

Minimizing Risks to the Environment from Norm Residues in Road Construction –11076

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ABSTRACT

This paper presents the results of a radiological environmental impact study related to the use of NORM residues generated by mining and processing as a basic material for the construction of roads in the internal areas of a phosphate fertilizer manufacturing plant. These materials include titaniferous hardpan, quartzite and soil, which are generated during the mining and beneficiation of phosphate rocks. Samples were collected and analyzed in order to determine U-238, Th-232, Ra-226, Ra-228, Pb-210, and K-40 content in raw samples and their leachate. The external and internal doses were estimated using conservative assumptions for some exposure scenarios such as external gamma exposure for members of the public that would use the roads and for the workers involved in the road construction, internal gamma exposure due to inhalation of suspended dust material and ingestion of contaminated water from an hypothetical well located near the road. A dosimetric model was developed to estimate effective dose assuming that gamma radiation emission comes from a polyenergetic source with a trunk geometric layout. The results indicated that utilization of quartzite and titaniferous hardpan as a raw material in road and access roadway construction would not result in workers and members of the public receiving a higher dose than the natural level in the region.

INTRODUCTION

On the last twenty years the need to evaluate the environmental and occupational radiological impact caused by the use of the NORM materials generated by industrial and mineral companies has increased. These types of companies typically generate raw materials rich in natural radionuclides, or by-products and residues rich in natural radionuclides. The radiological impact associated to the use of by-products/wastes generated by this type of companies generally depends on the management options that they adopt. Especially during the last decade, the best management option is to find some “marketable applications for the NORM residues” in order to transform them in a valuable resource instead of a useless and dangerous residues [1]. Obviously, the radiological, environmental, and health impact of these co-products in their possible applications should fall within the limitations imposed by the existing regulations at the national and/or international level.

According to the Brazilian Nuclear Energy Commission Regulation [2], from the point of view of radiological protection, practices that must be regulated includes:

a) handling, production, possession and utilization of sources, as well as transportation, storage and disposal of radioactive materials, encompassing all related activities that involve or could involve radiation exposure;

b) those that involve exposure to natural sources which CNEN considers the need to control.

In 2005 CNEN published the regulation CNEN/NN-4.01 “Requirements for Safety and Radiological Protection for Mineral-Industrial Installations” [3], in which the regulatory agency establishes obligatory monitoring criteria for installations that deal with radioactive materials or practices that could expose workers to conditions that are unhealthy because of the level of radiation exposure. The regulation states:

“Mineral-Industrial plants that deal with ore containing associated uranium or thorium or simply installations are locations in which raw materials containing radionuclides of the natural uranium or thorium series are treated, enriched or manufactured, including waste piles and waste storage areas”.

From the regulatory point of view, Brazil doesn't have a specific regulation for the utilization of naturally-occurring radioactive materials in construction of homes, roads, landfills, etc, which means that each case must be individually analyzed. Thus the evaluation of radiological impact associated with a specified practice must take place by means of radiation dose estimation to humans, taking different exposure scenarios into consideration.

Among the industries in which radiation exposure problems could be more significant, the cycle of mining and processing of minerals stand out. This is because when some minerals form, they incorporate uranium and thorium in greater concentration than is found in the earth's crust. The phenomenon takes place in mining industries, specifically those that mine coal, niobium, gold, iron, and heavy metals (such as zirconium and rare earths). The phosphate fertilizer industry should also be emphasized.

Concentrated phosphate material, containing 30% to 38% P_2O_5 , are the only sources of phosphorous on these fertilizers. Phosphate rocks, finely ground, while may be directly applied to soil as fertilizer, have low solubility and thus must be transformed into more soluble forms. Obtaining phosphate rocks takes place in the mineral extraction industry and various steps must take place in order to get to this product, which undergoes mining and processing of the ore.

The main natural phosphate deposits in Brazil come from two geological situations: igneous carbonated bodies (Catalao I: Goias, Tapira and Araxa, MG; Jacupiranga and Juquia, SP), which are responsible for around 80% of national production; and sedimentary formations (Patos de Minas/MG, Irece/BA, and Olinda/PE). Normally igneous phosphates have greater concentrations of thorium than sedimentary phosphates. The later, however, have greater uranium concentrations

Production of phosphate fertilizers generates great amounts of residues in all of its production steps, from mining to processing, moving through the industrial manufacturing processes of the fertilizer and the application of the fertilizer in soils. The tailings generated in the mining process, such as titanium hardpan and quartzite, are caused by weathering of alkaline rocks encountered around phosphate mineralization. Thus all of this material must be removed in order to carry out phosphate mining. In general, large volumes are generated, justifying a search for viable alternatives for utilization rather than simply directly disposing the material in piles. Both are considered naturally occurring radioactive material, that is, NORM.

The main objective of the present study is to evaluate the radiological impact coming from using waste tailings (titanium hardpan and quartzite), generated in the phosphate rock mining process, as a foundation for roads and access ways in the internal area of a future mining facility that will mine and process phosphate rock, as well as produce phosphate fertilizers. In order to carry out the study, five steps were defined:

- 1) sampling and analysis of natural radionuclide concentrations (U-238, Th-232, Ra-226, Ra-228, and K-40) present in tailing samples (quartzite and titanium hardpan), as well as soil samples from the region in order to understand the local natural radioactive levels in which the materials being studied will be used;

- 2) natural radionuclide (U-238, Th-232, Ra-226, Ra-228) concentration analysis present in the leachate of these materials;
- 3) application of a mathematical model to estimate the effective dose from external exposure to gamma radiation emitted by the naturally occurring materials (quartzite and titanium hardpan) and a comparison with the results obtained when using soil from the region;
- 4) estimation of the effective compromising dose from inhaling resuspended particulate matter during road construction and that from ingestion of water from underground wells located near the roads;
- 5) final analysis of the results obtained in comparison with the dose limits stipulated by CNEN radiological protection regulations [2].

MATHEMATICAL MODEL FOR CALCULATING EXTERNAL DOSE DUE TO GAMMA RADIATION

The model utilized to estimate the external dose received by individuals that will work in the roads or those that will use them is based on a model developed by Heilbron *et al* [4]. According to this research, in terms of natural external exposure, radionuclides from the U and Th series are responsible for the majority of human dose rates since the contribution of the radionuclides from the U-235 is negligible in dose determination.

The model proposed to calculate the external dose of the individuals includes the main uranium and thorium daughters gamma emitters in secular equilibrium (or not). The road is simulated as a cuted cone with the geometrical shape shown in Fig. 1.

The build-up factor is calculated using the Taylor's formula and the photons flux ϕ (photons/cm²s) at a specific distance x (cm) from the top of the source with height h (cm) and radius r (cm) can be determined by integrating a disk-shaped source according to the mathematical expression:

$$d\phi = \int (Sv/2) \{E_1[(b_1 + \mu_s * (h-x))] - E_1[(b_1 + \mu_s * (h-x)) \sec \theta]\} * [c * e^{-\alpha_1 * \mu_s * (h-x)} + (1-c) * e^{-\alpha_2 * \mu_s * (h-x)}] dx \quad (\text{Eq. 1})$$

In which:

$$E_1(x) = \int_x^\infty [e^{-y} / (y)] dy \quad (\text{Eq. 2})$$

And:

Sv = radioactivity of the source (Bq)/volume of the source (cm³);

$$b_1 = \mu_{ar} * z; \quad (\text{Eq. 3})$$

μ_{ar} is the air attenuation coefficient in cm⁻¹;
 μ_s is the soil attenuation coefficient, in cm⁻¹.

The following equation relates the photon flux ϕ (function of the radionuclides activity concentration) with the dose rates D at a distance z from the radioactive material surface:

$$D = \phi * E * P * (\mu/\rho)_{\text{tissue}} * f_c \text{ (rem/h)} \quad (\text{Eq. 4})$$

In which:

$$f_c = (1.6 \times 10^{-8} \text{ g. rem/MeV}) * (3,600 \text{ s/h});$$

(μ/ρ) = massic tissue absorption coefficient in cm^2/g ;

E = gamma energy in MeV;

P = percentage of gamma radiation emission by disintegration.

Thus, for a poly-energetic source, the total dose rate is the sum of the dose rates obtained for each energy. The solution for the integral equation is obtained by using the auxiliary integration formula:

$$\int e^{kx} E_1(b+ax) dx (1/k) \{(e^{kx}) * E_1(b+ax) - (e^{-kb/a}) * E_1[(b+ax)*(1-k/a)]\} \quad (\text{Eq. 5})$$

Heilbron *et al* [4] developed a “Mathematica” software for this model dose taking uranium and thorium’s decay chains into consideration.

The calculations were carried out considering:

- uranium and thorium are in secular equilibrium with their parents, resulting in 32 gammas for thorium and 23 gammas for uranium;
- the specific activity is $4.046 \times 10^3 \text{ Bq/g}$ for Th-232 and $1.244 \times 10^4 \text{ Bq/g}$ for U-238. A density of 1.8 g/cm^3 (approximately equal to that of the soil) and 1 Bq/cm^3 as concentration of uranium and thorium.

The density cited above is equivalent to:

- 1 Bq/cm^3 of U-238 is the equivalent of 44.22 ppm U;
- 1 Bq/cm^3 of Th-232 is the equivalent of 137.7 ppm Th.

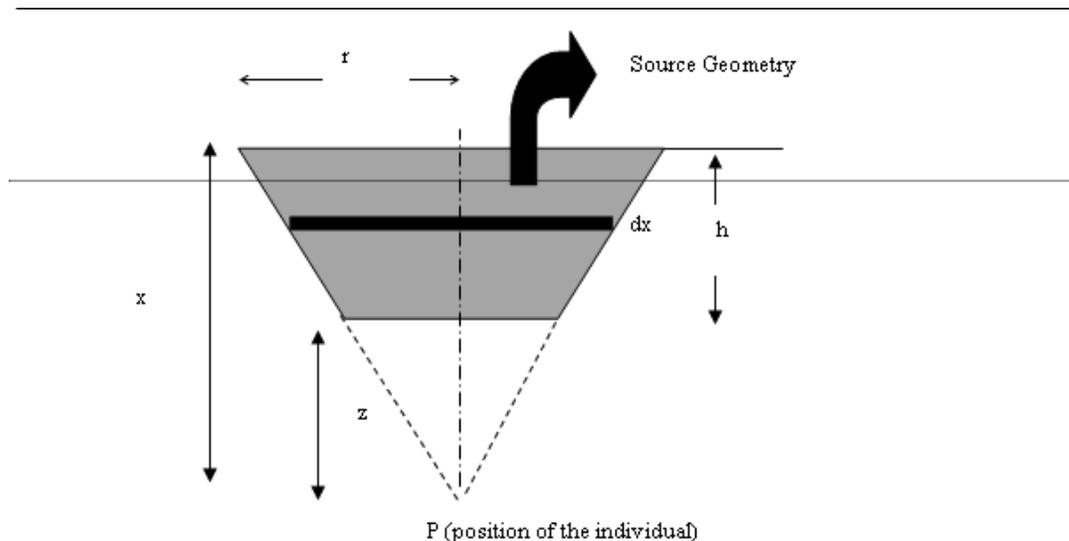


Fig. 1. Source Geometry.

Table I shows the dose rates obtained as a function of the source size and distance for uranium materials and Table II for materials associated with thorium. In both cases, the dose rate was estimated for a uranium concentration of 1 Bq/cm³ in secular equilibrium with its parents.

As can be seen, for materials containing uranium, the dose rate values converge for sources with a height of 1 m and a radius of at least 50 m, when distance from the surface z is 1 m, which means that a source of this size can be considered infinite. In this case, the dose rate at 1 m from a source surface of 1 ppmU is equal to 4.59 x 10⁻⁹ Sv/h (assuming that 1 Bq/cm³ U-238 is the equivalent of 44.22 ppm U). For materials containing thorium, the convergence of the dose rate values takes place for sources with 1 m height, radius of at least 50 m, and 1 m distance from the surface z. The source can also be considered infinite based on these values, resulting in a dose rate of 2.13 x 10⁻⁹ (Sv/h)/ppm Th at 1m from the source surface.

Table I. Dose rate results in function of the dimensions of a uranium source.

Height (m)	Radius (m)	Z (m)	Dose Rate (Sv/h) ^(*)
1	1	0.08	9.58 x 10 ⁻⁷
1	5	0.08	9.68 x 10 ⁻⁷
1	10	0.08	9.69 x 10 ⁻⁷
1	1	1	8.11 x 10 ⁻⁸
1	10	1	1.95 x 10 ⁻⁷
1	50	1	2.05 x 10 ⁻⁷
1	100	1	2.06 x 10 ⁻⁷
2	1	1	8.12 x 10 ⁻⁸

^(*)1 Sv ≈ 100 R

Table II. Dose rate results in function of the dimensions of a thorium source.

Height (m)	Radius (m)	Z (m)	Dose Rate (Sv/h) ^(*)
1	100	0.08	1.42 x 10 ⁻⁶
1	1	1	1.15 x 10 ⁻⁷
1	10	1	2.76 x 10 ⁻⁷
1	50	1	2.91 x 10 ⁻⁷
1	100	1	2.92 x 10 ⁻⁷
2	1	1	1.15 x 10 ⁻⁷

Equation 5 shows the relation between the dose rate TD expected at 1 m from the top of an infinite U and Th source as a function of their concentration in ppm (based on the 1 Bq/cm³ and a material density of 1.8 g/cm³).

$$TD = 4.59 \times 10^{-9} \text{ (Sv/h) /ppm U} + 2.13 \times 10^{-9} \text{ (Sv/h)/ppm Th} \quad (\text{Eq. 5})$$

It also stands out that for high densities (4.0 g/cm³) and fixed source geometry (radius = 11 m and thicknesses of 10 and 20 cm), the expected dose rate at one meter above the source per ppm of U and Th won't significantly vary with the source density. In this case, the greatest contributor to exposure

comes from the surface material because of the self-shielding source effect. This behavior is shown on Table III and IV.

As can be seen from table III the contribution of the first 10 cm layer of radioactive material is responsible for 93% of the dose rate 1 meter from the surface in the case of a uranium source and a density of 4g/cm^3 . For a thorium source, there are also no large differences (10%) in the expected dose rates at 1 m per ppm of Th. In other words, the contribution of the first 10 cm is responsible for 90% of the dose rate at 1 meter from the surface.

Table III. Influence of the density on the dose rate at 1 m for uranium sources with different thicknesses.

Density (g/cm^3)	Dose rate (Sv/h)	ppm U eq to 1 Bq U/cm^3	Dose rate/ppm U (Sv/h)/ppm
Radius = 11 m and thickness = 20 cm			
1.0	2.8×10^{-7}	80.4	3.5×10^{-9}
2.0	1.7×10^{-7}	40.2	4.2×10^{-9}
3.0	1.2×10^{-7}	26.8	4.5×10^{-9}
4.0	9.1×10^{-8}	20.1	4.6×10^{-9}
Radius = 11 m and thickness = 10 cm			
1.0	2.01×10^{-7}	80.4	2.51×10^{-9}
2.0	1.40×10^{-7}	40.2	3.49×10^{-9}
3.0	1.07×10^{-7}	26.8	3.98×10^{-9}
4.0	8.53×10^{-8}	20.1	4.24×10^{-9}

Table IV. Influence of the density on the dose rate at 1 m for thorium sources with different thicknesses.

Density(g/cm^3)	Dose rate (Sv/h)	ppm Th eq to 1 Bq Th/cm^3	Dose rate/ppm Th (Sv/h)/ppm
Radius = 11 m and thickness = 20 cm			
1.0	3.5×10^{-7}	246.3	1.4×10^{-9}
2.0	2.1×10^{-7}	123.2	1.7×10^{-9}
3.0	1.4×10^{-7}	82.1	1.8×10^{-9}
4.0	1.1×10^{-7}	61.6	1.8×10^{-9}
Radius = 11 m and thickness = 10 cm			
1.0	2.52×10^{-7}	246.3	1.03×10^{-9}
2.0	1.73×10^{-7}	123.2	1.41×10^{-9}
3.0	1.31×10^{-7}	82.1	1.59×10^{-9}
4.0	1.03×10^{-7}	61.6	1.68×10^{-9}

MODEL USED TO SIMULATE A ROAD

A road generally has a rectangular shape. Nevertheless, in this conservative model its shape will be considered as circular with the same surface area of a 50 m road wide (see Fig. 2). It is also important to emphasize that a 50 m-radius circular source is infinite, which means to say that the gamma dose rate contributions from radioactive materials outside of this range are negligible.

Thus:

Area of the circle = area of a rectangle 50 m long with width L, that is: $L \cdot 50 = \pi R^2$.

That is, a circle with radius R must be considered equal to:

$$\sqrt{(L \cdot 50) / \pi} \quad (\text{Eq.6})$$

Table V below presents R values in function of the length of the road L, such that the areas are equivalent.

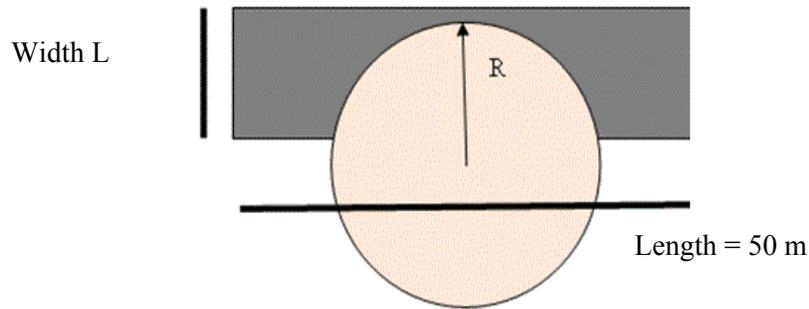


Fig. 2 – Model used to simulate the road.

Table V. Equivalent radius.

Road width (m)	Equivalent radius – R (m)
4	≈ 8
5	≈ 9
6	≈ 9.8
7	≈ 10.6

ROAD COMPOSED OF TWO DIFFERENT MATERIALS

For two different materials, the total dose rate estimate was determined considering the geometry in the calculations shown in Fig. 3.

Table VI below shows the results for dose rates A (20 cm), B (10 cm) and C(10 cm) per ppm at 1 m in function of U source density, considering radius of 11 m and Table VII below shows the results for dose rates A, B and C per ppm at 1m in function of Th source density, considering a 11 m radius

It should be noted that to calculate an expected final dose rate, resulting from one meter distance from the top of the road made of two combined material with a certain density ρ (g/cm^3), the procedure shown on Fig. 3 must be followed.

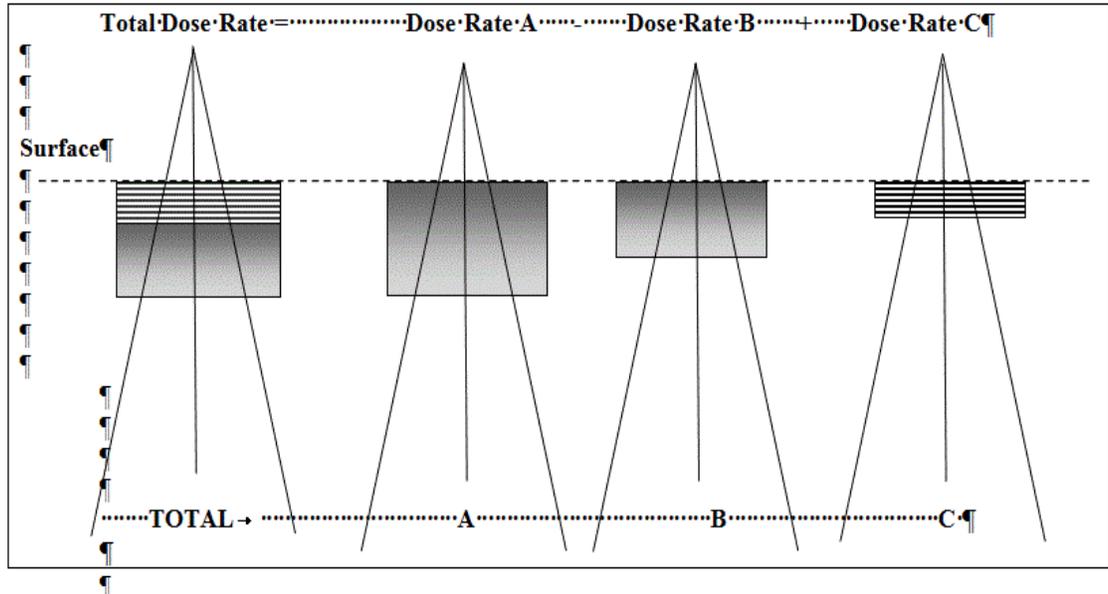


Fig. 3. Geometry for estimating external dose due to gamma emissions of a road constructed with materials of different compositions.

Table VI. Dose rate results in function of the density of a uranium source.

ρ (g/cm ³)	Dose rate A (Sv/h)/ppm U	Dose rate B (Sv/h)/ppm U	A-B (Sv/h)/ppm U	Dose rate C (Sv/h)/ppm U
1.0	3.5×10^{-9}	2.51×10^{-9}	0.99×10^{-9}	2.51×10^{-9}
2.0	4.2×10^{-9}	3.49×10^{-9}	0.71×10^{-9}	3.49×10^{-9}
3.0	4.5×10^{-9}	3.98×10^{-9}	0.52×10^{-9}	3.98×10^{-9}
4.0	4.6×10^{-9}	4.24×10^{-9}	0.36×10^{-9}	4.24×10^{-9}

Table VII. Dose rate results in function of the density of a thorium source.

ρ (g/cm ³)	Dose rate A (Sv/h)/ppm Th	Dose rate B (Sv/h)/ppm Th	A-B (Sv/h)/ppm Th	Dose rate C (Sv/h)/ppm Th
1.0	1.4×10^{-9}	1.03×10^{-9}	0.37×10^{-9}	1.03×10^{-9}
2.0	1.7×10^{-9}	1.41×10^{-9}	0.29×10^{-9}	1.41×10^{-9}
3.0	1.8×10^{-9}	1.59×10^{-9}	0.21×10^{-9}	1.59×10^{-9}
4.0	1.8×10^{-9}	1.68×10^{-9}	0.12×10^{-9}	1.68×10^{-9}

As expected, it stands out that for both uranium and thorium, when high density material (4.0 g/cm³) is used in both layers, the contribution of the exposure rate at 1 meter from the surface of the road due to the second layer is very small in relation to the contribution of the first layer, unless of course the concentration of radioactive material (in ppm) in the second layer is far superior than in the first one.

Density values of a rock vary between 2.5 g/cm³ and 2.7 g/cm³. For quartzite, density can vary between 2.42 g/cm³ and 2.65 g/cm³ [5]. On the other hand, ilmenite, FeTiO₃; leukoxene (a mineral neoformed by ilmenite alteration, called leukoxinated ilmenite); and rutile, TiO₂, are the main minerals in

titanium ore. There are other minerals that are potential sources of titanium: anatase, perovskite, and titanite. The density of titanite varies between 3.0 g/cm^3 and 3.5 g/cm^3 [6].

In the present work, the dose estimate was carried out for distinct scenarios. They are:

- 1) 10 cm of quartzite (upper layer) and 10 cm of titanium hardpan (lower layer);
- 2) 10 cm of titanium hardpan (upper layer) and 10 cm of quartzite (lower layer);
- 3) 20 cm of quartzite;
- 4) 20 cm of titanium hardpan;
- 5) 20 cm of soil collected from different areas.

Using considerations made earlier and the results of tables VI and VII, the dose rate expected at one meter from the top of the road for scenario 2, for example, was estimated as follows:

- 10 cm of titanium hardpan on top ($\rho \approx 3 \text{ g/cm}^3$)
 $3.98 \times 10^{-9} (\text{Sv/h})/\text{ppm U} + 1.59 \times 10^{-9} (\text{Sv/h})/\text{ppm Th}$
- 10 cm of quartzite on the bottom (conservative $\rho = 3 \text{ g/cm}^3$)
 $0.52 \times 10^{-9} (\text{Sv/h})/\text{ppm U} + 0.21 \times 10^{-9} (\text{Sv/h})/\text{ppm Th}$
- Total = $3.98 \times 10^{-9} (\text{Sv/h})/\text{ppm U} + 1.59 \times 10^{-9} (\text{Sv/h})/\text{ppm Th} + 0.52 \times 10^{-9} (\text{Sv/h})/\text{ppm U} + 0.21 \times 10^{-9} (\text{Sv/h})/\text{ppm Th}$

ANALYSIS OF NATURAL RADIONUCLIDE CONCENTRATIONS

As previously discussed, titanium hardpan and quartzite samples were collected, as well as soils from the region in which the industrial mining installation will be constructed. The facility will mine and process the ore and produce phosphate fertilizers. Phosphate rocks from the study region are of igneous origin.

Sample points were chosen randomly for the purpose of obtaining data that is representative of the studied area. A total of 20 sub-samples of each material to be studied were collected at the selected sites. They were sun dried, homogenized, and subsequently divided into representative quarters, with a small amount sent out for analysis.

The analyses for U-238, Th-232, Ra-226, Pb-210, and K-40 concentrations were carried out at the CNEN Poços de Caldas laboratory (LAPOC). The determination of U-238 and Th-232 concentration in each of the samples was carried out using the UV-Vis Spectrophotometer method with Arsenazo III [7]. The elements Ra-226, Ra-228, Pb-210, and K-40 were analyzed by the Gamma Spectrometry method [8].

Table VIII shows the results of the concentration analyses for radionuclides present in samples of titanium hardpan, quartzite, and soil from the study area. As can be seen, the results for U-238 and Th-232 concentration and their parent present in titanium hardpan are similar to those obtained in the regional soil samples (where today there are several coffee farms), with the exception of Ra-228.

Table VIII. Results of the radionuclide activity concentration presented in titanium hardpan, quartzite and soil samples.

Sample	Concentration (Bq kg ⁻¹) ¹					
	U-238	Th-232	Ra-226	Ra-228	Pb-210	K-40
Titanium Hardpan	1151 ± 148	694 ± 104	380 ± 28	578 ± 26	302 ± 46	<100
Quartzite	0.487 ± 63	<182	<20	48±3	<30	440 ± 45
Soil I (from the area in which the phosphate fertilizer production plant will be constructed)	566 ± 72	435 ± 72	108 ± 8	129 ± 8	80 ± 11	181 ± 25
Soil II (from the area in which the ore processing plant will be constructed)	1104 ± 143	575 ± 86	330 ± 20	330 ± 13	234 ± 23	<100
Soil III (from the area in which the tailing dam 1 will be constructed)	1801 ± 211	297 ± 59	140 ± 10	171 ± 8	114 ± 12	86 ± 19
Soil IV from the area in which the tailing dam 2 will be constructed)	1813 ± 212	299 ± 60	56 ± 5	70 ± 5	46 ± 8	541 ± 34

^(*) Background levels

DOSE DUE TO EXTERNAL EXPOSURE

The effective dose estimate, from external gamma radiation exposure caused by different materials, was carried out for two study scenarios. They were:

Scenario 1) an individual working in the road construction 40 hours per week, during 3 months. In this case, his total exposure time would be 480 hours per year.

Scenario 2) people using the road on foot, making a 30-minute trip one way and a 30 minute return trip each day, considering a 5-day work week (40 weeks per year) and thus being exposed for the equivalent of 200 hours per year.

Table IX shows a summary of all of the results for external dose rate for each of the study conditions. It's important to note that the results on this Table are overestimated since the dose estimate from external exposure began with the assumption that the elements U and Th are found in secular equilibrium (the activity of the parent is the same as that of the parent).

The hypothesis considered is very conservative; and it isn't true especially in the case of U as observed on Table VIII. For example, in titanium hardpan the ratio between Ra-226 and U concentration is

¹ 1 Bq U/g → 80 ppm U and 1 Bq Th/g → 247.4 ppm Th

0.3 (380/1151). Therefore, the activity of the parent is much smaller than that of the parent, indicating that the final dose due to direct exposure to gamma rays emitted by uranium parent should be less than 1/3 of the value obtained. In the case of thorium, note that the hypothesis of secular equilibrium is reasonable, since the ratio of Ra-228 to Th is 0.8. Therefore, the final external dose value from U and Th contribution in secular equilibrium with their parent will be approximately half of the value shown on Table IX. This is valid for all of the other scenarios considered.

The concentration of K-40 is low as can be verified below:

Scenario 1

$$1.2 \times 10^{-8} \text{ (Sv/h)/ppm K} * 1.7 \text{ ppm K} * 480 \text{ h/year} = 0.10 \text{ m Sv/year.}$$

Scenario 2

$$1.2 \times 10^{-8} \text{ (Sv/h)/ppm K} * 1.7 \text{ ppm K} * 120 \text{ h/year} = 0.02 \text{ mSv.}$$

Table IX. Results of the dose estimate of external exposure to gamma radiation for each study condition.

Materials used to build the road	Dose rate due to external exposure (mSv/year)	
	Scenario 1	Scenario 2
10 cm quartzite (upper layer) and 10 cm of titanium hardpan (lower layer)	0.44	0.18
10 cm of hardpan (upper layer) and 10 cm of quartzite (lower layer)	0.37	0.15
20 cm of quartzite	0.16	0.07
20 cm of titanium hardpan	0.35	0.15
20 cm of soil from the area where the phosphate fertilizer plant will be built	0.21	0.09
20 cm of soil from the area where the ore processing plant will be built	0.30	0.12
20 cm of soil from the area where waste dam 1 will be built	0.35	0.15
20 cm of soil from where waste dam 2 will be built	0.37	0.15

DOSE FROM INTERNAL EXPOSURE

Inhalation

For a conservative estimation of the internal dose to the workers that will build the road, the air concentration can be obtained using the dust load concept. Various factors influence the resuspension of air particles, including weather conditions, local wind speed, local terrain layout, time of deposition, particle size, etc.

Particle diameter is important for two reasons: the quantity that can be resuspended and the quantity that can be inhaled. Particles in the range of 1000-2000 μm diameters can roll or slide along the surface because of the wind while particle in the range 50-1000 μm in diameter can be vertically suspended in the air due to wind action and quickly settle because of gravity.

Particles smaller than 50 µm in diameter can be resuspended by the wind or other disturbances in the area and remain suspended for a long period of time. Very small particles (0.1 µm in diameter or less) are rarely resuspended alone (they are suspended in aggregates with other materials) [9].

From the point of view of radiological protection, this study deals with calculation of the dose for those particles that can be inhaled by humans, that is, below 10 µm in diameter.

The dust load CP is generally given in kg/m³ and is defined as the ratio between concentration of the radionuclide in the air C_{air} in Bq/m³ and the activity concentration of radionuclides in the soil C_m given in Bq/kg, that is:

$$CP = C_{air} (Bq/m^3) / C_m (Bq/kg) \quad (\text{Eq. 7})$$

In urban areas the dust load varies between 5 and 50 µg/m³ and in large industrialized areas in the range of 100 to 800 µg/m³ [9]. However, larger values can be encountered in specific areas due to disturbances caused by humans, mainly in very dry environments (370 – 65000 µg/m³), for example near vehicles that move such as tractors, earthmovers, etc.

In order to estimate internal dose, a conservative calculation can be made assuming that some of the material (10%) has a diameter smaller than 10 µ (breathable fraction) in the less dense material (quartzite, 2.4g/cm³) in such a way that results in a greater specific activity.

Considering the recommendation of Wilkins [10] for CP and the previous data that would result in maximum air concentrations due to uranium and thorium present in the quartzite, it can be inferred that:

- 10,000 µg/m³ * 0.789 Bq/g*10% = 0.00079 Bq U/m³
- and
- 10,000 µg/m³ * 0.182 Bq/g*10% = 0.00018 Bq Th/m³

The committed effective doses of the workers due to inhalation can be calculated by:

$$D (Sv) = C_{air} (Bq/m^3) * inhalation\ rate (m^3/h) * dose\ factor (Sv/Bq) * exposure\ time (h/year) \quad (\text{Eq. 8})$$

Dose factors taken from IAEA [11] are found reported on Table X below.

Table X. Conversion factors for dose by inhalation

Radionuclide	Conversion factor for committed effective equivalent dose due to inhalation (Sv/Bq)
Th-232+D	1.1x10 ⁻⁴
U-238 + D	8.0x10 ⁻⁶

Taking into account the previous data:

- For uranium: 0.00079 Bq/m³ * 1 m³/h * 8x10⁻⁶ Sv/h * 960 h/year = 0.0061 mSv/year
- For thorium: 0.00018 Bq/m³ * 1 m³/h * 1.1x10⁻⁴ Sv/h * 960 h/year = 0.019 mSv/year
- 0.025 mSv/year total internal dose

It must be mentioned that the expected doses for inhalation will be much less than calculated since the use of masks by the workers wasn't considered, which would reduce doses to almost zero.

Ingestion

One of the aspects that must be emphasized is the possible long-term impacts due to leaching of the material utilized as a road construction material and the consequence of its migration into the environment. Leaching takes place because of liquids present in the materials deposited and that are released during the process of decomposition and infiltration by rain water. This liquid can permeate the deep layers of the area of deposition and reach the ground water, in addition to be transported on the surface to the nearby waterways.

Table XI shows the concentration results of U-238, Th-232, Ra-226, Ra-228 and Pb-210 present in the leachate of each of the collected samples. As can be seen, the results of analysis of titanium hardpan leachate are similar to those obtained for the different types of soil that are found in the region where the facility will be constructed.

Table XI. Analysis results for natural radionuclide concentration present in leachate samples

Sample Identification	Leachate concentration (Bq L ⁻¹)				
	U-238	Th-232	Ra-226	Ra-228	Pb-210
Titanium Hardpan	0.12 ± 0.01	<0.006	0.55 ± 0.06	0.8 ± 0.1	0.083 ± 0.007
Quartzite	0.16 ± 0.02	<0.006	0.20 ± 0.03	0.32 ± 0.03	0.10 ± 0.01
Soil I	<0.1	0.007 ± 0.002	0.56 ± 0.06	0.66 ± 0.09	0.20 ± 0.02
Soil II	0.13 ± 0.01	0.007 ± 0.002	0.55 ± 0.05	0.65 ± 0.08	0.053 ± 0.006
Soil III	< 0.006	<0.1	0.47 ± 0.05	0.67 ± 0.08	0.08 ± 0.01
Soil IV	0.11 ± 0.01	0.007 ± 0.002	0.54 ± 0.06	0.66 ± 0.09	0.11 ± 0.01

Determining the concentration of radionuclides in groundwater coming from leaching of contaminants present in the soil can be estimated by using simplified models as well as by using more complex models that require numerical solutions [12]. In general two methods are used: one that utilizes the Dilution/Retardation Factor (DAF) to consider the mixture of the leachate contaminants with groundwater, and the other that utilizes a saturated zone flow model and transportation to calculate the resulting concentrations at a specified study point considering advective, dispersive, decay and biodegrading processes of the radionuclides in groundwater.

The first method conservatively assumes that there is a well for water consumption by members of the public located near the edge of the contamination source (that is, there is no dilution between the points of release of the contaminant and consumption).

In the present study, the estimate for pollution concentration in groundwater was carried out using the Dilution/Attenuation Factor (DAF) [13, 14]. This case comes from the hypothesis that even though the contaminants present in the soil solution undergo physical, chemical, and biological processes that tend to diminish the concentration that reaches the groundwater, only the physical dilution process was considered [13].

Once it is in the aquifer, dilution by clean underground water would further reduce the concentration before the contaminants reached the reception point (for example, wells where water is used for human consumption). The ratio between radionuclide present in the soil leachate solution (C_{soil}) and the concentration present in groundwater (C_{water}) is given by [13]:

$$C_{water} = C_{soil}/DAF \quad (\text{Eq. 9})$$

EPA adopted a DAF value of 20 and CETESB a value of 20 [13, 14]. As the DAF value increases, less contamination is expected in the aquifer. In this work, the DAF value of 10 was utilized as recommended by CETESB.

The committed effective dose by water ingestion from a hypothetical well dug beside the road is given by:

$$D (Sv) = C_{water} (Bq/L) * annual\ water\ consumption (L/year) * dose\ factor (mSv/Bq) \quad (Eq. 10)$$

Annual water consumption was defined as being 730 L/year [15]. Dose factors [16] for ingestion of radionuclides by an adult member of the public are shown on Table XII.

The final results of effective committed dose by ingestion, taking into consideration the concentration present in the leachate of each of the materials analyzed, are shown on Table XIII. As can be seen, all of the values estimated are below the limits adopted by CNEN for members of the public, that is, 0.3 mSv/year [2].

Table XII. Dose factor for ingestion of each of the studied radionuclides [15]

Dose factor (mSv/Bq)				
U-238	Th-232	Ra-226	Ra-228	Pb-210
4.51E-05	2.31E-04	2.81E-04	6.91E-04	6.91E-04

Table XIII. Effective committed dose due to water ingestion.

Sample composition	Effective Committed Dose (mSv/year)					Total (mSv/year)
	U-238	Th-232	Ra-226	Ra-228	Pb-210	
Titanium Hardpan	3.95E-04	1.01E-04	1.13E-02	4.04E-02	4.19E-03	0.056
Quartzite	5.27E-04	1.01E-04	4.10E-03	1.61E-02	5.27E-04	0.026
Soil I	3.29E-04	1.18E-04	1.15E-02	3.33E-02	1.01E-02	0.055
Soil II	4.28E-04	1.18E-04	1.13E-02	3.28E-02	2.67E-03	0.047
Soil III	1.98E-05	1.69E-03	9.64E-03	3.38E-02	4.04E-03	0.049
Soil IV	3.62E-04	1.18E-04	1.11E-02	3.33E-02	5.55E-03	0.050

CONCLUSION

The main objective of this work was to evaluate the radiological impact from using naturally occurring radioactive materials (titanium hardpan and quartzite) as a base for road construction in roads and access ways in an internal area of an industrial mining complex.

The effective dose estimates from external exposure by means of a mathematical model show that utilization of the materials analyzed in this work as a base for road construction in an internal area of an industrial mining complex wouldn't result in an increasing of the dose of individuals of the public above the natural radiation levels of the region. The effective dose from external exposure to gamma radiation received by individuals that would use these roads is the same as if they use other roads nearby made of the soils of the region.

The estimate of external dose for workers and individuals that will use the roads was made starting with a conservative hypothesis, that is, that uranium and thorium present in the materials being studied are in secular equilibrium with their parents. The results of the analyses for determination of the concentration of naturally-occurring radionuclides (U-238, Th-232, Ra-226, Ra-228, and Pb-210) showed that this isn't true. For example, in the case of utilization of titanium hardpan, the effective dose from external exposure shall be approximately half of the estimated value. This implies that final dose values for road workers for

480 hours of exposure per year, and for those that use the roads are under the restricted limit adopted by CNEN for members of the public, which is 0.3 mSv/year (CNEN, 1988).

Internal doses from inhaling the dust resuspended during road construction were also considered negligible. The dose from ingesting water from a hypothetical well dug beside the road was below the dose restriction limit adopted by CNEN for members of the public, that is, 0.3 mSv/year (CNEN, 1988).

References

1. M.J. GÁZQUEZ, J.P. BOLÍVAR, R GARCÍA-TENORIO, F. GALÁN, “Natural Occurring Radionuclide Waste in Spain: The Huelva Phosphogypsum Stacks Case”, Proceedings of the 1st Spanish National Conference on Advances in Materials Recycling and Eco – Energy, Madrid, Spain (2009).
2. COMISSÃO NACIONAL DE ENERGIA NUCLEAR, “Diretrizes Básicas de Radioproteção”, Rio de Janeiro, CNEN NE 3.01 (1988).
3. COMISSÃO NACIONAL DE ENERGIA NUCLEAR, “Requisitos de Segurança e Proteção Radiológica para Instalações Minerio-Industriais”, Rio de Janeiro, CNEN NE 4.01, (2005).
4. P.F.L. HEILBRON FILHO, A.M. XAVIER, J.P. GUERERO, R.N. ALVES, E.M. PONTEDEIRO, “Development of Radiological Basis for Transport Safety Requirements for Low Specific Activity Materials and Surface Contaminated Objects”, Proceedings of the III Meeting on the IAEA Co-ordinate Program, on LSA Radioactive Materials, Cable Town, South Africa (2001).
5. A. Andrew, “Densities of Common Rock Types”, http://geology.about.com/cs/rock_types/a/aarockspeggrav.htm. (accessed April, 04, 2010).
6. V.S. Urusov, N.N. Eremin, O.V. Yakubovich, O.V., “Electron-Density Distribution in Ca[TiO(SiO₄)] Titanite”, Crystallogr. Reports, v. 40, pp. 442-448 (1995).
7. M.C. SIQUEIRA, K.A.P. OLIVEIRA, A.A. MOZETO, M.H. TADDEI, F.F. DIAS, V.M. JACOMINO, “A comparison between spectrometry with Arsenazo III and neutron activation for determination of U and Th in Phosphogypsum”, Proceedings of the International Nuclear Atlantic Conference INAC 2007, Santos, Brazil (2007).
8. C.H.R. SAUEIA, B.P. MAZZILLI, B. P., “Distribution of Natural Radionuclides in the Production and Use of Phosphate Fertilizers in Brazil”, Journal of Environmental Radioactivity. v. 89, pp. 229-239 (2006).
9. T. GODISH, “Air Quality”, Lewis, New York, NY, 448p (1997).
10. B.T. WILKINS, B.T., “Influence of radionuclide bearing marine sediment on terrestrial exposure pathway”, NRPB-M461, Chilton, USA (1994).
11. INTERNATIONAL ATOMIC ENERGY AGENCY, “Exemption of Radiation Sources and Practices from Regulatory Control” Vienna, Austria, IAEA-TEC-DOC 401 (1987).
12. K.H. BIRDSSELL, A.V. WOLFSBERG, D. HOLLIS, “Groundwater Flow and Radionuclide Transport Calculations for a Performance Assessment of a Low Level Waste Site”, Journal of Contaminant Hydrology, v. 46, pp. 99 – 129 (2000).
13. US ENVIRONMENTAL PROTECTION AGENCY, “Soil Screening Guidance for Radionuclides: User’s Guide”, Washington, DC, EPA/540-R-00-007, (2000).
14. COMPANHIA DE TECNOLOGIA DE SANEAMENTO AMBIENTAL, “Relatório de Estabelecimento de Valores Orientadores para Solos e Águas Subterrâneas no Estado de São Paulo”, São Paulo, 73p (2001).
15. WORLD HEALTH ORGANIZATION, “Guidelines for Drinking Water Quality”, 3rd Edition, Incorporating the First And Second Addenda Vol. 1, Recommendations, Geneva, Suisse (2008).
16. INTERNATIONAL ATOMIC ENERGY AGENCY, “International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources”, Vienna, Austria, Safety Series No. 115 (1996).