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Analysis and risk estimates to workers of Brazilian granitic industries and sandblasters exposed to respirable crystalline silica and natural radionuclides

L. Estellita [a], A.M.A. Santos [b], R.M. Anjos [a,*], E.M. Yoshimura [c], H. Velasco [d], A.A.R. da Silva [c], J.G. Aguiar [b]

[a] Instituto de Física, Universidade Federal Fluminense, Av. Gal Milton Tavares de Souza, s/n, Gragoatá, 24210-340 Niterói, RJ, Brazil
[b] Fundação Jorge Duprat Figueiredo de Segurança e Medicina do Trabalho, Fundacentro, C.P. 11484, 05499-970 São Paulo, SP, Brazil
[c] Instituto de Física, Universidade de São Paulo, C.P. 66318, 05314-970 São Paulo, SP, Brazil
[d] Grupo de Estudios Ambientales - IMASL. Universidad Nacional de San Luis / CONICET. Ej. de los Andes 950, D5700HHW San Luis, Argentina

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ABSTRACT

Occupational exposure to respirable crystalline silica and to radiation emitted by natural radionuclides present both in rocks and sands was studied in the Brazilian extractive process and granite product manufacture. Respirable airborne dust samples were collected in working environments, where workers perform different tasks with distinct commercial granites types, and also in places where sandblasters work with sands from different origins. The free crystalline silica contents were determined using X-ray diffraction of the respirable particulate fraction of each sample. Dust samples from granite cutting and sandblasting ambient had the natural radionuclides concentrations measured by gamma spectrometry. Dust concentrations in the workplaces were quite variable, reaching values up to 10 times higher than the respirable particle mass threshold limit value (TLV) set by the American Conference for Governmental Industrial Hygienists of 3 mg m^{-3}. Also the free crystalline silica concentrations were high, reaching values up to 48 times the TLV of 0.025 mg m^{-3}. Additionally, our results suggest that the risk of radiation-induced cancer in the granite or marble industries is negligible. However, the combined exposure to dust, gamma radiation, and radon daughter products could result in the enhancement of lung cancer risks associated to sandblasting activities.

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1. Introduction

Brazil is an important producer and trading center of ornamental stones such as granites, marbles, limestone, quartz-based stones, slates and basalts. More than half of this production corresponds to over 200 different types of "commercial granites" with specific names, geological origins and mineral compositions. They are hard natural stones and, compared to marble, they require stronger tools to be extracted from a mountain, cut, shaped and polished. Concerning their compositions, granites are mixtures of multicolored mineral grains such as quartz, feldspar, hornblende, pyroxene and biotite (Anjos et al., 2005). Thus, granite is rich in silica, with contents up to 65%, varying in each kind of rough stone. In addition, it is well-known that the presence of natural radionuclides in its composition can produce high radiation levels (Anjos et al., 2005).

Due to the relative simplicity of the Brazilian ornamental stone productive process, low-investment small business, hiring a few unqualified employees, are very frequent in this commercial activity, mainly in granite and marble shops. In the extraction mines, workers use essentially pneumatic rock drillers. Removal of granite blocks of about 30 ton requires, for example, drilling holes into the rock formation to accommodate explosive charges or expansive mortars. Saws, drills, grinders and polishers are used in finishing works. Though the literature contains many reports about the occupational risks inherent to such kind of activity, the lack of information among the workers is still a big problem. Included in these risks are the high levels of exposure to crystalline silica dust, which increase respiratory diseases, mainly silicosis. Due to the importance and gravity of this disease and to the elevated number of workers exposed to dust in mines or shops of ornamental stones, one of the priority actions of the Brazilian Government concerning social health is the Brazilian Program to Eliminate Silicosis, in consonance with the ILO/WHO global program for elimination of silicosis (Algranti et al., 2004).

Understanding early pulmonary responses to inhaled particulates may lead to prevention and intervention approaches to help to avoid or at least to mitigate the development of irreversible occupational lung diseases (Kuempel et al., 2003). Several studies have established quantitative relationships between cumulative exposure to respirable crystalline silica dust and ornamental stone

* Corresponding author. Tel.: +55 21 26295843; fax: +55 21 26295887.
E-mail address: meigikos@if.uff.br (R.M. Anjos).

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workers' pneumoconiosis (Tharr, 1999; Santos et al., 2007). It is well documented that chronic and, in some cases, acute exposure to dust containing silica can cause serious health problems (Tharr, 1999; Marchiori et al., 2006). Smokers are at an increased risk; smoking and silica act synergistically in causing chronic obstructive disease in the lung (Hnizdo and Sluis-Cremer, 1991; Malmberg et al., 1993; Tharr, 1999). Silica exposure has also been associated with other lung diseases, such as tuberculosis and lung cancer (Tharr, 1999; Marchiori et al., 2006).

Another professional activity that can lead to silicosis and risk of deaths for workers exposed to respirable crystalline silica is sandblasting (Marchiori et al., 2006). In Brazil, this activity is mainly concentrated in surface cleaning (stone buildings, metal bridges, and other metal surfaces) and removal of painting and barnacles from ship hulls. Abrasive blasting involves forcefully projecting a stream of abrasive particles onto a surface, usually with compressed air or steam. Because sand was commonly used in this process, workers who perform abrasive blasting are often known as sandblasters. Like the stone dusts, sands contain silica and may contain natural radionuclides that contribute to ionizing radiation exposure of the workers. Due to the difficulties of controlling the amount of silica in the ambient air, the use of sand for blast cleaning operations has been prohibited in several countries, including Brazil (NIOSH, 1992; Marchiori et al., 2006).

There are regions in the world with high concentration of natural radioactive elements in rocks and soil, and they are regarded as anomalous (Anjos et al., 2004; UNSCEAR, 1993, 2000). The amount of natural radiation existing in a given area depends on the types of minerals present in soils and rocks (Anjos et al., 2004, 2007). For example, monazite and zirconite are minerals concentrated in sands and soils that contain higher than normal levels of thorium and uranium (Anjos et al., 2006). Therefore, areas rich in these minerals usually have high environmental radiation levels (Veiga et al., 2006).

The average annual effective dose from natural and man-made sources for the world's population is currently about 2.8 mSv. Nearly 85% of this dose (2.4 mSv) is due to natural radiation, of which 0.5 mSv is attributable to terrestrial gamma-ray sources (UNSCEAR, 1993, 2000). Among the world regions with considerably higher levels of natural radiation we may cite Yangjiang (China, 5.5 mSv y^{-1}) and the coastal belt of Kerala (India, with levels from 1.5 to 20 mSv y^{-1}). In various beaches of the Brazilian Atlantic coast, like Pitinga, Caetité, Buena and Guarapari (Brazil) the radiation dose levels ranges from 3.5 to 10 mSv y^{-1}. In these regions there are high concentrations of monazite and zirconite in the soils and rocks, and consequently in the sand (Veiga et al., 2006; Anjos et al., 2008). As a result, the use of these sands for sandblasting can enhance, unnecessarily the risks associated with the exposure to radiation. The sand processed into fine particles in air, is inhaled, irradiating the lung tissues with alpha particles, and increasing the risk of lung cancer (UNSCEAR, 1993, 2000; Anjos et al., 2008).

Among the various types of ornamental stones available, granite and sandstone show the highest potential for exposure to silica. Besides that, granitic rocks have the highest natural radiation levels. Contents of ^{232}Th, ^{238}U and ^{40}K are also intimately related to their mineral compositions and general petrologic features (Anjos et al., 2005, 2006, 2007). Then, it is possible to presume that the granite workers would be the employees of the ornamental stone industries with the highest risks of developing lung diseases. In the case of sandblasters, several studies have revealed high risks for silicosis due to the difficulty in controlling exposures (NIOSH, 1992; Marchiori et al., 2006) and, in our opinion, besides the concentration of silica in the sand respirable fraction, the concentration of natural radionuclides in the sand, which varies according to where the sand was collected, should also be taken into account in the estimation of lung disease risks. For instance, inhalation of black sands, which are naturally rich in monazite, increases radiation risks.

Therefore, with the aim of studying the main features of the dust occupational exposure in the Brazilian granite extraction process and manufacture of granite products and to identify and quantify the dust distributions in this kind of work environment, samples in the respirable particulate fractions were collected in these environments. Particularly, this study also has the goal to evaluate an occupational risk rate, taking into account both the exposure to crystalline silica and to natural radionuclides. In fact the exposure to radiation may contribute to long term increase of lung cancer incidence of workers exposed to dust in mines and granite shops and it is not commonly investigated in occupational health works. This estimate was also performed in sandblasting activities because some small businesses circumvent the laws and still use sand in the blast cleaning operations.

2. Materials and methods

2.1. Sampling strategy and sample analysis

For this study, two granite mines from Castelo District (South Espírito Santo State, Brazil) and eight granite shops from the São Paulo City (São Paulo State, Brazil) were selected. Sand samples from regions rich in monazites, such as the Areia Preta Beach (Guarapari, South coast of Espírito Santo State, Brazil) and Buena Beach (São Francisco de Itabapoana, North coast of Rio de Janeiro State, Brazil), were collected and inserted in a sandblasting device in order to be processed into fine particles.

Castelo District was selected because in this area it is still possible to find employees using both dry and wet processes for cutting granitic rocks. Moreover, the first mine was chosen for extracting gray granites (petrographically classified as biotite monzogranite), with thorium contents around 50 ppm, a typical concentration among Brazilian Granites. On the other hand, the second mine produces yellow granites (hornblende-bearing monzogranitic gneiss) with lower Th concentrations, around 5 ppm (Anjos et al., 2005). Eight male rock drillers participated in this study. Fig. 1 shows workers using pneumatic rock drill machines during the extraction of granites. With the dry drilling process (Fig. 1a), the rock driller works in the dust atmosphere created by airborne particles of silica. Similar problem occurs in granite shops, mainly in the finishing sectors. Despite the peculiar features of each of the granite product manufacturers, the activities and procedures applied in their workplaces were very similar. Each granite shop had on the average about nine male employees working in the cut and finishing sectors, of whom around three worked with wet sawing devices. The full shift of the rock drillers and granite shop workers was 8 h per day or 40 h per week. Each mine and granite shop was analyzed during 70% of both daily and weekly employee's full shift. Thus, granite drillers and granite shop workers were sampled to determine their time-weighted average (TWA) exposure to crystalline silica. Dust samplers were distributed to different groups of workers using the method proposed by Leidel et al. (1977) as a guideline. About 100 personal samples of airborne dust were collected in the breathing zones of the employees (clipped to the workers' lapels) during granite drilling and stone manufacturing operations. Forty stationary samples were collected near the workers at some work stations. These area samples were installed in common circulation places and close to the main dust sources, to evaluate the environmental concentration as an indicator of the indirect dust exposure of workers and the efficiency of existing dust control systems. Dust samples deposited in the ground were also collected in order to determine their particle size

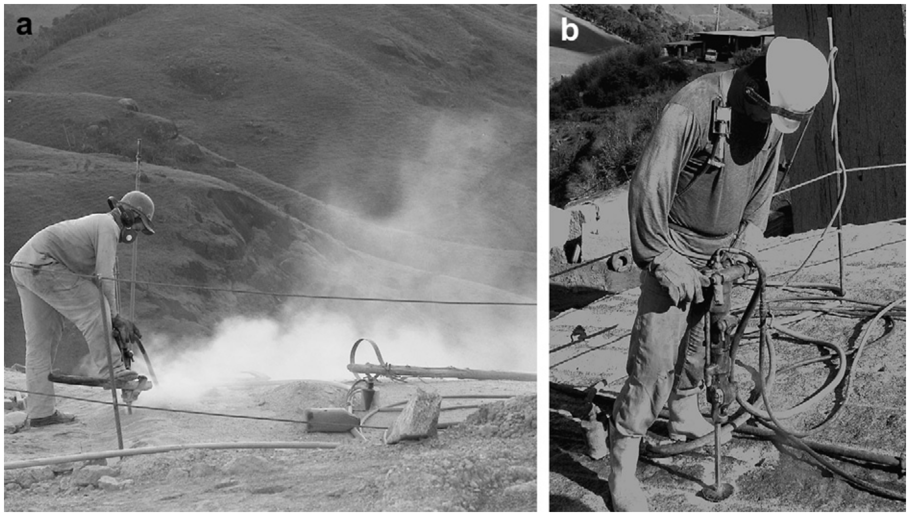

Fig. 1. Workers using pneumatic rock drills to extract granite blocks: (a) a dusty atmosphere is created by dry drillings; (b) the dust is practically eliminated in the wet process.

distributions and the respective concentrations of ^{238}U and ^{232}Th. Natural U and Th are the long lived parents of radioactive series of daughter radionuclides – part of them decay through alpha particle emission, which is of concern regarding internal exposure.

Personal and area respirable airborne dust samples were collected on 5 μm pore size, 37 mm PVC filters, using size-selective samplers designed so that the respirable dust fraction meets the 4 μm (50%) cutoff criteria (ACGIH, 1993–1994) with constant-flow personal sampling pumps. The filters were weighed before and after sampling and the dust concentration was determined using the difference in filter weights, after adjustment for blanks, and sampling volume. The results were expressed as $mg\,m^{-3}$. For the respirable dust sampler the detection limit was 0.05 mg. The free crystalline silica contents were determined as α-quartz by X-ray diffraction (XRD) of the respirable fraction of each sample (NIOSH, 1994; Santos, 1989). Measurements were performed in the XRD laboratory of Fundacentro. The lower limit of detection (LLD) for quartz was 0.005 mg.

The experimental uncertainties from the concentration measurement process were estimated taking into account basically the uncertainties in mass determination and in the sampled air volume. The maximum relative errors were: 5% for the personal sampling pumps, from 4% to 6% for the mass obtained by gravimetry and 5% for the silica mass obtained by XRD. The final uncertainties of the concentration values are estimated as 10% for respirable fractions and silica contents.

Dust samples from the granite cutting and the sandblasting were measured by the conventional technique of gamma spectrometry, using a 55% efficiency high-purity Germanium detector (HPGe), in order to obtain the concentrations of ^{238}U and ^{232}Th. Samples were prepared and analyzed at the Laboratory of Radioecology (LARA) of the Physics Institute of the Federal Fluminense University. The samples were oven dried at 110 °C for about 24 h (time required for the samples to reach a constant mass) and sieved in mesh screens. Different mesh sizes were used to evaluate the effects of grain size on the radionuclides concentration. The samples were packed into radon impermeable cylindrical plastic containers, dry-weighed and kept sealed during four weeks, in order to reach the equilibrium of the ^{238}U and ^{232}Th series and their respective progeny. About 100 g of sample material was used for individual sample measurements.

Energy spectra from each sample were accumulated during 4–24 h, depending on the heavy mineral concentrations present in the sample. The determination of ^{238}U and ^{232}Th were based on the measurements of photopeaks from ^{214}Bi (1764.5 keV) and ^{208}Tl (2614.4 keV), respectively. Technical details of sample preparation and gamma-analysis can be obtained elsewhere (Anjos et al, 2004, 2005, 2006, 2007, 2008). The lower limits of detection (LLD) for ^{232}Th and ^{238}U were estimated to be 4.0 $Bq\,kg^{-1}$ and 2.5 $Bq\,kg^{-1}$, respectively.

2.2. Data analysis

2.2.1. Characterizing occupational exposure

In this study, characterization of the occupational exposure was based on the method recommended by the American Industrial Hygiene Association (AIHA) Exposure Assessment Strategies Committee (Mulhausen and Damiano, 1998). Therefore, the log-normality, the average exposure level and its corresponding 95% confidence interval for each selected exposure group were calculated. The log-normality of the exposure profile for each exposure group was examined by using the *W*-test as suggested by Gilbert (1987). The geometric mean was used to describe the average exposure for a given exposure profile, since the value is directly related to its average and cumulative doses (Leidel et al., 1977; Hewett and Ganser, 1997). Traditional statistical procedures that consider variations in exposure concentrations caused by sampling, analysis, and environment were used to estimate the geometric mean (GM) and the resultant value was used to compare with the selected occupational exposure limit. These methods are suitable for sample sizes from 5 to 500 with geometric standard deviations (GSDs) from 2 to 5. Detailed calculating procedures for both GM and its 95% confidence interval have been described by Leidel et al. (1977) and Hewett and Ganser (1997). For each exposure profile, the point of estimate for the fraction of exposures exceeding the selected occupational exposure limit was calculated according to the method suggested by Leidel et al. (1977).

2.2.2. Doses to workers from intake of natural radionuclides

The Human Respiratory Tract Model (HRTM) for Radiological Protection (ICRP, 1994a,b) has been applied to assess the individual committed effective dose for the workers belonging to the selected exposure groups. The HRTM constitutes a comprehensive revision and updating of previous lung model developed by the International Commission on Radiological Protection (ICRP) that assesses organ doses and effective dose from intake of radionuclides. Two routes of intake are considered: ingestion and inhalation (Valentin, 2003). In this model, a new respiratory tract is proposed and the gastrointestinal tract is based on the biological model developed by Eve (1966). The respiratory tract is represented by five regions: **ET1**, Posterior Nasal Passage; **ET2**: Larynx; **BB**: Trachea and Main Bronchi; **bb**: Bronchioles; and **Al**: Alveolar duct and alveoli.

In this study, only the inhalation intake route was considered. The model evaluates the fraction of the inhaled particles deposited in each region, for aerosol sizes ranged 0.6–100 μm, requiring knowledge of the aerosol characteristics (geometric standard deviation, Activity Median Aerodynamic Diameter – AMAD) and the absorption of the material from lung to blood. Inhalation dose coefficients were assessed using the AMAD value of 5 μm. For absorption, three classes of absorption of the radionuclide in the body are recommended, according to whether the absorption is considered to be fast (Type F), moderate (Type M) or slow (Type S). Additionally the model considers several routes of clearance from the respiratory tract involving three processes: (a) To the environment: material deposited in ET1 is removed by extrinsic means such as nose-blowing; (b) Internal particle transport: movement of particles towards the gastrointestinal tract and lymph nodes; and (c) Absorption: particles are absorbed by body fluids. For each processes the model propounds clearance kinetics expressed in terms of fractional clearance rates.

In this study the HRTM parameter values to evaluate the committed effective doses were chosen considering the particular situation of Brazilian workers exposed to inhalation of natural radionuclides ^{238}U and ^{232}Th due to their activities in the granitic industries or as sandblasters. Although concentrations of ^{40}K can be high in dust and sand (even higher than those of thorium and uranium), its contribution to the committed dose by inhalation is usually negligible due to the characteristics of this radionuclide. ICRP inhalation dose coefficients for ^{40}K are three orders of magnitude smaller than for ^{238}U and four orders of magnitude smaller than for ^{232}Th (ICRP, 1994a,b). According to ICRP (1995a,b) recommendations, the worker exposure period was considered to be 6.5 h per day, 240 days per year. Type M behaviour is assumed as a default for environmental exposure of members of the public to radioisotopes in particulate form. Exceptions are made in a few cases, including thorium, where experimental data have indicated that many of the principal forms of the element likely to be encountered exhibit other behaviour characteristics: the analysis of tissues from a few workers at autopsy 30 years post-exposure showed a excess concentration of thorium in lung and lymph nodes, indicating Type S behaviour (ICRP, 1995b). Thus, for uranium, a default Type M for the absorption was assumed, and for thorium the slow absorption was considered.

2.2.3. *Cumulative risk estimates*

In the last years, a number of assumptions have been proposed by several authors to determine the relationship between silicosis and cumulative exposure to silica dust to workers (Muir et al., 1989). The same arguments are relevant to perform a statistical analysis and estimates of cancer incidence and fatality risks to workers exposed to natural radionuclides. Silicosis is treated as a disease that is acquired through exposure to silica dust. Similarly, tissues of the lung irradiated by alpha particles emitted by the radionuclides of Uranium and Thorium series deposited in the respiratory track have an increased risk of acquiring lung cancer. In both cases, there is no risk if there is no exposure. Moreover, in both cases, the risk does not cease if the exposure ceases: silicosis may progress, and the lung cancer may occur years after the initial exposure. Finally, it is the accumulation of silica or natural radionuclides over the years, and not the rate of exposure to these materials that is the important factor for the risk evaluation.

3. Results

3.1. *Exposure profiles for workers exposed to crystalline silica*

Table 1 shows exposure profiles of the respirable dust for the three selected exposure groups or environmental workplaces in granite shops. The geometric mean concentration values of the respirable dust exposures are larger in the dry finishing sector than in the area samples of the saw sector. Minimum and maximum concentrations show a large variability, confirming the *in situ* observation that workers of different exposure groups were subject to respirable dust with different exposure levels. Dust concentrations vary depending on several factors, such as the kind of task executed, the number of workers operating the grinders, the proximity between the workbenches, the working rhythm, ventilation, the relative position of the exhausting system (when there was one) and the dust sources, the proportion of wet tasks executed, and the size of the granite shop. Table 1 also shows exposure profiles of the respirable dust for the three selected exposure groups or environmental workplaces in granite mines. The existence of natural ventilation (the work activities are performed outdoors) favors the dispersion of dust, leading to geometric mean concentration values of the respirable dust exposures in the personal samples lower than in granite shops. Furthermore, the use of the pneumatic wet rock drill machines, which supply water to moisten the contact surfaces during the granite extraction, reduces significantly the dust in the atmosphere, and consequently the respirable dust fractions in the corresponding personal samples. We also found that the exposure profiles of the respirable dust for the above six selected exposure groups were consistently in the form of a log–normal distribution with GSDs ranging from 2.3 to 2.8.

The respirable crystalline silica exposure profiles, displayed in Table 1, show also that the respirable quartz exposure levels are dependent on the ventilation and on drilling type (wet or dry). The exposure profiles of the respirable quartz for the five exposure groups were all log-normally distributed with GSDs ranging from 2.6 to 3.5. Table 1 shows that 88–98% of the respirable quartz exposures in the observed exposure groups exceeded the current

Table 1
Respirable exposure profiles to dust and crystalline silica for the three selected exposure groups in granite shops, and the two in the granite producers; fractions of workers' respirable dust exposures exceeding the current TLV–TWA of 3.0 mg m^{-3} for the selected exposure groups and; fractions of workers' respirable quartz exposures exceeding the current TLV–TWA of 0.025 mg m^{-3} for the selected exposure groups. *N* is the number of samples analyzed.

Site	Exposure groups (sample type)	*N*	Agent	Dust and crystalline silica measured concentration (mg m^{-3})		Fraction above TLV (%)
				GM	Range	
Granite shops	Dry finishing	40	Dust	2.7	0.4–29	45
	(Personal)	40	Silica	0.2	ND–1.2	98
	Circulation area	41	Dust	1.3	0.1–8.8	21
	(Area)	41	Silica	0.1	ND–1.2	88
	Wet activities	38	Dust	0.9	0.1–4.4	10
	(Personal)	38	Silica	0.1	ND–0.5	90
Granite mining	Dry activities	8	Dust	0.8	0.4–1.1	6.0
	(Personal)	8	Silica	0.1	ND–0.2	89
	Circulation area	8	Dust	1.3	0.1–2.8	19
	(Area)	8	Silica	0.1	ND–0.3	90
	Wet activities	6	Dust	0.10	0.06–0.13	0.5
	(Personal)	6	Silica	–	ND	0

ND = silica content below the lower limit of detection (LLD).
GM = geometric mean value.

time-weighted average threshold limit value (TLV–TWA = 0.025 mg m^{-3}) set by ACGIH (2006). The above results suggest that respirable quartz exposures of granite workers are quite severe.

The results obtained in this study show that workers in Brazilian granite shops are characteristically exposed to high concentrations of extremely fine dust. Such dust is produced mainly by the tools used during the activities of dry finishing of ornamental stones, but workers from wet sawing sectors are also exposed to the dust due to the working procedures adopted. Dust concentration in these workplaces varies within a wide interval of values, following a log–normal distribution. Among all the analyzed samples there are values of the respirable particle mass up to 10 times higher than the TLV of 3 mg m^{-3}. Another important result refers to the free crystalline silica concentration, which in some samples reaches values as high as 48 times the TLV of 0.025 mg m^{-3} (ACGIH, 2006).

Such picture demonstrates that the control measures actions adopted by the granite shops were insufficient or inefficient in the mitigation of dust and elimination of the occupational exposure to this agent. Considering that the majority of Brazilian granite shops show similar behaviour, but there are several others that do not adopt any collective control measure, it can be inferred that the dust exposure situation in Brazil, and mainly to silica, is probably even more serious that the identified in this study. Additionally, the results on the exposure of rock driller operators to respirable dust indicate that, even in activities that are performed outdoors, like the granite extraction, it is essential to introduce wet drillings to diminish the workers' exposure to dust and silica.

Thus, this work confirm the common sense alerts proposed by the National Institute for Occupational Safety and Health (NIOSH) and the Brazilian Program to Eliminate Silicosis, which request actions to prevent silicosis morbidity and mortality risks to workers exposed to respirable crystalline silica. It urges to inform workers and employers of the granite extraction and manufacturing activities about the respiratory hazards associated with drilling and finishing operations. It is also important to promote the modernization of the Brazilian granite business, stimulating the implementation of wet tools able to decrease the dust production, and of ventilation devices that favors the dispersion of dust.

3.2. *Exposure profiles for workers exposed to natural radionuclides*

Mean values of activity concentrations of ^{40}K, ^{232}Th and ^{238}U obtained from two different commercial granite types of Castelo District and superficial sand samples from Guarapari and Buena beaches are shown in Table 2. Youngest, felsic and potassic members of igneous rocks are generally rich in uranium and thorium, and their abundances increase from basic to acidic rocks (Anjos et al., 2005, 2006, 2007). This feature distinguishes different granitic rock formations, justifying the different activity concentration values of ^{232}Th (160 Bq kg^{-1} and 22 Bq kg^{-1}) and ^{238}U (51 Bq kg^{-1} and 16 Bq kg^{-1}) observed in gray and yellow granites, respectively.

The formation of sedimentary strata is associated with the breakdown of source rocks, followed by the transport of the terrigenous matter, its re-deposition and sedimentation. Therefore, beach sands are weathering-resistant remainders of geological formations, transported by wind, rivers, and glaciers to the coast, and deposited on the beaches by actions of waves and currents (Anjos et al., 2006, 2010). In this way, the low K-activity observed in the coastal zones (Buena and Guarapari Beaches) shows that its sediments do not retain memory of Brazilian pre-Cambrian formations. This fact is supported by Table 2, which shows that beach sand samples present values of potassium concentrations significantly lower than granite samples.

Table 2
Mean values of ^{232}Th and ^{238}U concentrations obtained from granite and sand samples of Castelo District and Guarapari and Buena beaches, respectively.

Sample	^{40}K (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{238}U (Bq kg^{-1})
Yellow granite (hornblende-bearing monzogranitic gneiss)	1573 (163)	22 (13)	16 (9)
Gray granite (biotite monzogranite)	1481 (145)	160 (60)	51 (25)
Buena Beach	60 (29)	$3.8 (1.4) \times 10^3$	$3.7 (1.2) \times 10^2$
Guarapari Beach	63 (32)	$55 (21) \times 10^3$	$4.0 (1.7) \times 10^3$

Values in parentheses represent the standard deviation of the mean ($N = 15$).

Sands are defined as mineral grains with diameters between 63 μm and 2 mm and may be divided into light (quartz and feldspars) and heavy mineral fractions. Usually, heavy minerals are found scattered on coastal deposits, but in some favorable circumstances, they can be found in high concentrations, suitable for exploitation. Sea level changes can contribute to the re-working of alluvial and colluvial deposits, through current and wave actions, and so this mechanism can promote the re-concentration of dense minerals such as monazite and ilmenite (Anjos et al., 2006, 2007). Guarapari and Buena Beaches are Brazilian examples of this phenomenon. According to Table 2, the activity concentration of ^{232}Th in Guarapari beach sand is about 2–3 orders of magnitude higher than in granites.

In order to evaluate the effect of particle size on the activity concentration of ^{40}K, ^{232}Th and ^{238}U, a few dust samples were separated in grain size fractions: granite dust samples from granite drilling and finishing activities and sand samples from Guarapari and Buena beaches, transformed into fine particles using a sandblasting machine, had the activity concentration analyzed according to grain size. The distributions of ^{40}K, ^{232}Th and ^{238}U as a function of grain size were obtained, and the results for all samples are consistent with heavy mineral isolation in the fine fraction, as the radionuclide concentration has similar behaviour for different granite types and for sands from different beaches. The results shows that the K content from samples of granite (or sand) remained unchanged, since its values varied only slightly with grain size. On the other hand, both U and Th contents increase exponentially with decreasing grain size.

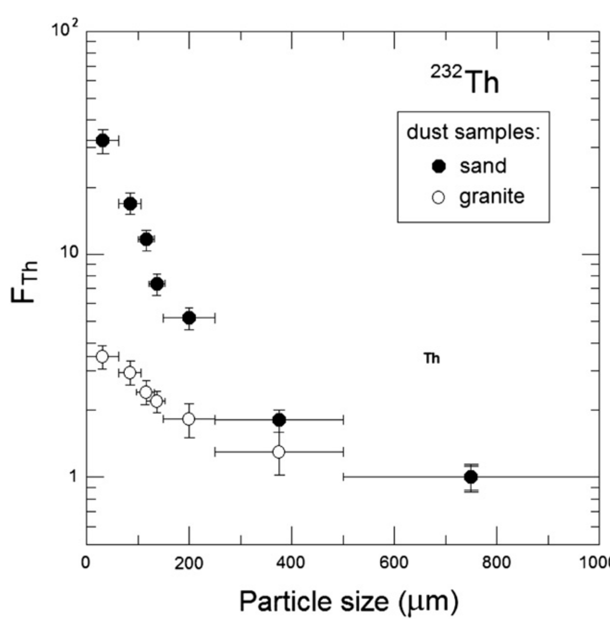

Fig. 2. Coefficient F_{Th} as a function of the particle size.

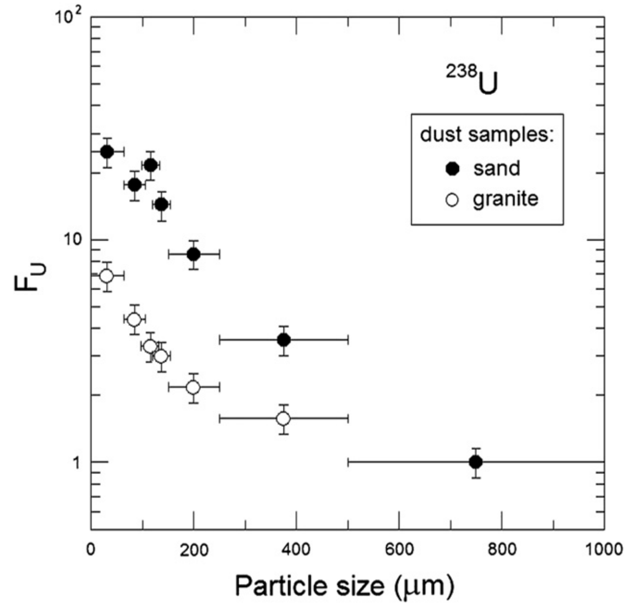

Fig. 3. Coefficient F_U as a function of the particle size.

With the aim of better understanding this behaviour, the values of ^{232}Th activity concentration were normalized by the higher grain size fraction value (500 μm ≤ φ ≤ 1000 μm), defining a coefficient (F_{Th}). This factor shows how many times the concentration of ^{232}Th for a given grain size fraction is higher than the concentration in grains with diameters between 500 and 1000 μm. Similarly F_U was obtained for the uranium relative concentrations. Figs. 2 and 3 show F_{Th} and F_U, respectively, as a function of particle diameter According to these graphs both U and Th contents increase exponentially with decreasing grain size. Nevertheless, this increase is much more pronounced for sand than granite samples, reaching relative concentrations one order of magnitude higher in the finest sand fractions. As the dust fraction deposited in the lungs correspond to the finer grains, these results suggest that natural radionuclides could present an additional respiratory hazard to workers, depending on the amount of particles deposited (^{232}Th and ^{238}U activities) in the lung tissues over the years.

Table 3 shows the parameter values of Human Respiratory Tract Model for Radiological Protection used in this work to calculate the committed effective doses due to the intake of ^{238}U and ^{232}Th from dust in a time period of one year (ICRP, 1995a). This table also includes an estimate of the inhaled dust (mg m^{-3}) for each one of the selected groups in different work sectors of granite shops and granite producers in terms of working time. This amount, multiplied by the daily volume inhaled (m^3) and by the dust activity concentration (kBq kg^{-1}) gives the activity inhaled per day. The calculations for sandblasting activities were developed using the same configuration of the dry finishing sectors in the granite shops. Table 4 shows values of the committed effective dose for adults (age ≥ 25 years), which corresponds to the effective dose that would be received by the worker during a 50-year period after the intake. For the granite samples the ^{40}K concentrations are at maximum two orders of magnitude higher than the thorium or uranium ones (Table 2), giving rise to negligible contributions to the committed effective doses, as previously mentioned (ICRP, 1994a, b). This way, they were not included in the calculations. From this table, one can notice that, after the first year of work, the values of committed effective doses for ^{238}U and ^{232}Th from granite samples are very low to produce additional damage to the lungs of the workers in the Brazilian granitic industries. For illustration, the mean effective dose received by the world population due to

Table 4
Values of committed effective dose (CED) from intake of ^{238}U and ^{232}Th during a period of one year. These values are estimated for adults (age ≥ 25 years), in which are computed for a 50-year period after intake.

Environment	Area	Granite type	CED [^{238}U, Type M] (mSv)	CED [^{232}Th, Type S] (mSv)
Granite shop	Dry finishing	Yellow	0.36×10^{-3}	6.75×10^{-3}
		Gray	0.92×10^{-3}	21.0×10^{-3}
	Circulation area	Yellow	0.18×10^{-3}	3.25×10^{-3}
		Gray	0.44×10^{-3}	10.1×10^{-3}
	Wet activities	Yellow	0.12×10^{-3}	2.25×10^{-3}
		Gray	0.31×10^{-3}	7.00×10^{-3}
Granite producers	Dry finishing	Yellow	0.11×10^{-3}	2.00×10^{-3}
		Gray	0.27×10^{-3}	6.22×10^{-3}
	Circulation area	Yellow	0.18×10^{-3}	3.25×10^{-3}
		Gray	0.44×10^{-3}	10.1×10^{-3}
	Wet activities	Yellow	0.01×10^{-3}	0.25×10^{-3}
		Gray	0.03×10^{-3}	0.78×10^{-3}
Sandblasting	Sand from Buena Beach		0.108	1.13
	Sand from Guarapari Beach		0.116	16.4

Table 3
Input parameter values of Human Respiratory Tract Model (HRTM) for Radiological Protection used in this work.

Parameters			^{238}U	^{232}Th
f_1: gastrointestinal uptake factor			0.02 (Type M)	5×10^{-4} (Type S)
Ventilation parameters: light exercise – adult male	f_R: breathing frequency (min^{-1})		20	20
	B: ventilation rate (m^3 h^{-1})		1.5	1.5
Daily time budget (h)			6.5	6.5
Daily air volume inhaled (m^3)			9.75	9.75
Inhaled dust (mg m^{-3})	Granite shop	Dry finishing	2.7	2.7
		Circulation area	1.3	1.3
		Wet activities	0.9	0.9
	Granite producers	Dry activities	0.8	0.8
		Circulation area	1.3	1.3
		Wet activities	0.1	0.1
	Sandblasting		2.7	2.7
Radionuclide concentration in the inhaled dust (kBq kg^{-1})	Granite	Yellow	0.036 (0.012)	0.090 (0.050)
		Gray	0.090 (0.045)	0.28 (0.10)
	Sand	Buena Beach	10.7 (2.9)	14.9 (5.5)
		Guarapari Beach	11.5 (4.9)	216 (82)

Values in parenthesis represent the standard deviation of the mean.

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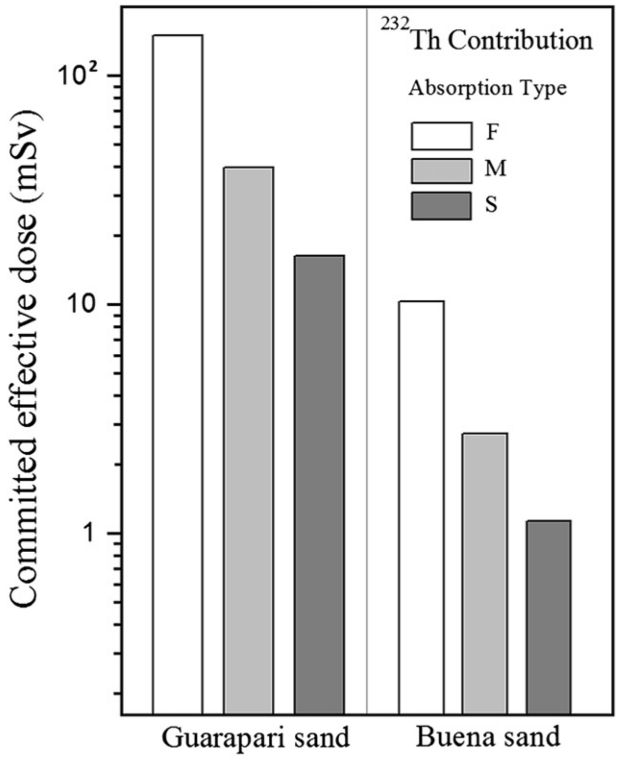

Fig. 4. Committed effective dose behaviour for sandblasting with sand from Guarapari and Buena beaches according to whether the absorption is considered to be fast (Type F), moderate (M) or slow (S) for ^{232}Th, according to the HRTM with the parameters shown in Table 4.

natural sources of radiation in the earth is about 2.4 mSv per year or 144 mSv in the 50-year period (UNSCEAR, 1993, 2000).

Studies on disease caused by inhalation of crystalline silica dust have shown that the mean time between the beginning of the exposure and the first proven profusion and conglomeration is 7 years (ranging between 4 and 10 years). Most of the patients used to work without breathing-mask, usually in orthostatic position. All patients presented conglomerate masses involving the upper lobes, and in about 90% of the cases the lesions were located in the posterior region of the lungs (Marchiori et al., 2006). Assuming, then, the worst situation of Table 4, which would be an employee of the finishing sector of granite, after a period of 10 years of continuous employment, this worker would have received a total committed effective dose of about 0.2 mSv. Even if there was a rapid absorption of the thorium into the body, the total committed effective dose does not exceed 2 mSv. According to ICRP (2008) there is, however, general agreement that epidemiological methods used for the estimation of cancer risk do not have the power to directly reveal cancer risks in the dose range up to around 100 mSv. Thus, the results suggest that the risk of radiation-induced cancer in the granite industries is negligible, being, then, the respirable crystalline silica the main causative agent of lung damage.

Studies about workers exposed to respirable crystalline silica during sandblasting have presented a higher risk for silicosis than rock drillers or finishing workers of granites due to the difficulty in controlling exposures (NIOSH, 1992). Additionally, the respirable-size freshly ground silica produced contains silicon-based radical that react with aqueous environments to produce OH radicals and this feature may lead to enhancement manifestations of lung injury (Vallyathan et al., 1988). However, this problem might become worse if natural radionuclides are introduced in the lung tissues. According to Table 4, a sandblaster who used sand from Buena Beach would receive a total committed effective dose of about 12 mSv after 10 years of continuous activity with the parameters shown in Table 3. The use of sand from Guarapari for sandblasting would give rise to higher doses: 10 years of exposure to natural radionuclides could result in 170 mSv in the 60 years of life following the first intake. If we take into account the fact that the distribution of heavy minerals in beach sands is not homogeneous and there is, therefore, places where concentrations of thorium and uranium are about seven times higher than their mean values observed, the total committed effective dose from blasting operations could reach values of about 1.2 Sv. According to ICRP (2008) the nominal risk of lung cancer (adjusted for lethality and quality of life) would be 126 cases per 10,000 persons per Sv for working age population (18–64 years).

This is an interesting fact, because the combination of radiation exposure and exposure to dust is quite common in industrial settings and in the environment, and these agents are reported in both animal studies and in vitro studies to act synergistically at high exposures (UNSCEAR, 1993, 2000). In this work, contributions of radon gas (a radioactive noble gas product of ^{238}U and ^{232}Th decays), which are present mainly in the indoor atmosphere, are not included. In fact, radon gas is a particularly significant source of natural radiation, being responsible for half of the natural effective dose to the population, and the determination of concentration of this gas in the granite shop and sandblaster ambient should be performed.

The parameters used to evaluate the committed effective doses, including the inhalation dose coefficients are important terms in the model. The HRTM assumes, by default, that particle transport rates are the same for all materials, and that the absorption rates are constant. It is recommended that material-specific rates of absorption are used whenever reliable human or animal experimental data exist. Even though the ICRP recommend to use absorption type S absorption rate for ^{232}Th, there are works in literature (Frelon et al., 2007) that have indicated type M thorium absorption for internal exposure to black sand (such as sands from Guarapari and Buena Beaches). This change would mean a considerable enhancement in the value of the committed effective dose. Fig. 4 illustrates the results for CED in sandblasting according to whether the absorption is considered to be fast (Type F), moderate (M) or slow (S) for ^{232}Th. Thus, further radiobiological research is necessary in order to better understand the chronic exposure to dust containing natural radionuclides. Besides that, we also believe that it is necessary to obtain experimental data about radon gas concentrations in the work environment of granite manufacturing.

4. Conclusions

The results obtained from this study have confirmed that workers of Brazilian granitic industries are characteristically exposed to high concentrations of extremely fine dust. Such dust is produced mainly by the activities of rock drilling and of dry finishing of ornamental stones. Workers from wet sawing sectors are also exposed to the dust due to the working procedures adopted, although in lower quantities. On the other hand, our results suggest that the risk of radiation-induced cancer in the granite industries is negligible, being, the respirable crystalline silica the main causative agent of lung damage. In this way, this risk in the other industries of ornamental stones (such as marbles, limestone, quartz-based stones, slates and basalts) would be even lower, since the concentration of natural radionuclides is even lower.

Concerning the sandblasters, as this work has shown that the occupational exposures to silica and natural radionuclides can be high, special attention must be taken in order to avoid potential risk

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enhancement: it is very important to avoid the use of sand for abrasive blasting activities. Besides the known risk of inhaling crystalline silica, the source of sand is also a very important factor because it can have high concentrations of radioactive minerals. Indeed, the combined exposure to both agents may increase the risks.

Like the Organization for Economic Co-operation and Development (OECD, 1979) proposes limits for the safe use of building materials taking into account the radiation hazards in urban environments, it is important to give some quantitative estimation of the maximum allowable activity concentrations of ^{232}Th and ^{238}U in the materials used for blasting operations or in granitic industries. This recommendation would be extremely useful for experts dealing with legislation issues. The recommendation of the Brazilian National Nuclear Energy Commission (CNEN, 2005) waives the requirements of radiological protection for materials containing concentrations of thorium and uranium higher than 1 and 10 Bq g^{-1}, respectively. According to our calculations, the work with materials with such activity concentrations would result in a CED of 0.6 mSv to the sandblaster per year of work. Higher risks should not be accepted, and the use of materials with activities higher than the exemption values must be avoided in blasting operations.

Additionally, it is possible to notice the urgent necessity of modernization of the Brazilian granitic industries. Nevertheless, such modernization should occur with the aggregation of technological capabilities for the execution of each specific task, and the implementation of tools that decrease the dust production. The concern with health and security should not be just a legal exigency, but the granitic industries might understand that, even in a small business, they could make a difference by improving the working conditions of their stores.

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