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Radioactivity assessment of the waste deposited in Candonga's lake after the Fundão dam's collapse

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ABSTRACT

The collapse of the Fundão dam on November 5th, 2015, caused impacts along the hydrographic basin of Rio Doce. Renova Foundation has the mission to repair, restore and reconstruct the regions affected by this disaster. After the collapse, a huge amount of waste was deposited in Risoleta Neves Hydroelectric Power Plant's lake (Candonga). This waste has been dredged out from the lake and deposited in a nearby pile. The use of the waste from this pile is being considered for civil construction. Samples of this waste pile were collected and they were characterized by X-rays diffraction and EDS analyses, which showed that it consists of 80% quartz, 11% hematite, 8% goethite and 1% clay, mainly kaolinite. There is also an organic matter consisting of branches, roots, and leaves of trees, and plants. Previous radiological analyses made before the collapse showed that the radionuclide content of the waste deposited in Fundão dam was very low. However, the waste deposited in Candonga's lake has a new composition and it is important to evaluate its radioactivity assessment. The analysis results of the Candonda's waste samples will be presented and compared to the results obtained in the Fundão dam samples, before its collapse.

Keywords: Fundão dam's collapse, waste, radioactivity assessment.

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1. INTRODUCTION

With the collapse of the Fundão dam, November 5th, 2015, 32 million cubic meters of waste were entrained by the waters of the rivers, causing impacts on several communities and riverside villages, reaching as far as the Risoleta Neves Hydroelectric Power Plant's lake (Candonga) [1]. The large volume of waste reached the districts of Bento Rodrigues and Paracatu Baixo, Mariana (Minas Gerais), and a part of the district of Gesteira, Barra Longa (Minas Gerais) [2].

80% of the wastes that came from the Germano Mine were concentrated between Fundão dam and the Risoleta Neves Usina Hydroelectric Power Plant's lake (Candonga) in an extent of 113 km (Figure 1). The hydroelectric dam reservoir has retained 10.5 million m³ of the waste. The remainder followed from the Rio Doce to the sea, causing disruption in districts of Minas Gerais and the north coast of Espírito Santo [1,3].

Figure 1: *Candonga reservoir and vicinity.*

Foundation Renova is responsible to develop socio-economic and socio-environmental programs to repair, restore and reconstruct the regions affected by this disaster. The dredging of the

first 400 meters of Risoleta Neves Hydroelectric Power Plant's lake (Candonga) started on July 4th, 2016. Two dredgers are being used to remove about 5000 cubic meters of waste from the lake per day [3]. Samarco, the Brazilian mining company founded in 1977 and currently controlled through a joint venture between Vale S.A. and Anglo-Australian BHP Billiton, has created areas for the deposition of these wastes by placing them on pile shape, located along the Candonga reservoir. It also acquired a privately owned land in the proximities of the power plant. The company began to use the area for deposition of sediments in January of 2017. The use of the waste from this pile is being considered for civil construction. Samples of this waste pile were collected and they were characterized by X-rays diffraction and EDS analyses, which showed that it consists of 80% quartz, 11% hematite, 8% goethite and 1% clays, mainly kaolinite.

Previous works executed by the researchers from CDTN, before the collapse, shown that the radionuclide content of the waste stored in Fundão dam was very low [4]. However, the waste deposited in the Candonga dam has a new composition and the radioactivity assessment must be considered. The industrial wastes containing radionuclides have been receiving considerable global attention, because of the large amounts of NORM-containing wastes and the potential long term risks of long-lived radionuclides [5]. In this context, this work presents an assessment of natural radionuclides contents of the waste deposited in the Candonga dam. The analyses results of this waste samples will be presented and compared to the results obtained in the Fundão dam samples, before its collapse.

2. MATERIALS AND METHODS

The waste sample, Figure 2, was collected at Candonga reservoir, in the state of Minas Gerais, Brazil. The cement was provided by a conventional company of construction materials. Both the samples were homogenized and packaged in bags containing 3 kg of material, approximately.

Figure 2: *Aspects of waste sample collected at Candonga reservoir, Minas Gerais, Brazil.*

This work presents results of concentration measurements of radionuclides such as 232Th, 226Ra, and 40K by gamma spectrometry in the waste and in cement to estimate the radiological hazard risk by using the radium equivalent activity as radiation hazard index. It also presents results of exhalation rate of soil-cement specimens made of waste in substitution of regular sand used as building construction material, for comparison.

2.1 Gamma Spectroscopy

The samples were dried for about 48h in an oven at 60°C and transferred to Marinelli beakers (500mL). Each recipient was sealed for at least four weeks to reach secular equilibrium between 226Ra and 228Ra and their respective immediate progeny [6,7].

The measurements of natural radioactivity were performed by gamma-ray spectrometry: a system from CANBERRA, consisting of a hyper-pure germanium detector (HPGe), coaxial geometry, 15% efficiency, with data acquired and treated with Genie 2000 software.

All the gamma emitters nuclides from natural series of U and Th were determined by Gamma Spectrometry. 226Ra (238U chain) and 232Th were determined by measuring the areas of 214Bi and 212Pb, respectively, because the aging time of at least thirty days after sealing the Marinelli beakers, which assures the secular equilibrium between these elements and their progeny [7,8]. This

period of time is also enough to restore the equilibrium in the 232Th chain, between 228Ac and 228Ra.

The specific gamma line used and the detection limits are shown in Table 1 [9,10].

Table 1. Radionuclides determined by Gamma Spectroscopy Analysis and detection limits.

Radionuclide	Used gamma lines [energy (keV) - emission probability]	Detection limit ($Bq.kg^{-1}$)
^{40}K	1460.8 (10.7%)	1.0
^{214}Bi (^{226}Ra) [a]	609.3 (46.3%)	0.4
^{228}Ac	338.3 (11.4%); 911.6 (27.7%); 968.2 (16.6%)	0.4
^{212}Pb (^{232}Th) [a]	238.6 (44.6%)	0.4

[a] The gamma lines represent ^{214}Bi and ^{212}Pb. ^{226}Ra and ^{232}Th were determined indirectly.

The gamma spectrometry laboratories have been participating regularly in intercomparisons programs like the National Intercomparison Program coordinated by IRD/CNEN (Institute of Radiation Protection and Dosimetry), in Brazil, and by the IAEA (International Atomic Energy Agency). The isotopes 60Co, 65Zn, 106Ru, 133Ba, 134Cs, 137Cs, 226Ra, Th and U are analyzed in the intercomparison programs.

Calibration

Energy calibration was performed in the energy range from using point sources 0.8 to 2 MeV. The following photon emissions and radionuclides were used: ^{241}Am, ^{133}Ba, ^{152}Eu, ^{137}Cs, and ^{60}Co. The efficiency curve for calibration was determined by standard solution prepared from SRM (Standard Reference Material) or CRM (Certified Reference Material).

2.2 External exposure evaluation

When comparing the specific activity of samples containing different amounts of ^{226}Ra, ^{232}Th, and ^{40}K, it is necessary to introduce the term radium equivalent activity (Ra_{eq}). It represents a single quantity that combines specific activities of ^{226}Ra, ^{232}Th and ^{40}K. It is developed as a numerical

indicator for the external dose to the public. The maximum allowed value for public dose considerations is 370 Bq kg^{-1} [9]. Equation 1 shows how radium equivalent activity is calculated.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K, \quad (1)$$

where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1} respectively [11, 12].

The external hazard index (H_{ex}) is a radiation hazard index defined to evaluate the indoor radiation dose rate due to the external exposure to γ-radiation from the natural radionuclides in the construction building materials of dwellings [11]. This index value must be less than unity to keep the radiation hazard insignificant, i.e. the radiation exposure due to the radioactivity from construction materials to be limited to 1.0mSv y^{-1}. It was proposed the conservative model for limiting the radiation dose to this value, based on infinitely thick walls without windows and doors, in accordance with equation 2:

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810) \leq 1, \quad (2)$$

where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in Bq kg^{-1} respectively. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}).

2.3 Radon exhalation rate measurement

To determine the radon exhalation rate of the test mortar specimens. The samples were inserted in the sealed closed circuit showed in Figure 1. After reaching the equilibrium, the integral flow of atoms entering the circuit arising from the exhalation of the mortar specimens is maintained equal to the number of radon atoms that are removed from the circuit by radioactive decay. Thus, the diffusive flux density at the surface of the test sample can be calculated by Equation 3.

$$J_D = C_{Rn}.V_{ar}.\lambda_{Rn}/S \quad (3)$$

Where:

J_D is the radon flux density ($Bq.m^{-2}.s^{-1}$),

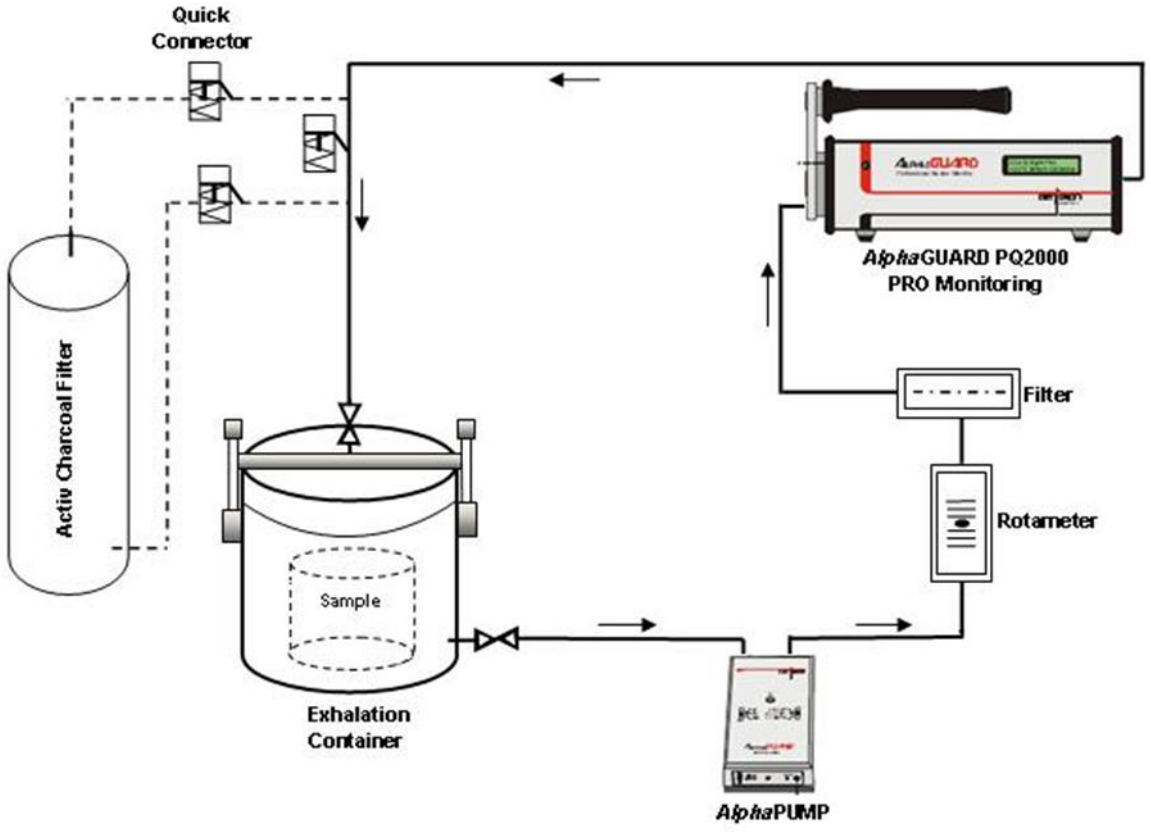
C_{Rn}. is the liquid equilibrium radon concentration indicated by the Radon Monitor AlphaGUARD PQ2000 PRO (Bq/m^3),

V_{ar} is the liquid internal air volume of the circuit (m^3),

S is the exhalation surface area of the sample brick (m^2), and,

$\boldsymbol{\lambda_{Rn}}$ is the radon decay constant (s^{-1})

Figure 3. *Closed-circuit used to determine the radon exhalation rate of the sample bricks.*



2.4 Evaluation method of internal exposure to radon and its daughters in environmental indoor

^{222}Rn and ^{220}Rn are the gaseous radioactive decay products of the direct decay of ^{226}Ra and ^{224}Ra, respectively. They are present in all terrestrial materials like that used in the walls, floor, and ceiling construction elements. By recoil, some of these radon isotopes are released from the solid matrix to the pore space of the material when radium decays. The fraction of atoms released of the material is called emanation coefficient or emanation power.

By diffusion and convection, radon atoms entering the pore space are then transported through this space until they decay or are released into the atmosphere, defined as exhalation. Thus the additional radon entry into dwellings or other buildings arising from the use of itabirite sand waste is evaluated by considering a model for reference building, presented by The United Nations Scientific Committee on Effect of Atomic Radiation – UNSCEAR presented in 1988 and 1993 Reports [13].

The model consists of masonry construction with a volume of 250 m^3, a surface area of 450 m^2, and the air exchange rate of 1h^{-1}. It is carried out to illustrate the effects of the several mechanisms of radon entry, including diffusion and convection from the ground and from building materials and other sources.

Based on the model reference building, the rate of radon entry into the building is calculated. For this calculation, the radon exhalation rate from mortar specimens made of itabirite waste, mortar specimens of Candonga waste, and a regular commercial were determined by measuring in a closed circuit as presented in 2.3.

2.5 Radon entry into buildings

The rate of radon entry from building elements into a house, U, assumed to be equivalent to the reference house assessed by the Committee [13], is given by Equation 4.

$$U = 3.6x10^3 \, x \, S_b \, x \, J_D \, / V \qquad (4)$$

Where,

U is the total rate of radon entry from the building elements into the house (Bq. m^{-3}. h^{-1}),

S_b is the surface areas of the walls (m^2),

J_D is the diffusive flux density ($Bq.m^{-2}.s^{-1}$), and

V is the volume of the house (m^3).

Considering the reference house, where the surface area of radon emitting walls is approximately 450 m^2 and the volume of 250 m^3.

3. RESULTS AND DISCUSSION

3.1 External exposure

The specific activities of individual nuclides determined in each sample are presented in Table 2 for two kinds of cement samples, 2 samples of different sand suppliers, the samples of itabirite waste, and the sample of waste deposited in Candonga. Cement and Sand samples were collected from industrial construction materials in Belo Horizonte and were analyzed to compare.

The shown results are the mean values of at least three measurements and the standard deviation of the mean. In Table 2, it is also presented the equivalent radium activity, Ra_{eq} (Bq/Kg) calculated by the equation (1) and the Radiation Hazard Index - H_{ex}.

The comparison of these values with the worldwide average concentration of ^{232}Th, ^{226}Ra and ^{40}K in construction material reported by UNSCEAR (1993) as 50 Bq kg^{-1}, 50 Bq kg^{-1} e 500 Bq kg^{-1} show: regular sand sample A has ^{232}Th and ^{40}K activity concentration higher than reference values; the regular sand sample B has only ^{232}Th activity concentration above the world average value in construction material; the cement A and B have a significant ^{226}Ra activity concentration; and in the samples wastes analyzed, ^{226}Ra, ^{232}Th, ^{40}K activity concentrations are close to the normal radiation background although of the natural radionuclides activity concentrations to be enhanced in the waste deposited in Candonga. This can be explained because with collapse the environmental conditions were changed and it has had preferential adsorption of the natural radionuclides in clay and

hematite presents in the waste deposited in Candonga. As shown in Table 2, in none samples the Ra_{eq} and the H_{ex} is above the limit proposed of the 370 Bq kg^{-1} and of the unity, respectively.

Table 2. *Specific activities of individual nuclides determined in each sample by gamma spectrometry and the calculated values of radium equivalent activity and Radiation Hazard Index - H_{ex}.*

Sample	Specific Activities and Radium Equivalent Activity (Bq kg^{-1})						(H_{ex})
	^{238}U series		^{232}Th series				
	^{214}Pb	^{214}Bi (^{226}Ra)	^{212}Pb	^{228}Ac (^{232}Th)	^{40}K	Ra_{eq}	
Regular Sand A	32.1±1.6	34.8±1.7	68.9±3.5	88.7±4.5	1305±20	262±7	0.71
Regular Sand B	38.1±0.6	39.9±0.8	65.6±0.6	84.1±1.7	335.3±6.0	186±3	0.50
Cement A	151.1±3.6	156.7±3.7	26.7±2.5	35.6±2,8	142±10	219±6	0.59
Cement B	215.8±3.1	204.5±2.2	37.0±0.8	41.8±4.1	160.8±5.1	277±6	0.75
Itabirite Waste (before collapse)	2.2±0.2	2.0±0.2	0.5±0.2	0.2±0.1	1.6±0.2	2.4±0.3	0.01
Waste Deposited in Candonga	7.4 ± 0.3	7.0 ± 0.3	4.6 ± 0.2	5.71 ± 0.2	33.0 ± 1.0	17.7 ± 0.5	0.02

3.2 Internal exposure to radon

The diffusive flux density at the surface of the test sample determined for the 3 mortar specimens (one made with 75% of regular sand, other with 75% of itabirite sand waste and other with 75% of waste deposited in Candonga) are presented in Table 3. It is also presented the calculated rate of radon entry from the building elements, and the additional radon concentration due to the use of the itabirite waste and the waste deposited in Candonga in the model reference building presented in the UNSCEAR 1988 and 1993 Reports [13].

The diffusive flux density (J_D) range from 0.001 to 0.074 mBq. $m^{-2}.s^{-1}$ and the total rate of radon entry from the building elements into the house (U) range from 0.007 to 0.480 Bq. $m^{-3}.h^{-1}$. Accord-

ing to Table 3, it was also possible to verify that the sand sample has higher J_D and U than waste samples. The sample of waste deposited in Candonga (after the collapse) present J_D and U higher than Itabirite waste (before collapse). Just for comparison, the values of J_D and U to the reference house are J_D = 0.0008 Bq. $m^{-2}.s^{-1}$ and U = 1.2 Bq. $m^{-3}.h^{-1}$ to floor slab and J_D = 0.0016 Bq. $m^{-2}.s^{-1}$ and U = 10 Bq. $m^{-3}.h^{-1}$ to surface areas of the walls (concrete). Considering an air exchange rate of the $1h^{-1}$, the indoor radon concentrations in the reference house from floor slab and from surface areas of the walls are about 10 and 1 $Bq.m^{-3}$, respectively [13].

Table 3. *Surface of the test samples, calculated rate of radon entry into the UNSCEAR reference house, and additional contribution to the radon concentration* ΔC_{Rn} *(Bq.* m^{-3}*).*

Mortar specimens (aggregate)	Net-CRn ($Bq.m^{-3}$)	S-Total Tested (m^2)	JD ($mBq.\ m^{-2}.s^{-1}$)	U ($Bq.\ m^{-3}.h^{-1}$)
Regular sand	346	1.96 x10-2	0.074	0.480
Itabirite Waste (before collapse)	123	3.92x10-2	0.001	0.007
Waste Deposited in Candonga (after collapse)	217	3.86 x 10-2	0.016	0.101

4. CONCLUSION

The samples of itabirite waste and the waste deposited in Candonga analyzed showed that the radionuclide content of this kind of waste even after some mixture with clay material is very low and even much lower than conventional materials used as building material, although it was possible to have occurred some relatively big increase in the radioactive content of the waste mixture with clay, especially in area of the Iron Quadrangle of Minas Gerais, where it is expected a high content of iron oxide and hydroxide, that efficiently scavenge for uranium. The results showed that

the replacement of regular sand by the itabirite waste or waste deposited in Candonga strongly reduces the radon exhalation and internal and external exposures.

Based on this preliminary radiological evaluation, the itabirite wastes and waste deposited in Candonga may be used in replacement of regular sand. However, more evaluation analysis must be carried out, considering that, due to geological and geochemical reasons, large variations may occur. This is preliminary work and must be continued, especially attempting for higher representatively of the samples.

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REFERENCES

[1] HEINECK, A.; VALADARES, L.; ANDRADE, M. **Samarco.** Belo Horizonte, Minas Gerias, Brasil. 2016. Available at: <http://www.samarco.com/wp-content/uploads/2016/11/Samarco-intensifies-construction-works-at-Candonga.pdf> Last accessed: 17 Nov. 2017.

[2] RIOS, F. **Samarco**. Belo Horizonte, Minas Gerias, Brasil. 2016. Available at: <http://samarco.com/wp-content/uploads/2017/01/Book-Samarco_final_baixa.pdf> Last accessed: 17 Nov. 2017.

[3] STOFELA, D.; PINTO, H.; DONÉ, A.; HEINECK, A.; VALADARES, L.; ANDRADE M. **Samarco.** Belo Horizonte, Minas Gerias, Brasil. 2016. Available at: <http://www.samarco.com/wp-content/uploads/2016/08/11072016-–-Samarco-intensifies dredging-at-Risoleta-Neves-Hydroeletric-power-plant's-lake-Candonga.pdf> Last accessed: 17 Nov. 2017.

[4] FREIRE, C.B.; CUCCIA, V.; SANTOS, T.O.; TELLO, C.C.O.; LAMEIRAS, F.S.; ROCHA, Z. Radioactivity assessment of pavement blocks made with Itabirite ore waste. **Rev Bra de Pesq e Desenvol**, v. 14, p. 55-61, 2013.

[5] IAEA International Atomic Energy Agency. **Technical Reports Series no. 419:** Extent of environmental contamination by naturally occurring radioactive material (NORM) and technological options for mitigation. Vienna: IAEA, 2003.

[6] NADA, A. Evaluation of natural radionuclides at Um-Greifat area, eastern desert of Egypt, **Applied Radiation and Isotopes**, v. 58, p. 275-280, 2003.

[7] BRUZZI, L. et al. "Radioactivity in raw materials and end products in the Italian ceramics industry." **Journal of Environmental Radioactivity**, v. 47, p. 171-181, 2000.

[8] CUCCIA, V. et al. Distribution of radionuclides in Bayer process. **INTERNATIONAL NUCLEAR ATLANTIC CONFERENCE**, 2007, Belo Horizonte. Annals Belo Horizonte: Comissão Nacional de Energia Nuclear, 2007.