Radionuclide activity concentration in soil, granites and water in a fluorosis endemic area of India: An oral health perspective

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ABSTRACT

Background: The idea that hilly areas have a high background radiation which in turn is linked to dental fluorosis is widely held by many in India. There is little evidence to confirm this.

Objective: The study aimed to investigate the level of environmental radionuclide in order to determine any interrelationship between radionuclide and dental fluorosis in Pavagada, a fluorosis endemic area of Karnataka, India.

Methods: Gamma radionuclides (Th232, Ra226 and K40) were determined by high purity germanium (HPGe) gamma-ray spectrometer (Bq Kg⁻¹) using a 50% relative efficiency p-type broad energy HPGe detector. Fluoride was estimated using fluoride Ion-selective electrode (ISE). Fluorosis was assessed using WHO diagnostic criteria.

Results: The average mean $\pm$ SD activity concentration in soil for K40 was detected between 416.6 ± 9.3 and 769.1 ± 15.0 . Th232 was estimated between 13.6 ± 0.6 and 57.6 ± 1.3 ; and Ra226 was found between 6.4 ± 0.4 and 21.6 ± 0.7 . Similarly, the mean activity concentration detected (Bq kg⁻¹) in granite for K40 ranged between 259.8 ± 6.9 and 1608.5 ± 26.9 . The activity concentration of Th232 ranged between 26.4 ± 0.8 and 57.6 ± 1.3 ; and that of Ra226 was between 6.6 ± 0.3 and 21.6 ± 0.7 . Drinking water contains 2.98 ppm of fluoride. Dental fluorosis was diagnosed as mild to moderate.

Conclusion: The radionuclide activity in water, soil and granites were below the detection limit. Hence, the study revealed no association of fluorosis and radionuclide level in Pavagada, Karnataka, India. This helps resolve an ambiguity.

1. Background

Naturally occurring radiation exposing living and non-living objects is known as 'background radiation' and radionuclide.¹ The earth is exposed to naturally radioactive substances, i. e, radionuclide and nearly 82% of human species absorb radiations naturally.¹ Several studies revealed different values of background radiation from radionuclide are exposed to human species.¹ The highest known level of background radiation affecting a population in India was recorded in the Kerala and Tamil Nadu states of the country.² In these places nearly

1 in 40,000 people receive doses which are more than an average of 15 mSv per year, (the maximum permissible dose (MPD) is 27,000 mSv annually) from gamma radiation in addition to a similar dose from radon.^{1,2} The association between increased fluoride content in water and the level of background radiation in Kerala such as- Karangapally, Kerala and other places in India is not reported, although it has been an ambiguity remains among the researchers and population regarding an interrelationship between of high level of fluoride and that of radionuclide, needs to be addressed to clear with factual evidence.

The earth's crust such as soil, sands and rocks contain uranium and

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thorium.² The concentrations of radionuclide in soil, granite and rocks depend on the geographical location.^{1,2} For instance, the concentration of potassium (K+) ranges between 1000 and 30,000 ppm but the level increases in rocky areas.² Therefore, terrestrial background radiation depends on the level of potassium and uranium series (238U–206 Pb), including thorium series (232Th to 209 Pb), and actinium series (235U–207 Pb). Each of these series consists of many α , β , and γ emitters. The concentration of these radioactive isotopes in the soil and water varies greatly.^{1,2}

The soil and rock in southern India are rich in fluoride and may contribute high level of fluoride in the groundwater of Pavagada Taluk where mean value of Fluoride in drinking water is 2.98 ppm.³ Pavagada is an administrative area of a district, typically comprising a number of villages and a small town of Tumkuru district of Karnataka state in India, the area is surrounded by high rocky hills and mountains. Pavagada is widely covered with granites and corundum over 60 kms.^{3,6} The granite contains uranium; thorium and the radioactivity are evident.^{2,4} However, there are conflicting reports about the relationships between an increased amount of nucleosides and the high content of fluoride in granite, soil and water.^{5,7}

A recent case study by A. Gordon (2016) has discussed about the potential risk of skeletal fluorosis from background radiation.⁸ The author also emphasized that some bottled water may contain radionuclide which are collected from rocky or mountainous areas may affect health.⁸ The author recommends screening of drinking water for any gross alpha and beta radiation activity annually, based on certain standards (IBWA, 2012; NSF, 2015).⁸

But the World Health Organization (WHO) says that the levels of activity above the suggested target values does not necessarily indicate a serious health risk to the consumers (WHO, 2006).^{6,7}

There are discussions on whether level of increased nucleosides radioactivity (which is a determinant of background radiation) influences fluoride content and vice versa).^{6,8} Nevertheless, this study collected samples of rocks, granite, sand and water to examine the level of radionuclides activity in the areas of Pavagada; where a high proportion of population is badly affected with dental fluorosis.³ The objective of the study was to determine the radionuclide activity in soil, granite and water in the Pavagada fluorosis endemic area of India.

2. Materials and methods

2.1. Study location

The soil, granite and water were collected from seven villages of the Pavagada region of Tumkuru District, Karnataka state in India.

2.2. Method of sample collection

The Soil, granite and water samples were collected by a random sampling method from Pavagada Taluk of the Tumkuru district located in Karnataka State of India. The collection of samples was performed based on equidistance and from an area of 1 m² separated from each other by a distance of 300 m to cover the study site and to assess a significant local spatial variation in terrestrial radioactivity.³ All soil samples were collected from the locations away from high buildings, roads or any man-made structures or trees, as these could affect the level of background radiation.

3. Preparation of the samples

3.1. Soil

The area of about 1 sq. meter open surface at ground level, and those were away from public road and buildings were selected for sample collection. The soil samples were collected from all the four corners and centre of the marked area. The top layer of vegetation and

root was removed and digged the soil up to 6 cm depth and placed the sample inside in a polythene bag.^{9,10} The soil samples in the laboratory were air dried for a week and then oven dried at 105 °C for 24 h. The samples of soil was pounded into powder, and sieved out through 2 mm wire, so that a uniform particle size was obtained. In order to maintain radioactive equilibrium between all three radionuclide, the soil samples were then packed into a 120 mL air tight polyethylene cylindrical container, dry-weighed, and stored for a period of 32 days.¹⁰

3.2. Granites

Firstly, the samples were dried at 110 °C and were pulverized, homogenized and sieved through a 1-mm mesh.¹¹ Five hundred gms of each meshed sample was placed in a Marinelli beaker and then stored for four weeks to reach an equilibrium state between radium and its decay products.^{11,12}

3.3. Water

Immediately after the collection of each water sample, the water was acidified with 50 ml of 11 M HCl to prevent adsorption of radionuclide on the container walls (IAEA, 1970).¹³ The collected water samples were filtered through the filter paper to remove very fine undissolved particles. For measuring radionuclide, each of the Fifty one (n = 51) water sample was evaporated to reduce its volume to 500 ml and packed into 500 ml cylindrical containers, sealed tightly and wrapped with thick vinyl tape around their screw necks, and kept for at least 25 days to ensure that equilibrium was achieved between Ra226, K40 and Th232 and their respective progeny.^{13–15}

4. Measurement of radionuclide

The concentrations of gamma radionuclide were determined by gamma-ray spectrometer using a 50% relative efficiency p-type broad energy HPGe detector (Canberra Industries Inc., USA),¹⁰ having FWHM of 2.00 keV at 1.33 MeV. The detector was shielded in an ultra-low background graded lead shielding of 150 mm thickness with a nitrogen gas purging arrangement. The spectrum was acquired and analysed by a 32 K multichannel analyser (Lynx, Canberra Industries Inc., USA) and a Genie 2000 advanced gamma analyses software. The activity concentrations (Bq Kg⁻¹) of Th232, Ra226 and K40 in soil (N = 4), granite (N = 3) and water (N = 2) were determined).^{6–9,9,10,10–18,22}

5. Results

The activity concentration for all radionuclides including Radium (226Ra) were detected within normal range, as per Table 1. The level of activity of the concentration of Th232, Ra226 and K40 in the water was

Table 1
Activity concentration of Bq kg⁻¹ in Soil and Granite of different regions at Pavagada.

Source N = 3	Location	Activity concentration in Bq kg ⁻¹		
		⁴ K	²³² Th	²²⁶ Ra
		Mean ± SD	Mean ± SD	Mean ± SD
Soil	Kumudu	566.9 ± 15.0	54.4 ± 1.8	14.9 ± 0.8
	SimaralaKunte	416.6 ± 9.3	13.6 ± 0.6	6.4 ± 0.4
	Venkatapura	769.1 ± 15.0	53.5 ± 1.4	13.6 ± 0.7
	Kodamagadu (Pavagada)	652.5 ± 17.0	33.7 ± 1.9	14.4 ± 1.1
Granite	Kanavanghalli	1608.5 ± 26.9	57.6 ± 1.3	21.6 ± 0.7
	Kodamagadu (Pavagada)	259.8 ± 6.9	26.4 ± 0.8	6.6 ± 0.3

** The values are almost within the permissible limit BDL means Below Detection Limit.

Table 2Activity concentration of Bq kg⁻¹ in Water of different regions at Pavagada.

Sl.No	Location N = 3	Source of Water	Activity concentration in Bq L ⁻¹		
			⁴ K	²³² Th	²²⁶ Ra
1	SimkarlaKunte	Bore Well	BDL**	BDL**	BDL**
2	Venkatapura	Bore Well	BDL**	BDL**	BDL**

** The values are almost within the permissible limit, BDL means Below Detection Limit.

below the detection limit (BDL), as per Table 2. The average mean $\pm$ SD activity concentration (Bq kg⁻¹) in soil for K40 was between 416.6 $\pm$ 9.3 and 769.1 $\pm$ 15.0; Th232 was between 13.6 $\pm$ 0.6 and 57.6 $\pm$ 1.3; and for Ra226 was between 6.4 $\pm$ 0.4 and 21.6 $\pm$ 0.7. Similarly, mean activity concentration detected (Bq kg⁻¹) in granites for K40 was between 259.8 $\pm$ 6.9 and 1608.5 $\pm$ 26.9; the amount of Th232 was between 26.4 $\pm$ 0.8 and 57.6 $\pm$ 1.3; and that of Ra226 was between 6.6 $\pm$ 0.3 and 21.6 $\pm$ 0.7. The radionuclide activity in water was below detection limit (BDL). The grain size of the prepared sample ranged between 80 μ and 120 μ . The mean fluoride concentration of drinking water in fluorosis endemic area (mostly dental) was detected 2.98 ppm.

6. Discussion

Pavagada Taluk is draught-prone area, and environmentally it has high temperature and dry weather condition, Pavagada is surrounded by rocky hills and hillocks, less greens comparing to other areas of the state of Karnataka. The rocks contain mostly granite. Granite has high fluoride content. Usually fluoride from granite leaches into the underground water, the main source of drinking water in the area. In the study. The study aimed to measure radionuclide. The Gamma spectrometry was used to measure radionuclide as a standard method, and it identifies the intensity of gamma radiation versus the energy of each photon, used in other studies^{9–11}

Our results revealed that, the activity concentration was low in the soil for K40, Th232 and Ra226. The results were similar to the study conducted by Nafee et al. (2017).²⁰ The activity concentration in granite for K40 was higher in the Kanavangahalli village (1608.5 $\pm$ 26.9 Bq kg⁻¹). The Th232 and Ra226 activity concentration was lower. These findings also are similar to the results of Nafee et al. (2017).²⁰ The radiation results are reported in Tables 1 and 2 The radiation released in water was found below the level of detection (Table 2). The soil and rocks showed variations in their radiation activity (Table 1).

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) is a division established by the General Assembly of the United Nations in 1955 has a directive to recommend permissible limit of environmental radiation.²¹ The UNSCEAR has a specific objective is-to assess and report levels and effects of exposure to ionizing radiation and contents of trace elements including fluoride.²¹ Policy departments and the organizations in many countries agrees with the expert committee's guidance and the authorities control the level of radiation for a protective measure of health and environment.²⁰ The activity concentration (Bq kg⁻¹) estimates given by UNSCEAR 2000 for K40 was 500 Bq kg⁻¹, Th232 was 50 Bq kg⁻¹ and Ra226 was 50 Bq kg⁻¹.^{21,22} Based on our results, the granites have relatively high activity of radionuclide K40 (1608.5 $\pm$ 26.9 Bq kg⁻¹), which is similar to other reports (¹⁹,2p), our samples were ranged within the permissible limit and some of the samples were estimated below detection limit. However, it has been seen that there is no interrelationship between high level of fluoride in drinking water associated dental fluorosis and the level of radionuclide in the environment of Pavagada area of India.

7. Conclusion

The level of radionuclide activity, except the granite samples which expressed relatively high level of radionuclide activity, but the level was not exceeding the maximum tolerated dose (MTD). In the study, it has been revealed that, apparently there is no obvious relationship between the high concentration fluoride in drinking water associated dental fluorosis and the level of radionuclide in the environment, which is safe for the species, nevertheless, an extensive and in-depth study is recommended. Misconceptions and ambiguity regarding the links with environmental radiation levels serve to obfuscate the issue and hamper any potential public health strategies to deal with the endemic levels of fluorosis in this area. Hence, an evidence based information needs to be addressed for the concerned population, researchers and the clinicians.

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Declaration of competing interest

No Conflict of Interest.

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