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Phosphogypsum recycling in the building materials industry: assessment of the radon exhalation rate

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ABSTRACT

Phosphogypsum can be classified as a Naturally Occurring Radioactive Material (NORM) residue of the phosphate fertilizer industry. One of the main environmental concerns of its use as building material is the radon exhalation. The aim of this study is to measure the radon exhalation rate from plates and bricks manufactured with phosphogypsum from three installations of the main Brazilian producer, Vale Fertilizantes, in order to evaluate the additional health risk to dwellers. A simple and reliable accumulator method involving a PVC pipe sealed with a PVC pipe cover commercially available with CR-39 radon detector into a diffusion chamber was used for measuring radon exhalation rate from phosphogypsum made plates and bricks. The radon exhalation rate from plates varied from 0.19 ± 0.06 Bq m^{-2} h^{-1}, for phosphogypsum from Bunge Fertilizers, from 1.3 ± 0.3 Bq m^{-2} h^{-1}, for phosphogypsum from Ultrafertil. As for the bricks, the results ranged from 0.11 ± 0.01 Bq m^{-2} h^{-1}, for phosphogypsum from Bunge Fertilizers, to 1.2 ± 0.3 Bq m^{-2} h^{-1}, for phosphogypsum from Ultrafertil. The results obtained in this study for the radon exhalation rate from phosphogypsum plates and bricks are of the same order of magnitude than those from ordinary building materials. So, it can be concluded that the recycling of phosphogypsum as building material is a safe practice, since no additional health risk is expected from the radiological point of view.

1. Introduction

In the last years, there has been an increased interest in measuring radionuclides concentration in building materials and radon levels in the air at homes, due to their potential risk to human health (Campos and Pecequilo, 2003; Cazula et al., 2015; EC, 1999, Todorović et al., 2014; Turhan et al., 2008; Kovler, 2009). Exposure to radon accounts for more than 50% of the annual effective dose from natural radioactivity (UNSCEAR, 2000).

Radon in a house comes mainly from the soil adjacent to the foundation. However, radon exhalation from building materials is another potential source of the gas in indoor environment.

Building materials are an important radon source in houses since they can contain small amounts of natural radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K. The radionuclides content is usually low, but some materials, like phosphogypsum, may contain higher concentrations depending on the origin of the raw material used. The presence of radionuclides puts restrictions on the use of phosphogypsum in building materials and in soil amendments. The Brazilian regulatory body ruled that phosphogypsum would only be permitted for use in agriculture or in the cement industry if the concentration of ^{226}Ra and ^{228}Ra does not exceed 1 Bq g^{-1} (CNEN, 2014). Recently, a working group was established at the national regulatory level in Brazil, aiming to define a policy for using phosphogypsum as construction material.

Phosphogypsum is a waste of the phosphate fertilizer industry. It is formed by the chemical attack of the phosphate rock with sulphuric acid to produce phosphoric acid. This waste is generally stored in piles near the fertilizer factory. Phosphogypsum can be classified as a NORM residue, *Naturally Occurring Radioactive Material,* which means that its reuse may pose risks to human and environment from the radiological protection point of view (El Afifi et al., 2007; IAEA, 2013a). The radiological impact assessment of NORM residues on the environment should comprise the release of radon isotopes into the atmosphere (IAEA, 2013b).

The residues containing NORM altered by human activities has received considerable amount of global attention over the last decades. The main concern is the large amount of this residue and consequent long-term risk due to the presence of radionuclides

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with long half-lives and high radiotoxicity. According to TECDOC 1712 "Management of NORM Residues" from International Atomic Energy Agency (IAEA, 2013c), it is necessary changing attitudes towards NORM residues. This document emphasizes that "… *there is an overall trend worldwide towards greater recycling of NORM residues and their use as by-products. This is being driven by sustainability issues such as concerns over the depletion of non-renewable resources, by more stringent environmental protection legislation, by a growing recognition that the amounts of NORM disposed of as waste need to be minimized in order to make their disposal manageable, and sometimes simply by economic considerations, some of which become evident only when the true costs and liabilities of NORM residue disposal as waste are taken into account. Some countries are now making specific provision in their regulatory systems for NORM residue recycling and use*". Therefore, there is an overall trend in recycling phosphogypsum, provided that such practice will not cause additional radiological health risk to the public.

Phosphogypsum has been used in cement industry, as sub-base of roads, as building material and as soil amendment. Phosphogypsum has become an item of commercial interest in many countries, with a well-established market value in agriculture and construction. However, the commercial use of phosphogypsum has been restricted because of concerns about its radioactivity content. Relatively high radioactivity, particularly ^{226}Ra activity concentration (Dueñas et al., 2007, Mazzilli et al., 2000), among other impurities, can prevent its reuse for several purposes.

The worldwide phosphogypsum production is estimated to be at 160 million t (IAEA, 2013a) per year and only 14% is recycled and used as building material (Máduar et al., 2011). Production is increasing worldwide and could reach up to 250 million t within the next twenty years. Approximately 3 billion t of phosphogypsum are stored in piles of various sizes in well over 50 countries (IAEA, 2013a).

The Brazilian annual production of phosphogypsum reaches 12 million tons (DNPM, 2014), and Brazil, like other countries that produce this residue, is trying to find safe solutions for its reuse. One possible application of phosphogypsum is in the manufacturing of building materials. However, an evaluation of its radiological impact is required, mainly due to the radionuclides content and radon exhalation rate. Several studies were published concerning the radiological impact of using Brazilian phosphogypsum as building material, comprising the measurement of the radionuclides content, as well as the internal and external exposure of dwellers (Campos et al., 2011; Máduar et al., 2011, Mazzilli and Saueia, 1999; Nisti et al., 2013; Rabi and Silva, 2006). Since most of the Brazilian phosphogypsum contains traces amounts of radium isotopes, a main concern to the dwellers is the inhalation of radon and consequent internal exposure. The radon exhalation rate of is defined as the amount of activity released per unit surface area per unit time from the material. It depends on the ^{226}Ra content of the material, emanation coefficient, gas diffusion coefficient in the material, porosity and density of the material (Dueñas et al., 2007; Sharma and Virk, 2001).

The aim of this study is to determine experimentally the radon exhalation rate from bricks and plates made of phosphogypsum, in order to evaluate if these building materials pose no additional health risk to dwellers due to radon exhalation. A practical approach to measure radon exhalation rate directly from the surface of the material is to allow the radon build up in a container, known as accumulator, over time (Chen et al., 2010; Kotrappa and Stieff, 2009; Sharma et al., 2003; Shweikani and Raja, 2009) and measure the radon content by passive method with solid state nuclear track detectors (SSNTD). The phosphogypsum bricks and plates evaluated in this study were manufactured by UCOS (humidification, compaction and drying) method (Novogesso® Technology) (Kanno et al., 2007), with phosphogypsum provided by Vale Fertilizers, the main Brazilian phosphate fertilizer producer.

2. Materials and methods

Several methods to measure radon exhalation rate from building materials make use of accumulators associated with alpha particle detectors, such as, continuous gas monitor, scintillation cell, ionization chamber and solid state nuclear track detector (Kobeissi et al., 2013; Lettner and Steinhäusler, 1988; Righi and Bruzzi, 2006; Tuccimei et al., 2006). In this study, a simple and reliable accumulator method with CR-39 into diffusion chambers was used for measuring radon exhalation rate from phosphogypsum building materials.

Eighteen samples of plates and fifteen samples of bricks manufactured with phosphogypsum provided by Vale Fertilizantes were analyzed for the ^{222}Rn exhalation rate. The phosphogypsum producers are namely Ultrafertil located in Cubatão, Fosfertil located in Uberaba and Bunge Fertilizers located in Cajati. Five samples of brick manufactured with natural gypsum were also analyzed.

A Polyvinyl chloride (PVC) pipe sealed with a PVC pipe cover commercially available of know dimensions (7.2 cm diameter and 30 cm height) was used as the accumulator. As reported by Tuccimei et al. (2006), PVC can be considered a radon-tight material, unless exposed to high levels of gamma radiation that can affect its radon permeability. The accumulator was placed on the phosphogypsum bricks and plates and the contact between the accumulator and samples was sealed with silicone adhesive. A CR-39 in a plastic diffusion chamber (passive radon detector) was placed inside the accumulator at a distance of 25 cm from the surface of the sample, in order to count only the ^{222}Rn contribution and to avoid the role of ^{220}Rn from the surface of phosphogypsum building materials samples (Faheem and Matiullah, 2008; Shafi-ur-Rehman et al., 2006). This passive radon detector assures the discrimination of radon decay products by allowing only the gas to enter the diffusion chamber (Bartlett et al., 1988). The exhalation rate was determined through the radon concentration at the accumulator.

At the accumulator closed system (Faheem and Matiullah, 2008; Rahama et al., 2007), the passive radon detector is exposed to variable concentrations, starting from zero to equilibrium concentration. So, in order to calculate the radon concentration at the accumulator, it is necessary to determine the effective exposure time. The effective exposure time of the passive detectors to radon was calculated through the following equation (Shweikani and Raja, 2009):

$$t_{eff} = t - \frac{1}{\lambda}\left(1 - e^{-\lambda \cdot t}\right) \tag{1}$$

where: t is the real exposure time and λ is ^{222}Rn decay constant.

The detectors were exposed to radon for 30 days. After exposure, CR-39 detectors were etched in a KOH (30% mass weight) solution at 80 °C for 5.5 h in a constant temperature bath (Máduar et al., 2011; Manocchi et al., 2014). After etching, CR-39 detectors were washed, dried and counted under a Carl Zeiss microscope for track density measurements. The background was 12 ± 2 tracks per cm^2. The track density was related to radon concentration in the environment, by using a calibration factor of 2.14 ± 0.17 tracks cm^{-2} per kBq m^{-3} h, obtained with Pylon model RN-150 calibrated radon gas source. Radon concentration in the accumulator was calculated through the following equation (Shweikani and Raja, 2009):

$$c = \frac{D}{k \cdot t_{eff}} \tag{2}$$

where C is the radon concentration ($\text{Bq} \cdot \text{m}^{-3}$), D is the track density ($\text{tr} \cdot \text{cm}^{-2}$), k is the calibration factor ($\text{tr} \cdot \text{cm}^{-2}$ per $\text{Bq} \cdot \text{m}^{-3} \cdot \text{d}$), t is the effective exposure time (d).

Radon exhalation rate was calculated using the following equation (Faheem and Matiullah, 2008; Shafi-ur-Rehman et al., 2006):

$$E = \frac{c \cdot (\alpha \cdot A + V \cdot \lambda)}{A\left[1 - e^{-t(\alpha A/V+\lambda)}\right]} \tag{3}$$

where E is the radon exhalation rate (Bq m^{-2} h^{-1}), C is the radon concentration in the accumulator (Bq m^{-3}), V is the air volume of the accumulator (m^3), A is the surface area of the sample (m^2), α is the back diffusion constant, λ is the decay constant of radon (h^{-1}), t is the exposure time (h).

3. Results and discussion

Table 1 presents the results of the radon exhalation rate from plates and bricks analyzed in this study. The radon exhalation rate from plates varied from 0.19 ± 0.06 Bq m^{-2} h^{-1}, for the phosphogypsum from Bunge Fertilizantes, to 1.3 ± 0.3 Bq m^{-2} h^{-1}, for the phosphogypsum from Ultrafertil. As for the bricks, the results ranged from 0.11 ± 0.01 Bq m^{-2} h^{-1}, for the phosphogypsum from Bunge Fertilizers, to 1.2 ± 0.3 Bq m^{-2} h^{-1}, for the phosphogypsum from Ultrafertil.

It can be seen from Table 2 that the experimental results obtained in this study for phosphogypsum plates and bricks from Bunge Fertilizers, Fosfertil, and Ultrafertil are of the same order of magnitude than those from ordinary building materials. So, it can be concluded that these phosphogypsum building materials pose no additional health risk to dwellers due to radon exhalation rate.

It is expected that the radon exhalation should depend on the radium activity concentration in the samples. In a previous work, Nisti et al. (2013) evaluated the average activity concentration of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K, as well as, the radium equivalent activity for the same phosphogypsum plates and bricks from Ultrafertil, Fosfertil and Bunge Fertilizers analyzed in this study. A positive correlation can be observed between radium contents and radon exhalation rates, with correlation coefficients 0.91 and 0.82 for bricks and plates, respectively (Figs. 1 and 2).

Several authors observed that the ^{222}Rn exhalation rate from phosphogypsum depends not only on the ^{226}Ra content, but also on other factors such as density, porosity and manufacturing process of the building material. Dueñas et al. (2007) conducted a study of radon exhalation from phosphogypsum piles and showed that there is a statistically significant relationship between the exhalation of ^{222}Rn and the ^{226}Ra activity concentration and the porosity and density. A multiple regression analysis was carried out in order to determine to which extent these factors could influence the radon exhalation. The R-squared statistic of the model developed by them indicated that 80% of the ^{222}Rn exhalation variability depend on these factors.

Table 1
^{222}Rn exhalation rate (value ± standard deviation) from phosphogypsum plates and bricks from different producers.

Samples	^{222}Rn exhalation rate (Bq m^{-2} h^{-1})	
	Plates	Bricks
Ultrafertil phosphogypsum	1.3 ± 0.3	1.2 ± 0.6
Fosfertil phosphogypsum	0.41 ± 0.07	0.47 ± 0.15
Bunge Fertilizers phosphogypsum	0.19 ± 0.06	0.11 ± 0.01
Natural gypsum	ND	0.18 ± 0.08

ND – not determined.

Table 2
Radon exhalation rate from bricks and plates made of phosphogypsum and other building materials (Bq m^{-2} h^{-1}).

Building Material	^{222}Rn exhalation rate	References
Phosphogypsum Brick	0.11–1.2	This study
Phosphogypsum Brick	0.41–5.7	Nisti et al., 2013
Phosphogypsum Brick	6–10	Fournier et al., 2005
Gypsum Plate	0.2–18[a]	Folkerts et al., 1984
Phosphogypsum Plate	0.19–1.3	This study
Phosphogypsum Plate	0.16–4.3	Nisti et al., 2013
Phosphogypsum Plate	2.2–4.8	Lettner and Steinhäusler. 1988
Crude Brick	0.16	Sharma and Virk. 2001
Granite	0.16–1.4	Kotrappa and Stieff. 2009
Cement	0.27–0.66	Lettner and Steinhäusler. 1988
Cement	0.18–0.91	Sharaf et al., 1999
Concrete	4.3	De Jong et al., 2006
Soil	2.2–2.8	Shafi-ur-Rehman et al., 2006
Sand	3.9–17	Shafi-ur-Rehman et al., 2006
Slate	0.36–1.9	Chen et al., 2010

[a] Phosphogypsum.

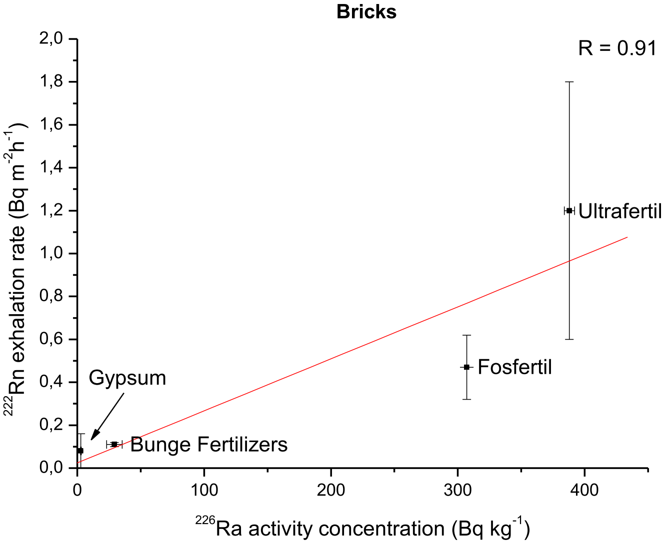

Fig. 1. Correlation between radon exhalation rate and radium activity concentration from phosphogypsum bricks of different producers.

The variation observed in the ^{222}Rn exhalation rates from phosphogypsum plates and bricks cannot be explained only by the Ra content, but depends also on other factors such as density, porosity and manufacturing process of the building material. Considering only the ^{226}Ra activity concentration of phosphogypsum brick and plates (Nisti et al., 2013), it is expected that the radon exhalation rates would be of the same order of magnitude. Nevertheless, the radon exhalation rate from phosphogypsum bricks and plates from Ultrafertil is three to four times higher than those from Fosfertil. This difference can be explained by the hypothesis that radon exhalation depends on several properties of the material and not only on the radium activity concentration, as corroborated by Dueñas et al. (2007). In this study, the influence of porosity on ^{222}Rn exhalation rate from phosphogysum bricks and plates was evaluated. The total porosity of the phosphogypsum brick and plates was calculated from the bulk density and article

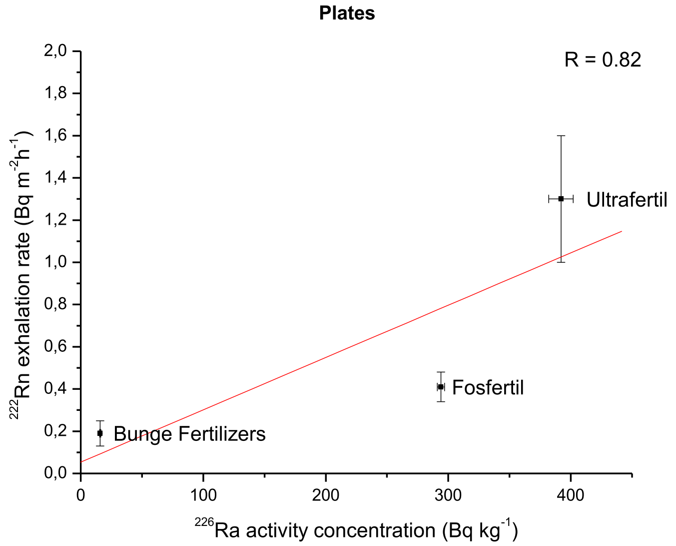

Fig. 2. Correlation between radon exhalation rate and radium activity concentration from phosphogypsum plates of different producers.

density (Rutherford et al., 1995). Figs. 3 and 4 show the correlation between ^{222}Rn exhalation rate per unit of ^{226}Ra activity concentration and total porosity.

It can be observed a correlation between radon exhalation rates per unit of radium activity concentration and total porosity, with correlation coefficients −0.92 and 0.94 for phosphogysum bricks and plates, respectively. However, plates and bricks behave differently; while radon exhalation decreases linearly with increase in porosity for phosphogysum bricks, an inverse behavior was observed for the plates. This can be explained by the manufacturing process of these building materials. Although both are manufactured by UCOS method, a resin is added to the plates, forming a kind of coating film on the plates that can affect the radon exhalation rate.

The effect of internal wall covering or painting on radon exhalation rate from phosphogypsum bricks and plates was also evaluated. The results obtained showed decreases of 64% and 70% in the radon exhalation rate for painted and covered wall, respectively.

Nisti et al. (2014) evaluated the natural radionuclides content and the radon exhalation rate from phosphogypsum piles from Ultrafertil and Fosfertil fertilizer industries. The radon exhalation rates varied from 341 to 562 Bq m^{-2} h^{-1} for Fosfertil and Ultrafertil, respectively. These results are two orders of magnitude higher than those from plates and bricks, giving evidence that the porosity, density and gas diffusion coefficient in the building material play an important role in the radon exhalation.

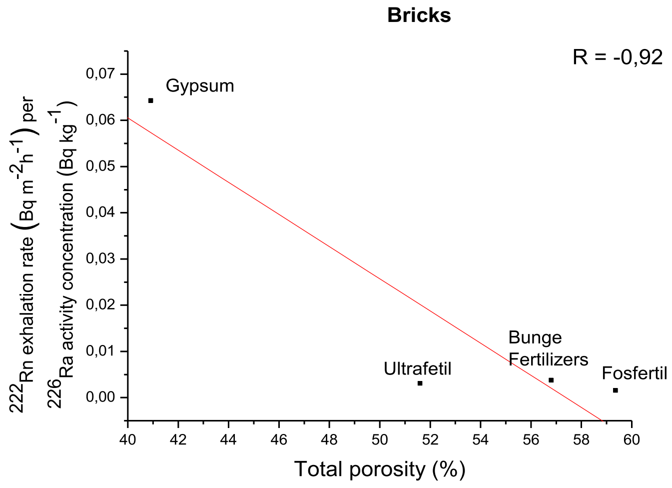

Fig. 3. Correlation between ^{222}Rn exhalation rate per unit of ^{226}Ra activity concentration and total porosity from phosphogysum bricks of different producers.

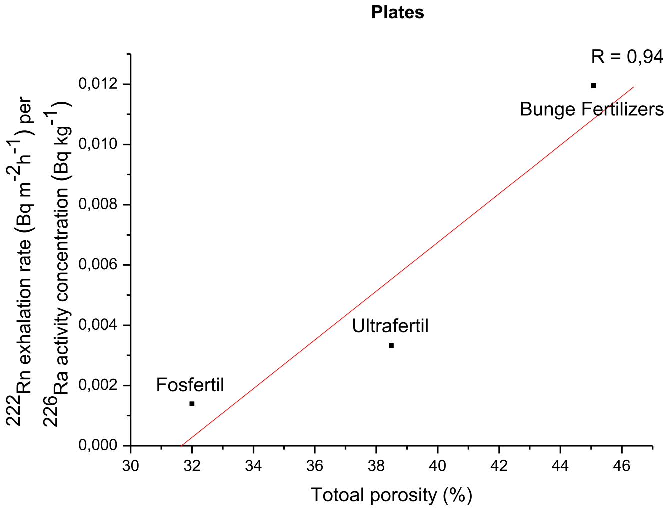

Fig. 4. Correlation between ^{222}Rn exhalation rate per unit of ^{226}Ra activity concentration and total porosity from phosphogysum plates of different producers.

4. Conclusions

The PVC pipe sealed with a PVC pipe cover commercially available with NRPB radon detector was a convenient method for measuring radon exhalation rates from phosphogypsum made building materials.

From the results obtained for radon exhalation rate from phosphogypsum bricks and plates from Bunge Fertilizers, Fosfertil and Ultrafertil it can be concluded that the use of these building materials for house construction pose no additional health risk due to exhalation of radon. Building components, such as paint and plaster could be considered as insulating materials that further improve the reduction of radon exhalation from phosphogypsum bricks and plates.

Building materials should be classified on the basis of their, radon exhalation rate, besides the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, and the radium equivalent activity; as proposed by Tuccimei et al. (2006) and Steger et al. (1992).

It can be concluded that the recycling of phosphogypsum as building material will pose no additional internal exposure due to inhalation of radon and external exposure from gamma emitters in dwellers. Therefore, the practice of recycling phosphogypsum as building material can be a safe alternative, since no additional health risk is expected from the radiological point of view. Furthermore, this practice will be driven by sustainability issues such as concerns over the depletion of non-renewable resources of natural gypsum, by more stringent environmental protection legislation, and by minimizing the amounts of NORM disposed of as waste.

The results obtained in this study can contribute for the establishment of guidelines by the Brazilian regulatory agency for the safe use of phosphogypsum as building material.

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