Influence of a component of solar irradiance on radon signals at 1 km depth, Gran Sasso, Italy

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Exploratory monitoring of radon is conducted at one location in the deep underground Gran Sasso National Laboratory (LNGS). Measurements (15-min resolution) are performed over a time span of ca 600 days in the air of the surrounding calcareous country rock. Using both α- and γ-ray detectors, systematic and recurring radon signals are recorded. Two primary signal types are determined: (i) non-periodic multi-day (MD) signals lasting 2–10 days and (ii) daily radon (DR) signals—which are of a periodic nature exhibiting a primary 24-h cycle (θ = 0.48). The local ancillary environmental conditions (pressure, temperature) seem not to affect radon in air monitored at the site. Long-term patterns of daytime measurements are different from the pattern of night-time measurements indicating a day–night modulation of γ-radiation from radon in air. The phenomenology of the MD and DR signals is similar to situations encountered at other locations where radon is monitored with a high time resolution in geogas at upper crustal levels. In accordance with recent field and experimental results, it is suggested that a component of solar irradiance is affecting the radiation from radon in air, and this influence is further modulated by the diurnal rotation of the Earth. The occurrence of these radon signals in the 1 km deep low-radiation underground geological environment of LNGS provides new information on the time variation of the local radiation environment. The observations and results place the LNGS facility as a high-priority location for performing advanced investigations of these geophysical phenomena.

1. Introduction

Radon ($^{222}\text{Rn}$) is a radioactive inert gas formed by disintegration from $^{226}\text{Ra}$ as part of the $^{238}\text{U}$ decay series. The combination of its noble gas character and its radioactive decay makes it a unique ultratrace component for tracking temporally varying natural processes in subsurface systems.

Numerous works have dealt with radon variations in the subsurface geological environment. Large temporal variations of radon are often encountered in air in the geological environment, at time scales from diurnal to annual. Interpretations as to the nature of these variations, unique to Rn, often invoke either above surface atmospheric variations or the influence of subtle active geodynamic processes. It is assumed that the temporal patterns of Rn in the geogas phase are owing to processes affecting its exhalation from the country rock and/or gas transfer processes in the complex consisting of rock porosity and subsurface air space. Environmental influences, particularly atmospheric pressure and temperature, have been proposed for the origin of the periodic signals observed in Rn time series [1–4]. However, other studies indicate that a consistent meteorological influence cannot be identified as giving rise to variability in Rn time series and suggest gravitational tides as an influencing factor on Rn variability [5–9]. Despite the presumed advantages of Rn as a geophysical proxy, understanding of the origin of the different Rn signals remains a challenging task.

The occurrence of radon signals has also been observed at considerable depth, at sites which are within rock systems. At these levels, the air pressure regime is related to the local atmospheric barometric pressure while temperature tends to show stable patterns reflecting the rock temperature at depth. Radon signals characterized by systematical recurrence of patterns in the temporal, geological and geographical domains have been reported from tunnels and boreholes in France [10], from Tenerife at a depth of more than 800 m [11–13] and from the desert in Israel at a depth of more than 100 m [14–17].

The setting of Laboratori Nazionali Gran Sasso (LNGS) offers a unique opportunity to investigate the temporal signals owing to a combination of qualities: (i) underground, at a depth in the order of 1000 m and (ii) low-background radiation environment. First radon measurements and monitoring results in air and the local hydrologic system of LNGS were reported by Bella & Plastino [18,19]. This contribution describes patterns and characteristics of radon signals in air of the immediate surrounding country rock of LNGS. Site parameters and the unsuccessful outcome of an examination as to the influence of local environmental parameters combined with the identification of further exceptional geophysical characteristics in the time series lead us to suggest again the influence of a component in solar irradiance as a driver of radon variation.

2. Experimental set-up

The underground facilities are located in a side tunnel of the 10 km long freeway tunnel crossing the Gran Sasso Mountain ridge from L’Aquila to Teramo. The mountain ridge towering above the laboratory results in an average depth of 1400 m of the laboratory. The laboratory is built within a thick sequence of folded and faulted Mesozoic to Tertiary sedimentary units of the Gran Sasso massif in the central Apennines [20]. The LNGS facility is excavated in limestone and dolomite with 238U activities in the range of 0.42–6.8 ppm [21]. The hydrological situation is such that the laboratory is situated below the regional water table. The facility consists of three large experimental halls, each about 100 m long, 20 m wide and 18 m high and interconnecting service tunnels. The tunnels and rooms of the LNGS facility are lined with concrete walls (60 cm thick), serving as a shield for the laboratory from mechanical, hydrological and environmental radiation [22]. Radon measurements in experiment halls with concrete lining are in the range of 20–150 kBq m$^{-3}$, depending on the ventilation regime [23].

Radon monitoring is performed in a side room, roughly $4 \times 2 \times 2$ m, along one of the service tunnels of the facility (figure 1). The country rock, composed of steeply dipping beds of limestone and marl, is exposed at the side walls and ceiling of the site. The room is separated from the
Figure 1. Layout of LNGS underground facility. Radon measurements in air are conducted at the monitoring site which is located next to a service tunnel (hatched line — fault trace).

service tunnels by a metal partition and tight door (both fitted with a rubber seal), limiting the exchange of air in the room with air in the service tunnel while allowing a good contact with the air of the surrounding country rock.

Radon measurements are performed using α- and γ-ray detectors. Utilization of γ-detector systems for monitoring Rn is based on the detection of γ-radiation from the $^{214}\text{Bi}$ and $^{214}\text{Pb}$. Owing to the short half-lives of the Rn daughters, equilibrium of the Rn and its daughters is achieved after a short time (approx. 100 min). Two $2'' \times 2''$ NaI detectors fitted with a single channel analyser in the range of 50–3000 keV (PM-11 detector; ROTEM Inc., Israel) are placed in parallel and horizontally at the monitoring site to record the γ-radiation emitted from the walls and from radon in the air of the room. The advantages of using these detectors for monitoring of radiation from radon are given by Zafrir et al. [24]. Varying radon levels are recorded by the count rate of the γ-detectors, connected to a datalogger. The direct measurement of radon by α-radiation is performed with a calibrated Alphaguard system (Genitron Gmbh). Ancillary environmental parameters (pressure, temperature, relative humidity) are recorded by the Alphaguard system in the initial phase and ongoing pressure and temperature monitoring was installed in November 2006. Integration times are set to 15 min for the γ-system and 10 min for the α-system. The elapsed time is shown on a decimal-day scale (relative to zero day in 1 January 2005).

The bulk of the results rely on data obtained using two PM-11 γ-sensors operated in tandem during more than 500 days (figure 2 and the electronic supplementary material). The Alphaguard system was operated during an initial stage and also for several intervals in parallel to the γ-detectors. The temporal variation of radon recorded by the γ-detectors was preprocessed in the following manner:

- the observations in the time series of the two γ-sensors were summed as one γ-sensor with an improved sensitivity;
- the resulting time series was smoothed using a 25-h sliding average. Such a decomposition of the time series highlights the different signals contained in it; and
- the residual time series was denoised using a further nine-sample sliding average in order to emphasize some aspects.
Figure 2. Range of radon monitoring intervals using $\gamma$-detectors and coregistration with $\alpha$-detectors. Data gaps are indicated. (Online version in colour.)

3. Results

The main results of the $\alpha$-signal of radon variation and ancillary parameters in the air of the site obtained in the initial stage are shown in figure 3. Upon the closing of the site door, the following features are observed:

- a fast decrease in temperature from a level of 17–18°C to a stable level around 11.5°C with a variation less than $\pm 0.3^\circ$C,
- a parallel increase in humidity to a stable level of around 93%,
- a time varying pressure, and
- a rapid and significant build-up of radon from around 100\,Bq\,m$^{-3}$ to around 450\,Bq\,m$^{-3}$ followed by temporal variation.

The new conditions at the site are attained within 2 days after isolating the site from the laboratory atmosphere and environment. The attained overall radon level of around 400\,Bq\,m$^{-3}$ is relatively low compared with most subsurface geogas in geological environments. Monitoring results (around 40 days) show that the temperature and humidity levels are very stable over time. Small variations in temperature (less than 0.5$^\circ$C) are probably reflecting temperature variations in the adjoining service tunnel. The temporal pattern of radon indicates the occurrence of daily and multi-day (MD) signals, which vary following a pattern that differs from those of pressure and temperature.

Figure 4 shows the temporal variation of radon measurement with $\alpha$-detector (AlphaGuard system) and the simultaneous record from the two $\gamma$-detectors. The two $\gamma$-detectors exhibit very similar variation patterns which are parallel to the $\alpha$-signal. Measurements using the two $\gamma$-sensors and the $\alpha$-detector are instrumentally independent. Thus, operation of the AlphaGuard system in parallel with the $\gamma$-detectors substantiates that the variation in the $\gamma$-radiation represents variation of radon in the air of the room. The recorded $\gamma$-radiation originated from two sources: (i) a constant component owing to the radioactive elements in the wall rock (and concrete), and (ii) a varying component emitted from radon decay products in the air of the site. The count rate of both $\gamma$-sensors is similar, and the systematic difference among them can be attributed to a slight difference in sensitivity and/or owing to their slightly differing positions. The higher count rate of the $\gamma$-sensor, relative to the $\alpha$-detector, reflects the large difference of the sampled volume (owing to the different range of $\alpha$- and $\gamma$-radiation in air; see also [24]). The high correspondence of the radiation pattern recorded by the two $\gamma$-sensors allows referring to them as a single detector by summing the two count rates, thus improving the signal-to-noise ratio.

Taking into consideration the complete dataset, systematic temporal variations in the radon level are observed. The primary variation is manifested as signals lasting one to several days.
Figure 3. Time series of radon (using a $\alpha$-detector) in air at experimental site and local environmental parameters (pressure, temperature and relative humidity). (Online version in colour.)

Figure 4. (a) A 25-day interval of parallel measurements by two $\gamma$-detectors and a $\alpha$-detector displaying a high concordance of the variation. (b) A 10-day detail of the time series. The concordance is further manifested by denoising each time series using a nine-point sliding average (shown as solid lines). Fifteen-minute resolution and legend—for both Figure a,b. (Online version in colour.)
Figure 5. A typical example of MD signals during a time interval of around 100 days. The MD signal is obtained using a 25-h sliding average. The temporal patterns shown in the plot represent the combination of the signals from both of the $\gamma$-sensors (using a 25-h sliding average).

A very high concordance is observed among the time series of these detectors (figure 4a), which is enhanced by denoising each time series using a nine-point sliding average (figure 4b). This concordance enables the following conclusions:

— The radon concentration in the room air varies in a systematic pattern.
— Variations of the order of 50 Bq m$^{-3}$ ($\alpha$-sensor) and around 500 counts/15-min are above the statistical uncertainty and reflect variations owing to environmental and/or geophysical processes.

The measured signal was decomposed using a 25-h sliding average revealing two types of signals in the radon time series—daily radon (DR) signals and MD signals, similar to those encountered in several other sites and locations [15–17,25]. The MD signals, with amplitudes of over 10 kcounts/15-min, span 2 or more days and are non-periodic. A typical representation of the MD signal is given in figure 5 using the smoothed time series showing that they have both symmetric (i.e. similar absolute rate of the rising and decreasing limbs) and asymmetric forms.

The DR signal occurs as a periodic signal but with highly irregularly varying daily amplitude (figure 6). In some days, it appears as if a minor semidiurnal component is also present in the time series. Generally, no relationship can be determined between the MD and DR signals (figure 6a) but in some time intervals, lasting several tens of days, an apparent relationship is observable (figure 6b).

Further insight into the DR signal is gained by spectral analysis. Fast Fourier transform (FFT) spectrum was calculated for the combined $\gamma$-time series for 222 days—the longest continuous dataset available (days 403–625). In order to enable a long time series, a jump in the background level, occurring at 565.6, was rectified by adjusting the following data to the previous ones. To this end, 10-day long averages were calculated prior to and following the offset, and the difference (=206.6 counts 15 min$^{-1}$) was used to adjust the subsequent data. Linear detrending was applied to the dataset in the course of the FFT analysis.

The FFT results (figure 7) show that a small but well-distinguished diurnal constituent occurs in the dataset. The primary diurnal cyclic constituent (=0.99997 cycles d$^{-1}$; $\theta = 0.48$) represents a frequency of 1 cycle d$^{-1}$. The occurrence of a further semidiurnal constituent with a frequency of 2 cycles d$^{-1}$ is also apparent in figure 7. The observation of a semidiurnal periodicity in this
Figure 6. Examples of relationships between concurrent MD (solid) and DR (grey) signals (15-min resolution). MD and DR signals are obtained by decomposition with a 25-h sliding average. DR signal is further denoised using a nine-point (2.25 h) sliding average. (a) The daily amplitude of the DR signals varies in an irregular fashion. (b) An apparent relationship is observed between the amplitude of the DR signal and the MD signal.

Figure 7. FFT of the radon signal measured using the combined γ-detector signal during a 222-day period (from 564 to 776). A distinct diurnal component (1 cycle d$^{-1}$) is observed and possibly also a semidiurnal one (2 cycles d$^{-1}$). See text.
The measurement site is stable (figure 3). The plot in figure 8 demonstrates the temporal variation to reflect an atmospheric—pressure and/or temperature—driving mechanism. Temperature at adjacent to the laboratory from the surface outside the LNGS, which dilutes the indoor radon owing to its low level. Radon variation, a generally well-known pattern. The latter is affected by ventilation driving fresh air into the spectra of radon and pressure (see below; figure 10).

Different time intervals show both similar and dissimilar covariation patterns among pressure and radon. Concurrent environmental parameters measured at the radon monitoring site include temperature, humidity and air pressure. Humidity and temperature show very stable patterns. Temperature variation is less than 0.5°C during longer intervals (table 1). Pressure shows an MD variation, a generally well-known pattern. The latter is affected by ventilation driving fresh air from the surface outside the LNGS, which dilutes the indoor radon owing to its low level. Radon level at the monitoring site reflects primarily the content of radon in the air of the country rock adjacent to the laboratory.

In numerous cases, it is reported that variation patterns in radon time series seem, at first hand, to reflect an atmospheric—pressure and/or temperature—driving mechanism. Temperature at the measurement site is stable (figure 3). The plot in figure 8 demonstrates the temporal variation of γ-radiation from radon with the variation of pressure measured both at the subsurface site and at the above surface. The pressure in the laboratory follows the surface pressure, but it is probably somewhat affected by the ventilation system. The γ-radiation pattern follows a different pattern. Different time intervals show both similar and dissimilar covariation patterns among pressure and γ-radiation. Simultaneous measurement of pressure and radon by the Alphaguard system allows a comparison of the radon level and the local air pressure. Figure 9 shows the relationships between the radon level and the indoor air pressure (figure 9a) and the relationship between the gradients (difference between consecutive measurements) of pressure and radon level (figure 9b). The apparent absence of correlation, particularly between radon values and pressure gradient, suggests that pressure variations cannot be considered as a straightforward and direct driver of the radon variation at the confined conditions of the site.

In these two examinations, the dispersed patterns (figure 9; days 193–235) among pressure and α-radiation indicate that the variation of radon is not related to the pressure variation pattern. Similar dispersed patterns are encountered in correlation diagrams (not shown) in four additional intervals of continuous measurements spanning 285 days (table 1). Detrended and normalized hourly measurements of pressure versus γ-radiation and of pressure difference (among consecutive data points) versus γ-radiation were used.

The statistical significance of these dispersion patterns of the five datasets is examined using the Kendall rank correlation test (table 1). This test was previously applied to examine the significance of the relationship between temperature variation and radon [26]. In the case of the dataset of pressure and radon (α) shown in figure 9, the outcome yields p-values < 0.1.

### Table 1. Results of the Kendall rank tests for correlation between pressure and radon. Within each interval, the measurements are continuous. τ, Kendall rank correlation coefficient; p, probability for rejecting the null hypothesis of mutual dependence.

<table>
<thead>
<tr>
<th>days (relative 1 Jan 2005)</th>
<th>τ</th>
<th>p-value</th>
<th>temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>193–235 (figure 9)</td>
<td>−0.2047</td>
<td>0.145 × 10^{-17}</td>
<td>(figure 3)</td>
</tr>
<tr>
<td>Δ pressure and radon</td>
<td>−0.0662</td>
<td>0.00185</td>
<td></td>
</tr>
<tr>
<td>669–781</td>
<td>−0.2382</td>
<td>0.163 × 10^{-25}</td>
<td>14.32 ± 0.38</td>
</tr>
<tr>
<td>Δ pressure and gamma</td>
<td>0.1093</td>
<td>0.203 × 10^{-16}</td>
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</tr>
<tr>
<td>787–842</td>
<td>−0.2353</td>
<td>0.161 × 10^{-26}</td>
<td>13.24 ± 0.20</td>
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<tr>
<td>Δ pressure and gamma</td>
<td>0.0801</td>
<td>0.132 × 10^{-4}</td>
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<tr>
<td>843–919</td>
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<td>0.359 × 10^{-23}</td>
<td>12.82 ± 0.21</td>
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<td>Δ pressure and gamma</td>
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<td></td>
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<tr>
<td>920–954</td>
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<td>0.838 × 10^{-24}</td>
<td>12.1 ± 0.03</td>
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<td>Δ pressure and gamma</td>
<td>0.0936</td>
<td>0.640 × 10^{-4}</td>
<td></td>
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**Figure 8.** Time series (50 days) of air pressure both in the subsurface laboratory and at surface along with concurrent variation of the $\gamma$-radiation owing to radon in the air of the room. All time series are dominated by MD signals. The subsurface pressure varies according to the above surface pressure. The variation of radon shows a different pattern. (Online version in colour.)

**Figure 9.** Relationship between: (a) radon level and air pressure values inside the site and (b) radon level and pressure difference between consecutive measurements. The data are hour averages of the values measured during 42 days (days 193–235) using the $\alpha$-detector system. The Pearson correlation coefficient between radon and pressure data in (a) is $-0.442$. The statistical significance of these distribution patterns was investigated with the Kendall rank correlation test (see text).

The Kendall rank correlation test to the four further intervals (table 1) yield $p$-values which are extremely low, especially in the case of the measured values of pressure and radon. Based on the outcome, the null hypothesis that a dependency exists between temperature and radon is rejected. The conclusion from these tests on the independent datasets is that the variation of radon at this site is not related to the pressure variation pattern.

Time series of atmospheric pressure measurements typically contain diurnal (24-h cycle) and semidiurnal (12-h) periodic variations [27], with relative amplitudes that change from place to place and in time. This allows also investigating the eventual connection between radon and pressure in the frequency domain. Using the data of the four intervals in the range of day 669–954 (table 1), the spectra of radon and pressure at the monitoring site are shown, per interval,
Figure 10. Spectra (FFT) of (a) the radon and (b) pressure signals during four time intervals in the range of days 668–954, using data at a resolution of 1-h. The values of each time series was linearly detrended and normalized for calculating the spectra. A 24-h periodic constituent is not present in the case of the pressure signal, whereas it occurs in the radon signal. See text. (Online version in colour.)

in figure 10. The resulting spectra indicate that a 24-h periodic constituent is observed with amplitude of 0.2–0.3 (normalized) in the radon signal while such periodicity does not occur in pressure. A 12-h periodicity with amplitude 0.5–0.1 (normalized) occurs in pressure.

The long-term variation in the frequency–time domain of subsurface pressure and radon were analysed using the Continuous Wavelet Transform (CWT). The two time series show different patterns of variation (figure 11). The variation of pressure in the frequency–time domain is uniform. In comparison, the variation of radon is irregular in the same frequency–time domain. This is indicating, again, that variation of pressure is not driving the variation in radon.
Figure 11. CWT analysis of the site air pressure timeseries (a) and the normalized $\gamma$-ray timeseries (b). Significantly different patterns between pressure and radon are observed in the frequency–time domain. (Online version in colour.)

Figure 12. CWT analysis of the site air pressure timeseries (a) and the normalized $\gamma$-ray timeseries (b) decomposed to daytime and night-time measurements. Very similar patterns are obtained for pressure and significantly different patterns are observed for radon. (Online version in colour.)

An advanced reanalysis [28] of the $\gamma$-radiation from the primary radon in the air experiment of Steinitz et al. [29] demonstrated, among other things, that different temporal variation patterns occur depending on whether daytime or night-time measurements are considered. The existence of this feature in the LNGS data was investigated by applying a similar approach using the normalized hourly $\gamma$-ray time series and performing two analysis steps:

(i) The time series was decomposed into two time series of (i) daytime (08.00–16.00 h) measurements and (ii) night-time (20.00–04.00 h) measurements. The ensuing gaps
in these time series were filled by linear interpolation. Figure 12b shows a CWT (Morlet wavelet) analysis of the decomposed $\gamma$-ray time series, presented as a time-integrated power spectrum. Different patterns are obtained for the daytime and nighttime measurements. On the other hand, the same kind of analysis performed on the decomposed time series of pressure (figure 12a) results in similar and uniform patterns both for the daytime and for the night-time measurements, as is expected.

(ii) The 24-h periodic component in the time series was filtered and reconstructed (FFT; figure 13). The reconstructed time series is composed of 24 values per each day. This allows tracking per hour-in-day (HID) values over the whole time interval. In other words, one is observing the long-term variation pattern of measurements taken at a
specific hour within the daily cycle. Using such a measurement scheme (a specific hour in each day), the anticipation is that no systematic pattern occurs. Figure 14 shows the decomposed set of 24 HID time series, showing that in difference with the expected the time series, vary in a regular interwoven pattern. Further examination shows that this internal HID structure of the S1 periodic component is actually composed of 12 pairs of inverse time series with a 12-h difference per pair. Figure 15 presents two examples of such pairs. The overall pattern of the HID variation is presented as a three-dimensional diagram in figure 16 showing that the normalized level of the 24-h component is strongly positive around 04.00 h and highly negative 12 h later, around 16.00 h.

The results of both analyses indicate that a day–night differentiation occurs in the γ-radiation from radon at LNGS, similar to that observed by Sturrock et al. [28]. Furthermore, the 12-h asymmetry indicates that this feature is connected to the rotation of the Earth around its axis.
4. Discussion and conclusions

Generally, the interpretation of radon signals, especially at shallow levels, often invokes above surface atmospheric influences. Particularly, atmospheric pressure and temperature have been proposed for the origin of the periodic signals observed in Rn time series [1–4]. Reviewing the abundant literature on radon variation in semiconfined and subsurface situations, one notes that a large span of different views exists as to the eventual influence of pressure and/or temperature. Furthermore, other studies indicate that a consistent meteorological influence cannot be identified and suggest other influences. The evolving picture is that this is an ‘unexplained’ issue.

Being a component of air, the intuitive inclination is to ascribe radon variation to atmospheric (pressure and temperature) variations. Atmospheric (pressure, temperature) and radon variation are composed of signals of similar style and especially of waveforms of comparable duration. The common approach in analysis of radon time series leads to uncertainty in interpretation. Generally, the variation of radon is addressed only in the time domain, sometimes also in the frequency domain. Frequently, only relatively short-time intervals are used. In the time domain, this leads to intervals of correspondence and/or inverse conformity. The resemblance can also lead to similar spectra in the frequency domain. Therefore, establishing such correspondence only in the time and/or frequency domains is ambiguous. Furthermore, such relationships do not imply a physical connection. Here, it should be noted that so far no explanation has been put forward as to the specific property of radon which enables its unique behaviour. In this respect, it is conspicuous that similar variation patterns are not known for other noble gas trace components in air (He, Kr, Xe). Therefore, we maintain that solving this quandary requires additional statistical criteria which are more indicative. This refers especially to the analysis in the combined frequency–time domain as demonstrated in this contribution.

The concordant radon signal measured at LNGS using two independent radiation detector systems confirms the exactness of the absolute radon content and its variation in the air inside the monitoring site, which is in contact with the geogas system of the locally exposed rock face. The overall environmental features of the measurement site limit the option of mass transfer to explain the variations in radon. Radon originates in the surrounding rock system from where it diffuses into the experiment room. The level of radon in the adjacent rock porosity and crevices is certainly higher than that in the experiment room, and thus does not constitute a sink for radon. Fast decrease in the level of radon in the order of 10% within a day occurs from time to time. Such a decrease cannot be accounted for by mass transfer out of the room. The tightness of the room and the stable temperature and humidity patterns in the room indicate that there is no significant exchange of air mass between the room and the laboratory, especially at a daily scale. Furthermore, there is no reason to assume selective transfer of radon from the air in the room.

Two types of signals occur in the temporal variation pattern of radon in the geogas at LNGS—a non-periodic MD signal and a DR signal. The periodic character of the DR signal enables a deeper insight and a further comparison of characteristics with those of local environmental parameters. Local temperature, governed by the surrounding rock temperature, is apparently not related to the variation of radon, as it is very stable. Pressure gradient is often raised as a driver of radon variation in geogas. Analysis for correlation in the time and frequency domains shows that temporal variation of radon is most probably unrelated to the variation of pressure. Additional analysis in the frequency–time domain, which is more indicative, yields very different patterns for radon and pressure. The complicated pattern obtained for radon cannot be accounted for by the uniform pattern of pressure. In addition, by using this approach, a day–night differentiation is observed in radon which does not exist for pressure. Furthermore, this day–night feature in radon implies a connection with the rotation of the Earth. This situation indicates that variation of air pressure also cannot be considered as the primary driver of the cyclic DR signals, and probably also not of further components of the radon variation. In turn, this implies that a different periodic process is operating which produces the 24-h cyclic variation in the radon signal.
The following features and considerations may shed light on the processes driving the radon signals in air at LNGS:

— incompatibility of above surface atmospheric processes—both temperature and pressure—as primary driving mechanisms;
— a clearly defined non-periodic MD signal occurs in the radon time series of LNGS. Elsewhere [30], such signals have been associated with subtle geodynamic transients affecting the local rock system, based mainly on a statistical correlation of such signals with earthquakes in a geologically related domain. Determining a connection between radon emission and active geodynamics is not possible owing to lack of information on the spatial distribution of such signals around LNGS;
— the periodic DR signal at LNGS is not influenced by solid earth gravity tide processes. The main diurnal constituent is only a 24-h period (S1), whereas the constituents typical for gravitational interaction (primarily M2; [31]) are lacking. This probably indicates that the weak tidal-mechanical effects are not influencing the radon signal. Similar conclusions have been arrived at from recent investigations at other locations (see below);
— the apparent partial association between the MD signal and the amplitude of the diurnal variation (peak-to-peak) opens the possibility for some sort of coupling; and
— a long-term difference among daytime and night-time measurements is observed, a feature which is not known for other geophysical time series (atmospheric, solid earth).

Radon signals in geogas within rock systems in upper crustal levels have been intensely studied in Israel [25,30,32] and Tenerife [32,33]. Geophysical analysis of these relatively long time series measured with a high time resolution (within 1 h) demonstrates that Rn signals in air confined within rock units are characterized by systematic recurrence of signal patterns in the temporal, geological and geographical domains, at a depth of more than 100 m. The main types recognized are multi-year, annual radon (AR; periodic), MD and daily radon (DR; periodic) signals [14,15,25,34] as well as intense variation lasting several hours [16,17]. Examination of the patterns suggested that

— variation patterns in the subsurface regime cannot be accounted for by simple and direct time varying processes in the gas system such as emanation, diffusion, absorption and advection;
— absence of above surface atmospheric influence, in particular pressure; and
— periodicity in the DR signal is characterized by solar tide frequencies S1 (24-h), S2 (12-h) and S3 (8-h) [31], implying an external above surface driving mechanism.

Based on the accumulated data and phenomenology, it was suggested that a component of solar irradiance is influencing the radon signal, primarily the periodic signals. Further insight on the issue was recently gained by experimental simulation of radon signals in confined volumes of air [29]. In contrast to the expectation, the results demonstrate nuclear radiation patterns that are non-uniform in space and in time. The variations (20%) were both periodic and non-periodic, spanning several orders of temporal magnitude—from annual to daily. The results, obtained under static and isolated conditions, are in disagreement with the expected radioactive equilibrium and its spatially uniform expression within and around the experimental volume. The characteristics of the prominent periodic signals indicate forcing by a component of solar tide which can traverse 5 cm of lead. These experimental results are in line with the observations from the geological environment and conform to the implied geophysical consequences. Reanalysis by Sturrock et al. [28] of the long experimental radon time series could (i) corroborate the results, (ii) present evidence of solar rotational frequencies (which are independent of the Earth) in the nuclear radiation time series and (iii) show a 24-h modulation of the γ-radiation of the radon system in the annual and solar frequency bands. In conformity with the view in previous works, Sturrock et al. [28] suggested that the decay is influenced by solar radiation, and solar neutrinos were considered as a possible particle involved.
The radon signals observed within confined air of the rock system at NLGS, at 1 km depth, show similarities with the observations in the above works—in terms of the radon patterns and the dilemmas versus eventual atmospheric drivers. Therefore, it is suggested that other geophysical processes drive the nuclear radiation from radon at LNGS. If this is the case, then the possibility is raised that the driver of the daily periodic component at LNGS is also owing to a component of solar irradiance. The occurrence of a day/night difference in the DR signal at LNGS supports the notion that solar irradiance is influencing the nuclear radiation from radon in air. The day/night effect is a modulation of this influence owing to the rotation of the Earth around its axis.

Identification of geophysical radon signals in the air in the realm of Gran Sasso has specific implications for research in this field and on related subjects, as follows:

— the occurrence of such geophysical signals in a further geographical location (central Apennines),
— observing such phenomena in a calcareous sequence with a low-uranium content,
— the phenomena are detected at a depth in the order of 1000 m below the surface,
— observing radon signals at a level situated below the regional water table,
— providing new information on the environmental radiation phenomena at LNGS, including aspects of its local specific temporal variation, and
— setting ground for the exceptional option of performing advanced physical and geophysical investigations on the radon system based on the infrastructure of LNGS.

The observations and results mark the LNGS facility as a high-priority location for performing advanced investigations of geophysical phenomena of the radon system, owing to its location and infrastructure [18,19,22,35–37]. The present investigation was hampered by the combination of the overall low radon signal and the limitations imposed by the analytical system employed. To overcome this situation and to explore the remaining questions such as to the influence of local environmental influences, controlled active experiments as devised and described by Steinitz et al. [29] should be performed at deep underground laboratories such as LNGS (IT), LSC (ES), Sanford (US) and Sudbury (CN).

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