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Periodicity of Diurnal Variation of Soil Radon Concentration Levels and Temperature

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Abstract: Concentration levels of soil radon at various depths (25, 50 and 75 cm), over a period of two days, in a phosphatic formation site located in Irbid, Jordan were measured using an active detector, the RAD7® from Durridge Company, USA.

Periodicity of diurnal soil radon concentration levels is observed. Data were fitted using a cosine function. This fit produces a pattern similar to that of the variation of temperature of the soil with time obtained by the solution of the heat equation. The angular frequency of the periodicity is consistent with that of the temporal variation of the temperature of the soil for a given depth. Periodicity is clearly marked for the depths 25 cm and 50 cm, which is consistent with previous studies.

Keywords: Soil radon; Heat equation; Diurnal variation, RAD7®; Active detector; Phosphatic formation; Irbid.

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Introduction

Radon gas isotopes, ^{219}Rn (actinon), ^{220}Rn (thoron) and ^{222}Rn (radon) occur in the environment, being produced in the natural decay chains of ^{235}U, ^{232}Th and ^{238}U, respectively, all decaying by alpha emission. Because of its relative high life-time ($t_{½}$ = 3.82 days), the ^{222}Rn isotope is considered important in applied nuclear and environmental studies.

In addition to radon, soil gas measurements will normally also register the presence of thoron, depending on soil composition. It is well known that exposure of population to high concentrations of radon and thoron and their solid daughters for long periods leads to pathological effects like respiratory functional changes and lung cancer [1]. In this work, only soil radon will be considered. At least 80% of the radon emitted into the atmosphere comes from the top few meters of the ground [2]. The radon emanation rate varies from one place to another due to differences in radium concentration and soil parameters such as moisture content, porosity, permeability and grain size [3, 4]. The exhalation of radon from soil involves two mechanisms: diffusion and convection. These two mechanisms are affected by many factors including the properties of the soil [5].

The health hazards of radon and its decay products have led to study radon transport in soils and into buildings [6]. A thorough understanding of gas transport is required to evaluate these issues.

Diffusion of radon is characterized by the (effective) diffusion coefficient. The diffusion coefficient of ^{222}Rn in air is 0.11 cm^2 s^{-1} [7]. The effective diffusion coefficient is about a factor of four smaller than the free air diffusion coefficient in fairly dry soils [6, 8]. Effective diffusivities in saturated systems are about four

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orders of magnitude less than those in dry soils [9]. Models explaining the diffusion of soil radon are, in general, based on the solution of the diffusion equation in the quasi-homogenous approximation with boundary conditions assuming a semi-infinite medium [8, 10].

Many studies of diurnal variation of indoor radon [11, 12], radon near the ground [13, 14], soil radon [15, 16] and even in the atmosphere [17 – 20] exist in the literature. The case of soil radon is particularly interesting for the studies of earthquakes [21 - 23].

Neves *et al*. studied the indoor radon periodicities and their physical constraints in the inland Coimbra region in Portugal [11]. They reported a well marked periodicity in radon activity with maximum values occurring more frequently in the morning between 9 and 10 a.m. Their principle conclusion is that daily variations are shown to have no relation with earth tides and their amplitudes exhibit a significant correlation with outdoor temperature; no dependence on barometric pressure was found. Rainfall disturbs the observed daily radon cycles through a strong reduction of their amplitude, but has no effect on the long-term variability of the gas concentration.

Diurnal radon variations in the upper soil layers and at the soil-air interface related to meteorological parameters were studied by Schubert and Schulz [16]. In their work, conducted in the laboratory using a column consisting of a homogenous mixture of dry sand and uranium tailings, measurements of radon concentration levels were performed in order to obtain information on the radon transport under well defined conditions

The dependence of radon concentration has been exclusively studied on the soil/air temperature gradient and on the wind speed. The soil moisture content has been kept constant. Significant diurnal variations of the radon concentration were detected in the uppermost soil layer and at the soil/air interface. Such a behavior was not found deeper than 30 cm in soil layers. It is argued that the diurnal radon variation in the uppermost soil layer is mainly associated with the diurnal inversion of the soil/air temperature gradient giving rise to a convective soil gas migration additional to the common upward diffusion processes, whereas the diurnal variation of the radon concentration at the soil/air interface is caused by the interplay of the temperature gradient and the wind speed. No impact of atmospheric pressure variation on the radon migration has been observed [16].

Dueňas and Fernandez [22] measured soil radon concentration at different depths at two sampling sites, in the region of Malaga, Spain, to determine if these exhibited significant temporal and quantitative relationships with a number of earthquakes. The magnitudes of the earthquakes ranged from 2 to 4. While both positive and negative precursory perturbations of radon concentrations were observed, in some cases the perturbations were detected after the earthquakes, and several earthquakes occurred without any perturbation of the radon signal being observed, either before or after the events. The study was also aimed at evaluating exhalation of soil radon as an early warning of imminent earthquake activity but meteorological factors were observed to interfere with, or mask the signal so that radon exhalation alone does not appear to be a reliable parameter for predicting earthquakes.

The aim of this work is to study the diurnal variation of soil radon concentration levels, and to correlate it with the variation of temperature with time using a mathematical model based on the heat equation. The influence of atmospheric pressure on soil radon diffusion as well as that of the soil properties: moisture, water saturation and porosity, are not considered in this work. This is justified by the fact that the measurements have been performed over a period of two days where the effects of the variation of these parameters can be neglected.

Methodology and Experimental Procedure

A calibrated active radon device, RAD7®, (Durridge Company, USA) was used for the measurement of soil radon activity levels. RAD7® is a portable radon gas surveyor (PRS) which is suitable for continuous, or grab, sampling measurements of soil radon gas by a solid state detector. RAD7® uses a silicon detector that converts alpha radiation directly to an electrical signal. One important advantage of solid state detectors is their ability to electronically determine the energy of alpha particle. Every nucleus of ^{222}Rn decays through the sequence ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po and ^{210}Pb. With each transformation the nuclei emit alpha, beta or gamma radiation. RAD7® is designed to detect alpha radiation only [24].

RAD7® can be used in a continuous monitoring mode, which is the most appropriate for this study. A group of commands configure RAD7® to perform tests according to the study's requirements. The used probe, a simple transparent plastic tube, is connected to the device for the measurement of soil radon concentration at different depths (25, 50, 75 cm) below the soil/air interface. The RAD7® pack contains a drying unit which is connected to the probe (Fig. 1).

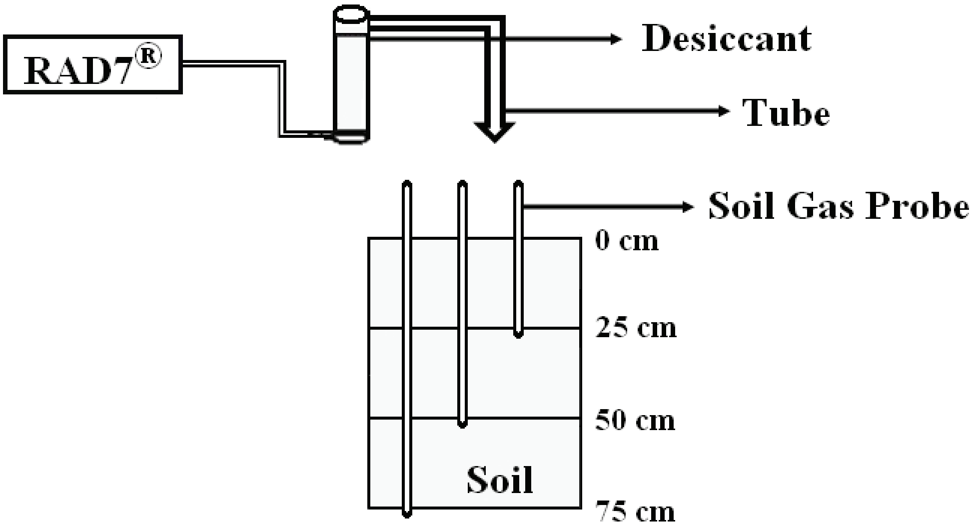

FIG. 1. Field work - Schematic.

It is necessary first to purge the RAD7® for one hour or more with dry, fresh air, before connecting the probe. The purging procedure is achieved with the probe disconnected but the drying unit hooked up to RAD7®. The used measurement protocol is a 2 hours cycle, 24 recycles, auto mode thoron off and auto pump. The associated printer prints out a header for each measurement. The test is started after checking the header to make sure that the setup is what is required [25].

The total duration of a run was determined by multiplication of cycle time by recycle number; so each depth was automatically measured for 48 hours. In auto pump setting, the pump is switched on for 4 minutes and the air containing radon gas is pumped to the detector through the drying unit. There will be a reading after every two hours cycle, though the first reading will be low because the ^{218}Po decay rate in the detector takes more than 10 minutes to reach equilibrium with the radon concentration in the measurement chamber. Readings are stored in the RAD7® memory for later use.

The site chosen is located near the campus of Yarmouk University, Irbid - Jordan. The soil formation is phosphatic [26]. Data were taken in autumn during the period Nov. 15-21, 2011, just before the beginning of the rainfall season in the region, which means that variation of water saturation can be neglected. A hole was dug and the probe was inserted at depths of 25, 50 and 75 cm (Fig. 1). When measurements were taken at a depth of 25 cm, the external temperature varied between 23.1 °C and 24.4 °C for the first day and between 22.1 °C and 23.7 °C for the second day. For the other depths, the minimum temperature registered was 20.7 °C and the maximum one was 25.8 °C.

RAD7® is calibrated by the manufacturer against a master instrument, which, in turn, is calibrated against a standard maintained by the National Radiological Protection Board (NRPB). The overall calibration accuracy is estimated to be about ±5%. In most circumstances, the precision of individual RAD7® measurements of radon concentration is limited by counting statistics. The error values in Table 1 are those given by the device itself. In the figures, an overall error of ±5% is considered for each individual measurement.

The first remark is the fact that the highest values were obtained in the morning period (between 7 and 10 am). The lowest values were obtained in the late afternoon period (depth 25 cm) or in the evening (depths 50 and 75 cm).

TABLE 1. Highest and lowest soil radon concentration levels (C_{Rn} in Bq/m^3) for all depths.

Depth (cm)	First day		Second day	
	Highest value	Lowest value	Highest value	Lowest value
25	6335 ± 156 at 07:00 am	1855 ± 105 at 5:00 pm	4504 ± 135 at 05:00 am	697 ± 85 at 05:00 pm
50	19088 ± 251 at 08:00 am	12336 ± 332 at 10:00 pm	19394 ± 2 at 12:00 pm	14782 ± 225 at 08:00 pm
75	28823 ± 305 at 10:00 am	19568 ± 374 at 12:00 am	31768 ± 324 at 12:00 pm	21114 ± 267 at 12:00 am

Results and Data Analysis

Table 1 shows the highest and lowest measured values for all depths.

Figures 2, 3 and 4 show the results of measurements, over the two days period, of relative soil radon concentration levels (C/C_{max}) as a function of time, for the depths: 25, 50 and 75 cm, respectively. C_{max} is the maximum concentration value for a given depth over 24 hours. The vertical axis is thus unitless. The very first measurement for each set of data is neglected, thus the number of data points considered is 23 for each depth.

The time scale in Fig. 2, for the depth 25 cm, is such that t = 0 h corresponds to the reading taken the first day at 1:00 am. For the depths 50 and 75 cm, t = 0 corresponds to the reading taken the first day at midnight and 02:00 am, respectively. The time interval between two measurements is two hours. The figures will be discussed later in this paper.

A preliminary non-linear least squares fit using a cosine function ($A\cos(\omega t + \delta) + B$) was used in order to smooth the data showing the periodic behavior of soil radon concentration with time. The fit parameters are: A and B are dimensionless offset parameters, ω is the angular frequency and δ is a phase constant. The value of these parameters for data at depths 25, 50 and 75 cm for the two days period are shown in Figures 2 to 4, respectively.

The periodicity of the soil radon levels is clearly observed for the depths 25 and 50 cm which is consistent with previous studies [16].

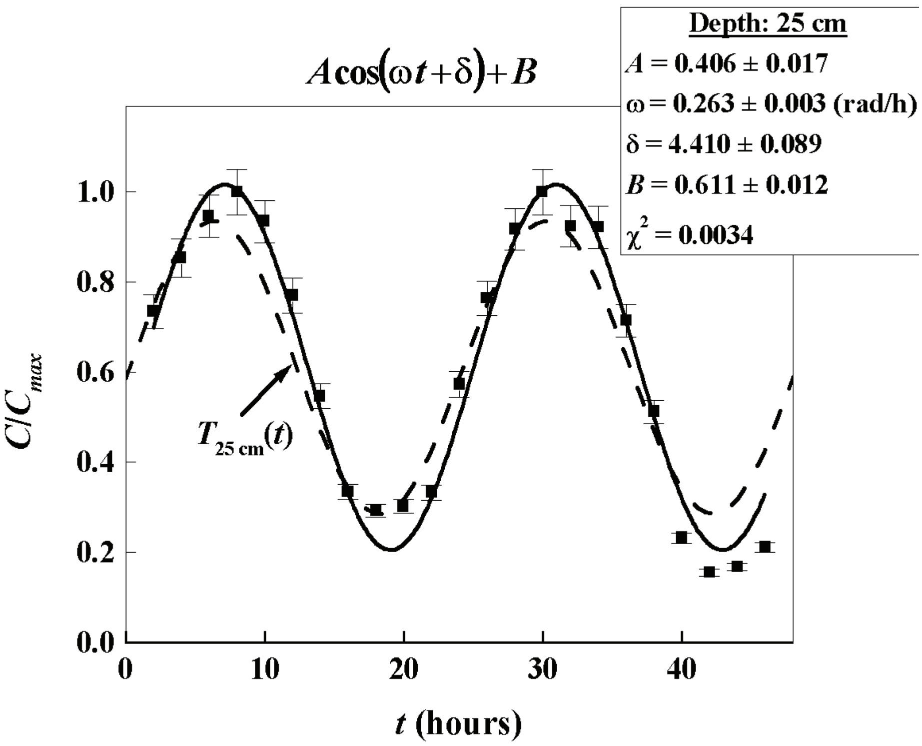

FIG. 2. Diurnal variation of soil radon concentration levels at depth 25 cm over two days (48 h cycle). See text for $T_{25\,\mathrm{cm}}(t)$.

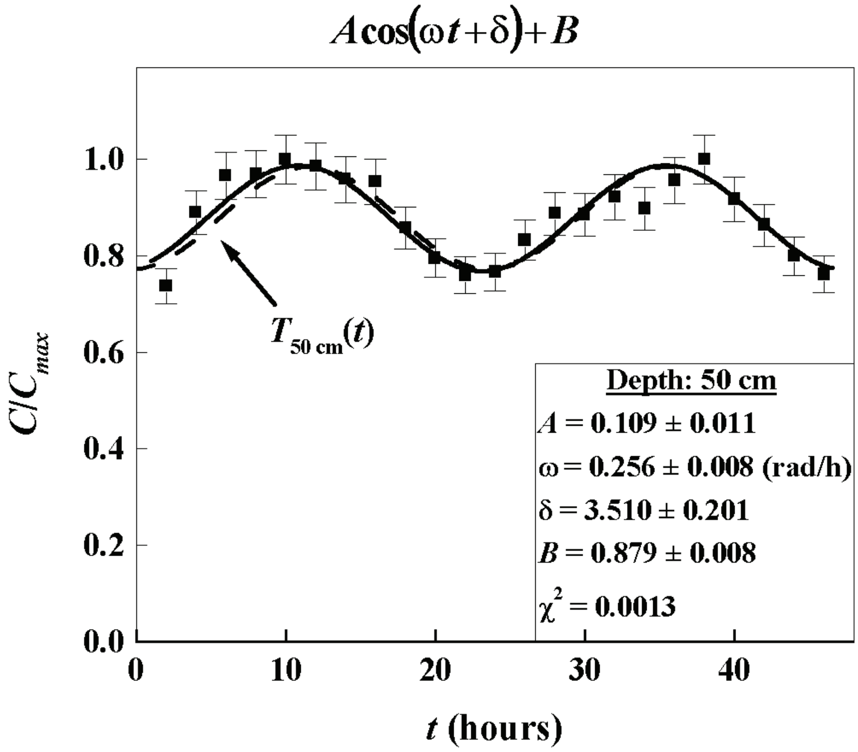

FIG. 3. Diurnal variation of soil radon concentration levels at depth 50 cm over two days (48 h cycle). See text for $T_{50\ cm}(t)$.

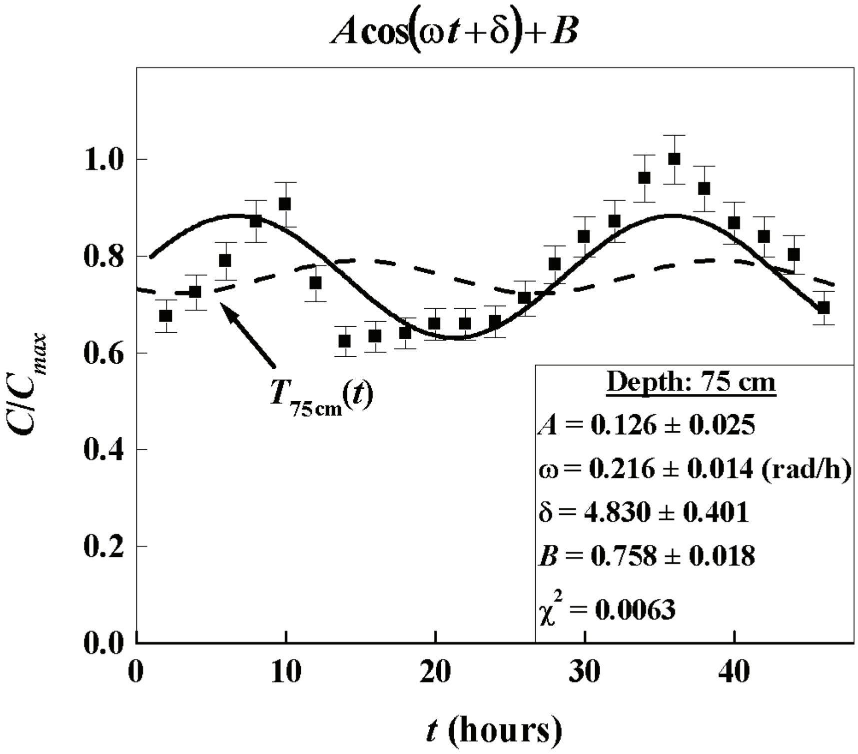

FIG. 4. Diurnal variation of soil radon concentration levels at depth 75 cm over two days (48 h cycle). See text for $T_{75\ cm}(t)$.

Tentative explanation of the choice of a cosine fitting function

The fact that the cosine function fits well the data was thought of being related to the periodic variation of the temperature of the soil. The following assumptions support this idea:

1- The soil is considered as a semi-infinite homogenous medium. Porosity of the soil η is considered to be constant (η ≈ 0.5 as confirmed by a separate study). Since measurements in this study were done just before the beginning of the rainfall season in the region, soil moisture variation is neglected.

2- Dependence of soil radon diffusion on pressure is also neglected. For relatively short periods of measuring time, atmospheric pressure in the study region does not vary. In addition, various previous studies, in particular that of Schubert and Schulz [16], support this assumption.

3- The temperature of the soil can be obtained from the solution of the heat equation:

$$\nabla^2 T = -k \frac{\delta T}{\delta t} \quad (1)$$

with a heat wave, originating from the sun and hitting the soil, given by:

$T(t) = T_0 \cos(\omega t + \delta)$ at $z = 0$. (2)

k is the thermal diffusivity. k is related to the Fourier coefficient, a, of a medium of density ρ and specific heat capacity at constant pressure c_P, by the relation:

$$k = a \frac{\rho}{c_P} \quad (3)$$

T_0 is the maximum amplitude of the heat source (the sun) generally taken to be 15 °C, i.e. the average temperature of the earth, ω is the angular frequency of the earth (ω = 7.27 10^{-5} rad/s = 0.262 rad/hour) and δ is a phase constant.

4- The temperature field, $T(z,t)$, at depth z below the soil/air interface is the solution of the heat equation with the boundary conditions:

$$T(0,t) = T_{av} + T_0 \cos(\omega t) \quad (4)$$

$$T(\infty,t) = T_{av} \quad (5)$$

where T_{av} is the average temperature of the soil-air interface (around 20 °C in this study). This solution has the form:

$$T(z,t) = T_{av} + T_0\, e^{-F z} \cos\left(\omega\left(t - \frac{z}{v}\right)\right) \quad (6)$$

Taking a, the Fourier coefficient for soil (0.018 $cm^2\,s^{-1}$),

$$F = \sqrt{\omega/(2a)} = 0.0449\,\mathrm{cm}^{-1},$$

$$v = \sqrt{2a\omega} = 0.0016\,\mathrm{cm\,s}^{-1} = 5.827\,\mathrm{cm\,h}^{-1}$$

The fitted values of ω, δ and B are used in order to plot the dimensionless function:

$$\left.\begin{aligned} T_z(t) &= \frac{T(z,t) - T_{av}}{T_0} \\ &= B + e^{-Fz} \cos\left(\omega\left(t - \frac{z}{v}\right)\right) \end{aligned}\right\} \quad (7)$$

for the depth z = 25 cm. The offset parameter B obtained by the fit is used here to scale the function with the data. The origin for $T_z(t)$ is shifted by two hours. The dimensionless functions $T_{25\mathrm{cm}}(t)$, $T_{50\mathrm{cm}}(t)$ and $T_{75\mathrm{cm}}(t)$ are, respectively, shown in Figures 2, 3 and 4 (dashed line) – no units needed.

For clarity we give below these functions,

$$T_{25\mathrm{cm}}(t) = 0.611 + 0.325 \cos\big(0.262\big((t-2) - 4.290\big)\big) \quad (8a)$$

$$T_{50\mathrm{cm}}(t) = 0.879 + 0.105 \cos\big(0.262\big((t-2) - 8.580\big)\big) \quad (8b)$$

$$T_{75\mathrm{cm}}(t) = 0.758 + 0.034 \cos\big(0.262\big((t-2) - 12.870\big)\big) \quad (8c)$$

The diurnal variation of soil radon concentration levels follows a pattern similar to that of the temperature variation with time for the depths 25 and 50 cm.

The cosine behavior is less marked for the depth 75 cm, and as can be seen in Fig. 4 the diurnal variation of temperature shows no correlation with the variation of C/C_{max}. The incoming solar radiation energy is utilized as heat as it travels down the soil profile. Thus, available energy decreases with depth. An amplitude damping is observed. This damping is due, as expected, to the factor

e^{-Fz} in Eq. (7). This damping causes a distortion which is visible in Fig. 4. In addition, there is a delay in the time at which any specific location on the temperature cycle reaches a given point in the soil. This time lag becomes more pronounced with increasing depth [27].

Conclusion

The present study shows that the measured periodicity of soil radon concentration levels can be fitted using a cosine function. This fit produces a pattern similar to that of the variation of temperature with time at the depths 25 cm and 50 cm. The variation of temperature is obtained by resolving the heat equation for a heat wave, originating from the sun, hitting the soil which is approximated as a semi-infinite homogenous medium. Besides its theoretical importance, this work may help in the study of earthquakes. An application of this work would be the following: permanent monitoring of the soil radon concentration levels in real time and their pattern similar to the diurnal variation of the temperature of the soil, could indicate, in the case of sensitive perturbations in the cosine pattern, changes of the structure of the soil due to tremors following an earthquake.

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