

Soil-gas radon monitoring in an active granite quarry from central Portugal

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Abstract. Seven soil-gas radon monitoring stations were placed along the active front of a granite quarry in Canas de Senhorim, Central Portugal, recording continuously for 81 days. Important differences in the radon concentration were found between stations, with average values comprised between 102 and 2982 Bq m⁻³, which can be explained by the local presence of uranium anomalies in the regional late-orogenic Hercynian granite, usually associated with faults. One of the boreholes exhibits large radon anomalies lasting for several days, and two, contrary to the others, show a clear daily periodic behaviour, with minima around 19:00 LT and maxima around 07:00 LT. The different patterns observed in stations placed at such a short distance (< 100 m) has no clear explanation and deserves further investigation. Data analysis shows no evidence of soil-gas radon concentration changes during explosions carried out at the quarry. This is likely to result from the absence of a progressive stress field affecting the rock, as typically occurs before an earthquake.

1 Introduction

The isotope ²²²Rn (hereafter designated by radon) is a radioactive noble gas with a half-life of 3.8 days. It is produced in uranium-bearing materials by alpha decay from radium (²²⁶Ra) as part of the uranium (²³⁸U) series.

Radon is considered a potential earthquake precursor since micro-fracturing and increased stress levels associated with an earthquake event are thought to enhance the release of radon gas from rocks (Thomas, 1988). Several studies have reported anomalous radon concentrations associated with seismic events (e.g. Igarashi et al., 1995; Steinitz et al., 1999; Planinic et al., 2001; Steinitz et al., 2003; Hartmann and

Levy, 2005; Yasuoka et al., 2006; Crockett et al., 2006) but the corresponding physical mechanisms are poorly understood. Since direct experiments on the effect of earthquakes on radon release are difficult to conduct, a possible alternative is to use man-made explosions as an earthquake surrogate and investigate the corresponding effect on radon concentration (e.g. Zafrir et al., 2009). In principle the pressure gradient associated to an explosion is capable to enhance radon transport in the fractured media, leading to increased soil-gas radon concentration.

This study addresses the effect of man-made explosions on soil-gas radon concentration. In order to assess the influence of explosion events, radon monitoring was carried out in an active granite quarry located in central Portugal.

2 Geological setting

The Agrepor quarry produces high quality granite aggregates for use as building materials. The quarry is located in the vicinity of the town of Canas de Senhorim, Central Portugal (Fig. 1), and actually extends for about 1 km², including extraction areas, milling facilities, and administrative buildings. The local granite corresponds to the dominant facies of the late-orogenic Hercynian pluton known as the Beiras batholith, which is a medium to coarse-grained porphyritic biotite granite. Other minor varieties also occur in the batholith, ranging from fine to medium grained and from muscovite to muscovite-biotite granites. The batholith was emplaced in a complex of metasedimentary schists and greywackes (CSG) of late Precambrian to lower Palaeozoic age (Pereira et al., 1998). A few veins and lenses of aplites and pegmatites, usually a few meters thick, cut the granites and the country rock metasediments.

The Beiras granite is associated with an important event of uranium mineralization, as indicated by the presence in the region of 60 old mines that were exploited until the end



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Fig. 1. Location of the Agrepor quarry, close to the town of Canas de Senhorim, Central Portugal. Hercynian granites are represented with grey colour.

of last century. A total of 4000 t of U_3O_8 were produced, with an important contribution from the Urgeiriça mine, located close to the Canas de Senhorim town and the Agrepor quarry. Uranium prospecting work carried out in the region allowed to identify the presence of thousands of radiometric anomalies, related to the local presence of uranium, without economic interest due to insufficient ore grade or small total volume. These anomalies, however, can present an important environmental risk due to their high levels of radon production, as shown by Pereira et al. (2010). Most anomalies occur in association with faults, namely of the families $N75 \pm 5^\circ E$, $N55 \pm 5^\circ E$, and $N35 \pm 5^\circ E$, where uranium can reach concentrations of several hundred ppm in excess of the granite background (6–15 ppm). These faults were active since Hercynian times, behaving as normal, inverse or strike-slip faults in response to variations of the stress field, particularly during the Alpine orogeny (Pereira et al., 2010). Uraninite is apparently the primary mineralogical support of uranium in the Beiras batholith. Taking into consideration that this mineral can be readily leached in contact with meteoric water, and that groundwater in granite massifs preferentially moves through fractures, it is not surprising that fault-filling materials (brecciated granite and quartz veins) can provide an ade-

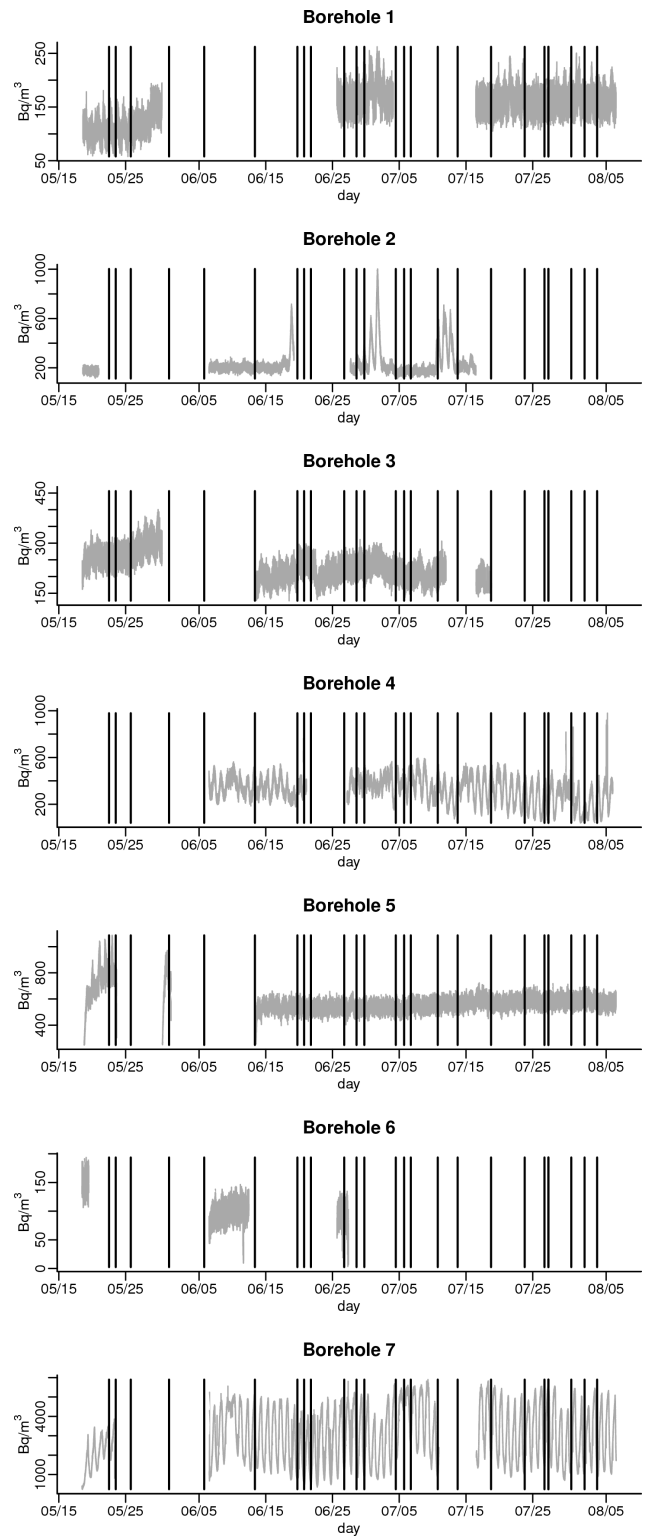


Fig. 2. Time series (1-min rate) of radon concentration over the complete data acquisition period (16 May to 6 August). The vertical solid lines indicate the time of explosions in the quarry.

Table 1. Data summary: minimum (min), mean, maximum (max), and standard deviation (stdev).

	min	mean	max	stdev
1	57.9	148.9	262.2	29.5
2	112.2	231.2	1002.6	107.2
3	127.5	228.2	455.8	37.6
4	40.15	321.9	979.0	108.2
5	248.0	575.4	1086.5	77.7
6	2.4	102.1	194.7	23.3
7	239.7	2982.0	5898.9	1428.5

quate environment for uranium deposition. Secondary phosphates, like autunite and torbernite, and iron oxo-hydroxides can be observed in these materials. The presence of such mineralization is usually easily recognized with the use of portable gamma-ray scintillometers, due to its superficial nature, and secondary phosphates can be macroscopically observed in the most intense ones. Indeed, the majority of the referred uranium mines were exploited as open pits and rarely below 30 to 50 m. An important exception is the underground Urgeiriça mine, related with a deep vein with strike N60E, with pitchblende mineralization, where works were carried out until a depth of 500 m. As expected from the geological setting described, soil-gas radon concentrations are quite high in the local granite: Pereira et al. (2003) report an average concentration of 110 kBq m^{-3} at a depth of 0.80 m in the region of Canas de Senhorim. However, these concentrations can be much higher along specific faults, as already referred, reaching in this case several hundred to a few million Bq m^{-3} (21 MBq m^{-3} were obtained in a measurement carried out over the Urgeiriça vein). Average uranium concentration of the granite was estimated by the same authors to be 8.6 ppm, reaching a few dozen to several hundred ppm in some fault-filling materials (Pereira et al., 2010). As a consequence, indoor radon in dwellings of the towns of Canas de Senhorim and Urgeiriça is quite high, on average 298 and 285 Bq m^{-3} , respectively, (geometric mean), with a significant proportion exceeding the recommended annual average of 400 Bq m^{-3} . The same pattern can be observed in other towns of Central Portugal with similar geology, like Guarda and Oliveira do Hospital (Pereira et al., 2003).

3 Methods

A total of 7 monitoring stations were placed close to the active exploitation front of the quarry, approximately distributed along its extension: 5 were installed outside the perimeter and 2 inside the working area. The distance between stations was typically close to 100 m, and the distance to programmed explosions in the quarry ranged from a few dozen meters up to ca. 250 m. In each place a hole with a few meters depth was drilled directly in the rock with appropriate

equipment available at the quarry. A PVC tube was inserted in each hole carrying an RM-70 detector (Aware, USA), sensible to alpha, beta, and gamma radiation/X-rays above 10 keV, with counting sensitivity to high energy charged particles approaching 100%. This is a GM pancake detector, with an effective diameter of 1.125 inches and thickness of 0.06 inches. A fan-filter system was used to increase detection limits. Background gamma radiation arriving to the detector is approximately constant in place, thus counting variations are due to correlative changes of radon concentration. The detectors were typically positioned at a depth comprised between 1 and 2 m, and connected through wires to an assembly placed at the surface and protected inside a box, composed of a palmtop computer for data registration and power supply batteries. Monitoring was carried out during 3 months, in spring/summer conditions with low moisture, and the exact time of each explosion was registered manually. Radiation levels were measured in each station with 1 min intervals. Radiation levels, expressed as counts per minute (CPM), were subsequently converted to Bq m^{-3} through a calibration procedure carried out in the Laboratory of Natural Radioactivity of the Department of Earth Sciences of the University of Coimbra, with the use of an appropriated calibration chamber (Genitron-Saphymo, Germany), a certified radon source NIST SRM-4973 (National Institute of Standards & Technology, USA), and a high precision radon monitor AlphaGUARD Pro (Genitron-Saphymo, Germany). The procedure is described in detail in Neves et al. (2009).

A survey of the quarry with a portable gamma-ray spectrometer GR-130G (Exploranium, Canada) equipped with a NaI detector of 2×1.5 inches was also carried out in order to characterize background gamma radiation levels and the presence of possible anomalies. The results of the survey carried out with the gamma-ray spectrometer show that the heterogeneity of uranium distribution in the Beiras granite is reflected in the quarry. Background values of $0.20\text{--}0.25 \text{ nGy h}^{-1}$ were observed for the absorbed dose (directly in contact with the rock), with a significant increase in some fractures of the $N55 \pm 5^\circ \text{ E}$ and $N35 \pm 5^\circ \text{ E}$ systems that crosscut the quarry, up to 0.80 nGy h^{-1} . These faults are subvertical and submetric and do not occur frequently, the higher absorbed dose reflecting some degree of uranium enrichment.

4 Results and discussion

The experimental (1-min) data at the 7 boreholes and over the complete period of acquisition from 16 May to 6 August 2001 (81 days) are displayed in Fig. 2. A statistical summary of the data is given in Table 1. The most striking characteristic of the data is the big diversity of the type of signals obtained from boreholes only a few tens of meters apart. Neighbouring sites exhibit very different average values, ranging from ca. 100 up to ca. 3000 Bq m^{-3} . The temporal patterns

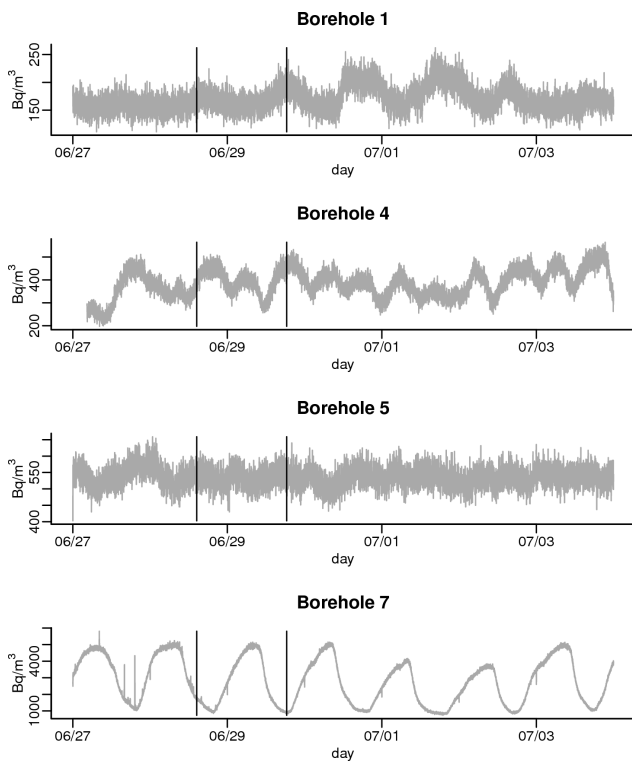


Fig. 3. Time series (1-min rate) of radon concentration for the period from 16 July to 4 August. The vertical solid lines indicate the time of explosions in the quarry.

are also very different from one place to another, with some sites exhibiting a non-periodic temporal pattern while others display a clear periodic behaviour (boreholes 4 and 7) with minima occurring in the afternoon, around 19:00 LT (local time) and maxima in the morning, around 07:00 LT. Furthermore, borehole 2 exhibits a few large anomalies in radon concentration (e.g. in early and mid July) lasting for several days which are very similar to the radon signals described by Steinitz et al. (2007) and Barbosa et al. (2010). The results obtained are globally lower than usually observed for soils derived from the regional granite. This likely results from the fact that the monitoring holes were drilled directly in unaltered rock, with a very thin soil cover (< 20 cm). Moreover, the granite of the quarry shows in some sections a low density of fractures, namely joints, presenting suitable characteristics for ornamental stone.

Radon monitoring was hindered by issues of power supply and problems in the field instrumentation (e.g. the upper assembly of one of the stations was destroyed by falling rocks from a close-by explosion and had to be replaced). These field issues led to data loss and some discontinuities in the records (visible in Fig. 2). Therefore, two distinct periods of continuous measurements at boreholes 1, 4, 5, and 7 are considered hereafter (Figs. 3 and 4).

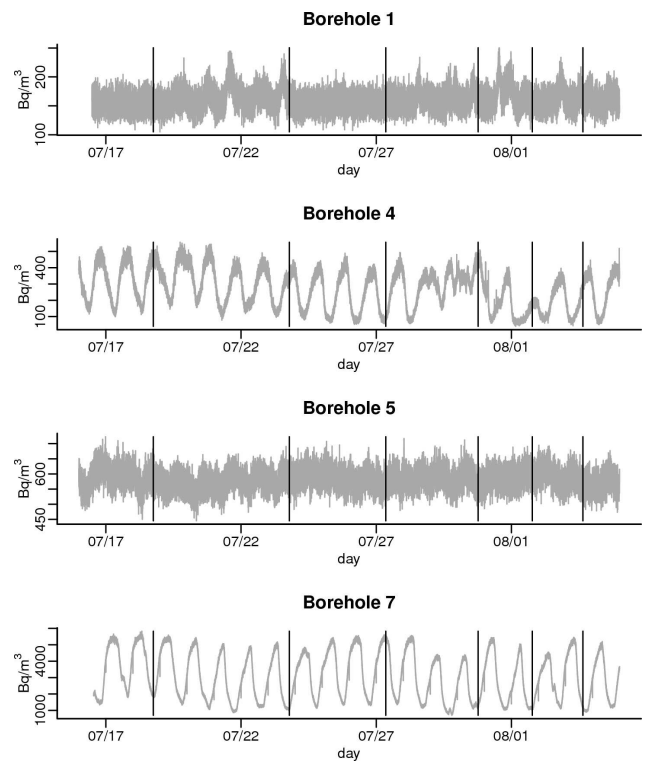


Fig. 4. Time series (1-min rate) of radon concentration for the period from 27 June to 3 July. The vertical solid lines indicate the time of explosions in the quarry.

In order to assess the influence on radon concentration of explosions occurring in the quarry, continuous observations from 27 June to 3 July and from 16 July to 4 August 2001 are considered. Visual inspection of the time series in Figs. 3 and 4 does not reveal any prominent features in soil-gas radon concentration at the moment of the explosions or in the following minutes. This is confirmed by the separate statistical analysis of the radon concentration values before and after the explosions. The boxplots of the data 24 h before and after the explosions for the two periods considered (Fig. 5) don't show any significant change in the mean or variance of radon concentration before and after the explosions.

5 Conclusions

The results from continuous radon monitoring in this active quarry show significant differences in the soil-gas radon concentrations from nearby sites. The average radon concentration ranges from ca. 100 to ca. 3000 Bq m⁻³. These differences can be due to the heterogeneity of uranium distribution in the rock and increased local permeability. Furthermore, two of the sites exhibit a clear daily periodic behaviour,

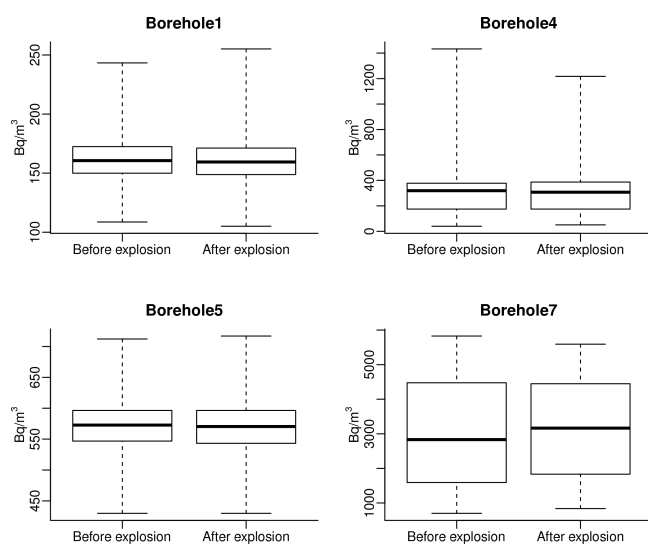


Fig. 5. Boxplots of radon concentration values 24 h before and after the explosion events marked in Figs. 3 and 4.

which is absent at the other locations. The explanation of this effect deserves further investigation.

The main conclusion of this work is that no significant changes in soil-gas radon concentration associated with explosion events are observed at any of the sites. A similar behaviour was observed in a seismic calibration explosion experiment performed in Israel (Zafir et al., 2009).

The exhalation of radon is thought to be enhanced when rocks are subject to stress, which would justify the role of radon as a potential earthquake precursor. Although these results seem to contradict that hypothesis, that is not necessarily so. In fact, in the case of man-made explosions, a progressive stress field effecting the rock, as likely occurs before an earthquake, is absent. Therefore, the earthquake preparatory stage seems to be crucial for the role of radon as a potential precursor.

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References

- Barbosa, S. M., Zafir, H., Malik, U., and Piatibratova, O.: Multi-year to daily radon variability from continuous monitoring at the Amram tunnel, southern Israel, *Geophys. J. Int.*, 182, 829–842, doi:10.1111/j.1365-246X.2010.04660.x, 2010.
- Crockett, R. G. M., Gillmore, G. K., Phillips, P. S., Denman, A. R., and Groves-Kirkby, C. J.: Radon anomalies preceding earthquakes which occurred in the UK, in summer and autumn 2002, *Sci. Total Environ.*, 364, 138–148, 2006.
- Hartmann, J. and Levy, J. K.: Hydrogeological and gasgeochemical earthquake precursors – A review for application, *Nat. Hazards*, 34, 279–304, 2005.
- Igarashi, G., Saeki, S., Takahata, N., Sumikawa, K., Tasaka, S., Sasaki, Y., Takahashi, M., and Sano, Y.: Ground-water radon anomaly before the Kobe earthquake in Japan, *Science*, 269, 60–61, 1995.
- Neves, L. J. P. F., Barbosa, S. M., and Pereira, A. J. S. C.: Indoor radon periodicities and their physical constraints: a study in the Coimbra region (Central Portugal), *J. Environ. Radioactiv.*, 100, 896–904, 2009.
- Pereira, A. J. S. C., Carter, A., Hurford, A. J., Neves, L. J. P. F., and Godinho, M. M.: Evidence for the unroofing history of Hercynian granitoids in Central Portugal derived from Late Palaeozoic and Mesozoic sedimentary zircons, *Solid Earth*, Kluwer Academic Publishers, 173–186, 1998.
- Pereira, A. J. S. C., Neves, L. J. P. F., Godinho, M. M., and Dias, J. M. A.: Natural radioactivity in Portugal: influencing geological factors and implications for land use planning, *Radioproteção*, 2, 109–120, 2003.
- Pereira, A. J. S. C., Godinho, M. M., and Neves, L. J. P. F.: On the influence of faulting on small scale soil-gas radon variability – a case study in the Iberian Uranium Province, *J. Environ. Radioactiv.*, 101, 875–882, 2010.
- Planinic, J., Radoli, V., and Lazanin, Z.: Temporal variations of radon in soil related to earthquakes, *Appl. Radiat. Isotopes*, 55, 267–272, 2001.
- Steinitz, G., Vulkan, U., and Lang, B.: Radon flux at the northwestern segment of the Dead Sea (Dead Sea rift) and its relation to earthquakes, *Israel, J. Earth Sci.*, 48, 283–299, 1999.
- Steinitz, G., Begin, Z. B., and Gazit-Yaari, N.: A statistically significant relation between Rn flux and weak earthquakes in the Dead Sea rift valley, *Geology*, 31, 505–508, 2003.
- Steinitz, G., Piatibratova, O., and Barbosa, S. M.: Radon daily signals in the Elat Granite, Southern Arava, Israel, *J. Geophys. Res.*, 112, B10211, doi:10.1029/2006JB004817, 2007.
- Thomas, D. M.: Geochemical precursors to seismic activity, *Pure Appl. Geophys.*, 126, 241–265, 1988.
- Yasuoka, Y., Igarashi, G., Tokonami, S., and Shinogi, M.: Evidence of precursor phenomena in the Kobe earthquake obtained from atmospheric radon concentration, *Appl. Geochem.*, doi:10.1016/j.apgeochem.2006.02.019, 2006.
- Zafir, H., Steinitz, G., Malik, U., Haquin, G., and Gazit-Yaari, N.: Response of Radon in a seismic calibration explosion, *Israel, Radiat. Meas.*, 44, 193–198, doi:10.1016/j.radmeas.2009.01.002, 2009.