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PRIMORDIAL RADIONUCLIDES DISTRIBUTION AND DOSE EVALUATION IN UDAGAMANDALAM REGION OF NILGIRIS IN INDIA

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Abstract - The activity concentration of primordial radionuclides i.e., ^{238}U series, ^{232}Th series and ^{40}K, in soil samples collected from Udagamandalam environment, have been measured by employing NaI (Tl) Gamma ray Spectrometer. The absorbed gamma dose rate has also been simultaneously measured by using both Environmental Radiation Dosimeter at each soil sampling location (ambient gamma dose) as well as from the gamma dose derived from the activity concentration of the primordial radionuclides. The results of activity concentration of each radionuclides in soil, absorbed dose rate in air due to soil activity and possible cosmic radiation at each location along with human effective dose equivalent for Udagamandalam environment are presented and discussed.

INTRODUCTION

The most important places among the well documented high background radiation areas of the world inhabited by sizable population are Brazil, China, India and Iran [1-4]. The source of high background radiation is monazite deposits in the first three cases and radium in soil/water and radon in air are the source of high background in Iran [1-4].

In India, monazite placer deposits have been found along its long coastal line: Ullal (Karnataka), Chavara (Kerala), Manavallakuruchy and Kalpakkam (Tamil Nadu) and Chatrapur (Orissa) [5-7]. The external radiation level in the monazite region of India is similar to that reported in Brazil. But the monazite deposits of South West Coast (Kerala and Karnataka) and certain places of East Coast (Tamil Nadu) of India are more extensive than those in Brazil [8]. High content of thorium and traces of uranium are reported from these areas mainly due to the presence of Monazite sand found as beach placer. These Thorium and Uranium might have got dispersed or redistributed during igneous, sedimentary and metamorphic cycles of geological evolution, which might have resulted in small concentrations of deposits under favorable geological processes. Investigation on the distribution of these radioactive elements, and to some extent radiation levels have been carried out in these areas in order to trace the source and nature of the associated minerals covering radioactivity.

Information in literature indicates that the monazite deposits on the coastal area of Kerala and Tamil Nadu in India are formed due to the weathering of rocks in Nilgiri hills and Western ghats system [8]. A similar argument says that the monazite deposits in the coastal area of

Brazil are formed from the progressive weathering of primary rocks and erosion of the sedimentary peneplains by transportation and successive concentration of monazite sand, which adds support to the above literature information [9].

Also most of the river confluenting in Arabian sea of south west and Bay of Bengal of south east coast have their source in Western ghats system and Nilgiri hills. Radiation survey is being carried out in the western part of Western ghats system to find the exact origin of monazite deposits. However, there has been no radiation study available for Nilgiri hills, which form the eastern side of Western ghat system. Hence a study has been undertaken in Nilgiri hills which consists four taluks namely Udagamandalam, Coonoor, Kotagiri and Gudalur. The present paper discusses about the results radioactivity measurements of Udagamandalam region.

STUDY AREA

The Nilgiris biosphere is well known for its splendidly beautiful environment. This biosphere is one of the oldest and most important Eco systems in India. The Nilgiris is a highly populated (7,10,214 - 1991 census data) area, spreading over a wide area of 2549 square km.

This biosphere lies between 11° 5′ and 11° 45′ N latitude and 76° 13′ and 77° 2′ E longitude and is situated in eastern part of Tamil Nadu state in Southern India. The Nilgiris are well defined massif that forms the southern limit of the main Western ghat system that stretches unbroken from Mumbai in the North India. The Nilgiris is hardly a plateau in the strict sense of the word, in which valleys drop to 1,600 metre and peaks rise to 2,600 metre. The highest peak of South India 'Dodabetta' (2,648 metres above mean sea level) is situated in Udagamandalam region of the Nilgiri hills. The average elevation of the biosphere is 1,981 m above mean sea level. The annual average rainfall is 1800 mm. The annual temperature variation is from around 4° C to 24° C [10]. The eastern and southern sides of the Nilgiris are surrounded by other districts of Tamil Nadu State whereas the western side is surrounded by Kerala State and the northern side by the Karnataka State. The rivers originating from Udagamandalam and Kotagiri are confluenting in Kerala and Tamil Nadu coasts.

The Nilgiris is one of the important tourism areas in India, with a very large moving population from in and around the country. The study area, Udaga- mandalam in Nilgiris, is shown in Fig.

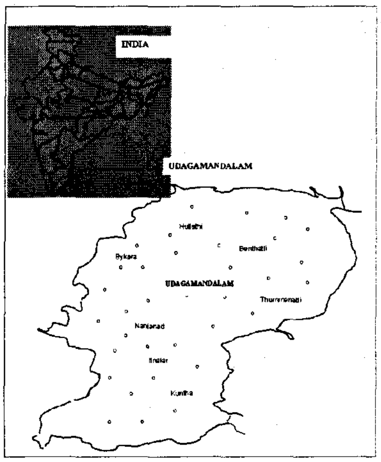

Fig. 1. Study area - Udagamandalam

AMBIENT SURVEY

The radioactivity status of the environment is defined mainly by gamma radiation in a particular region since gamma rays contribute most of the external exposure to population. Environmental radiation dosimeter (ERD), type ER 705, supplied by Nucleonix System PVT Ltd., Hyderabad, India has been used in the present study for the measurement of gamma radiation level at all the sampling locations. The dosimeter is designed to read exposure rate in two accuracy ranges of 0.1(R/h and 1(R/h. The survey meter is calibrated regularly using standard source (^{137}Cs), before starting survey work.

The gamma dose rate measurement has been done at 1metre above the ground level. At each location, a total of 5 readings have been recorded. Geometric mean value of the measured readings is calculated to reduce the small-scale variations of the level at a site. Finally, the survey meter readings, recorded as exposure rate (μR/h), have been converted into nGy/h using appropriate conversion factor. Since the study area is at an altitude of around 2,000 meters, the cosmic radiation also contributes a significant dose to the background radiation level. Hence, at each location, the ambient radiation level includes cosmic radiation dose to the gamma radiation, which is relatively varying to the altitude of the location.

SOIL SAMPLING AND ANALYSIS

In the natural environment, rocks undergo a continuous process of weathering owing to several geological processes which eventually results in soil formation. The resulting soil type usually has the characteristic of the parent rock i.e., the radioactivity of soil is that of the rock from which the soil has been derived. Hence, soil becomes the best representative for radioactivity in any environment. In the present study, the activity concentration of ^{238}U series, ^{232}Th series and ^{40}K has been measured from soil samples using gamma spectrometric analysis.

The soil samples for gamma spectrometry analysis have been collected from natural, undisturbed and uncultivated ground surfaces in conformity with the IAEA recommendations [11]. Each soil sample has been obtained from nine sub samples collected in an area of approximately 100 m^2 and upto a depth of 10 to 20cm. Extraneous materials like plant parts, pebbles and stones etc have been removed from the soil samples and oven-dried at 105° C for 24 hours to remove the water content from soil. The dried samples are crushed in mortar and allowed to pass through micro sieves to maintain the uniform soil grain size. The fine samples are then packed in a 250 ml polythene vessel and weighed to obtain the activity concentration of radionuclides in Bq/kg. The bottles containing processed soil samples are sealed hermetically and externally, so that the over pressure produced inside by the ^{222}Rn decay should not result in leakage of gas. These samples are kept for one month period so as to ensure secular equilibrium between ^{226}Ra and its daughter products and are subjected to gamma spectrometric analysis.

NaI(Tl) scintillation gamma spectrometer, one of the most efficient method for measuring activity concentration of radionuclides from soil samples, has been used in the present study. The spectrometer is calibrated regularly both in terms of energy response and counting efficiency using the standard sources (from Environmental Survey Laboratory, Kalpakkam, India) recommended by International Atomic Energy Agency, Vienna (IAEA). The standard sources used are (1) ^{40}K (2) ^{226}Ra for ^{238}U series and (3) ^{228}Ra for ^{232}Th series. The activity concentration of various radionuclides of interest is determined from the count spectrum of the corresponding samples. The peaks corresponding to 1.46 MeV (^{40}K), 1.764 MeV (^{214}Bi) and 2.614 MeV (^{208}Tl) are considered in arriving at the activity levels of 40K, 238U series and ^{232}Th series respectively. The concentrations of ^{232}Th, ^{238}U, and ^{40}K are estimated in Bq/kg from the counts in the respective energy region and the details are given elsewhere [12, 13]. The gamma dose rate due to primordial radionuclides present in the soil samples at one m above ground level is also calculated. The conversion factor given by UNSCEAR is used in this study and it is given below [14].

$$D = (0.623\ C_{Th} + 0.427\ C_U + 0.043\ C_K). \quad nGy.h^{-1}$$

where C_{Th}, C_U and C_K are the activity concentrations of primordial radionuclides viz., ^{232}Th series, ^{238}U series and ^{40}K existing in the soil in Bq.kg^{-1}.

RESULTS AND DISCUSSION

1. Soil Activity

The spectral measurements of activity concentration of ^{232}Th series, ^{238}U series and ^{40}K in $Bq.kg^{-1}$ in soil samples of Udagamandalam environment clearly revealed the presence of thorium in almost all the soil samples. The observed spectral photo peaks at 238.6 keV, 373.3 keV, 510.7 keV, 727.3 keV, 911.2 keV, 916 keV, 1587 keV and 2614 keV are due to ^{212}Pb, ^{228}Ac, ^{208}Tl, ^{208}Tl, ^{212}Bi, ^{228}Ac, ^{212}Bi and ^{208}Tl, which are the daughter radionuclides of ^{232}Th series. This confirms the abundance of ^{232}Th series radionuclides in soil of Udagamandalam.

The activity concentration of primordial radionuclides ^{232}Th series, ^{238}U series and ^{40}K in the soil of Udagamandalam lies in the range of 26.3 - 226.4 $Bq.kg^{-1}$, Bdl* - 87.5 $Bq.kg^{-1}$ and 95.7 - 443.9 $Bq.kg^{-1}$ with a mean of 114.6 $Bq.kg^{-1}$, 43.2 $Bq.kg^{-1}$ and 272.6 $Bq.kg^{-1}$ respectively. The minimum activity concentration (26.3 $Bq.kg^{-1}$) for ^{232}Th series has been measured at a place Thummanatti, which is situated, in eastern slope of Udagamandalam where as the maximum activity concentration (226.4 $Bq.kg^{-1}$) for the same series is observed at Nanjanad, which is a small plateau region in south western side. The minimum, ^{238}U series activity concentration in below detectable limit has been observed at Udagamandalam town area and the maximum activity (87.5 $Bq.kg^{-1}$) is observed at Bykara, which is also a plateau region situated in the western side of Udagamandalam. For ^{40}K, the minimum concentration (95.7 $Bq.kg^{-1}$) has been measured in the soil sample collected at a place Kallatti, situated in the western slope region and the maximum (443.9 $Bq.kg^{-1}$) at Kadanad, a place situated in the eastern part of Udagamandalam.

The maximum activity of the studied radionuclides has been measured in plateau or valley locations and the minimum in slope regions. This may be due to the continuous erosion and weathering of soil/rocks in the sloppy hill areas due to rain [15, 16] and successive transport through rivers and the accumulation of radionuclides in the plateau and valley regions [17]. In Udagamandalam, weathering of soil/rock is occurring because of rain through out the year and hence such results in activity concentration of radionuclides are observed. This mobility of radionuclides by erosive and weathering processes in large water flow areas in other parts of world has specifically been studied by several investigators [18, 19].

It has been observed from the data that, there exists a wide range of variation in the activities of the ^{232}Th series, ^{238}U series and ^{40}K radionuclides. This fact is attributable to the wide variety of lithological components existing in the environments under study, as observed by Baeza et al in the soil radionuclide distribution study carried out in Caeres environment of Spain [20]. The range is much broader for ^{232}Th series and ^{40}K than ^{238}U series in both the environments. This is quite understandable from the very large fluctuations in the activity of ^{232}Th series and ^{40}K from region to region, which is also attributable to the heterogeneous characteristics of soil as reported for Kalpakkam, coastal area in Tamil Nadu [21].

The arithmetic mean of ratio of activity of ^{232}Th series to ^{238}U series comes to 3:1 and the range differs from (17:1 to as low as 1:1 in Udagamandalam. A correlation has been arrived between the ^{232}Th series and ^{238}U series activities and a correlation co-efficient of 0.44 is observed. From the ratio and correlation co-efficient between ^{232}Th series and ^{238}U series activities, it can be concluded that the ^{232}Th series activity is widely varying than ^{238}U series activity and there exists no equilibrium between them in the soil samples of Udagamandalam environment. This non-equilibrium between ^{232}Th series and ^{238}U series is possibly again due to transportation of radionuclides in soils. Transportation is mainly due to the movement of ground and surface waters. Ground water movement is greater through open textured soils like sandy than through compact soils like clayey soil [22]. All the three radionuclides viz., ^{232}Th series, ^{238}U series and ^{40}K are oxidised and in the oxidised state ^{232}Th is quadrivalent and ^{238}U is divalent. This results in sharp non-equilibrium of soil borne radioactivity as ^{232}Th is strongly absorbed and ^{238}U is removed by leaching. Moreover, ^{232}Th is one of the radionuclides of rare earth's listed for their immobile character in soil [23, 24].

Thus, the gamma spectrometric analysis of activity concentration of ^{232}Th series, ^{238}U series and ^{40}K radionuclides in soil samples of Udagamandalam has showed the abundance of ^{232}Th series radionuclides in soil. The ^{232}Th series activity concentration in Udagamandalam environment is higher than the activity value of ^{232}Th series in soil proposed for Indian and world averages [25,14]. The compilation of the results of abundance of thorium in soil analysed from various environments around the world shows typical higher concentration of ^{232}Th series in soil where underlying granite and igneous type rocks has been found [26, 27].

Also, the high content of monazite in soil/granite and igneous rock leads to the higher concentration of thorium in soil samples [28,29]. It may be concluded from these results that the higher activity concentration of ^{232}Th series in soil samples anlaysed in Udagamandalam is because of the monazite occurrence in Nilgiri hills. Due to the geological processes like thermal expansion and contraction, expansion of water freezing in interstices, chemical attack and leaching, erosion by water and wind lead to the weathering and transportation of rocks and soil containing monazite placer from Nilgiris and get accumulated in the coastal areas of Kerala and Tamil Nadu [30, 31, 6]. Most of the river confluenting in Kerala and Tamil Nadu seas have their origin in Nilgiri Mountain, which supports the above statement. A similar argument has been given by Moraes that the monazite deposits in the coastal areas of Brazil are formed from the weathering of rocks, transportation through rivers and successive accumulation of monazite sand, which confirms the results discussed in the present investigation [9].

2. Dose Evaluated from Soil Activity

The gamma absorbed dose in air at a height of one metre above ground surface is estimated from the activity concentrations of gamma emitting isotopes present in the soil. The absorbed gamma dose rate due to the contribution from ^{232}Th series activity in soil varies from 17.4 to 149.9 nGy.h^{-1} with the mean value being 75.8 nGy.h^{-1}. The dose due to ^{238}U series varies from 2.7 to 37.4 nGy.h^{-1} (neglecting BDL) with a mean value of 18.5 nGy.h^{-1} and of ^{40}K from 4.1 to 19.1 nGy.h^{-1} with mean value of 11.8 nGy.h^{-1}. The computed values of gamma absorbed dose rates from all the three radionuclides in Udagamandalam environment is in the range from 27.6 to 189.1 nGy.h^{-1} with a mean of 106.1 nGy.h^{-1}. According to Green et al., the areas having a dose rate more than 40 to 50 nGy.h^{-1} can be included in the "Radiation Atlas" of the world [32]. The study area, Udagamandalam, could be included in the "Radiation Atlas" of the world since the dose rate are much greater than 40 to 50 nGy.h^{-1}.

The percentage contribution of individual radionuclides to the total dose due to ^{232}Th series, ^{238}U series and ^{40}K are 70.4%, 17.5% and 12.1%. This result indicates that ^{232}Th series activity accounted for the most of the absorbed dose in air. The reported values for relative contribution of ^{232}Th series, ^{238}U series and ^{40}K in Indian soil to the absorbed dose are 33.6%, 17.7% and 48.7% respectively, in which ^{40}K accounted for the half of the total activity for India [25]. However, the relative contribution of absorbed dose published for all over the world are 40% for ^{232}Th series, 25% for ^{238}U series and 35% for ^{40}K [33]. All the three radionuclides contribute more or less equal dose, with the ^{232}Th series being the maximum dose contributor to the total absorbed dose and ^{238}U series being the minimum contributor with ^{40}K in the mid. But, in Udagamandalam, the gamma dose rate is mainly due to ^{232}Th series i.e, 70.4%. Also, it can be seen from the gamma dose rate results of both India and all over the world that, ^{40}K accounted for significant dose that is comparable to ^{232}Th series contribution, where as in Udagamandalam, the contribution due to ^{40}K to the total absorbed dose is very low.

The evaluated values of absorbed dose in air from soil would not be the exact dose to the human beings, since man does not spent whole time outdoors continuously in a day. To approximate closely to more real dose values to human, the occupancy factor of human in indoor and outdoor and the indoor/outdoor dose rate ratio have been used [14]. The indoor and outdoor occupancy factors used are 0.8 and 0.2 respectively, and the indoor-to-outdoor ratio is 1.3, since, on an average, man spend 20% of the total time in a day at outdoor [14, 20]. Under these suppositions, the total dose rate to population (taking into account both indoor and outdoor stays) ranges from 34.2 to 234.4 $nGy.h^{-1}$, the mean value being 131.5 $nGy.h^{-1}$.

On following the recommendation published by UNSCEAR, the absorbed dose rate in air has been converted into human effective dose equivalent by using the conversion factor of 0.7 Sv Gy-1 [14]. The mean effective dose equivalent arising from the corrected mean absorbed dose rate in air for Udagamandalam is 806.3 $\mu Sv.y^{-1}$. These values are just about 80% of the 1.0 $mSv.y^{-1}$ recommended by the International Commission on Radiation Protection as the maximum permissible effective dose equivalent for members of the public [34].

3. Ambient Survey Results

Ambient dose measurement by Environmental Radiation Dosimeter has been carried out at all the soil sampling locations at one metre above the ground. The absorbed dose rate has been found to vary from 84.5 to 263.9 $nGy.h^{-1}$ with the mean value being 159.3 $nGy.h^{-1}$. The ambient dose results have indicated that the values of absorbed dose rates measured by environmental radiation dosimeter are higher in magnitude than computed from the soil activity in almost all the locations. This is due to the contribution of cosmic radiation in the study environment, since the altitude is varying from 1,500 m to 2,600 m. The cosmic ray component at sea level is 32 $nGy.h^{-1}$ and the cosmic dose doubles for every increment of 1500 m from the mean sea level [33, 35, 36]. Hence, the cosmic ray dose at 1500 metre could be 64 $nGy.h^{-1}$ and at 2,600 metre it is to be 110 $nGy.h^{-1}$.

Dodabeta, the peak of Nilgiris (altitude 2,646 metre) showed the maximum value of ambient dose of 263.9 $nGy.h^{-1}$ measured by environmental radiation dosimeter. The value of absorbed dose computed from soil activity at this place is 148.2 $nGy.h^{-1}$. On subtracting the dose computed from soil activity, from the dose measured by environmental radiation dosimeter, the cosmic dose contribution has been calculated and the value is found to be 115.7 $nGy.h^{-1}$, which is equal to the dose derived for the altitude of 2,600 metre from the literature values i.e. around 110.1 $nGy.h^{-1}$ [33, 35, 36]. The minimum absorbed dose of 84.5 $nGy.h^{-1}$ has been measured at a place Tummanatti at which the dose contribution from soil is 27.6 $nGy.h^{-1}$. The cosmic ray contribution at this place is calculated to be 56.9 $nGy.h^{-1}$. Since, the place is at an altitude of about 1,500 metre, the calculated value coincides approximately with the literature value i.e. 64 $nGy.h^{-1}$.

The cosmic radiation dose has also been measured in another way by using environmental radiation dosimeter. The detector has been carried in a lake on a boat in Udagamandalam and confirming the distance from the shore and depth of the water level are enough such that the contribution from soil is very much neglected, the dose is noted, which is wholly due to cosmic radiation. The dose level has been noted at five points and the average is taken as the mean cosmic dose and is given as 82.6 $nGy.h^{-1}$. The observation is done at an altitude of about 2,000 metres. The result is almost the same as calculated from the subtraction between environmental radiation dosimeter value and soil activity dose.

At few locations, on calculating the cosmic radiation component from dose measured by environmental radiation dosimeter and the dose computed from soil activity, the cosmic dose found to be less than sea level value of 32 $nGy.h^{-1}$. This is because, on collecting the soil samples at grass covered locations and even though the grass are removed during sample collection, the effect of shielding by the grass for gamma radiation from the soil to the air should not be avoided. At such locations, the dose measured by environmental radiation dosimeter would not totally include the gamma radiation from soil and the dose measured by environmental radiation dosimeter is mainly due to cosmic radiation at these locations. If the dose computed from soil activity (which has not been actually observed by environmental radiation dosimeter) is deducted from the environmental radiation dosimeter dose, then it leads to the negative value of cosmic dose. Hence, the cosmic dose with much lower values at some locations is an obscure result calculated from the environmental radiation dosimeter and the dose computed from soil activity.

Because of the significant contribution of cosmic radiation to the absorbed dose rate in Udagamandalam, the absorbed dose rate and human effective dose equivalent given in the previous section needs modification. Taking into account the indoor and outdoor occupancy factors, the total absorbed dose (soil+cosmic) in Udagamandalam is calculated and the values are in the range from 94.8 to 327.3 $nGy.h^{-1}$ with the mean being 197.3 $nGy.h^{-1}$. The mean effective dose equivalent arising from the absorbed dose (soil+cosmic) to the population is 1209.8 $\mu Sv.y^{-1}$.

The values exceed the 1.0 $mSv.y^{-1}$ recommended by the International Commission on Radiation Protection as the maximum permissible effective dose equivalent for members of the public (ICRP, 1991).

Hence, the important conclusion derived from the results of dose to population in Udagamandalam and Kotagiri environment is when considering the soil activity dose alone, the effective dose to population is within the limit proposed by ICRP. However, because of high altitude of both the study areas, the cosmic radiation pose additional significant dose to population, which in turn exceeds the dose limit proposed by ICRP [34]. The data obtained in this study will serve as baseline data for proper assessment of health hazard due to natural background radiation on the inhabitants of Udagamandalam as well as the whole Nilgiris.

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