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# Assessment of occupational exposure in a granite quarry and processing factory

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## Abstract

Workers in the granite industry face an occupational hazard: silicosis due to the crystalline silica present in inhalable dust. As granite can also present a variable, and occasionally significant, content of naturally occurring radionuclides, they may also face a radiological hazard. In order to assess the risk, a granite industry with a quarry and processing factory was selected to assess the occupational exposure. Three main potential pathways were observed: external irradiation, inhalation of granite dust, and radon exposure. The external dose rate was similar to that in a nearby farming area. A slight increment ( $0.016\text{--}0.076\text{ mSv yr}^{-1}$ ) was observed in the quarry and stockpile, due to quarry faces and granite blocks. The effective dose due to granite dust inhalation was  $0.182 \pm 0.009\text{ mSv yr}^{-1}$  in the worst case scenario ( $3\text{ mg m}^{-3}$  dust load in air and no use of filter masks). Thus, the mean value of the effective dose from these two pathways was  $0.26\text{ mSv yr}^{-1}$ , lower than the reference level of  $1\text{ mSv yr}^{-1}$  for the general population. The annual mean value of radon concentration in the indoor air was  $33\text{ Bq m}^{-3}$ . However, during granite processing works the radon concentration can increase up to  $216\text{ Bq m}^{-3}$ , due to mechanical operations (sawing, polishing, sanding, etc). This radon concentration was below the  $600\text{ Bq m}^{-3}$  reference level for action in working places. Therefore, workers in this granite factory face no significant additional radiological exposure, and no-one needs to be designated as occupationally exposed and subject to individual dosimetry.

Keywords: granite, dose, inhalation, dust, NORM, radon, worker

(Some figures may appear in colour only in the online journal)

## 1. Introduction

Workers in the granite industry face an important health and safety hazard. The procedures for extracting and processing granite can generate a large quantity of granite dust in the workers' environment. The inhalation of this dust poses a hazard due to its high crystalline silica content, which, when deposited in the lungs, gives rise to silicosis. Crystalline silica has also been classified as a human carcinogen by the Agency for Research on Cancer and is related to lung cancer [15]. Due to this hazard, there is legislation regulating the load of dust in indoor air, in particular the inhalable fraction ( $\text{Ø} < 50 \mu\text{m}$ ). In the case of Spain, the environmental limit value for daily exposure to inhalable dust is  $3 \text{ mg m}^{-3}$ , and for the free silica content, lower than  $0.1 \text{ mg m}^{-3}$ .

The ornamental rock industry, and in particular that of granite, can be considered as a potential NORM (naturally occurring radioactive materials) activity, since naturally occurring content in granites can be variable and occasionally higher than the average content of other rocks and soil [1, 3, 16]. Therefore, workers can be exposed to an additional health hazard, related to the occurrence of naturally occurring radionuclides. This exposure also includes a potential radon hazard, as part of the natural decay series of uranium, due to the exhalation of radon in natural conditions [4, 11], or enhanced by mechanical processes such as compression [14]. The latter is of special relevance in the case of the granite industry, since these processes are commonly used in extracting granite from the quarry and later processing and shaping the granite blocks (cutting, shaping and finishing). In addition, as the inhalation of granite dust particles is involved in the development of silicosis in workers, it generates the question of whether an additional radiological risk is also involved.

The goal of the present study is to assess the occupational exposure in a granite quarry and processing industry. Therefore, a representative industry of one of the most important production areas in Spain was selected, which has a quarry and a processing factory. The extracted granite falls into the category of granite *s.s. (sensu stricti)* with an approximate content of 33% quartz, 34% alkali-feldspar, and 32% plagioclase [11]. Then, the different working locations, and the corresponding potential exposure pathways were identified. These pathways are external irradiation, granite dust inhalation, and radon exposure. The effective annual doses for external irradiation, dust inhalation, and radon concentration in indoor air were compared with reference values in the legislation to identify any possible health hazard.

## 2. Material and methods

### 2.1. Description of the granite quarry and processing factory

The granite quarry was located in Quintana de la Serena (Badajoz, Spain), which is one of the most important granite extraction and processor centres in Spain. The selected company is a perfect representative of this industrial sector in the region. It possesses a quarry and a processing factory, in which the work is carried out using different machines and diamond tools, but also ornamental stone handcraft is done with manual tools. In the quarry, granite blocks are extracted with diamond tools and hydraulic perforation, thus reducing the quantity of dust and silica exposure to workers. This factory had 24 employees at the time of the study, working 40h per week. Each worker carried out specific work, although they could be assigned to the quarry or the processing factory according to production requirements. Table 1 lists the main working environments observed in the factory and their placement, either indoors or outdoors.

**Table 1.** Classification of the different work environments observed in the granite factory, their emplacement (indoors/outdoors), and potential exposures identified in each of them.

Work environment	Emplacement	Potential exposure		
		External	Internal (dust)	Radon
Office	Indoors	X		X
Quarry	Outdoor	X		
Processing factory	Indoors/outdoors <sup>a</sup>	X	X	X
Handcraft workshop	Outdoors	X	X	

<sup>a</sup>The processing factory is a module with big doors to allow the passing of heavy machinery, and the doors are open during workdays.

## 2.2. External exposure

The external irradiation dose rate was determined using a dose rate meter monitor, FAG FHZ600A. The monitor comprises a 54.2 cm<sup>3</sup> pressurized proportional counter, designed to measure dose rates in the range 0.005  $\mu\text{Sv h}^{-1}$ –1 mSv h<sup>-1</sup>. The monitor was calibrated using the ‘shadow field’ technique, which is a slight modification of the ‘free field’ technique. It uses <sup>60</sup>Co and <sup>137</sup>Cs sources of certified activity in terms of a primary reference in air kerma rate. The detector and the source were placed at a height of 1 m above ground and collimated to allow only the primary beam to reach the detector. Calibration was performed for different source-to-detector distances [9]. The mean value of the associated uncertainty for dose rate measurement with this monitor was about 5%. The monitor was placed 1 m above soil and at least 1 m from other surfaces.

## 2.3. Radionuclide determination

To determine the radionuclide content in resuspendable dust, about 200 g of dust was collected from the dust extraction systems in the factory. They were sieved using air current, and a fraction lower than 45  $\mu\text{m}$  was collected, about 63% of the bulk material. Aliquots of granite samples were ground to particle sizes between 0.1 and 0.25 mm. Then they were dried at 100 °C for at least 48 h to remove moisture.

The <sup>40</sup>K and <sup>226</sup>Ra in the aerosol sample was determined by  $\gamma$ -spectrometry. An aliquot of 150 g was placed into 191 cm<sup>3</sup> Petri-type capsules and sealed to avoid loss of any <sup>222</sup>Rn emanations. After 28 d, to allow <sup>226</sup>Ra to reach secular equilibrium with its descendants (<sup>214</sup>Bi and <sup>214</sup>Pb), the samples were assayed by gamma spectrometry using an HpGe detector of 43% relative efficiency.

An aliquot of 2 g was acid digested with a mixture of HNO<sub>3</sub>, HCl, and HF (9:3:6 ml) in a microwave oven (Ethos Pro Milestone Ltd) at 180 °C for 20 min prior to the corresponding radiochemical procedure. After digestion, the samples were evaporated to dryness, and H<sub>3</sub>BO<sub>3</sub> was added to eliminate fluorides. To determine the uranium and thorium content, <sup>232</sup>U and <sup>229</sup>Th were first added as tracers. Then, the uranium and thorium content was co-precipitated with Fe(OH)<sub>3</sub>. The precipitate was re-dissolved in HCl 9M, followed by separation in a column with Dowex 1  $\times$  4 resin. Thorium was not retained in the column and was collected. The uranium was retained in the column, and subsequently eluted with HNO<sub>3</sub> 8M. Thorium and uranium fractions were evaporated and converted to a HCl medium. The alpha sources for these two radionuclides were prepared by co-precipitation with NdF<sub>3</sub> [17].



**Figure 1.** External dose rate in winter/summer, expressed in  $\mu\text{Sv h}^{-1}$ , at the quarry.

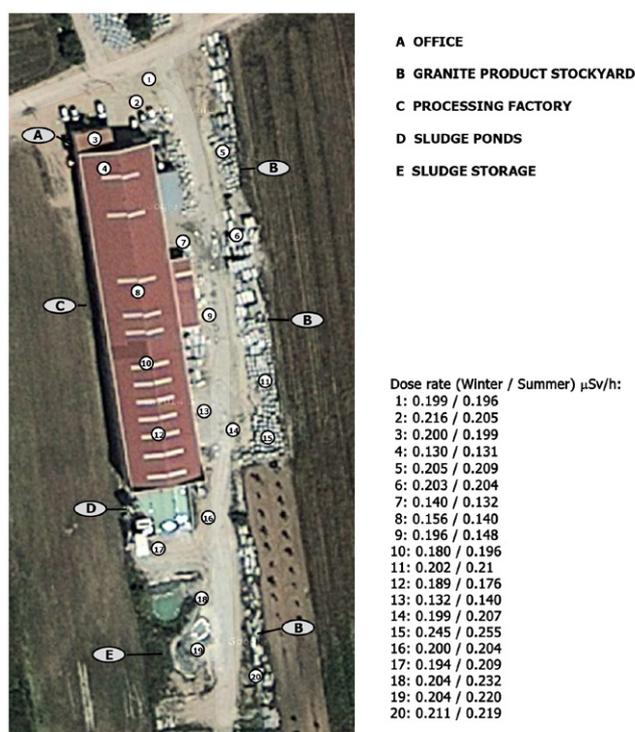
For  $^{210}\text{Po}$ , an aliquot of 1 g d.w. of sediment was digested with  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$  (8:2 ml) in a microwave oven at 180 °C for 20 min. The sample was filtered and converted to a HCl medium.  $^{209}\text{Po}$  was used as tracer. The polonium content was co-precipitated with  $\text{Fe}(\text{OH})_3$ . Then the precipitate was dissolved in HCl 8 M and diluted to HCl 1.5 M. The polonium was autodeposited onto silver planchets, and measured by alpha spectrometry [8].

Alpha spectrometries of uranium, thorium, and polonium samples were carried out using twelve different silicon detectors with a mean efficiency of 23.2% and a resolution of 38.7 keV for a source–detector distance of 6 mm. Associated uncertainties were estimated using the  $2\sigma$  criterion, and they accounted for measurement uncertainty, chemical recovery and calibration efficiency.

The overall quality control of these radiochemical procedures is guaranteed by the accreditation of the laboratory to carry out radioactivity assays in environmental samples according to UNE-EN ISO/IEC 17025 (ISO 2005). Different reference materials were also used to check the quality of the measurements: IAEA-381 for uranium, IAEA Soil 6 for  $\gamma$ -spectrometry, and IAEA-327 for thorium and polonium.

#### 2.4. Radon determination

The radon content was determined using charcoal canisters according to the EPA procedure [10]. Each month two canisters were placed in each location and exposed for about 2 d, recording start and end times. In the office area (first floor), they were placed on a filing cabinet, and in the factory processing area (groundfloor) on a shelf in a store room, as it was the only closed room in the area. Then, they were sealed and transported to the laboratory (3 h after end of exposure) to be measured by  $\gamma$ -spectrometry using the equipment previously described. The  $^{214}\text{Pb}$  (295.22 and 351.93 keV) and  $^{214}\text{Bi}$  (609.31 keV) were systematically analysed. The calibration of the canister geometry was carried out by spiking three blank canisters with a



**Figure 2.** External dose rate in winter/summer, expressed in  $\mu\text{Sv h}^{-1}$ , in the processing factory area.

known amount of  $^{226}\text{Ra}$  in each of them. Then, they were sealed, and efficiency determined one month later. The validity of the results was checked using a reference canister, reproducing its reference value. This measurement technique using passive detectors gives the average radon content for the exposure time (2 d, and nights, when the quarry was closed). However, some works in the quarry involved sawing, hammering, etc, which generates a lot of dust. Therefore, at two different times, the radon content was determined using an active detector, AlphaGuard, for short time measurements (10 min) in working conditions. In order to compare the radon results from these two techniques, a large amount ( $\sim 69$  kg) of granite from this quarry was placed in plastic container with airtight closure, and left alone for a month. Then, two canisters and the Alphaguard were placed inside the container for 2 d. The ratio between the average radon measurement from the canisters and the Alphaguard was  $0.87 \pm 0.11$  in this setup under closed conditions.

### 3. Results and discussion

#### 3.1. Assessment of external exposure

In order to assess properly the external dose to workers, the external dose rate was measured at several locations: (i) outside the granitic batholith; (ii) inside the granitic batholith, but in a farming area not exploited by the granite industry; (iii) different working areas within the selected factory (office, quarry, processing factory, handcraft workshop, and stockpile area). The external dose rate in each of them was measured at two different times, summer and



**Figure 3.** External dose rate in winter/summer, expressed in  $\mu\text{Sv h}^{-1}$ , in the handcraft workshop and stockpile area.

**Table 2.** Mean value, standard deviation and range of variation of external dose rate, expressed in  $\mu\text{Sv h}^{-1}$ , in the different locations in the granite factory and outside it.

Location	External dose rate ( $\mu\text{Sv h}^{-1}$ )	
	Winter	Summer
Outside batholith area	$0.15 \pm 0.06$ (0.152–0.160)	$0.170 \pm 0.08$ (0.164–0.175)
Inside batholith (farming area)	$0.203 \pm 0.020$ (0.188–0.218)	$0.20 \pm 0.03$ (0.184–0.221)
Office	$0.208 \pm 0.011$ (0.200–0.216)	$0.202 \pm 0.005$ (0.199–0.205)
Quarry area	$0.22 \pm 0.03$ (0.191–0.281)	$0.24 \pm 0.05$ (0.193–0.274)
Processing factory area	$0.17 \pm 0.03$ (0.131–0.216)	$0.17 \pm 0.03$ (0.132–0.207)
Handcraft workshop area	$0.211 \pm 0.003$ (0.196–0.215)	$0.213 \pm 0.005$ (0.193–0.220)
Stockpile area	$0.23 \pm 0.05$ (0.199–0.352)	$0.24 \pm 0.05$ (0.204–0.358)

winter, reflecting the extremes of the Mediterranean climate in the area. Figures 1–3 show the mean values of the external dose rate (summer/winter) at different locations over the map of the factory. Table 2 lists the range of external dose rate in these locations. Moving from outside the granitic batholith into it slightly increased the dose rate, from 0.152–0.175  $\mu\text{Sv h}^{-1}$  up to 0.184–0.221  $\mu\text{Sv h}^{-1}$ . Worker exposure to external irradiation in the office, processing

**Table 3.** Naturally occurring radionuclide content in granite from the quarry and resuspendable dust (<45  $\mu\text{m}$ ), expressed in  $\text{Bq kg}^{-1}$ , dose coefficients for inhalation of radionuclides for members of the public,  $e$ , expressed as  $\text{Sv/Bq}$  [12], and the corresponding annual dose, expressed in  $\mu\text{Sv yr}^{-1}$ .

Radionuclide	Granite ( $\text{Bq kg}^{-1}$ )	Resuspendable dust ( $\text{Bq kg}^{-1}$ )	$e$ (Sv $\text{Bq}^{-1}$ )	$D$ ( $\mu\text{Sv yr}^{-1}$ )
$^{234}\text{U}$	$89 \pm 9$	$138 \pm 23$	$9.4 \cdot 10^{-6}$	$7.4 \pm 1.2$
$^{235}\text{U}$	$4.0 \pm 0.4$	$4.8 \pm 2.6$	$8.5 \cdot 10^{-6}$	$0.23 \pm 0.13$
$^{238}\text{U}$	$88 \pm 9$	$141 \pm 24$	$8.0 \cdot 10^{-6}$	$6.5 \pm 1.1$
$^{226}\text{Ra}$	$99 \pm 9$	$159 \pm 18$	$9.5 \cdot 10^{-6}$	$8.6 \pm 1.0$
$^{210}\text{Po}$	$98 \pm 12$	$119 \pm 24$	$4.3 \cdot 10^{-6}$	$2.9 \pm 0.6$
$^{210}\text{Pb}^{\text{a}}$	$98 \pm 12^{\text{a}}$	$119 \pm 24^{\text{a}}$	$5.6 \cdot 10^{-6}$	$3.8 \pm 0.8$
$^{228}\text{Th}$	$79 \pm 7$	$92 \pm 8$	$4.0 \cdot 10^{-5}$	$21.0 \pm 1.8$
$^{230}\text{Th}$	$98 \pm 9$	$135 \pm 11$	$1.0 \cdot 10^{-4}$	$77 \pm 6$
$^{232}\text{Th}$	$70 \pm 7$	$86 \pm 8$	$1.1 \cdot 10^{-4}$	$54 \pm 5$
$^{40}\text{K}$	$1174 \pm 15$	$1312 \pm 45$	$2.1 \cdot 10^{-9}$	$(1.58 \pm 0.05) \cdot 10^{-2}$

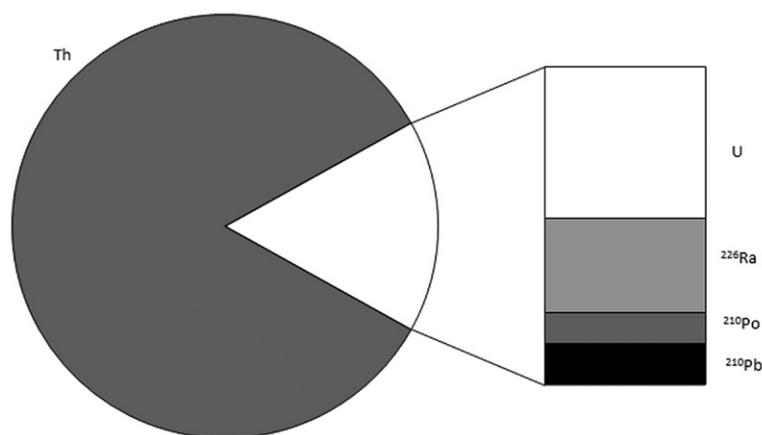
<sup>a</sup>Equilibrium between  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  was assumed.

factory and handcraft workshop were approximately the same as that found in the farming area on the batholith, which served as the background. The dose rate in the processing factory was lower than that observed in the farming area due to the presence of a concrete layer on top soil laid for supporting heavy machinery. The dose rate in the different locations within the quarry and factory were very close to those observed in the farming area outside it. The difference between the average dose rates in the nearby farming area and these working places was in the range  $0.008\text{--}0.037 \mu\text{Sv h}^{-1}$ . The annual effective dose, estimated assuming permanent residence during annual working hours (2080 h, assuming 40 working hours per week and 52 weeks), was  $0.016\text{--}0.076 \text{mSv yr}^{-1}$ , which is below the world mean value of  $2.4 \text{mSv yr}^{-1}$  for exposure to natural sources [18].

Only in some points in the granite block stockpile and close to the faces in the granite quarry, were higher values observed due to the presence of granite blocks and/or quarry faces surrounding the dose rate monitor. However, the presence of workers in those areas is very limited, as the granite walls are exploited to extract blocks, and workers only neared the block stockpile to select the one to process. In these two locations, heavy machinery is used to move and manipulate the granite blocks, which also serves as an extra protection for workers due to the extra distance provided between the operator and the granite blocks and shielding by the machine itself, as it poses additional material, and the driving place is closed, thus avoiding external dust. The mean values in quarry and stockpile areas were 10–18% higher than the background. However, this difference is not statistically significant as those values overlap when associated uncertainties are considered.

### 3.2. Assessment of internal exposure (dust inhalation)

The presence of dust in the processing factory and handcraft workshop is unavoidable, since granite blocks are processed in them. In the processing factory, the diamond tools used for sawing and cutting are refrigerated with water, reducing the amount of dust in the indoor air. However, in the handcraft workshop all processes involve the use of manual tools in dry conditions. Thus, the amount of dust is considerably higher. This is the reason why it is placed at a different location within the factory in an open module (no walls), so it can be



**Figure 4.** Contribution of the naturally occurring radionuclides to the annual dose due to dust inhalation. U and Th contributions are the sum of  $^{234,235,238}\text{U}$  and  $^{228,230,232}\text{Th}$ , respectively.

considered to be outdoors. According to health and safety recommendations, workers must use personal protection equipment, which is protection masks regarding exposure to dust. These mask are able to retain 99% of solid and liquid particles higher than  $1\ \mu\text{m}$  [2]. The occurrence of dust in indoor air is regulated by Spanish legislation, and is not allowed to be higher than  $3\ \text{mg m}^{-3}$  [6].

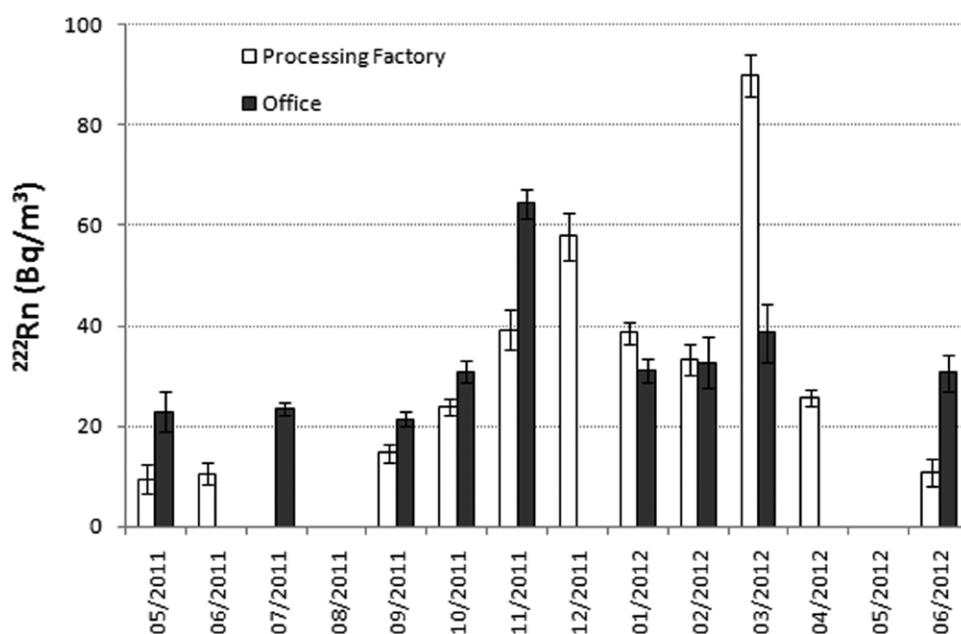
The radionuclide content of granite dust is related to the content in granite. Table 3 shows the naturally occurring radionuclide content of granite ( $0.1 < \varnothing < 0.25\ \text{mm}$ ) and resuspendable granite dust ( $\varnothing < 45\ \mu\text{m}$ ). The activity levels in granite dust are higher than those of granite. This can be attributed to particle size effect, since small particles usually present higher radionuclide content, probably due to a higher surface/volume ratio [5, 11]. Regarding the radionuclides in the different natural series, it can be observed they are in equilibrium in the granite and in its dust. The  $^{210}\text{Pb}$  content was not determined in the measurements, but it can be considered in equilibrium with  $^{210}\text{Po}$ .

The dose due to the inhalation of granite dust by workers was assessed using equation (1).

$$D\ (\text{mSv yr}^{-1}) = \sum_i A_i\ (\text{Bq kg}^{-1})e_i\ (\text{Sv Bq}^{-1})d\ (\text{kg m}^{-3})V\ (\text{m}^3\ \text{yr}^{-1}) \quad (1)$$

where  $A_i$  is the activity level of radionuclide  $i$  in dust, expressed in  $\text{Bq kg}^{-1}$ ;  $e_i$  is the dose coefficient, expressed in  $\text{Sv Bq}^{-1}$ , for inhalation of the general adult population [12];  $d$  is the quantity of granite dust in air, expressed in  $\text{kg m}^{-3}$ ; and  $V$  is the annual volume of air inhaled, expressed in  $\text{m}^3\ \text{yr}^{-1}$ . The uncertainty associated with the effective dose for dust inhalation was calculated by uncertainty quadratic propagation from the experimental values.

The dose assessment was carried out considering the worst case scenario. The air dust load,  $d$ , was considered to be the maximum legal amount,  $3\ \text{mg m}^{-3}$ , which is higher than the mean value  $1.6\ \text{mg m}^{-3}$ , reported in Spain for this type of industry [13]. Workers were considered not to wear any personal protection (filter mask). All the radionuclide content in dust was considered to have its maximal effect after inhalation, i.e. all dust particles were assumed to be of  $1\ \mu\text{m}$  and the highest  $e_i$  values were considered [12]. The annual volume inhaled,  $1906\ \text{m}^3\ \text{yr}^{-1}$ , was calculated using  $22\ \text{m}^3\ \text{yr}^{-1}$  for adults [12], considering 5 work days (8 h each day) per week and 52 weeks per year. In these conditions, the maximum effective dose due to dust inhalation in the worst case scenario was



**Figure 5.** Mean value and standard deviation of radon concentration, expressed in  $\text{Bq m}^{-3}$ , in indoor air at the office and processing factory areas. The absence of data represents activity levels below the detection limit.

$0.182 \pm 0.009 \text{ mSv yr}^{-1}$ , which is much lower than the reference level of  $1 \text{ mSv yr}^{-1}$  above the background for the general population [12]. Considering the use of filter masks of type FF3 and the mean value air dust load,  $1.6 \text{ mg m}^{-3}$ , a more realistic effective annual dose estimate by inhalation would be  $0.97 \mu\text{Sv yr}^{-1}$ . Figure 4 shows the contribution of the naturally occurring radionuclides analysed to the dose by inhalation, which decreased in the following order:

$$^{228,230,232}\text{Th} > ^{234,235,238}\text{U} > ^{226}\text{Ra} > ^{210}\text{Pb} > ^{210}\text{Po} > ^{40}\text{K}.$$

### 3.3. Radon exposure

Radon concentration in the indoor air in the office and processing factory areas was determined on a monthly basis with activated charcoal canisters. Figure 5 shows the annual variation of the radon concentration in these two areas. The mean value of radon concentration in the office (first floor) and processing factory (groundfloor) were  $(33 \pm 13 \text{ (S.D.)}) \text{ Bq m}^{-3}$  within the range  $(22\text{--}64) \text{ Bq m}^{-3}$  and  $(32 \pm 24 \text{ (S.D.)}) \text{ Bq m}^{-3}$  within the range  $(9.6\text{--}90) \text{ Bq m}^{-3}$ , respectively. An increase in radon concentration was observed during winter. The radon concentration was also assessed in other areas in the processing factory module and in the handcraft workshop. This was only carried out once (in summer), as the dust load was higher. The mean values were  $(11 \pm 5 \text{ (S.D.)})$  and  $(5 \pm 3 \text{ (S.D.)}) \text{ Bq m}^{-3}$  in the factory module and handcraft workshop, respectively, which were lower than those in figure 5 due to the higher ventilation in those areas.

As the use of passive detectors (canisters) give a mean value of the radon concentration during the exposure time (2 d), this value takes into consideration long periods of time in which no work is carried out (mainly night). Thus, during a sampling campaign, the radon (in summer) concentration was determined using an active detector (Alphaguard) during short time

**Table 4.** Range of variation of  $^{222}\text{Rn}$  concentration, expressed in  $\text{Bq m}^{-3}$ , in different locations in the factory.

Location	$^{222}\text{Rn}$ ( $\text{Bq m}^{-3}$ )		Location	$^{222}\text{Rn}$ ( $\text{Bq m}^{-3}$ )	
	<i>N</i>	Range		<i>N</i>	Range
Office	6	27–56	Quarry	7	9–16
Factory (module)	3	65–93	Handcraft workshop	4	118–216
Factory (store room)	4	73–120	Sludge stockpile	2	93–133

*Note:* The office and factory (store room) are indoors, the factory (module) is indoors/outdoors, and the rest of the locations are outdoors. *N*: number of 10 min measurements carried out.

exposures (10 min) and at working conditions in the factory and quarry. Table 4 gives the range of radon content in different areas. The ranges in the office and store room are of the same order as those detected by the canister methodology. Due to the low occupancy factor of the store room (maximum about  $1 \text{ h d}^{-1}$ ), the dose to workers would be lower than for office workers. However, the ranges for the factory module and handcraft workshop were higher than those obtained by the canisters. This can be attributed to workings in process during the determination with the Alphaguard, and daily and seasonal variations of radon concentration. The radon content in the quarry is very low, corresponding to the outdoors. But in the sludge stockpile area, it was higher, although it was outdoors (see figure 2).

In all the areas, radon concentration was well below the reference level of  $600 \text{ Bq m}^{-3}$  in Spanish legislation regarding radon in work environments [7]. Therefore, the workers' exposure to radon can be considered as negligible.

#### 4. Conclusion

Granite can present variable content of naturally occurring radionuclides, and its extraction and processing may pose a health hazard from the radiological point of view, additional to that of silicosis. The assessment of the occupational exposure to naturally occurring radionuclides was made in a selected granite industry, which had a quarry and an associated processing factory. Three main potential exposure pathways were identified: external irradiation, granite dust inhalation, and radon exposure. The first one affected all working environments, since the factory is located on a granitic batholith. The inhalation of granite dust was considered in places where the extraction and processing of granite blocks occurred (processing factory and handcraft workshop). Finally, radon exposure was considered in closed environments, such as the office and processing factory.

The dose rate due to external irradiation in the factory was of the same order as that detected in a nearby farming area not used in granite industry. In the quarry and in the stockpile areas, a slight increase in the mean value of the dose rate ( $0.016\text{--}0.076 \text{ mSv yr}^{-1}$ ) was observed due to the granite faces and/or stockpiled blocks surrounding the dose rate monitor. Workers in these areas use heavy machinery to move and manipulate the granite blocks, which serve as an additional shield/protection. The mean value in each considered area was below the world mean dose rate of  $2.4 \text{ mSv yr}^{-1}$  due to natural sources [18]. Therefore, the external irradiation pathway seems to have little influence on the occupational exposure. The radionuclide content of granite dust ( $\text{Ø} < 45 \mu\text{m}$ ) was higher than that of the original granite, due to the smaller particle size. In the worst case scenario, the maximum dust load allowed by law with no use of personal protection equipment (filter masks), the maximum effective dose by inhalation, was  $0.182 \pm 0.009 \text{ mSv yr}^{-1}$ . Thorium radionuclides were the main contributors, about 77%.

The use of filter masks can reduce this dose by two orders of magnitude, since they are able to retain 99% of particles higher than 1  $\mu\text{m}$ . The contribution of these two exposure pathways to workers is as much as 0.26  $\text{mSv yr}^{-1}$  in the worst case scenario, which is lower than the reference level of 1  $\text{mSv yr}^{-1}$  for the general population.

The mean annual radon concentration in indoor air was measured to be about 33  $\text{Bq m}^{-3}$ , using passive detectors (activated charcoal canister). An active detector was also used for short integration times (10 min) in working conditions. An increase in the radon content was observed when granite was being processed, which usually involves sawing, polishing and sanding. The maximum radon content was 216  $\text{Bq m}^{-3}$ . As all radon content was below the intervention level of 600  $\text{Bq m}^{-3}$  for work places according to Spanish legislation, no action or control is required.

As a consequence of these results, no workers in this quarry need to be designated as occupationally exposed and subject to individual dosimetry

## Acknowledgment

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## References

- [1] Aarafa W 2004 Specific activity and hazards of granite samples from the Eastern Desert of Egypt *J. Environ. Radioactiv.* **75** 315–27
- [2] AENOR 1998 UNE-EN 136:1998. Equipos de protección respiratoria. Máscaras completas. Requisitos, ensayos, marcado, Madrid (in Spanish)
- [3] Ahmed N K 2005 Measurement of natural radioactivity in building materials in Qena city, Upper Egypt *J. Environ. Radioactiv.* **83** 91–9
- [4] Allen J G, Zwack L M, MacIntosh D L, Minegishi T, Stewart J H and McCarthy J F 2013 Predicted indoor radon concentration from a Monte Carlo simulation of 1000 000 granite countertop purchase *J. Radiol. Prot.* **33** 151–62
- [5] Baeza A, Del Rio M, Miró C and Paniagua J 1992 Natural radioactivity in soils of the province of Cáceres (Spain) *Radiat. Prot. Dosim.* **45** 261–3
- [6] BOE 2007 Orden ITC/2585/2007. Instrucción Técnica Complementaria 2.0.02. Protección de los trabajadores contra el polvo, en relación con la silicosis, en las industrias extractivas. Reglamento general de normas básicas de seguridad minera. B.O.E. no. 215. 7 de septiembre de 2007, Madrid (in Spanish)
- [7] BOE 2012 Instrucción IS-33, de 21 de diciembre de 2011, del Consejo de Seguridad Nuclear, sobre criterios radiológicos para la protección frente a la exposición a la radiación natural. B.O.E. no. 22, 26 de enero de 2012, Madrid (in Spanish)
- [8] Bolívar J P, García-Tenorio R, Mas J L and Vaca F 2002 Radioactive impact in sediments from an estuarine system affected by industrial wastes releases *Environ. Int.* **27** 639–45
- [9] Bøtter-Jensen L 2000 Testing of environmental radiation monitors using the Risø low-level radiation measurement stations *Radiat. Prot. Dosim.* **92** 109–14
- [10] EPA 1987 U.S. Environmental Protection Agency. EPA 520/6-87-005. EERF Standard operating procedures for Radon-222 measurement using charcoal canisters, Montgomery, AL (USA) 1–30
- [11] Guillén J, Tejado J J, Baeza A, Corbacho J A and Muñoz J G 2014 Assessment of radiological hazard of commercial granites from Extremadura (Spain) *J. Environ. Radioact.* **132** 81–8
- [12] ICRP 2012 Compendium of Dose Coefficients based on ICRP Publication 60. ICRP Publication 119 *Ann. ICRP* **41** 87–119
- [13] INS (Instituto Nacional de Silicosis) 2001 *Memoria de Actividades 2001*, Technical Report, Madrid (in Spanish)
- [14] Koike K, Yoshinaga T, Suetsugu K, Kashiwaya K and Asaue H 2015 Controls on radon emission from granite as evidenced by compression testing to failure *Geophys. J. Int.* **203** 428–36

- [15] Liu Y, Steenland K, Rong Y, Hnizdo E, Huang X, Zhang H, Shi T, Sun Y, Wu T and Chen W 2013 Exposure-response analysis and risk assessment for lung cancer in relationship to silica exposure. A 44 year cohort study of 34018 workers *Am. J. Epidemiol.* **178** 1424–33
- [16] Pavlidou S, Koroneos A, Papastefanou C, Christofides G, Stoulos S and Vavelides M 2006 Natural radioactivity in granites used as building materials *J. Environ. Radioact.* **86** 48–60
- [17] Sill C W 1987 Precipitation of actinides as fluorides or hydroxides for high-resolution alpha spectrometry *Nucl. Chem. Waste Manage.* **7** 201–15
- [18] UNSCEAR 2008 United Nations scientific committee on the effects of atomic radiation *UNSCEAR 2008 Report to the General Assembly with Scientific Annexes* United Nations, New York