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Applied Radiation and Isotopes 64 (2006) 520–524

Applied Radiation and Isotopes

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Multi-day radon signals with a radioactive decay limb—Occurrence and geophysical significance

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Received 23 May 2005; received in revised form 14 October 2005; accepted 14 October 2005

Abstract

Multi-day signals, generally with duration of 2–10 days, are a prominent temporal variation type of radon (Rn) in geogas in the unsaturated zone. Rare multi-day Rn signals have been found which are characterized by: (a) a declining limb lasting up to 10 days which conforms to the radioactive decay of Rn, (b) recurs at the same location and (c) is recorded in diverse situations—volcanic and seismogenic. It suggested that a Rn blob is injected at a lower level on a steady upward flow of geogas whereby the rise and final fall of the signal are attributed to the edges of the blob while the central Rn-decay segment records the passing of the decaying blob itself. Rn-decay signals are a small subset of multi-day Rn signals which are considered as highly irregular and unusable for the understanding of geophysical processes. In difference, it is concluded that multi-day Rn signals are probably proxies of subtle geodynamic processes at upper crustal levels and are therefore significant for studying such processes.

Keywords: Radon; Geodynamics; Dead Sea rift; Tenerife

1. Introduction

The utilization of Rn in geogas and in hydrogeologic systems as an indicator of geodynamic processes has been discussed in a variety of geographic and geologic regions, mainly in seismogenic and volcanic settings. Complex signals and results are obtained. The limited time-series data that can be collected and the time-resolution used add to the difficulty of discerning systematic features.

Rn (^{222}Rn), a radioactive inert gas formed by disintegration from ^{226}Ra, occurs at varying concentrations in geological environments. Its noble character and its radioactive decay make it a unique ultra-trace component in subsurface systems. Numerous works relating to this subject suggest that both environmental and geophysical processes affect temporal changes of Rn in geogas. Identifying the nature of these influences is crucial for understanding and utilization of Rn as a geophysical tracer.

Few recent works describe results of high-resolution Rn monitoring in the unsaturated zone. Trique et al. (1999), measuring in a tunnel within massive gneiss, associated multi-day Rn bursts with transient deformation due to loading and unloading of a local water reservoir. Reddy et al. (2004) describe a multi-day Rn signal with a Rn-decay decreasing limb, measured in granite within the stable continental platform, and associated it with a specific micro-earthquake. Using an 8-year record of Rn, obtained next to a major active boundary fault of the Dead Sea rift (DSR), a statistically significant relation between multi-day Rn signals and earthquakes in the nearby sector of the DSR was demonstrated by Steinitz et al. (2003).

Systematic high-time resolution (<1 h) monitoring of Rn has been performed for several years in Israel and Tenerife (Canary Islands), in upper crustal rock systems. Systematic variation patterns of the Rn signal in the geogas

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doi:10.1016/j.apradiso.2005.10.004

are observed, in terms of recurrence at a site and among different locations within areas: (a) multi-year variation, (b) annual-seasonal variation, (c) multi-day signals, generally with duration in the range of 2–20 days long, and (d) diurnal variations. This study focuses on a specific multi-day Rn signal type characterized by: (a) an unambiguous temporal signature part of which can be indisputably interpreted in terms of nuclear physics, (b) recurs at the same location, (c) similar specific signals are recorded in diverse geographic, geological and geodynamic situations. These serve as a basis for drawing conclusions pertaining to the overall significance of these specific signals as well as other associated Rn signals.

2. Experimental setup (field sites)

Long time series of Rn in geogas were obtained at three sites (Table 1) in two different geodynamic settings—an active volcanic edifice in Tenerife (Canary Islands, Spain) and in the seismically active DSR (Israel). In Tenerife Rn monitoring is performed in the AJM tunnel (gallery), about 10 km SE of the active Teide volcano. The monitoring site is in a horizontal tunnel dug into massive lava flows of the central volcanic complex (Cañadas Edifice) of Tenerife, at a depth of 75 and 400 m from the entrance, and having an ambient temperature close to 18 °C around the year. Martín (1999) identified this tunnel as one of the subsurface locations of anomalous Rn levels (attaining $>100\,\mathrm{kBq/m^3}$) in Tenerife. In the DSR, two monitoring sites are located along its western margin. The main monitoring site 17W is located at Enot Zuqim, NW of the Dead Sea, next to the scarp of the main western active fault of the DSR. The site, at which Rn levels of $>550\,\mathrm{kBq/m^3}$ are attained, is situated within a 20 km long regional Rn anomaly located in the rift next to the western boundary fault (Steinitz et al., 1999). The sensor at 17W is placed in unconsolidated gravel at a depth of 1.5 m. The Amram site, some 200 km to the south, is located in massive granite in an uplifted basement block of the western margin of the southern segment of the DSR. Two Rn sensors are placed at a distance of 140 m inside the N–S tunnel, separated from the external atmospheric environment by two doors.

3. Methods

Nuclear detectors used consist of alpha and gamma detectors. At the AJM site the detectors are placed horizontally and parallel to the tunnel, 20 cm from the floor and the sidewall. The alpha detector used is an Alphameter 611 (AlphaNuclear Inc., Canada), based on a $400\,\mathrm{mm^2}$ silicon junction diode, immersed in a sensing volume open to the geogas. Utilization of gamma detector systems for monitoring Rn is based on the detection of gamma radiation from the ^{214}Bi, and to a lesser extent those from the ^{214}Pb. Due to the short half-lives of the Rn daughters equilibrium of the Rn and its daughters is achieved after a short time (~25 min). At the AJM site a 2″ × 2″ NaI detector (PM-11 detector, ROTEM Inc., Israel) is immersed in a 1.5–2 l measurement cell, open at its lower end. The detector and the measuring volume are shielded with Pb plating (5 mm thick), reducing the background gamma-ray radiation emitted from the surrounding lithology. At site 17W a similar shielded gamma sensor setup is placed at the lower end of a vertical tube equipped with gaskets and stoppers thereby separating it from the atmosphere. The detector in this case is a 1.5″ × 1.5″ NaI detector (SRAT, France). Measurement at the Amram site is performed simultaneously with an alpha detector ($400\,\mathrm{mm^2}$ Si diode, Barasol BT45N, Algade Inc., France) and a bare gamma detector (2″ × 2″ NaI detector, PM-11) placed parallel to the tunnel, 50 cm from the floor and the sidewall. Integration times are set to 15 min at all sites. Time is shown on a decimal-day scale (time since 1.1.1992 for DSR; 1.1.1998 for Tenerife).

Table 1
Comparison of site features at which Rn-decay signals have been observed

Site	Tenerife	Israel—Dead Sea rift	
	AJM	Amram	17W
Geodynamic context	Active volcanic island	Plate boundary—active rift (DSR)	
Structural/tectonic situation	Flank of volcanic edifice	Raised structural marginal block	Adjacent to a large active boundary fault
Background Rn level	Local/regional Rn anomaly	Normal Rn level	Very high regional Rn anomaly, associated with boundary fault
Age of host rock	Plio-Pleistocene	Precambrian	Sub recent–recent
Lithology	Massive phonolitic flow	Massive granitic intrusion	Unconsolidated gravel
Distance from surface	Horizontal: 400 m, vertical: 75 m	Horizontal: 100 m, vertical: 70 m	Vertical: 1.5 m
Elevation (asl)	+1575 m	+220 m	−388 m
Distance to groundwater	475 m (estimate)	Not known. Probably large, if at all	~5 m?
Climate	Temperate	Arid	Arid
Temperature at subsurface site (mean)	18.5 °C	28 °C	27 °C
Approximate radon background level (Bq/l)	0.1–1.5	1.5	3.7

4. Results

Multi-day signals, generally with duration of 2–20 days, and which are most roughly symmetric (similar rising and decreasing limbs) or asymmetric, are the most prominent temporal variation type at the discussed locations. Unique multi-day Rn signals (Rn-decay) occur in which a relatively fast rise to peak of the Rn signal is followed by a gradual decrease, lasting 2–10 days, that conforms to the radioactive decay of Rn (half-life of 3.82 days). In some cases the falling limb reaches Rn levels that are close to the background or, in other cases, it is replaced by a fast final fall of the signal (Fig. 1, left). The occurrence of such signals is rare relative to the occurrence of other types. Fig. 1 shows an example from the AJM site recorded simultaneously by independent alpha and gamma detector systems as well as the Rn-decay curve fitted from the peak. The close-to-identical patterns recorded by both detectors confirm that Rn levels are indicated. Fig. 2 shows similar patterns for Rn-decay signals from the Amram site, also recorded simultaneously by two different detectors. Fig. 3 exhibits a large multi-day Rn signal with a very long Rn-decay limb (~10 days) from site 17W.

5. Discussion

The specific physical features of the described Rn signals suggest some constraints on the local geogas flow pattern. Ample evidence exists supporting the rise of geogas in the flanks of the volcanic edifice of Tenerife, including the area of AJM. This includes the fumarolic field at Teide volcano, large areas with thermal anomalies (up to 50 °C) and diffused CO_2 emissions in the subsurface (Valentín et al., 1990; Soler et al., 2004), and in soils of Las Cañadas Caldera (Hernández et al., 2000). Geochemical indications for rise of gases from depth along the shores of the Dead Sea, in proximity of the major boundary faults of the rift include manifestations of H_2S observable at springs and in their vicinity.

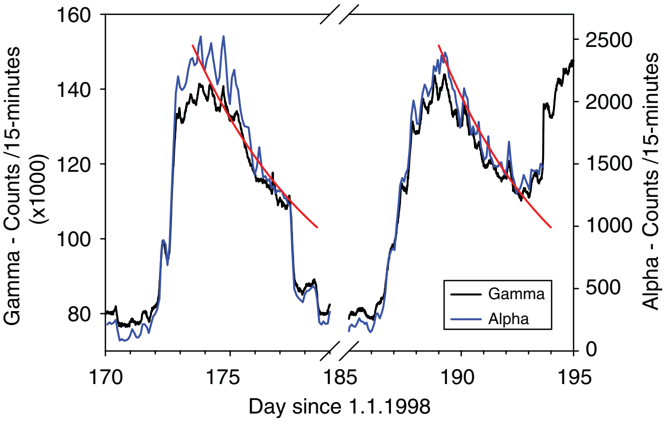

Fig. 1. Two multi-day radon signals from AJM site in Tenerife. Alpha and gamma detectors record the radon signal simultaneously. Radon decay, lasting some 4 days, is presented based on modeling the decay of the alpha counting.

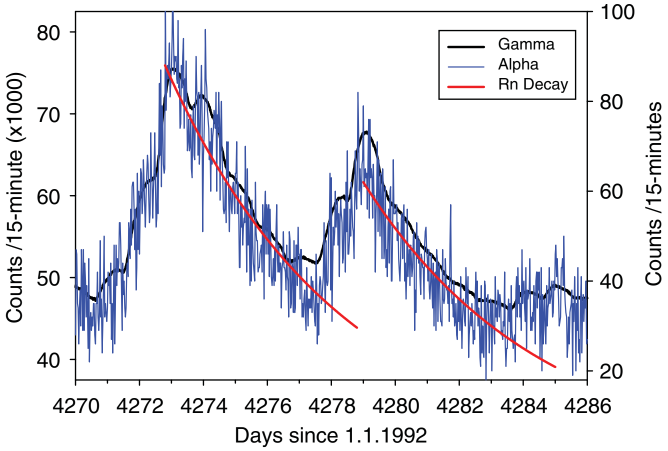

Fig. 2. Two consecutive multi-day radon signals from Amram site in the DSR. Alpha and gamma detectors record the radon signal simultaneously. Radon decay, lasting some 4–5 days, is presented based on modeling the decay of the alpha detector.

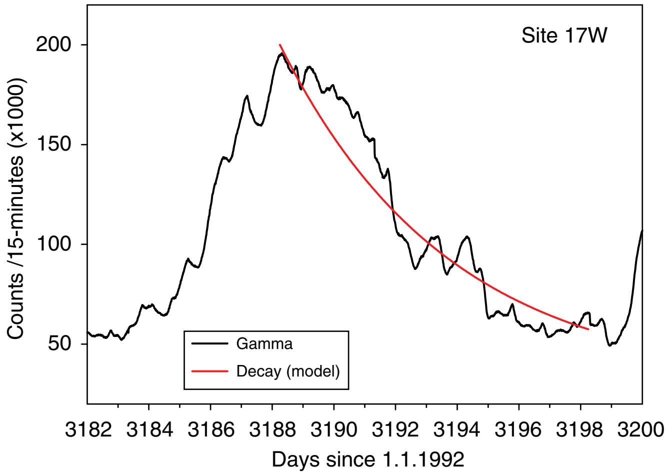

Fig. 3. A large multi-day radon signal from site 17W in the NW Dead Sea, DSR. The duration of the radon-decay-governed decreasing limb of the radon signal is around 10 days. A rough calibration yields 1 count per 15 min which is approximately equivalent to 9.6 Bq/m^3.

In the three sites discussed the local and immediate geology is well known. In all cases, the anomalous Rn levels are not supported by an anomalous uranium or radium content in the rocks of the immediate local vicinity. In the case of the 17W site a possible source is identified as uranium-bearing Senonian phosphorites that are buried below the site at a depth on the order of several hundred meters (Steinitz et al., 1992, 1999). This example is based on well-established considerations, concerning the stratigraphic and structural pattern of the region, and substantiated by the encounter in boreholes of such rocks in structurally equivalent down-faulted tectonic blocks of the DSR, some 30 km to the south (Grossowicz, 1979). A 60-m borehole next to site 17W penetrated gravel with no uranium (radium) anomaly to its bottom, suggesting a greater depth for a source rock.

The involvement of advection of geogas is also supported by the pattern of the Rn signals themselves. The fast rates of rise and decrease of the Rn signal are unlike the form of a diffusion-controlled process and are inconsistent with the diffusion range of Rn, which is on the order of 5–10 m in air. Therefore, it can be assumed that Rn is carried up from lower levels by a rising geogas flow that passes the detectors. Furthermore, it can also be assumed that velocities that are relatively stable over longer periods of time characterize such upward flow, occurring in permeable zones and especially along fracture systems. The extent of the system (size, volume; the concordance of signals over distances of 15 km along a fault) implies this assumption (Steinitz et al., 1992, 1999).

The above suggests three possible causes for the observed variations in Rn concentrations:

(1) a geogas with a uniform distribution of a relatively high content of Rn is rising from depth to the surface on a carrier whose velocity varies considerably,
(2) a geogas with an initially uniform distribution of a relatively high content of Rn, rising on a carrier gas at a stable velocity, is mixed between the source and the sensor with a Rn deficient geogas (air?) arriving from a different direction, and
(3) Rn is intermittently injected at depth onto a carrier geogas rising with a stable velocity. We tend to adopt this possibility.

A complementary line of consideration can be drawn for the three different time segments noted in the multi-day Rn-decay signals: a fast rise, a decay segment, and eventually a fast decrease (in some cases). The several days long decay segment indicates a unique stable equilibrium time interval in which no significant addition or dilution of Rn occurred, implying that addition or depletion of Rn by diffusion and advection are not operative at this time (excluding minor fluctuations). At the time intervals of fast increase and decrease (sometimes within hours) a diffusion-controlled addition or removal process is too slow to explain the observed rates. In these cases, geogas flow must be assumed, whereby Rn-rich geogas is introduced into the vicinity of the sensor at the rise time and is flushed away by Rn-depleted geogas at the final decrease.

The pattern of multi-day Rn-decay signals can thus be examined in terms of two adverse situations concerning advection of geogas—flow or no-flow conditions of the subsurface geogas system, suggesting two end-member scenarios:

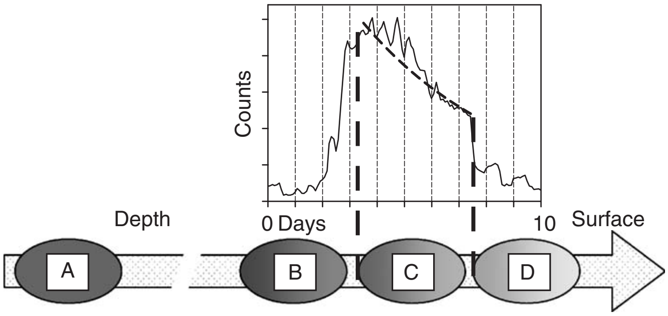

Fig. 4. Model of the record of a decaying radon blob on a carrier gas passing across the nuclear detector system (steady flow scenario). Gray level represents the concentration of Rn in the geogas.

1. Steady flow situation (Fig. 4): A continuous upward flow of Rn-deficient geogas flow system in the subsurface is assumed to be of a steady nature. This system is a carrier gas that brings, from time to time, a blob of Rn that had been injected onto the geogas flow (Fig. 4A) at a location distant from the measurement setup. This location is probably at the source of the Rn at a lower subsurface level. Once deposited as a uniform blob on the carrier (i.e. uniform Rn concentration, Fig. 4A) and separated from the source region the Rn in the blob decays according to its half-life. Upon arrival of the leading edge of the blob to within the detection range of the sensor (Fig. 4B), a fast rise of the signal is recorded as the steeply rising flank of the Rn signal. The time interval required for the blob to pass the detector region is recorded as the gradual decrease governed by the decay (Fig. 4C). The passing of the trailing edge of the blob is recorded as the fast decrease of the Rn signal (Fig. 4D). In this model the time interval of the decay segment is a function of the velocity carrier gas and the size of the blob in the direction of the flow.
2. Intermittent flow: In this scenario the inflow of Rn-rich geogas is discontinuous and comes to a standstill, enabling the observed decay pattern. This is followed, after several days, by renewed geogas flow—this time an influx of geogas with low Rn content (air?)—which flushes away the remaining Rn. Such a scenario results in an alternation between a Rn-rich and a Rn-deficient record. The Rn-rich geogas flow originates, most likely, from a lower level while the Rn deficient geogas must be envisaged from a different source, location and direction. An extreme alternative could mean downward flow of atmospheric air ("reverse flow"). This implies a varying flow pattern for the geogas in terms of direction and velocity.

In terms of geogas flow systems at upper crustal levels, the intermittent flow scenario is complicated relative to the steady flow. It requires a flow system changing its regime within hours. Furthermore, the steady flow model is in conformity with the simpler assumption of stable flow patterns of geogas and of long time constants, relative to the half-life of Rn, of geogas flow in most subsurface environments.

Multi-day Rn signals exhibiting a Rn-decay limb are observed in diverse settings (Table 1), although their occurrence is rare (~1% of multi-day signals). It can be assumed that similar exceptional conditions are responsible for this specific Rn signal in the different settings. It is suggested that the model of a Rn blob on a steady geogas flow is an appropriate mechanism applicable to these cases. This allows to suggest that:

1. the specific Rn-decay signals are a small subset of multi-day Rn signals recorded at each site. Assuming they represent specific conditions, the other multi-day Rn signals can be considered as representing variations and irregularities in terms of the injection of the Rn as a blob and the flow parameters of the same carrier system.
2. In the case of site 17W, preliminary considerations can be presented concerning the rate of geogas flow. We assume that a Rn blob having a vertical dimension on the order of 100 m is released on to the carrier gas from the underlying 100 m thick uranium enriched phosphorite sequence. The characteristic time interval of the Rn-decay limb of about 10 days is the time it takes the blob to pass the detector (Fig. 3). Hence a vertical velocity of 10 m/d is derived. Assuming that the primary Rn signal will practically decay within 10–20 half-lives of Rn suggests that the source rock is at depths not greater than 400–800 m, which is compatible with the known geological setting.

Rn signals have been measured and monitored for several decades at different locations, spanning a large range of environmental, geographic, geological and geodynamic conditions. The prevailing impression is that such signals are highly irregular and are not prone to a systematic treatment, and are hence useless for the understanding geophysical processes in upper crustal levels. In comparison, the sites investigated here are also in different rock systems, in the unsaturated zone, and measurements were performed utilizing electronic detector systems with a time resolution of less that 1 h. Combining this with the occurrence at different locations of a multi-day Rn signal which exhibits a feature which is simply related to a known physical rule implies that systematic parameters and communalities can be established and thus geophysical information may be gained from such investigations. Moreover, this approach allows setting new constraints for modeling of radon release and transport in the subsurface.

6. Conclusions

It is demonstrated that geophysical processes can be directly associated with multi-day Rn signals having a Rn-decay limb. It is argued that other, more complex types of multi-day Rn events also reflect geophysical processes that deserve investigation and modeling in order to yield information on subtle geophysical processes in the upper crust.

Multi-day Rn signals observed at upper crustal levels, in the unsaturated zone, have forms that are related to the release, transport and mixing processes that govern them. Using end-member types of signals, comparing them and tracing their occurrence in relation to geological factors allows the drawing of conclusions on the whole set of such signals, which includes more complex types. This supplements other approaches such as correlation with other concurrent temporal phenomena.

Both recurrence and observation of Rn signals with a decay limb, which are a key element in understanding the investigated system, are possible only by acquiring long and continuous time-series of Rn obtained in the frame of systematic monitoring programs.

Acknowledgments

We acknowledge the support of EC grant EVRI-CT-1999-40010 and the support of the Spanish Project PGC2000-2177-CE. The authors are grateful to Dr. H. Woith for his critical review.

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