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Observations of the relationship between directionality and decay rate of radon in a confined experiment

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Abstract. Radon (^{222}Rn) is a radioactive inert gas with an accepted half life of 3.8235 days. Its unique, systematic and complex variation in the geological environment and in simulation experiments combined with lack of understanding of the underlying drivers lead us to conduct tests of its apparent half life. A primary test took into account experimental observations indicating anisotropy of the gamma radiation from radon in air, which is related to global orientation. Using a goniometric configuration radon diffuses into two identical cylinders oriented along Earth axis of rotation and in a vertical and perpendicular direction to the latter. Detectors placed on cylinder ends measure gamma radiation sub parallel to these directions. At steady state and confined conditions different patterns of daily signals are observed in the two directions. Isolating the cylinders from the source leads to an exponential decrease on which similar daily signals are superimposed, having amplitudes proportional to the level of the remaining radon. The indicated apparent half-lives are in significant difference from the accepted value: 0.861 ± 0.003 days in the pole direction and 2.308 ± 0.008 days in the vertical direction. The outcome is in conformity with observations on radon signals in confined conditions and their different manifestation at different directions.

1 Introduction

Radon (^{222}Rn) is a radioactive inert gas formed by disintegration from ^{226}Ra as part of the ^{238}U decay series. The combination of its noble gas character and its radioactive decay attract it as a unique ultra-trace component for tracking temporally varying natural processes. Using nuclear techniques, the measurement sensitivity for ^{222}Rn is extremely high and can be performed with a time resolution in the order of 1 hour or less. Temporal variation patterns of radon in geogas[1] (= subsurface air) have been investigated since the middle of the last century. Numerous works [1–17] tried to clarify the possible reasons of the variation patterns and their potential geodynamic

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[1] Geogas: gaseous phase in the unsaturated zone above the groundwater level, sometimes referred to as soil air or soil gas.

consequence. Due to its properties it is considered a unique trace component, in particular as a proxy of temporally varying processes in natural environments. So far, however, a comprehensive theory and physically sound explanation of the observed variation patterns remains elusive.

The paramount interest is in Rn as an eventual indicator and proxy for active mechanical seismogenic and volcanic geodynamic processes. Numerous studies have reported changes in radon concentrations associated with pre-seismic activity and events [1–5]. In parallel, transport of radon in soil and water has been investigated as a tool for monitoring volcanic activity [6–9]. In these scenarios radon is assumed to be a highly sensitive tracer of secondary geodynamic processes mainly mechanical and thermal. Only a few works address these issues using high resolution long term monitoring of Rn in the unsaturated zone [10].

Utilization of Rn as a proxy of active geodynamics is hampered by the disputed influence of above surface atmospheric dynamics on the transfer (advection) of radon in the subsurface fluid (geogas, water). This relates primarily to daily variations of radon which are prominent, frequent and well known from observations at shallow depths (for a summary see [11]). Their occurrence is generally attributed to atmospheric influences at the boundary between the atmospheric air and geogas. The influence of environmental factors, mainly atmospheric pressure, is suggested [12–15] as the driver of these daily variations. A different approach suggests daily variation of tides as the driver [16–18]. The overall picture evolving from the published works is that the understanding of the processes driving radon signals in subsurface geogas is uncertain and disputed. The diversity of the observations, the complexity of the phenomena, and the span of the suggested mechanisms render the overall picture unresolved, hampering utilization of radon as a significant proxy of geodynamic processes. So far the eventual geophysical drivers of the variation of ^{222}Rn, as well as its specific qualities enabling this temporal variation, are widely unknown.

Since 2007, simulation experiments have been performed by us using the Enhanced Confined Mode (ECM) principle [19–21]. A high level of radon in air is maintained in a tight container by diffusion via a tube from a radon source. Radon detectors alpha and/or gamma detectors – measure the nuclear radiation. A lead shield is added to minimize the effect of environmental gamma radiation on gamma detectors [19].

In the ECM systems, elevated radon levels are maintained in an airtight volume by diffusion from a radon source (Fig. 1). We expected that stable (constant) radiation patterns would be observed, once a steady state between release/diffusion/decay is attained in the ECM volume. However, to our surprise, we found non-uniform radiation patterns in space and in time, composed of periodic and non-periodic signals which are analogous to those observed in the geological environments.

2 The temporal variation of radon in geogas

Similar and comparable variation patterns and characteristics are obtained from radon measurements in the geological environs and in the experimental ECM approaches. The primary outcome concerning the ^{222}Rn signal in these situations is that the compounded variation is composed of periodic annual radon (AR) and semi-annual (SAR) signals, a periodic daily radon (DR) signal containing diurnal (S1; 24-hour), semi-diurnal (S2; 12-hour) and ternary diurnal (S3; 6-hour) periodic constituents; and non-periodic multi-day (MD) signals. At geological sites these signals recur at a single site and with different combinations at different sites to depths of $>$ 150 meters [22–26]. Similar occurrences are recorded from the measurements in Tenerife to depths of 350 and 850 meters [27] and at the Gran Sasso laboratory (LNGS; [28], down to a depth of 1000 meters. An extreme variant of the DR signal

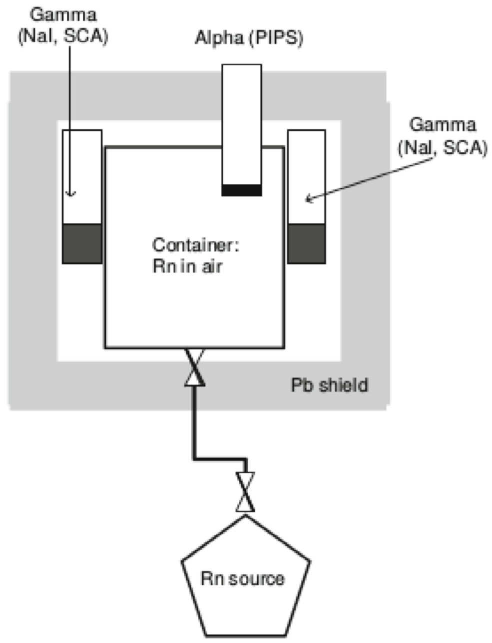

Fig. 1. The ECM principle.

is the sub-daily radon (SDR) signal. These signals, which occur in some days, vary from background levels to high peak values and back to background within 6 to 12 h. Their occurrence indicates very fast variations of radiation from radon at the point of measurement. At two subsurface sites (depth: 10 and 85 m) SDR signals occur in some days [29,30] and occur preferentially within the daily cycle i.e. exhibit a non-regular diurnal periodicity.

Multi-year data sets shows that the amplitudes and phases of the periodic components in the diurnal band (DR signal) are modulated annually (AR). In ECM experiments, the periodic phenomena were attributed to the influence of solar-tide, which is due to the rotation of the earth around its axis and around the sun. The diurnal periodicity was dominated by solar components S1, S2 and S3, exhibiting dissimilar relative amplitudes and different phases at the different detectors [19]. The latter results, obtained under static and isolated conditions, are in flagrant disagreement with the expected radioactive equilibrium and its spatially uniform expression within and around the experimental volumes. New experimental results [20,21] have demonstrated that both MD and DR signals exhibit spatial directionality related to the global east-west and north-south directions. It appears that in the case of the ^{222}Rn system, the periodic AR and especially the DR signals, in both the subsurface and in the confined experiments, are generated – to an extent to be determined – by an above-surface periodic process. The characteristics of the latter features in the time and frequency domains led to our suggestion that a component of solar irradiation influences directly the ^{222}Rn system [19–21,25,28–30]. A primary criterion was the lack of tidal gravity periodicity. Diurnal periodic components typical of gravity tide are M2, O1 (etc.; [31]), which were absent.

In recent years, the widely held view that nuclear decay rates are fundamental constants of nature has been challenged by reports (in the physics community) of periodic variations in nuclear decay rates [32–36]. Despite long-standing evidence supporting the conventional view that the decay rate of an unstable isotope is an

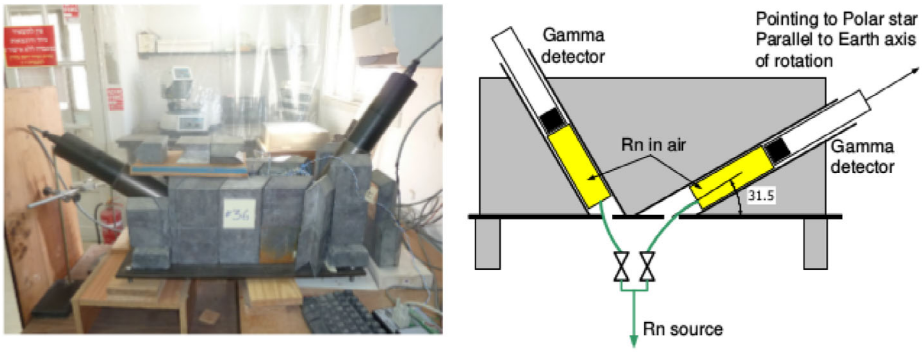

Fig. 2. The goniometer experiment. In the diagram north is to right, and in the photo (left) north is to the left. Pb shield is indicated schematically (grey) in the diagram and is shown in photo. See text.

intrinsic property of that isotope, there are growing indications of small variations (of the order of a fraction of 1%) for some nuclei. The suggestion that the observed periodic effects could be due to seasonal "environmental" variations in temperature, pressure, or humidity, is countered by the fact that some of the periodicities are not seasonal (e.g. 11 cycles per year), and may possibly be associated with solar events [34]. Although there is as yet no detailed mechanism to account for the observed variations in decay rates, it has been suggested that they are being influenced by the Sun, possibly via neutrinos. The strongest evidence for this conjecture is the discovery of periodicities that may be attributed to internal solar rotation and to internal oscillations [32,35,37]. To date similar periodicities have been observed in more than 20 experiments.

The unique, systematic and complex variation of radon in the geological environment and in ECM experiments combined with a lack of understanding of the underlying drivers motivated the investigation of its apparent half life. A series of tests on radon in air have been performed at the GSI since 2008 based on the ECM experimental concept. Different ECM configurations and different detectors are applied. A first description of a key test is given below.

3 Directionality and apparent half-life (AHL) in ECM-Goniometer experiment

In previous works [20,21] we showed that directionality related to global orientation occurs in the temporally varying radiation from radon in confined conditions, suggesting that nuclear radiation from radon is anisotropic. This assumption is further investigated using an ECM in a semi-goniometric configuration (Fig. 2). Radon from a source (103 kBq; Pylon Electronics Inc., Canada) is diffused (via tubes and three valves) into two identical cylindrical canisters (ø70 mm; 570 cc), which are fixed onto a frame. The cylinders contain air at atmospheric pressure and the applied system components, similar to those previously used [19–21], provide air tightness. The long axis of one canister is oriented parallel to the rotation axis of Earth (pointing to the north pole – Pole) and the other perpendicular (vertical; V_2_P) to the axis of rotation of earth. Two gamma detectors are placed at the upper faces of the two cylinders, along their axes. The PM-11 (Rotem Industries Ltd., Israel) gamma detector contains a NaI(Tl) scintillator, 2 "diameter and 2" thick, coupled to a 2″ photomultiplier tube and a 1 mm window (aluminium). It is fitted with a high voltage power supply (700±200 Vdc), single channel analyser (SCA, 50–3000 keV), and pulse shaper. In this configuration the detectors are influenced preferentially by gamma radiation which is parallel to the cylinder axes (the solid angle is around 8.5% of the 4π sphere). Pulses from the detectors are counted and summed per 1-minute using a data logger (CR-800; Campbell Scientific Inc., USA). The data logger is fed from a continuously charged battery (12 V) and a stabilizer maintaining a voltage of 12 ± 0.005 V, which supplies 5.0 ± 0.1 V to the detector.

The whole setup is shielded with Pb (5 cm) to reduce the influence of gamma radiation from the environment and the source. The background levels (counts/ 15-minute) are 23,800 at Pole and 22,500 at V_2_P, i.e. at a level of 4–6%. The level of the gamma radiation at the ENV detector is 93,000. The available Pb and structure of the system allowed us to enclose detectors in a Pb shield laterally and upwards. Most of the background gamma radiation originates from sector of the floor of the lab which is unshielded to the downward oriented detectors. This background, considered to be a relatively constant contribution, was not subtracted. It is taken into account by the non-linear exponential regression (3 parameters) of the decay. A further gamma detector, placed on the table about 50 cm apart, monitors the environmental gamma radiation in the lab. Gamma radiation from the experiment was blocked (Pb, 10 cm) to this detector.

The relative count rate of goniometer detectors is inter-calibrated using the gamma radiation from Rn at Pole position, leading to a sensitivity relation of Pole/V_2_P = 1.18. The measurements are adjusted accordingly for difference in sensitivity prior to analysis. Data collection is at a resolution of 1-minute. The count rates are summed for 15-minute intervals and hour averages of 15-minute counts are used below. A decimal time scale is used where Day 1 = 1.1.1992.

To date (November 2014) more than 10 trials (tests) have been performed with this setup since February 2009. The outcome of the different tests is similar. Each test consists of three stages: a) opening source and canister valves thereby enabling the diffusion of radon into the two cylinders leading to a buildup of the gamma signal; b) attaining a quasi-stable level of radon in the cylinders; c) closing the three valves and thereby terminating further diffusion of radon to the cylinders and also isolating them from each other. At this stage only nuclear decay is occurring independently in each of the cylinders.

Figure 3 shows a typical test in which buildup was initiated at Day 7951 and isolation at Day 8022. Different signal rise patterns occur in the two cylinders (Fig. 4) where the rise in direction Pole is fast relative to the rise in V_2_P. In both directions the gamma radiation from radon attains a maximum at different times which is followed by a semi-exponential decrease to a quasi-stable level. The peak of the V_2_P lags the peak at Pole by around 1 day. The decrease to the plateau is fast at Pole relative to V_2_P. The plateau level at direction V_2_P is around 50% higher than at Pole. At the quasi-stable level (plateau) the inter-calibrated count-rate at the V_2_P detector is 6×10^5 (per 15-minutes) compared with 4×10^5 at detector Pole. This indicates (again) the effect of directionality of the gamma radiation.

Daily Radon (DR) signals are superimposed on the decrease and the following plateau (Figs. 3, 5) and are absent in the environmental gamma radiation (Fig. 5). These DR signals contain a primary 24-hour periodicity (S1) and a minor 12-hour (S2) periodicity. The pattern of the DR signal is different at the two directions, in term of both signal form and phase.

Closing the valves on the source and on each cylinder initiated independent decay runs in the two cylinders. This results in a well behaved and statistically significant exponential decrease, which is different in the two directions. Figure 6 shows the dissimilar exponential decrease of the gamma radiation in the two directions once a decay run is initiated on Day 7900. In both canisters the calculated apparent-half-life

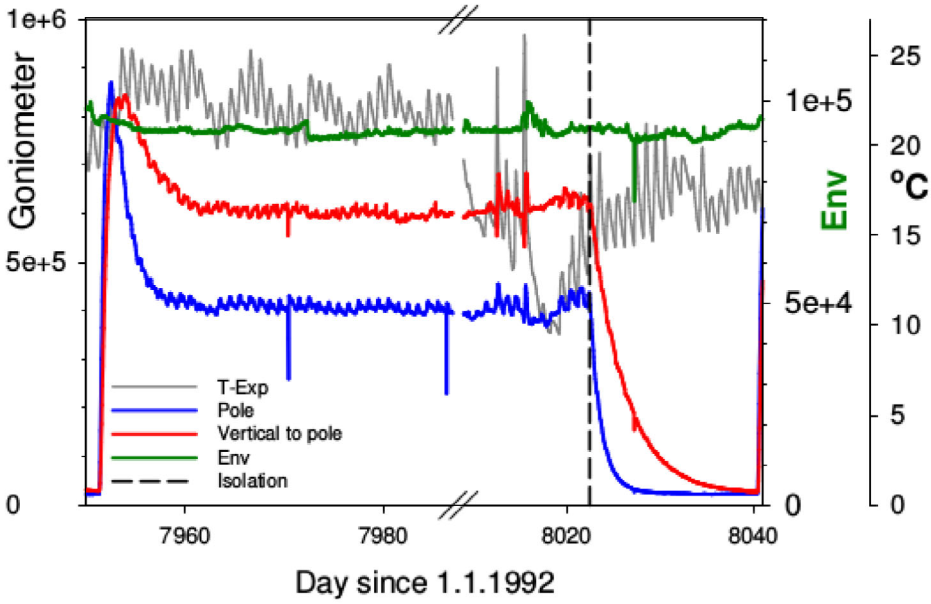

Fig. 3. A test cycle in the experiment consisting of a) opening the radon source leading to rise in the signal; b) attaining a maximum which is followed by a decrease to a c) steady state level with DR signals, and d) an exponential decrease once isolated from the source (vertical line). Temperature on experiment and environmental gamma radon in lab are also shown.

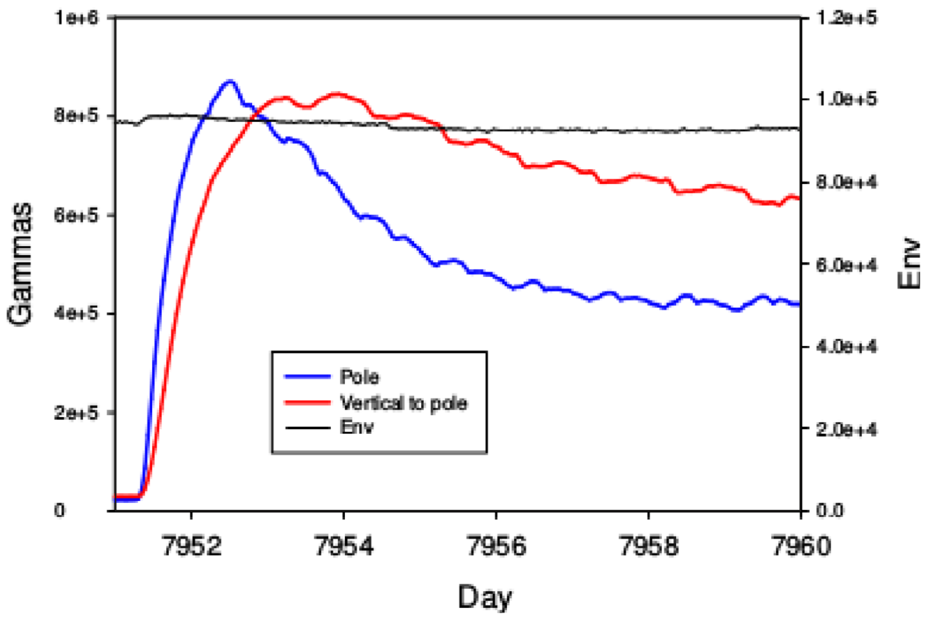

Fig. 4. Detail of Fig. 3 showing the different patterns at the two direction during: a) the rise of the signal (after opening the source), and b) the stabilization to the plateau level. DR signals are superimposed in both directions. The pattern of the environmental gamma radiation is also shown.

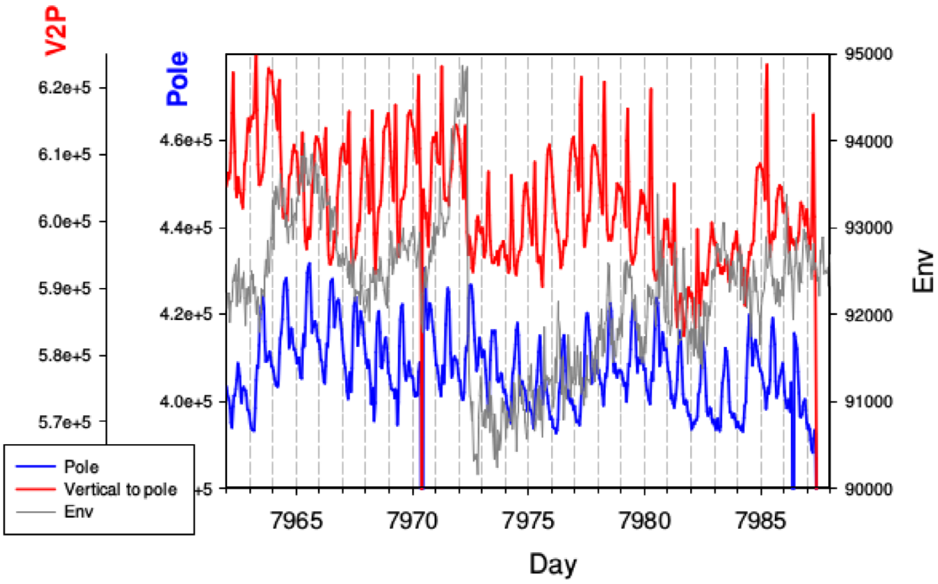

Fig. 5. 26-day interval at the steady state level demonstrating the DR signals. Different patterns of the DR signal occur in the different directions. S1 and S2 signals are observed in the time series. The pattern of the environmental gamma radiation is also shown (grey).

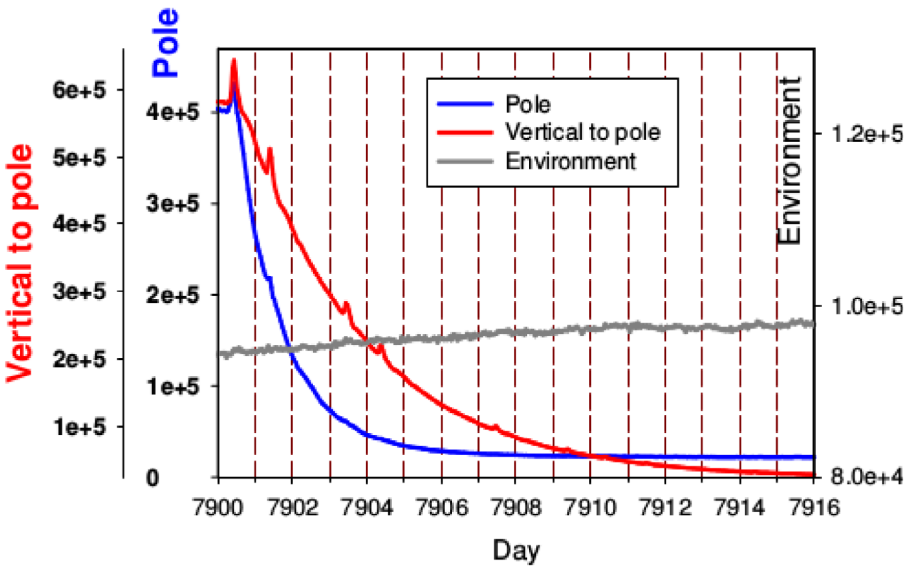

Fig. 6. Exponential decrease during a decay run (#5) performed in the goniometric experiment. AHL values are: Pole = 0.861 ± 0.003 days; Vertical to pole = 2.308 ± 0.008 days.

(AHL) differs from the accepted half-life of radon (3.82 day). The AHL along the Pole direction is 0.86 days and along the V_2_P direction it is 2.3 days. Similar AHL values have been obtained in further (12) decay runs performed with this setup.

With the total isolation of the cylinders, DR signals are superimposed on the decay pattern (Fig. 7). The DR signals are different in the two directions and their amplitudes are related to the overall remaining radon level. Such features have been

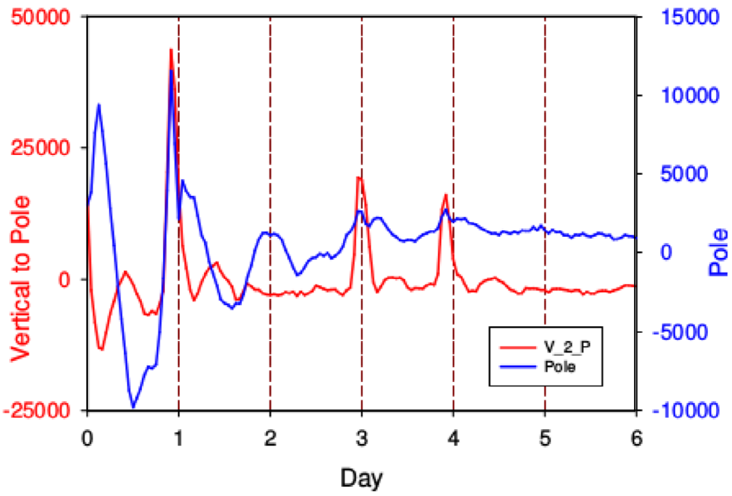

Fig. 7. Residuals of the exponential regression calculated in the decay run (#5) shown in Fig. 6. Time (day) is shown relative to isolation from the source. DR signals are observed in both directions exhibiting: a) decreasing amplitude related to the decay, b) S1 and S2 periodicities, and c) different phase in the two directions.

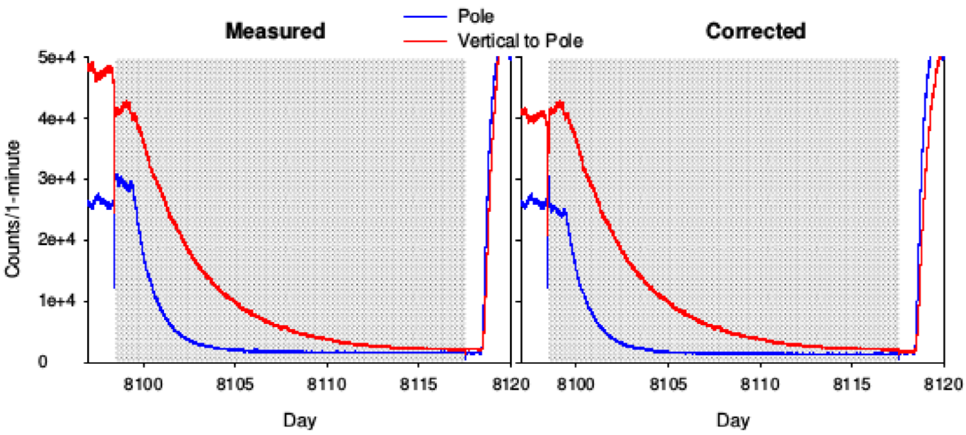

Fig. 8. Decay run #9 preformed using goniometer detectors at interchanged positions (hatched). Count rate is per 1-minute. Left: the measured signal at each detector; Right: signals corrected for difference in sensitivity. AHL values are: Pole $= 0.817 \pm 0.002$ days; Vertical to pole $= 2.493 \pm 0.006$ days.

reported in previously ECM experiments [19].

In one of the decay runs (#9) the two detectors of the goniometer were interchanged. Similar different AHL are obtained at the two directions (Fig. 8).

The results from this experiment confirm that gamma radiation from radon is spatially anisotropic. The configuration connects this anisotropy to the global directions of Earth.

More than 120 decay runs were performed during 2009-2014 in more than 20 experiments using different ECM configurations, varying host gas conditions, and with different alpha and gamma detectors. The obtained AHL values of radon range from 0.8 to around 4 days. Such variations are incompatible with common knowledge on radioactive decay.

4 Discussion

The results of the test are not in conformity with the presently accepted half-life of ^{222}Rn (3.8235 days). The latter was determined by [38] using simultaneously two radon samples of very different activity which are enclosed within gold capsules. Once a day the capsules were alternately inserted and measured in a NaI detector system having a 4π configuration. Our experiment differs from this in several respects: a) radon is contained in a larger air volume; b) a limited and separate angular sector of the 4π radiation field is recorded by each gamma detector; c) application of a sub-hour time resolution of the measurements, allowing the revealing of daily variations. Our result on the AHL of radon in this goniometric test, which deviates significantly from the accepted value, is representative of other tests using this setup. Furthermore, it is also comparable with further deviations we obtained in a series of experiments using other configurations (to be published separately). This leads to the impression that further factors may play a role.

The described test deals with two aspects of radon in air: a) apparent decay rate of radon as reflected by the AHL, and b) directionality, which is related to global orientation. These two aspects seem to be due the same influencing factor which is reflected at the pole direction by a relatively higher rate of decay (lower AHL) and a lower steady-state radiation level. The features of the goniometric experiment are in conformity with previous observations from ECM experiments in which radon signals occur in radon in air under confined conditions. We find different manifestation at different directions in terms of amplitude and phase of the periodic components of the DR signal. The observed association between directionality, global orientation, DR signal and variability in AHL is a further consideration for linking the phenomena to the influence of a component in solar radiation on nuclear processes in radon.

The outcome of the experiment links to recent observations and conclusions derived from investigations of geological radon and from simulation experiments. Apparently a gap in our understanding of the nuclear decay of radon in air is indicated, which should be addressed by the relevant physics community. However the identification of this issue resulted from earth science observations and considerations. Hence this topic requires a joint track combining physics and earth science investigations.

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