DISTRIBUTION OF ^{238}U, ^{232}Th, ^{40}K, AND ^{137}Cs CONCENTRATIONS IN SOIL SAMPLES NEARBY A NUCLEAR LABORATORY, CAPAO ISLAND, BRAZIL

by

Luciano S. R. OLIVEIRA [1], ***Celio J. V. OLIVEIRA*** [1], ***Bruno M. R. CARVALHO*** [1], ***Paulo A. M. CABRAL*** [1], ***Helio C. VITAL*** [1, 2], ***and Edson R. ANDRADE*** [1, 2, 3 *]

[1] Biological and Nuclear Defence Division, Chemical, Brazilian Army Technological Center, Rio de Janeiro, Brazil
[2] Nuclear Engineering Graduate Program, Military Institute of Engineering, Rio de Janeiro, Brazil
[3] Institute of Radioprotection and Dosimetry, Rio de Janeiro, Brazil

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Absolute soil concentrations of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs samples were measured using high-resolution gamma spectrometry. The area of interest encompasses an embankment in a mangrove swamp in Guaratiba, Rio de Janeiro, called *Capao Island*, where nuclear, chemical and biological defense laboratories of the Brazilian Army Technology Center are in operation for more than 30 years. In order to ensure that no significant environmental impact has resulted from neutron physics experiments performed in a graphite exponential pile in addition to the operation of two cesium-driven irradiating facilities, radiation monitoring of the isotopes was carried out. A total of eight 250 ml soil samples were extracted within an area of 300 m 300 m. No trace of ^{137}Cs was detected and the measured levels of ^{238}U were found to be close to the global mean. However, some data that slightly exceeded the expected normal range for ^{232}Th (60 % of samples) and ^{40}K (20 % of samples) should be attributed to the construction debris (cement, rocks, and sand) used in the embankment at the site. Since there is no handling of those isotopes at that site or adjacent facilities that could affect their presence, it was concluded that no detectable contamination has occurred.

Key words: gamma-ray spectroscopy, soil, ^{238}U, ^{232}Th, ^{40}K, ^{137}Cs

INTRODUCTION

The Chemical, Biological and Nuclear Division of the Technological Center of the Brazilian Army (CTEx) is located on the Capao Island, within an exceptionally well-preserved mangrove region of Guaratiba in Rio de Janeiro. The area is regarded as a part of an important natural reserve due to its unique characteristics and large number of species it encompasses. Moreover, the installation is considered an operational unity since it includes a subcritical nuclear assembly (a graphite exponential pile) and two research gamma irradiation facilities.

Therefore, due to its nuclear and radioactive characteristics, procedures aimed at ensuring the radiological safety of its rich environment have been implemented in the form of pre- and post-operational monitoring activities that include the assessment of soil concentrations of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs throughout the Capao Island via high-resolution gamma-ray spectroscopy analyses. Rather than providing highly accurate data, the measurements were intended to be added to the recorded history of the radiological characterization of its soil.

The technique allows both qualitative and quantitative analysis of gamma-emitting elements, providing simultaneous identification of multiple radionuclides [1]. The advantages of using high-purity germanium (HPGe) detectors for gamma spectroscopy include high-energy resolution, linear response over a wide energy range and a large amount of output information from a single analysis [2].

Gamma-ray spectroscopy measurements of specific soil activities of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs at the CTEx have been carried out in order to ensure that no significant radioactive contamination has occurred over the years due to research activities developed at its Nuclear Defence Section where a natural-uranium subcritical assembly (a graphite exponential pile) and a high-activity ^{137}Cs cavity-type irradiating facility are routinely operated.

The measurements performed in this work were focused on the radiological characterization of the embankment where buildings of sectors A, B, C, and D of

* Corresponding author; e-mail: fisica.dna@gmail.com

the CBRN Defence Division of CTEx are located. Radioisotopes such as ^{238}U, ^{232}Th, ^{40}K are found in nature, while any significant amount of ^{137}Cs could be a sign of contamination from irradiation facilities [3-5]. The sectors are designated by a letter, according to their azimuthal (clockwise) position with respect to the geographical center of the institute, sector A being due south. The Transportation sector would thus correspond to the B sector, but it is an exception, since in practice it has not been designated by a letter.

MATERIAL AND METHODS

The soil samples were collected, treated and remained stored in isolation for 30 days in order to reach secular equilibrium [6]. The 250 ml sealed cylindrical recipients containing the samples were wrapped in aluminum packages.

Calibration and energy efficiency curves were generated by using CANBERRA Genie 2000 spectroscopy software developed for the analysis of soil samples inside cylindrical containers of standard dimensions, namely 5 cm in height and of a 4 cm radius. Standard point sources were used for energy calibration. In addition, efficiency calibration was done by employing acid-solution sources provided by the National Radiation Metrology Laboratory of the Radioprotection and Dosimetry Institute. They accurately displayed well- known compositions and activities, encompassing several radionuclides and exhibiting the same cylindrical geometry as the 250 ml soil samples.

Listed in tab. 1 are the values of the gamma sources used in the calibration process and their associated energies. The correlation coefficient (r^2) for the energy calibration curve was 0.998. In addition, the extrapolation for detection efficiency at ^{40}K was estimated to have an uncertainty lower than 2 %, based on error propagation.

Soil analysis included the determination of concentrations of isotopes belonging to the natural series such as ^{238}U, ^{232}Th, and ^{40}K, in addition to ^{137}Cs. Peak energies and emission yields used in the calculation of absolute concentrations of ^{238}U and ^{232}Th, by assuming the secular equilibrium and considering the associated decay schemes, are listed in tab. 2 [7], along with those for ^{40}K. A few other possible decay channels that could lead to larger uncertainties in the estimates of concentrations have not been included. The decays not included were those pertaining to very low energies (susceptible to strong attenuation and large background correction factors) and those with very small yields (and, consequently, poorer statistics) [8].

Table 1. Data used for determination of the efficiency calibration curve of the system

Radionuclide	Energy [keV]	Fitted efficiency	Measured efficiency	Uncertainty [%]	Error [%]
^{210}Pb	46.54	0.05489	0.05489	4.06	1.49
^{241}Am	59.54	0.06029	0.06163	4.05	2.17
^{109}Cd	88.03	0.06030	0.06230	6.01	3.21
^{57}Co	122.06	0.05363	0.05271	2.59	1.76
	136.47	0.05048	0.05076	2.55	0.54
^{137}Cs	661.66	0.01154	0.01154	0.28	0.01
^{60}Co	1173.23	0.00648	0.00648	0.11	0.07
	1332.49	0.00578	0.00578	0.09	0.01

Table 2. Peak energies and decay yields for the ^{238}U and ^{232}Th series, ^{40}K and ^{137}Cs

^{238}U			^{232}Th		
Radionuclide	Energy [keV]	Yield [%]	Radionuclide	Energy [keV]	Yield [%]
^{214}Bi	609.31	45.49	^{228}Ac	338.320 911.196 968.960	11.40 26.2 15.9
			^{212}Pb	238.63	43.60
^{214}Pb	351.93	35.60	^{212}Bi	727.330	6.65
^{40}K	1460.81	10.55	^{137}Cs	661.66	84.99

Samples were collected in duplicate at the following sectors of the CTEx CBRN Divisions: D, A, C, and Transportation Sector (23°01'49.0"S, 43°34'49.6"W).

Eight soil samples from Capao Island were collected, prepared and analyzed as environmental samples at the CTEx Radiological Agents Identification Laboratory (LIAR). The constructed area, shown in fig. 1, spans some 400 m 400 m. The retrieved soil samples were stored at room temperature (on shelves) for 30 days, so as to allow for secular equilibrium. HPGe gamma spectrometry was then performed by counting times of the order of 10^5 seconds or longer so that high-resolution energy spectra of the samples could be determined with appropriate statistics. GENIE 2000 software was used in spectral analysis. The correction for the background spectrum was also performed so that the absolute specific activities of the radionuclides of interest, namely ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs, were determined by accounting for the net overall energy-dependent counting efficiency of the system.

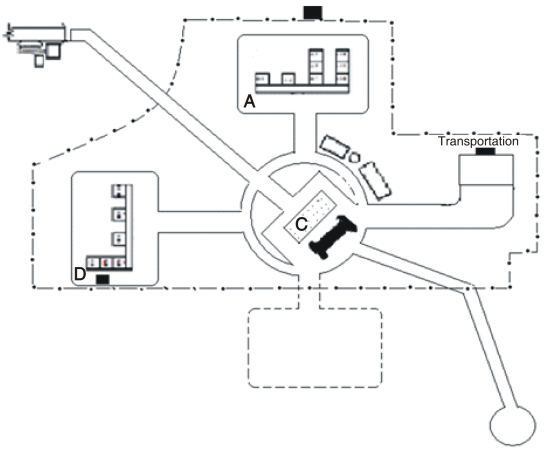

Figure 1. Locations of soil sample collection on Capao Island, CTEx

By using the GENIE2000 program, calibration curves were adjusted to the data points as expressed in eq. 1, for efficiency.

The radioactive sources and their associated peak energies [keV] used for the determination of the energy calibration curve were: (a) ^{241}Am (26.34, 59.54); (b) ^{155}Eu (86.55, 105.31); (c) ^{152}Eu (121.78, 244.70, 344.28, 778.90); (d) ^{137}Cs (661.65), and (e) ^{60}Co (1173.22, 1332.49). The data in tab. 2 have been used for determination of the fitted efficiency calibration curve that was used for the calculation of absolute soil concentrations of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs.

In order to calculate the activities (A) from the number of gamma rays detected by the HPGe spectroscopy system, eq. 1 was used, where $\varepsilon(E)$, S, and T are the efficiency, net number of counts, and counting time, respectively; k is decay correction factor that accounts for the mass loss of the radioisotope of interest due to its radioactive decay from the time the sample was collected until it was counted and i is the emission fraction of the isotope at the energy of interest [4]. The use of samples having the same geometry as that of the calibration sources eliminated the need for corrections, due to variations in the solid angle.

$$A \quad \frac{S}{T.i.\varepsilon(E)k} \tag{1}$$

RESULTS

Table 3 lists the data measured for the specific activities of ^{232}Th and ^{238}U in sectors A, C, D and Transportation. Errors listed in all tables are those statistically estimated at a 68 % (1σ) confidence level.

Two soil samples were collected and analyzed for each of the four areas surveyed (Sectors D, A, C, and Transportation Sector), as discriminated in the tables. The analyses performed in this work showed no significant amount of ^{137}Cs in any of the soil samples collected as no photopeak whatsoever was formed at the energy of ^{137}Cs after 10 days of counting. For comparison purposes, the reported average soil concentrations of naturally occurring radionuclides are presented in tab. 4 for Argentina, the USA, and the world [9].

Soil concentration data measured in this work are listed in tab. 5 as ratios to the world average figures for naturally occurring isotopes.

Table 3. Measured specific activities of ^{40}K ^{232}Th, and ^{238}U in Sectors A, C, D, and Transportation

Radionuclide	Sector average activity [Bqkg^{-1}]							
	A		C		D		Transportation	
^{232}Th (^{228}Ac, ^{212}Pb, ^{212}Bi)	141.2	0.9	341	10	198	12	9.2	0.4
^{238}U(^{214}Pb, ^{214}Bi, ^{234}Th)	32.0	2.2	56.0	5.5	34.2	2.9	5.1	0.7
^{40}K	772	8	147	2	1007	14	70 ± 5	

Table 4. Reported soil activities of naturally occurring radionuclides

Region	Soil activity of radionuclide [Bqkg^{-1}]					
	^{40}K		^{238}U		^{232}Th	
	Average	Range	Average	Range	Average	Range
Argentina	650	540-750				
USA	370	100-700	35	4-140	35	4-130
World mean	400	140-850	35	16-110	30	11-64

Table 5. Ratio of measured soil concentrations to the world average figures for naturally occurring radionuclides

Sector	Relative soil concentration (world mean = 1)					
	^{232}Th		^{238}U		^{40}K	
D	6.6	0.4	0.87	0.06	2.52	0.04
A	4.7	0.1	0.84	0.02	1.93	0.02
C	11.4	0.3	1.37	0.03	0.37	0.01
Transportation	0.31	0.01	0.12	0.01	0.18	0.01

DISCUSSIONS

Regarding ^{40}K, its concentrations were found to be close to the world average in sectors A and C, moderately above the upper limit of that range in sector D and about half of the world`s lower limit in the Transportation sector.

By inspection of tab. 5, it can be concluded that the ^{238}U concentrations determined in this work approached the world average across the embankments that lay the foundation for sectors D, A, and C, becoming an order of magnitude lower in the Transportation sector.

In addition, it can be easily gathered that the lowest concentrations of natural radionuclides are consistently found in the Transportation sector. That is to be expected considering that its mangrove soil has been kept unchanged and, for the most part, remains in equilibrium with its surrounding swamp environment.

In contrast, sectors D, A, and C have received great quantities of embankment consisting of concrete fragments, rocks, clay and other soil components in order to elevate them above the surrounding mangrove swamp level. As a consequence, in those sectors, the measured soil concentrations for ^{232}Th are significantly higher than the world average. It has been found that the measured concentrations of environmental radionuclides in sectors D and A do not differ significantly, in spite of the different materials handled and processed in them, which is an indicator of no significant environmental impact from such activities. Another finding was that the data measured for sector C apparently exhibits a different trend. A possible explanation for that could be the difference in the composition of materials used to form its older embankment whose origin was different from the one used to build sectors D and A.

CONCLUSIONS

Absolute gamma spectroscopy measurements have been performed, providing specific soil activities for ^{238}U, ^{232}Th, and ^{40}K across the Capao Island, CTEx, Guaratiba, some 50 km west of downtown Rio de Janeiro. Previous concentration data for those radionuclides in Brazil are not available in literature and that fact highlights the importance of the present data set.

Significant differences in soil concentrations among the different areas or sectors of the site have been attributed to different compositions of the embankments used to build them. Therefore, data from sectors D and A, both located on the same embankment, have been found to exhibit roughly the same trends, although no nuclear or radiological activities are performed in sector A, in contrast to sector D. In addition, their data differ considerably from those of sector C, situated on an older embankment of a different composition.

In addition, concentrations of one order of magnitude below the world average have been consistently found for all radionuclides surveyed in soil samples from the nearby Transportation sector, where the mangrove soils from the original composition still remain mostly intact and construction debris has not been dumped as part of the embankments.

Regarding ^{238}U, a good match of the experimental data with the world average concentrations has been found for the sectors on the embankments. In contrast, the higher ^{232}Th concentrations found probably result from the large concentrations of cement and rocks present in the embankments built in sectors D, A, and C. Finally, no sign of contamination by ^{137}Cs has been found.

AUTHOR CONTRIBUTIONS

Measurements and experimental set up were carried out by L. S. R. Oliveira, C. J. V. Oliveira, and B. M. R. Carvalho under supervision and guide lines of H. C. Vital and E. R. Andrade. All authors discussed the result. Manuscript was written by H. C. Vital and E. R. Andrade and reviewed by P. A. M. Cabral. Figures were prepared by L. S. R. Oliveira and C. J. V. Oliveira.

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Лусиано С. Р. ОЛИВЕИРА, Селио Ј. В. ОЛИВЕИРА, Бруно М. Р. КАРВАЉО, Пауло А. М. КАБРАЛ, Елио С. ВИТАЛ, Едсон Р. АНДРАДЕ

ДИСТРИБУЦИЈА КОНЦЕНТРАЦИЈА ^{238}U, ^{232}Th, ^{40}K И ^{137}Cs У УЗОРЦИМА ЗЕМЉИШТА У БЛИЗИНИ НУКЛЕАРНЕ ЛАБОРАТОРИЈЕ НА ОСТРВУ КАПАО У БРАЗИЛУ

Применом гама спектрометра високе резолуције измерене су апсолутне концентрације ^{238}U, ^{232}Th, ^{40}K и ^{137}Cs у земљишту. Област истраживања обухвата насип у мочвари мангрова у Гуаратиби, Рио де Женеиро, под називом острво Капао, где су у употреби преко 30 година лабораторије за нуклеарну, хемијску и биолошку одбрану Бразилског војнотехничког центра. Спроведен је мониторинг изотопа како би се осигурало да експерименти из области нуклеарне физике, обављени на графитној експоненцијалној инсталацији и у два радијациона постројења са цизијумским изворима, немају значајног утицаја на животну средину. Укупно је прикупљено осам узорака од по 250 ml земљишта на површини од 300 m 300 m. Није пронађен траг ^{137}Cs, а вредности уранијума су упоредиве са глобалним средњим вредностима. Међутим, неке податке који прелазе очекиване вредности за ^{232}Th (60 % узорака) и ^{40}K (20 % узорака) треба приписати грађевинском материјалу (цемент, камење, песак) коришћеном приликом изградње насипа. Пошто нема употребе ових изотопа у овој области нити у суседним постројењима, који могу утицати на њихову појаву, закључак је да није утврђена контаминација земљишта.

Кључне речи: спектроскопија гама зрачења, ^{238}U, ^{232}Th, ^{40}K, ^{137}Cs