

## Analysis of the $^{40}\text{K}$ Levels in Soil using Gamma Spectrometry

José Araújo dos Santos Júnior<sup>1\*</sup>, Jorge João Ricardo Ferreira Cardoso<sup>1,2</sup>, Cleomacio Miguel da Silva<sup>1</sup>, Suêlto Vita Silveira<sup>1</sup> and Romilton dos Santos Amaral<sup>1</sup>

<sup>1</sup>Grupo de Estudos em Radioproteção e Radioecologia (GERAR); Department of Nuclear Energy; Federal University of Pernambuco; Av. Prof. Luiz Freire, 1000; Cidade Universitária; 50740-540; jaraujo@ufpe.br; Recife - PE - Brasil.

<sup>2</sup>Department of Geology; Federal University of Pernambuco; Av. Acadêmico Hélio Ramos, s/n; Cidade Universitária; 50740-530; jcardoso@ufpe.br; Recife - PE - Brasil

### ABSTRACT

Potassium-40 was determined in soil in an area with 40,000 m<sup>2</sup> situated in the western State of Pernambuco, Brazil. For radiometric measurements, the gamma spectrometry method with a high purity germanium (HPGe) detector was used. Sampling of 78 soil samples has been performed at intervals of 25 m. The specific activities of  $^{40}\text{K}$  were calculated based on the photopeak of 1.46 MeV. Values from 541 to 3,572 Bq kg<sup>-1</sup> were obtained (mean of 1,827 Bq kg<sup>-1</sup>). These values allowed the determination of the elemental concentrations as well as the absorbed dose rates in air, 1 m above the ground. The values varied from 1.7 to 11.5% (mean of 6%) and from 23.4 to 154.3 nGy h<sup>-1</sup> (mean of 79 nGy h<sup>-1</sup>), respectively.

**Key words:**  $^{40}\text{K}$  in soil;  $^{40}\text{K}$  gamma spectrometry;  $^{40}\text{K}$  in environmental radiological protection.

### INTRODUCTION

The Brazilian Northeast region, despite its interesting geological structure (Malanca et al., 1993), has not been sufficiently explored in terms of environmental radiological studies. Since 1974, the Brazilian Nuclear Enterprise (NUCLEBRAS) performed geological and radiometric mapping studies in that region, where a small anomalous area of uranium was discovered in the Pernambuco sylvan. The first radiometric-geological studies occurred after an accord between NUCLEBRAS and the Federal University of Pernambuco (UFPE) in 1975 (Magalhães, 1977).

Natural environment radionuclides are responsible for the most of the external exposures of gamma radiation. This contribution is mainly due to the

radionuclides of the natural series  $^{238}\text{U}$ ,  $^{235}\text{U}$ , and  $^{232}\text{Th}$ , followed by  $^{40}\text{K}$ , universally present in the Earth (Shenber, 1997; Tzortzis; Tsertos, 2004). The world mean specific concentration of  $^{40}\text{K}$  (activity per unit soil mass) is 370 Bq kg<sup>-1</sup>, varying from 100 to 700 Bqkg<sup>-1</sup> (Mcaulay; Moran, 1988). Radioactivity levels of the environment depend on geological aspects, mainly on rocks and soil, where they are found in varying concentrations (Tzortzis; Tsertos, 2004).

The average absorbed dose rate in air at a 1 m distance from the  $^{40}\text{K}$  source is 15.54 nGy h<sup>-1</sup> (Selvasekarapandian et al., 2000). The United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR (1993), establish that, the world mean dose from natural radiation sources of normal areas is estimated to be 2.4

\* Author for correspondence

mSv.a<sup>-1</sup>, while that for all man-made sources, including medical exposure, is about 0.8 mSv.a<sup>-1</sup> (Ghiassi-Nejad et al., 2001). Thus, 75% of the radiation dose received by humanity is come from natural radiation sources. Based on these radiation levels, one can certify that the knowledge of primordial radionuclides, such <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K and <sup>87</sup>Rb, is an important pre-requisite for evaluation of the rate of exposure and the absorbed dose by the population (Kannan et al., 2002). This knowledge ensures radiological control, since a predominant part of the environmental radiation is found in the upper soil layer (Kannan et al., 2002). Potassium is an essential element of human metabolism and can be found in all living cells, mainly in the muscular tissue. Natural potassium is composed of three isotopes: <sup>39</sup>K, <sup>40</sup>K and <sup>41</sup>K, of which only <sup>40</sup>K is radioactive. This radionuclide occurs at the approximate abundance of 0.012% in relation to the whole mass of potassium (Pires, 1995). <sup>40</sup>K has a half-life of 1.28 x 10<sup>9</sup> years and is a beta and gamma emitter (89% and 11% of its radiation, respectively) with respective energies of 1.3 MeV and 1.46 MeV (Peixoto et al., 1995; Erdtmann; Soyka, 1979). The relevance of the potassium-40 study is mainly due to its long half-life, its importance for living organisms with a uniform distribution, and because it is responsible for 98% of the gamma emission of the primordial radionuclides present in the earth. The present work is important because there is no reference regarding the area concerning the <sup>40</sup>K concentrations in the soil, which were found to be high.

## MATERIALS AND METHODS

### *Sampling and preparation*

The soil samples were collected in an area of approximately 40,000 m<sup>2</sup>, located in the Sylvan region of the state of Pernambuco, Brazil (Fig.1). The sampling was carried out at 25 m intervals, and the samples were collected from the C horizon (average depth of 35 cm), mainly due to the kind of climate. In the laboratory, the samples were dried, sieved at a granulometry lower than 63 µm and homogenized. Each sample of 200 grams was stored in standardized polyethylene containers.

### *Measure of the radioactivity*

The activities of the <sup>40</sup>K in the soil were determined by gamma spectrometry HPGe of 4.54 cm of diameter, active volume of 41.1 cm<sup>3</sup>, and efficiency of 27%, coupled to a Canberra MCA (multi-channel analyzer) with 8,192 channels. This system presented a resolution of 1.77 keV in 1,332 keV for <sup>60</sup>Co. To reduce the background effect, the detector was enveloped by nearly 6.5 cm lead covering. The measurement time for each sample was standardized at 43,200 seconds (12 hours). The counting efficiency of <sup>40</sup>K in the energy of approximately 1.46 MeV was determined with <sup>152</sup>Eu, multi-gamma emitting, with energies varying from 39 keV to 2 MeV, certified by the Institute for Radioprotection and Dosimetry (IRD), Rio de Janeiro, Brazil. Based on the calculations of the counting efficiency of the <sup>152</sup>Eu standard, it was possible to calculate the specific activities of the potassium 40, with the energy 1.46 MeV, based on equation 1 (Tzortzis et al., 2003):

$$A = \frac{C}{\varepsilon \times \gamma \times m \times t} \quad (1)$$

where A is the specific activity of the radionuclide (Bq kg<sup>-1</sup>); C is the liquid count; ε the counting efficiency; γ the percentage of gamma emission probability of the radionuclide under consideration; t the counting time (s) and m the mass of the sample (kg).

Using the above equation, it was possible to calculate the specific activities of the <sup>40</sup>K in the study area through the energy of 1.46 MeV. For the detection limit calculation (LD), equation 2 was used, where the factor 4.66 corresponded to one level of confidence of 95% (Silva; Mazzilli, 2005).

$$LD = \frac{4.66 \sqrt{C}}{\varepsilon \times \gamma \times m \times t} \quad (2)$$

The parameters of equation 2 for the calculation of the LD were applied to the measurement of the background in the photopeak of 1.46 MeV of the <sup>40</sup>K.

### *Calculation of elemental concentrations of <sup>40</sup>K*

Through the measurement of the activities of potassium 40, calculated from its gamma emission in the photopeak of 1.46 MeV (equation 1), it was

possible to calculate the elemental concentrations reported in units of  $\mu\text{g.g}^{-1}$ , using equation 3 (Tzortzis; Tsertos, 2004).

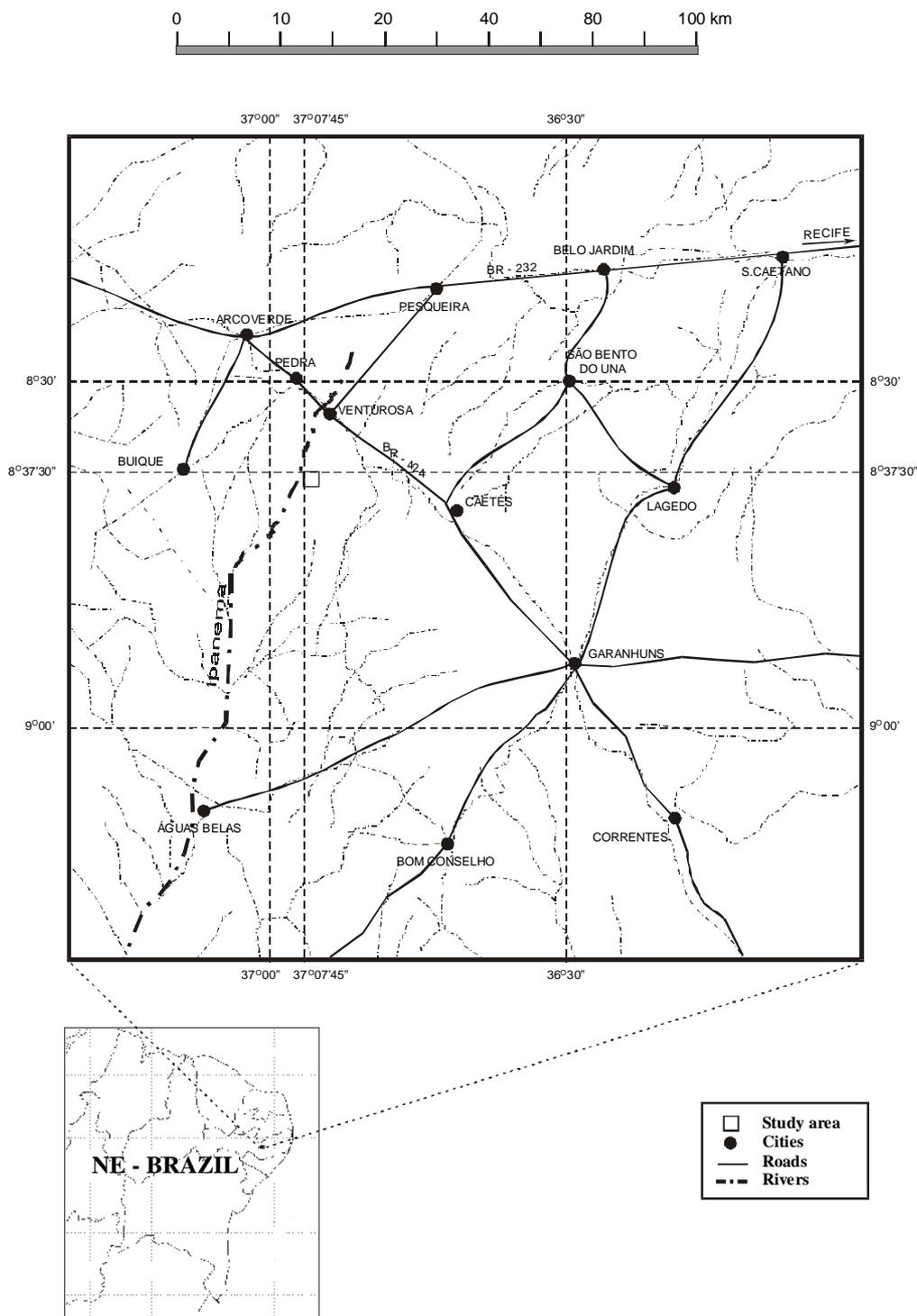


Figure 1 - Localization of the study area access routes.

$$F_E = \frac{M_E \times C}{\lambda_E \times N_A \times f_{A,E}} \times A_E \quad (3)$$

where  $F_E$  is the fraction of element E in the sample,  $M_E$ ,  $\lambda_E$ ,  $f_{A,E}$  and  $A_E$  are the atomic mass ( $\text{kg mol}^{-1}$ ), the radioactivity decay constant ( $\text{s}^{-1}$ ), the fraction atomic abundance in nature and the measured activity concentration ( $\text{Bq kg}^{-1}$ ), respectively, of the corresponding radionuclide considered ( $^{40}\text{K}$ ),  $N_A$  is Avogadro's number ( $6.023 \times 10^{23}$  atoms  $\text{mol}^{-1}$ ), and  $C$  is a constant with a value of 100 for K. These elemental concentrations are presented in percentages for potassium.

#### Calculation of the absorbed dose in air

Equation 4, published in UNSCEAR (1988), was used to calculate the absorbed dose rate (D) in outdoor air at one meter above ground, proceeding from the gamma emissions of  $^{40}\text{K}$  (Shenber, 1997; Selvasekarapandian et al., 2000; Kannan et al., 2002):

$$D = (4.32 \times 10^{-3} C_K) \times 10^{-8} \text{ Gy h}^{-1} \quad (4)$$

where  $C_K$  is the specific activity of  $^{40}\text{K}$  ( $\text{Bq kg}^{-1}$ ) and  $10^{-8}$  is the conversion factor from the absorbed dose (D).

## RESULTS AND DISCUSSION

The Figs. 2, 3 and 4 show the frequency distributions based on the calculations of the specific activities, elemental concentrations and absorbed dose rates in air 1 m above the ground surface, respectively, for the  $^{40}\text{K}$  in the soil of the Sylan region of the state of Pernambuco, Brazil. The non-parametric test of Kolmogorov-Smirnov at a reliability level of 95%, showed that the best distribution of these data for all the tests is the log-normal one, with asymmetry on the right.

The map of the activity distribution of the  $^{40}\text{K}$  in the study area is shown in Fig. 5. The highest values obtained for the  $^{40}\text{K}$  activities were determined in NE portion of the area, probably a result of the presence of granite and, consequently, higher elemental concentrations and absorbed doses.

The specific activities of  $^{40}\text{K}$  in soil varied from 541 to 3,572  $\text{Bq kg}^{-1}$  (mean 1,827  $\text{Bq kg}^{-1}$ ), with a detection limit of approximately 62  $\text{Bq kg}^{-1}$ .

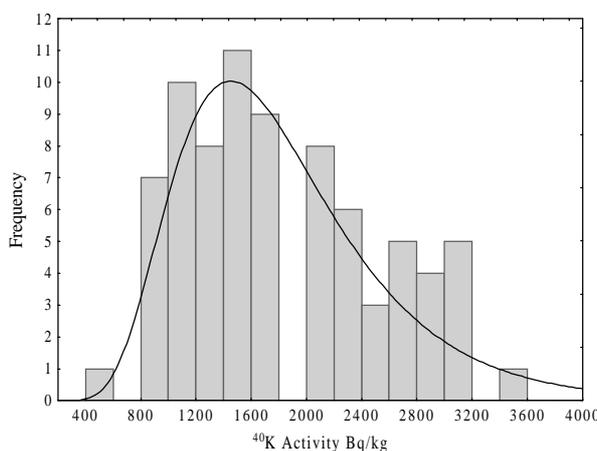
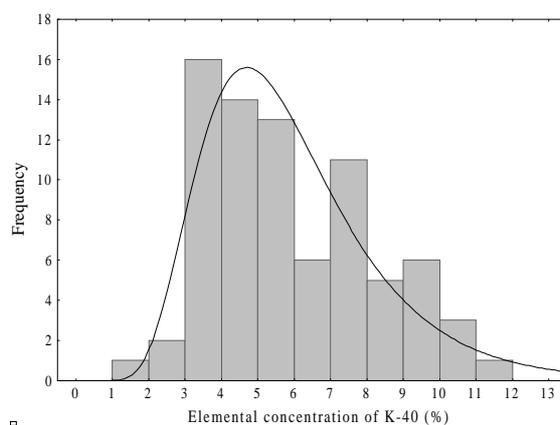
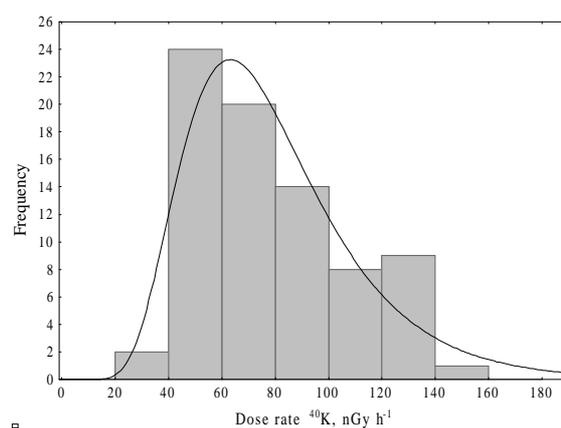


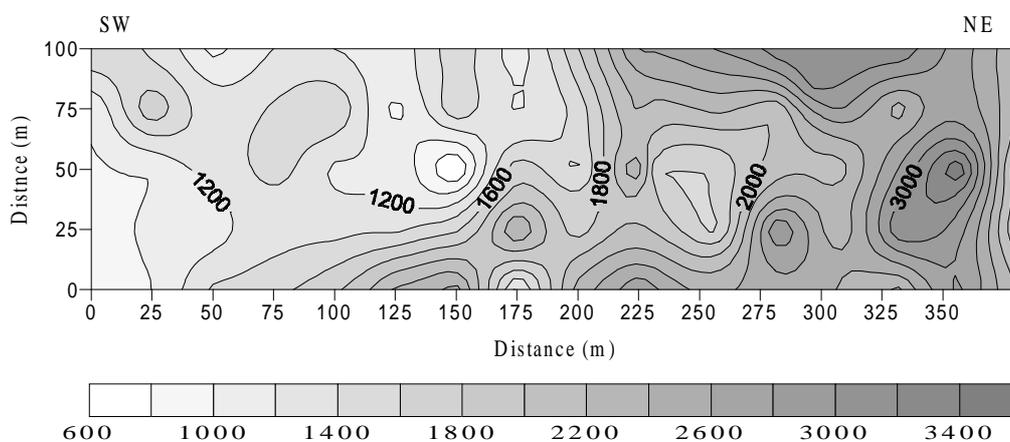
Figure 2 - Frequency distributions of  $^{40}\text{K}$  ( $\text{Bq kg}^{-1}$ ) in soil.



**Figure 3** - Frequency distributions of  $^{40}\text{K}$  (%) in soil.



**Figure 4** - Frequency distributions of  $^{40}\text{K}$  (nGy h<sup>-1</sup>) in soil.



**Figure 5** - Map of the activity distributions of  $^{40}\text{K}$  (Bq kg<sup>-1</sup>) in the study area.

Table 1 shows the statistical parameters obtained for  $^{40}\text{K}$  in soil of the study area.

The elementary concentrations of  $^{40}\text{K}$  varied from 1.7% to 11.5%, with a mean of approximately 6%. The worldwide average for the elemental concentration of  $^{40}\text{K}$  in soil is 1.3%. The UNSCEAR 2000 report (Tzortzis; Tsertos, 2004), presents  $f_c$  varying from 1.3 to 9, approximately ( $f_c$  mean about 5). The absorbed doses in air 1 m above the surface were calculated using equation 4 UNSCEAR report (Shenber, 1997; Selvasekarapandian et al., 2000; Kannan et al., 2002), where values varying from 23.4 to 154.3  $\text{nGy h}^{-1}$ , with a mean of 79  $\text{nGy h}^{-1}$ , were obtained.

The elemental concentration of the  $^{40}\text{K}$  in soil samples from work conducted in various

countries (Tzortzis; Tsertos, 2004) is presented in Table 2.

When the values for the concentrations of  $^{40}\text{K}$  in the present study are compared with those obtained in other regions of the world, it can be seen that the ratio of the variations of the present study to those of Italy, Spain, India, Jordan and Cyprus are 0.9-4.6; 8.5-2.2; 8.5-23; 17-11.5; >1.7-6, respectively. Thus, only the minimum concentration of Italy (1.9) is higher than the minimum concentration in the present study (1.7). All the other elemental concentrations are lower.

The specific activities and absorbed doses in the air one meter above the ground for  $^{40}\text{K}$  obtained from the study, and the work conducted in other regions, according to Tzortzis and Tsertos (2004) can be seen in Table 3.

**Table 1** - Statistical data of  $^{40}\text{K}$  in soil samples from the area of study.

Statistical	A ( $\text{Bq kg}^{-1}$ )	[K] (%)	D ( $\text{nGy h}^{-1}$ )
Range	541-3,572	1.7-11.5	23.4-54.3
Arithmetic mean	1,827	5.9	78.9
Geometric mean	1,696	5.5	73.3
Median	1,662	5.3	71.8
Mode	1,250	4.0	54.0
Mean error (%)		4.2	
Skewness		0.481	
Kurtosis		-0.714	
Frequency		Log-normal	

[K] = elemental concentration of  $^{40}\text{K}$ ; D = rate absorbed dose.

**Table 2** - Elemental concentrations of  $^{40}\text{K}$  in soil samples from the present study and studies conducted in other regions (Tzortzis; Tsertos, 2004).

Region	Range $^{40}\text{K}$ (%)
Pernambuco - Brazil	1.7 - 11.5*
World average	1.3
Italy	1.9 - 2.5
Espanha	0.2 - 5.2
Rajasthan, India	0.2 - 0.5
Russaifa, Jordan	0.1 - 1.0
Cyprus	< 1 - 1.9

\*Present study

**Table 3** - Specific activities and absorbed doses in air at one meter above the ground for  $^{40}\text{K}$  in different localities of the world.

Location	$^{40}\text{K}$		Reference
	Activity ( $\text{Bq kg}^{-1}$ )	D ( $\text{nGy h}^{-1}$ )	
PE-Brazil	1,827 (541-3,572)	79 (23.4-154.3)	present study
World *	370 (100-700)	15.54	Mcaulay; Moran, 1988
RN-Brazil	677.8 (56.4 -1,972)	29.3 (2.4-85.2)	Malanca et al., 1993
Tripoli	270 (265-282)	12 (11.4-12.5)	Shenber, 1997
Gudalore-India	195.2 (77.5-595.9)	73 (27.5-203.9)	Selvasekarapandian et al., 2000
Kalpakkam-India	406 (200-854)	17.4 (9-37)	Kannan et al., 2002

\* Worldwide average; PE = Pernambuco; RN = Rio Grande do Norte.

## CONCLUSIONS

The results show that gamma spectrometry is an important tool for environmental monitoring of  $^{40}\text{K}$ . By this method, the distribution the elemental concentration, as well as the absorbed dose in air, one meter above the ground surface, could be determined. The measured activities in the studied area are five fold higher than the world mean, and, consequently, the elemental concentrations and absorbed dose are five fold higher than the world average. The highest concentrations were found in the NE region of the area.

## ACKNOWLEDGEMENTS

The authors thank the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq) for the scholarship; to Professor Suêlto Vita da Silveira (through FADE - Fundação de Apoio ao Desenvolvimento da Universidade Federal de Pernambuco) for the financial support; and Professor Carlos Adolpho M. Baltar for making the laboratory under his management available to us (Laboratory of Mineral Technology of the Federal University of Pernambuco).

## RESUMO

A radioatividade do  $^{40}\text{K}$  foi determinada em solo em uma área de aproximadamente 40.000 m<sup>2</sup> localizada no Estado de Pernambuco, Brasil. Para as medidas radiométricas foi usado o método de espectrometria gama com detector de germânio de alta pureza (HPGe). A amostragem perfaz um total de setenta e oito amostras de solo coletadas em intervalos de 25 m. As atividades específicas do  $^{40}\text{K}$  foram calculadas com base no fotopico gama

de 1,46 MeV. Foram obtidos valores que variaram de 541 a 3.572  $\text{Bq kg}^{-1}$  (média aritmética de 1.827  $\text{Bq kg}^{-1}$ ). Com base nestes cálculos, obteve-se concentrações elementares e doses absorvidas no ar a 1 m, derivadas da contribuição do  $^{40}\text{K}$  no solo, onde os valores obtidos variaram respectivamente de 1,7 a 11,5% (média 6%) e 23,4 a 154,3  $\text{nGy h}^{-1}$  (média 79  $\text{nGy h}^{-1}$ ).

## REFERENCES

- Erdtmann, G. and Soyka, W. (1979), *The gamma of the radionuclides: tables for applied gamma ray spectrometry*. New York : Verlag Chemie.
- Ghiassi-Nejad, M.; Beitollahi, M. M.; Fallahian, N.; Amidi, J. and Ramezani, H. (2001), Concentrations of natural radionuclides in imported mineral substances. *Environment International*, **26**, 557-560.
- Kannan, V.; Rajan, M. P.; Iyengar, M. A. R. and Ramesh, R. (2002), Distribution of natural and antropogenia radionuclides in soil and beach sand samples of Kalpakkam (India) using hyper pure germanium (HPGe) gamma ray spectrometry. *Applied Radiation and Isotopes*, **57**, 109-119.
- Magalhães, C. A. P. (1977), *Projeto Venturosa: estudos geológicos e radiométricos na folha venturosa - área I, sub-área F. Monografia (Graduação)*, Universidade Federal de Pernambuco, Recife, Brazil.
- Malanca, A.; Pessina, V. and Dallara, G. (1993), Assesment of the natural radioactivity in the Brazilian State of Rio Grande do Norte. *Health Physics*, **65**, 298-302.
- Mcaulay, I. R. and Moran, D. (1988), Natural radioactivity in soil in the republic of Ireland, *Radiation Protection Dosimetry*, **24**, 47-49.
- Peixoto C. M.; Auler, L. M. L. A. and Fonseca, R. C. M. (1995), Interferência do  $\text{K}^{40}$  na atividade beta total em amostras ambientais. In: Encontro de Física de Reatores e Termo-hidráulica, 10.; Encontro de Aplicações Nucleares, 3., Águas de Lindóia. *Proceedings... Águas de Lindóia*.

- Pires A. C. B. (1995), Identificação geofísica de áreas de alteração hidrotermal, Crixás-Guarinos, Goiás. *Revista Brasileira de Geociências*, **25**, 61-68.
- Selvasekarapandian, S.; Sivakumar, R.; Manikandan, N. M.; Meenakshisundaram, V.; Raghunath, V. M. and Gajendran, V. (2000), Natural radionuclide distribution in soils of Gudalore, India. *Applied Radiation and Isotopes*, **52**, 299-306.
- Shenber, M. A. (1997), Measurement of natural radioactivity levels in soil in Tripoli. *Applied Radiation and Isotopes*, **48**, 147-148.
- Silva, P. S. C. and Mazzilli, B. P. (2005), U and Th series nuclides in Cubatão River sediments. In: International Conference on Nuclear Analytical Methods in the Life Sciences, 8., Abstract, Rio de Janeiro *Proceedings...* Rio de Janeiro, Brazil.
- Tzortzis, M. and Tsertos, H. (2004), Determination of thorium, uranium and potassium elemental concentrations in surface soils in Cyprus. *Journal of Environmental Radioactivity*, **27**, 325-338.
- Tzortzis, M.; Tsertos, H.; Christofides, S. and Christodoulides, G. (2003), Gamma-ray measurements of naturally occurring radioactive samples from Cyprus characteristic rocks. *Radiation Measurements*, **37**, 221-229.

Received: July 05, 2005;

Revised: July 14, 2005;

Accepted: August 01, 2005.