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TECHNICAL NOTE

METHODOLOGY STUDY ON EVALUATION OF RADON FLUX FROM SOIL IN CHINA

Qiuju Guo[1,*], Kainan Sun[2] and Jianping Cheng[2]
[1]Department of Technical Physics, School of Physics, Peking University, Beijing 100871, P. R. China
[2]Department of Engineering Physics, Tsinghua University, Beijing 100084, P. R. China

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Radon flux rate from soil is one of the most important factors for the evaluation of environmental radon levels. The objective of our study is to make a rough estimation of the nationwide radon flux rate from soil in China. Considering the applicability to the large area and complex distribution of soil types in China, a simple model was established. To test the model, field measurements on radon flux rate totally in 47 points from different areas were carried out from spring to summer in 2003. Laboratory experiments of each soil sample on related soil properties, such as radium contents, water contents, soil dry bulk density and soil texture were performed simultaneously. Approximately 30% of the samples had very consistent calculated values with their measured values, their relative errors were less than 0.25. In spite of the large uncertainties in the model which is influenced by so many factors, the considerable validation of the model can be shown. Based on the discussion of distribution of the errors as well as the reasons causing the errors, a trial modification of the model was made. A modification function looked necessary when soil water content was <10%.

INTRODUCTION

Over the past few decades, there has been a large scientific interest in the study of environmental radon. One of the main reasons is its associated health hazard, another is its widespread use as an environmental tracer(1).

Soil is a source of radon. The infiltration of radon gas (^{222}Rn) from soil has been identified as one of the main mechanisms influencing indoor radon levels in many buildings. It was reported that a worldwide average of 60.4% of indoor radon comes from the ground and the surrounding soil of buildings(2). Information on the spatial variability of radon exhalation rate would be useful for identifying areas with a risk of high radon exposure. On the other hand, the well-understood chemical behaviour (inert gas) of ^{222}Rn and its convenient half-life (3.82 d) make radon a useful tracer in the studies of air mass transportation. For example, it is often used in validating global atmospheric transport models(3,4).

Several studies on mapping radon exhalation rate from soil have been reported in local, regional or national scale with different methods. For example, Ielsch *et al.*(5) proposed a methodology based on radon exhalation rate quantification, starting from a precise characterisation of the main local geological and pedological parameters. In China, because of the large area and complex distribution of soil types, a large number of field measurements on a national scale is extremely difficult to perform. Therefore, it is essential to develop an effective, economical and practical method for the classification and mapping of radon exhalation rate from soil in China. In this work, a simple model was established based on soil radon diffusion theory, which included the influences of soil radium content, emanation coefficient, soil porosity and moisture saturation. To test the model, both field measurements on radon flux rate and laboratory experiments on related soil properties, such as soil radium content, soil texture, soil porosity and water content were carried out totally in 47 points from different areas (but most of them were in Beijing area). The distribution of the model errors as well as the reasons causing the errors were discussed, and a trial modification was made in the original model to improve the consistency of calculated radon flux rate values with measured ones, especially for the samples in which soil water content was <10%.

MATERIALS AND METHODS

A simple model for estimating radon flux rate from soil

Since the influence of the most meteorological parameters on advection is usually temporary, instantaneous and difficult to be modelled (6,7), only diffusion mechanism is considered in this work for estimating long-term averaged values. Based on the conservation of mass, an idealised, one-dimensional, steady-state model for the transportation and distribution of radon in soil can be expressed as the following differential equation:

$$D_e \frac{d^2C}{dx^2} - \lambda C + \frac{A}{p_{\mathrm{eff}}} = 0, \qquad (1)$$

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[*] Corresponding author: qjguo@pku.edu.cn

where D_e is the effective radon diffusion coefficient ($m^{-2}\ s^{-1}$); C represents the radon concentration in pore air of the soil ($Bq\ m^{-3}$); x is the distance from the ground surface with its positive direction downward (m); λ is the radon decay constant (s^{-1}); p_{eff} is the effective porosity of the soil; and A is defined as Equation 2 is the production rate of radon gas into the pore space.

$$A = \lambda\rho RE, \tag{2}$$

where ρ represents the dry bulk density of the soil ($kg\ m^{-3}$); R is the ^{226}Ra activity concentration in the soil particles ($Bq\ kg^{-1}$); and E is the radon emanation coefficient.

Given the boundary conditions: $C|_{x\to 0} = 0$ and $C|_{x\to\infty}$ is finite, the solution for Equation 1 is:

$$C = \frac{A}{\lambda p_{\mathrm{eff}}}\left[1 - \exp\left(-\frac{\sqrt{\lambda D_e}}{D_e}x\right)\right]. \tag{3}$$

On the other hand, as radon flux is continuous at the ground surface between soil and atmosphere, the flux rate (F) of radon from ground surface can be expressed as follows:

$$F = p_{\mathrm{eff}} D_e \left.\frac{\mathrm{d}C}{\mathrm{d}x}\right|_{x=0}. \tag{4}$$

Combined into Equation 3, Equation 4 can be rewritten as:

$$F = \sqrt{\lambda D_e}\rho ER. \tag{5}$$

It is known that the effective radon diffusion coefficient (D_e) in soil has been experimentally studied, which can be expressed as the following equation(8):

$$D_e = pD_0 \exp\left(-6mp - 6m^{14p}\right), \tag{6}$$

where p is the total soil porosity which can be estimated by Equation 7(9); D_0 is the radon diffusion coefficient in open air with a constant of 1.1×10^{-5} $m^{2-}\ s^{-1}$; and m is the volume fraction of water saturation which can be calculated from water mass content (w) by using Equation 8.

$$p = (93.947 - 32.995\rho)/100. \tag{7}$$

$$m = wp/1000p. \tag{8}$$

The above equations indicate that the radon flux rate can be calculated when the parameters of R, w, ρ and E in the soil are known.

To test the model, the parameters R, w, ρ of each soil sample in all the field measurement points were measured in our laboratory. For the parameter of the radon emanation coefficient (E), reported typical radon emanation coefficients for different types of soil texture(10) were referenced, and Table 1 shows some related data.

Table 1. Typical values of radon emanation coefficient for different soil textures.

Soil types	Emanation coefficient
Sand	0.14
Sandy loam	0.21
Loam	0.24
Silty loam	0.25
Clay	0.28

Field measurements

Field measurements of radon flux rate from soil were carried out in totally 47 points from different soil texture in Beijing, Guiyang (Guizhou province) and Huhhot (Inner Mongolian Municipality) from June to August, 2003. The majority of the points were around Beijing area. The measurement points were usually selected at farms, gardens or schools that were representative of soil texture in the local area. At each point, the radon flux rate was measured and at least two soil samples were taken by an annular steel sample container. Then all the soil samples were taken back to the laboratory for the analysis and measurement on relative soil parameters.

The radon flux rate measurements were performed with the device ERS-2 (TRACERLAB company, Germany), which is an Electrostatic-Radon-Sampler for the determination of the ^{222}Rn gas concentration and for the determination of the flux rate. The ERS-2 was operated with an Alpha-Spectroscopy detector and multichannel analyser (MCA) with 256 channels. For the determination of the radon flux rate, the ERS-2 was placed on the soil surface with sealed condition. Usually, at each point, the measurement was performed for 4–5 cycles and the cycle time was 10 min. The ERS-2 gave the radon gas concentration of each cycle in the unit of $Bq\ m^{-3}$. The radon concentration data for each cycle were automatically stored in the memory of ERS-2. To calculate the flux rate, first the data in ERS-2 were read into an external PC-system, and then the data were linear simulated to derive the flux rate with the result of $mBq\ m^{-2}\ s^{-1}$ by using the Tracerlab-Spectrum-Software. Figure 1 shows the schematic diagram of ERS-2.

The radium content of each soil sample was measured using a high purity germanium (HPGe) detector with a relative efficiency of 48.3% ($E_\gamma = 1.33$ MeV). First, each soil sample was baked for ~24 h at a constant temperature of 105 ± 2°C in an oven. Then the dry sample was grind and sifted through a special soil sifter with 0.25 mm aperture. Third, the sifted dry soil was placed into a standard plastic container ($\Phi 75 \times 50$ mm) and sealed for at least 20 d before the spectrometric measurement. Finally, the sample was measured with the HPGe

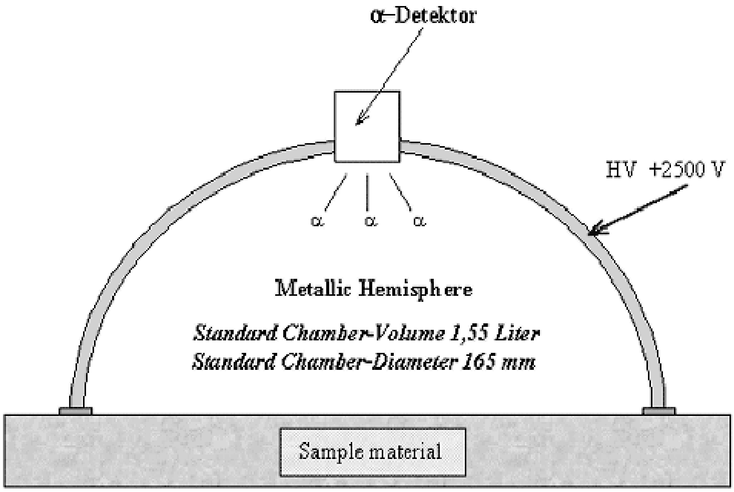

Figure 1. Schematic diagram of ERS-2.

Table 2. Radon soil flux rate and some soil parameters in three areas in China.

City	Beijing	Guiyang	Huhhot
Number of samples	38	6	3
Dry bulk density (g cm^{-3})	1.41 (1.09–1.63)	1.32 (0.96–1.54)	1.51 (1.39–1.63)
Water content (%)	10.3 (2.1–18.7)	27 (18.1–55.4)	13 (9.3–18.1)
Emanation coefficient	0.22 (0.21–0.25)	0.26 (0.24–0.28)	0.22 (0.21–0.24)
Radium content (Bq kg^{-1})	23.5 (15.0–31.4)	132 (32.4–493.1)	17.9 (15.3–21.2)
Measured radon flux rate (mBq m^{-2} s^{-1})	13.2 (3.1–49.9)	34.8 (4.6–128.5)	14.0 (2.0–29.0)
Calculated radon flux rate (mBq m^{-2} s^{-1})	15.4 (9.4–22.6)	60.4 (8.2–265.5)	10.2 (7.8–13.6)
Relative error	1.2 (0.07–4.2)	1.6 (0.08–5.5)	1.2 (0.17–2.9)

for ∼6–8 h to achieve the spectrum for the determination of radium content.

The soil dry bulk density and soil water content were measured with the annular steel sample container (Φ70 × 52 mm), an oven (105 ± 2°C) and an electrical balance (scale: 0–500 g, Precision: 0.1 g). The soil texture of each sample was commercially measured with a laser granularity detector in China University of Geosciences.

RESULTS AND DISCUSSION

The majority of the soil samples in Beijing and Huhhot areas are sandy loam, which is typical soil texture in the north of China. While the soil samples in Guiyang area are usually clay and have higher radium content and higher radon flux rate as well. The results are in accordance with the results of the investigation on natural radionuclide contents in soil in China(11), which indicated that Guizhou was the province with the highest average radium content in China.

In order to estimate the discrepancy between the calculated radon flux rate value and the measured one for each sample, the relative error of the model is introduced which is defined as:

$$\text{Relative\_error} = \frac{|\text{Calculated} - \text{Measured}|}{\text{Measured}}$$

The main soil parameters of the three areas and radon flux rate results for both measured and calculated are shown in Table 2.

In Table 3, more information on the results in Beijing area are provided in detail.

After a complete study on the distribution of relative errors in all the samples, it was found that the samples with lower soil water content (<10%, totally 18 samples) usually had much lower measured radon flux rates than calculated results. (Table 4).

It is well known that the relationship between the radon exhalation and the soil water content is rather complex. Considering soil total porosity, the effective radon diffusion coefficient has an inverse proportion with the soil moisture saturation according

Table 3. Radon flux rate from soil and related soil parameters in Beijing area.

	Range	Arithmetic mean	Median
Dry bulk density (g cm^{-3})	1.09–1.63	1.41	1.4
Total porosity	0.4–0.58	0.48	0.48
Water content (%)	2.1–18.7	10.3	10.3
Moisture saturation	0.05–0.7	0.32	0.29
Effective diffusion coefficient (10^{-6}m^2 s^{-1})	0.4–5.3	2.4	2.4
Emanation coefficient	0.21–0.25	0.22	0.21
Radium content (Bq kg^{-1})	15.0–31.4	23.5	22.6
Measured radon flux rate (mBq m^{-2} s^{-1})	3.1–49.9	13.2	9.9
Calculated radon flux rate (mBq m^{-2} s^{-1})	9.4–22.6	15.4	15.7
Relative error	0.07–4.2	1.2	0.58

Table 4. Relative errors in different soil water contents.

Soil water content	<10%	10–20%	>20%	Total
Number of samples	18	24	5	47
Measured/calculated (arithmetic mean)	0.59	1.15	1.29	0.95
Relative error (original model)	1.7	1.06	0.86	1.29
Relative error (modified model)	0.82	1.04	0.86	0.93

to Equation 6. On the other hand, the emanation coefficient increases with the increasing moisture content until certain moisture saturation is reached, and after that it keeps constant with continuously increasing moisture content(8). A further study shows that in sand, the radon emanation reaches its maximum at a water content of ~2–10%, and in clay at 10–15%(12). Moreover, as the adsorption of radon atoms on solid surface generally decreases rapidly with increasing water content; in dry soils, the radon exhalation rate has the tendency to increase with increasing water content. When a certain moisture saturation is reached, the adsorption becomes negligible(8).

The total discrepancies between the calculated radon flux rates and measured ones are mainly caused by two types of factors. First, the calculating model itself is based on diffusion theory, which neglects some of the other important mechanisms, such as advection and adsorption. The original model does not consider the adsorption of radon atoms on dry solid surface, which cannot be neglected for dry soils(8). The original model also does not include the dependence of emanation coefficient on soil moisture saturation. These two factors can partly explain why the samples with low-water content usually had much lower measured radon flux rates than the calculated ones. Second, the errors both in field measurements and laboratory experiments on radon flux rate and soil parameters may result in the final relative error.

Based on the above study of the experimental data and theoretical analysis of the dependence of the radon exhalation on the sample water content, a trial modification of the model was made. The approach was to derive the ratio of measured to calculated flux rate as a function of the soil water content by the use of linear simulation of the samples with soil water content <10%. After the ratio reached 1 according to the linear function, a constant function 1 was used. The procedure to develop the modification function is shown in Figure 2. Also, Table 4 shows that the average of relative errors was reduced after the modification especially for the samples with soil water content <10%.

Table 5 shows the comparative results of distribution of relative errors according to our measured results of radon flux rate values during our field measurements, which indicates that the modification obviously decreases the total relative error, especially the errors in samples whose measured radon flux rates are rather low (e.g. <10 mBq m^{-2} s^{-1}). The comparative result of the frequency distribution of samples according to relative errors is shown in Figure 3. Figure 3 shows that the number of samples with relative error <0.5 is largely increased after modification.

Table 5 suggests that the model is most valid when the measured radon flux rate values are in the range of 10–40 mBq m^{-2} s^{-1}, which is the typical range in the north of China. When the measured radon flux rate values are <10 mBq m^{-2} s^{-1} or >40 mBq m^{-2} s^{-1}, the relative errors become rather high.

CONCLUSIONS

Different from former studies on radon flux from soil, this work did not build the soil column artificially to control all the soil parameters(13). All the measurements were performed on natural soil so that the data could suggest a real trend of radon flux from natural soil; however at the same time, the method led to some difficulties in analysing the relationship among the many parameters quantificationally. The

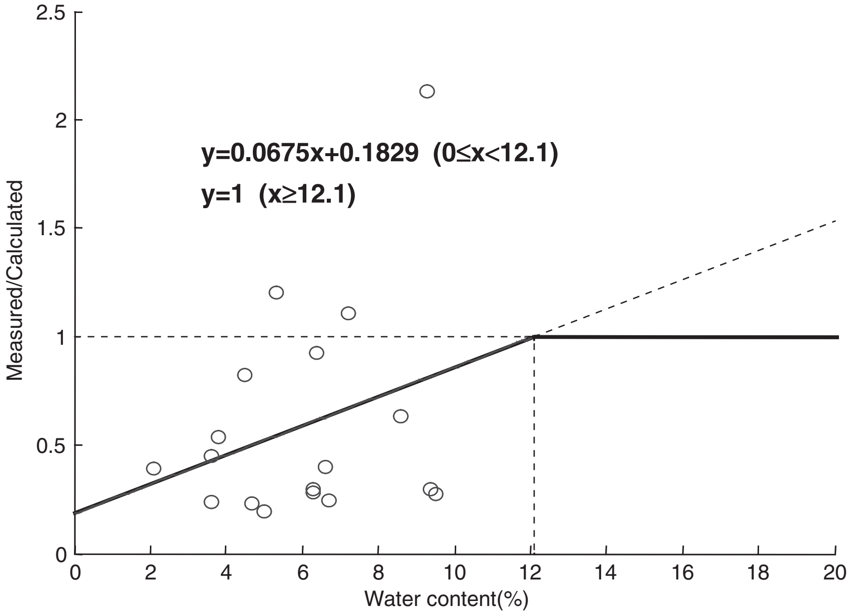

Figure 2. Modification function of the model.

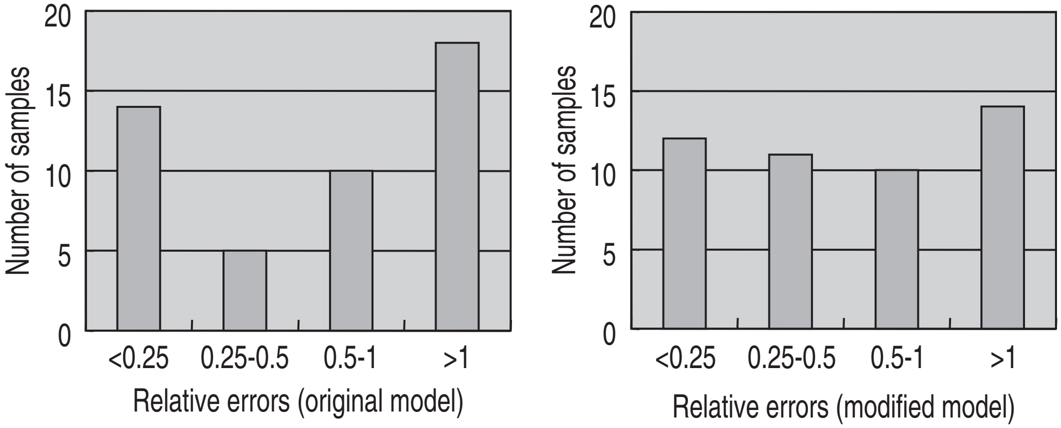

Figure 3. Distribution of samples according to relative errors.

Table 5. Distribution of relative errors according to measured radon flux rate.

Radon flux rate (mBq m^{-2} s^{-1})	<10	10–20	20–30	30–40	>40	Total
Number of samples	22	15	5	1	4	47
Relative error (original model)	2.12	0.27	0.34	0.56	1.88	1.29
Relative error (modified model)	1.33	0.28	0.47	0.59	1.88	0.93

measured radon flux rate result is the synthesis of many influencing parameters, some of which are not even considered.

Totally 47 points in different areas were measured to validate our model. It was suggested that a modification function was necessary and effective to the original model especially in the case of soil water content <10%. Table 5 also suggests that the model is most valid when the measured radon flux rate values are in the range of 10–40 mBq m^{-2} s^{-1}.

Although this work includes limited number of samples, the measured data on comprehensive parameters such as radon flux rate from soil, soil radium content, soil porosity and moisture saturation, and soil texture will provide valuable references for future researches. It should be pointed out that the errors in the calculated values could be much larger due to seasonal changes in the radon flux, e.g. in winter, it is not possible to measure the radon flux during conditions of snow and freezing. In spite of the large uncertainties in both the models and the measurements which are influenced by so many factors, the considerable validation of the model can be seen. In Addition, the discussion on the distribution of errors as well as the reasons causing the errors will show direction for the improvement of the model. What we have done in this paper to try to modify the model is an example. All the above work is the foundation for the establishment of a practical mathematical model for calculating radon flux rate from soil in China.

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