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Measurement and Calculation of Radon Releases from NORM Residues

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FOREWORD

The mining and milling of uranium ore produce large quantities of residues containing natural decay series radionuclides. Although such residues are relatively small in magnitude compared with those from metal mining and extraction processes, their present worldwide production exceeds several million tonnes annually. In addition, a number of other resource extraction industries (such as phosphate, mineral sands, oil and gas, and coal) produce residues with similar radionuclide profiles. In recent years, these materials have become increasingly interesting from the point of view of radiological impact assessment. There is thus a need to reduce the environmental and health risks from these materials to an acceptable level.

In response to the needs of its Member States, the IAEA has for many years supported efforts to publish information on the environmental behaviour of radionuclides. In 1992, the IAEA published Technical Reports Series No. 333 (TRS 333), Measurement and Calculation of Radon Releases from Uranium Mill Tailings, which reviewed the major aspects of radon release, control and monitoring as they relate to the management of uranium mill tailings.

Since the publication of TRS 333, a number of developments have taken place in techniques for the measurement, modelling and prediction of radon exhalation, and practical experience has been gained with various residue repositories. It was therefore considered timely to produce a revision of TRS 333 and to expand its scope to include other NORM residues. The present report provides a comprehensive overview of the prediction, measurement and monitoring of radon releases from NORM residues, including uranium mining and milling residues.

The IAEA wishes to express its gratitude to P. Martin (Australia) for his assistance in chairing the meetings, and to K. Lange (Canada) for her assistance in editing this report, as well as to those experts who contributed to the development and completion of this report.

The IAEA officer responsible for this report was M. Phaneuf of the IAEA Environment Laboratories.

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CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

Residues from the mining and milling of uranium ores as well as from a number of other resource extraction industries (for example, phosphogypsum from the phosphate industry and residues from mineral sands processing and from the oil, gas and coal industries) contain uranium or thorium series radionuclides or both [1] and are therefore naturally occurring radioactive materials (NORM). One important pathway for radiological impact arising from NORM residues is the release of radon isotopes into the atmosphere. As a result, the prediction of such releases, including the effect of various covers, is an important element in rehabilitation planning. In addition, measurement of releases from existing residue repositories is often necessary to provide an input parameter to models of radon dispersion in the atmosphere for final assessment of radiological impact and for the investigation of any required ameliorative actions.

In response to the need of its Member States, in 1992 the IAEA published Technical Reports Series No. 333 (TRS 333), Measurement and Calculation of Radon Releases from Uranium Mill Tailings [2]. TRS 333 reviewed the major aspects of radon release, control and monitoring as they relate to the management of uranium mill tailings.

Since the publication of TRS 333, a number of developments have taken place in techniques for the measurement and modelling of radon exhalation, and practical experience has been gained with various types of residue repository. Additionally, the IAEA has organized a series of projects from 1992 to the present aimed at improving environmental assessment and remediation. Through these projects, the environmental behaviour of radionuclides has been considered in many other IAEA publications, mainly in the context of contaminated site characterization and environmental remediation. These publications include: the characterization of contaminated sites [3], technical and non-technical factors relevant for the selection of the preferred remediation strategy and technology [4–6], an overview of applicable technologies for environmental remediation [7–9], options for the cleanup of contaminated groundwater [10] and planning and management issues [11]. Other IAEA publications on related aspects include reports on the remediation of uranium mill tailings [12] and of dispersed contamination [13, 14], on the decontamination of buildings and roads, on the characterization of decommissioned sites and on radiation protection and the management of radioactive waste in the oil and gas industry [15]. In view of these new publications as well as of new findings regarding radon, it was deemed necessary to produce a revision of TRS 333.

This report covers radon releases from repositories of NORM residues. For simplicity, in this report the term 'residue' includes tailings from processing, residues from heap leaching, waste rock, sludges, filter cakes and scales. Specific examples include uranium mill tailings, phosphogypsum, coal fly ash, and thorium refinery residues.

Such residues may be present in the environment in repositories in various physical forms, such as tailings ponds, dumps or stacks. Rehabilitated waste is typically covered with natural and engineered materials such as layers of clay, soil, water, rock, geosynthetics, vegetation, a combination of any of these or it may be exposed to the atmosphere.

The factors affecting radon releases from residue repositories will vary considerably with the residue type, the repository characteristics and the radon isotope of interest. This report describes approaches to the measurement, calculation and monitoring of radon releases that can be applied to many situations, together with specific examples of important representative scenarios.

There are three radon (Rn) isotopes naturally present in the environment: ^{222}Rn, a member of the uranium decay series, ^{220}Rn, a member of the thorium decay series, and ^{219}Rn, a member of the actinium decay series. ^{220}Rn and ^{219}Rn are often referred to as thoron and actinon, respectively. In the case of actinon, its short half-life (3.98 s) and its very low activity concentration relative to ^{222}Rn in the environment mean that for all practical situations its radiological impact is negligible compared with that of ^{222}Rn. The impact of ^{220}Rn is minimal, as its half-life is 55.6 s and it has a higher activity concentration than ^{219}Rn but still much lower relative to ^{222}Rn. Consequently, the primary focus of this report is ^{222}Rn, and to some extent ^{220}Rn, wherever applicable.

1.2. OBJECTIVE AND SCOPE

This report covers the prediction, measurement and monitoring of radon releases from NORM residues, including uranium mining and milling residues. The intention is to support environmental investigations and assessments, as well as remediation in areas contaminated by NORM, by presenting state of the art concepts, models and parameters.

The primary focus of this report is the release of ^{222}Rn, although ^{220}Rn and ^{219}Rn are also discussed, and the material herein discussed can also be applied to those isotopes, with modifications. The report covers methods of prediction, measurement and monitoring of radon emanation from grains into interstitial spaces, transport within the residue and soil profile and exhalation into the atmosphere from the surface. The related topics of emanation and exhalation from non-residue materials, the behaviour of radon in the atmosphere following exhalation, implications for dose assessment and applications of radon as a tracer of environmental processes are all outside the scope of this report.

This report is intended to be used in conjunction with other IAEA publications related to the assessment of the radiological impact of radioactive waste, such as Safety Reports Series No. 19, Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment [16], IAEA Safety Standards Series No. WS-G-2.3, Regulatory Control of Radioactive Discharges to the Environment [17], and other related publications.

1.3. STRUCTURE

This report is comprised of nine sections. Section 2 concerns factors controlling radon releases from residue materials. Sections 3 and 4 describe the measurement and estimation of variables affecting the radon exhalation rate. Section 5 considers measurement methods for radon concentrations in soil gas and for radon exhalation flux density from a surface in detail, including instrumentation and its calibration. Section 6 provides methods for predicting radon exhalation flux density. Section 7 addresses waste repository cover characteristics and meteorological conditions. Section 8 briefly discusses aspects of radon monitoring programmes including data collection, factors in design and implementation and quality assurance. Section 9 outlines a case study in which multiparametric measurements at a uranium waste repository are compared with model predictions.

The report also includes two appendices presenting the main mathematical development of the radon diffusion and radon flux equations.

2. FACTORS CONTROLLING RADON RELEASES

2.1. GENERAL

The three naturally occurring radon isotopes ^{222}Rn, ^{220}Rn and ^{219}Rn are formed on the alpha decay of their radium parents ^{226}Ra, ^{224}Ra and ^{223}Ra, respectively (Figs 1–3, and Table 1).

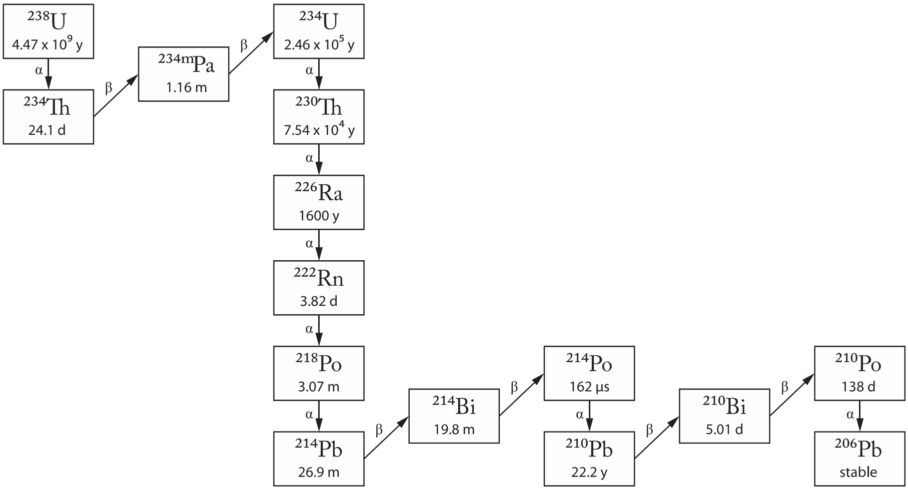

FIG. 1. Uranium decay series.

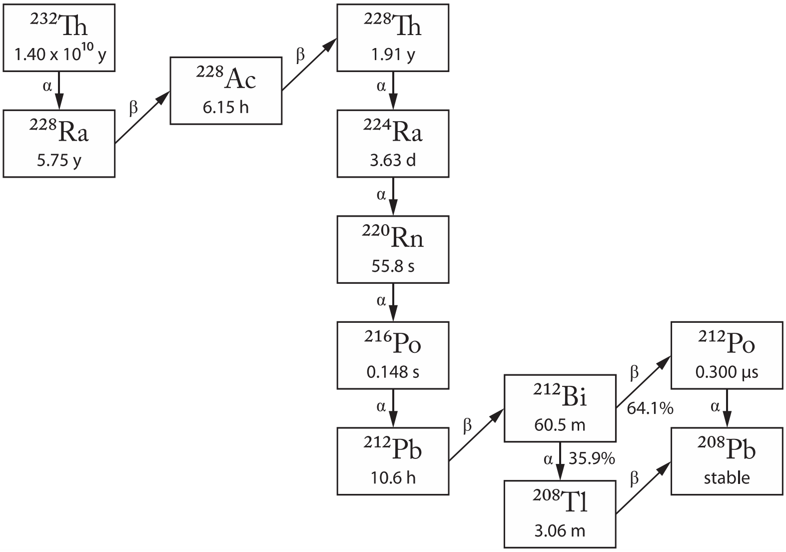

FIG. 2. Thorium decay series.

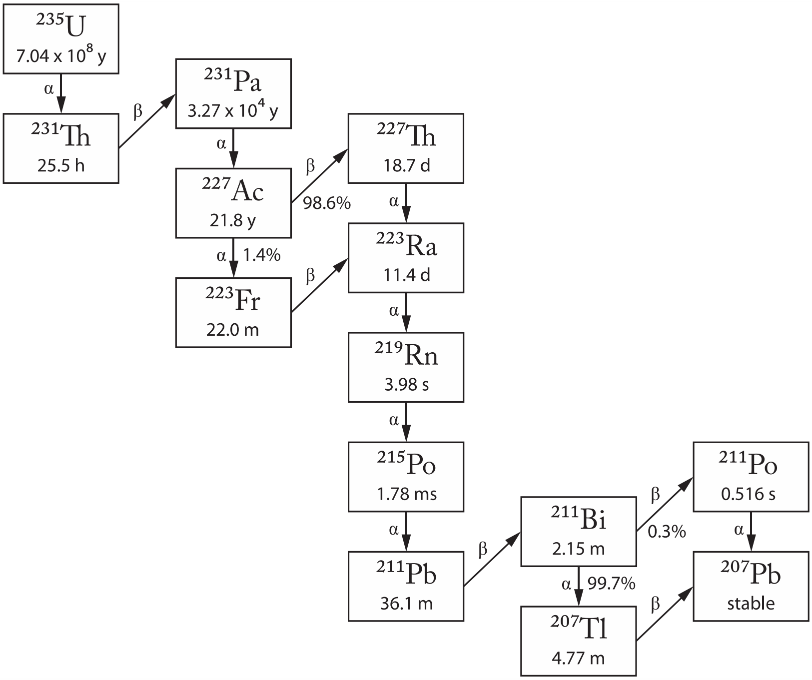

FIG. 3. Actinium decay series.

TABLE 1. RELEVANT PHYSICOCHEMICAL PROPERTIES FOR ^{222}Rn, ^{220}Rn AND ^{219}Rn

Parameter	Symbol	^{222}Rn	^{220}Rn	^{219}Rn
Half-life	$T_{1/2}$	3.8232 (8) d	55.8 (3) s	3.98 (3) s
Decay constant	λ	2.0984×10^{-6}/s	1.242×10^{-2}/s	1.74×10^{-1}/s
Average recoil energy on formation	E_r	86 keV	103 keV	104 keV
Diffusion coefficient in air	D_{MA}	1×10^{-5} m^2/s		
Diffusion coefficient in water	D_{MW}	1×10^{-9} m^2/s		

Note: Decay data are taken from Ref. [18].

Radon atoms located within solid grains are unlikely to become available for release to the atmosphere, owing to their very low diffusion coefficients in solids. However, if they are located in the interstitial space between grains, they may diffuse to the surface. Therefore, releases of radon from a residue repository to the atmosphere take place by the following series of processes [19]:

(a) Emanation — radon atoms formed from the decay of radium escape from the grains (mainly because of recoil) into the interstitial space between the grains.
(b) Transport — diffusion and advective flow cause the movement of the emanated radon atoms through the residue or soil profile to the ground surface.
(c) Exhalation — radon atoms that have been transported to the ground surface and then exhaled to the atmosphere.

These processes are illustrated in Fig. 4.

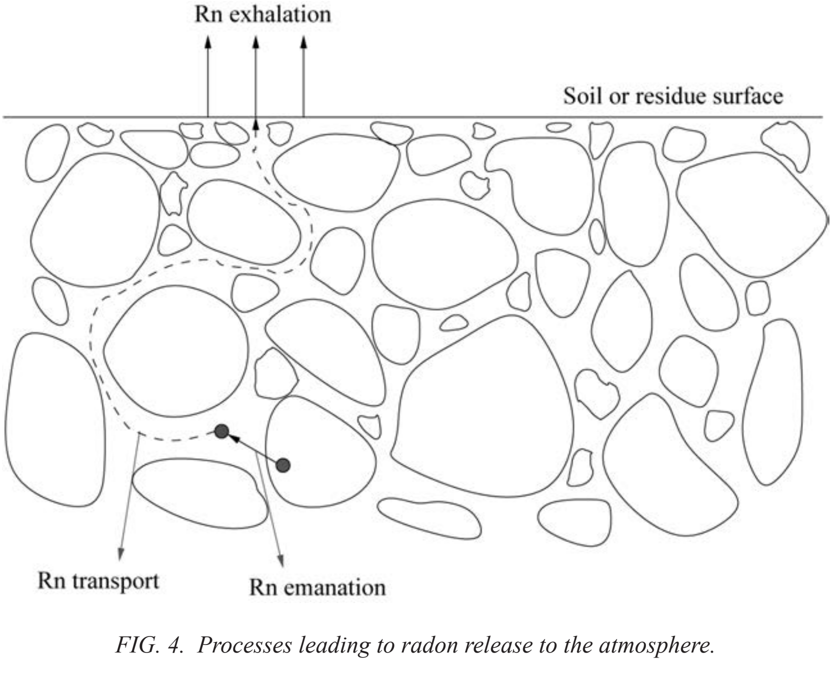

FIG. 4. Processes leading to radon release to the atmosphere.

The amount of radon available for transport to the surface is given by the radon production rate per unit volume of the residue material, P ($\mathrm{Bq \cdot m^{-3} \cdot s^{-1}}$):

$$P = \lambda E R \rho_b \tag{1}$$

where

λ is the decay constant for radon ($\mathrm{s^{-1}}$);
E is the emanation coefficient (dimensionless);
R is the radium activity concentration in the residue (Bq/kg);

and ρ_b is the bulk density ($\mathrm{kg/m^3}$).

This is the source term of radon for a particular residue material. It follows that the source term increases with the emanation coefficient, the radium activity concentration and the bulk density.

Radon in the interstitial space may be transported to the surface by diffusion and advective flow. Advective flow may be significant in some cases owing to surface cracks or holes, gas production, transport in water or the presence of large voids. However, in most cases, the dominant transport mechanism is diffusion, and therefore the transport of radon is commonly referred to as diffusion.

Following emanation and diffusion to the surface, radon may be released from the surface into the atmosphere. This release is referred to as radon exhalation, and is characterized by the exhalation flux density ($\mathrm{Bq \cdot m^{-2} \cdot s^{-1}}$).

2.2. EMANATION COEFFICIENT

The emanation coefficient is defined as the fraction of radon atoms generated that escape the solid phase in which they are formed and become free to migrate through the bulk medium. This term has also been referred to as the emanation fraction or the emanating power.

It is generally accepted that radon escapes from a grain as a result of its recoil when its parent, radium, decays [20]. As the recoil range in solids is small, typically less than 0.05 μm, most of the recoiling atoms remain within the mineral grain lattice. Further movement of the atoms from the lattice does not significantly contribute to radon release because of the very small diffusion coefficient through minerals, 10^{-25}–10^{-27} $\mathrm{m^2/s}$ [21, 22]. If the recoil terminates outside the grain or in an open pore, radon is able to migrate (Table 2 [23] and Fig. 5 [24]).

TABLE 2. ALPHA RECOIL RANGES OF ^{222}Rn AND ^{220}Rn IN VARIOUS ENVIRONMENTS

Medium	Density (g/cm^3)	Projected range (nm)	
		^{222}Rn	^{220}Rn
Air	1.58×10^{-3}	53×10^3	60×10^3
Water	1.00	77	87
Quartz (SiO$_2$)	2.65	34	38

Note: Alpha recoil ranges were calculated using SRIM-2006 [22], assuming 86 and 103 keV of recoil energies for ^{222}Rn and ^{220}Rn, respectively.

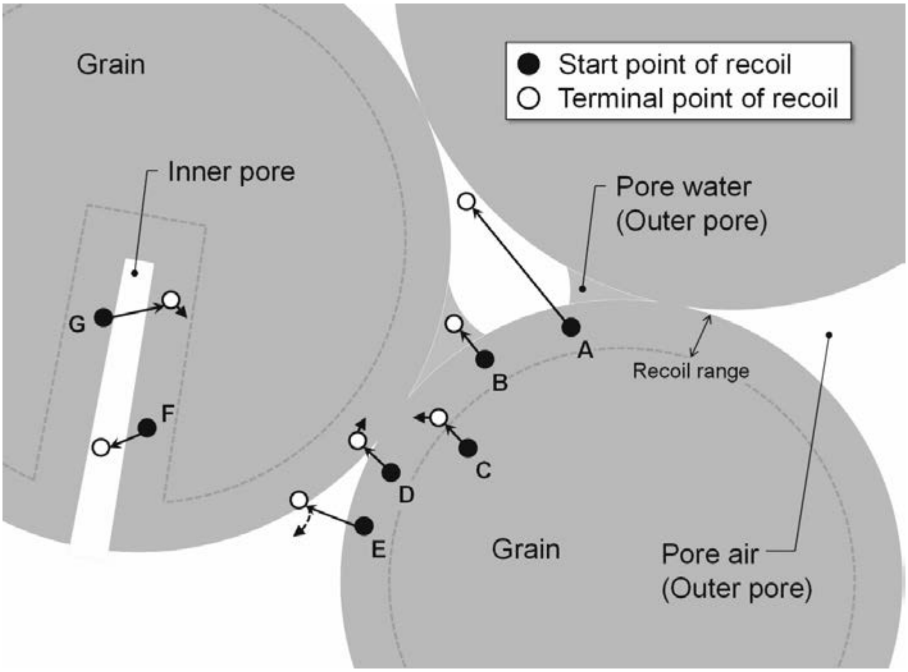

FIG. 5. Schematic diagram of radon emanation [23].

2.2.1. Factors affecting radon emanation

Variations encountered in the emanation coefficients and the complex effects of host rock and uranium mineralogy, uranium ore grade, particle size and moisture content have been described qualitatively and by some models [24–30]. In practice, the emanation coefficient has to be measured for each material being studied. The effects of radium distribution and particle size, and of moisture and mineralogy are briefly described in Sections 2.2.1.1–2.2.1.3.

2.2.1.1. Radium distribution, particle size and shape

The particle size and shape determine in part how much uranium and radium is close enough to the surface of the grain to allow the radon to escape into the interstitial pores. If radium were uniformly distributed throughout a grain, the emanation coefficient would approach an inverse proportion to the particle diameter if this were greater than approximately 0.1 μm. On the other hand, if radium were mainly distributed on the grain surface, the emanation coefficient would be constant regardless of the particle diameter (Fig. 6).

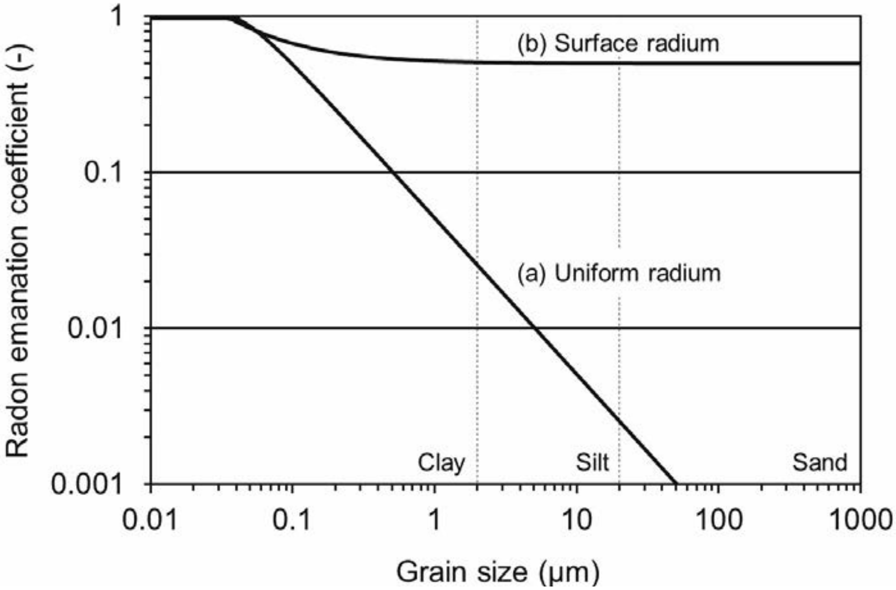

FIG. 6. Calculations of radon emanation coefficients in the single grain model. Radium is assumed to be distributed (a) uniformly in a spherical grain, and (b) on the surface of a spherical grain. This model does not take into account the embedding of radon into a second grain. The range of radon with a recoil energy of 86 keV in a common mineral (quartz, SiO_2) was set to 34 nm, based on the SRIM-2006 calculation [22].

Uniform distribution is often a characteristic of primary minerals [31–33], including some process residues such as fly ash [34], while surface distribution is a characteristic of secondary minerals such as sedimentary deposits and residues such as sand filters [29, 35].

There are more complex theories to explain radon emanation in situations that lie between the extremes presented above (uniform distribution vs. surface only). These theories consider factors such as fractal lattice structure [21] and adjacent particle effects [29, 30].

2.2.1.2. Moisture content

Moisture content is well known to have a strong effect on the emanation coefficient [32, 35–39]. This is because the typical ranges for recoil in water are much less than in air (see Table 2) and therefore water is more effective at stopping radon atoms within the pore space.

Figure 7 shows this effect for one type of uranium mill tailings [37]. In dry residues, the emanation coefficient is relatively low because most radon atoms escaping from particles bury themselves in adjacent particles. As the moisture content increases, the pores contain more water and a recoiling atom is more likely to terminate its recoil in the pore. At higher moisture contents (above 5% by volume), few of the atoms can penetrate into an adjacent particle and the emanation coefficient remains nearly constant with increasing moisture up to saturation. In general, emanation coefficients in saturated residues are 2–6 times higher than in dry residues. The higher value corresponds to residues with smaller particle sizes.

2.2.1.3. Mineralogy

The emanation coefficient of radon depends on a number of mineralogical aspects such as lattice structure, porosity, grain shape, and elemental composition. NORM residues are generally the product of a processing and milling process, which can have a significant effect on the mineralogy of the residue. Knowledge of the mineralogical characteristics of the residue can be important in understanding its emanation behaviour.

The emanation coefficient of soil grains is variable because soil is generally composed of many kinds of mineral grains of different rock origins. Weathering may also influence the characteristics of soils by increasing the specific surface area (area to volume ratio) of the matrix.

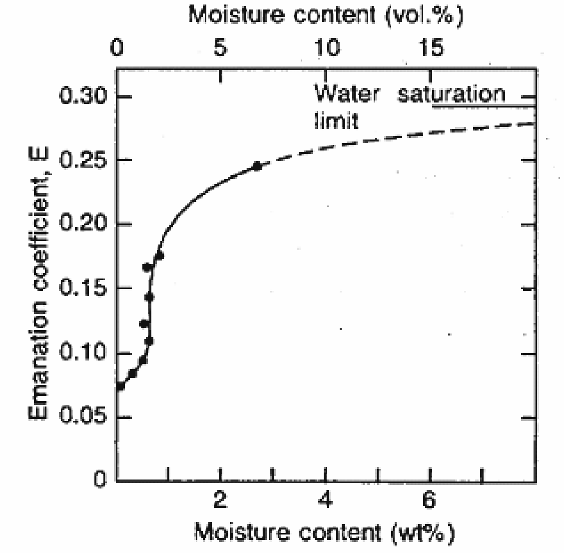

FIG. 7. The effect of moisture content on the ^{222}*Rn emanation coefficient for a sample of uranium mill tailings [37].*

In the specific case of high grade uranium ore, uranium minerals are often present as thick bands or as large grains which are normally associated with low emanation coefficients. On the other hand, these high grade ores have been exposed to much higher radiation doses and this can lead to high emanation coefficients as a result of radiation damage to the crystal lattice [40]. Therefore, uranium ore grade can affect the emanation coefficient of crushed materials and their residues because of these two different and competing effects.

Residues may undergo chemical and physical changes in the repository. Over time, this can result in changes in composition and grain size [41, 42]. As a result, the method of attachment of radium within the matrix can change, which may affect the radon emanation [43, 44].

2.2.2. Radon emanation coefficients for natural materials

A large number of studies have reported radon emanation rates for natural materials including uranium mine and mill tailings, rocks, soils and minerals. As is shown in Fig. 8, radon emanation coefficients vary considerably between and within material groups. In particular, the reported coefficients for fly ash and minerals were low, generally less than 0.1, whereas they were higher for rocks, soils and mill tailings, generally lying in the range of 0.1–0.3.

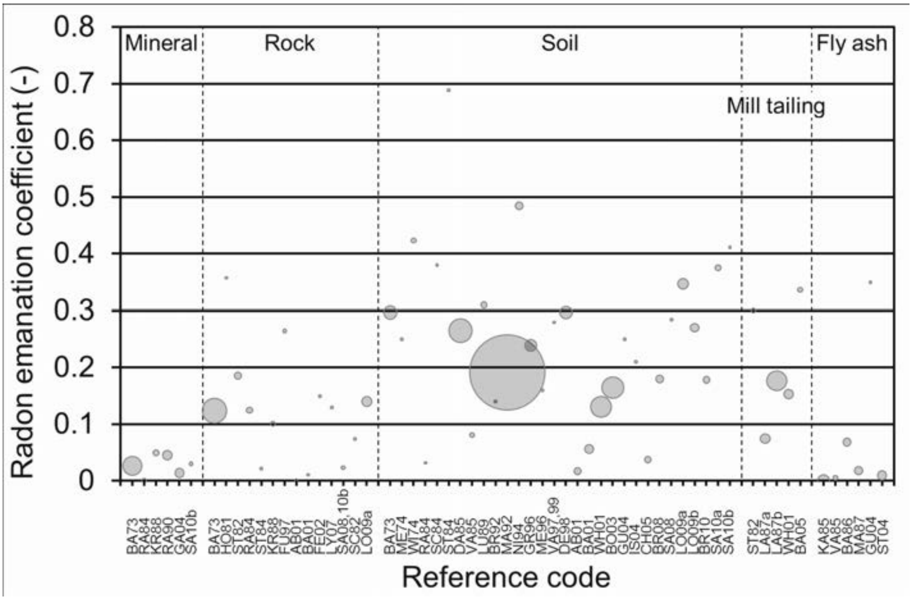

FIG. 8. Radon-222 emanation coefficients measured for various materials. The area of each plot depends on the number of samples [23].

Higher coefficients reported for soil and tailings suggest that radium in these two materials tends to be enriched near the grain surface. Cracks and fissures on the surface of the grain, from previous radioactive decays and chemical or weathering effects, can also increase the surface area of the grain, thus increasing the emanation coefficient.

Emanation coefficient data for ^{220}Rn is much sparser than for ^{222}Rn. Table 3 [45–49] shows a summary of data available in the literature. In general, it can be seen that ^{220}Rn emanation coefficients are lower than, but of a similar order of magnitude to, reported ^{222}Rn emanation coefficients.

2.3. DIFFUSION COEFFICIENT

The molecular diffusion coefficient of radon is defined by Fick's first law, which states that radon flux density is linearly proportional to its concentration gradient:

TABLE 3. RADON-220 EMANATION COEFFICIENT FOR VARIOUS MATERIALS

Material	Sample number	Mean Rn-220 emanation coefficient	Reference
Mineral	2	0.0005 ± 0.0001	[45]
	11	0.011 ± 0.016	[46]
Average	13	0.01 ± 0.02	
Rock	5	0.157 ± 0.086	[45]
	32	0.105 ± 0.145	[47]
Average	37	0.11 ± 0.14	
Soil	12	0.11 ± 0.02	[48]
	16	0.16 ± 0.06	[49]
Average	28	0.14 ± 0.05	

Note: Moisture content may differ among the samples although test temperature was around room temperature.

$$f = -D_M \nabla C \qquad (2)$$

where

f is the radon flux density ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$);
D_M is the molecular diffusion coefficient (m^2/s);

and ∇C is the gradient of radon activity concentration (Bq/m^4).

The negative sign arises from the fact that radon diffuses from high to low concentrations. The molecular diffusion coefficients for radon in air (D_{MA}) and in water (D_{MW}) are approximately 1×10^{-5} m^2/s and 1×10^{-9} m^2/s, respectively (see Table 1), and can be substituted for D_M in Eq. (2).

In a porous medium such as soil, radon moves by diffusion in the pore space between the soil particles. The rate of radon movement or flux through a soil may be slower than by diffusion in a homogeneous medium such as pure air for two main reasons:

(a) Smaller fluid volume limiting flow (porosity, n);
(b) Tortuous flow path around particles (tortuosity, τ).

Taking these factors into account, the bulk flux density of radon through a porous medium is given by:

$$f = -n_s \tau D_M \nabla C \tag{3}$$

where

f is the radon flux density ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$);
n_s is the porosity of the soil;
τ is the tortuosity factor, which equals unity for a pure solution and is typically less than unity in soils (e.g. a typical value for τ is 0.66, as obtained for closely packed uniform spheres);
D_M is the molecular diffusion coefficient (m^2/s);

and ∇C is the gradient of radon activity concentration in the pore space (interstitial) (Bq/m^4).

The terms τ and D_M are often lumped together to define the diffusion coefficient of the pore fluid such that:

$$D = \tau D_M \tag{4}$$

Substituting this in Eq. (3) yields:

$$f = -nD\nabla C \tag{5}$$

The quantity of nD may be represented by the notation D_e and referred to as the effective bulk diffusion coefficient of the soil. In this report, the radon diffusion coefficient D is used rather than D_e, because it is directly related to the diffusion length of radon in the matrix and is therefore the more fundamental parameter. In the simple case of a homogeneous bare residue surface, the flux is solely dependent on D and does not depend on n.

Radon diffusion coefficients in soil may be position dependent and are generally dependent on the soil type, its pore size distribution, its water content, and the degree and method of its compaction [50–52]. Because of the large difference between D_M in air and in water, moisture effects generally dominate over other physical factors. Consequently, a number of researchers have studied the dependency of radon diffusion coefficients on moisture content for different soil types [37, 53]. Such studies commonly demonstrate that the diffusion of radon is seriously hampered when soil moisture exceeds a certain threshold, which depends on the geometry of the soil pore space. Figure 9 presents measured values for the radon diffusion coefficient, D, as a function of moisture content. The results show that:

(a) For moisture saturations of less than 0.25, D is relatively constant, varying between approximately 9×10^{-7} and 7×10^{-6} m^2/s.

(b) As moisture saturation increases, the spread of D increases by approximately 2 orders of magnitude at intermediate saturation and by approximately 4 orders of magnitude at total saturation.

(c) D decreases to values typical of saturated materials at saturations between 0.8 and 1.0.

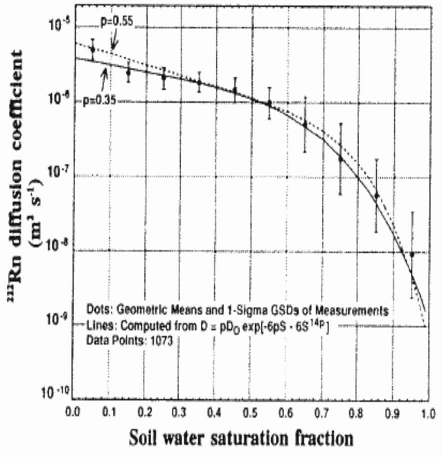

FIG. 9. Comparison of measured radon diffusion coefficients with a simple correlation function.

The effect of moisture saturation on the diffusion coefficient can be explained qualitatively. For dry soils, the diffusion coefficient can be calculated as $D = \tau D_{MA}$. When moisture content is low, radon diffusion is not hindered by the presence of small amounts of water that form thin, discontinuous films around the soil particles, and therefore this equation still applies. At higher moisture contents, free water appears in the interstitial space between particles and radon particles must either take a longer route through the air filled pore space or else diffuse partially through water. In this region, the diffusion coefficient is not uniquely determined by the percentage moisture by weight, but depends on how the water and air phases are spatially distributed. As the porous medium approaches full saturation, the diffusion coefficient approaches the value given by $D = \tau D_{MW}$.

In the intermediate region, the diffusion coefficient is best determined experimentally, although both empirical relationships and mathematical models such as a pore distribution model can be used.

2.4. RADON EXHALATION

The exhalation flux density, denoted by f, describes the release of radon over a specific area ($\mathrm{Bq \cdot m^{-2} \cdot s^{-1}}$). The total release rate of radon, or the total radon exhalation flux, F (Bq/s), for a NORM residue facility is therefore obtained by multiplying the area averaged exhalation flux density by the total surface area of the repository. The main variables directly affecting the exhalation flux densities are:

R	the radium activity concentration in the pile (Bq/kg);
ρ_b	the bulk density of the matrix ($\mathrm{kg/m^3}$);
E	the emanation coefficient (non-dimensional);
D	the diffusion coefficient of radon ($\mathrm{m^2/s}$);

and Z, the thickness of the covering material (m).

Values of these parameters are generally obtained by direct measurements, some of which are described in Section 3. If data for any of the parameters listed above are not available, then the radon exhalation flux density may be estimated by choosing default or approximate values for these parameters.

3. MEASUREMENT OF VARIABLES AFFECTING RADON EXHALATION FLUX

3.1. MEASUREMENT OF THE EMANATION COEFFICIENT

The emanation coefficient can generally be evaluated by two methods: the combination of measurements of radium and radon, and gamma spectrometry under different conditions.

In the former, for example, sample material in a cylindrical container is held in a closed vessel for more than 4 weeks to ensure the establishment of radioactive equilibrium between ^{222}Rn and ^{226}Ra (Fig. 10). After that, the total activity of radon released into the air from the sample material is evaluated by a measurement of radon concentration and the effective volume of the measurement system.

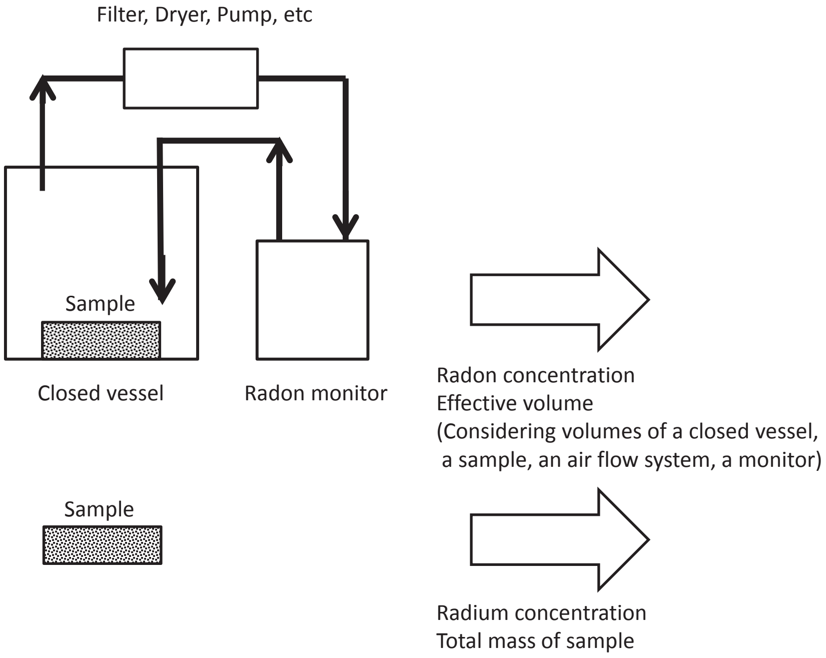

FIG. 10. Schematic diagram of an example process of emanation coefficient measurement.

The effective volume is determined by considering the volume of the closed vessel, of all material inside the closed vessel, of the loop system for radon measurement and of the radon monitor. The total activity of radium in the sample material can be determined by various methods such as alpha spectrometry, gamma spectrometry, liquid scintillation spectrometry and mass spectrometry [54]. The emanation coefficient is calculated by:

$$E = \frac{VC}{MR} \tag{6}$$

where

E is the emanation coefficient;
V is the effective volume of the sampling device (m^3);
C is the radon concentration (Bq/m^3);
M is the total mass of the sample (kg);

and R is the radium activity concentration (Bq/kg).

Another approach used for radon and radium measurements is the flow through method (Fig. 11).

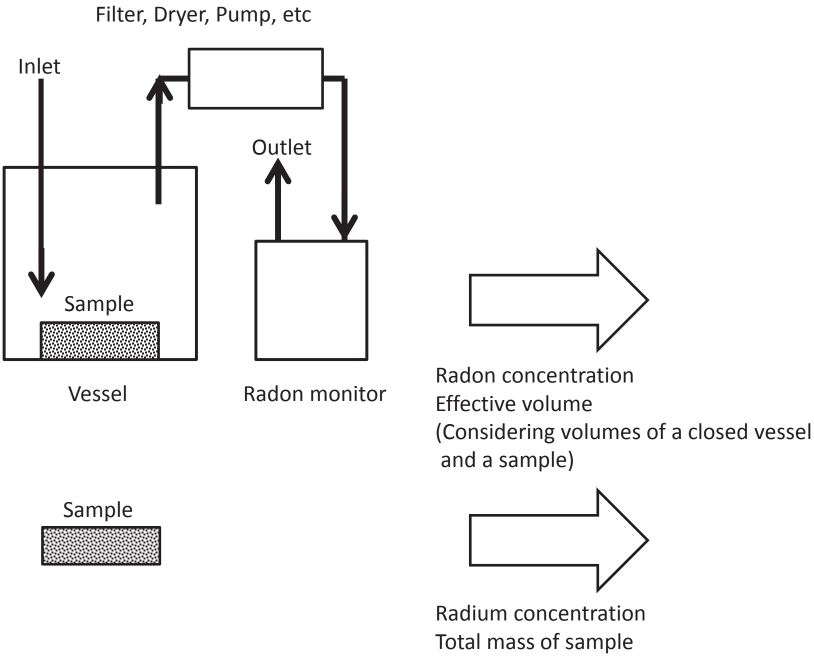

FIG. 11. Schematic diagram of an example process of emanation coefficient measurement by flow through.

For evaluation, the effective volume is determined by considering the volume of a vessel and all material in the closed vessel. In addition, several hours only may be required for radon measurement because the steady state is immediately established by ventilation. Using radon free air or air with a negligible amount of radon as a carrier gas, the emanation coefficient is calculated by:

$$E = \frac{\left(v + \lambda V\right)C}{\lambda M R} \tag{7}$$

where

E is the emanation coefficient;
v is the flow rate (m^3/s);
λ is the decay constant of ^{222}Rn (s^{-1});
V is the effective volume (m^3);

C is the radon concentration (Bq/m^3);
M is the total mass of the sample (kg);

and R is the radium activity concentration (Bq/kg).

In the gamma spectrometry method, the sample is sealed in a container and gamma rays emitted from ^{214}Pb or ^{214}Bi are measured several times before and after the establishment of radioactive equilibrium between ^{226}Ra and its progeny (Fig. 12).

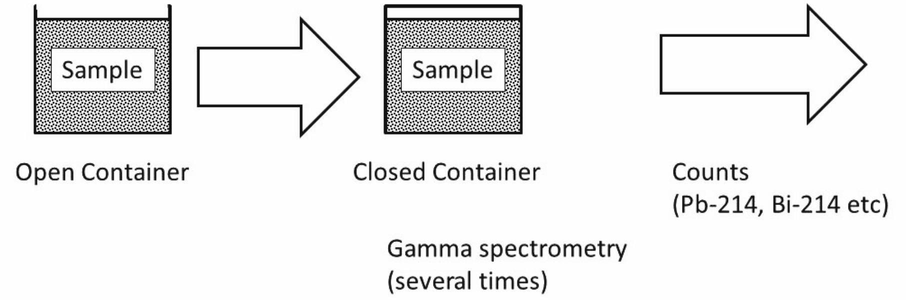

FIG. 12. Schematic diagram of an example process of emanation coefficient measurement.

It is assumed that radon activities are always equal to those of its progeny. Gamma spectrometry is a useful non-destructive method and indirect measurement using progeny nuclides is commonly applied for radium isotopes. In addition, calibration may not be required in this method because only relative counts are needed. The emanation coefficient is calculated by:

$$E = \frac{N_{eq} - N_0}{N_{eq}} \tag{8}$$

where

E is the emanation coefficient;
N_{eq} is the specific counts selected from peaks of ^{214}Pb and ^{214}Bi in equilibrium condition;

and N_0 is the number of counts corresponding to N_{eq} in initial condition.

Analysis of multiple peaks such as 295 keV (^{214}Pb), 352 keV (^{214}Pb), 609 keV (^{214}Bi) and 1120 keV (^{214}Bi) improves precision.

In preparation for measurement, it is necessary to remove radon from the sample pores. In the case of a sample kept in a closed bottle for a long period prior to analysis, the radon progeny attached to the sample grains can also cause imprecision in the first measurement. Thus, it is common to leave such a sample (with a thickness of a few centimetres at the most) open for several hours, or to avoid the use of data measured within several hours after placing and sealing a sample into a container [55, 56].

If the ^{214}Pb or ^{214}Bi counts seem to reach equilibrium within a month, the measurement result should be corrected by the estimation of leakage impact.

3.2. MEASUREMENT OF THE DIFFUSION COEFFICIENT

Several methods are available to measure the radon diffusion coefficient in porous materials such as soil, building materials, NORM residue, etc. [19, 57–62]. Some of the methods are sample based measurements while others are of the in situ type. Since radon emissions are directly governed by in situ diffusion coefficients, the measurement of these coefficients are described below. The most popular method in the in situ category is the soil radon depth profiling method [19, 57, 58]. In this method, radon concentrations at different depths of the residue from the surface are measured by inserting a soil probe equipped with a radon monitor to generate depth profile data of radon concentration in the matrix. Concentration data collected at various depths are least square fitted to the following equation derived from diffusion theory [19]:

$$C(z) = C_\infty \left(1 - e^{-\frac{z}{L_r}} \right) \tag{9}$$

where

$C(z)$ is the radon concentration in pore-air at depth z (Bq/m^3);

C_∞ is the radon concentration at large depth of the residue repository (>2 m) (Bq/m^3);

z is the depth (m);

and L_r is the radon diffusion length (m).

The parameters C_∞ and L_r are obtained as fitting parameters. From the value of L_r obtained from fitting, the diffusion coefficient D_r is calculated as $D_r = \lambda L_r^2$.

Direct measurement of the effective diffusion coefficient (the product of the porosity and the diffusion coefficients) may be important for predicting radon flux, especially if information is available on the radon concentration at a great depth within the NORM residue. It can be directly estimated using the in situ accumulator technique (described in Section 5.3.1) in the context of radon flux measurements. Following the deployment of the accumulator on the residue surface, the radon concentration in the headspace increases approximately as a saturating exponential with time [63]. The time required to attain saturation in the chamber is governed by a time constant (τ_e) of the accumulator–residue system. In contrast to one dimensional diffusion models that do not predict saturation [64, 65], application of a two dimensional diffusion theory predicts saturation and indicates that the time constant is a function of the effective diffusion coefficient of radon in the matrix and the dimension of the accumulator [66, 67]. The relation is given by the following equation:

$$\tau_e = \frac{1}{\lambda + \frac{nD_r}{aH} k \chi_v} \tag{10}$$

where *a* is the chamber radius (m) and *H* is the effective height of the accumulator, i.e. the ratio of total volume (sum of volume of accumulator headspace, internal volume of ^{222}Rn monitor and tubing volume) to base area of the accumulator (m).

In Eq. (10)

$$\chi_v = \frac{\Gamma(v)\Gamma(v+2)}{\Gamma(v+0.5)T(v+1.5)} \tag{11}$$

in which $\Gamma(\nu)$ is the gamma function, and

$$\mathrm{v} = \frac{1}{\pi}\arccos\left[\frac{\mathrm{D_{MA}}}{\mathrm{D_{MA}} + \mathrm{nD_r}}\right] \tag{12}$$

where D_{MA} is the molecular diffusion coefficient of radon in air; and nD_r is the effective diffusion coefficient (D_e) of radon in the residue.

In view of the somewhat complicated nature of the mathematical relationship above, a look-up table is provided (Table 4) to find the effective diffusion coefficient (D_e) from the observed value of the time constant (τ_e) of accumulator set-ups of various volume to radius ($V/\pi a$) ratios. To illustrate the use of the look-up table, consider a chamber of $V/\pi a$ ~150 cm^2 for which the time constant (τ_e) is observed to be ~140 min. The effective diffusion coefficient is found by locating the nearest value of the observed time constant, 140 min, in the column representing the $V/\pi a$ ~150 cm^2 from the look-up table (i.e. 141 min in the column $V/\pi a$ = 150 cm^2), which corresponds to an effective diffusion coefficient of 0.002 cm^2/s. For intermediate values of the parameters, D_e can be obtained by linear interpolation.

TABLE 4. LOOK-UP TABLE FOR FINDING THE EFFECTIVE DIFFUSION COEFFICIENT ($D_e = nD_r$) OF RADON IN THE RESIDUE MATRIX FROM THE OBSERVED TIME CONSTANT (τ_e) OF RADON BUILDUP IN ACCUMULATOR SET-UPS WITH VARIOUS VALUES OF $V/\pi a$ (cm^2)

	$V/\pi a$ (cm^2)								
	50	75	100	125	150	200	250	300	450
D_e (cm^2/s)	Time constant, τ_e (min)								
2.0E-06	1329	1840	2278	2657	2989	3542	3984	4335	5110
6.0E-06	849	1208	1534	1829	2099	2572	2975	3312	4113
1.0E-05	677	974	1248	1502	1736	2159	2527	2842	3618
2.0E-05	495	720	932	1132	1321	1670	1984	2259	2969
4.0E-05	356	522	681	834	980	1256	1511	1740	2354
6.0E-05	292	431	564	693	818	1055	1277	1478	2030
8.0E-05	254	375	493	607	717	929	1129	1311	1819
1.0E-04	228	337	443	547	647	840	1024	1192	1666
2.0E-04	162	240	317	393	467	611	749	878	1250
4.0E-04	112	166	220	273	326	429	529	623	901
6.0E-04	92	137	181	225	269	354	438	518	753
8.0E-04	79	118	157	196	234	309	382	452	660
1.0E-03	71	106	141	175	209	277	343	406	595

TABLE 4. LOOK-UP TABLE FOR FINDING THE EFFECTIVE DIFFUSION COEFFICIENT ($D_e = nD_r$) OF RADON IN THE RESIDUE MATRIX FROM THE OBSERVED TIME CONSTANT (τ_e) OF RADON BUILDUP IN ACCUMULATOR SET-UPS WITH VARIOUS VALUES OF $V/\pi a$ (cm^2) (cont.)

	$V/\pi a$ (cm^2)								
	50	75	100	125	150	200	250	300	450
D_e (cm^2/s)	Time constant, τ_e (min)								
2.0E-03	48	71	94	118	141	187	232	276	407
4.0E-03	34	50	67	84	100	133	165	197	292
6.0E-03	27	41	54	67	81	107	134	159	237
8.0E-03	23	35	46	58	69	92	115	137	203
1.0E-02	20	31	41	51	61	81	101	121	180
2.0E-02	14	20	27	34	41	54	68	81	121
4.0E-02	9	14	18	23	27	36	45	54	81

4. ESTIMATION OF VARIABLES AFFECTING RADON EXHALATION FLUX

4.1. RADON EMANATION COEFFICIENT (*E*)

As shown in Fig. 7 (Section 2.2.1.2), the degree of saturation has a strong effect on the radon emanation coefficient in a material. However, there are some cases where materials can be expected to have significantly lower emanation coefficients, as discussed in Section 2.

The following empirical equation relates the radon emanation in dry conditions to moisture saturation [68]:

$$E = E_0\left[1 + 1.85\left(1 - \exp(-18.8m)\right)\right] \tag{13}$$

where

E is the emanation coefficient;
E_0 is the radon emanation coefficient of residues in dry conditions;

and m is the fraction of pore space filled with water.

The emanation coefficients for ^{220}Rn are generally lower than those for ^{222}Rn (see Section 2.2.2).

4.2. DIFFUSION COEFFICIENT (D_r)

The diffusion coefficient D_r is also a sensitive function of moisture. Several correlations have been developed to evaluate this quantity from the information on moisture content. These correlations are shown in Fig. 9 (Section 2.3).

The radon diffusion coefficient in residue (D_r) may be also estimated using the empirical formula provided by Rogers and Nielson (1991) [51] that is given as:

$$D_r = D_{MA} n_T \exp(-6mn_T - 6m^{14n_T}) \tag{14}$$

where

D_{MA} is the molecular diffusion coefficient of radon in air;
n_T is the total porosity which may be calculated from $n_T = 1 - (\rho_b/\rho_g)$;

ρ_b is the dry bulk density;
ρ_g is the grain density of the residue matrix (~2700 kg/m^3);

and m is the fraction of pore space filled with water and is given by the relation $m = \rho_b\theta_d/100\rho_w nT$, where θ_d is the moisture content on a dry weight basis and ρ_w is the density of water (kg/m^3).

4.3. RADIUM ACTIVITY CONCENTRATION (R)

In most instances, the radium activity concentration in the original ore matrix will be in equilibrium with the long lived parent in that matrix. Therefore, in the absence of other data, radium concentration in the residue can be estimated from the grade of the original raw material, and any dilution or enrichment brought about by the milling and extraction processes.

An example is given here for uranium mill tailings. After processing for uranium extraction in the mill, nearly all the ^{226}Ra will be contained in the mill tailings. There will be a certain amount of dilution in the effluent slurry that will be discharged into the impoundment owing to the addition of various process chemicals. As a result, the ^{226}Ra concentration (Bq/kg) in the pile will be less than that in the host ore. If a dilution factor w (kg of solid waste for kg of uranium ore processed) is assumed, then a rough estimate of the radium concentration in the residues can be calculated from the ore grade (i.e. percentage uranium). Since 1% of uranium in the ore is equal to 1.24×10^5 Bq/kg, R can be obtained from:

$$R = \frac{1.24 \times 10^5 \mathrm{G}}{w} \tag{15}$$

where G is the average ore grade.

5. MEASUREMENT OF RADON CONCENTRATION AND EXHALATION FLUX DENSITY

5.1. GENERAL

Before a description of methodologies for the determination of radon flux is given, it is necessary to have an understanding of the available techniques for the measurement of radon gas. The theory supporting the radon flux measurement is, for the most part, common to the various field techniques. In simple terms, a container (also called a chamber) of suitable size is placed with the open end on the surface of the residues and the concentration increase or steady state concentration of radon in the chamber is measured. For a soil gas measurement, a hollow rod is inserted into the surface. For both the radon flux and the soil gas measurements, the radon concentration increase in the chamber or rod is measured for a specific time period.

The method selected for measuring the actual radon gas concentration depends upon a number of factors, including availability of resources and support facilities (e.g. availability of equipment), the scope of the project, the physical characteristics of the site to be investigated, the likely levels of radon concentration to be measured, time requirements and the available financial base.

5.2. MEASUREMENT OF RADON CONCENTRATION

As radon is an odourless, colourless, inert gas with isotopes of relatively short half-lives, there are no chemical or mass spectrometric techniques with sufficient sensitivity to measure it. Consequently, it must be determined using techniques that depend on the radioactive properties either of radon itself or of its decay products.

Figure 1 shows, within the uranium decay series, the radioactive decay sequence from ^{222}Rn into stable ^{206}Pb. Although there are several techniques for the detection of ^{222}Rn and its short lived progeny, the majority of them use detection of the alpha particles from ^{222}Rn, ^{218}Po , ^{214}Po or a combination of any of these, or detection of gamma rays arising from the decay of ^{214}Pb and ^{214}Bi.

An important consideration is whether or not the detection method needs to be capable of distinguishing between ^{222}Rn and ^{220}Rn. Some detection techniques such as alpha or gamma spectrometry inherently provide this capability, while techniques such as nuclear track detectors do not. In the latter case, the radon isotopes can only be distinguished through the equipment design, such as the membrane for ^{220}Rn decay used in twin cup dosimeters [69].

5.2.1. Mode of measurement

Radon measurement techniques may be classified based on their electrical power requirements. Passive detectors do not require an electrical power source and generally rely upon the diffusion of the radon through a small gap or filter into a detection chamber. An active device requires some form of electrical power source to operate pumps, detectors or both or to create an electrical potential to enhance the collection efficiency of the detector.

Radon collection and measurement methods may also be classified into instantaneous, semi-integrating (also known as continuous real time or continuous on-line) and fully integrating (also known as time averaging or time integrating) modes.

Instantaneous mode. In this mode, a sample of gas is taken on a short timescale. The sample may either be stored for later radon measurement (for example, if the sample has been taken into a previously evacuated scintillation flask), or have its radon measured immediately. An example of the latter approach is available on radon measurement equipment used in a so-called 'sniffer' mode, in which radon is typically present with minimal ingrowth of its progeny. These methods allow for a large number of measurements to be taken in a relatively short period of time.

Semi-integrating mode. In this mode, the sampling and counting are done simultaneously, and the radon concentrations are evaluated at regular intervals. Generally, the sampling is performed for periods of a few minutes to a few hours, depending on the arrangement of the measurement system. Sample collection may be carried out either by diffusion mode or by pump based flow through mode. Detection can be carried out by alpha spectrometry, ionization chamber methods or by gross alpha counting techniques. An example of a semi-integrating mode is flow through scintillation cells. Such methods are used to obtain information on rapid changes of radon concentrations in a given environment. In addition, they may be preferred over instantaneous modes owing to their superior sensitivity, reduction in the magnitude of systematic errors or their ability to measure a time varying signal (e.g. owing to diurnal variations in the system under study).

Fully integrating mode. This mode of measurement provides a time integrated radon concentration for the period of sampling (typically of the order of weeks or months). Sampling techniques operating in this mode must maintain an integrated record of each alpha particle impacting on the measurement medium. Once removed from the sampling site, they must maintain the exposure information until it is analysed. The passive methods are useful for obtaining long term averages of low levels of radon concentrations. They also have the advantage of enabling large numbers of measurements over extended regions at a relatively low cost. Examples of these methods include nuclear track detectors, electrets and the collection of radon on activated charcoal prior to measurement using techniques such as gamma spectrometry or liquid scintillation. All three types of methods discussed above consist of primary elements such as nuclear track detectors, solid surface barrier detectors, scintillation cells, electrets, activated charcoal and ionization chambers. These are briefly discussed in Sections 5.2.2–5.2.7.

5.2.2. Nuclear track detector

Nuclear track detectors are passive, integrating, lightweight and relatively inexpensive radon detectors [70, 71]. Their operation is based on the use of a selected film material, such as cellulose nitrate, polycarbonate or allyl diglycol carbonate (CR39). Alpha particles impinging on the film leave microscopic damage trails or 'tracks'. Following exposure, the film is etched in an alkaline solution (typically NaOH or KOH) to increase the size of the tracks so that the track density (number of tracks per unit of area) can be easily determined by optical microscopy or by automated scanning and counting technology. Owing to their relatively low sensitivity on short integration timescales, nuclear track detectors are more suited to soil gas radon measurements than to soil flux density measurements.

5.2.3. Solid surface barrier detectors

Use of solid state surface barrier detectors for detecting radon using alpha spectrometry is an active technique [72]. A common arrangement is to draw air through a cylindrical system with two filters in line. The purpose of the first filter is to remove the particulate activity, in particular the radon progeny present in the air. There is then a delay between the two filters in which ingrowth of radon progeny occurs. These progeny are then collected on a second filter, which faces the surface barrier detector.

Another approach involves collecting the decay products onto a metal surface placed inside a metallic chamber and maintained at a negative potential [73]. The positively charged ^{218}Po activity collected on the metal surface is then counted using the surface barrier detector.

In either case, knowledge of the time that the measurement system has allowed for ingrowth of the progeny in the air enables the calculation of the radon concentration.

An advantage of alpha spectrometry is the ability to spectrally separate the signals of the ^{222}Rn and ^{220}Rn progeny as these have different alpha energies. Surface barrier detectors have very low intrinsic backgrounds, enabling a low detection limit to be attained. Other advantages include their compact size, high energy resolution and ruggedness. Disadvantages include the necessity of an electrical power supply and associated electronic equipment and the relatively high cost of alpha spectrometry detection systems.

5.2.4. Scintillation cell

Scintillation cells are enclosed cylinders internally coated with a scintillation material, generally silver activated zinc sulphide powder, with a transparent end window. Interaction of alpha particles with the scintillation material causes production of light photons, which are detected and amplified by a photomultiplier adjacent to the end window, and converted to a count rate by an electronical device.

Scintillation cells can be operated in a closed or flow through arrangement. Commonly, the air passes through a filter prior to entry to the cell, especially in flow through systems; this is to remove dust as well as radon progeny present in the inlet air. The removal of the short lived radon progeny allows a reliable calculation of their subsequent ingrowth in the measurement system.

It is possible to distinguish ^{222}Rn and ^{220}Rn using scintillation cells, either by the use of two cells with a delay line for ^{220}Rn decay [74], by programmed counting [75] or by use of the pair counting technique to identify the successive alpha particles produced from ^{220}Rn and ^{216}Po [76].

Scintillation cells are amenable to good control of measurement uncertainties, meaning that high accuracy can be achieved. Cells are also robust and relatively low cost, and they allow the rapid assessment of ^{220}Rn concentration. A disadvantage of scintillation cells is that with use the background slowly increases owing to buildup of ^{210}Pb and its alpha emitting progeny ^{210}Po within the cell. This may be solved by periodically replacing the zinc sulphide screen. Another disadvantage is that dual measurement of ^{222}Rn and ^{220}Rn using the pair counting approach is limited to situations where the radon concentration is much lower than the ^{220}Rn concentration.

5.2.5. Electret

The electret radon detector is an electrically charged Teflon disc serving both as a source of an electrostatic field and as a sensor. Alpha decays in the detector chamber cause ionization of the air in the chamber and a consequent decrease of charge on the electret. The electret voltage drop over the measurement period is used to quantify the radon concentration.

Electrets are passive, integrating, lightweight and relatively inexpensive radon detectors. Their disadvantages include the necessity of allowing for the discharge of voltage caused by gamma radiation [77] and the potential influence of humidity condensation in the detector chamber on electret discharge [78, 79].

5.2.6. Activated charcoal

Activated charcoal is used as a passive medium on which radon can be adsorbed. The charcoal is weighed into a canister and sealed until it is exposed to the air to be tested. The period of exposure can range from a few hours to several days, depending on the anticipated level of radon to be encountered and the required detection limit. Following exposure, the canisters are again sealed and the radon activity determined, most commonly by gamma spectrometry, although liquid scintillation can be used following radon extraction [80].

Charcoal canisters are simple, passive, robust and low cost, and allow a relatively large number of measurements to be made over a reasonable time period (i.e. of a few days). Their capacity to adsorb radon is reduced by adsorption of water, so they are less suitable for wet or humid environments.

5.2.7. Ionization chamber

Ionization chambers used for measuring radon are enclosed metal cylinders constructed with a central collecting anode. They are accurate measurement systems that operate on the principle that the alpha particle energy dissipated during the natural radioactive decay of various radon daughters contained within the cylinder is collected on electrodes and measured as an ion current by means of an electrometer. These units can be operated in closed or flow through mode and, depending on the background and the chamber volume, they can achieve detection limits as low as 4 Bq/m^3.

5.3. MEASUREMENT OF EXHALATION FLUX DENSITY

The three basic approaches for the measurement of radon exhalation flux density are accumulation, flow through systems and adsorption. Each of these is described below.

5.3.1. Accumulation

The accumulation method is commonly used for measuring the exhalation flux densities of ^{222}Rn, ^{220}Rn and also of stable trace gases such as CO_2 and CH_4 [81]. It involves the placement of a chamber known as an accumulator [63, 82, 83], closed at one end and open at the other, inverted on the surface of interest. In order to prevent superficial leakage of accumulating radon, the mouth of the chamber is sealed onto the surface by inserting a short length of its cylindrical wall into the residue matrix. The radon atoms exhaled from the residue surface underlying the chamber enter the headspace resulting in a gradual buildup of the radon concentration. This concentration is measured at several regular intervals during the deployment period or, in some cases, just once at the end of the deployment period [84]. From these measurements, the exhalation flux density is quantified. The radon concentration is measured using on-line monitors or instantaneous methods discussed in Section 5.2, such as scintillation cells or alpha spectrometry.

Figure 13 shows a schematic diagram of the set-up for a cylindrical accumulator. The concentration of radon inside the accumulator at a time t after deployment depends on the volume, V in m^3 (= $V_1 + V_2$, where V_1 is the accumulator headspace volume and V_2 is the volume of the measuring device connected on-line), the surface area of the opening of the accumulator A(m^2), the radon isotope decay constant λ (s^{-1}), the radon exhalation flux density f (Bq·m^{-2}·s^{-1}), the degree of back diffusion of radon and the degree of ventilation in flow through mode operations. Back diffusion refers to the loss of radon from the accumulator via the soil to the atmosphere at the soil–accumulator interface and is especially significant for smaller volume (≤1 L) accumulators [66]. Possible leaks and adsorption on the accumulator material may also affect the concentration measurement.

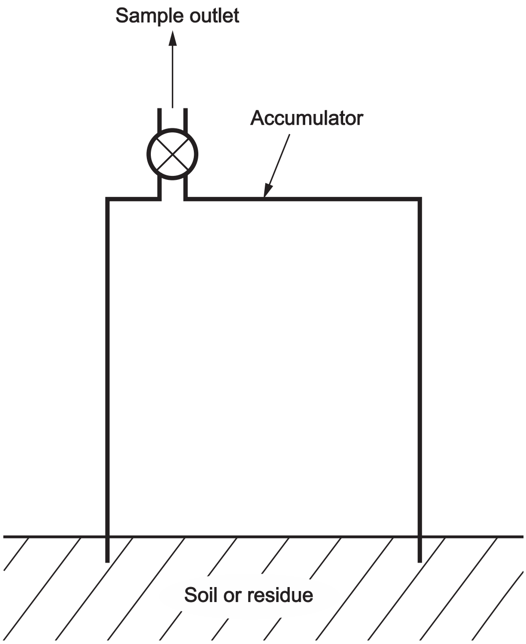

FIG. 13. Schematic diagram of the use of an accumulator.

5.3.1.1. Selection criteria of the accumulator

An accumulator can be a single chamber device that is directly inserted into the surface or it can be a two part device — a collar that is inserted into the soil and a cover that is then sealed onto the collar [83]. The use of radon absorbing materials should be avoided. The accumulator dimensions can vary depending upon the flux levels, portability requirements, time available for a measurement and desired resolution to map the flux across a given surface. A large base accumulator (~20–50 cm in diameter) is useful for obtaining representative data over larger areas and allows better coverage of a heterogeneous surface (rocks, burrows, vegetation, etc.). In addition, larger bases allow more radon to enter the chamber and hence are useful when the flux levels are small. However, large accumulators are not convenient from the point of view of portability, especially for large scale field studies.

Smaller accumulators (around 5–10 cm in diameter) are useful for high resolution spatial measurements to explore local variations of fluxes. However, they have larger back diffusion effects that affect the flux; these effects need to be accounted for through process based models. Similar considerations also exist regarding the height of the accumulator. For taller accumulators, radon may not be mixed uniformly at all heights and it may be necessary to employ a small fan inside the chamber for mixing. On the other hand, back diffusion effects will be stronger in chambers with smaller heights. In actual practice, the choice of the accumulator diameter and height should be based on considerations of portability, flux levels, spatial representativeness of measurements and the sensitivity of the monitoring instruments.

5.3.1.2. Model for interpreting concentration data

The mixing scenario in the air space bounded by the accumulator changes from a state of uniform mixing prior to deployment (due to ever present atmospheric turbulence) to a state of diffusive mixing (due to isolation from the open atmosphere) post-deployment. This causes an initial flux drop by a factor $k = [1 + n\,(D/D_a)^{0.5}]^{-1}$, (where n, D are the porosity and radon diffusion coefficient in the residue matrix and D_a is the molecular diffusion coefficient of radon in air). The value of k is typically about 0.88. Further, the back diffusion effect brings about a gradual drop in the flux as the concentration builds up inside the chamber. This is modelled by two dimensional diffusion theory [67] according to the formula:

$$C(t) = C_0 + C_m\left(1 - \exp\left[-t/\tau_e\right]\right) \tag{16}$$

where

C is the radon concentration;
t is the measurement time;
Cm is found by $k\,f\,(A/V)\tau_e$, where f is the radon flux density per bulk area, A is the surface area of the opening of the accumulator, V is the effective volume of the sampling device (m^3);
C_0 is the initial radon concentration inside the accumulator (at $t = 0$), which is assumed to be the same as the steady atmospheric radon concentration (Bq/m^3);

and τ_e is the time constant of radon buildup.

τ_e has a complicated dependence upon the dimensions of the chamber and the radon diffusion coefficient D in the residue. It can vary from a few minutes for small chambers to several hours for large chambers. Upon fitting the data on radon concentration measured at regular intervals (at least 6 points) to Eq. (16) the parameters C_0, C_m and τ_e can be estimated, from which the flux, f, may be estimated.

For measurement durations where t is significantly lower than τ_e, Eq. (16) approximates to a linear formula:

$$C(t) \approx C_0 + k\frac{A}{V}ft \tag{17}$$

The flux may be estimated from the slope obtained by linear regression analysis of the concentration data with respect to time. Equation (17) is ideally suited for short duration accumulator deployment during which only a few concentration measurements have been made.

The method of data analysis for measurement of ^{220}Rn flux is somewhat different from that for ^{222}Rn. In view of its short half-life, the ^{220}Rn concentration in the accumulator will attain a steady state within approximately 5 min. If C_1 is the steady state concentration, the flux may be estimated with Eq. (18).

$$f_{\mathrm{Rn-220}} = \frac{\mathrm{V}\lambda_{\mathrm{Rn-220}}}{A}\left(C_1 - C_0\right) \tag{18}$$

5.3.2. Flow through method

In the flow through method, a chamber working on a similar principle to an accumulator is placed over the surface to be investigated. However, in this case, the air in the chamber is continuously removed at a constant rate and the radon concentration in the exhaust stream is measured by a semi-integrating technique such as a flow through scintillation cell.

The flow rate of air through the chamber must be sufficiently high to prevent buildup of radon in the headspace of the chamber (i.e. a steady state should be achieved). However, if the airflow rate is too high, the radon concentration in the exhaust air will be low and cause increased uncertainties in the radon measurements. The air pressure in the chamber should be the same as the ambient pressure outside. Care should be taken to ensure that restricted replacement of withdrawn air or use of a pump at the inlet do not result in lower or higher pressure, respectively.

At steady state, the exhalation flux density is proportional to the concentration of radon in the exhaust stream and to the exhaust stream flow rate, and inversely proportional to the active surface area of the chamber. Other factors in the calculation of radon flux density are dependent on the details of the system and its calibration.

For non-spectroscopic radon detection such as scintillation cells, the presence of ^{220}Rn and its decay products must be allowed for, often by use of a delay line for ^{220}Rn decay prior to filtration and counting. Double cell systems have been developed for simultaneous measurement of both radon and ^{220}Rn exhalation flux densities [74]. Figure 14 shows a schematic diagram of such a double cell flow through system. In this example, a filter is used to remove radon and ^{220}Rn progeny from the airstream, and radon and ^{220}Rn together with their subsequently ingrown progeny are counted in the first flow through scintillation cell. The delay line allows the decay of most of the ^{220}Rn. A second filter then removes the radon and ^{220}Rn progeny, and the second scintillation cell counts radon and its subsequently ingrown progeny together with a small component of the remaining ^{220}Rn and ^{220}Rn progeny.

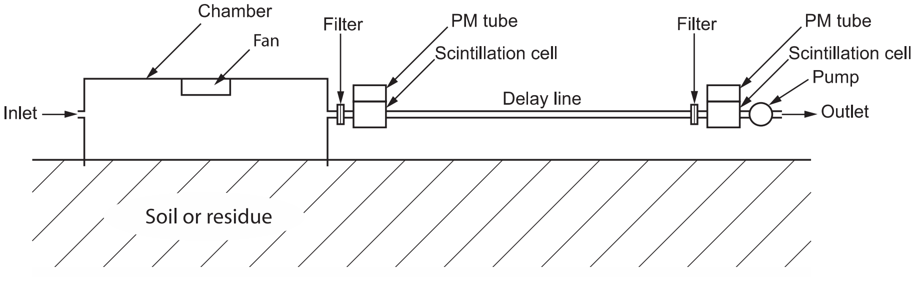

FIG. 14. Schematic diagram of the use of a double cell flow through system.

5.3.3. Adsorption

The adsorption method for ^{222}Rn exhalation flux involves the use of an adsorption medium (usually activated charcoal) placed in close proximity to the surface being investigated.

Figure 15 shows a schematic diagram of a charcoal adsorption canister. Before use, the charcoal is heated in an oven to remove any radon, moisture and other contaminants, which may have been adsorbed previously. This drying may be carried out when the charcoal is already in the canister or prior to it being weighed into canisters. Once prepared, the canister is sealed to prevent adsorption of ambient radon or moisture onto the charcoal.

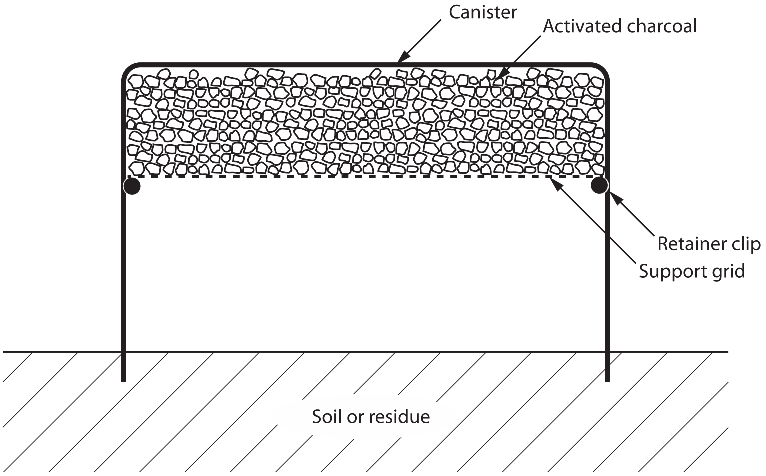

FIG. 15. Schematic diagram of the use of a charcoal adsorption canister.

Following exposure, the canisters are again sealed and the activities of the radon progeny ^{214}Pb and ^{214}Bi measured, most commonly by gamma spectrometry, following a short (~3 h) ingrowth period for the progeny. Liquid scintillation counting may be used as the measurement technique if a higher counting efficiency is required [80]. The measurement should take place as soon as practicable after the progeny ingrowth period in order to minimize loss due to radon decay.

The radon exhalation flux density over the period of exposure can be estimated using the expression:

$$f = \frac{N \cdot t_c \cdot \lambda^2 \cdot \exp(\lambda t_d)}{\varepsilon \cdot A \cdot \left[1 - \exp(-\lambda t_e)\right] \cdot \left[1 - \exp(-\lambda t_c\right]} \tag{19}$$

where

f is the radon flux density ($\mathrm{Bq \cdot m^{-2} \cdot s^{-1}}$);
N is the net count rate, after background subtraction, obtained during the counting period (counts per s, or $\mathrm{s^{-1}}$);
t_c is the counting period (s);
λ is the radioactivity decay constant for $^{222}\mathrm{Rn}$ ($\mathrm{s^{-1}}$);
t_d is the delay period from the end of the exposure to the beginning of the counting interval (s);
ε is the counting efficiency of the system relative to the activity of adsorbed radon (Bq/s);
A is the area of the canister ($\mathrm{m^2}$);

and t_e is the period of exposure of the charcoal in the canister(s) [85].

When exposed to humid or wet environments, the charcoal will adsorb moisture, which can lead to reduction in the radon collection efficiency [80, 86]. To ensure that water adsorption is not problematic or to allow any required correction, water adsorption can be quantified by weighing the canisters before and after exposure.

5.3.4. Measurement of soil gas concentration

Soil gas measurement is normally carried out using a tube inserted into the surface. This probe is open at the end, or has an opening at the side, to allow the diffusion of radon into the inside volume. The radon concentration inside the probe is measured either in situ (for example, using nuclear track detectors) or by removal of a sample of the air inside the probe for measurement of radon by an instantaneous or continuous on-line method [87, 88].

Depending upon the capabilities and requirements of the measurement technique employed, the system may need to include a porous membrane to ensure removal (by decay) of $^{220}\mathrm{Rn}$, a dehumidifier or a filter, or both to remove radon progeny. As radon concentrations in soil gas are generally in the range of 10^3–10^5 $\mathrm{Bq/m^3}$, and concentrations in NORM residue may be higher than this, the upper end of the calibration range of the measurement instrument may need consideration.

It is important that a good seal is obtained between the probe and the soil or residue being tested. This may be more difficult to achieve where stones are present. One study [88] indicated that passive methods may give lower results than active methods for largely impermeable clay soils, due to decay of radon during diffusion from the soil pores.

Soil gas radon concentrations exhibit seasonal changes [89–91]. Diurnal variations have also been observed for soil layers close to the surface [92].

5.3.5. Measurement of mass exhalation rate

The mass exhalation rate is defined as the radon activity released into the air per unit time from the mass of the matrix and is measured by enclosing the sample in a closed chamber and monitoring the buildup of radon concentration in the chamber at regular time intervals. Typically, about 350–500 g of residue sample is enclosed in a leaktight metallic chamber coupled to a continuous ^{222}Rn monitor. The radon concentration $C(t)$ at time t since the closing of the chamber builds up according to the formula [93–95]:

$$C = \frac{J_m M}{V\lambda_e}\left[1 - e^{-\lambda_e t}\right] + C_0 e^{-\lambda_e t} \tag{20}$$

where

J_m is the mass exhalation rate ($\text{Bq}\cdot\text{kg}^{-1}\cdot\text{h}^{-1}$);
C_0 is the ^{222}Rn concentration present in the chamber volume at $t = 0$ (Bq/m^3);
M is the total dry mass of the sample (kg);
V is the effective volume (volume of chamber + internal volume of ^{222}Rn monitor — volume of sample) (m^3);
λ_e is the effective decay constant for ^{222}Rn, which is the sum of the leak rate (if existing) and the radioactive decay constant of ^{222}Rn (s^{-1});

and t is the measurement time (s).

Upon least square fitting of the data to the above formula, J_m may be obtained from the fitted parameters with the knowledge of the dry mass M of the sample. The dry mass is estimated by drying the sample at about 110°C until it attains constant weight.

5.4. CALIBRATION AND UNCERTAINTIES

The assurance of accuracy expected in measurement results is realized by traceability. That is, the status of the calibration method for utilized monitors should be in line with a national or international standard. In addition, procedures for calibration and performance tests should be described clearly and objectively in the written quality assurance plan established by each organization.

Generally, the instrumentation for field measurements should be tested and calibrated using a STAR (System for Test Atmospheres with Radon) operated by reference organizations or manufacturers. Since a STAR provides a reference concentration of radon in the air, this represents a testing and calibration of the equipment's system for measuring radon concentration only. The radon concentration in the reference atmosphere is usually determined by a secondary standard, which is calibrated by the primary standard. The calibration is performed under atmospheric standard conditions, i.e. 20°C, 60% relative humidity and 101.3 kPa atmospheric pressure.

The air sample should be filtered to remove ^{222}Rn and ^{220}Rn decay products before entering the detector. This is a requirement not only for continuous methods but also for grab sampling methods, owing to the restraint of increasing background.

Recently, radon measured absolutely has become popular as a primary standard in some reference organizations [96, 97]. However, radon released from an aqueous solution of a soluble ^{226}Ra salt is also available and may have the benefit of reproducibility.

For ^{222}Rn exhalation flux density measurements, the greatest measurement uncertainty is the degree of uniformity of mixing in the collection chamber and measurement instrument. Therefore, in this case, it is extremely important to include a mixing element in the uncertainty budget.

For accurate field measurements, instrumentation should be protected against more severe environmental conditions than are found in laboratories. In addition, performance tests must be carried out in expected environmental conditions because it is difficult to remove all confounding factors. The major considerations in special requirements for radon flux density measurements are given below.

Detection limits. The measurement plan should not only consider the lower detection limit but also the upper one. The radon concentration in the accumulation chamber can be expected to sometimes be higher than several hundred thousand Bq/m^3. For example, for a typical radon exhalation flux density from uranium mill tailings of 10 Bq·m^{-2}·s^{-1}, the radon concentration in an accumulation chamber of 10 cm in height reaches approximately 4×10^5 Bq/m^3 after 1 h.

Background. Even in the continuous measurement of indoor radon, ^{210}Pb, a long lived decay product of radon, accumulates in the detector and may cause uncertainty in the radon measurement. For radon exhalation flux density measurement, the ^{210}Pb in the detector should be checked periodically and cautiously owing to higher radon concentrations in the accumulation chamber than in the indoor environment.

Detector volume. The volume of the detector, including sampling devices such as pumps and tubes, may also cause uncertainty in estimation of the activity concentration of radon, which is accumulated within a given time in a container with a known volume.

Sensitivity to ^{220}Rn. Generally, ^{222}Rn and ^{220}Rn are separated by using their very different half-lives. Therefore, especially in continuous measurements, special attention should be paid to the half-life of ^{220}Rn. Alternatively, ^{222}Rn and ^{220}Rn can be separated using the alpha particle energies emitted from them or their decay products or both.

Humidity. The humidity inside the accumulation chamber may affect the efficiency of some detectors or detection systems, such as ionization chambers, activated charcoal, electret, etc. Air drying systems may be available for some instruments.

Gamma dose. The ambient gamma dose may affect the efficiency of some detectors or detection systems, such as ionization chambers and electrets.

A quality assurance plan may be established, applied and maintained to objectively show the consistency of measurement results or the compliance with relevant laws, regulations and other requirements or both. Such a plan should make clear its scope, satisfy laws and regulations, and be accepted by all interested parties. The plan could also include a standardized system in writing, such as a procedure for measurement, handling and for the maintenance of instruments used, recording and so on. These should be included in the monitoring plan described in Section 8 in detail.

6. SIMPLIFIED EXPRESSIONS FOR ESTIMATING RADON EXHALATION FLUX DENSITY

6.1. GENERAL

The control of radon releases from radium bearing NORM residue repositories begins with an evaluation of the radon source term and the effectiveness of any cover introduced to attenuate radon migration to the surface.

The main factors affecting the generation, transport and control of radon in residue matrices have been discussed in general terms in Section 2. In the present section, mathematical expressions are provided to enable simple calculations for estimating the radon exhalation flux density.

The simplest approach for estimating exhalation flux density is based on the application of diffusion theory to radon transport in the repository matrix, under the following idealized assumptions:

(a) The residue properties are homogeneous.
(b) Radium is uniformly distributed in the residue matrix.
(c) Transport owing to pressure induced advection is negligible.

Although certain variations in the residue properties and radium content are inevitable, representative values of these parameters are used to evaluate local fluxes. If these variations are essentially limited only with respect to depth (homogeneous laterally), the multilayer approach can be used to evaluate the fluxes. Further, assumption (c) is generally justified under the premise that it is difficult to sustain appreciable pressure differentials between the soil and the open atmosphere for a significant length of time. Given these assumptions, expressions are provided for three situations, namely: radon flux from a bare residue repository, a repository covered with homogeneous material (with negligible radium content) and a repository with a multilayer cover. The development of the radon diffusion equation is advanced in Appendices I and II.

6.2. RADON EXHALATION FLUX DENSITY FROM BARE RESIDUE

The mean radon diffusion length, which may simply be referred to as the diffusion length, L_r, is a measure of the mean distance travelled by radon atoms in the residue matrix before their radioactive decay. It is defined as:

$$L_r = \left(\frac{D_r}{\lambda}\right)^{1/2} \tag{21}$$

where D_r is the radon diffusion coefficient of the residue (m^2/s); and λ is the decay constant of ^{222}Rn (s^{-1}).

L_r is a measure of penetration by a plume into a zone with diffusion constant D_r owing to a constant concentration source at the origin, with a larger L_r corresponding to greater penetration. If the concentration source is unity, L_r is the square root of the centroid of that plume with respect to the origin.

For bare residues, there are two general cases. In the first case, the residue thickness is much greater than the radon diffusion length and the residue may be regarded as being of infinite thickness for the purpose of the estimation of radon flux density. In the second case, the thickness of the residue repository is comparable to or less than the diffusion length.

6.2.1. Residue thickness much greater than radon diffusion length

In the case of the residue thickness being much greater than the radon diffusion length, diffusion theory predicts that the radon flux density is directly proportional to the radium activity concentration, the emanation coefficient and the bulk density, and has a square root dependence on the radon diffusion coefficient:

$$f_r^{\infty} = R\rho_b E\left(\lambda D_r\right)^{1/2} \tag{22}$$

where

f_r^{∞} is the radon flux density;
R is the radium activity concentration;
ρ_b is the bulk density;
E is the emanation coefficient;
λ is the decay constant;
D_r is the radon diffusion coefficient;

and the subscript ‘*r*’ refers to the residue material and the superscript ‘∞’ refers to a residue whose thickness is large (the expression is strictly true only in the limit as the thickness approaches infinity).

Equation (22) is also applicable to ^{220}Rn, in which case the flux is essentially contributed by the ^{224}Ra content present in the top layer of a few cm thickness, which is about the order of a diffusion length for ^{220}Rn. For a typical top residue with a D_r value of 2×10^{-6} m^2/s, the diffusion length for ^{220}Rn is approximately 0.013 m.

6.2.2. Residue thickness comparable to or less than the diffusion length

When the thickness of the residue repository is comparable to or less than the diffusion length L_r (~1 m for ^{222}Rn), Eq. (22) requires modification. For a residue repository of thickness z_r, diffusion coefficient D_r (diffusion length L_r) and porosity n_r resting on a soil with porosity n_s and diffusion coefficient D_s, the solution of the multilayer radon diffusion equation (see Appendix I) yields the following expression for the flux density, f_r, of radon on top of the repository:

$$f_r = f_r^{\infty} \frac{2\sinh\left(\frac{z_r}{2L_r}\right)\left[\sinh\left(\frac{z_r}{2L_r}\right) + k\cosh\left(\frac{z_r}{2L_r}\right)\right]}{\sinh\left(\frac{z_r}{L_r}\right) + k\,cosh\left(\frac{z_r}{L_r}\right)} \tag{23}$$

where

$$k = \frac{n_r}{n_s}\left(\frac{D_r}{D_s}\right)^{1/2}$$

Three limiting formulas can be obtained from Eq. (23).

If k is much lower than 1 ($k << 1$), i.e. if the matrix below the residue repository is porous, and has a diffusion coefficient and porosity that are much greater than those of the repository, the following expression is obtained:

$$f_r = f_r^{\infty}\tanh\left(z_r/2L_r\right) \text{ for a porous base } (k = 0) \tag{24}$$

On the other hand, if the base of the repository is totally impervious to radon ($k = \infty$), the equivalent expression is (see Appendix I):

$$f_r = f_r^{\infty}\tanh\left(z_r/L_r\right) \text{ for an impervious base } (k = \infty) \tag{25}$$

The impervious base is the most conservative estimate that yields the highest flux since penetration of radon into the soil–water matrix below the repository will always reduce the emission. The variation of the flux as a function of $z_r/2L_r$ for different k values is shown in Fig. 16.

As may be seen from Eq. (23), residue thickness beyond $2L_r$ (~2–4 m) has little influence on flux. For shallow repositories ($z_r << L_r$) with impervious bases, Eq. (24) suggests that all the radon escapes to the surface. When z_r is much greater than L_r ($z_r >> L_r$), Eq. (23) is equivalent to Eq. (21).

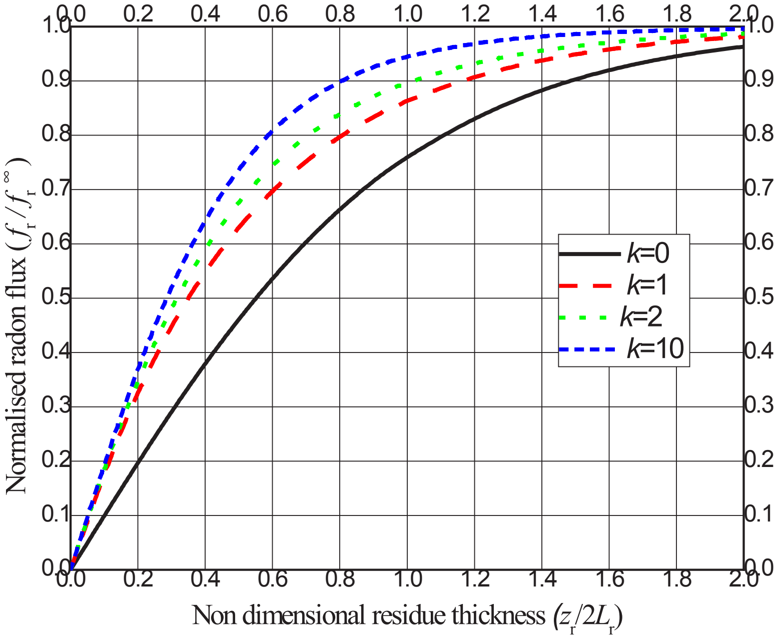

FIG. 16. Variation of the normalized radon flux (f_r/f_r^{∞}) from a residue repository as a function of non-dimensional residue thickness ($z_r/2L_r$) for different values of the parameter k.

6.3. RADON FLUX DENSITY FROM RESIDUE REPOSITORIES WITH A SINGLE COVER

Earthen materials with a low radium content are effective covers for attenuating radon exhalation from residue repositories. If radon emission from the cover materials is neglected, the flux density from a residue surface covered with soil of thickness z_c may be approximated by:

$$f_{cr} \approx f_r \exp\left[-z_c/L_c\right] \tag{26}$$

where L_c is the radon diffusion length in the cover material. Since L_c decreases significantly with increasing moisture content in the material, a cover material that compacts well and retains moisture is the most effective for controlling radon releases.

The following example illustrates the application of Eqs (22) and (26). For a moist uranium residue pile with $R = 4 \times 10^4$ Bq/kg, $\rho_b = 1500$ kg/m^3, $E = 0.2$ and $D_r = 1.0 \times 10^{-6}$ m^2/s, which corresponds to $L_r = 0.69$ m, the radon flux density is estimated using Eq. (22) as 17 Bq·m^{-2}·s^{-1}. This calculation is applicable for repositories of thickness exceeding about 2 m.

For the example above, the ratio of the surface flux to radium activity concentration (R) is equal to 4.3×10^{-4} kg·m^{-2}·s^{-1} or, if R is expressed in Bq/g, this ratio is 0.43 g·m^{-2}·s^{-1}. For moist uranium residues with $D_r = 2.5 \times 10^{-7}$ m^2/s, the ratio is 0.22 g·m^{-2}·s^{-1}. It has been generally found that the ratio of flux (Bq·m^{-2}·s^{-1}) to radium concentration (Bq/g) increases with decreasing moisture, ranging from 0.2 for wet residues to 0.6 for moist residues and 1.2 g·m^{-2} ·s^{-1} for dry residues [98].

If the same repository is covered by 1.5 m of soil with $D_c = 4 \times 10^{-7}$ m^2/s, the ^{222}Rn flux density decreases by a factor of approximately 31 (i.e. to a value of 0.6 Bq·m^{-2}·s^{-1}).

6.4. RADON FLUX DENSITY FROM RESIDUE REPOSITORIES WITH MULTIPLE COVERS

It may be advantageous or necessary to use different materials to cover a residue repository. This situation requires multilayer analysis. Multilayer analysis is also useful to address situations where the moisture varies significantly with depth for a single material cover, thereby causing drastic variation in the diffusion coefficients with depth. A three layer system is shown schematically in Fig. 17. Each layer is characterized by a diffusion coefficient (and equivalently by a

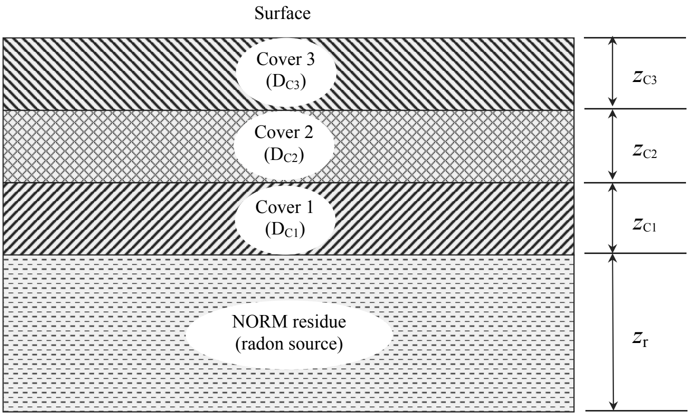

FIG. 17. Multilayer cover system on the surface of the NORM residue.

diffusion length) and thickness. The value of the diffusion coefficient depends upon the moisture content, compaction and type of material.

As a first approximation, Eq. (26) can be applied to each cover layer successively considering the radon flux density from the lower layers to be the source flux into the cover layer being analysed. This gives:

$$f_{c1} \approx f_r \exp\left[-z_{c1}/L_{c1}\right]$$

and

$$f_{c2} \approx f_{c1} \exp\left[-z_{c2}/L_{c2}\right] \approx f_r \exp\left[-\{z_{c1}/L_{c1}\} - \{z_{c2}/L_{c2}\}\right]$$

This may easily be generalized to an n-layered system as

$$f_{c1} \approx f_r \exp\left[-\sum_{j=1}^{i}\left\{-z_{cj}/L_{cj}\right\}\right] \tag{27}$$

The factor multiplying f_r on the right hand side of Eq. (27) is the effective attenuation factor for a combination of multiple covers. The usefulness of these equations may be demonstrated by considering the previous single layer example to be actually stratified into two layers ($i = 2$) of widely differing moisture content and hence differing diffusion coefficients. If the lowest metre of the cover has a diffusion coefficient of 4×10^{-7} m^2/s and the uppermost half metre has a diffusion coefficient of 2×10^{-6} m^2/s, then, from Eq. (27), the exhalation flux density is calculated as $f_{c2} = 1.1$ Bq·m^{-2}·s^{-1}.

7. EFFECTS OF METEOROLOGICAL CONDITIONS AND THE REPOSITORY COVER

7.1. METEOROLOGICAL EFFECTS

The weather parameters of air pressure, temperature, rainfall and wind speed affect radon exhalation flux from the surface of residue repositories. The physical and hydrological properties of the cover and residue and the length of time for which radon fluxes are reported are important in assessing the extent of meteorological effects. In particular, diurnal and seasonal variations in the weather parameters can lead to variations in flux over these time periods.

Usually, the duration of atmospheric pressure changes is much less than the half-life of ^{222}Rn. The exhalation flux density from the soil surface can be expected to increase if the atmospheric pressure decreases. Since atmospheric pressure changes are cyclic, the increases and decreases in the flux tend to compensate for each other. Thus, the long term fluxes are not strongly dependent on atmospheric pressure changes.

Strong winds can cause erosion and are of particular concern for exposed residues, where contaminated particles may be carried to surrounding locations. In cold regions, strong winter winds have contributed to freeze-drying effects on covers and residues. Where residue has been isolated under a water cover, wind induced wave action may increase radon transport to the surface by mixing or reducing the cover thickness.

Longer term seasonal variations may influence the annual average radon fluxes. For example, a snow covering and frozen ground can decrease the annual surface flux. Rainfall directly affects soil moisture content. In one study in a tropical region, radon exhalation flux densities were observed to increase as the soil started turning from dry to moist, followed by a rapid decrease as the rainy season started. Average radon exhalation flux densities over the wet season were lower than in the dry season, but had greater variability [99].

Similarly, long term temperature changes may alter the flux. For example, higher temperatures can slightly increase the diffusion coefficient, which slightly increases the flux. Although the surface temperature of a large residue pile can fluctuate widely, the temperature within the pile is typically more uniform. However, depending on the physical and chemical properties of the pile, other processes may lead to high internal temperatures (e.g. bacterial reactions).

Covers that are used to reduce radon emissions are often based in part on consideration of local meteorological conditions. Covers that depend on evapotranspiration to help control infiltration rely on seasonal fluctuations in precipitation and temperature to perform effectively. Higher temperatures in a waste rock pile may lead to early degradation of geomembrane liners. Long term modelling of cover systems should consider the impact of a changing climate on the integrity of a cover system.

7.2. ASPECTS OF REPOSITORY COVER DESIGN

7.2.1. Surface cover effects

During the remediation of mining and industrial sites, covers or caps are often placed on the surface of the residue to control the transport of contaminants to the environment. One important design consideration for cover systems is their ability to adequately reduce radon exhalation. Regulations may vary, requiring for example that a cover achieve a prescribed exhalation flux density value, or that radon levels be reduced to their background levels.

The performance of a cover system component depends on two factors: (1) the performance of the component itself and (2) the way individual components interact as a system. Owing to the long half-life of the radon progenitors, covers are typically expected to perform over extended time frames. Several factors such as wind and soil erosion, changes in moisture content and intrusion by tree roots and burrowing animals may result in changes in performance over time. Given the absence of observations and performance data in the long term, models are used to predict the long term performance of residue containment systems.

Covering layers reduce radon flux density by increasing the time taken by radon atoms produced in the residue to be transferred to the surface, allowing for radon to decay before reaching the atmosphere. Radon migration through the cover material is largely governed by diffusion, although advective processes may also assist in the transport, e.g. if pressure differentials exist across the material. Thus, decreasing the radon gas diffusivity and permeability of the cover media can increase cover efficiency. The diffusivity and permeability of the soil generally depend on orientation, soil type, pore size distribution, water content and the degree and method of compaction.

The degree of diffusive transport of radon through any material can be characterized by a quantity known as the mean diffusion length, or relaxation length, which can be deduced from the ratio of the diffusion coefficient to the radioactive decay constant (see Ref. [100] for an exact characterization), with greater radon penetration at steady state. This quantity can vary greatly between materials. For fine grained moist soils, the diffusion length for ^{222}Rn ranges from 0.2 to 0.5 m, whereas for coarse, dry sands, the value may be as high as 1.5 m. Conversely, in dense material (e.g. dense granite), the diffusion length is much smaller, of the order of 0.1–0.15 m. In air, the diffusion length is 2.2 m. The diffusion length of ^{220}Rn is about 77 times smaller than that of ^{222}Rn for any given material.

In the following subsections, considerations for radon migration related to different aspects of repository cover design will be described.

7.2.2. Surface cover materials

Cover materials that have been effective in reducing radon emissions include water, earthen materials, geosynthetics such as geomembranes and geosynthetic clay liners, and evapotranspirative barriers. Simple covers may contain one type of material; however, combinations of different materials are often required. Factors that influence the selection of cover material include climate, availability of materials, properties of the residue, regulatory requirements and other site specific needs or issues (e.g. generation of acid rock drainage, water infiltration, erosion).

7.2.2.1. Water covers

As the diffusion coefficient of radon is much lower in water than in air, water covers can significantly reduce radon emissions. Subaqueous disposal is more common at uranium mine waste facilities than at other NORM facilities. Underwater disposal is typically achieved by flooding a residue impoundment or relocating the residues or waste rock to a storage basin such as a former open pit mine. In shallow water covers, diffusion may become enhanced by natural convection, wind action and evaporation. Ref. [101] examines the issue of surface water effects in predicting radon emissions from water covered uranium residue impoundments, where residue characteristics and local topography can vary significantly. For the shallow (<1 m) impoundment water, water movement was sufficient to remove radon from the residues–water interface and transport it to the atmosphere in a short time (several hours), resulting in virtually no radon containment by the surface water itself compared with similar bare saturated residues. Nevertheless, the radon flux from residues saturated with water is still much less than that from dry residues [37]. Environments with snow cover and freezing have also been shown to reduce radon emissions.

Example: In Northern Saskatchewan, Canada, mineralized waste rock has been placed in former open pit mines; several metres of water cover provide an effective barrier to oxygen and radon.

7.2.2.2. Earthen cover materials

Low permeability soil covers are generally made from fine grained silts and clays. These materials can achieve very low permeabilities of less than 1×10^{-9} m/s [102] and are very effective for limiting radon transport. However, permeability may increase for unprotected clay covers as a result of wetting and drying, freezing and thawing, and of deformation processes [103], which can lead to increased radon transport. A conventional cover system designed to limit infiltration and radon emissions has a lateral drainage layer consisting of either coarse sand or gravel, to limit head buildup on top of the clay, and a top layer of durable rock for erosion protection. Depending on the climate and environment, this rock may also support a vegetative cover.

Use of rock mulch and vegetation and avoidance of compaction may be strategies used for limiting erosion by rainfall [104], although using vegetation and avoiding its compaction may mean higher radon flux densities than would otherwise be the case. In such cases, the design must be optimized, taking into account the competing requirements and the specific characteristics of the situation.

Native soil covers may provide adequate resistance to wind and water erosion, and may also encourage indigenous plant species; for long term design, weather resistant natural materials are favoured. However, even if compacted, sandy native soils are much less effective in reducing radon emissions than other kinds of native soil, unless a considerable soil depth is used.

As discussed in Section 2.3, soil moisture can significantly affect radon transport. Moist and saturated soil conditions in cover materials tend to slow the rate of diffusion to the surface. The diffusion coefficient through a saturated soil may be several orders of magnitude smaller compared with that of a dry soil (Fig. 9).

Example: The decommissioning of the Cluff Lake Uranium Mine site in Canada involved placing a 1 m thick layer of locally available glacial till (the till was classified as a silty sand material with grading lying within the following envelope: 12–30% gravel, 58–64% sand, 16% silt and clay, 0–8% boulders and cobbles) on top of a residue impoundment containing 2.6 Mm^3 of residues covering an area of 55 ha. After placement of the cover, radon concentration measurements were equal to background levels.

7.2.2.3. Geosynthetics

Manufactured from synthetic polymeric materials, geosynthetics are increasingly being used as covers in containment facilities, such as low level radioactive waste landfills. The two main types of low permeability geosynthetics used in cover design are geomembranes and geosynthetic clay liners. Other types of geosynthetics such as geonets and geotextiles are used to serve other functions in a cover system (e.g. used as filters to separate fines) and help to protect low permeability materials.

Geomembranes composed of high density polyethylene are most commonly used for lining residue facilities, while the more flexible polymers, such as low density polyethylene, polypropylene and polyvinyl chloride, are often used in final covers to accommodate settlement. These membrane materials have a very low hydraulic conductivity to water (less than 10^{-13} m/s); however, leakage can occur through defects such as tears or inadequate seams in the geomembrane. An assessment of leakage through holes in the geomembrane requires an estimate of the size and frequency of such holes. Ref. [105] provides guidance on estimating defects per hectare and Ref. [106] reviews gas migration through geomembranes. Long term covers should consider the degradation concerns for geomembranes. The estimated service life of a geomembrane can vary widely and is strongly dependent on the temperatures to which the geomembrane is exposed, ranging from about 1000 years at 10°C to as little as 15 years at 60°C. For this reason, geomembranes are often part of composite cover systems, where a second layer (of clay, for example) limits releases from membrane imperfections and thus extends the life of the cover system.

Geosynthetic clay liners (GCLs) consist of a layer of bentonite supported by one or two layers of fabric material. The bentonite layer is about 6 mm thick, and has a mass per unit area of 3.6–4.3 kg/m^2 and a water content of 10–20% at the time of manufacture. Although GCLs have a very low hydraulic conductivity, they may not be thick enough to provide sufficient time for radon to decay; GCLs are therefore also more effective when combined with other layers.

Example: The cover for a 32 ha low level radioactive waste landfill in Fernald, Ohio (1.0 Mm^3 of mixed industrial, construction and low level radioactive waste), consists of a 2.9 m thick composite final cover system. The cover includes a geomembrane underlain by a geosynthetic clay liner and a compacted clay layer. The 0.9 m biointrusion layer placed on the surface of the cap system is designed to further limit the release of radon [103].

7.2.2.4. Evapotranspirative cover layers

Evapotranspirative cover layers rely on seasonal removal by evaporation and transpiration to control infiltrating surface water. They are becoming more common in arid or semiarid regions where potential evapotranspiration significantly exceeds actual precipitation [107]. Capillary barriers can be placed below the evapotranspirative cover. This would create a storage reservoir to enhance the available water capacity and limit unsaturated flow [108]. Available water storage capacity has been defined as the difference between the total amount of water remaining when the soil dries and the permanent wilting point for plants. A conservative choice would be to use the wilting point to determine how dry conditions predict the maximum exhalation of radon gas from a waste facility.

7.2.3. Effects of vegetation on the cover

Surface vegetation can be used to protect cover materials; it is used for erosion control, stabilization and limiting infiltration. However, investigations of cover systems have identified several mechanisms by which surface vegetation could compromise design criteria for mitigating radon release by cover materials:

(a) Plants may act to alter soil moisture conditions, which can affect radon flux density. For example, plants may remove moisture from the soil during transpiration, which would result in a drier soil, thus increasing the radon flux density.
(b) Root intrusion can increase the permeability of the underlying soil layers, resulting in an increased radon flux density. Roots from woody plants and deep shrubs have been observed to penetrate the compacted clay layer at several uranium mine waste sites [109].
(c) Plants that are rooted deeper within the cover and waste system may act as conduits for radon. Ref. [110] shows that aqueous radon and radium are readily extracted from the soil into vascular plants via the transpiration stream and that radon is eventually released into the atmosphere through the leaf cuticle.

Numerical models have been developed to simulate radon and radium transport in a multiphase soil system that may or may not include vegetation. For more information, see Ref. [111].

8. MONITORING RADON RELEASES FROM NORM FACILITIES

8.1. GENERAL

Monitoring for radionuclides is undertaken at NORM facilities to demonstrate that adequate measures have been taken to protect the environment and keep radiological doses as low as reasonably achievable (ALARA). Many monitoring programmes are required by regulations, but the collected data are also beneficial for other reasons, such as for effective control of discharges and for public information.

Comprehensive information for the design and operation of source and environmental monitoring programmes and systems relating to the release of radioactive material to the environment from authorized practices can be found in Safety Reports Series No. 64 [112]. This section focuses on monitoring radon exhalation from NORM residue repositories.

A radon exhalation monitoring programme consists of a set of activities implemented to sample, measure and analyse radon in and around the area of a residue repository for the purpose of, but not limited to:

- Verifying compliance with any authorized limits or safety standards;
- Determining the source term for predictive modelling of radon levels;
- Obtaining data for the design and assessment of the performance of mitigation strategies such as waste covers.

The purpose, design and implementation of a radon monitoring programme are dependent on the facility's characteristics, environmental characteristics and anticipated effects. In particular, the monitoring programme will change according to the operational phase of the residue repository.

A monitoring programme should be planned in advance in order to ensure proper progression of the activity and the appropriate selection, use and optimization of resources.

Planning should define:

- The purpose of the monitoring plan;
- The design of the programme;
- Monitoring techniques and methods;
- Survey implementation;
- Data handling, analysis and maintenance;
- Calibration, quality control and quality assurance.

Sections 5 and 6 discussed the methods and tools available for calculating and measuring radon releases from NORM residues. This section briefly reviews the factors that should be considered in setting up a radon exhalation monitoring programme. The guidance given here is general in nature and relates only to the monitoring part of the overall monitoring programme. Since the design and implementation are site specific, expert advice on carrying out such activities should be obtained, especially if the information is to be used to design covers for residue repositories or to assess health effects to workers or nearby residents as well as to non-human biota.

8.2. DATA COLLECTION AT VARIOUS STAGES

Knowledge of the radon exhalation flux densities of a site prior to operation is a very important factor in assessing the above background radiological impact of a NORM residue repository during and after operation. Collection of suitable data prior to site operation is crucial as such data cannot be collected once the site is disturbed. For example, in the case of one uranium mining operation in northern Australia, it is possible that mining operation reduced the radon exhalation flux from the site because the original ore body outcropped at the surface, but the small pre-mining dataset does not enable the determination of a sound value for the true pre-mining exhalation flux [113].

Radon monitoring is conducted during the operational stage for periodic impact assessments along with the assessment of incremental changes in exhalation flux densities. These data will be used to guide decommissioning activities.

Post-operational monitoring is used to evaluate the adequacy of decommissioning activities, such as the ability of cover materials to limit radon exhalation.

Periodic and systematic review of the monitoring programme is necessary to ensure that the information being collected is still relevant and is fulfilling required objectives.

8.3. FACTORS IN DESIGN AND IMPLEMENTATION

A number of factors need to be considered in setting up a programme to assess the releases of radon from residue repositories. Many of these factors are site specific. To be effective, the monitoring programme should consider the following [112, 114]:

- Location and number of sampling stations;
- Location and number of control sites;
- Timing and frequency of measurements, taking into account diurnal or seasonal variations;
- Regulatory requirements for the accuracy of instruments and the design of the programme;
- Resource considerations;
- Physical, hydrogeological and radiological characteristics of the residue repository;
- Effect of meteorological parameters, such as precipitation, wind speed and direction, temperature, relative humidity and barometric pressure;

- Seasonal and ageing effects, such as changes in moisture content of the residue and the cover.

8.4. QUALITY ASSURANCE

Incorporating quality assurance activities into each aspect of the radon monitoring programme (project design, sampling, sample handling, data interpretation) will provide confidence in the results of the programme. The monitoring programme should address the following issues: instrument calibration and maintenance, the establishment of sampling and measurement protocols, adherence to relevant standards and best practices, and quality control.

Development of standardized documentation for recording and reporting results will facilitate the availability and comparability of data and documentation over a long period, which is normally required to track the performance of a residue repository.

9. CASE STUDY OF RADON EXHALATION FROM A URANIUM RESIDUE REPOSITORY

9.1. INTRODUCTION

This section presents a case study of multiparametric measurements carried out on a uranium waste repository at Jaduguda, India. The residues produced at the uranium mill at Jaduguda are segregated into coarse and fine fractions at the ore processing facility before disposal. The coarse fraction is used for backfilling the mine, while the fine fraction (slurry form) is placed into a tailings pond. The residues consolidate and form a compact mass of low permeability. These conditions favoured the use of a diffusion based model for predicting radon releases.

This study illuminates some of the results presented in the previous sections, by comparing predicted flux by means of empirical equations to direct measurements using an accumulator.

The radon flux densities were predicted using two approaches, namely:

(1) Using in situ measured values for the radon emanation coefficient (E) and radon diffusion length in the residues;

(2) Using the empirical correlations given for the radon diffusion coefficient (D_r (m^2/s)) in Ref. [51] and for the radon emanation coefficient (E) in Ref. [68].

9.2. MEASURING RADON FLUX DIRECTLY AT THE RESIDUE SURFACE

In situ radon fluxes from the residue surface were measured using the accumulator technique. The set-up (Fig. 18) consisted of a cylindrical accumulator (15 cm in diameter and 15 cm in height) attached to a continuous radon monitor.

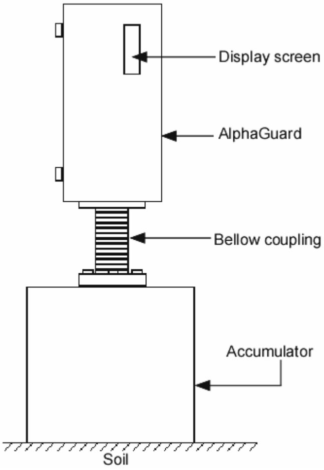

FIG. 18. Accumulator with AlphaGuard continuous radon monitor set up for radon flux measurement.

The effective volume of this measurement system is 3.4 L. The accumulator was inserted into the residue surface to a depth of approximately 1 cm to prevent superficial leakage. Radon concentrations inside the accumulator were monitored at 10 min time intervals for about 3 hours at each location. The concentration data were fitted to an exponential growth model to simultaneously obtain the in situ radon flux and diffusion coefficient by following the methods outlined in Sections 3.2 and 5.3.1.

Typical data on the buildup of radon concentration in the accumulator deployed at one location at the site are shown in Fig. 19.

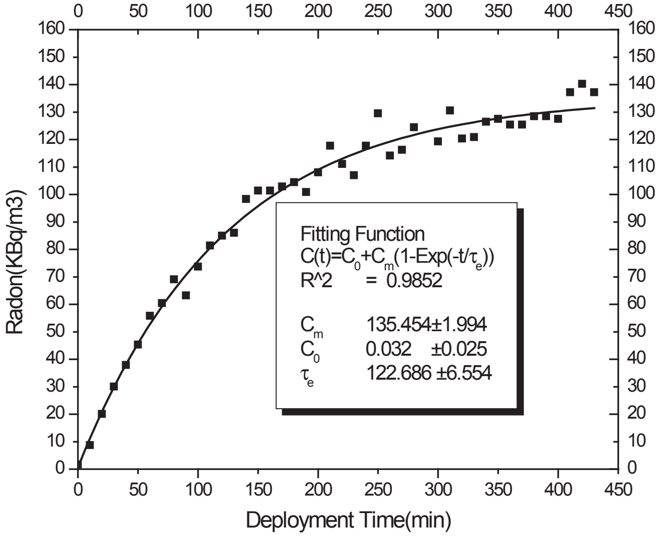

FIG. 19. Typical buildup data of radon concentration in the chamber with least square fitting of Eq. (16) in Section 5.3.1.2. for an accumulator of radius a = 7.5 cm and effective volume V = 3.4 L deployed on the uranium residue repository.

9.3. ESTIMATING FLUX BY MEASUREMENT OF L_r AND E

Residue samples were collected from 40 locations of the residue pile by inserting a 1000 cm^3 stainless steel cylindrical tube into the residue matrix. The samples were processed and dried in an oven at 110°C to attain constant weight. Then, the samples were packed in a 300 cm^3 leakproof plastic container and stored for approximately 30 d to ensure secular equilibrium between ^{226}Ra and its decay products. Gamma spectrometry [115] was used to measure the dry weight specific ^{226}Ra content in the samples. The samples were subjected to radon mass exhalation rate measurement [94] as outlined in Section 5.3.5. The dry sample radon emanation coefficient was then determined by the equation:

$$E = J_{\mathrm{m}}/\lambda R \tag{28}$$

where

E is the dry sample radon emanation coefficient;
J_m is the mass exhalation rate (Bq·kg^{-1}·s^{-1});
R is the dry weight radium activity concentration (Bq/kg);

and λ is the decay constant of ^{222}Rn (s^{-1}).

The soil porosity was estimated using the equation:

$$n_T = 1 - \rho_b/\rho_g \tag{29}$$

where

n_T is the soil porosity;
ρ_b is the measured dry bulk density of the residue (kg/m^3);

and ρ_g is the grain density of the residue material (2700 kg/m^3) [116].

The moisture saturation fraction (*m*), the fraction of the pore space filled with water, was estimated from the measured soil moisture as $m = \rho_b\theta_d/100\rho_w n_T$ where θ_d is the moisture content on a dry weight basis and ρ_w is the density of water (kg/m^3). The in situ temperature *T* at a depth of 10 cm from the residue surface was measured by a sensor.

9.3.1. In situ radon diffusion length (L_r)

The in situ radon diffusion lengths were determined by analysing the depth profile of radon concentration in the residue matrix. The schematic diagram of the measurement set-up is shown in Fig. 20.

Radon concentrations were measured at different depths from 20 to 150 cm, using a soil probe with a flow rate of approximately 0.5 L/min. According to diffusion theory, the radon concentration *C*(*z*) at depth *z* follows the equation [19]:

$$C(z) = C_\infty\left(1 - \exp\left(-z/L_r\right)\right) \tag{30}$$

where

C_∞ is the radon concentration at great depth (Bq/m^3);
z is the thickness of the material (m);

and L_r is the radon diffusion length (m).

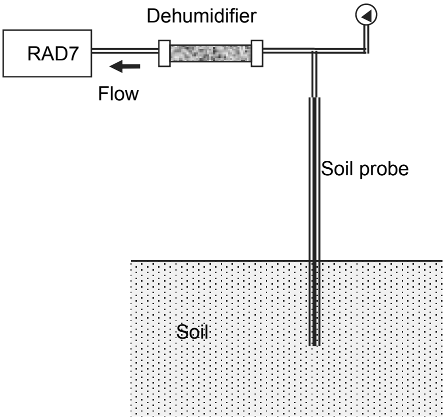

FIG. 20. Schematic diagram of soil probe method for the measurement of radon concentration in residues using a continuous radon monitor.

At the end of the experiment, the radon concentration data at different depths were fitted to Eq. (30) by the method of least squares and the in situ radon diffusion length was obtained from the fitting coefficients. A typical fit to the data is shown in Fig. 21.

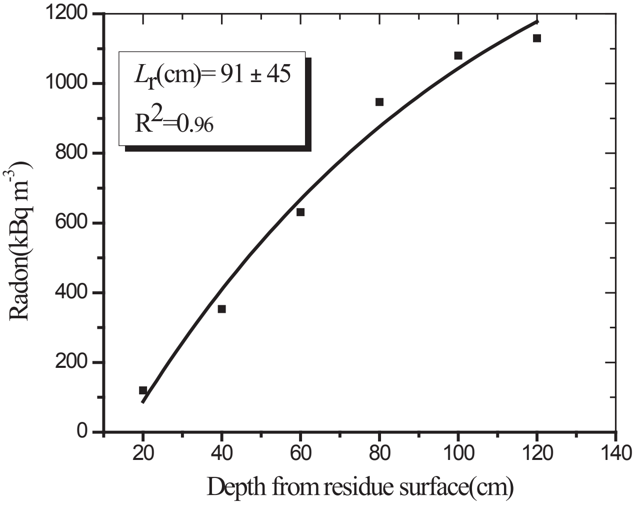

FIG. 21. Depth profile of radon concentration in soil gas and the fitted curve.

9.3.2. In situ radon emanation coefficient (*E*)

This process was performed along the lines outlined in Section 3.1. The in situ radon emanation coefficient (*E*) in the residue was determined by measuring the mass exhalation rates using fresh samples without subjecting them to drying. These experiments were performed at the site itself immediately after collecting the samples and hence the results are expected to be close to the values corresponding to in situ moisture conditions.

9.3.3. Estimation

According to Ref. [116], the radon flux at the residue surface may be written as:

$$f_r = \lambda L_r R \rho_b E \tag{31}$$

where

f_r is the radon flux ($\mathrm{Bq\cdot m^{-2}\cdot s^{-1}}$);
λ is the decay constant of ^{222}Rn ($\mathrm{s^{-1}}$);
R is the radium activity concentration in uranium residue (Bq/kg);

and ρ_b is the bulk density of residues ($\mathrm{kg/m^3}$).

9.4. ESTIMATING FLUX BY CALCULATING L_r AND E VIA EMPIRICAL FORMULAS

The third, and least exact, approach to estimating the radon flux at the surface uses Eq. (31), but, instead of using experimentally measured values of L_r (and thus D_r) and *E*, it uses empirical relations for D_r and *E*. The diffusion coefficient was estimated using Eq. (14), including a temperature correction:

$$D_r = D_{MA} n_T \exp(-6mn_T - 6m^{14n_T}) \left(\frac{T}{273}\right)^{0.75} \tag{32}$$

The following empirical equation was employed to estimate *E*:

$$E = E_0\left[1 + 1.85\left(1 - \exp\left(-18.8m\right)\right)\right] \tag{33}$$

The variables in the above equations are defined as follows:

D_{MA} is the radon diffusion coefficient in air at ambient temperature and pressure (1.1×10^{-5} m^2/s);
n_T is the soil porosity;
m is the fraction of pore space filled with water;
T is the absolute temperature (K);

and E_0 is the radon emanation coefficient for uranium residue in dry conditions.

Once these equations were computed, they were simply substituted into Eq. (29) to generate the flux estimate.

9.5. ILLUSTRATIONS OF ESTIMATION AND MEASUREMENT RESULTS

9.5.1. Determination of exhalation flux

Table 5 presents a summary of various parameters measured for the repository. Figure 22 shows the radon fluxes measured and predicted (by the two approaches mentioned above) over 10 locations of the residue. There is an overall agreement, within deviations of approximately 20%, between the measured and the predicted radon fluxes. A closer statistical analysis (not shown) indicates that the radon fluxes predicted using the in situ diffusion lengths and emanation coefficients are closer to the measured values than the fluxes predicted from the moisture corrected empirical functions given in Eqs (32) and (33). This could be due to the fact that the moisture content estimated from the top layer of the repository may not represent values down to a depth of approximately 1 m, thereby affecting the parameter values based on empirical correlations.

9.5.2. Determination of radon diffusion length

Sometimes, it is of interest to estimate the diffusion length, given a known exhalation flux. For this purpose, the relevant equations can be reversed and the known values of exhalation flux can be used to estimate *Lr* using measurement based and empirical formulas.

TABLE 5. THE MEAN OF MEASURED RESIDUE PARAMETERS INCLUDING STANDARD DEVIATION IN WINTER, SUMMER AND RAINY SEASONS FOR 40 LOCATIONS OF THE URANIUM WASTE RESIDUE REPOSITORY

Parameters	Winter	Summer	Rainy	Average
Radium, R (Bq/kg)	5124 ± 2348	5144 ± 2256	5081 ± 2244	5166 ± 2282
In situ ^{222}Rn diffusion length, L_r (m)	0.81 ± 0.15	0.90 ± 0.14	0.72 ± 0.14	0.81 ± 0.14
In situ ^{222}Rn emanation coefficient, E	0.24 ± 0.05	0.20 ± 0.02	0.28 ± 0.07	0.24 ± 0.05
Dry ^{222}Rn emanation coefficient, E_0	0.09 ± 0.04	0.10 ± 0.03	0.09 ± 0.03	0.09 ± 0.03
Dry bulk density, ρ_b (kg/m^3)	1790 ± 66	1786 ± 52	1801 ± 37	1792 ± 52
Total porosity, n_T	0.34 ± 0.02	0.34 ± 0.03	0.33 ± 0.04	0.34 ± 0.03
Moisture saturation fraction, m	0.54 ± 0.05	0.35 ± 0.04	0.74 ± 0.06	0.54 ± 0.05

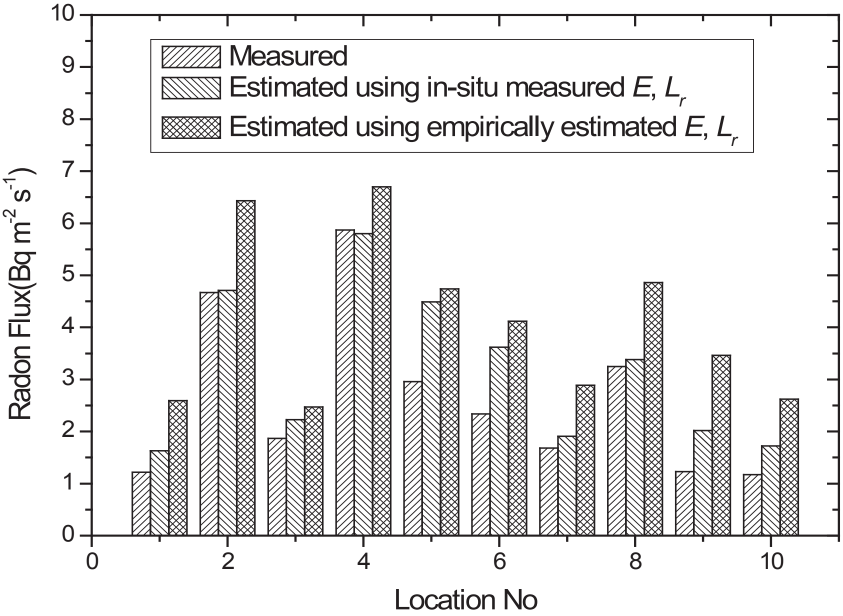

FIG. 22. Comparison between measured and predicted radon flux densities at 10 locations.

Figure 23 shows the comparison of radon diffusion lengths obtained from the three methods: the accumulator method, the depth profile analysis of soil gas radon and the empirical relation given in Eq. (32). Overall agreement between the three methods was within 20%. The diffusion lengths estimated from the empirical correlation method were generally higher than the other two, thereby supporting the moisture content effect mentioned in Section 9.5.1. The study also demonstrates that the accumulator method is a useful tool, not only for deriving exhalation flux, but also for estimating the in situ diffusion coefficient in soil.

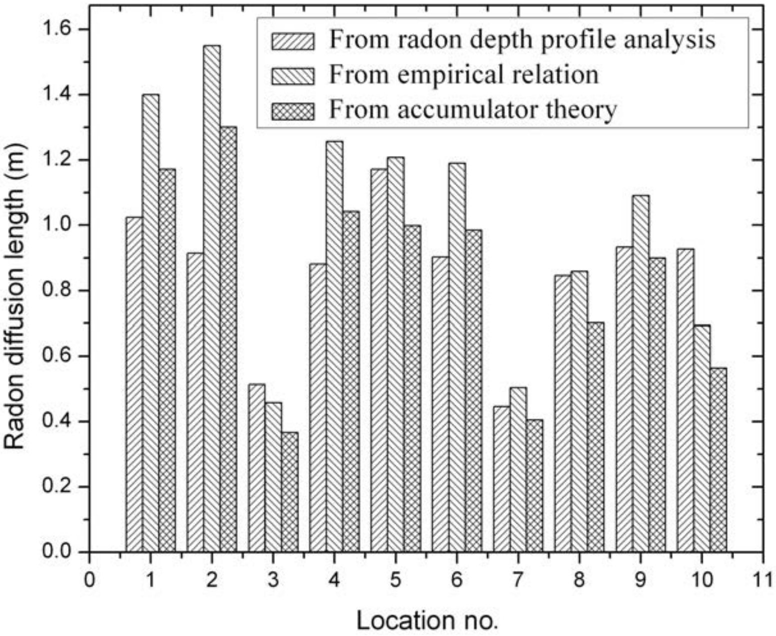

FIG. 23. Comparison of radon diffusion length in uranium residue matrix obtained by three different methods at 10 locations.

9.5.3. Relating radium content to radon flux

Separately from the above analysis, Fig. 24 shows the relationship between radium content and the seasonally averaged radon fluxes at over 40 locations on the repository. The linear fit shows a strong correlation between radium content and radon flux in the repository. The slope yields a ratio of (8.3 ± 0.4) × 10^{-4} kg·m^{-2}·s^{-1}. This ratio also represents the product of the radon decay constant λ (2.1×10^{-6}) and average values of the radon diffusion length (L_r), the soil density (ρ_b) and the in situ radon emanation coefficient (E). Using the measured value of the above parameters (Table 5), this ratio works out to be 7.3×10^{-4} kg·m^{-2}·s^{-1}. The slope $(8.3 \pm 0.4) \times 10^{-4}$ kg·m^{-2}·s^{-1} may be used as a conversion factor to quickly estimate the radon flux (Bq·m^{-2}·s^{-1}) using the ^{226}Ra content (Bq/kg) for the uranium waste residue repository at Jaduguda.

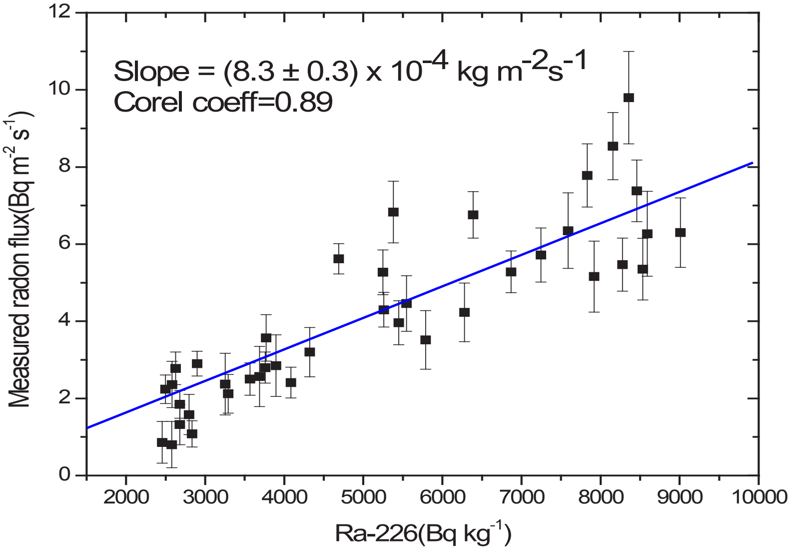

FIG. 24. Correlation between measured radon flux densities and ^{226}Ra content at 40 locations.

Appendix I

MATHEMATICAL DEVELOPMENT OF RADON DIFFUSION EQUATIONS

I.1. FORMULATION OF RADON DIFFUSION EQUATION FOR A POROUS MEDIUM

Consider a matrix in which radon is continuously released to the pore volume of the matrix owing to emanation from the grains containing Ra. Let P_p be the radon activity released into the unit volume of the pore space per unit time. A local flux density f_p (a vector quantity) may be defined representing the activity of radon crossing per unit pore area per unit time. Let C_p be the pore space radon concentration (i.e. radon activity divided by volume of pore space) at any point at a given time. The general transport equation is obtained by a limiting process of the time rate of change of radon activity in an infinitesimal pore volume as being due to the difference between the generation rate and losses due to leakage rate and radioactive decay (with decay constant λ) [19]:

$$\frac{\partial C_p}{\partial t} = P_p - \nabla \cdot f_p - \lambda C_p \tag{34}$$

In general, f_p has two components: (a) pressure driven advection flux and (b) gradient driven diffusion flux. Generally, in most situations, the pressure difference sustained between the pore air and the atmosphere is too small to cause significant pressure induced flux as compared with diffusion driven flux and, hence, it is neglected. For a one dimensional system, the diffusion flux is expressed in terms of the concentration gradient by applying Fick's first law of diffusion as follows:

$$f_p = -D \frac{\partial C_p}{\partial z} \tag{35}$$

where D is the radon diffusion coefficient in the pore space in the matrix. In a completely dry material, D is the product of the molecular diffusion coefficient (D_{MA}) in the pore space air and tortuosity (τ) that accounts for the tortuous paths traversed by the radon atoms along the pores [117]. However, in general, residues contain some moisture and thus radon atoms will diffuse both in the air and water phases resulting in a lower diffusion coefficient (D) in the pore space [51].

Substitution of Eq. (35) into Eq. (34) yields the following diffusion equation:

$$\frac{\partial C_p}{\partial t} = P_p + D\frac{\partial^2 C_p}{\partial z^2} - \lambda C_p \tag{36}$$

If R is the radium activity concentration of the matrix (i.e. the radium activity in the soil grain divided by the mass of the matrix), ρ_b is the bulk density of the matrix (i.e. the mass of the matrix divided by the bulk volume of the matrix), E is the radon emanation coefficient and n_T is the total porosity of the matrix (i.e. the total pore volume divided by the bulk volume), then P_p can be expressed as:

$$P_p = \frac{\lambda R \rho_b E}{n_T} \tag{37}$$

With this, Eq. (36) becomes:

$$\frac{\partial C_p}{\partial t} = D\frac{\partial^2 C_p}{\partial z^2} - \lambda C_p + \frac{\lambda R \rho_b E}{n_T} \tag{38}$$

While the flux f_p in Eq. (35) refers to unit pore area, the exhalation flux at the residue surface refers to the bulk area that includes both the pore area and the area covered by the solid materials. Assuming the fractional pore area at the surface is the same as the fractional pore volume of the bulk of the residue, the exhalation flux density may be related to the pore space concentration as follows:

$$f = n_T f_p = -n_T D\frac{\partial C_p}{\partial z} \tag{39}$$

In dry residues, the pores will be filled only with air and, hence, C_p will be the same as the concentration C_a in pore air. In the case of residues containing moisture, the radon atoms in the pore volume will be distributed partly in air and partly in water in proportion to the water–air partition coefficient of radon. It is more convenient to recast the diffusion equation in terms of C_a to develop consistent formulations and interface boundary conditions to address multilayer problems involving covers of different moisture contents and material properties. If C_w and C_a denote the radon concentration in water and in air, and if n_w and

n_a are water filled and air filled porosities respectively, then mass conservation [117] requires that:

$$n_T C_p = n_a C_a + n_w C_w \tag{40}$$

The law of equilibrium partitioning relates C_w to C_a as follows:

$$C_w = K C_a \tag{41}$$

where K is the water–air partition coefficient. Combining Eq. (41) with Eq. (40) gives:

$$n_T C_p = C_a (n_a + K n_w) = n_e C_a \tag{42}$$

where

$$n_e = (n_a + K n_w) \tag{43}$$

The quantity n_e denotes the partition corrected porosity. It may be expressed more conveniently in terms of the volumetric moisture saturation (m), defined as $m = n_w / n_T$, as follows :

$$n_e = n_T \, [1 - (1 - K) m] \tag{44}$$

With this, Eq. (38) may be recast in terms of C_a as follows:

$$\frac{\partial C_a}{\partial t} = D \frac{\partial^2 C_a}{\partial z^2} - \lambda C_a + \frac{\lambda R \rho_b E}{n_e} \tag{45}$$

Under steady state conditions, Eq. (45) becomes:

$$D \frac{\partial^2 C_a}{\partial z^2} - \lambda C_a + \frac{\lambda R \rho_b E}{n_e} = 0 \tag{46}$$

The corresponding expression for flux density is:

$$f = -n_e D \frac{\partial C_a}{\partial z} \tag{47}$$

The formulation of the diffusion equation in terms of radon concentration in the air phase (C_a) and partition corrected porosity (n_e) enables a direct application to a multilayer problem since both C_a and f will satisfy continuity requirements at the interface of the two layers even if their moisture content and porosities differ from one other.

I.2. GENERAL SOLUTION AND BOUNDARY CONDITIONS

The general solution to Eq. (46) is:

$$C_a(z) = A\exp(z/L) + B\exp(-z/L) + (R\rho_b E/n_e) \tag{48}$$

where

$$L = \sqrt{\frac{D}{\lambda}} \tag{49}$$

and is known as the diffusion length of radon in the given matrix. *A* and *B* are unknown coefficients to be determined using the boundary conditions of the problem.

The associated radon flux density, obtained from Eqs (47) and (48), is:

$$f = \frac{n_e D}{L}\left[B\exp(-z/L) - A\exp(z/L)\right] \tag{50}$$

For simplicity of notation in multilayer problems, it is convenient in Eq. (48) to drop the subscript '*a*' denoting the radon concentrations in the air of the pore space and replace it with layer (i = 1, 2, 3…) specific radon concentrations. Hence, the radon concentration in the air space of the ith layer will be denoted by C_i. Similarly, the subscript '*e*' may be dropped from the partition corrected porosity since this is the only porosity that actually appears in all the expressions. With this, the boundary conditions of the problem may be stated as:

(a)	$f(0) = 0$	the flux is zero at the base of the residues (impervious base);
(b)	$C_i(z_i) = C_{i+1}(z_i)$	the concentration is continuous across the interface at z_i;
(c)	$f_i(z_i) = f_{i+1}(z_i)$	the flux is continuous across the interface at z_i;

(d) $C_n(z_n) = C_{atm}$ the concentration at the surface of the top layer (medium n) is equal to a specified value, usually the atmospheric concentration.

I.3. SOLUTION FOR RADON FLUX ACROSS THE SURFACE OF BARE RESIDUES

The application of boundary condition (a) to Eq. (50) yields $A = B$. The application of boundary condition (d) for $C_{atm} = 0$ at the top surface of the residues of thickness z_r yields values for A as a function of z_r.

Finally, the flux at the surface of the bare residues is given by:

$$f(z_r) = f_r = R\rho E L_r \lambda \tanh\left[\frac{z_r}{L_r}\right] \tag{51}$$

where subscript ‘r’ refers to residues.

I.4. SOLUTION FOR COVERED RESIDUES

The solution of the diffusion equation for a two region problem applies to a residue pile covered with a homogeneous material. For simplicity, the source term in the cover is assumed to be zero, and C_{atm} at the surface of the cover is also assumed to be zero.

Application of the boundary conditions yields values for A_c and B_c as functions of z_c, where the subscript ‘c’ refers to the cover material. Substitution of A_c and B_c into Eq. (50) yields the flux as a function of the position z in the cover. Finally, the flux at the surface of the cover is given by substituting $z = z_c$:

$$f = \frac{2 f_r \exp\left[-\frac{z_c}{L_c}\right]}{\left[1 + \frac{n_r L_r}{n_c L_c}\tanh\frac{z_r}{L_r}\right] + \left[1 - \frac{n_r L_r}{n_c L_c}\tanh\frac{z_r}{L_r}\right]\exp\left[-2\frac{z_c}{L_c}\right]} \tag{52}$$

The terms n_i and L_i account for moisture, porosity and diffusion coefficient differences at the residue–cover interface. If the moisture and porosity differences are small, the residue and cover diffusion coefficients are approximately equal and residue thickness is large compared with z_c, then Eq. (52) becomes:

$$f_c \approx f_r \exp\left[-\frac{z_c}{L_c}\right] \tag{53}$$

This implies that the free surface flux decreases exponentially as the thickness of the cover increases. This relationship is useful for a quick estimation of the cover thickness required to achieve a given exhalation flux density.

Appendix II

REFINED METHODS OF RADON FLUX CALCULATION

The equations in Section 6 (simplified expressions for estimating radon exhalation flux density) can be modified to increase the accuracy of the calculations and extend their range of application. This is shown here for the cases of uncovered and single layer systems. Exact analytical solutions for up to four layer systems have been developed [118–120], but they are cumbersome to use. In general, multilayer systems are addressed by solving the differential equations directly using numerical models.

II.1. FLUX CALCULATION FOR UNCOVERED LAYERED RESIDUES

Some residue situations require that the vertical heterogeneities in a residue pile be considered explicitly. Such heterogeneity may be due to major variations in the radium concentration or in the diffusion coefficient as a function of depth. The heterogeneities are considered explicitly by dividing them into layers that have homogeneous properties within each layer. Then, each layer is both a source layer and a cover layer to layers beneath it. Combining Eqs (51) and (53) gives, for the flux at the surface of a pile with *n* layers:

$$f_r = \sum_{i=1}^{n} R_i \rho_i E_i \lambda L_{ri} \tanh\left[\frac{z_{ri}}{L_{ri}}\right] \exp\left[-\sum_{j=i+1}^{n} \frac{z_{ri}}{L_{ri}}\right] \tag{54}$$

where

$$L_{ri} = \sqrt{\frac{D_{ri}}{\lambda}} \tag{55}$$

is the diffusion length of radon in the *i*th layer of the residue material.

In the formula given above, the following assumptions have been made:

(a) For a given layer, the layer beneath it is treated as impervious to radon;
(b) The approximation given in Eq. (53) is used to represent the effect of a cover layer above the given residue layer.

For example, consider a three layer residue pile. The properties of the pile are described in Table 6. The layers are heterogeneous both in depth and in diffusion coefficient (D_r). An exact calculation performed with the RAECOM [120] computer code yields a randon flux density (f_r) of 17 Bq·m^{-2}·s^{-1}. Using these same variables in Eq. (54) yields an f_r of 18 Bq·m^{-2}·s^{-1}. However, if the D_r were deemed homogeneous throughout the three layers using the D_r of the middle layer, the resulting f_r would be 12 Bq·m^{-2}·s^{-1}. If the D_r were deemed homogeneous with the top layer value, the resulting f_r would be 17 Bq·m^{-2}·s^{-1}. This highlights the higher influence of the properties of the top layer of residue on the f_r of the pile.

TABLE 6. PROPERTIES OF AN EXAMPLE THREE LAYER RESIDUE PILE

Layer	Layer depth (m)	Diffusion coefficient (D_r)	Radium activity concentration (Bq/kg)	Emanation coefficent (E)	Density (kg/m^3) (ρ)	Radon flux density (f_r)
All three layers	6		4×10^4	0.2	1.5×10^3	
Top	1	1×10^{-6} m^2/s				18 Bq·m^{-2}·s^{-1}
Middle	2	5×10^{-7} m^2/s				
Bottom	3	1×10^{-7} m^2/s				
Layers homogeneous with middle D_r	6	5×10^{-7} m^2/s				12 Bq·m^{-2}·s^{-1}
Layers homogeneous with top D_r	6	1×10^{-6} m^2/s				17 Bq·m^{-2}·s^{-1}

For calculating ^{220}Rn fluxes from thorium residues, the properties of the top layer (a few centimetres deep) are sufficient.

II.2. FLUX CALCULATION FOR A SINGLE COVER

Apart from the thickness of the cover material, the properties of the cover and the residue matrices have an important bearing on the radon flux reduction factor. This is illustrated below.

The exact solution of the radon diffusion equation for a single cover in Eq. (52) is:

$$f_c = \frac{2f_r \exp\left[-\frac{z_c}{L_c}\right]}{\left[1+\frac{n_r L_r}{n_c L_c}\tanh\frac{z_r}{L_r}\right]+\left[1-\frac{n_r L_r}{n_c L_c}\tanh\frac{z_r}{L_r}\right]\exp\left[-2\frac{z_c}{L_c}\right]} \tag{56}$$

where

$$n_i = n_{Ti}\left[1-(1-K)m_i\right],\ i = r \text{ or } c \tag{57}$$

denotes the partition corrected porosity for residue *r* or cover *c*, and

$$L_i = \sqrt{\frac{D_i}{\lambda}},\ i = r \text{ or } c \tag{58}$$

denotes the radon diffusion length in residue *r* or cover *c*.

K denotes the radon distribution coefficient between water and air (about 0.26 at 20°C), and m_i denotes the volume fractional moisture saturation in residues or cover.

If the residue pile thickness is much greater than the diffusion length of radon, then the hyperbolic tangent term is unity and Eq. (56) becomes:

$$f_c = \frac{2f_r \exp\left[-\frac{z_c}{L_c}\right]}{1+\frac{n_r L_r}{n_c L_c}+\left[1-\frac{n_r L_r}{n_c L_c}\right]\exp\left[-2\frac{z_c}{L_c}\right]},\ z_r >> z_c \tag{59.a}$$

If the moisture and porosity differences between the residue and cover layers are small and if corresponding diffusion coefficients are approximately equal, Eq. (59.a) further reduces to:

$$f_c = f_r \exp\left[-\frac{z_c}{L_c}\right] \tag{59.b}$$

which is the simple exponential attenuation shown by A in Fig. 25.

However, if $n_r L_r$ is much less than $n_c L_c$, Eq. (56) becomes:

$$f_c = \frac{2f_r \exp\left[-\frac{z_c}{L_c}\right]}{1+\exp\left[-2\frac{z_c}{L_c}\right]} \tag{60}$$

If the value of z_c is low, the value of f_c is approximately equal to f_r and very little flux attenuation occurs, as shown by B in Fig. 25. However, as z_c increases, f_c decreases exponentially in the same manner as in A, but retains twice the magnitude.

The case in which $n_r L_r$ is much greater than $n_c L_c$ is shown as C in Fig. 25. In this case, a thin cover is relatively effective; f_c again decreases, but its magnitude is $2(n_c L_c / n_r L_r)$ times the value in curve A. However, in most situations, it is very difficult to maintain a cover with an $n_c L_c$ value less than the $n_r L_r$ value of the residue. Hence, this condition has limited application.

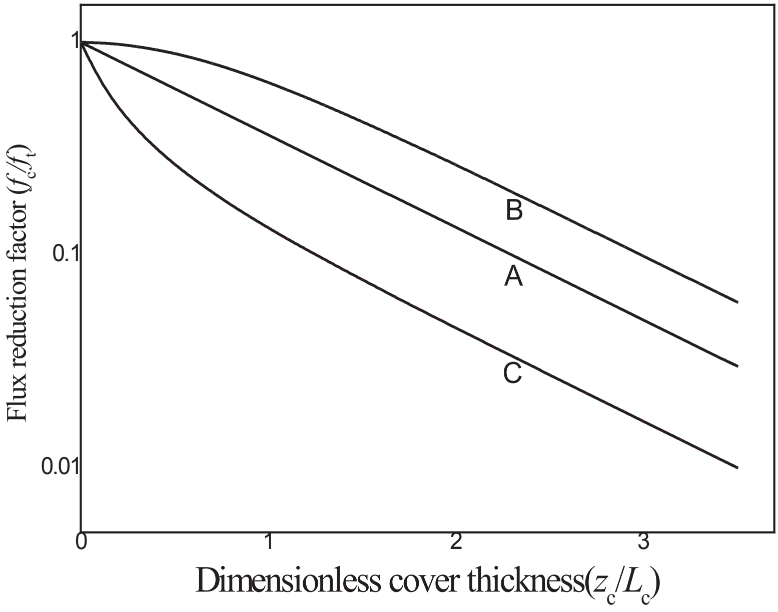

FIG. 25. Radon flux reduction factor for various cover thicknesses. (A: $n_r L_r = n_c L_c$; B: $n_r L_r << n_c L_c$; C: $n_r L_r >> n_c L_c$).

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NOTATION

A	surface area of an opening (m^2)
a	chamber radius (m)
C	radon concentration (Bq/m^3)
D	radon diffusion coefficient (m^2/s)
D_M	molecular diffusion coefficient (m^2/s)
D_{MA}	molecular diffusion coefficient in air (m^2/s)
D_{MW}	molecular diffusion coefficient in water (m^2/s)
D_e	effective (bulk) diffusion coefficient (m^2/s)
D_r	diffusion coefficient
E	emanation coefficient
ε	counting efficiency of the system relative to the activity of radon adsorbed (counts·s^{-1}·Bq^{-1})
f	radon flux density (Bq·m^{-2}·s^{-1})
f_p	local flux density representing the activity of radon crossing per unit pore area per unit time (Bq·m^{-2}·s^{-1})
F	total radon exhalation flux (Bq/s)
G	average ore grade
H	height (m)
J_m	mass exhalation rate (Bq·kg^{-1}·h^{-1})
K	water–air partition coefficient
L_r	radon diffusion length (m)
λ	decay constant (s^{-1})
λ_e	effective decay constant for ^{222}Rn, which is the sum of the leak rate (if existing) and the decay constant (s^{-1})
M	mass (kg)
m	moisture saturation fraction (vol./vol.)
N	net count rate, after background subtraction, obtained during the counting period t_C (counts/s)
n	porosity
n_s	soil porosity
P	radon production rate (Bq·m^{-3}·s^{-1})
P_p	radon activity released into the unit volume of the pore space per unit time (Bq·m^{-3}·s^{-1})
R	radium activity concentration (Bq/kg)
ρ_b	bulk density (kg/m)
ρ_g	grain density (kg/m)
ρ_w	density of water (kg/m)
T	absolute temperature (K)
t	measurement time (s)

t_c	counting period
t_d	delay period from the end of exposure to the beginning of the counting interval (s)
t_e	period of exposure of the charcoal in the canister (s)
τ	tortuosity factor
τ_e	time constant of radon buildup
v	flow rate (m^3/s)
V	effective volume (m^3)
w	dilution factor (kg of solid waste for kg of uranium ore processed)
z	depth or thickness of the material (m)
θ_d	moisture content on a dry weight basis (vol./vol.)

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