Bio-indicators for Impact Assessment of Radionuclides Contamination

Prakash V[1*], Narayana Y[2]

[1] Department of PG Studies and Research in Physics, Payyanur College Kannur - 670 327, Kerala

[2] Department of Studies in Physics, Mangalore University, Mangalagangotri D.K. - 574 199, Karnataka

Email: *prakashamv@gmail.com

Abstract: *The activity of important radionuclides ^{232}Th, ^{226}Ra and ^{40}K were analysed in twelve medicinal plants collected from Mangalore and surrounding region by gamma spectrometry. The activity in the representative soil sample is also measured in order to study the transfer of radionuclides from soil to plant. In plant the average values of ^{232}Th, ^{226}Ra and ^{40}K activity were found to be 0.8 $Bqkg^{-1}$, 5.0 $Bqkg^{-1}$ and 37.1 $Bqkg^{-1}$ respectively. In soil the average values of ^{232}Th, ^{226}Ra and ^{40}K activity were found to be 54.7 $Bqkg^{-1}$, 64.2 $Bqkg^{-1}$ and 384.3 $Bqkg^{-1}$ respectively. The average values of transfer coefficient for ^{232}Th, ^{226}Ra and ^{40}K were found to be 0.02, 0.08 and 0.10 respectively. The ^{232}Th activity was below detection level for most of the plant samples, though the activity was significant in soils associated to these plants. The significant activity of ^{226}Ra in both plant and associated soil shows the higher root uptake of this radionuclide from soil. All the plants and associated soils showed significant ^{40}K activity. The plant Mamia suregia showed higher transfer coefficient for all the three radionuclides. The plant can be used as bioindicator for the future monitoring of these radionuclides. The absorbed gamma dose rates prevailing in the study area were also measured using portable scintillometer. The results of these systematic investigations are presented and discussed in this paper.*

Key Words: *Radionuclide, ^{232}Th, ^{226}Ra, ^{40}K, Soil, Scintillometer, Transfer Coefficient.*

Introduction

Mangalore city and the nearby areas of the coastal Karnataka are heading to become a region of major industrial activity centre. In view of this detailed studies on the radioactivity and trace element concentration in terrestrial, aquatic and atmospheric environs of the region has been carried out. As part of the program the activities of important radionuclides ^{232}Th, ^{226}Ra and ^{40}K in twelve medicinal plants which are commonly found in the region were analysed by gamma spectrometry. Medicinal plants are important for pharmacological research and drug development, not only when plant constituents are used directly as therapeutic agents, but also when they are used as basic materials for the synthesis of drugs.

To study the transfer of radionuclides from soil to plant, the activity in the representative soil sample was measured. The uptake of radionuclides within the soil to plant is a part of the biochemical cycling. The mobility and availability of radionuclides depend on several factors such as geochemical, biological and climatic conditions. The transfer factor, defined as the ratio of the concentration of the radionuclide in plant to the concentration of the radionuclide in soil was calculated. This factor is also known as the plant root uptake factor. From the concentration of radionuclides in soil the radium equivalent activity Ra_{eq} (single quantity which represents the specific activities of ^{232}Th, ^{226}Ra and ^{40}K) was estimated. Detailed gamma radiation survey was also been carried out in the environment using portable plastic scintillometer. The results of these systematic investigations are presented and discussed in this paper.

Materials and Methods

1. Sampling

In the present study, 12 ayurvedic medicinal plants and associated soils collected from Moodabidri, near Mangalore, were analyzed for the concentration of natural radionuclides ^{232}Th, ^{226}Ra and ^{40}K. About 1 kg of soil sample was collected from the plant sampling site and brought to the laboratory. All the samples were carefully processed following standard procedure (EML procedure manual, 1983). The soil samples were oven dried, ground, sieved and filled in air tight plastic containers and stored for about one month to ensure secular equilibrium between ^{226}Ra and its short lived daughters. The containers were sealed carefully to avoid the escape of gaseous ^{222}Rn and ^{220}Rn (Narayana et al., 2001). These samples were subjected to gamma spectrometry analysis.

Approximately 5 kg of each plant (leaf, stem, root and fruits if any) were collected in a big polythene bag, brought to the laboratory, washed with running water to free from pollutants and cut into pieces. The samples were then ashed at 500^0C in a muffle furnace. The ashed vegetation samples were filled in air tight plastic vials and stored for about one month to ensure secular equilibrium between ^{226}Ra and its short lived daughters. These samples were subjected to gamma spectrometric analysis.

2. Experimental Set-up

Gamma spectra from the samples are recorded using a high efficiency 5" x 5" NaI (Tl) detector coupled to a 4K Multi-channel analyzer. The gamma ray spectrometer was calibrated using different standards and the efficiency of the detector was determined experimentally (Narayana et al., 2001). The standards for uranium and thorium were prepared using known weight of analyzed ore samples that are in radioactive equilibrium and mixing them with silica powder simulating the sample matrix. ^{40}K standard was prepared by taking oven dried analar potassium chloride. The spectrum obtained from NaI (Tl) is more complex to analyze. The Compton continuum of higher energy peak contributes to the count rate observed in the lower energy peak. Therefore Compton contribution from the higher energy peak to the lower energy peak should be evaluated to determine the activity. In the present work simultaneous equation method (Abani, 1994) was employed for the analysis of the spectrum and to determine the concentration of various radionuclides. The activity of ^{232}Th was evaluated from 2614 keV gamma line of ^{208}Tl, the activity of ^{226}Ra from 1764 keV gamma line of ^{214}Bi and that of ^{40}K from its 1461 keV photopeak.

From the concentrations of naturally occurring radionuclides in the soil, the total absorbed gamma dose rates in air were estimated. The conversion factors (dose coefficients) used to compute the total dose rate in air per unit specific activity concentration in soil (1 Bq kg^{-1}) were 0.623 nGy h^{-1} for ^{232}Th, 0.461 nGy h^{-1} for ^{226}Ra and 0.0414 nGy h^{-1} for ^{40}K (UNSCEAR, 2000). Inorder to compare the specific activities of materials containing different concentrations of radium, thorium and potassium, the radium equivalent activity concentration index Ra_{eq} was calculated (Beretka and Mathew, 1985). The radium equivalent activity is the single quantity which represents the specific activities of ^{232}Th, ^{226}Ra and ^{40}K. The equation used to find out the radium equivalent activity is based on the estimate that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} of ^{232}Th or 13 Bq kg^{-1} of ^{40}K generate the same gamma ray dose rate.

The Minimum Detectable Activity (Narayana et al, 2001) at 95% confidence level for 36,000 seconds of counting time and 275g sample weight was found to be 0.4 Bq kg^{-1} for ^{232}Th, 1.8 Bq kg^{-1} for ^{226}Ra and 5.4 Bq kg^{-1} for ^{40}K. Each sample weighing about 275g were counted for sufficiently long time (36,000 seconds) to reduce the counting error. The activity reported corresponds to dry weight of the sample.

Results and Discussion

The results of activity of ^{232}Th, ^{226}Ra and ^{40}K in plant and associated soil samples are given in Table 1. From the table it is clear that the level of activity of these radionuclides in plant is much lower when compared to soil. In plant the activity of ^{232}Th varies in the range BDL – 3.3 Bq kg^{-1} with an average value of 0.8 Bq kg^{-1}, the activity of ^{226}Ra varies in the range 3.3 - 7.8 Bq kg^{-1} with an average value of 5.0 Bq kg^{-1} and that of ^{40}K varies in the range 26.1 - 58.3 Bq kg^{-1} with an average value of 37.1 Bq kg^{-1}. In soil the activity of ^{232}Th varies in the range 41.6 – 68.9 Bq kg^{-1} with an average value of 54.7 Bq kg^{-1}, the activity of ^{226}Ra varies in the range 58.2 - 72.5 Bq kg^{-1} with an average value of 64.2 Bq kg^{-1} and that of ^{40}K varies in the range 335.3 – 448.8 Bq kg^{-1} with an average value of 384.3 Bq kg^{-1}.

The ^{232}Th activity was below detection level in most of the plant samples analyzed, though the activity was significant in soils associated to these plants. This shows that the uptake of this radionuclide by plant is negligible even though the soil on which the plant grows has significant amount of ^{232}Th activity. The variation of activity of ^{226}Ra shows near uniform trend in almost all plant samples. The observed ^{226}Ra activity was higher compared to ^{232}Th activity in all plants. The significant activity of ^{226}Ra in both plant and associated soil shows the higher root uptake of this radionuclide from soil. It is clear from the table that all the plants and associated soils show significant ^{40}K activity. The higher activity of ^{40}K in plants may be due to the continuous accumulation of ^{40}K through root uptake over a period of time. It is known that as an essential element of metabolism, plants take up potassium from soil in varied amounts depending upon the metabolism.

The activities of ^{232}Th, ^{226}Ra and ^{40}K in soil samples are compared with the reported values of other environs in Table 2. The average values obtained for ^{232}Th (54.7 Bq kg^{-1}) is higher in comparison with the population weighted world average value of 45 Bq kg^{-1}, the average value obtained for ^{226}Ra (64.2 Bq kg^{-1}) is approximately double the population weighted world average of 32 Bq kg^{-1}. The higher ^{232}Th and ^{226}Ra activity observed in the soil samples may be traced to the laterite type of soil prevailing in the region (Narayana et al, 2001). The average activity of ^{40}K (384.3 Bq kg^{-1}) in the present study is less in comparison with the population weighted world average of 420 Bq kg^{-1} (UNSCEAR, 2000).

From the activity of naturally occurring radionuclides the radium equivalent activity was calculated. The results of radium equivalent activity are given in Table 3. The radium equivalent activity varies in the range 143.5 Bq kg^{-1} to 189.1 Bq kg^{-1} with an average value of 172.1Bq kg^{-1}. The average radium equivalent activity in the soil samples lies much below the recommended limit of 370 Bq kg^{-1} (UNSCEAR, 2000).

Table 3 also gives the results of gamma absorbed dose rate measured at 1m above the ground level using portable plastic scintillometer. The absorbed gamma dose rate measured in the study area varies 34.8 nGy h^{-1} to 52.2 nGy h^{-1} with an average value of 43.5 nGy h^{-1}. The radiation level follows almost a uniform pattern with dose rates close to the mean value except in some sampling stations. The present value lies between the dose rate ranges of 18 - 93 nGy h^{-1} given for the world (UNSCEAR, 2000). The measured dose rates were less in comparison with the all India average value of 88.5 nGy h^{-1} projected by Nambi et al. (1987). The absorbed dose rates in air due to naturally occurring radionuclides in coastal Karnataka are related to the local geology of the region (Narayana et al., 2001).

The soil-to-plant transfer coefficients (Table 3) vary in the range 0.03 - 0.06, 0.05 - 0.13 and 0.06 - 0.15 for ^{232}Th, ^{226}Ra and ^{40}K, respectively with corresponding average values of 0.02, 0.08 and 0.10. It is clear from table that, the transfer coefficient for ^{40}K is significantly higher compared to ^{232}Th and ^{226}Ra, suggesting higher levels of uptake of ^{40}K. Similar findings were reported by Avadhani (2002) for Goa environs. According to ANL (1993), the transfer coefficient values for Th, Ra and K are 0.001, 0.04 and 0.3 respectively.

The soil to plant or soil to vegetation transfer factor, for a given type of plant and for a given radionuclide can vary considerably from site to site with season and time after contamination. These variations depend on such factors as the physical and chemical properties of the soil, environmental conditions and chemical form of the radionuclide in the soil. Furthermore, soil management practices such as ploughing, liming, fertilizing and irrigation can also effect the uptake of radionuclide by the vegetation. It is interesting to note that the plant *Mamia suregia* showed higher transfer coefficient for all the three radionuclides though the soil associated with *Mamia suregia* showed low activity when compared to the soil activity associated with other plants. This indicates that the uptake of radionuclides by the plant is moderately high compared to other plants. Therefore the plant can be used as bioindicator for the future monitoring of these radionuclides contamination and for impact assessment.

Table 1. The Activity of ^{232}Th, ^{226}Ra and ^{40}K in Plants and Associated Soils

Sample ID	Name	Activity in plant (Bq kg^{-1})			Activity in soil (Bq kg^{-1})		
		^{232}Th	^{226}Ra	^{40}K	^{232}Th	^{226}Ra	^{40}K
MP1	*Putranjeeva roxburghii*	2.6 ± 0.6	5.3 ± 1.1	37.8 ± 2.9	58 ± 0.7	62.2 ± 1.2	358.2 ± 3.1
MP2	*Mamia suregia*	2.1 ± 0.9	7.8 ± 1.6	50.4 ± 4.1	41.6 ± 0.7	58.2 ± 1.2	335.3 ± 3.1
MP3	*Mesua nagassarium*	BDL	4.8 ± 1.1	35.7 ± 2.9	68.9 ± 0.8	61.4 ± 1.2	358.8 ± 3.1
MP4	*Saraca indica*	BDL	3.3 ± 0.7	26.4 ± 2.1	52.2 ± 0.7	58.3 ± 1.2	369.1 ± 3.2
MP5	*Syzygium jambolanum*	BDL	4.8 ± 1.1	39.5 ± 3.1	55.8 ± 0.8	63.7 ± 1.3	366.0 ± 3.2
MP6	*Garcinia indica*	BDL	3.7 ± 0.8	27.3 ± 2.2	48 ± 0.7	61.2 ± 1.3	356.7 ± 3.2
MP7	*Ficus benghalensis*	BDL	4.6 ± 1.1	41.2 ± 3.1	62.4 ± 0.8	64.8 ± 1.3	362.6 ± 3.2
MP8	*Flacartia montana*	BDL	3.7 ± 0.9	31.0 ± 2.4	59.9 ± 0.8	58.3 ± 1.2	361.5 ± 3.2
MP9	*Nyctanthes arbor-tristis*	BDL	7.6 ± 1.7	58.3 ± 4.7	48 ± 0.9	72.5 ± 1.5	431.0 ± 3.8
MP10	*Morinda citrifolia*	BDL	3.3 ± 0.7	26.1 ± 2.0	47.2 ± 0.9	70.5 ± 1.5	428.6 ± 3.8
MP11	*Ficus recemosa*	1.9 ± 0.7	5.4 ± 1.1	39.5 ± 3.1	60.4 ± 0.9	69.3 ± 1.5	435.3 ± 3.8
MP12	*Barringtonia acutangula*	3.3 ± 0.6	5.4 ± 1.0	32.3 ± 2.6	54.4 ± 0.9	70.3 ± 1.5	448.8 ± 3.8
Mean		**0.8**	**5.0**	**37.1**	**54.7**	**64.2**	**384.3**
Median		**2.4**	**4.8**	**36.7**	**55.1**	**63.0**	**364.3**
Range		**BDL-3.3**	**3.3-7.8**	**26.1-58.3**	**41.6-68.9**	**58.2-72.5**	**335.3- 448.8**

BDL-Below Detection Limit

Table 2. Comparison of ^{232}Th, ^{226}Ra and ^{40}K Activity in Soil with Other Environs.

Activity (Bq kg^{-1})			Region	Reference
^{232}Th	^{226}Ra	^{40}K		
41.6 - 68.9 (54.7)	58.2 - 72.5 (64.2)	335.3 - 448.8 (384.3)	Moodubidiri, Coastal Karnataka	Present work
14 - 160 (64)	7 - 81(29)	38 - 760 (400)	India	UNSCEAR (2000)
10 - 200(60)	3 - 60 (26)	40 - 800 (350)	Irland	McAulay& Moran (1988)
18 - 135 (50)	35 -228 (90)	281 - 711 (524)	Chaina	Ziqiang et al., (1988)
29.8	20.1- 62.3	61 - 316.7	Karnataka	Narayana et al., (2001)
45	32	420	Popln. Wtd. world avg.	UNSCEAR (2000)

Table 3. Radium Equivalent Activity, Absorbed Dose and Transfer Coefficient

Sample ID	Ra_{eq} (Bq kg^{-1})	Dose rate (nGy h^{-1})	Transfer Coefficient		
			^{232}Th	**^{226}Ra**	**^{40}K**
MP1	172.7	34.8	0.04	0.09	0.11
MP2	143.5	43.5	0.05	0.13	0.15
MP3	187.5	52.2	-	0.08	0.10
MP4	161.3	34.8	-	0.06	0.07
MP5	171.7	43.5	-	0.07	0.11
MP6	157.3	43.5	-	0.06	0.08
MP7	182.0	34.8	-	0.07	0.11
MP8	171.8	52.2	-	0.06	0.09
MP9	174.3	52.2	-	0.11	0.14
MP10	171.0	34.8	-	0.05	0.06
MP11	189.1	52.2	0.03	0.08	0.09
MP12	182.6	43.5	0.06	0.08	0.07
Mean	**172.1**	**43.5**	**0.02**	**0.08**	**0.10**
Median	**172.3**	**43.5**	**0.05**	**0.08**	**0.10**
Range	**143.5 – 189.1**	**34.8 – 52.2**	**0.03 - 0.06**	**0.05 - 0.13**	**0.06 - 0.15**

Conclusion

The ^{232}Th uptake by plant is negligible when compared to the uptake of ^{226}Ra and ^{40}K by medicinal plants. The average activity concentrations for ^{232}Th and ^{226}Ra in soil in the environment of Moodabidri are higher than the population weighted world average value. The radium equivalent activity in the soil samples lie below the recommended limit. The radiation level follows almost a uniform pattern in the study area. The transfer coefficient for ^{40}K is significantly higher compared to ^{232}Th and ^{226}Ra. The transfer coefficient for a given plant and for a given radionuclide can vary considerably from site to site with season and time after contamination. The plant *Mamia suregia* can be used as bioindicator for the future monitoring of radionuclide contamination and for impact assessment.

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