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Using ^{222}Rn as a tracer of geodynamical processes in underground environments



D.L. Valladares^a, A.A.R. da Silva^{b,d}, T. Lacerda^c, R.M. Anjos^{c,*}, M. Rizzotto^a, H. Velasco^a, J.P. de Rosas^a, G. Tognelli^e, E.M. Yoshimura^b, J. Juri Ayub^{a,c}

^a GEA, Instituto de Matemática Aplicada San Luis (IMASL), Universidad Nacional de San Luis, Consejo Nacional de Investigaciones Científicas y Técnicas, Ej. de los Andes 950, D5700HHW San Luis, Argentina

^b Instituto de Física, Universidade de São Paulo, P.O.Box 66318, 05314-970, São Paulo, SP, Brazil

^c Instituto de Física, Universidade Federal Fluminense, Av. Gal Milton Tavares de Souza, s/nº, Gragoatá, 24210-340, Niterói, RJ, Brazil

^d Serviço Especializado em Engenharia de Segurança e Medicina do Trabalho, Departamento de Saúde, Universidade de São Paulo, Rua da Reitoria, 109, 05508-900, São Paulo, SP, Brazil

^e Departamento de Geología, Universidad Nacional de San Luis, Ej. de los Andes 950, D5700HHW San Luis, Argentina

HIGHLIGHTS

- ^{222}Rn levels in two old mines in San Luis, Argentina
- CR-39 nuclear track detectors were used for this purpose
- higher concentration values of ^{222}Rn were observed in summer than in winter
- radon pattern distribution appear as a good method to trace air currents
- it localizes unknown ducts, fissures or secondary tunnels in subterranean environments.

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ABSTRACT

Radon levels in two old mines in San Luis, Argentina, were measured and analyzed, with the aim to assess the potential use of this radioactive noble gas as a tracer of geological processes in underground environments. La Carolina gold mine and Los Cóndores tungsten mine are today used as tourism mines. CR-39 nuclear track detectors were used for this purpose. Measurements were performed during both winter and summer seasons. The findings show that in these environments, significant radon concentrations are subject to large seasonal fluctuations, due to the strong dependence on natural ventilation with the outside temperature variations. For both mines, higher concentration values of ^{222}Rn were observed in summer than in winter; with an extreme ratio of 2.5 times between summer and winter seasons for Los Cóndores mine. The pattern of radon transport inside La Carolina mine revealed, contrary to what was believed, that this mine behaves as a system with two entrances located at different levels. However, this feature can only be observed in the winter season, when there is a marked difference between the inside and outside temperatures of the mine. In the case of Los Cóndores mine, the radon concentration pattern distribution is principally established by air current due to chimney-effect in summer and winter seasons. In both cases, the analyses of radon pattern distribution appear as a good method to trace air currents, and then localize unknown ducts, fissures or secondary tunnels in subterranean environments.

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1. Introduction

The analysis of temporal and spatial variations on the flux of soil gases across the soil–air interface can be a useful tool to study geodynamical processes occurring in the rock mass due to earthquakes, volcanic and seismic activities, tremors, shocks, and coal or rock bursts. One of these gases is the radioactive ^{222}Rn (half-life 3.82 days, alpha emitter),

commonly known as radon. It is a naturally occurring noble element from the decay of ^{238}U series, with ^{226}Ra as its immediate parent nuclide. Radon atoms are continuously generated in the rock matrix and emanate into the air-filled pore space, from where some of them reach the ground surface and escape into the atmosphere (Atkinson et al., 1983; Nazaroff, 1992; Hakl, 1997; Hakl et al., 1997; Kies et al., 2002; Viñas et al., 2007; Anjos et al., 2010a; Estellita et al., 2010; Steinitz et al., 2011). The generation of radon depends on the uranium concentration and the nature of the parent mineral. However, the transmission to atmosphere is largely independent of these.

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

Uranium is very seldom homogeneously distributed throughout rocks and soils. Most of the uranium in rocks can be attributed to discrete uranium bearing minerals even when there is only a few parts per million of uranium present (Ball et al., 1991; Anjos et al., 2005, 2006, 2007, 2010b, 2011; Veiga et al., 2006). Clearly, as radon is a gas with a short half-life, its chance to escape from the mineral matrix to the environment is higher when it is generated near the superficial margin of that mineral. The openness and imperfections in the lattice are important controls, as is the specific surface area of the mineral. Once the radon is released from the parent mineral to the intergranular region several factors take over, such as the fluid transmission features of the rock, the nature of fractures and disaggregation features (Ball et al., 1991). As variations in the flux of radon are only caused by physical factors, since it is not a reactive species, it could give valuable information on distribution and dynamics of airflows inside of underground environments (Viñas et al., 2007). Then, the patterns of radon concentration into this kind of environments would allow studying geodynamic processes, such as air mass movements.

Among the underground environments of scientific and technological interest are the caves, quarries, and mines. In these places, variations in the concentration of ^{222}Rn might be caused by the presence of ducts and galleries as well as interconnected sets of fissures, fractures and voids. One of the most appropriate methods to trace airflows in these systems, which carry gas radon, is the use of nuclear track detectors. These detectors have desirable features for in situ measurements in large scales (Grossi et al., 2011).

In this study, we investigated the transport process of natural radon in two exhausted and abandoned mines in San Luis Province, Argentina, with the aim to evaluate the potential use of this gas as a tracer or marker of geodynamic processes. These mines were chosen because they have different physical configurations in their cavities, features that can affect the airflow patterns and radon concentrations. La Carolina gold mine is currently a blind end system, corresponding to a horizontal excavation into the side of a mountain, with only a main adit. Los Córdobes tungsten mine is also a horizontal excavation into the side of a mountain, but has a vertical output (a shaft) at the end of the main gallery.

2. Material and methods

2.1. Study area and its geological features

The study area comprises a portion of the Pampa Range of San Luis Province in west-central Argentina, a complex of Early Paleozoic metamorphic rocks intruded by Paleozoic granitoids and pegmatites related to tungsten and gold deposits (Miller and Singewald, 1919; Da Silva et al., 2011). Geologically this sector is composed of high-grade granulite facies metamorphic rocks, mainly gneisses and migmatites, with intercalated igneous mafic-ultramafic bodies (Rastall and Wilcockson, 1920). Fig. 1 shows a geological illustration of the study area.

La Carolina gold mine ($32^{\circ} 48' 0'' \text{ S}$, $66^{\circ} 60' 0'' \text{ W}$) belonged to West Argentine Gold Company, a British company that, in 1882, drove a 380 m adit from a point on the western side of the Carolina Mountain to the eastern direction, intersecting the four principal veins of mineral. In total about 500 m of tunnels was excavated and several shafts were sunk. The mine was operational until 1894, when a catastrophic collapse of the main adit killed 30 miners, resulting in the closure of the mine (Hoskold, 1904). After its abandonment, several secondary tunnels have been buried. Currently, this mine shows a horizontal excavation into the side of a mountain, with a main adit (Fig. 2a).

In Argentina, tungsten deposits are located partly in the province of Catamarca in the north, and in the provinces of Córdoba and San Luis in the center. In this last region, the most important mines are distributed in the ranges of Sierra de Córdoba and Sierra de San Luis. The ore was chiefly wolfram rich, with lower contents of scheelite, hubnerite and tungstic ochre accompanied by pyrites, copper

ores, galena, bismuthinite, a little molybdenite, occasionally cassiterite, and, in some places, notable amounts of niobium and tantalum minerals. The gangue is mainly quartz, sometimes with white mica and occasionally topaz and fluor spar (Rastall and Wilcockson, 1920; Werner et al., 1998). Los Córdobes mine was the major tungsten quarry in the country and the second one in South America, being exploited by German and American miners from 1898 to 1965. The lode was brecciated and composed of anastomosing veins. This mine is situated in the Sierra de San Luis, near a small town called Concarán in San Luis Province ($32^{\circ} 33' 25'' \text{ S}$, $65^{\circ} 15' 20'' \text{ W}$). The lode was cropped out on the hill side and was worked by adits. At the level of the main adit, it was displaced by a series of step faults, which necessitated frequent cross-cutting excavations. The ore was, then, exhausted down to this level, and further was developed other extraction shaft to a depth of 90 m. Also in this case, after the mine abandonment, several secondary adits have been buried. Currently, what remains of the mine appears as a 450 m main adit with a shaft at the end of this adit. Fig. 2b shows the topographical illustration of the Los Córdobes mine.

2.2. Experimental procedure

Radon concentration measurements were performed using CR-39 nuclear track detectors. The sampling was performed in both the winter and summer seasons. The field works started in the summer. During this season, radon monitors were exposed for 105 days in La Carolina mine and 42 days in Los Córdobes mine. The exposure time in Los Córdobes, which was monitored after La Carolina, was reduced in order to facilitate the track detector analysis, since the CR-39 detectors exposed in La Carolina mine showed very high track densities. In winter season, radon monitors were exposed for 36 days in La Carolina mine and 96 days in Los Córdobes mine. The exposure time intervals in winter season were decided taking into account the statistics associated with results obtained in the summer measurements, and the difficulty of carrying out field work during the winter. The mean value of the outside temperature at La Carolina mine was 15° C in summer and 6° C in winter. The mean value of the outside temperature at Los Córdobes mine was 20° C in summer and 9° C in winter (REM, 2011). The internal temperature remained stable around 16° C at two mines during summer and winter seasons.

The CR-39 plastic track detector (1.7 cm^2 area and 0.9 mm thick) was enclosed in a radon diffusion chamber (NRPB/SSI type monitors) (Orlando et al., 2002). During summer measurements, detectors were deployed at 14 points within the adits of La Carolina gold mine and 20 points of Los Córdobes wolfram mine. For winter measurements, detectors were deployed at 20 points in La Carolina and Los Córdobes mines. Fig. 2a and Table 1 illustrate the sites where the detectors were placed in the La Carolina mine. Fig. 2b and Table 2 illustrate the sites where the detectors were placed in Los Córdobes mine. The detectors were placed at a distance of 20 cm from the wall and 1 m above the ground. The choice of measurement sites was based on the criteria of accessibility and in order to cover equally well the adits of the mines. The determination of radon concentrations was performed at the Dosimetry Laboratory of Nuclear Physics Department of the University of São Paulo (Da Silva and Yoshimura, 2005). For this determination, two main assumptions are used: radon in the environmental air rapidly reaches equilibrium with the air inside the monitor; and both radon progeny and shorter half life radon isotopes are prevented from entering the diffusion chamber (Shweikani and Durrani, 1995). The mean radon concentration during the whole period of exposure is directly proportional to the track density in the plastic detector and the time of exposure. The conversion factor of track density to radon concentration was experimentally determined and its value is known to be very constant for this combination of monitor and plastic detector (Orlando et al., 2002; Howarth and Miles, 2003). To determine it in our etching and measurements conditions, a separate group of monitors was exposed in a 583 L chamber to a radon source (^{226}Ra of 96.57 kBq, model 2000A, Pylon Electronics Inc.) for

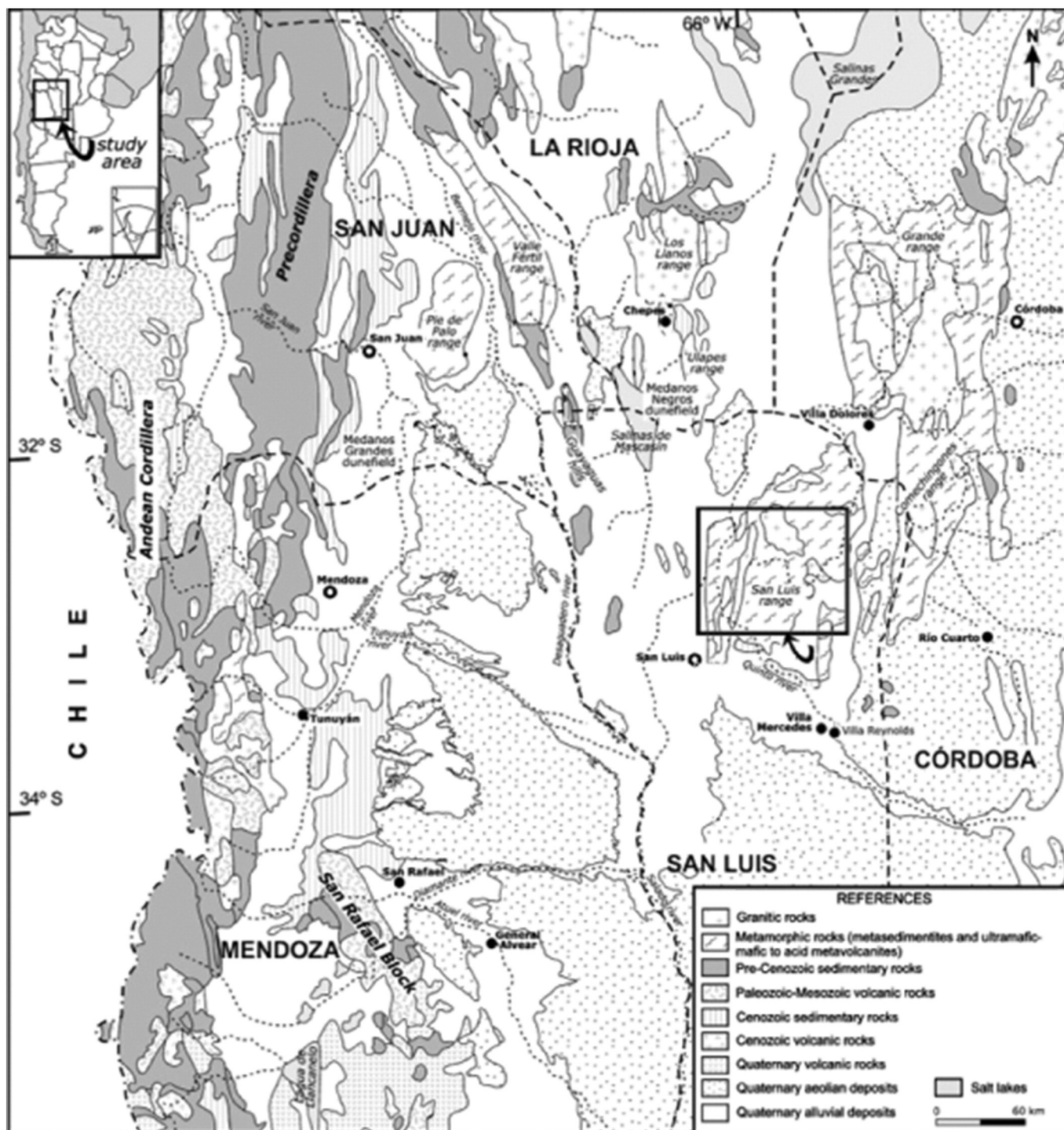


Fig. 1. Geologic setting and location of the studied area (modified from Tripaldi et al., 2010).

known periods of time, and the value of 2.8 ± 0.2 (track cm^{-2})/($\text{kBq m}^{-3} \text{ h}$) was obtained, being compatible with the already published values (Orlando et al., 2002; Howarth and Miles, 2003). The radon concentration inside the 583 L chamber was checked with a Lucas cell detector (model 110A, Pylon Electronics Inc.). The detector was calibrated by the Institute for Energy and Nuclear Research of the Brazilian Nuclear Energy Commission (IPEN-CNEN). After a chemical etching (in a 30% KOH solution at 80 °C for 330 min) CR-39 detector track densities were evaluated with a semi automated optical microscopy analysis system described in a previous work (Da Silva and Yoshimura, 2005). The uncertainties of the concentration activities are

derived from the counting statistical error, that is, the square root of the number of observed events (Poisson statistics). In this work, the uncertainties were estimated to be of the order of 8% for the radon concentrations.

2.3. Analysis of radon concentration patterns inside subterranean environments to trace of geodynamical process

Caves are usually considered as static environments where physical properties as temperature and humidity remain stable during the year. But inside the caves, some parameters have very pronounced variations

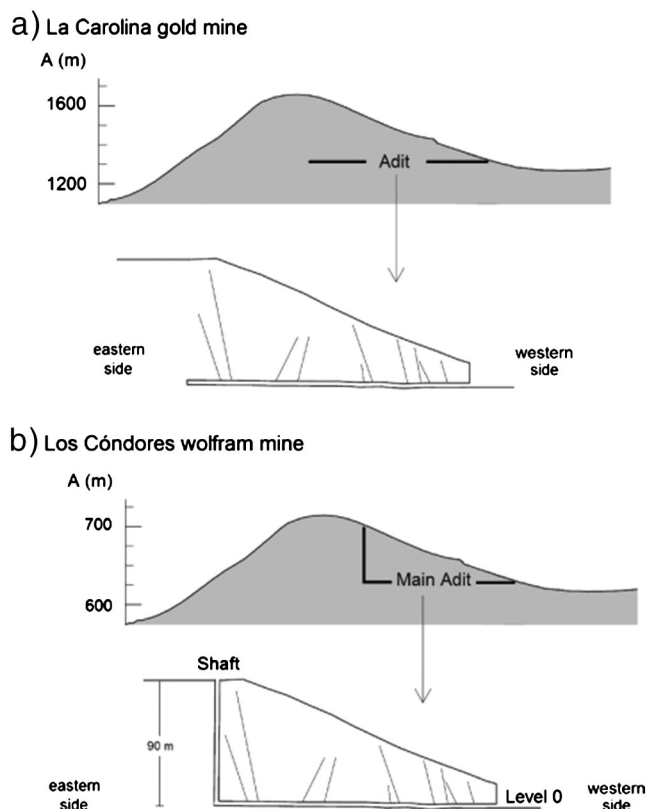


Fig. 2. Illustrations of the topographical scale diagram of the (a) La Carolina gold mine and (b) Los Cóncores wolfram mine. A (y axis) represents the altitude, indicating that the maximum depths are approximately 300 and 90 m for La Carolina and Los Cóncores mines, respectively.

due to exchanges between the cave air, the fracture system, the outside, the sediments and the water in the cave. Air radon concentration is one of these parameters which have important variations related to changes in environmental parameters.

The term “emanation” is used in reference to the release of radon from the mineral particle, while the term “exhalation” refers to release of radionuclide from the surface to the environment. Emanation and exhalation are complex processes; in part because they are influenced by a wide number of factors, being the location of uranium and radium isotopes a main factor. Other factors that can influence the release of radon to the environment are particle size and rock morphology,

Table 1

Distribution of ^{222}Rn in the La Carolina Mine during the summer and winter seasons. The position of the CR-39 detectors is measured from the main entrance of the mine (see Fig. 2a).

Summer		Winter	
CR-39 position (m)	^{222}Rn (Bq m^{-3})	CR-39 position (m)	^{222}Rn (Bq m^{-3})
20	1700 ± 140	21	695 ± 58
85	3840 ± 310	33	657 ± 54
99	4580 ± 370	47	655 ± 55
118	4510 ± 360	51	675 ± 55
118	4530 ± 360	61	707 ± 58
132	5000 ± 400	81	680 ± 56
164	5190 ± 420	106	819 ± 67
164	5120 ± 410	126	836 ± 72
243	5600 ± 450	146	1056 ± 86
		168	2376 ± 186
		182	3488 ± 281
		189	3049 ± 249
		209	3123 ± 252
		218	3952 ± 321
		250	4968 ± 406

Table 2

Distribution of ^{222}Rn in Los Cóncores Mine during the summer and winter seasons. The position of the CR-39 detectors is measured from the main entrance of the mine (see Fig. 2b).

Summer		Winter	
CR-39 position (m)	^{222}Rn (Bq m^{-3})	CR-39 position (m)	^{222}Rn (Bq m^{-3})
17	446 ± 36	4	149 ± 13
50	469 ± 39	14	201 ± 17
75	442 ± 37	57	179 ± 15
103	457 ± 38	107	165 ± 14
150	504 ± 42	153	196 ± 17
198	483 ± 40	220	181 ± 15
251	689 ± 55	226	265 ± 22
300	648 ± 53	239	235 ± 20
357	529 ± 44	286	256 ± 21
400	652 ± 43	322	262 ± 21
425	580 ± 47	355	257 ± 21
432	620 ± 50	403	325 ± 27
		420	670 ± 55
		450	473 ± 39

moisture content of the rock, pore structure, barometric pressure, temperature, radium distribution and vegetation effects (Mudd, 2008; Sharma, 1997).

The transport of radon through the pore space is due to diffusion and advection mechanisms. The molecular diffusion is the dominant mechanism in the intergranular channels, capillaries and small pores, while the advection by air flow is the principal mechanism in macropores. The heterogeneity of the geological material is a source of large variation of radon diffusion in a porous medium, with respect to the theoretical assumptions. Mica and vermiculite, which are flaky minerals, have a shape factor that causes the diffusion coefficient to be one-half to one-third of the theoretical value. Clay and shales contain significant proportions of flaky minerals, usually oriented so as to hinder vertical movement. They retard diffusion to a greater extent than a porous medium having the same porosity but consisting of spherical particles (Cigna, 2005). The advective transport process, resulting from temperature differences between the underground and the surface, is of greater importance than molecular diffusion, because it gives to ^{222}Rn the chance to travel over large distances in the subsurface before its decay (Sharma, 1997). In addition to exhalation and emanation rates, the crucial factor that determines the radon concentration inside caves and tunnels of subterranean environments is the air motion, because the advective air flow is usually the dominant transport mechanism of the gas at this scale.

In caves with two or more entrances at different levels, the air movements inside the cave are due to difference of external and internal air densities only caused by varying external temperatures. This usually leads to consider that the air temperature in caves is nearly constant. This is a temperature-induced air movement named “chimney-effect” (Atkinson et al., 1983).

The chimney-effect occurs when the pressure exerted at the entrance of a mine, by the column of air inside, differs from the pressure of external air. Since the density of the air depends upon temperature, in first approximation the pressure difference is proportional to the temperature difference between the inside and the outside of mine:

$$p_{in} - p_{out} \approx -gh \frac{\rho_{in}}{T_{out}} (T_{in} - T_{out}) \quad (1)$$

Here, h is the elevation difference between the upper and lower cave entrances, g is the acceleration due to gravity and the subscripts *int* and *out* refer to the mean internal and external air pressure (p), air density (ρ) and temperature (T), see Fig. 3 (Atkinson et al., 1983; Hakl, 1997; Hakl et al., 1996, 1997). According to this relationship, when the interior air is colder than the outside air ($\Delta T < 0$), the pressure exerted by the tunnel air will be greater than the pressure outside ($\Delta p > 0$) and the

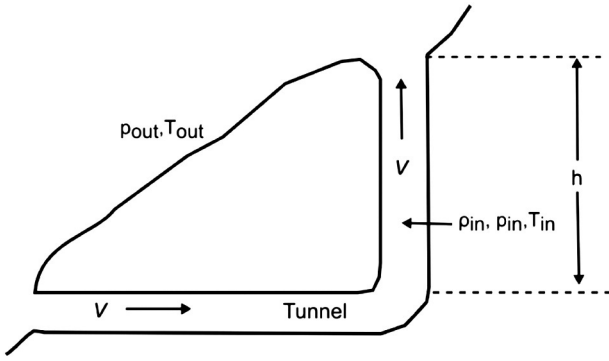


Fig. 3. Illustration of the chimney-effect wind in a simple cave.

air will blow out through the lower entrance. When the outside conditions are colder than inside ($\Delta T > 0$), the air movement is from the lower to the upper entry ($\Delta p < 0$). Chimney-effect airflow reverses its direction seasonally and also might reverse daily at some times of the year. At this approximation level, which is excellent for $h < 1000$ m, variations in atmospheric pressure have no an important effect on the advection process (Atkinson et al., 1983; Haki et al., 1996, 1997).

The seasonal pattern of radon concentration produced by chimney-effect airflow depends on the configuration and connection of underground cavities, passages, fissures and fracture system. For horizontal caves with two entrances and for caves with most passages and openings to the exterior located above the entrance level, a typical seasonal change in radon concentration is obtained, with a maximum in summer and minimum in winter (Kies and Massen, 1995; Haki et al., 1992, 1997). For caves where there is a main gallery with openings located below the entrance level the seasonal radon pattern has winter maxima and summer minima, as the airflow enters into the cave from below. As usually this air is radon enriched, it leads to higher radon concentrations in winter than in summer, when fresh air entering through the main entrance is loaded gradually, along with radon during its movement trough the cave (Kies and Massen, 1995; Haki et al., 1996).

The radon concentration pattern in caves with only one entrance (blind end systems) is more complex to analyze as the airflow can have opposite directions on the entrance cross section, and then, the radon concentration will depend of the vertical coordinate (Atkinson et al., 1983). In spite of this difficulty, simple considerations about the airflow stream allow to explain the seasonal changes in radon distribution patterns. When the cave air is warmer than the outside air (winter), the lighter air of the fracture system, which is radon-enriched, rises, and pushes cave air out through the exit. During this movement, the air can be enriched in radon from the fracture system and from the sediments. As higher temperature differences move a greater air volume, the radon concentration of the air entering to the cave galleries from below decreases when the speed of air flowing through the fracture system is increased. In summer, when the cave air is colder than the outside air, the described process reverses, the direct inflow of fresh air from outside into the cave leads to the lowest radon concentrations.

Many subterranean mines are typical examples of one or other described cave configurations, where the openings are entrances, long fractures of the rock or secondary tunnels. Despite the complexity of radon exhalation and transport processes inside the mine tunnels, the analysis of radon concentration distribution can be made with the help of models like the advective reacting diffusion equation, (Socolofsky and Jirka, 2005; Eff-Darwich et al., 2008; Wigley, 1967; Atkinson et al., 1983; Nazaroff, 1992) which in the one dimensional case is represented by the expression:

$$\frac{\partial C(x,t)}{\partial t} - D \frac{\partial^2 C(x,t)}{\partial x^2} + v \frac{\partial C(x,t)}{\partial x} = \frac{RL}{S} - \lambda C(x,t) \quad (2)$$

where C [Bq m^{-3}] is the radon concentration, D [$\text{m}^2 \text{h}^{-1}$] is the diffusion coefficient of radon in open air, v [m h^{-1}] is the air velocity, $\lambda = 7.54 \cdot 10^{-3} \text{ h}^{-1}$ is the decay constant of radon, R [$\text{Bq m}^{-2} \text{h}^{-1}$] is the exhalation rate, S [m^2] is the cross section and L [m] the peripheral length of tunnel. The second term on the left hand side corresponds to normal diffusion, while the third describes convection or advection. On the right hand side, the terms describe the release of radon into the mine environment and the radon decay process.

In order to apply Eq. (2) to mine tunnels, some assumptions should be made. The conditions should be such that the tunnel can be treated as if it was a cylindrical tube with constant cross section and uniform air velocity. Furthermore, the value of radon diffusion length ($2\sqrt{Dt}$) for the measurement time, should be similar to the radius of cross section of the tunnel, to assure that the radon concentration is approximately uniform on the cross section, i.e. the radon concentration is only function of the x coordinate.

One special solution of Eq. (2) is appropriate to analyze the patterns of radon in tunnels. It is obtained disregarding the diffusion term of Eq. (2), as the advection process is the principal transport mechanism of radon. In this case and under steady conditions, the radon concentration at any distance x from the tunnel entrance can be described by the solution:

$$C(x) = C_{\infty} (1 - e^{-\lambda/x}) \quad (3)$$

where $C_{\infty} = RL/(S\lambda)$, the contour condition $C(0) = 0$ is used, and it is assumed that the air flux at entrance has zero radon concentration. This equation assumes a homogeneous medium where the diffusion takes place. The presence of secondary air flow tributaries, like secondary tunnels and cracks connected with the principal adit, modify the concentration pattern given by Eq. (3). The exact form of the radon concentration pattern depends on the velocity and radon content of the tributary stream air flow (Atkinson et al., 1983).

3. Results and discussion

Radon can be emanated from underground at a rate, which is influenced by several factors, such as the lithology of the bedrock, structural features such as faulting and jointing, soil physical and chemical properties (Sharma, 1997). Previous studies were performed by our research group on the lithology of the bedrock and structural features from the main adits of the mines. They showed that both La Carolina and Los C ndores have homogeneous mineral composition (Da Silva et al., 2011). Additionally, studies about the spatial variations of the radon concentration in these mines were conducted during the summer season. The results, shown in Table 1, revealed that the radon concentration varied significantly throughout the La Carolina gold mine. The lowest values can be observed near the mine entrance. Moving towards its interior, the concentration increased rapidly first, then more slowly, reaching a plateau at the end of the main adit. This value remained practically unchanged in its secondary adits. This behavior allows us to suppose that the main adit of La Carolina mine behaved as blind end system. However, the radon concentration from underground can also be influenced by the mode of transport (diffusion or fluid flow), and atmospheric influences such as precipitation, wind, and temperature (Sharma, 1997). Thus, it would be interesting to understand what happens to the radon concentration inside the mines in extreme conditions. These arguments led us to undertake new measurements in the mines during the winter season.

3.1. La Carolina Mine

Table 1 shows the distribution of the radon concentration inside La Carolina mine as a function of distance from its entrance, during summer and winter seasons. From this table, one can observe that ^{222}Rn

concentration values are higher in summer than in winter. In addition, it is observed that there is a very different behavior of the radon concentration pattern inside the mine during both seasons. This difference can be better understood with the graph shown in Fig. 4. In summer, the radon concentration grows from the entrance, roughly following a saturation curve. Whereas in winter, the radon concentration is nearly constant from the mine entrance to a point situated at 150 m, beyond this point the concentration rapidly increases. At the end of the main adit, both distributions tend to equalize.

This seasonal change in the radon concentration pattern could be explained by the existence of an air current produced by chimney effect inside of the tunnel (as the mean temperature differences, $\Delta T (T_{in} - T_{out})$, are $\Delta T_{winter} = 10^\circ\text{C}$ and $\Delta T_{summer} = 1^\circ\text{C}$). During the winter, the air stream would flow between two inputs at different levels, from the mine entrance on the lowest level to an unknown (dismantled) duct, which connects the main adit to the outside. This unknown duct would be located at 150 m from the mine entrance. In cold season, the entry of air with low radon content appreciably decreases the radon concentration inside the mine; and then from the point of view of air movement, the tunnel portion extending to a distance of 150 m behaves as a tunnel with two entrances at different levels. Beyond this point, the airflow is stagnant and then, the radon concentration increases like in a tunnel with only one entrance. We have not measured data of air flow in the tunnel, but the existence of air stream inside the tunnel was verified during winter season.

In summer season, chimney effect should also exist, likely generating an airstream in the opposite direction, i.e. towards the entrance of the mine. Possibly due to the small difference between outside and inside average temperature in the warm season ($\Delta T = T_{in} - T_{out} = 1^\circ\text{C}$), there is no airflow, and then the principal adit of La Carolina mine behaves as a blind tunnel during the summer season. The curve obtained fitting the summer concentration data of radon concentration in function of distance, with the solution given in Eq. (3) is shown in Fig. 4. The parameters obtained fitting the summer data are $C_\infty = 5468 \pm 253 \text{ Bq m}^{-3}$ and $v = 0.46 \pm 0.01 \text{ cm h}^{-1}$. This best-fit was obtained by the least squares minimization ($R^2 = 0.97$). The low value of air velocity obtained gives support to the hypothesis suggesting that there is not air movement during the summer in La Carolina mine, i.e. the air is stagnant. The same result is obtained if we assume that the mine entrance is moved to the distance of $x_0 = 150 \text{ m}$ in the winter season (see the dotted curve in Fig. 4).

After obtaining these results, the miners responsible for this mine performed a careful inspection and confirmed the existence of a secondary opening approximately 150 m from the mine entrance.

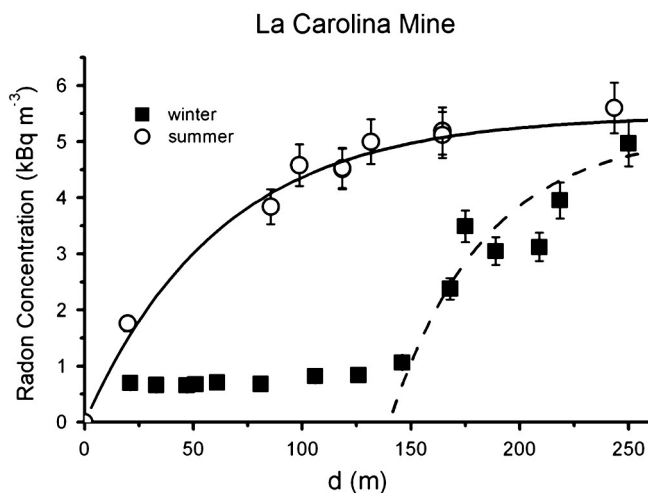


Fig. 4. Distribution pattern of radon concentration in the La Carolina mine. The solid and dotted curves represent the same fit using Eq. (3). The best-fit was obtained by the least squares minimization ($R^2 = 0.97$).

3.2. Los Cóncores Mine

The values of radon concentration along of principal adit of Los Cóncores tungsten mine during the summer and winter seasons are shown in Table 2. Similar to what happens to the case of La Carolina gold mine, the results show that the radon concentration values are higher in summer than in winter. However, there is a constant relationship between mean values of radon concentrations (summer/winter), being around 2.5. The particular arrangement of the main adit of Los Cóncores could explain the seasonal variation of radon levels. This mine is a clear example of underground environments which have openings to the outside, situated at two different levels at least (see Fig. 2b): the mine entrance at zero level and the shaft. In cold season, when the environmental conditions are favorable to chimney-effect ($\Delta T_{winter} = 7^\circ\text{C}$), the larger size of shaft cross-section allows considerable airflow in the main adit, with consequent entry of air from outside by the lower entrance, which significantly decreases the radon concentration inside the mine.

In summer, inside the principal adit of Los Cóncores, the chimney effect should produce an air flow in opposite direction to the winter one (from the shaft to the entrance at zero level). If this was the case, the radon concentration pattern should follow a saturation curve increasing from a low value near the shaft. As can be observed in Fig. 5, the radon concentration pattern does not follow this behavior. While the value of temperature difference ($\Delta T_{summer} = -4^\circ\text{C}$) could be enough to maintain the airflow by chimney-effect during the daylight hours, the distortion of this pattern can be caused by the existence of unknown (not taken into account) tunnels or entrances communicating the principal adit with the outside. These ducts could allow the flow into the mine from the direction of the radon-rich fracture system to the principal adit, resulting in high radon levels inside the mine, or introduce fresh air poor in radon producing the reverse effect.

As can be seen in Fig. 5, at a site localized at approximately 200 m of zero level entrance, the concentration distribution patterns of radon have a wide step, in both seasons. The existence of an unknown duct, which connects the main adit to the outside at this point, would explain this fact. If the opening of the duct were located below the opening of the shaft, during winter the chimney-effect would generate an air stream from the entrance at zero level (main flow) and from the opening of duct (secondary flow), towards the shaft. Both the main and the secondary air currents are loaded with radon on their way to the shaft, and they add their contributions giving rise to the jump that appears in the radon concentration pattern of winter.

During summer, the air current produced by chimney effect flows from the shaft to both entrances, as both are below the opening of

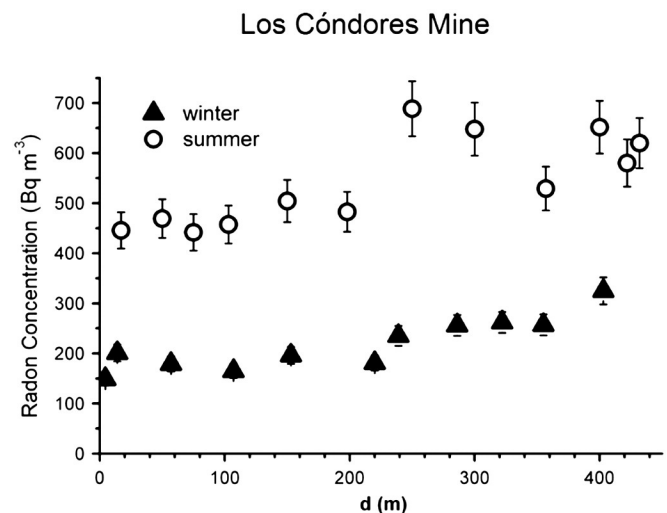


Fig. 5. Distribution pattern of radon concentration in the Los Cóncores mine.

shaft. The air stream would be split in two currents at the mentioned point (200 m entrance at level 0), yielding the abrupt change of the radon concentration value at this site. After the measurements of radon concentration inside of Los Códoreos mine, a careful inspection of the mine was made and it was found that about 200 m of the mine entrance there is indeed a tunnel large enough to sustain the hypothesis mentioned above, explaining the wide step in radon concentration patterns.

4. Conclusions

The results obtained from this study in La Carolina and Los Códoreos mines, show that the radon concentration values are higher in summer than in winter, and that the pattern distributions of radon concentration in summer are very different from the one in winter, for both mines. The studies performed during the summer season, suggest that the main adit of La Carolina gold mine behaves as a blind end system. The measurements of radon concentrations performed in winter season reveal that La Carolina gold mine behaves as a system with two entrances at different levels. This is due to the fact that the chimney effect is the main factor that sets the radon concentration pattern in winter. In the case of Los Códoreos mine, the radon concentration pattern distribution is principally established by the air current due to the chimney-effect in summer and winter seasons. In both cases, the analysis of radon pattern distribution appears as a good method to trace air currents, and then localize unknown ducts, fissures or secondary tunnels in subterranean environments.

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References

Anjos RM, Veiga R, Soares T, Santos AMA, Aguiar JG, Frascá MHBO, et al. Natural radionuclide distribution in Brazilian commercial granites. *Radiat Meas* 2005;39:245–53.

Anjos RM, Veiga R, Macario K, Carvalho C, Sanches N, Bastos J, et al. Radiometric analysis of Quaternary deposits from the southeastern Brazilian coast. *Mar Geol* 2006;229:29–43.

Anjos RM, Veiga R, Carvalho C, Macario K, Gomes PRS. Geological provenance of Quaternary deposits from the southeastern Brazilian coast. *Nucl Phys A* 2007;787:642c–7c.

Anjos RM, Umisedo N, da Silva AAR, Estellita L, Rizzotto M, Yoshimura EM, et al. Occupational exposure to radon and natural gamma radiation in the La Carolina, a former gold mine in San Luis Province, Argentina". *J Environ Radioact* 2010a;101:153–8.

Anjos RM, Macario KD, Lima TA, Veiga R, Carvalho C, Fernandes PJF, et al. Correlations between radiometric analysis of Quaternary deposits and the chronology of prehistoric settlements from the southeastern Brazilian coast. *J Environ Radioact* 2010b;101:75–81.

Anjos RM, Juri Ayub J, Cid AS, Cardoso R, Lacerda T. External gamma-ray dose rate and radon concentration in indoor environments covered with Brazilian granites. *J Environ Radioact* 2011;102:1055–61.

Atkinson TC, Smart PL, Wigley TML. Climate and natural radon levels in Castleguard Cave, Columbia Icefields, Alberta, Canada. *Arct Alp* 1983;15:487–502.

Ball TK, Cameron DG, Colman TB, Roberts PD. Behaviour of radon in the geological environment: a review. *Q J Eng Geol Hydrogeol* 1991;24:169–82.

Cigna AA. Radon in caves. *Int J Speleol* 2005;34:1–18.

Da Silva AAR, Yoshimura EM. Track analysis system for application in alpha particle detection with plastic detectors. *Radiat Meas* 2005;39:621–5.

Da Silva AAR, Valladares DL, Anjos RM, Velasco H, Rizzotto M, Yoshimura EM. Assessment the health hazard from ^{222}Rn in old metalliferous mines in San Luis, Argentina. *Water Air Soil Pollut* 2011;218:371–86.

Eff-Darwich A, et al. Natural air ventilation in underground galleries as a tool to increase radon sampling volumes for geologic monitoring. *Radiat Meas* 2008;43:1429–36.

Estellita L, Santos AMA, Anjos RM, Yoshimura EM, Velasco H, da Silva AAR, et al. Analysis and risk estimates to workers of Brazilian granitic industries and sandblasters exposed to respirable crystalline silica and natural radionuclides. *Radiat Meas* 2010;45:196–203.

Grossi C, Vargas A, Camacho A, López-Coto I, Bolívar, JPB, Xia Yu, et al. Inter-comparison of different direct and indirect methods to determine radon flux from soil. *Radiat Meas* 2011;46:112–8.

Hakl J. (Ph.D. thesis) Application of Radon-222, as a natural tracer in environmental studies. Lajos Kossuth University; 1997. [<http://ganymedes.lib.unideb.hu:8080/dea/bitstream/2437/80292/2/ertekezes.pdf>].

Hakl J, Hunyadi I, Csige I, Géczy G, Lénárt L, Töröcsik I. Outline of natural radon occurrences on karstic terrains of Hungary. *Radiat Prot Dosim* 1992;45:183–6.

Hakl J, Csige I, Hunyadi I, Várhegyi A, Géczy G. Radon transport in fractured porous media - experimental study in caves. *Environ Int* 1996;22:S433–7.

Hakl J, Hunyadi I, Csige I, Géczy G, Lénárt L, Várhegyi A. Radon transport phenomena studied in karst caves - international experiences on radon levels and exposures. *Radiat Meas* 1997;28:675–84.

Hoskold HD. Official report upon the mines, mining, metallurgy and mining laws of the Argentine Republic. Buenos Aires, Argentina: Ministry of Agriculture, Commerce and Industries; 1904.

Howarth CB, Miles JCH. NRPB Report W-44: results of the 2002 NRPB intercomparison of passive radon detectors. Chilton, UK: NRPB, National Radiological Protection Board; 2003.

Kies A, Massen F. Radon and underground climate in the Moestroff Cave. *Gas Geochem Northwood* 1995;63–71.

Kies A, Massen F, Tosheva Z. Influence of variable stress on underground radon concentrations. *Geofisica Int* 2002;41:325–9.

Miller BL, Singewald JT. The mineral deposits of South America. New York: McGraw-Hill Book Co.; 1919 [598 pp.].

Mudd GM. Radon sources and impacts: a review of mining and non-mining issues. *Rev Environ Sci Biotechnol* 2008;7:325–53.

Nazaroff WW. Radon transport from soil to air. *Rev Geophys* 1992;30:137–60.

Orlando C, Orlando P, Patrizii L, Tommasino L, Tonnarini S, Trevisi R, et al. A passive radon dosimeter suitable for workplaces. *Radiat Prot Dosim* 2002;102:163–8.

Rastall RH, Wilcockson MA. Tungsten ores: London, Imperial Institute Monograph on Mineral Resources with Special Reference to the British Empire, John Murray; 1920 [81 pp.].

REM. Red de Estaciones Meteorológicas. <http://www.clima.edu.ar/app/Index.asp>, 2011.

Sharma PV. Environmental and engineering geophysics. Cambridge: Cambridge University Press; 1997 [372 pp.].

Shweikani R, Durrani SA. Thoron contributions in radon measurements in the environments. *Radiat Meas* 1995;25:615–6.

Socolofsky SA, Jirka GH. CVEN 489–501: special topics in mixing and transport processes in the environment. 5th ed. Coastal and Ocean Engineering Division, Texas A&M University; 2005.

Steinitz G, Piatibratova O, Kotlarsky P. Possible effect of solar tides on radon signals. *J Environ Radioact* 2011;102:749–65.

Tripaldi A, Ciccioli PL, Alonso MS, Forman SL. Petrography and geochemistry of late Quaternary dune fields of western Argentina: provenance of aeolian materials in southern South America. *Aeol Res* 2010;2:33–48.

Veiga R, Sanches N, Anjos RM, Macario K, Bastos J, Iguatemy M, et al. Measurement of natural radioactivity in Brazilian beach sands. *Radiat Meas* 2006;41:189–96.

Viñas R, Eff-Darwich A, Soler V, Martín-Luis MC, Quesada ML, Nuez J. Processing of radon time series in underground environments: implications for volcanic surveillance in the island of Tenerife, Canary Islands. Spain *Radiat Meas* 2007;42:101–15.

Werner ABT, Sinclair WD, Amey EB. International strategic mineral issues summary report - tungsten. US Geological Survey Circular 930-O; 1998.

Wigley TML. Non-steady flow through porous medium and cave breathing. *J Geophys Res* 1967;72:3199–205.