samples varied from 0.32 to 12.56 mBq L^{-1} with an average of 3.21 mBq L^{-1}. In each taluk, tenfold variation in ^{210}Po activity was found in water samples. The ^{210}Po concentrations ranged between 1.61 and 9.35 mBq L^{-1} in Chamarajanagar taluk, 1.02 and 12.56 mBq L^{-1} in Gundlupet taluk, 0.56 and 4.64 mBq L^{-1} in Kollegal taluk, 1.14 and 5.75 mBq L^{-1} in Hanur taluk and 0.32 and 2.28 mBq L^{-1} in Yelandur taluk. ^{210}Po activity in surface water samples was lower than groundwater samples. In surface water samples the ^{210}Po concentration varied from 0.36 to 4.86 mBq L^{-1} with an average of 1.85 mBq L^{-1} in the study area. The ^{210}Po concentration ranged between 0.95 and 3.23 mBq L^{-1} in Chamarajanagar taluk, 1.62 and 4.86 mBq L^{-1} in Gundlupet taluk, 0.36 and 2.52 mBq L^{-1} in Kollegal taluk, 0.64 and 3.90 mBq L^{-1} in Hanur taluk and 0.47 and 3.00 mBq L^{-1} in Yelandur taluk.

A high concentration of 12.56 mBq L^{-1} was observed in the groundwater sample of Madapattana village of Gundlupet taluk. This may be due to the presence of granite rocks and phosphatic rocks in the region which are known to contain higher concentration of radionuclides. Bore wells drilled in this region are deeper (>150 metres from the surface) than in other taluks and effluents from stone crushing industries are also the reasons for higher concentrations of ^{210}Po in this region. The lower concentrations were observed at villages of Yelandur taluk. This taluk is rich in ground and surface water sources and depth of bore wells ranged between 30 and 50 meters below ground level. The concentration of ^{210}Po in all the water samples of Chamarajanagar district were within the safe guideline level of 100 mBq L^{-1} recommended by WHO (2011)(14).

WHO has adopted a pragmatic and conservative approach and set the ingestion dose coefficient of 0.1 mSv from a year's consumption of drinking water and recommended 0.01 mSv y^{-1} as a safer level of ^{210}Po in drinking water. The dose coefficient used for estimating ingestion dose represents <5% of the average annual dose attributable to radiation of natural origin(14).

The ingestion dose to the population of every taluk due to ^{210}Po in water samples was calculated considering the average ^{210}Po activity in each taluk and is presented in Table 1. The ingestion dose due to ^{210}Po in groundwater varied from 1.46 to 4.57 μSv y^{-1} with an average of 2.81 μSv y^{-1}. The ingestion dose due to ^{210}Po in surface water varied from 1.16 to 2.31 μSv y^{-1} with an average of 1.62 μSv y^{-1}. These values are below the recommended value of 0.01 mSv y^{-1} by WHO (2011)(14) and 1 mSv y^{-1} by ICRP (2000)(19). The concentration of ^{210}Po in ground and surface water, and the average ingestion dose due to ^{210}Po is represented in Figure 3. Hence the radiological effect due to ^{210}Po in drinking water is relatively lower in Chamarajanagar district.

^{210}Po activity in food samples and associated effective dose

The ^{210}Po activity in food sample that are commonly used by the local population such as milk, rice, wheat, millets, pulses and some animal origin food are shown in Table 2. The population of Chamarajanagar district consumes a variety of food from plant origin and animal origin. The food habit and quantity of food

Table 2. ^{210}Po concentration in food samples of Chamarajanagar district and their ingestion dose.

Sl. No	Samples	No. of samples analysed	Concentration of ^{210}Po (mBq kg^{-1}) Range	Average	W_{FV} (kg y^{-1})	W_{FG} (kg y^{-1})	Ingestion dose to vegetarians (μSv y^{-1})	Ingestion dose to general people (μSv y^{-1})
1	Natural water	160	0.32–12.56†	2.53†	730*	730*	2.81	2.81
2	Milk	6	2.6–10.3†	4.25†	91.25*	91.25*	0.47	0.47
3	Raw Rice	8	47–242	96	136.88	132.31	15.77	15.24
4	Wheat	8	28–52	41	54.75	45.63	2.69	2.24
5	Millets	12	72–465	157	73.00	63.88	13.75	12.03
6	Pulses	10	50–190	79	27.38	18.25	2.60	1.73
7	Lean & Poultry products	5	60–115	98	–	8.94	–	1.05
8	Fish (river)	4	1124– 3000	1500	–	3.19	–	5.75
9	Fish (marine)	4	2000—48,000	33,000	–	2.01	–	79.50
10	Prawn (river)	4	20,120– 35,600	26,000	–	0.55	–	17.08
11	Prawn (marine)	4	25,000– 52,000	44,000	–	0.82	–	43.36
12	Crab (river)	4	71,000– 200,000	90,000	–	0.55	–	59.13
13	Crab (marine)	4	300,000– 1,300,000	8,00,000	–	0.37	–	350.40
						Total ingestion dose	38.09	590.80

W_{FV} = Total annual consumption rate of food item by a vegetarian. W_{FG} = Total annual consumption rate of food item by the general public. † = mBq L^{-1}, * = L y^{-1}.

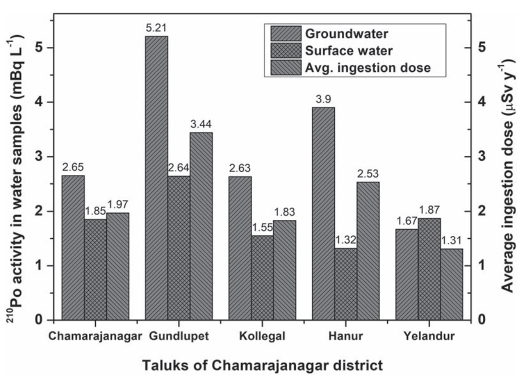

Figure 3. ^{210}Po activity in water samples and corresponding ingestion dose.

consumption by the people varies from one region to another depending on cultural practices, religious faiths and economical background.

Plant-derived foods were found to have lower activity concentration of ^{210}Po compared to animal origin foods. The ^{210}Po activity in milk samples varied from 2.6 to 10.3 mBq L^{-1} with an average of 4.25 mBq L^{-1}. A demographic survey was conducted to estimate the annual intake of food by the population using which the annual ingestion dose due to ^{210}Po in food samples were calculated (Table 2). Ingestion dose was estimated separately to the general public and to the people who are pure vegetarians. By considering an annual consumption volume of 91.25 L of milk per person, the ingestion dose due to ^{210}Po in milk samples has been estimated to be 0.47 μSv y^{-1}. In raw rice, the ^{210}Po activity varied from 47 to 242 mBq kg^{-1} with an average of 96 mBq kg^{-1} and the ingestion dose for vegetarian and general public are 15.77 and 15.24 μSv y^{-1}, respectively. In wheat, the ^{210}Po activity varied from 28 to 52 mBq kg^{-1} with an average of 41 mBq kg^{-1} and the ingestion dose are 2.69 and 2.24 μSv y^{-1} for vegetarians and general public respectively.

Millets like finger millet, pearl millet, foxtail millet, barnyard millet, proso millet, kodo millet and little millet and pulses like pigeon peas, green gram, black gram, horse gram and peas are commonly consumed in the form of side dishes along with main cereal/millet food items by the people in the study area. The ^{210}Po activity in millets varied from 72 to 465 mBq kg^{-1} with an average of 126 mBq kg^{-1} and in pulses, the ^{210}Po activity varied from 50 to 190 mBq kg^{-1} with an average of 79 mBq kg^{-1}. The ingestion doses due to the ^{210}Po activity in millets for vegetarian and general people were 13.75 and 12.03 μSv y^{-1}, respectively. The ingestion doses due to the ^{210}Po activity in pulses for vegetarian and general people were 2.60 and 1.73 μSv y^{-1}, respectively.

The public who consume both plant and animal origin food receive additional dose from meat and sea foods. The activity concentration in animal origin food varies across four orders of magnitude. The concentration of ^{210}Po in lean and poultry products varies from 60 to 115 mBq kg^{-1} with an average of 97 mBq kg^{-1} and the resultant average ingestion dose is 1.05 μSv y^{-1}.

Table 3. Concentration of ^{210}Po in water from different regions of the world.

Sl. No	Geographic region	Source of water	^{210}Po Activity (mBq L^{-1})	References
1	**Malaysia**	River	0.63–14.98	(15)
		Treated water	0.34–6.80	
2	**Karnataka, India**	Borewell	1.89–4.18	(21)
		River	0.86–4.49	
3	**Jaduguda, India**	Groundwater	< 0.08–7.41	(22)
4	**Kalpakam, India**	Groundwater	0.6–2.6	(3)
5	**Belgium**	Drinking water	< 0.1–3.51	(23)
6	**Italy**	Bottled drinking mineral water	<0.4–21.01	(20)
7	**Japan**	Bottled water	1–4.9	(24)
8	**North Vietnam**	Thermal waters	0.56–8.26	(25)
9	**Present study**	Groundwater	0.32–12.56	
		Surface water	0.36–4.86	

^{210}Po concentrations in marine organism are higher compared to fresh water organisms. The average activity of ^{210}Po in fresh water fish, prawn and crab are 1.5, 26 and 90 Bq kg^{-1}, respectively. Being continental location, the district receives more fresh water food than sea food. Concentrations of ^{210}Po in marine fish, prawn and crab varied from 2 to 48, 25 to 52 Bq kg^{-1} and 0.3 to 1.3 kBq kg^{-1} with a respective average levels of 33, 44 and 0.8 kBq kg^{-1} respectively. Sea food has higher ^{210}Po concentration because they receive radionuclides directly from the surrounding water and also from their food which contains higher quantities of minerals including radioactive elements compare to river water. In biota, contrasting uptake, excretion and bio distribution of polonium can also take place.

The ingestion dose due to activity of ^{210}Po in fresh water fish, prawn and crab are 5.75, 17.08 and 59.13 μSv y^{-1}, respectively. The ingestion dose due to ^{210}Po in marine fish and prawn are 79.50 and 43.36 μSv y^{-1}, respectively.

Marine crab showed the highest concentration of 1.3 kBq kg^{-1} among all the edible samples of the present investigation. Being a continental location, the study area gets marine crabs from the nearby seaside markets and hence consumption of marine food is generally lower than consumption of fresh water food. Crabs have got high nutritive value and low fat content. People consume marine food very occasionally and most of the families cannot afford marine food due to their economical background. Even though the consumption of marine crabs is less, people can receive higher dose of 350.4 μSv y^{-1} due to very high activity of ^{210}Po in marine crabs.

The concentration of radionuclides in water from different regions of the world is shown in Table 3. Ahmed et al. (2018) reported that ^{210}Po concentration in the Langat river varied from 0.63 to 14.98 mBq L^{-1} which is higher compared to other reported values. The elevated values are probably because of granite zone through which the river flows and the terrestrial primordial sources like ^{238}U and its progeny ^{210}Po along with atmospheric and anthropogenic sources in that region(15). Desideri et al. (2007) have reported the highest value of 21.01 mBq L^{-1} in potable mineral water which is an exception where 72.5% of the samples show lesser polonium activity of <1 mBq L^{-1} (20). The values obtained in the present investigation were much lower compared to other values reported globally.

Total ingestion dose of ^{210}Po in pure vegetarian population and general people are 38.09 and 590.80 μSv y^{-1}, respectively. People consuming food from both plant and animal origin receive a dose of 552.71 μSv y^{-1} due to consumption of lean, poultry products, fresh water and marine organisms which contain higher activity of ^{210}Po. The ingestion dose values of the present study lie within the reference level of 1 mSv y^{-1} (18, 19).

Conclusions

The ^{210}Po activity concentration was found to be higher in groundwater than in surface water. The average concentration of ^{210}Po in ground and surface water of Chamarajanagar district, Karnataka, India was 3.21 and 1.85 mBq L^{-1} respectively. The study examined ^{210}Po concentration in food samples including plant and animal-origin and estimated the radiation dose. The study revealed that the ^{210}Po concentration in organisms from marine origin was higher compared to that of river origin. Among the samples studied, highest ^{210}Po concentration of 1.30 kBq kg^{-1} was observed in marine crabs, whereas in the fresh water crab it was 200 Bq kg^{-1}.

The total estimated radiation dose due to ^{210}Po in food and water was found to be 38.09 and 590.80 μSv y^{-1} for the population consuming only vegetarian food and for the population consuming both vegetarian and animal origin food, respectively. The radiation dose due to consumption of rice and millets was 15.77 and 13.75 μSv y^{-1} respectively. Radiation dose due to consumption of natural water, milk, wheat and pulses was low compared to consumption of rice and millets. People consuming animal origin food were exposed to additional dose in μSv y^{-1} of 79.50 (marine fish), 17.08 (river prawn), 43.36 (marine prawn), 59.13 (river crab) and 350.40 (marine crab). The current study reports for the first time that the total radiation dose due to ^{210}Po ingested through water and food in Chamarajanagar district is within 1 mSv y^{-1}, the recommended level by ICRP.

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