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Comparative Study of Soil Radon Concentration Levels Using Active and Passive Detectors

N. M. Ershaidat, B. A. Al-Bataina and W. S. Al-Rayashi

Department of Physics, Yarmouk University, Irbid 21163, Jordan.

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Abstract: Passive radon diffusion dosimeters containing CR-39 detectors, and an active electronic device, RAD7[®], were used for measuring soil radon concentration levels, at different depths, in a phosphatic site in the city of Irbid, north of Jordan. Time-averaged values of the active detector readings based on the periodicity of soil radon concentration levels are compared with measurements of the passive detectors at different depths. An acceptable agreement is observed.

Highlights

- Soil radon measured using an active detector (RAD7[®]) and a passive one (CR-39).
- Diurnal periodicity of soil radon diffusion and time-averaged values of RAD7[®] readings.
- The “average” readings of the RAD7[®] are compared to the measurements of the CR-39.

Keywords: Soil; Radon; Active detector, RAD7[®], CR-39 dosimeters; Phosphate.

Introduction

Radon (²²²Rn) study and measurements is an important topic in the applied nuclear and environmental field. Health hazards due to the exposure of population to radon and its progeny for long periods are the motivation behind many studies in this field. The major source of radon is soil and rocks beneath it. At least 80% of the radon emitted into the atmosphere comes from the top layers of the ground (around 1.5 m) [1].

Radon emanation is associated with the presence of uranium and its daughter radium in soil. Different types of rocks have typically different concentrations of uranium, thus different concentrations of radon, in soil and in the air above it. In particular, phosphate deposits contain the naturally occurring radionuclides ²³⁸U and ²³²Th together with their decay progeny. The radon gas belongs to the ²³⁸U chain [2]. In Jordan, phosphate is an important national economic source. Many regions in the country are of the type of phosphatic formation. Radon environmental studies in many locations in Jordan, where the soil is phosphatic, were

initiated more than 20 years ago. A national effort for the assessment of radon in dwellings, soil, water sources, ...etc. had been carried out since the early 1990s [3-6]. The present work is a continuation of this effort.

There are several detection techniques for measuring radon concentration levels, among which are solid-state nuclear track detectors (SSNTD), ion chambers and solid-state detectors. They are mainly grouped into two categories: 1) passive diffusion radon dosimeters using CR-39 detectors [7-8] and the LR115 detectors [9]. 2) active detectors based on continuous radon sampling requiring an electric power [10] such as the RAD7[®] [11] and AalpaGuard[®] [20].

In this work, both CR-39 and RAD7[®], from Durridge Co., Bedford, MA 01730, USA, have been used. Time integrated average values for soil radon concentration levels are obtained using the CR-39 dosimeters. In contrast, the

active detector RAD7[®] gives multiple readings over a given period of time.

Criteria for comparing these two types are mainly related to measurement precision and time needed for performing measurements in different operational conditions: atmospheric, landscape, ... etc. [12].

The present comparative study is based on measurements of soil radon concentration levels in a phosphatic formation, using an active detector; RAD7[®], in NORMAL mode, and a passive one; the CR-39 dosimeter.

There are several similar studies in literature. It is worth mentioning that none of them was concerned with phosphatic formations.

S. Giammanco et al. [13] made a comparison between different detection systems for measuring soil radon concentration levels along an active fault: the case of the Pernicana fault system, Mt. Etna (Italy). Three techniques were used: SSNTDs (CR-39 type), active detector (RAD7[®]) and soil CO₂ efflux measurement devices. Their main conclusion was as follows: while spot measurements of soil radon using RAD7[®] are useful for the quick recognition of high emission sites to be later monitored for ²²²Rn variations in time, SSNTD (CR-39 type) allow for the temporal monitoring of a relatively large number of sites, but cannot distinguish short-term changes due to their long integration times.

M. Abo-Elmagd et al. [14] measured radon and its related parameters inside seven ancient Egyptian tombs of the Valley of the Kings in Luxor, using passive (CR-39) and active (Alpha-Guard analyzer) techniques. The measurements were performed throughout winter and summer seasons. They concluded that active measurements are precise and provide fast results, but cannot be used for a long time. The CR-39 can be used over long exposure periods; i.e., this technique is useful in *low-radioactivity* measurements.

G. Jönsson et al. [15], in the framework of an EU-radon project, measured radon levels with passive, solid state nuclear track detectors of the types LR 115, CR-39 and Makrofol and with active electronic devices. They showed that active detectors are able to give time resolved data, while passive detectors give time integrated data.

Finally, in a recent work, Espinosa et al. [10] published the results of an intercomparison of indoor radon data using NTDs (nuclear track detectors) and four different dynamic recording systems, including RAD7[®]. Measurements were carried out in a controlled one-room cellar carved into volcanic rock presenting an almost constant radon emanation throughout the four seasons. Averages of the sum of short-term measurements over a long period were used in order to make a comparison with the passive detectors' response. Their main conclusion was that the active electronic devices can give good account of short-term indoor fluctuations even if the average radon concentrations of different monitors differ by 35%. They attribute these fluctuations to sudden changes in temperature and humidity of the air in the cellar.

The previous results [10] were based on averaging over a long period of time (three months). In this study, the idea is to use the periodicity of diurnal soil radon concentration levels in order to find an average over a relatively short period of time (7 hours).

Methodology and Experimental Procedure

Active detectors are designed to be used in measurements of indoor radon, especially in dwellings for safety purposes. For measurements of radon in water samples or in soil, simple extra accessories are needed.

RAD7[®], recently acquired, was used for the first time in this work. It is calibrated by the manufacturer against a master instrument, which, in turn, is calibrated against a standard maintained by the British National Radiological Protection Board (NRPB), known as HPA (Health Protection Agency) since 2004. The overall calibration accuracy is estimated to be about $\pm 5\%$.

RAD7[®] is a versatile radon detector. It is a measuring instrument used in laboratories and research work by radon testers, mitigators and home inspectors, in various sites with different climatic conditions. RAD7[®] is a computer-driven electronic detector and easy to use, with pre-programmed set-ups for common tasks.

The RAD7[®]'s internal sample cell is a 0.7 liter hemisphere, coated on the inside with an electrical conductor. A solid-state, ion-implanted, planar, silicon alpha detector is at the

center of the hemisphere. The high voltage power circuit charges the inside conductor to a potential of 2000 to 2500V, relative to the detector, creating an electric field throughout the volume of the cell.

RAD7[®] is a sniffer that uses the 3-minute alpha decay of a radon daughter, without interference from other radiations, and the instantaneous alpha decay of a thoron daughter. It sniffs out entry points and radon gushers and recovers in minutes from high radon exposures.

The site chosen (latitude 32° 32', longitude 35° 51') is in the city of Irbid, Jordan, near the campus of Yarmouk University. The bedrock is a mixture of Al-Hisa Phosphatic (AHP) Limestone and Amman Silicified Limestone (ASL) formations [16].

A hole was dug and, for averaging purposes, two previously calibrated CR-39 dosimeters of closed can technique [6] were placed at 100, 80, 60, 40, 20 and 0 cm depths. Thus, a total of twelve dosimeters were used. Each time a dosimeter was planted, the depth was refilled with the soil extracted while digging. No external source of soil was used.

All dosimeters were left *in situ* for 22 days before being collected, so that radon gas can reach secular equilibrium with its parent (²²⁶Ra). In order to extract the soil radon concentration

levels, a standard procedure for the treatment of the dosimeters is followed in a similar manner as in [6]. A 15% error estimation is also made as in the previous reference.

When using the active detector, it is essential to collect radon samples without exposing them to outside air. For this purpose, a special probe was devised for this study. A flexible transparent tube, 0.45 inch in diameter and 10 m in length, is inserted into metallic pipes of half an inch diameter. The metallic pipes' lengths were (10, 30, 50, 70, 90 and 110 cm) used for the depths: 0, 20, 40, 60, 80 and 100 cm, respectively. Thus, each of the latter pipes appears 10 cm above the soil. The probes were inserted at the same time as the dosimeters were planted. To avoid leakages of all kinds, the metallic pipes were tightly closed. Fig. 1 shows a schematic representation of the setup. Measurements were carried out on a daily basis for each depth for eight hours from 09:00 am to 5:00 pm. Readings after every half-hour cycle were registered. Thus, the total number of readings taken is sixteen (16). To avoid the effect of deposition of solid particles (decay products of radon) on the active detector, RAD7[®] was put off for the rest of the day between two consecutive sets of measurement in order to get rid of these solid daughters.

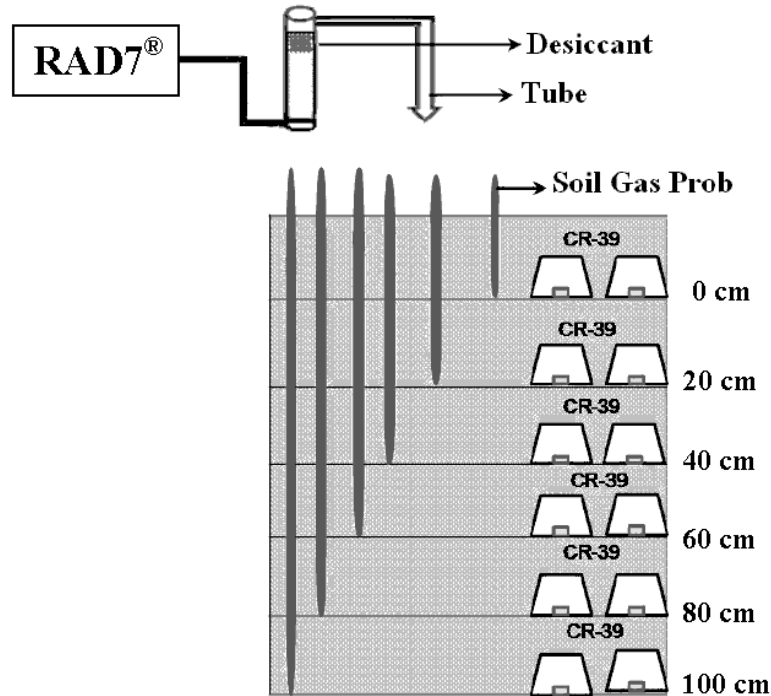


FIG. 1. Experimental setup - schematic.

Data were taken from Feb. 26 to March 3, 2011, during the rainfall season in the region. There was no rainfall during this period and temperatures of the air registered by RAD7[®] ranged between 15.5 °C and 19.4 °C. In addition, the studied soil represents an acceptable homogeneity and thus variations of moisture and water saturation will be neglected in this study [16].

The first two readings will be low, because the ²¹⁸Po 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. Thus, 14 of the 16 recorded readings, for each depth, will be used in this work.

Because of the high quality alpha semiconductor detector, and unique real-time spectral analysis, the RAD7[®] background is vanishingly small and is immune to the buildup of lead-210, which plagues other instruments. Intrinsic background may add less than 1 Bq/m³ to a typical measurement, far below the radon concentration of outdoor air (usually several Bq/m³ to few tens of Bq/m³). All the previous details as well as the technique of calculating the radon concentration levels separately from those of thoron and corrections due to absolute humidity and more technical specifications can be found in the RAD7[®] user's manual [11]. The practical timetable followed is shown in Table 1.

TABLE 1. Practical timetable for measuring soil radon concentration levels.

Depth (cm)	0	20	40	60	80	100
RAD7 [®] (8 hours daily)	Day 1	Day 2	Day 3	Day 4	Day 5	Day 6
CR-39	22 days					

Results and Data Analysis

The RAD7[®] Readings

Figures 2-6 show radon concentration levels using RAD7[®] (C_{RAD7}) for the depths 20, 40, 60, 80 and 100 cm. In most circumstances, the precision of individual RAD7[®] measurements of radon concentration is limited by counting statistics. In Figs. 2-6, an overall error of $\pm 5\%$ is

considered for each individual measurement instead of using the error given by the device itself. For the depth $z = 0$ cm, the maximum measured value is adopted; namely 1170 Bq/m³. As explained earlier, the first two readings, for each depth, are dropped.

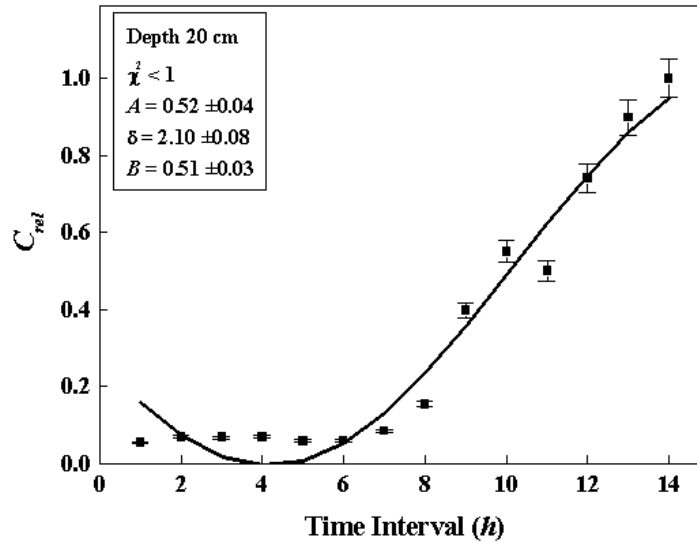


FIG. 2. RAD7[®] readings at a depth of 20 cm. See text for the fit details.

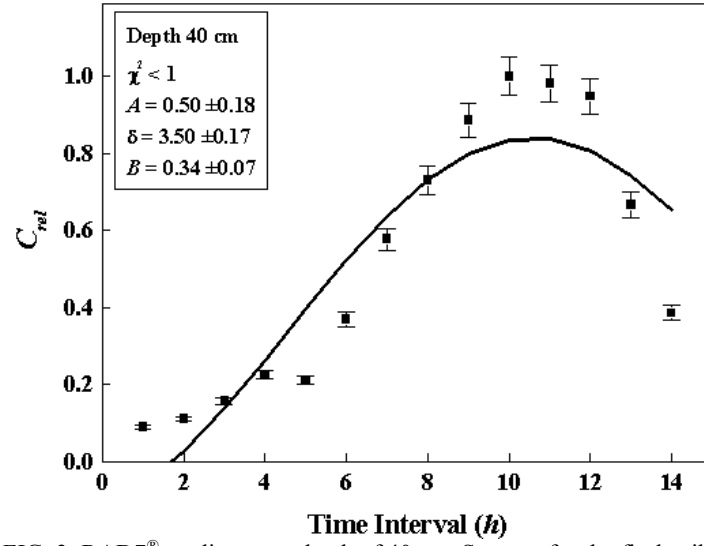


FIG. 3. RAD7[®] readings at a depth of 40 cm. See text for the fit details.

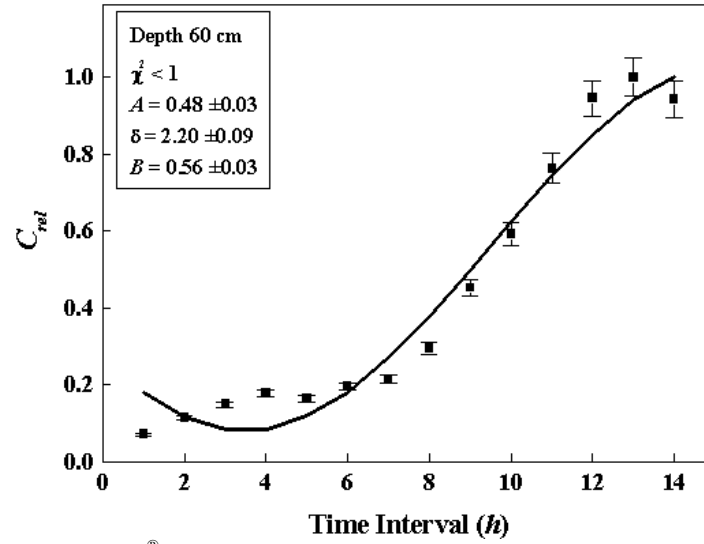


FIG. 4. RAD7[®] readings at a depth of 60 cm. See text for the fit details.

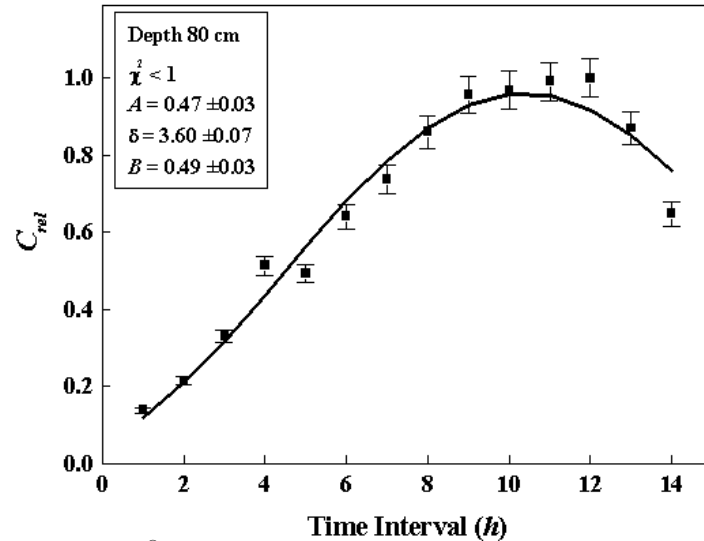


FIG. 5. RAD7[®] readings at a depth of 80 cm. See text for the fit details.

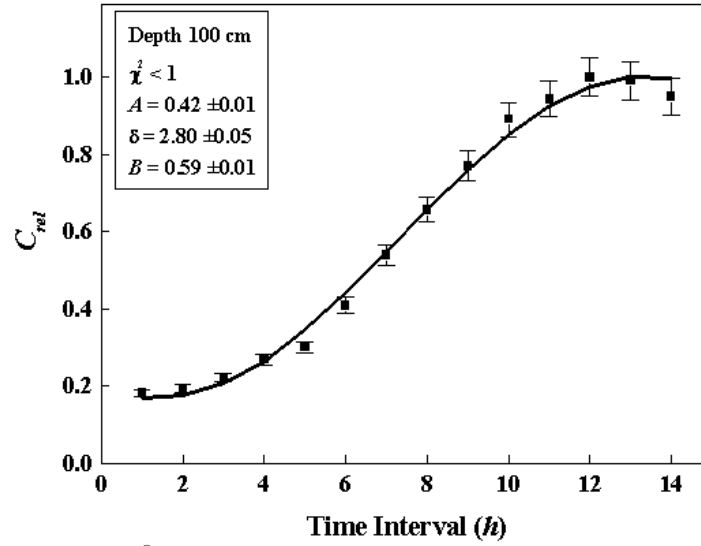


FIG. 6. RAD7[®] readings at a depth of 100 cm. See text for the fit details.

The CR-39 Readings

Table 2 shows the soil radon concentration levels measured using the CR-39 dosimeters (C_{CR39} in Bq/m³) for the depths 0, 20, 40, 60, 80 and 100 cm. Each of these measurements is the average reading of two dosimeters positioned at a given depth.

Fig. 7 shows a comparison between the two types of detectors, for the same depths mentioned above, where the averages of the RAD7[®] readings for each depth were considered.

Another possible comparison can be achieved where the RAD7[®] data is fitted to a cosine function. This choice is based on the well established periodicity of soil radon diffusion

[17-18]. The basic idea is that, for a given depth z , variation of soil radon concentration with time follows a similar pattern as the soil's temperature. In the appendix below, the major ideas are discussed.

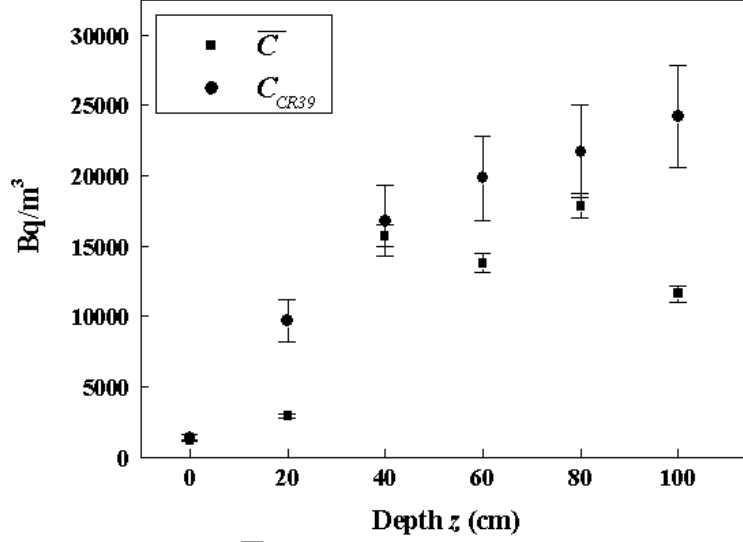
We actually use the relative quantity, for a given depth,

$$C_{rel} = \frac{C_{RAD7}}{C_{max}} \quad (1)$$

where C_{RAD7} and C_{max} are respectively the reading of RAD7[®] and the maximum value of the set of 14 readings of the active detector.

TABLE 2. C_{CR39} , C_{max} , $\bar{C}$ and $\bar{C}_{Fit}$ for all depths. See text for the definitions.

Depth z (cm)	0	20	40	60	80	100
C_{CR39} (Bq/m ³)	1390	9682	16778	19817	21720	24206
C_{max} (Bq/m ³)	1170	7904	25168	28359	23041	17328
$\bar{C}$ (Bq/m ³)	1170	2655	13197	12303	15412	10299
$\bar{C}_{Fit}$ (Bq/m ³)	-	2907	15714	13782	17852	11588
$diff_1 = \frac{ C_{max} - C_{CR39} }{C_{CR39}} \times 100\%$	16	18	50	43	6	28
$diff_2 = \frac{ \bar{C} - C_{CR39} }{C_{CR39}} \times 100\%$	16	73	21	38	29	57
$diff_3 = \frac{ \bar{C}_{Fit} - C_{CR39} }{C_{CR39}} \times 100\%$		74	18	39	27	57

FIG. 7. $\bar{C}$ (RAD7®) vs. C_{CR39} for all depths.

Eq. A7; namely $C_{fit} = A \cos(\omega t + \delta) + B$ with $\omega = 0.262$ rad/hour, is used to fit the data (C_{rel}). The fit parameters are: the dimensionless offset constants A and B and the phase constant δ . The pre-imposed confidence level is 90%.

Figures 2-6 also show the fitting curves, as well as the resulting parameters of the fit: A , B and δ .

Although the periodicity of soil radon diffusion is admitted to be valid for small depths (typically 50 cm), the fit here was done for all depths from 20 to 100 cm. It is being used as a mathematical tool in order to find a time-averaged value for the set of readings of RAD7® for each depth.

This average is defined as:

$$\bar{C}_{Fit} = C_{RAD7} \times \frac{1}{13} \int_1^{14} (A \cos(\omega t + \delta) + B) dt \quad (2)$$

The obtained values, called ($\bar{C}_{Fit}$), are shown in Table 2. The simple average ($\bar{C}$) as well as (C_{max}) for each set of the RAD7® measured values are also shown for comparison purposes. The last rightmost columns in the table give the following differences:

$$\left. \begin{aligned} diff_1 &= \frac{|C_{max} - C_{CR39}|}{C_{CR39}} \times 100\% , \\ diff_2 &= \frac{|\bar{C} - C_{CR39}|}{C_{CR39}} \times 100\% , \\ diff_3 &= \frac{|\bar{C}_{Fit} - C_{CR39}|}{C_{CR39}} \times 100\% . \end{aligned} \right\} \quad (3)$$

Finally, Fig. 8 shows $\bar{C}_{Fit}$ compared to the readings of the CR-39 passive detectors.

Discussion

The following remarks can be drawn from the previous results:

- 1- Both types of measurements, using the RAD7® active detector and the CR-39 passive dosimeters, show that soil radon concentration levels increase exponentially with depth as expected and confirmed by many previous studies [19].
- 2- Data fitting to a time-dependent cosine function, inspired by the variation of the soil's temperature, is valid for small depths $z \leq 60$ cm. Extrapolating to larger depths give acceptable results.
- 3- Averages, over a short period of time, obtained using this fit, give values of soil radon concentration levels which are comparable, and in the right range, to measurements obtained using the passive detectors.

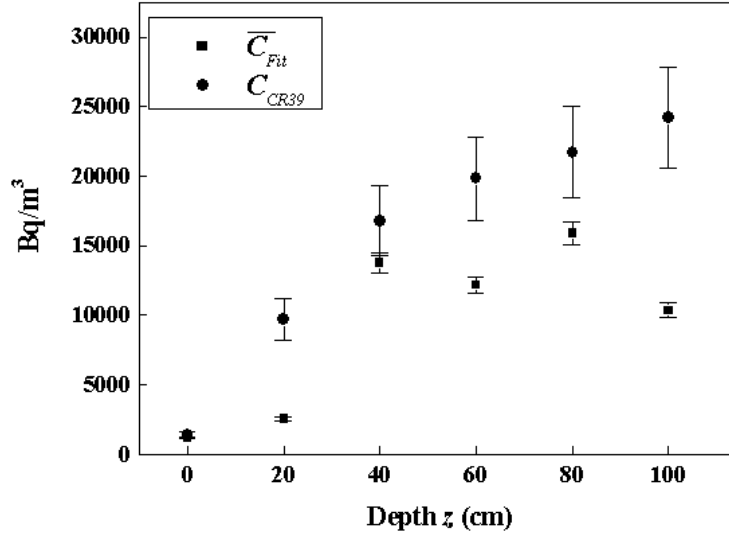


FIG. 8. $\overline{C}_{Fit}$ (RAD7®) vs. C_{CR39} for all depths.

The percent differences, for each depth, between C_{max} , $\overline{C}$ and $\overline{C}_{Fit}$, shown in Table 2, indicate that the average-time values can represent well the set of readings of RAD7® over the time period of measurement (7 hours).

Conclusion

Measurements using the RAD7® active detector and the CR-39 passive radon diffusion dosimeters show that soil radon concentration levels increase with depth as expected and confirmed by many previous studies [19]. The differences between the two types of detectors are mainly due to their function modes.

Taking the simple average of a set of readings of RAD7® over a short period of time gives the differences ($diff_2$) 16%, 73%, 21%, 38%, 29% and 57% for the depths 0, 20, 40, 60, 80 and 100 cm, respectively.

A standard non-linear least squares fit using a cosine function ($A \cos(\omega t + \delta) + B$), where ω is the angular frequency of the rotation of the earth around the sun, was used in order to smooth the RAD7® data using the expected periodic behavior of soil radon concentration with time. The time-average of this function is taken as being the “reading” of RAD7® for a given depth. Comparing this time-average to its corresponding measurement by the passive CR-39 dosimeters gives acceptable results.

The differences in this case ($diff_3$) are 16%, 74%, 18%, 39%, 27% and 57% for the depths 0, 20, 40, 60, 80 and 100 cm, respectively.

Comparing these differences with those in the previous paragraph indicates that the time-averaged values of the RAD7® readings “inspired” by the cosine behavior can be well considered instead of the simple averages over a short period of time.

Appendix

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

$$\frac{\delta T}{\delta t} = a \nabla^2 T \quad (A1)$$

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

$$T(t) = T_0 \cos(\omega t + \delta) \text{ at } z = 0. \quad (A2)$$

a is the Fourier coefficient, which is related to the physical properties of the diffusing medium as follows. For a medium of density ρ (g cm^{-3}) and specific heat capacity at constant pressure c_p ($\text{J g}^{-1} \text{K}^{-1}$) and thermal conductivity (k (usual units: $\text{W m}^{-1} \text{K}^{-1}$), we have:

$$a = \frac{k}{(\rho c_p)}. \quad (A3)$$

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 ($\omega = 7.27 \times 10^{-5}$ rad/s = 0.262 rad/hour) and δ is a phase constant.

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:

$$T(0, t) = T_{av} + T_0 \cos(\omega t) \quad (A4)$$

$$T(\infty, t) = T_{av} \quad (A5)$$

where T_{av} is the average temperature of the soil-air interface (around 18 °C in this study). This solution has the form - see details in [18]:

$$T(z, t) = T_{av} + T_0 e^{-Fz} \cos\left(\omega\left(t - \frac{z}{v}\right)\right), \quad (A6)$$

where F is the inverse of the damping width which is the width at which the amplitude of the wave is reduced by a factor $1/e$ and v is the diffusion velocity.

Taking a , the Fourier coefficient for soil ($0.018 \text{ cm}^2 \text{ s}^{-1}$), $F = \sqrt{\omega/(2a)} = 0.0449 \text{ cm}^{-1}$, $v = \sqrt{2a\omega} = 0.0016 \text{ cm s}^{-1} = 5.827 \text{ cm h}^{-1}$.

The periodic function used for the fit of the RAD7® data is:

$$C_{fit} = A \cos(\omega t + \delta) + B. \quad (A7)$$

See text for more details on the fit parameters.

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