Migration of radionuclide in soil and plants in the Western Ghats environment

P.K. Manigandan^{1*} and N.M. Manikandan²

 $^{\rm 1}$ Dhafir Institute, Abu Dhabi, UAE $^{\rm 2}$ Department of Physics, Bharathiar University, Coimbatore-641046, India

Background: A study on the migration characteristics of ²³⁸U, ²¹⁰Po, ²³²Th and ⁴⁰K within the soil to the wild plants in the Western Ghats were carried out. Materials and Methods: Concentrations of these radionuclides were measured in different depths of soil and from different species by employing gamma ray spectrometer and alpha counter. Results: The activity concentration of these radionuclides varied faintly within the soil but widely varied within the plant and between the species. Conclusion: The CR (Concentration ratio) showed wide variation in different species, while a few species of wild plants such as Evodia roxburghiana, Eleaocarpus oblangus and Glochidion neilgherense indicated preferential uptake of these radionuclides. Iran. J. Radiat. Res., 2008; 6 (1): 7-12

Keywords: Western Ghats, primordial radionuclides, monazite, depth profile, concentration ratio.

INTRODUCTION

The Nilgiri hill station is part of the Western Ghat of India known to have some of the significant distribution of monazite (1, 2). In order to know the radiation exposure of the population, it is very important to estimate the potential dose from the primordial and natural fallout radionuclides. The most common pathways for these radionuclides to reach humans usually involve direct ingestion or inhalation of contaminated dust particles. However these radionuclides can also be transmitted through the food chain and attendant doses are additive to those received from other pathways. In the case where ingestion or inhalation of dust is minimized as a result of humid environment or continuous soil cover, the food chain contribute to the total doses will become predominant. Thus the behavior or mobility of U and Th series is a major determinant of the plant uptake. Food chain

model in which soil/plant relationship is depicted, plants can be viewed as a hydraulic conduit for the water stored in the soil to travel upward and evaporate from the leaves. This stream of water carries radionuclides dissolved in the soil water to the roots. Obviously, the flow of water is dependent on soil moisture retention and supply. It is also a mechanism linked with the size and growth rate of a plant. This relation is further complicated by the effects of weather, growth conditions and multiple soil properties. Attempts to model all these processes in a mechanistic manner have not evolved to a broadly applicable level (3-4). Because of this, root uptake is often treated at an empirical level such as with the CR model. The CR model has been universally recommended by regulatory agencies and most radiological assessments rely on it.

MATERIALS AND METHODS

Sampling

Different species of plants were chosen to study the migration of these radionuclides to plant from uncultivated soil. Different fraction of samples like leaves and bark were collected from these plants at different locations within the forest of Long wood. Soil samples were also collected from four different places under the host trees; mixed thoroughly and about 2 kg of composite sample was collected in a polythene bag. In order to study vertical migration of

*Corresponding author:

Dr. P.K. Manigandan, Dhafir Institute, Abu Dhabi, UAE.

Fax: +97 12 6276678 E-mail: pkmgs@yahoo.com radionuclides in soil, soil samples were collected from different places at different depth. Vegetation samples were dried in an oven at 110°C and about 30g samples were taken for the wet ashing and subsequent analysis of ²¹⁰Po. The remaining samples were charred over a low flame and converted into uniform white ash using a muffle furnace at 400°C and similarly soil samples were dried in an oven at 110°C and taken for the analysis.

Gamma ray spectrometer

The primordial radionuclides activities were estimated using a γ ray spectrometer, which consisted of '3×3' NaI (Tl) detector. The soil samples were analyzed by NaI (Tl) spectrometer, which was coupled with TNI PCA II Ortec model 8K multichannel analyzer. A 3"× 3" NaI (Tl) detector was employed with adequate lead shielding which reduced the background by a factor of 95. The efficiency of various energy was arrived at using IAEA standard source and the required geometry. The system was calibrated both in terms of energy response and also for counting efficiency. The density of the sample used for the calibration was 1.3 gm/cm³ which was same as average of soil sample analyzed (1.24 gm/cm³) with the counting time of 20000 sec for each sample and a very good shielding to the detector the minimum detectable concentration was 7 Bq/kg for ²³²Th series, 8.4 Bg/kg for ²³⁸U series and 13.2 Bg/kg for ⁴⁰K at 3σ confident levels. The concentrations of various radionuclides of interest were determined using the counting spectra of each sample. The peaks corresponding to 1.46 MeV (40K), 1.76 MeV (214Bi) and 2.614 MeV (208Tl) were considered for the evaluation of the activity levels of 40K, 238U series and ²³²Th, respectively. The resolution of the crystal detector was 6% for $^{\scriptscriptstyle 40}K,\,4.4\%$ for $^{\scriptscriptstyle 232}Th$ series and 5.5% for the ²³⁸U series. The activity analysis of gamma spectra obtained for each soil sample was performed with dedicated software and the choice of reference was made so that they were sufficiently discriminated. Details of the detector and the calibration of the system were presented in the previous paper (5, 6).

To determine the concentration of ²¹⁰P.

about 30g of dried samples were taken. To start with, the samples were digested with 4N HNO₃ then with 8N HNO₃ and with a mixture of concentrated HNO₃ and H₂O. The digested samples were brought to the chloride medium by adding 0.5N HCl solution. Then ²¹⁰Po was deposited on a background count brightly polished silver disc through electro chemical exchange method ⁽⁷⁻¹¹⁾. Then it was counted in ZnS [Ag] alpha counter of background 0.2cpm and efficiency 30%. Polonium- 210 activity was estimated using the standard methods ⁽¹¹⁾.

RESULTS AND DISCUSSION

Migration of radionuclides in soil

In studying the variation of the radioactivity concentration as a function of depth, uncultivated soil samples were collected from the long wood forest. Table -1 lists the concentration of ²³⁸U, ²¹⁰Po, ²³²Th and ⁴⁰K at different depth in uncultivated soils.

It can be seen from the table, in most of the sample collected from the different location in the long wood forest, the activity concentration was found to increase in the first two profiles but afterwards the values are unpredicted (figure 1). The increase in the first two layers can be attributed to the concentration of the organic matter in the top layers (12). Schulz (13) had classified the radioactive elements occurring in the soil into two groups via immobile and mobile radioactive elements. Even though U and Th series belongs to the former category mobility of these radionuclides within soil is due the formation of organic complexes association with the colloids which can increase the mobility of the uranium but in the case of thorium which is adsorbed in the soil in a very immobile form, showed increase of activity in first two profiles is due to the high enrichment of Thorium in the soil than the Uranium.

But the fallout radionuclide ²¹⁰Po strongly adsorbed in soil will move through soil under the influence of water. Not only that the presence of earthworms turn over the top soil which leads to the distribution of fallout radionuclides into deep soil. At the same time

Table 1. Activity concentration radionuclides in different depth profile.

Locations	Different layer	Activity concentration [Bq/kg]						
		Th-232	U-238	Po-210	K-40			
L-1	0-10cm	39.17	15.12	20.12	198.79			
	10-20cm	55.37	28.45	24.33	213.10			
	20-30cm	51.36	30.27	19.23	200.11			
L-2	0-10cm	45.89	21.03	26.32	205.37			
	10-20cm	49.64	24.22	30.62	224.8			
	20-30cm	52.88	27.03	26.83	211.17			
L-3	0-10cm	47.76	19.99	24.13	202.77			
	10-20cm	55.45	29.76	29.48	210.73			
	20-30cm	48.53	24.28	20.83	229.13			
L-4	0-10cm	48.91	21.42	27.03	195.39			
	10-20cm	49.55	28.79	33.73	217.89			
	20-30cm	51.53	23.51	28.11	181.17			
	0-10cm	53.55	20.19	24.42	209.67			
L-5	10-20cm	57.63	21.01	29.44	221.22			
	20-30cm	54.42	26.95	22.36	227.28			
L-6	0-10cm	51.86	27.9	32.2	218.06			
	10-20cm	52.65	31.23	37.02	232.26			
	20-30cm	44.23	16.77	35.01	226.99			
L-7	0-10cm	46.96	18.57	22.1	201.14			
	10-20cm	49.97	19.20	26.77	233.97			
	20-30cm	48.31	20.30	20.00	186.54			
L-8	0-10cm	48.67	24.38	29.89	148.89			
	10-20cm	51.29	34.02	34.99	159.65			
	20-30cm	48.54	15.17	32.90	169.16			
L-9	0-10cm	44.41	18.56	22.81	211.19			
	10-20cm	52.07	22.12	29.66	219.78			
	20-30cm	48.72	33.68	23.07	178.88			
L-10	0-10cm	46.50	19.99	23.72	127.54			
	10-20cm	41.59	23.81	26.81	155.10			
	20-30cm	46.83	25.34	21.92	154.28			

certain portion of ²¹⁰Po in the upper layer may get washed out due to heavy rain, and deep soil keeps on accumulating without any movement. This attributes to the increase in the activity of ²¹⁰Po in the top two profiles. A good correlation is observed between U and its daughter product ²¹⁰Po in the soil with the regression of coefficient of 0.9604 (figure 2).

Expectedly, potassium belongs to the later group and shows some degree of mobility, which leads to variation of activity concentration in different layers of soil. The

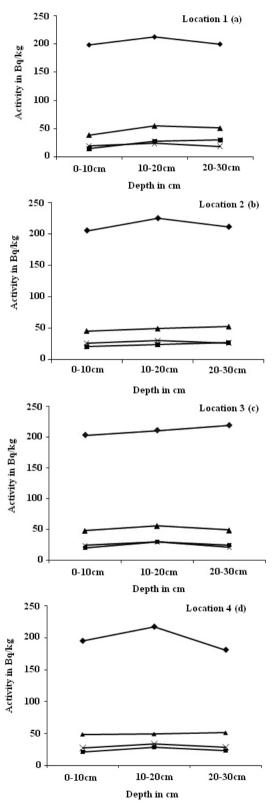


Figure 1. Variation of activity concentration of different radionuclides in the different depth of soil (a-Location 1, b-Location 2, c- Location 3, d-Location 4): Lines with the symbols ◆, ▲, ■,×, represents the activity variation of ⁴⁰K, ²³²Th, ²¹⁰Po and ²³⁸U with depth respectively.

same was observed by Karunakara et~al. 2001 and Shetty et~al. 2005 $^{(14, 15)}.$

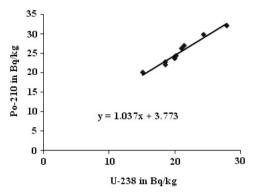


Figure 2. Relation between the activity concentration of ²³⁸U and ²¹⁰Po in soil.

Migration of radionuclide's in plants

In the soil, each radioactive element follows complex dynamics in which a part of its concentration is transported into the soil solution, while another part gradually becomes strongly bound to the particles of the soil. The portion of these radionuclides, which is in the soil solution, can be incorporated via the root into the plants. In some cases this is facilitated by their chemical similarity with other elements that the plant normally uses for its growth.

The same radionuclides were analyzed in samples of different plant species, whose results are presented in table 2. It is clear from the table that the activity concentration of ²³⁸U and ²³²Th are below detectable limit i.e. very low in most of the plants. According to CRPrinciples, plant radionuclide should concentration reflect soil concentration. However, this may be not true because of sorption on soil, which may render radionuclides less available for uptake (16). Further more, radionuclide belonging to physiologically regulated elements, or their analogues, may be selectively adsorbed, where as others may be excluded. Root uptake of radionuclides is a complex phenomenon, especially for primordial nuclides. The low activity concentration of radionuclides in plants can be observed clearly in most of the plant species except, Evodia roxburghiana, Eleaocarpus oblangus and Glochidion neilgherense. Significant difference in radioactivity concentration of these radionuclides between plant species is likely caused by physiological difference and related factors.

The activity of 210Po varies from 5.86 to 9.81Bg/kg in bark and in the younger leaves 7.88 to 12.96 Bg/kg. It is quite obvious that the concentration of ²¹⁰Po is higher in the younger leaves than bark because it is in the leaves where the requirement of nutrition is high and also the activity concentration of ²¹⁰Po is higher than the U in most of the samples. In general, the highest activity concentration in plants was found in those collected from areas with the highest radioactivity concentration in soil substrate but the activity concentration in the plants are not linearly related to the activity in soil. In all result, the small amount of activity concentration of these radionuclides may result from (a) higher mobility of ²¹⁰Po within the plant absorbed form soil in colloidal form (b) atmospheric deposition of dust on plants, from splash of soil onto plants during rainfall and from dust created during food processing. Washing plant samples may be insufficient to remove all of the extraneous contamination as some of it may have been absorbed into the plant.

Not unexpectedly, ⁴⁰K levels varied within and between plant species than others. It can be seen from the table that the activity concentration of ⁴⁰K in the bark varies from 19.53 to 171.97 Bq/kg and in younger leaves it varies from 121.26 to 219.25Bq/kg. The uptake of potassium increased as the ⁴⁰K concentration in soil increased, but not linearly, this indicates that the availability of ⁴⁰K to plant is not directly proportional to total ⁴⁰K present in soil. All the species seems to have similar requirement for ⁴⁰K, although there was significant difference in levels of potassium depending upon their metabolism.

Concentration ratio

From the results activity concentration of radionuclides in soil and plants, values of CR [CR=Activity of radionuclide in plant (Bqkg⁻¹ dry weight)/ Activity of radionuclides in soil (Bqkg⁻¹ dry weight)] has been calculated. The results are presented in the table 2 for the primordial and fallout radionuclides. CR

Table 2. Activity concentration radionuclide in the different fraction of plant and host soil.

Diame C	Type of sample	Activity Concentration [Bq/kg]			CR				
Plant Species		Th-232	U-238	Po-210	K-40	Th-232	U-238	Po-210	K-40
Evodia roxburghiana	Soil	68.12	33.53	37.71	203.9	0.230	0.324	0.292	0.801
	Bark	9.83	8.68	8.93	171.97				
	Leaves	15.69	10.87	11.02	163.32				
	Soil	49.39	23.97	28.31	213.63	BDL	BDL	0.319	0.568
Ilex wightiana	Bark	BDL	BDL	8.13	19.53				
	Leaves	BDL	BDL	9.02	121.26				
	Soil	50.47	24.35	28.54	213.93	0.203	0.350	0.332	0.926
Turpinia pomifora	Bark	BDL	BDL	7.63	146.66				
	Leaves	10.23	8.52	9.48	198.16				
	Soil	49.98	24.38	29.03	197.58	BDL	BDL	0.307	0.824
Isonandra candolleana	Bark	BDL	BDL	6.83	78.32				
	Leaves	BDL	BDL	8.92	162.85				
	Soil	55.17	22.53	26.83	219.27	BDL	BDL	0.336	0.097
Myrsine wightiana	Bark	BDL	BDL	7.13	100				
	Leaves	BDL	BDL	9.01	202.2				
	Soil	49.43	24.45	29.71	225.69	0.199	0.347	0.305	0.971
Eugenia arnottiana	Bark	BDL	BDL	8.03	77.04				
	Leaves	9.86	8.49	9.06	219.25				
	Soil	58.24	39.12	43.34	230.1	0.278	0.303	0.299	0.884
Eleaocarpus oblangus	Bark	10.88	9.32	9.81	165.25				
	Leaves	16.2	11.86	12.96	203.43				
	Soil	48.4	19.34	24.98	206.29	0.173	BDL	0.315	0.806
Cinnamomum wightii	Bark	BDL	BDL	5.86	140.98				
	Leaves	8.36	BDL	7.88	166.23				
	Soil	48.2	24	30.46	202.49				
Glochidion neilgherense	Bark	9.76	BDL	8.63	76.51	0.293	0.354	0.296	0.945
	Leaves	14.17	8.5	9.03	191.38				
Michelia nilagirica	Soil	38.59	22.93	28.46	213.45	BDL	BDL	0.332	0.680
	Bark	BDL	BDL	8.03	105.05				
	Leaves	BDL	BDL	9.46	145.1				

values for U, Th, Po and K were found to have the range of BDL to 0.354, BDL to 0.293, 0.292 to 0.336 and 0.568 to 0.971. CR value for ²¹⁰Po and ⁴⁰K is considerably higher than other radionuclides, which suggest higher

levels of uptake of these radionuclides. It is interesting to note that although all the tree species are grown in soils of similar physical chemical characteristic and similar concentration of these radionuclides, the CR

value are different for different species. This indicates that the some plant species concentrate higher 210Po and 40K radionuclides than the others, Karunakara et al. (2001) (17) observed the same. As discussed earlier, plants may take up potassium from soil as an essential element of metabolism and other radionuclides may be taken as a homologue of an essential element (16). It is interesting that the uptake of these radionuclides is relatively higher in the plant species like Evodia roxburghiana, Eleaocarpus oblangus and Glochidion neilgherense in most of the samples.

CONCLUSION

The study has provided date on primordial and natural fallout radionuclides activity in some of the predominant plant species of the Western Ghats region. The concentration of U, Th, Po and K in the soil faintly varied within the soil up to the depth of 20cm. There exists a secular equilibrium between U and Po in the soil but not in all the plants species, which had been studied. From the plants fraction analysis, the younger leaves have the highest levels of the activity of the aforementioned radionuclides than the bark. Uptake of the natural radionuclides of U and Th is low in all the plant species except few. All the plants species shows significant concentration of 40K and also 210Po but the level of ⁴⁰K is relatively higher than the ²¹⁰Po.

ACKNOWLEDGEMENT

The authors are thankful to Dr. A. Natarajan. Head, HASL, IGCAR, Dr. A.R. Lakshmanan. HASL, IGCAR, Dr. A.R. Iyengar, Head, ESL, Kalpakkam for their constant encouragement throughout the period of work.

REFERENCES

1. Mishra UC (1993) Exposure due to the high natural

- radiation and radioactive springs around the world. In proceeding of the international conference on high-level radiation areas (Ramsar) Iran-1990. IAEA publication series (IAEA), Vienna, p. 29.
- 2. Sunta CM, David M, Abani MC, Basu AS, ManiKandan MK (2000) Natural radionuclide distribution in soils of Gudalore, India. Applied Radiation and Isotopes, 52: 209-306.
- 3. Baldwin JP, Nye PH, Tinker PB (1973) Uptake of solutes by multiple root systems from soil. III-A Model for calculating the solute uptake by a randomly dispersed root system developing in a finite volume of soil. Plant and Soil, 38: 621-35.
- Barber SA, Bouldin DR., Karal DM, Hawkins SL (1984) Roots, nutrient and water influx, and Plant growth. American Society of Agronomy, Madison, WI.
- Manigandan PK, Selvasekarapandian S, Muguntha Manikandan N (2007) Distribution of Radionuclides in the Forest soils (Western Ghats -India). IJRR, 5: 17-22.
- 6. Selvasekarapandian S, Muguntha Manikandan N, Sivakumar R (2000) Natural radiation distribution of soil at Kotagiri taluk of Nilgiris biosphere in India. Eighth International Conference, Ibaraki, Japan.
- Berger KC, Erhardth WH, Francis CW (1965) 210 Po analysis of vegetables cured and uncured tobacco and associated soil. Science, 150: 1738-9.
- Abe S and Michiko A (1973) Chemical properties of ²¹⁰Po in the atmosphere: Proc Symp, IAEA-SM-181, 13: 367-73.
- Khandekar RN (1977)²¹⁰Polonium in Bombay diet. Health Physics, 33: 150-4.
- 10. Anand SJS and Rangarajan C (1990) Studies on the activity ratio of 210Po to 210Pb and their dry deposition velocities at Bombay in India. Journal Environmental Radioactivity. 11: 235-50.
- 11. Iyengar MAR, Ganapathy S, Kannan V, Rajan MP, Rajaram S (1990) Procedure Manual, Workshop on Environmental Radioactivity, Kiga, India.
- 12. Sheppard SC and Evenden WG (1988) Critical complication and review of Plant/soil concentration Ratio for uranium, thorium and lead, Journal of Environmental Radioactivity, 8: 255-285.
- 13. Suchulz RK (1965) Soil chemistry of radionuclides. Health physics, 11: 1317-1324.
- 14. Karunakara N, Avadhani DN, Somaseherappa HM, Narayana Y, Siddappa K (2001) 226Ra, 232Th and 40K concentration in soil samples of Kaiga of south-west coast of India. Health Physics, 80: 470-476.
- 15. Shetty PK, Narayana Y, Siddappa K (2006) Vertical profiles and enrichment pattern of natural radionuclides in monazite areas of coastal Kerala. Journal Environmental Radioactivity, 86: 132-142.
- 16. Sheppared SC and Evenden (1988) The assumption of linearity in soil and plant concentration ratio: an experimental evaluation. Journal Environmental Radioactivity, 7: 221-47.
- 17. Karunakara N, Avadhani DN, Somaseherappa HM, Narayana Y, Siddappa K (2003) 226Ra, 40K and 7Be activity concentrations in plant in the environment of Kiaga, India. J Environ Radioact, 65: 255-266.