Determination of radon exhalation rates from soil around buildings in Lagos environments using passive measurement technique

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Abstract

Purpose In this study, measurements of radon concentrations and estimation of exhalation rates were carried out in soil around buildings within Lagos State in order to determine the contribution to indoor radon concentrations from the soil, and determine the influence of soil moisture on the exhalation rates.

Methods Fifty-four samples were collected randomly with 27 measured as wet samples and 27 dried before measurements so as to account for the moisture content. Passive measurement method, using cover cup technique with solid state nuclear track detectors, CR-39, was employed.

Results The results showed weak correlations between the concentrations of radon emanated from the soil samples and the indoor radon concentrations. The results obtained suggested lower concentrations of radon emanated from wet soil than dry soil indicating the influence of moisture. The results further indicate that the highest and lowest values as well as the highest mean for both wet and dry soil samples were obtained from the same environment, suggesting that the soil in that environment are of anomalous petrophysical property.

Conclusion Concentrations of radon emanated from dry soil are higher than in wet soil, suggesting that the presence of moisture may results in reduction of radon concentrations in soil samples. The result of the surface exhalation rates and the mass exhalation rates are in congruent with results obtained for the concentrations of radon emanated from wet and dry soil samples.

Keywords Radon exhalation · Passive measurement · Soil · Petrophysical property

Introduction

Radon is found everywhere in varying amounts in the earth's crust [1]. Hence the major contributor to indoor radon concentrations is soil [2]. The phenomenon of how soil gas near buildings moves in through cracks and holes in the foundation provides some explanation for the observed variations at different floors of a building. Some investigations on radon concentrations in soil samples have recorded elevated or near

recommended limit [3–6]. The world average has however been estimated to be 30 Bqm^{-3} [7].

The factors that influence high levels of indoor radon in some houses relative to nearby houses are primarily due to the geology of radon which includes the factors governing the emanation and transport of radon [6]. The efficiency at which radon is released by soil grains is generally expressed as radon emanation coefficient; a measure of the fraction of radon released to gas phase. This fraction, also called emanating power, is lower in soils with very low water saturation percentage and soils that are young relative to the half-lives of ^{226}Ra (1600 years) [8]. Contrariwise, in some old soils, equilibrium may have been established between the deposition and decay of ^{226}Ra , thus maximizing the radon source at the surfaces of soil pores [9]. Most of the emanated radon originates from radium found in shallow surface layers, up to a depth of the recoil range [10]. However, due to diffusion, these flows are notably reduced in wetter soils [11]. The rate at which the emanated radon escapes into the atmosphere is measured as the exhalation rates.

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Radon moving through soil pores and rock fractures near the surface of the earth often migrates toward the foundation of a building because of the differences in air pressure between the soil and the building, the presence of openings in the building's foundation, and increases in permeability around the basement [12]. For instance when constructing a house with a basement, hole is dug, footings are set, and coarse gravel is normally laid down as base for the basement slab. Once basement walls have been built, the gap between them and the ground outside is filled with material that often is more permeable than the original ground. This gap is called disturbed zone. Radon moves from surrounding soil into disturbed zone and gravel bed. The backfill material in disturbed zone is usually made up of rocks and soil from the site. These also generate radon. The amount of radon in the disturbed zone and gravel bed depends on the amount of uranium present in the rock at the site, the type and permeability of soil surrounding the disturbed zone and underneath the gravel bed, and the soil's moisture content. The air pressure in the ground is often greater than that inside the building. Thus, air tends to move from the disturbed zone and gravel bed into the building through openings in the foundation and other routes of entries [1].

If the presence of a building causes the soil below the building slab and subslab aggregate to drain, the ability of radon to move upward the subslab aggregate will increase. In layered soils and those containing platy minerals such as clays, lateral movement of soil gas is favored and vertical movement of soil gas is inhibited. Where a low-permeability layer of soil is present above layers of higher permeability, permeability inversion exists. A building foundation can then offer a path of less resistance to atmospheric pressure changes than the natural soil, forcing radon-bearing soil gas into the backfill, subslab aggregate, and foundation. The subslab aggregate and the backfill, if it is permeable, allow under-pressure in a building to draw radon-bearing soil gas through routes of entry.

Passive measurement techniques are required in determining the long term average concentrations of radon which can be instituted as the baseline data. This technique has been found an effective and reasonable alternative in determining concentrations of radon and its decay products in soil samples [13–15]. The objectives of this study were to determine the concentrations of radon migrating from soil around buildings within Lagos State using passive measurement technique, determine the contribution to indoor radon concentrations from the soil, determine radon exhalation rates from soil and the influence of soil moisture on the exhalation rates, thereby establishing baseline data for the study area.

Materials and methods

The sampling locations in this study are within the coastal areas. Soil samples were collected from buildings whose

external surroundings were not treated with any coverings. The soil samples were randomly collected in replicate to make a total of fifty four (54) samples. The sample collection was carried out in each sampling location at spots very close i.e. within 2 m to the building. In order to ensure adequate protection in a residential setting, the surface soil depth (SSD) ranging from 5 to 10 cm was taken into consideration. Pebbles, stones, sticks and other macro-objects were removed from the samples after which they were packed into polyethylene terephthalate (PET) container of desired geometry, which has been reported to have low diffusion coefficient, no/ low affinity for radon, and affords the required source-to-detector distance [16–20]. Prior to collection of samples, the containers were washed with dilute HNO_3 and thoroughly rinsed with distilled water so as to decontaminate them. A digital weighing balance (ACB plus 1000, AE310F00439) was used to measure the mass of the samples. Each sample, properly packed into the PET containers, were tightly sealed, and recorded in the sampling register and then transferred to the Central Research Laboratory (CRL), Bells University of Technology, Ota (BELLSTECH).

Measurement commenced in 27 samples immediately after collection. In order to investigate the effect of moisture content on radon emanation from the soil, the other 27 samples collected from the same sampling location were weighed, dried in an oven at approximately 110 °C and weighed again until constant weight was obtained [3]. The soil moisture content was noted as the difference in weight of the initial moist soil sample and the dried sample (Table 1). The samples were thereafter crushed, homogenized using 2 mm-sieve and packed into the PET container and treated as the first set of samples. Covered cup technique was employed for all measurements with the detector fastened to the base of the PET's cap and kept at an approximate distance of 27 cm from the sample in order to avoid the contribution from thoron to the measurement (Fig. 1). The PETs used for measurements of both (wet and dry soil) samples were cylindrical containers with height of 30 cm and diameter of 7.5 cm. The detectors were exposed for 6 months, during which more than 99% of the equilibrium level was reached between ^{226}Ra and ^{222}Rn and progenies [20]. The indoor radon concentrations in each sampling location were simultaneously measured with the detectors mounted at the breathing height, an approximate distance of 2 m from the floor. The buildings assessed were bungalow type and measurements were taken within the bedroom for a period of 6 months so as to obtain the long term average. The rooms with average dimension of $4 \times 3.5 \times 3$ m had two windows of average size of 1.2×1.2 m and one door, and were observed to be of good ventilation. After exposure, the detectors were kept in air-tight aluminum foil prior to etching. The detectors were etched with 6.25 M solution of NaOH for 3 h at 70 °C, rinsed, dried, and the tracks were counted.

Table 1 Moisture contents of soil samples

Site	Sample	Weight (g)			MC (%)
		BD $\pm$ 0.005	AD (DW) $\pm$ 0.005	MC	
BADAGRY	1B	951.31	829.34	121.97	13
	3B	913.09	802.28	110.81	12
	8B	979.17	835.69	143.48	15
	9B	994.22	903.5	90.72	9
	10B	916.49	762.5	153.99	17
	12B	910.28	795.41	114.87	13
	16B	939.98	792.07	147.91	16
	21B	858.6	742.55	116.05	14
EPE	1E	709.97	625.1	84.87	12
	3E	963.12	860.1	103.02	11
	5E	734.7	646.97	87.73	12
	6E	787	734.14	52.86	7
	7E	855.94	742.63	113.31	13
	8E	951.83	837.76	114.07	12
	10E	982.31	802.24	180.07	18
	15E	941.2	809.75	131.45	14
	17E	942.6	838.24	104.36	11
	20E	966.97	852.08	114.89	12
IKORODU	1 K	662.62	596.44	66.18	10
	2 K	652.38	582.03	70.35	11
	4 K	680.06	630.18	49.88	7
	6 K	605.64	542.76	62.88	10
	11 K	595.24	538.08	57.16	10
	15 K	523.9	450.41	73.49	14
	17 K	874.97	787.08	87.89	10
	19 K	685.15	611.73	73.42	11
	21 K	545.09	507.51	37.58	7
	28 K	612.16	495.13	117.03	19

BD – BEFORE DRYING, **AD** – AFTER DRYING, **MC** – MOISTURE CONTENT, **NA** – NOT AVAILABLE.

Determination of radon concentrations

Radon concentration, C_{Rn} (Bqm^{-3}) were computed using equation 1, where CF ($\text{kBqh m}^{-3}/\text{track.mm}^{-2}$) represents the calibration factor, T (h) is the exposure time and T_D (track.mm^{-2}) is the track density [20]:

$$C_{Rn} = \frac{T_D(\text{track.mm}^{-2}) \times CF(\text{Bqhm}^{-3}/\text{track.mm}^{-2})}{T(h)} \quad (1)$$

Effective time of measurement T_{eff} was determined using the relation in equation 2 [20, 21] and was employed for the soil samples instead of time of exposure, T . This was required because measurements commenced before equilibrium was reached between ^{226}Ra and ^{222}Rn , resulting in the exposure of detectors to

variable levels of radon concentrations (i.e. from zero concentrations to equilibrium concentration level).

$$T_{eff} = T - \tau(1 - e^{-\lambda T}) \quad (2)$$

where τ = mean life of radon (5.5 days or 132 h), λ = decay constant of radon ($7.55 \times 10^{-3} \text{ h}^{-1}$).

The track density, T_D , is the ratio of the average number of tracks to the area of (field of view) FOV which was determined using equation 3:

$$T_D = \frac{\sum_i N_i}{nA} \quad (3)$$

where A is the area of FOV, N_i is the total number of tracks per detector and n is the total number of FOVs.

Fig. 1 The covered cup technique showing detector-to-sample distance

The associated statistical error was then determined using equation 4:

$$\sigma_{T_D} = \frac{\sqrt{\sum N_i}}{nA} \quad (4)$$

Exhalation rates in soil samples

The surface exhalation rate (E_s) in soil samples from this study was determined in unit of $\text{Bqm}^{-2} \text{h}^{-1}$ using equation 5 [11]:

$$E_s = \frac{V \times C_{Rn} \times \lambda}{A_s [T + \lambda^{-1}(e^{-\lambda T} - 1)]} \quad (5)$$

The mass exhalation rate (E_m) in units of $\text{Bqhk} \text{g}^{-1}$ on the other hand is given by equation 6 [4]:

$$E_m = \frac{V \times C_{Rn} \times \lambda}{M_s [T + \lambda^{-1}(e^{-\lambda T} - 1)]} \quad (6)$$

Where: V (m^3) is the effective volume of the measuring container, M_s (kg) is the mass of the sample and T (h) is the period of exposure. A_s (m^2) the surface area of the sample was determined with

$$A_s = 2\pi r^2 + 2\pi rh \quad (7)$$

Results and discussion

The results obtained are recorded in Table 2 which shows the concentrations of radon emanated from the

wet and dry soil samples, and the corresponding indoor environment. The concentrations of radon emanated from the wet soil samples range from 2.45–12.80, 6.36–8.90 and 5.18–6.26 Bqm^{-3} with mean of 8.38 ± 0.09 , 7.18 ± 0.09 and 5.66 ± 0.08 Bqm^{-3} respectively for Badagry, Epe and Ikorodu. However its concentrations in the dry soil samples vary from 5.27–11.26, 5.99–11.17 and 5.36–10.53 Bqm^{-3} with corresponding mean of 8.54 ± 0.10 , 7.93 ± 0.09 and 6.98 ± 0.09 Bqm^{-3} respectively for Badagry, Epe and Ikorodu. These suggest lower concentrations of radon emanated from wet soil than dry soil indicating the influence of moisture. The results further indicate that the highest and lowest values as well as the highest mean was obtained from Badagry from both wet and dry soil samples, suggesting that the soil in Badagry are of anomalous petrophysical property. Hence further study on the soil in this environment is suggested. However the plots of the concentrations of radon emanated from all soil samples and the indoor radon concentrations from these environments, shown in Fig. 2 indicate a weak negative correlation with correlation coefficients of -0.3020 and -0.3198 between radon concentrations from the indoor environment and its emanated concentrations from the wet and dry soil samples respectively. These suggest that indoor radon concentrations has tendency to increase with decreasing concentrations of radon emanated from soil samples; whether wet (moist) or dry soil. This relationship may not represent causation between the indoor radon concentrations and the concentrations of radon emanated from the soil samples, but it does describe an existing pattern for the study area. Moreover, studies have reported strong correlations between radon concentrations in the indoor air and soil around the building, values of 0.66 [22] and 0.68 [23].

Effect of moisture on radon exhalation rates from soil samples

The radon exhalation rates (surface and mass) for the soil samples (wet and dry) and the percentage moisture content are recorded in Table 2. The surface exhalation rates in wet samples range from 0.51–2.65 $\text{mBqm}^{-2} \text{h}^{-1}$, 1.32–1.84 $\text{mBqm}^{-2} \text{h}^{-1}$ and 1.07–1.26 $\text{mBqm}^{-2} \text{h}^{-1}$, with corresponding mean of 1.74 ± 0.68 , 1.49 ± 0.09 and 1.17 ± 0.06 $\text{mBqm}^{-2} \text{h}^{-1}$ respectively for Badagry, Epe and Ikorodu environments. The surface exhalation rates range for dry samples from 1.09–2.33 $\text{mBqm}^{-2} \text{h}^{-1}$, 1.32–2.31 $\text{mBqm}^{-2} \text{h}^{-1}$ and 1.11–2.18 $\text{mBqm}^{-2} \text{h}^{-1}$, with corresponding mean of 1.77 ± 0.41 , 1.64 ± 0.38 and 1.45 ± 0.29 $\text{mBqm}^{-2} \text{h}^{-1}$ respectively for Badagry, Epe and Ikorodu. The results show that the highest and lowest surface exhalation rates in the wet and dry soil samples occurred in Badagry. Highest mean surface exhalation rate value

Table 2 Radon concentrations (Bqm^{-3}) in soil samples, surface (E_s in $\text{mBqkg}^{-1} \text{h}^{-1}$) and mass (E_m in $\text{mBqkg}^{-1} \text{h}^{-1}$) exhalation rates

Location	Wet Soil (Bqm^{-3})	Dry Soil (Bqm^{-3})	Indoor Value (Bqm^{-3})	% Moisture content	Radon exhalation			
					Wet soil		Dry soil	
					E_s	E_m	E_s	E_m
10B	NA	5.27 ± 0.08	5.90 ± 0.08	NA	NA	NA	1.09	0.20
21B	9.71 ± 0.10	NA	NA	NA	2.01	0.36	NA	NA
9B	12.80 ± 0.12	11.26 ± 0.11	5.90 ± 0.08	9	2.65	0.47	2.33	0.42
1B	12.53 ± 0.11	10.71 ± 0.11	4.72 ± 0.07	13	2.60	0.47	2.22	0.40
12B	2.45 ± 0.05	8.90 ± 0.10	6.02 ± 0.08	13	0.51	0.09	1.84	0.33
8B	6.45 ± 0.08	9.35 ± 0.10	4.90 ± 0.07	15	1.34	0.24	1.94	0.35
16B	6.36 ± 0.08	5.72 ± 0.08	9.90 ± 0.10	16	1.32	0.24	1.19	0.21
MEAN	8.38 ± 0.09	8.54 ± 0.10	6.22 ± 0.08	13	1.74 ± 0.68	0.31 ± 0.12	1.77 ± 0.42	0.32 ± 0.08
6E	NA	6.63 ± 0.09	6.97 ± 0.09	NA	NA	NA	1.37	0.25
10E	7.08 ± 0.09	NA	7.85 ± 0.09	NA	1.47	0.26	NA	NA
17E	6.81 ± 0.08	NA	10.53 ± 0.11	NA	1.41	0.25	NA	NA
20E	7.08 ± 0.09	NA	NA	NA	1.47	0.26	NA	NA
3E	7.26 ± 0.09	7.54 ± 0.09	6.08 ± 0.08	11	1.51	0.27	1.56	0.28
5E	8.90 ± 0.10	11.17 ± 0.11	6.67 ± 0.09	12	1.84	0.33	2.31	0.41
1E	7.17 ± 0.09	6.63 ± 0.09	6.08 ± 0.08	12	1.49	0.27	1.37	0.25
8E	6.36 ± 0.08	11.17 ± 0.11	7.51 ± 0.09	12	1.32	0.24	2.31	0.41
7E	6.54 ± 0.08	5.99 ± 0.08	7.26 ± 0.09	13	1.35	0.24	1.24	0.22
15E	7.45 ± 0.09	6.36 ± 0.08	8.17 ± 0.09	14	1.54	0.28	1.32	0.24
MEAN	7.18 ± 0.09	7.93 ± 0.09	7.46 ± 0.09	12	1.49 ± 0.09	0.27 ± 0.02	1.64 ± 0.38	0.29 ± 0.07
1 K	6.08 ± 0.08	NA	5.27 ± 0.08	NA	1.26	0.23	NA	NA
21 K	5.90 ± 0.08	NA	9.81 ± 0.10	NA	1.22	0.22	NA	NA
28 K	5.27 ± 0.08	NA	NA	NA	1.09	0.20	NA	NA
4 K	5.18 ± 0.08	6.36 ± 0.08	6.17 ± 0.08	7	1.07	0.19	1.32	0.24
11 K	5.54 ± 0.08	10.53 ± 0.11	7.35 ± 0.09	10	1.15	0.21	2.18	0.39
6 K	5.63 ± 0.08	6.17 ± 0.08	6.61 ± 0.08	10	1.17	0.21	1.28	0.23
17 K	6.26 ± 0.08	8.25 ± 0.10	8.99 ± 0.10	10	1.30	0.23	1.73	0.31
2 K	5.18 ± 0.07	5.36 ± 0.08	5.99 ± 0.08	11	1.07	0.19	1.11	0.20
19 K	5.90 ± 0.08	6.81 ± 0.09	9.17 ± 0.10	11	1.22	0.22	1.41	0.25
15 K	5.63 ± 0.08	5.36 ± 0.08	8.35 ± 0.09	14	1.17	0.21	1.11	0.20
MEAN	5.66 ± 0.08	6.98 ± 0.09	7.52 ± 0.09	10	1.17 ± 0.06	0.21 ± 0.01	1.45 ± 0.29	0.26 ± 0.05

was also obtained for both wet and dry soil samples in Badagry. This is in congruent with results obtained for radon concentrations in wet and dry soil samples. The mass exhalation rate somewhat follows the same trend ranging from 0.09 – $0.47 \text{ mBqkg}^{-1} \text{h}^{-1}$, 0.24 – $0.33 \text{ mBqkg}^{-1} \text{h}^{-1}$ and 0.20 – $0.23 \text{ mBqkg}^{-1} \text{h}^{-1}$, with corresponding mean of 0.31 ± 0.12 , 0.27 ± 0.02 and $0.21 \pm 0.01 \text{ mBqkg}^{-1} \text{h}^{-1}$ in wet soil samples for Badagry, Epe and Ikorodu respectively; and varied from 0.20 – 0.42 , 0.22 – 0.41 and 0.2 – $0.39 \text{ mBqkg}^{-1} \text{h}^{-1}$, with corresponding mean of 0.32 ± 0.07 , 0.29 ± 0.07 and $0.26 \pm 0.05 \text{ mBqkg}^{-1} \text{h}^{-1}$ respectively for Badagry, Epe and Ikorodu in dry soil samples. The mean surface and mass exhalation rates are higher in dry soil than wet soil.

The moisture content in the soil samples range from 7 to 19% (Table 1) lying within what is classified as low to moderate moisture content which range from 5 to 25% [3, 24], when it is expected that emanation of radon in the pore spaces of the soil will be enhanced, indicating the influence of soil moisture content on radon emanation coefficients. Contrariwise, the concentrations of radon emanated from the dry soil are higher (in more cases) than in wet soil, suggesting that the presence of moisture within this range may result in reduction of the concentrations of radon emanating from the soil samples. Since there is a direct proportionality between radon emanation and radon exhalation, this range of moisture may result in reduction of the radon exhalation rates.

Fig. 2 Variation in radon concentrations in soil samples and indoor air

Conclusion

Measurement of radon concentrations in soil and the indoor environment were carried out within environments of Lagos Nigeria in order to investigate the variation of radon concentrations in the indoor air with its emanated concentrations from the soil around buildings and determine the effect of moisture on radon exhalation rates from the soil. Samples were collected within 2 m from the building at SSD ranging from 5 to 10 cm. Measurement was carried out on 27 samples beginning from the field while the remaining samples were dried in the laboratory to determine the moisture content before measurement. Covered cup technique was employed for all measurements of radon in soil samples. The moisture content of the soil samples ranged from 7 to 19% lying between the range (5–25%) classified as low to moderate moisture content for which it is expected that the moisture will enhance emanation of radon from soil. Contrariwise, the present study suggest that the presence of moisture within this range may result in reduction of the concentrations of radon emanating from the soil samples, as the concentrations of radon emanated from the dry soil are higher (in more cases) than in wet soil. The mean surface and mass exhalation rates are higher in dry soil than wet soil, which also suggest that the moisture content within the range observed in this study may reduce the exhalation rates.

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Compliance with ethical standards

This study does not involve human subject

Conflict of interest None.

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