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Determination of the Specific Activityof Soil and Fertilizers in Sergipe - Brazil

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***Abstract*—**Measurements of radioactivity in the environment is of great importance to monitor and control the levels of radiation to which man is exposed directly or indirectly. It is necessary to show that regardless of working or being close to nuclear power plants, people are daily in contact with some amount of radiation from the actual environment and food that are ingested, contradicting the view of most of them. The aim of this study was to analyze the rate of natural and artificial radiation from radionuclides present in cement, soil and fertilizers used in Sergipe State – Brazil. The radionuclide activitiesmeasured all samples arebelow the Brazilian limit of the exclusion and exemption criteria from the requirement of radiation protection.It was detected Be-7 in organic fertilizers that means a short interval between the brewing processes for use in agriculture. It was also detected an unexpected Cs-137 in some samples; however its activities does not represent risk for the population. Th-231 was also found in samples of soil and cement in the state of Sergipe that is an unprecedented result.

***Keywords*—**Cs-137, Be-7, Th-231radiation dose, radioisotopes

I. Introduction

EVERY form of life on Earth is exposed to natural radiation field. The man's exposure to natural radiation originates mainly from domestic sources to the human body (incorporated radionuclides) and external sources of geological origin (presence of U, Th and K in the soil) and cosmic (cosmogenic radionuclides, neutron, gamma).

Since the Chernobyl accident, reinforced by the recent accident in Fukushima, the fear of the population is concentrated on artificial radiation sources, particularly nuclear facilities. However, most of the average collective exposure of the world population is due to natural causes [1].

Before the 1960s the natural radiation was seen as one whose control by man was unnecessary because it was considered almost constant and equal to 1 mSv/year. But according toSantos[2], concerns related to exposures resulting from natural radiation emerged in the 1970s, becoming the object of intense research, resulting in the publication of several standards and guidelines by the regulatory organizations worldwide. In recent decades has been a growing interest in determining the levels of radiation to which the population of a given region is exposed. In this context, exposure to environmental radioactivity is one of the basic objectives of any environmental monitoring program.

It is known that the environment there is an average rate of radiation around 2.4 mSv/year [3], and yet, mankind survives today. Therefore it is concluded that it is very likely that there is a threshold of radiation dose below which there is no permanent damage to the human body, due to the body's ability to recover and heal. However, we can say scientifically that any dose level of radiation is harmful to human body.

The radiation that causes very high doses can destroy the cells of organisms; induce cancer or genetic defects by altering the characteristics of the cells. However, when the doses are low, there is no fully known relationship between effect and dose. The question of the biological effect caused by low dose levels is quite complex. In these cases, there are several competing factors besides radiation to cause harm to human body, such as cancer.There is still no sufficient statistical data to a conclusion. Therefore, measuring radioactivity in the environment is of great importance to monitor and control the levels of radiation to which man is exposed directly or indirectly and to correlate the dose received to the effects caused by it.

The crust is composed of natural radioactive materials, including uranium and thorium and their radioactive daughters, which are in balance in nature and constitute, along with the other primordial radionuclides and cosmogenic radionuclides, the radiation background. These radionuclides are dispersed to greater or lesser concentrations throughout the environment, but only those with half-life comparable to the age of the earth exist in significant concentrations. Irradiation of the human body by these natural sources is mainly due to radionuclides series of U-238 and Th-232; the isotope K-40;being gamma radiation the most important component of exposure to external radiation.

Yet, scientists must show that regardless of working or being close to nuclear plants, people are in daily contact with a small amount of radiation from the actual environment and food that are ingested, contradicting the view of most of them.

In the world, there are only few data on rates of natural environmental exposure range to which the population is subjected. In general, those regionshave the highest natural radioactivity levels in the gamma rate exposure. Higher

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environmental gamma levels are due to the presence of minerals of uranium and thorium in higher concentrations in the soil.

In Brazil, the areas with the highest index are located in the coastal region of the country, where are the principal deposits of heavy concentrates, such as monazite, ilmenite, zircon and others and also within the territory of which occurred alkaline intrusions and pegmatite uranium and associated thorium in high levels [4,5,6,7,8,9].

An extensive radiometric survey for characterizing areas of high environmental radioactivity was made in Brazil [5,6,10,4,11,12].Worldwide, the hot spots are located in Ramsar in Iran and Mallat[13,14,15], Yangjiang in China [16,17], Badgastein in Austria [18], several areas in the United States and Canada[19].

Part of environmental radiation is also coming from artificial radioactive elements found today throughout the globe due to fallout, which is the precipitation or deposition of radioactive elements on the surface after a nuclear explosion. The fallout that living beings are exposed today comes from nuclear weapons tests that took place between 1952 and 1963, when it was signed a treaty banning nuclear tests on the planet surface.

The aim of this study was to analyze the rate of natural and artificial radiation in soil, cement and fertilizersin Sergipe State, northeastern Brazil by gamma spectrometry and contribute with new data about region with low radiation levels. This location was chosen also because in the near future the Brazilian government intends installing a new nuclear plant close to thatregion[20,21].

II. Materials and Methods

A. Local

This study was performed with samples from two separate locations in Sergipe State, situated in northeastern Brazil(Fig. 1). Some soil samples were collected at the campus of Federal University ofSergipe (UFS), located at São Cristóvão, a city close to Aracaju, the main city in this State. Other samples were collected approximately 90 km away, in Neópolis Plateau, a region that produces many fruits and vegetables consumed by the population.

Neópolis is located beside the São Francisco River, near to the area where the Brazilian Government intends to build a Nuclear Power Plant. The climate in the region is tropical with rainy and dry summer rainfall around 1200 mm per year, with rainfall concentrated in the months April to September.

B. Samples

Two kilograms of each sample were collected since October 2008 until February 2010. Soil samples were collected from 16 different sites;one sample of cement (the most used in Sergipe's buildings);and11 different samples of fertilizers. The samples were divided into two groups: 1) soil and cement, 2) fertilizers.

Fig 1.Geographic Location Neopolis Plateau (Platô de Neópolis) and UFS(orange circles) in Sergipe State-Brazil[22].

B1. Samples of soil and cement

Five soil samples were collected at five different points within the campus of the Federal University of Sergipe (UFS): near to the Department of Physics (point 1);the Department of Chemistry (point 2); the Rectory (point 3); the Library (point4); and the Soccer field (point 5).

11 soil sampleswere collected randomlyfrom 11 different points of Neópolis Plateau, near to the plantations of fruits and vegetables. Each soil sample received the name correlated to the vegetable (or fruit) that where near collected.

One sample of nonproductive soil was also collected.

The cement sample chosen was from the brand most used in the construction industry in the state of Sergipe.

B2. Samples of fertilizers

Fertilizers can change the radionuclides concentration in the soil. So, the most used fertilizers in the study area were chosen to be analyzed in this work. 11 different samples of chemical and natural fertilizers were collected in Neópolis Plateau direct from the fruit and vegetable producers.

C. Gamma spectrometry

All these samples were placed in plastic bags previously identified and taken to the Environmental Monitoring Laboratory (AML) Eletronuclear, in Angra dos Reis,city in Rio de Janeiro State. In AML samples were sieved with a 16-mesh sieve, and then transferred to a Marinelliplastic beakerof

one liter (Figure 2).The assembly was weighed to determine the mass. The beaker was sealed with silicone glue and identified to be analyzed by gamma spectrometry.

The preparation process was performed similarly for all samples, which were previously mixed and placed in beakers carefully not to change their densities with possible compacting.

Measures of activities of the samples were performed using an experimental setup consisting of an intrinsic germanium detector HP(Ge) with a relative efficiency greater than 40% of a 4096 channel multichannel analyzer, nuclear electronics standard gamma spectrometry and shielding of lead, aluminum and acrylic. The spectra of background and samples were taken for a period of 60,000 seconds for each measurement, and then found the average result of the measures and their variances.

It must be emphasized that procedures to thegamma spectrometry were performed keeping all conditions unchanged.It was possible to identify the radionuclides present in every sample analyzed, with their specific activities.

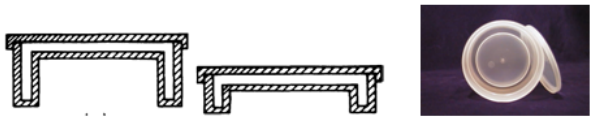

Fig 2 Geometry of Marinelli plastic beaker used in gamma spectrometry

III. RESULTS AND DISCUSSION

A.Samples of soil and cement

Soil samples collected at the Campus of the UFS are numbered from 1 to 5 and those collected in Plateau Neópolis were named according to the fruit or vegetable where they were collected. Of the 17 samples analyzed, 15 have only three radionuclides from natural radioactive series and potassium, as shown in Tables 1 and 2.

It is observed that the uranium and thorium concentrations are highly variable. However, they can be considered normal soil (1-8 ppm), i.e., they are not the potentially uraniferous soil, which can reach up to 50 ppm.Boylecites that the average content of uranium in normal soils is 1 ppm[23]. In his results, the uranium in soil presented activities ranging from less than 24.1 Bq/kg to 192.6 Bq/kg with an average of 24.1 Bq/kg.The uranium content in soil depends primarily on the material that gave rise to them, with higher values those developed in granitic rocks, gneisses and alkaline igneous rocks, while the lowest are found in basic igneous rocks and carbonate rocks.Based on the results presented in Table 1, the major activity detected for U-235 in soil was (2.19 ± 0.29) Bq/kg, which represents only 9.02% of the lowest activity recorded in the literature.

Boyle (1982) also reportedthe concentration of Th-232 in soils ranging from 0.41 Bq/kg to 203.5 Bq/kg, with a low average at about 20.35 Bq/kg[23]. As for uranium, the content of this radionuclide in soils depends mainly on the type of source material that originates. The highest concentration of Th-232 is derived from granitic rocks, alkaline igneous rocks, schists and gneisses, and lowest in basic igneous rocks and carbonate rocks.Th-232 is one of the most abundant radioisotopes in nature; however, it was not detected in the samples here studied.

TABLE I
NATURAL AND ARTIFICIAL RADIONUCLIDES DETECTED IN SAMPLES OF SOIL AND CEMENT

	Specific Activity (Bq/kg) for the radionuclides				
Samples	Ac-228	U-235	Th-231	Ra-226	Cs-137
Point 1	0.24± 0.02	0.11± 0.01			
Point 2	6.10± 0.22	1.33 ±0.01			
Point 3	0.15± 0.27		2.39± 0.13		0.18± 0.07
Point 4	7.02± 0.33	0.95± 0.13			
Point 5	6.57 ± 0.31	0.89± 0.12			
Cocon. (A)	9.31± 0.35	1.47± 0.12			
Cocon. (B)	7.94± 0.55	1.11± 0.19		18.2± 0.3	
Cocon. (C)	8.76± 0.64	1.47± 0.26		24.2± 0.4	
Mango	6.31± 0.39	1.6± 0.15			
Lemon	12.09± 0.02	1.82± 0.16			
Pas. fruit	13.49± 0.46				
Tomato	15.51± 0.67	2.19± 0.28			
Manioc	8.54± 0.57				
Cassava	17.57± 0.47				
Unproduct.	10.08± 0.48	1.35± 0.14			
Garden	15.33± 0.35	1.73± 0.16			0.45± 0.11
Cement	14.34± 0.39	3.46± 0.22	7.64± 0.76		

TABLE II
NATURAL AND ARTIFICIAL RADIONUCLIDES DETECTED IN SAMPLES OF SOIL AND CEMENT

	Specific Activity (Bq/kg) for the radionuclides				
Samples	K- 40	Bi-212	Bi-214	Pb-212	Pb-214
Point 1	3.33± 0.32	1.48± 0.04	2.02± 0.01	0.11± 0.07	0.17± 0.08
Point 2	0.11± 0.84	3.70± 0.51	5.61± 0.20	6.14± 0.30	5.65± 0.14
Point 3	0.33± 0.15	8.32± 0.67	0.13± 0.28	0.12± 0.28	0.13± 0.26
Point 4	0.22± 0.16	2.92± 0.78	7.83± 0.29	6.46± 0.34	7.53± 0.23
Point 5	0.22± 0.16	4.59± 0.91	6.82± 0.28	6.61± 0.50	7.01± 0.24
Cocon. (A)	30.06± 0.18	4.70± 0.10	6.82± 0.28	8.31± 0.24	6.97± 0.26
Cocon. (B)	36.42± 0.25		7.62± 0.37	10.97± 0.51	7.32± 0.34
Cocon. (C)	112.58± 0.44		8.98± 0.47	12.78± 0.55	8.68± 0.41
Mango	15.13± 0.15		5.68± 0.28	5.27± 0.32	5.98± 0.29
Lemon	21.13± 0.19	5.73± 0.04	2.02± 0.01	9.10± 0.49	7.50± 0.40
Pas. fruit	8.74± 0.12	8.74± 0.12	10.54± 0.46	1.14± 0.44	11.71± 0.39
Tomato	44.27± 0.28	11.66± 0.14	10.92± 0.46	18.69± 0.10	9.91± 0.43
Manioc	22.98± 0.33		11.02± 0.56	12.97± 0.54	10.87± 0.46
Cassava	27.03± 0.27	12.57± 0.13	11.98± 0.47	15.92± 0.55	13.80± 0.38
Unproduct.	41.78± 0.25	9.06± 0.14	9.13± 0.42	9.55± 0.51	8.79± 0.28
Garden	29.22± 0.17	9.13± 0.73	9.29± 0.28	11.56± 0.41	9.10± 0.23
Cement	204.44± 0.57	8.96± 0.12	23.54± 0.53	14.19± 0.37	25.47± 0.47

The radionuclide Th-231 found in point 3 and in cement can be considered an unprecedented result becausenone Th-231 in Brazilian soil and cement was related in literature. However, Bonotto et al.mentions that they found Th-231 in waters of the Guarani aquifer located in southeastern Brazil[24].

The activities determined for the other natural radionuclides in the samples are at or below the minimum detection limit. The lowest activity was found for K-40 with (0.11 ± 0.84) Bq.kg^{-1} in the soil near the Chemistry Department (point 2, in Table 1).

Because of its similarity to calcium, radium-226 can be fixed in the bones. This radioactive element emits alpha

particles, quite damaging when emitted inside the body. Also, when a radionuclide disintegrates, turns into another radionuclide, which may disintegrates if it is also radioactive. The process repeats until you finally get a stable isotope. In general, the natural radioactive elements are closely related to minerals and are not generally dangerous to human health. But when they involve the emission of gaseous radioactive elements can be very damaging because they can be inhaled.

Ra-226 when decays generatethe gaseous Rn-222. Rn-222 together with its non-gaseousdaughters, Po-218 and Po-214, are responsible for approximately 50% of the equivalent dose produced by natural ionizing radiation [25,26]. The radioactive gas Rn-222, when present in the air, can bind to dust particles and water droplets which are then inhaled and lodge in the lungs. So it is very important to determine the content of Ra-226 into the samples.

In soil samples of coconut plantation in areas B and C it was verified the activity of Ra-226 (18.22 ± 0.32) Bq/kg and (24.18 ± 0.42) Bq/kg, respectively, but they accounted for only 0.2% of the criteria for exclusion and exemption from the requirement of radiation protection given by the Brazilian National Commission of Nuclear Energy (CNEN), which is responsible for regulation in Brazil [27].Therefore, they are values very low.

Two samples,point 3 (near to UFS rector) and garden soil, besides presenting the radionuclides from the three natural series and K-40, presentedanunexpected artificialradionuclide, Cs-137. Meanwhile, its activity is below 0.05% of the limit of the exclusion and exemptioncriteria from the requirement of radiation protection; limit of 10 Bq/g,[27].

Fertilizer samples

Brazilian agriculture is a sector that has contributed most to the growth of the economy. As a result, the search techniques and tools that contribute to an increase in the production of different crops has been a major objective of modern agriculture, as the fertilizer industry.

It is known that chemical fertilizers mainly phosphate, may contain variables amount of uranium and thorium and their radioactive decay products and thus, they may contribute to increased levels of natural radionuclides on plants, causing an increase in dose to which consumers are exposed [28]. Therefore, 11 samples of fertilizers commonly used in the study area were analyzed. Of these, eight had only three radionuclides of natural decay series and K-40, as can be seen in Table 3 and 4.

The K-40 was the radionuclide that showed the highest activities in fertilizers, being the highest activity $(1636 \pm 36) \times 10$ Bq.kg^{-1} on KCl fertilizer.

The samples of organic fertilizers from cow and sheep manure contain the radionuclide Be-7, however their activitiesare only 0.05% below the limit of 1000 Bq/g[27].

Be-7 is a cosmogenic radionuclide produced by the process of spallation of oxygen atoms and nitrogen molecules in the troposphere and stratosphere. According to Appoloni andRios[29], the nuclear reaction produces BeO or Be $(OH)_2$, which diffuses through the atmosphere to form an aerosol that is deposited on the surface by fallout.

In most environments, the Be-7 binds rapidly and strongly in the surface soil, decaying by electron capture with emission of gamma rays of 477.56 keV and half-life of 53 days.

TABLE III
NATURAL AND ARTIFICIAL RADIONUCLIDES DETECTED IN FERTILIZERS

	Specific Activity (Bq/kg) for the radionuclides				
Samples	K- 40	Bi-212	Bi-214	Pb-212	Pb-214
KCl-U_1	(1617±34)×10				
KCl-F_1	(1636±36)×10				
U_2	3908±90				
Urea-F_2	24±2.0		1.29±0.21		
Phosphate-U_3	538±14	24.8±3.1	370.1±3.6	41.8±1.4	357.2±5.4
Phosphate-F_3	610±14	5.93±0.86	75.90±0.94	10.06±0.43	71.3±1.4
Legume-U_4	43.5±1.8			0.61±0.12	
Manure-sheep	629±16	4.08±0.52	4.08±0.52	8.32±0.44	4.70±0.39
Manure-caw	224.6±7.6	5.7±1.6	6.74±0.52	13.47±0.60	4.41±0.74
Manure-chicken	1334±28	11.6±1.8	15.20±0.54	17.86±0.51	16.12±0.46

TABLE IV
NATURAL AND ARTIFICIAL RADIONUCLIDES DETECTED IN FERTILIZERS

	Specific Activity (Bq/kg) for the radionuclides				
Samples	Ac-228	U-235	Th-231	Ra-226	Cs-137
KCl-U_1					
KCl-F_1					
U_2		1.51±0.28			
Urea-F_2					
Phosphate-U_3	43.3±1.1	26.1±1.7	99.0±6.0	633±40	
Phosphate-F_3	8.99±0.41	10.18±0.71	18.6±1.2	104±14	
Legume-U_4		0.57±0.08			
Manure-sheep	11.79±0.72	2.17±0.30			1.97±0.11
Manure-caw	17.56±0.78	17.56±0.78			
Manure-chicken	31.38±0.78	3.32±0.24			

In sheep manure, in addition to natural radionuclides, which were detected in the other samples, there was also the artificial radionuclide Cs-137.

The artificial Cs-137 radionuclide found in some samples of soil and fertilizershere analyzed is produced by the fission of uranium. It disintegrates emitting beta particles, electrons and gamma rays. The gamma rays resulting from the disintegration of Cs-137 is a very energetic radiation, so very penetrating, compared to other type of radiations, as alpha and beta particles. This fact should be taken into account, since it can greatly affect the population from the effects of interaction radiation on the human body. As the half-life of Cs-137 is 30 years, the effects of their pollution are felt over several decades.

Some years ago, the Cs-137 entered the atmosphere through nuclear weapons explosions, as tests conducted outdoors. Another way, more recent, was the chemical explosion and fire that occurred in the accidents with the Chernobyl and Fukushima nuclear plants. In all of them, the high temperatures volatilized large amount of materials, including radioactive. Soon after, it began the formation of particles, first with the less volatile materials. Since the initial concentration of the materials was high and located, these particles grew rapidly to reach a size that led to its deposition in the form of a fallout a few miles from its origin [30]. The

more volatile materials, including Cs-137, condensed after a significant drop in temperature. The overall concentration of materials was reduced by dispersion and as a result, the particles eventually formed were of smaller size and diameter, with little likelihood of rapid deposition in the absence of an official charger, like rain. Thus the amount of Cs-137, and other volatile radionuclides, which penetrate above the troposphere to the stratosphere, as a result of these explosions, could be distributed worldwide by the winds and ocean currents, slowly entering the biosphere through diffusion and deposition on land, through the rain [31,32].

Since the tropospheric exchange between North and South is lengthy, some Cs-137 derived from the Chernobyl accident and nuclear bomb explosions may arrive in Brazil [33]. Still, the Cs-137, found in tiny quantities in the soil and fertilizers studied here must be the result of atmospheric nuclear tests conducted in the 1950s.

IV. CONCLUSIONS

The soil had the lowest activity for K-40, with (0.11 ± 0.84) $Bq.kg^{-1}$.

The presence of Be-7 in organic fertilizers (half-life of 53.17 d) shows a short interval between the brewing processes for use in agriculture.

Although it was detected Cs-137 and Be-7 activities, they were within the limit of detection and do not represent risk for the population.

Th-231 found in samples of soil and cement in the state of Sergipe is an unprecedented result.

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