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Uranium in drinking water from the south coast districts of Kerala, India

S. Ben Byju[1], A. Sunil[1], M.J. Reeba[1], E.P. Christa[1], V.K. Vaidyan[1], R. Prasad[2], P.J. Jojo[1,3*]

[1]Centre for Advanced Research in Physical Sciences, Department of Physics, Fatima Mata National College, Kollam – 691001, Kerala, India
[2]Department of Applied Physics, ZakirHussain College of Engineering & Technology, Aligarh Muslim University, Aligarh-202002, India
[3]Department of Physics, University Malaya, Kuala Lumpur-50603, Malaysia

Background: The South-west coast of India is known to have very high levels of natural background radiation due to the monazite beach sand. Uranium is the heaviest trace element found in all terrestrial substances at varying levels with chemical and radio toxicities. It supports several short-lived radioisotopes in its decay series including radium. Uranium in drinking water is important in terms of the ingestion dose. ***Materials and Methods***: The present study reports the results of uranium analysis of 346 drinking water samples from the three costal districts of Kerala using fission track registration technique. ***Results:*** Results obtained show that uranium concentrations vary from 0.31 µg/l to 4.92 µg/l equivalent to the specific activity of 3.9 Bq/m^3 and 62 Bq/m^3, respectively. ***Conclusion:*** The estimated daily intake of uranium through drinking water is lower than the recommended limits. The distribution of uranium in water bodies shows a heterogeneous nature of distribution. **Iran. J. Radiat. Res., 2012; 10(1): 31-36**

Keywords: *High background radiation area, uranium, drinking water, fission track registration technique.*

INTRODUCTION

The largest proportion of human exposure to radiation comes from natural sources – from external sources of radiation, including cosmic and terrestrial radiation, and from inhalation or ingestion of radioactive materials [(1)]. Of all naturally occurring elements uranium is heaviest and radiotoxic with a very long half-life. It is a ubiquitous radioactive trace element found in almost all terrestrial substances in different levels of concentration. It also supports several short lived radioisotopes in its decay series including radium with potential radiological importance. Water plays an important role in the geophysical and geochemical processes, which slowly recycles the trace elements to and biosphere. Intake of higher levels uranium can lead its accumulation in the organs like kidney and can be carcinogenic [(2)]. Nephritis is the primary chemically induced effect of uranium on human health [(3)].

The Southwest coast of the Kerala State in India is known to have very high levels of natural background radiation owing to the rare earths rich monazite sand present in large amount. The major sources responsible for exposure are naturally occurring radio nuclides in the earth's crust such as ^{232}Th, ^{238}U, ^{40}K which occur in abundance in minerals such as monazites and zircons. These radionuclides impart not only the external dose to the human beings but also causes ingestion and inhalation doses to human body through intakes of air, water and food [(4)]. The monazite sand in the region contains about 9% thorium oxide, 0.35% uranium oxide along with phosphorus pentoxide, rare earths, and oxides of titanium, cerium, iron and silicon [(5)]. Therefore, the ingestion doses comprise of the long-lived radionuclides through the intake of water and the vegetables grown locally. There is evidence from both human and animal studies that radiation exposure at

Corresponding author:
Dr. Panakal John Jojo,
CARPS, Fatima Mata National College, Kollam-691001, India.
Fax: *+603 79674146*
E-mail: *jojo@jojopanakal.com*

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low to moderate doses may increase the long -term incidence of cancer. Animal studies in particular suggest that the rate of genetic malformations may be increased by radiation exposure. According to World Health Organisation (WHO) no deleterious radiological health effects are expected from consumption of drinking-water if the concentrations of radionuclides are below the guidance levels equivalent to a committed effective dose below 0.1mSv/year [1]. A systematic study has been conducted to evaluate the levels of uranium in drinking water from the natural resources in the coastal south Kerala, India.

MATERIALS AND METHODS

Estimation of uranium in water first involves the selection of sampling sites and sampling procedure. The region selected for the study was the coastal strip extending about 90 km from Varkala to Ambalappuzha. The sampling locations were selected towards the north of Varkala Beach (Trivandrum district) up to Navarakkal Temple, Ambalappuzha (Allappey district) with the locations along the coastal line including the High Background Radiation Area (HBRA) as shown in the figure 1. A total of 58 locations were selected and five to seven samples were collected from each location making a total of 346 samples. Samples were coded as 'W' (well), 'P' (pond) and 'B' (bore well) according to their sources. Drinking water samples were collected from 125 ponds, 137 wells and 84 bore wells in the study area including the HBRA (double shaded in the figure 1). All samples were collected between the coastal line and the National Highway 47 (NH47) passing almost parallel to the western coastal line. The study area is bound by latitude 8°43'N and 9°20'N and longitude 76°22'E to 76°43'E approximately.

collected only from the natural water bodies namely pond, well and bore well, which were used for drinking. Care was taken to have a geographically uniform distribution in choosing locations for sample collection. Small and clean plastic bottles (20 ml capacity) were used for sample

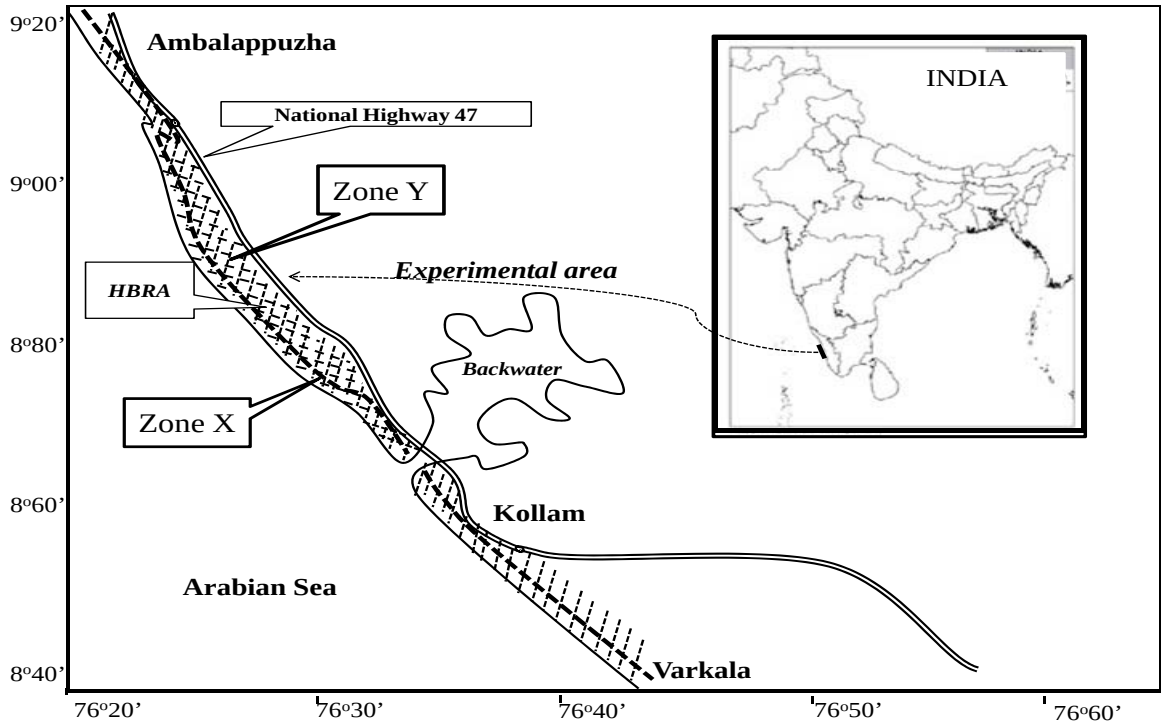


Figure 1. Experimental area including the high background radiation region.

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collection. Bottles were pre-rinsed with distilled water and then with the experimental water at the time of sample collection. Each sample brought to the laboratory was slightly acidified by adding a drop of nitric acid to minimize the loss of uranium through absorption in the bottles. Water samples were collected mostly from the surface of the water bodies in the case of pond and well. Altogether 346 samples were collected in six phases from various sources for analysis.

Fission track registration technique is a very sensitive and reliable method for analyzing uranium. This method is capable of determining uranium levels even in sub-ppb (particles per billion) levels and is relatively cheaper also. Experimental technique known as 'dry method' has been used for the analyses [6].

Dielectric fission track detectors (Makrofol – KG) cut in the form of circular discs of 1.3 cm diameter were properly washed and rinsed with the double distilled water. They were carefully numbered and were arranged on a tray. Each drop of water samples (0.05ml) was placed on each disc using a micropipette. Then, they were allowed to evaporate in a hot air oven at about 60° C to leave a circular residue of non-volatile substances in the water samples including natural uranium. Each detector disc with the non-volatile residue, including uranium, is then covered with another identical detector disc. The pair of discs is then sealed with polypropylene tapes to form a pellet of the sample [7].

These pellets were encapsulated in an aluminum can of about 5 cm length and 1.5 cm diameter. A blank pellet, without any water sample residue in it, was also placed in the can to assess the background tracks, if any. A pair of circular pieces of micro-slides, which acts as neutron flux dosimeters, was also kept in the can. This capsule was sent for irradiation at Bhabha Atomic Research Centre (BARC), Bombay in the thermal column of the APSARA Reactor at a flux of 10^{16} nvt for 3 h. After receiving the irradiated samples, from BARC the detector discs were separated and washed thoroughly with water. Then they were etched in 6.25N KOH solution at 60° for 20minutes so that the fission tracks were developed into clearly visible size [6].

The tracks were seen in a circular region where the evaporated water drop had left a residue of non-volatile matter including uranium in the water sample. The distribution of tracks was such that the circular region had a non-uniform distribution of tracks. The rim of the circular region had a higher track density and the interior portion had almost a uniform distribution of tracks. This is due to the fact that the water drop leaves maximum non-volatile residue along the rim during evaporation. To find the total number of tracks on the detector, the rim as well as the interior was scanned separately using an optical transmission type research microscope at a magnification of 400X. Using optical scanning total number of tracks was determined on the detector discs following the standard protocol [6, 8]. Deducting the background tracks recorded in the blank pellet, the actual number of tracks was found for both the detector discs of each pellet. For assurance of accuracy of results at least three pellets of each sample were used for analysis and only the congruent values were accepted. The average was taken as the total number (N) of tracks for the sample. To obtain the thermal neutron dose, the irradiated standard glass placed in the capsule was cut to form a fresh surface. The glass piece was etched in 48% Hydrogen Fluoride at 23°C for 5 seconds to make the tracks visible and is scanned to obtain the fission track density. The neutron dose was calculated using the relation [7]:

$$\Phi = K \rho \qquad (1)$$

where K is a constant ($=1.028 \times 10^{11}$) depending on the material of the standard glass used as flux dosimeter and ρ is the fission track density in the standard glass. With these known factors, uranium concentrations in water samples were

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calculated using the equation [6]:

$$C_w = [(NM)/(V G N_a E \sigma \Phi I] \quad (2)$$

where C_w is the concentration of uranium in water samples, N the total number of tracks, M the atomic weight of the fissile material (235), V the volume of the water droplet, N_a Avogadro's number, E the etching efficiency of the Makrofol-KG detector, σ fission cross section of the fissile isotope (580 barns), Φ the neutron flux used and I the isotopic abundance ratio [6].

RESULTS AND DISCUSSION

For the purpose of analysis, the experimental area was divided as 'X' and 'Y' zones based on their location with respect to coastal line and NH 47. 'X' zone represents the region proximate to the sea with 100 m distance from the west coast line. The remaining region was labeled as zone 'Y'.

Results of the analyses of the 346 water samples collected from different sources in the south west coast of India show that uranium concentration vary from 0.31 µg/l to 4.92 µg/l equivalent to the specific activity of 3.9 Bq/m^3 and 62 Bq/m^3, respectively. The arithmetic mean (AM), geometric mean (GM) and geometric standard deviation (GSD) of the measured values for the two zones are presented in the table 1. In general there is a clear elevation in the uranium concentration in the well and pond water samples collected from the zone X as compared with those from zone Y as indicated by the AM and GM values. This can be attributed to the abundance of monazite sand available in plenty in the coastal region. It is reasonable to assume that uranium is transferred to local water bodies through leaching. The skewness and Kurtosis of the measured data are also shown in the table 1.

For the zone X, the concentration of uranium for well water is slightly positively skewed meaning that majority of the data are slightly lower than the average value. The data for pond showed a highly left (negatively) skewed distribution with most the values concentrated on the right of the mean, with extreme values to the left. This indicates that the concentrations of uranium in these samples are generally higher than the mean value presented in the table 1. For bore well the data is somewhat symmetric about the mean. For the zone Y, both well and pond water showed a highly right skewed distribution indicating the exuberance of values lower than the mean. In this case also the bore water showed an approximately symmetric distribution about the mean.

For the zone X, uranium concentrations in well water showed a leptokurtic distribution with a well-defined central peak. The pond water samples showed a platykurtic curve with a broad centre. The mesokurtic distribution of bore well indicated a normal distribution. For the zone Y, water samples from well, pond and bore well showed platykurtic distribution with different degrees of kurtosis.

Table 1. Measured mean values of uranium concentration for the two zones.

	Zone X			Zone Y		
Source	Well	Pond	Bore well	Well	Pond	Bore well
Sample size	64	46	38	73	79	46
AM (µg/l)	2.19	3.46	1.09	1.09	1.12	0.98
GM (µg/l)	2.02	3.28	0.98	0.96	1.02	0.86
GSD	1.41	1.34	1.18	1.18	1.21	1.12
Skewness	0.54	-1.76	0.14	1.07	1.45	0.26
Kurtosis	3.45	1.26	2.98	2.38	1.85	2.43

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The obtained data was analyzed statistically to find any correlation between the levels of uranium in various sources using Pearson's method [9]. The analysis was done between the average values of uranium concentrations from the three types of sources for the fifty eight locations. It showed good positive correlation (r = 0.865 at 0.05 level of significance) between the levels of uranium in pond and well for the zone X. No such correlation could be obtained among the samples for zone Y. The ponds and wells in the zone X are relatively shallow and must be sharing same water table with the similar geophysical conditions.

WHO suggests a guidance value of Tolerable Daily Intake (TDI) of 15 mg/l equivalent to 370 mBq/kg assuming 60 kg adult consuming 2 l water per day [1]. The concentrations of uranium measured in the present work shows that they are well within the suggested limit. A recent study conducted in the packaged drinking water samples in India found that the range of uranium in water between 0.04 µg/l to 3.88 µg/l [10]. A very recent study conducted in India in ground water shows that the activity of radium (^{226}Ra) which can be treated as the activity of uranium was found to vary from 3.5 mBq/l to 208 mBq/l[11]. Analysis of uranium in drinking water in Punjab and Himachal Pradesh regions in India using laser induced fluorimetry was found to vary from 1.39 ± 0.16 to 98.25 ± 2.06 ppb with a mean value of 19.84 ± 0.87 ppb [12]. Uranium concentration analysis in drinking water using the same technique employed in the present study held in Muzaffarabad and hilly areas of Reshian in Pakistan were reported to vary from 0.03 ± 0.01 µg/l to 6.67 ± 0.14 µg/l with an average of 1.36 ± 0.05 µg/l [13].

While comparing with similar studies in other parts of the world, the levels in drinking water were reported to vary from 1.0 to 10.90 µg/l in Iran [14] and the mean value of uranium in drinking water was 9.6 ± 7.1 Bq/m^3 in Poland[15]. Another study conducted in 10 areas in Korea with about 500 drinking water samples reported a geometric mean of 0.17 µg/l [16]. All of these reported values in the literature are in very good agreement with the results of our analysis.

CONCLUSION

From the present observations it can be concluded that the concentration of uranium level vary considerably from natural source to source and place to place. There is no alarmingly high uranium concentration in any of the samples analyzed. The measured values are in good agreement with the reported values in the contemporary literature. The distribution of uranium in water bodies shows a heterogeneous nature of distribution. It is observed that the water bodies near to the sea have higher levels of uranium concentration. There is a positive correlation between the uranium levels in water collected from the pond and well samples along beach area. The daily intake of uranium through drinking water in the region is much less than the tolerable intake limit.

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