International Journal of Engineering, Mathematical and Physical Sciences
ISSN: 2517-9934
Vol:5, No:8, 2011

Dose due the Incorporation of Radionuclides Using Teeth as Bioindicators nearby Caetité Uranium Mines

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***Abstract*—**Uranium mining and processing in Brazil occur in a northeastern area near to Caetité-BA. Several Non-Governmental Organizations claim that uranium mining in this region is a pollutant causing health risks to the local population,but those in charge of the complex extraction and production of"yellow cake" for generating fuel to the nuclear power plants reject these allegations. This study aimed at identifying potential problems caused by mining to the population of Caetité. In this, work,the concentrations of ^{238}U, ^{232}Th and ^{40}K radioisotopes in the teeth of the Caetité population were determined by ICP-MS. Teeth are used as bioindicators of incorporated radionuclides. Cumulative radiation doses in the skeleton were also determined. The concentration values were below 0.008 ppm, and annual effective dose due to radioisotopes are below to the reference values. Therefore, it is not possible to state that the mining process in Caetité increases pollution or radiation exposure in a meaningful way.

***Keywords*—**bioindicators, radiation dose, radioisotopes incorporation, uranium.

I. Introduction

It is generally believed that some damage will occur to a person if one has any contact with radioactive materials. In fact, the radioactivity is present on Earth since its formation and nature, with all its living organisms, evolved in this background activity, natural and continuously exposed to ionizing radiation.

Many radioactive elements, natural or artificial, can be found in the environment, as in soil and rocks, water, food and beverages, construction materials, or even in the gases such as air we breathe, animals and plants, in short, everything that surrounds us and even inside human body [1]. However, some human activities such as mining may alter the distribution and concentrate the naturally occurring radioactive materials, increasing the radioactivity of a given area to values that may jeopardize the health and safety and the environment.

The radioactive elements more abundant in nature are ^{40}K, ^{238}U, and ^{232}Th, and their concentrations may vary in rocks and soils. In this context, Brazil has the 6th largest global geological reserve of uranium, the main reserves are in the Bahia (BA), Ceará (CE), Paraná (PR) and Minas Gerais (MG) states. Currently, the uranium mine of Caetité - BA is the only one explored in Latin American [2].

Some Non-Governmental Organizations (NGOs) such as Greenpeace [3] state that uranium mining in the district of Lagoa Real (Caetité-BA) is dangerous and polluting. However, INB, the company in charge of the complex extraction and production of yellow cake(and also for generating fuel for nuclear power plants in Brazil), rejected these accusations.

The orientation of Brazilian government is to continue the operations trying to minimize the impact on the environment. Despite new technologies that emerge each year, the contamination of the environment is a constant concern.

It is well known that once released into the environment, uranium may enter the human food chain through contaminated water or food like milk and vegetables. The continuous ingestion of uranium can cause several health hazards. Studies have shown that low concentrations of uranium ingested chronically lead to an accumulation of the element in bones, kidneys and throughout the volume of bone marrow hematopoietic stem cells, which may be exposed to alpha radiation [4].

Some radionuclides such as ^{238}U may be deposited in the teeth by replacing the calcium. Thus they have been long used as biomarkers of chronic exposure to heavy metals, for example, and provide a permanent record of exposure to the element [5]-[7]. Therefore, it is possible to estimate the incorporation of some radionuclides by determining its concentration in the teeth. By determining the concentration of radioisotope ^{238}U, ^{232}Th and ^{40}K, it is possible to estimate the radiation dose received, since these are the most abundant radionuclides in nature and the main contributors to the

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increase in background radiation.

To ascertain the actual effects of uranium exploration, this work was focused on the determination of ^{238}U, ^{232}Th and ^{40}K concentration in teeth of the Caetité population,as bioindicators, in order to assess the cumulative dose of radiation due to natural radioactivity of radioisotopes incorporated, and hence to know the impact of mining on the health of people living around it.

II. Materials and Methods

A. Sample Preparation

Fourteen teeth samples, extracted for orthodontic reasons from residents of the municipalities of Caetité (BA), were analyzed. Initially the teeth were cleaned with brush and running water in a dental office. Then they were dried with paper towels at room temperature. Subsequently, each tooth was cut with a dental diamond burs pen at low speed with constant water irrigation. Gloves, masks and goggles were used for individual protection, since the samples were pulverized and could be inhaled. Each tooth was separated into crown and root.

The determination of the isotopes contained in the samples was made from the roots of the teeth. All samples were crushed in an agate container and sieved to achieve particle size of 75 μm. In test tubes,1.5 mL of HNO_3wereadded to each200 mg sample. The solubilization was performed in a closed microwave oven, model DGT 100 plus – Provecto Analítica. After solubilization, it was necessary to wait 20 minutes to cool down the tubes. Next, the content of each tube was transferred into 10ml volumetric flasks and topped with purified water (18MΩ, Gehaka Master System).

B. Analytical Methods

Each digested subsample was analyzed using an inductively coupled plasma mass spectrometer(ICP-MS) system (Perkin Elmer ELAN 6100). This is a highly sensitive analytical technique used for the determination of the macro, micro and trace concentrations. To evaluate the analytical process and also the ICP-MS performance a statistical comparison between the elemental concentrations using the certified reference material IAEA Soil-7 Soil Trace Elements in Soil was done. After measuring each sample at least three times, mean and standard deviation were obtained.

This technique is regarded as a universal technique for analyzing solid samples, liquid and gas. It can also detect and separate the species in the presence of complex matrices [8]. The concentration of ^{238}U, ^{232}Th and ^{39}K were determined directly. The concentration of ^{40}K was calculated according to the natural isotopic distribution, 93.26% to^{39}K and 0.0117% to ^{40}K [9].

C. Dose Calculation

From the concentrations of ^{238}U, ^{232}Th and ^{40}Kradioisotopes in the samples, it was possible to calculate the equivalent dose received by teeth by (1):

$$H_{TOTAL} = H_U + H_{Th} + H_K \quad (1)$$

where H_U, H_{Th} and $H_K$are the equivalent dose (H) for each radionuclide^{238}U, ^{232}Th and ^{40}K, respectively. The equivalent dose H for each radionuclide is the average absorbed dose in the organ or tissue (D) multiplied by the radiation quality factors (w_R), given by ICRP [10].

With the equivalent dose it was also possible to calculate the effective dose (E), which is dependent on the organ affected. E can be obtained multiplying H by weighting factor of the respective tissue (w_T). For E received by skeleton, as teeth are similar in composition to bone tissue [10], it was used the weighting factors w_T bone marrow (W_{T1} = 0.12) and bone surface (W_{T2} = 0.01). The total effective dose rate, E_{TOTAL} was calculated by adding the effective doses due to each radionuclide (E_U, E_{Th} and E_K) due to radioisotopes ^{238}U, ^{232}Th and ^{40}K, respectively, according to (2):

$$E_{TOTAL} = E_U + E_{Th} + E_K \quad (2)$$

III. Results and Discussion

The quadrupole ICP-MS technique cannot provide concentrations of the elements of mass number 40 and its multiples. Thus, ^{40}K cannot be determined directly in the samples. However, it is possible to determine the concentrations of ^{39}K and, once the isotopic concentrations of potassium in nature is constant, it was possible to determine the concentration of ^{40}K using (3).

$$C_{K40} = (0{,}0117/93{,}26) \times C_{K39} \quad (3)$$

C_{K39} and $C_{K40}$are the concentrations of ^{39}K and ^{40}K, respectively. The concentrations of ^{238}U, ^{232}Th, ^{39}K and ^{40}K found in teeth and their respective uncertainties are presented in Table 1. Figure 1 shows the concentration of ^{238}Udetermined in this work compared with the reference concentration published by UNSCEAR [1] to the bones, which is 0.008 ppm, and corresponds to a specific activity of 0.1 Bq/kg.

By analyzing the samples isolated C 2, C4, Ca1, M4 and M11 it is noteworthy that incorporation of ^{238}U is above the reference value. However, all the other nine samples showed no incorporation of ^{238}U at all.

The average result obtained by Prado [11], who conducted a study on the levels of^{238}U incorporation by the inhabitants of the same region of Caetité also using teeth as bioindicators, was 0.0523 ppm, i.e., around 7 times greater than the reference value [1]. In this study, the average uranium embedded (0.007 ± 0.010 ppm) was approximately 8 times lower than that reported by Prado [11]. Also for Prado results, the majority of samples showed no incorporation of ^{238}U, but because some samples presented high rate of incorporation, the average was above the reference value. And this difference can be result of chemical or matrix interference existing on the analytical method.

TABLE I
CONCENTRATION THE ^{238}U, ^{232}Th, ^{39}K AND ^{40}K INTEETH SAMPLES EXTRACTED FROM THE CAETITÉ POPULATION

Samples	^{238}U (ppm)	$^{232}Th \times 10^{-3}$ (ppm)	^{39}K %	^{40}K %
C 2	0.029±0.004	58±9	60±15	0.0064±0.0015
C 3	0	53±8	49±12	0.0052±0.0013
C 4	0.030±0.004	59±9	50±13	0.0054±0.0013
Ca 1	0.009±0.001	16.6±2.5	35±9	0.0038±0.0009
Ca 2	0	54±8	47±12	0.0050±0.0012
PM 3	0	14.2±2.1	7.4±1.9	0.0008±0.0001
PM 4	0	58±9	45±11	0.0048±0.0012
PM 6	0	59±9	68±17	0.0072±0.0018
M 4	0.027±0.004	55±8	54±13	0.0058±0.0014
M 11	0.009±0.001	15.1±2.3	6.9±1.7	0.0007±0.0001
M 14	0	55±8	50±13	0.0054±0.0013
DL 2	0	53±8	47±12	0.0050±0.0012
DL 1	0	27±4	7.5±1.9	0.0008±0.0002
DL 3	0	58±9	66±17	0.0071±0.0017
Mean	**0.007±0.010**	**45±17**	**42±20**	**0.0045±0.0021**

C = Central incisor; Ca = Canine; PM = Pre-molar, M = Molar; DL=DeciduousLateral.

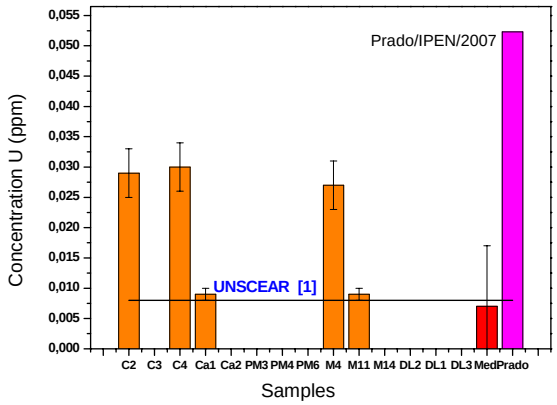

Fig. 1 Concentration the ^{238}U measured in samples of the teeth extracted from the Caetité population.

The average value of these cases is not a good index, since it can lead to erroneous conclusions. Severe cases should be further investigated. Some wells used by the population in locations not too close to the mining company have high concentration of ^{238}U, possibly due to natural causes [12]. Prado [11] concluded that the Caetité population is under a much higher radiobiological risk than other populations in the world. However, perhaps it is true only in some very specific locations in Caetité and maybe if the water consumption is restricted to certain sources there will be no problem for them. It must be clearly investigated.

Thus, for a more accurate conclusion about a possible population contamination due to U mining, the tooth sample should be carefully investigated, especially in relation to the origin of food and water consumed by this person. With the results presented here, it was not possible to conclude that U mining increases the concentration of U absorbed by the people surrounding the Caetité U mine, as stated by Greenpeace [3], mainly because the majority of teeth did not present ^{238}U incorporation at all.

Considering that the average weight of an adult is 70 kg (caucasian standard man), and knowing that about 10% of the total weight of the human body corresponds to the skeleton, then 7 kg of an adult is the weight of the skeleton [13]. From this, it was possible to estimate the average cumulative total of ^{238}U, ^{232}Th and ^{40}K for the human skeleton using the following expression (4):

$$C_{inc} = 7000 \cdot C_K \quad (4)$$

where $C_k$is the average concentration (in mg/g) of the radioisotopes found in teeth samples analyzed by ICP-MS. The values of incorporation (C_{inc}) found are (49 ± 70) g, (32 ± 12) g, (32 ± 15) g for ^{238}U, ^{232}Th and ^{40}K, respectively. The equivalent dose rate and total effective dose for the radionuclides in the study are presented in Table 2.

TABLE II
DOSE DUE TO THE INCORPORATION OF ^{238}U, ^{232}Th and ^{40}K I IN TEETH SAMPLES OF THE CAETITÉ POPULATION

Dose	^{238}U mSv/y	^{232}Th mSv/y	^{40}K mSv/y
H	(0.2±0.3)	(0.28±0.01)	(0.005±0.002)
E	(0.02±0.03)	(0.04 ±0.01)	(0.0006±0.0003)

H= Equivalent dose; E= Effective dose.

It is known that urine and/or feces eliminate over 95% of uranium entering the body. Wrennet al. [14] and Fisene and Wenford [15] observed by autopsy that bone is the site of greatest accumulation of uranium, with values between 2 and 62 mg. This variation is attributed to geographical factors, diet and analytical uncertainties. According to Eisenbud [16], the human skeleton contains 25 mg of uranium, which is equivalent to 0.296 Bq of activity and is responsible for a dose of 0.003 mSv/year. The concentration of ^{238}U obtained in the samples studied here is above the value found by Eisenbud [16], and also results in a higher accumulated dose. But the concentration determined is within the range established by Wrenn [14] and by Fisene and Wenford [15].

According to the ICRP [10], it is estimated that uranium absorbed into the blood tends to accumulate in the skeleton at an approximate value of 70 mg for adults. Therefore, in this case, the average concentrations of ^{238}U determined here is below this estimation.

According to the WHO [17] the human body incorporates by ingestion or inhalation of air, on average, 90 g of uranium. Approximately 66% of this element is found in bone (59.4 mg), 16% in liver (14.4 mg), 8% in kidney (7.2 mg) and 10% in other tissues. As the skeleton is the organ that most absorbs metals, the individual will always be exposed to radiation from radioactive isotopes of those metals that have been absorbed into the bone structure. The higher the incorporation, the higher is their exposure. From the value of ^{238}U embedded in the teeth evaluated in this work one cannot say that U mining is causing a greater accumulation of isotope in the Caetité population.

The contribution to the effective dose from ingestion and inhalation of the ^{238}U isotope is 50 μSv [18]. Since66% of uranium incorporated by a person is found in the bones, then, this would correspond to about 33 μSv annual effective dose in the skeleton due to the ^{238}U. Therefore, the annual effective

dose to the skeleton for samples in this work is below the reference value established by UNSCEAR [18].

The average concentration of ^{232}Th embedded in the teeth of the population of Caetité determined here was approximately five times higher than that found by Prado [11] (0.0094 ppm) in analysis of teeth by ICP-MS in the same locality. However, the reference range published by UNSCEAR [1] for the concentration of ^{232}Th in bones is from 0.001 to 0.006 ppm.Thus, all the teeth samples of Caetité residents analyzed here have ^{232}Th concentration well above the reference range, as well the average value found by Prado [11].

Also according to UNSCEAR [1], the concentration range of reference for the ^{232}Th in the human body corresponds to 6 to 24 mBq/kg. Assuming 7000 grams is the weight of the skeleton and converting these limits, the reference range for accumulated^{232}Th in skeleton is from 7 to 42 mg (42-168 mBq). Therefore the average concentration determined for ^{232}Th accumulated in the skeleton (32 ± 12) g is well above the upper limit of the reference.

However, it is reported in literature that^{232}Th and its decay products contribute in average of 90 μSv to the annual effective dose due to internal exposure from natural radionuclides [18]. As 70% of these elements are embedded in the bones, about 63 μSv of annual effective dose would be deposited in this organ. Thus, the calculated effective dose rate due to ^{232}Th(40 ±10) μSv/y is below the reference value.

Potassium, along with calcium and phosphorus, is one of the most abundant minerals in the human body. It is a necessary element to the internal functioning and metabolism of cells and it is also fundamental to maintain the dynamic balance presented by the membranes. Consequently, its radioactive isotope, the ^{40}K is the largest contributor to the natural radioactivity inside a human body, corresponding to 60 - 70% of the total dose [18].

Potassium is incorporated into the body through food. The homeostatic balance keeps its levels in the body, regardless of ingestion from food or water, whether radioactive or not. It is known that approximately 0.0118% of the concentration of potassium in the Earth is composed of the radioisotope ^{40}K. According to the NCRP [19], a man of 70 kg (reference man) contains approximately 140 g of potassium distributed in his body, with the majority in the muscles. Of these 140 g of potassium, 0.02 g would be ^{40}K, which corresponds to an activity of 3.7 kBq (0.1 μCi). The content of ^{40}K to the body weight is approximately 0.18% for adults and 0.2% for children [19].

According to UNSCEAR [18], ^{40}K leads to an annual equivalent dose of 170 μSv to the body for adults, with the largest contribution is due to β-particle rather than gamma radiation, since the beta radiation emitted by radioisotopes incorporated is almost completely absorbed by the body, while the gamma radiation can pass through the organs with very low interaction. About 95% of potassium in the human body is located inside the cells (mostly muscle), it is expected that ^{40}K contributes with small annual effective dose due to the skeleton, as really was found in this work.

The question of the biological effect caused by low dose levels is very complex since several factors such as food, drugs and chemicals contribute to damage the human body and even cancer. These doses of radiation, even though small, can potentially contribute to health problems in the population, but it is not possible to assign any health problem in the Caetité population to a greater incorporation of radionuclides due to mining of U Caetité according to the results of this work.

IV. Conclusion

The average concentration of^{238}U detected in the teeth extracted from the Caetité population is below the reference value set by WHO [17]. Some samples should be more detailed investigated because they presented high concentration above the reference value. The concentrations of ^{232}Th in samples of teeth are above the reference value indicated by the UNSCEAR [1], however the calculated effective dose rate is below the reference value. For ^{40}K, the values were low because the samples analyzed here are the teeth, and potassium accumulates, mostly in the muscles.

It was not detected any problems with the effective dose rate found in skeleton from population of Caetité and no problems resulting from the U mine exploration was detected. Therefore, it is not possible to affirm the mining process in Caetité is increasing pollution or exposition to radiation to the population.

Acknowledgment

The authors gratefully acknowledge the Fundação de Apoio a Pesquisa e a Inovação Tecnológica de Sergipe, Coordenação de Aperfeiçoamento de Pessoal de Nível Superior, Conselho Nacional de Desenvolvimento Científico e Tecnológico, theDepartmentof Nuclear PhysicsUniversityof São Paulo (USP), the INCT andcollaborators, Maria Célia Magalhães, Ionah Tavares, Gisele Gonçalves Costa.

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