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ARTICLE

Preliminary Study on a Regional Radon Concentration in Surface Soil Prediction Method

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A reformulated mathematical equation for predicting radon concentration in surface soil has been established based on the diffusive and convection theory. The parameters in the equation, such as radon emanation coefficient, convection velocity, diffusion coefficient, porosity of soil and so on, are closely related to the soil types. In order to determine these parameters, field measurement were carried out in totally 31 sites covering four types of soil in Chengdu Area, China. And least squares fitting algorithm and inversion fitting computation were also used. The prediction equation is ultimately applied to map the radon concentration in surface soil of Chengdu Area along with the geochemical databases of uranium content distribution in soil and soil texture distribution. According to the field measurement results on 17 sites in Chengdu, the prediction was verified to work property.

KEYWORDS: radon transport equation, uranium content, soil classification, moisture content, mapping

I. Introduction

About 50% of the natural radiation dose which people accepted is contributed from radon and its short-life progenies. And more and more attentions are paid on the issue of the health risks from them. In fact, radon in the ambient air is mainly released from the radioactive elements uranium and radium in the soil and rocks underground. Therefore, it is of significance for radon control to delineate the potential high radon zone combining the local geological background. Especially in China, the environmental radon survey started relatively late. A large-scale indoor radon measurement has not been conducted yet. However, the database based on regional geological survey has been built in many localities. A great number of geochemical data such as the uranium content and the distribution of soil texture are available. It is a feasible way to predict the regional radon distribution quickly and efficiently by combining these data with radon migration theory. Based on a steady-state radon transport model in porous materials[1)], combined with the local geological and pedological parameters, several methodologies were also developed for predicting a regional radon concentration[2–4)].

In this paper, a methodology for predicting the radon distribution in surface soil of Chengdu Area was introduced. First, a reformulated simplified mathematical equation for the prediction was established by using a new boundary condition, i.e., the flux density of radon on ground-air interface is continuous and equal. Second, the parameters in the radon concentration equation, such as radium content, soil porosity and so on, were measured by field samples. The theoretical radon value was calculated and compared with the measured value. The ultimate prediction equation was derived by corrected and verified by field measured results. Finally, according to the geological mapping on a scale of 1:250000, the uranium content data and the prediction equation, the radon distribution in Chengdu Area was predicted and mapped.

II. Theoretical Formula

In order to obtain the radon transport equation at the surface soil, assuming that

1) The soil column is homogeneous;
2) The elements of uranium and radium distribute uniformly in surface soil and do not exist in the air;
3) Radon transport in the soil is vertical and only due to diffusion and convection in the pore space.

If only diffusion effect is considered, the steady-state radon transport in the soil is governed by (x vertical axis, directed downward on the ground side)[1)]

$$D_1 \frac{d^2 N_1}{dx^2} - \lambda N_1 + A = 0 \qquad (1)$$

Where N_1 is the radon concentration (Bq·m^{-3}). D_1 is the pore average radon diffusion coefficient (m^2·s^{-1}). λ is the radioactive decay constant of radon (2.1×10^{-6}·s^{-1}). A is emanation concentration from Uranium decay through unit thickness in unit time (Bq·cm^{-3}·sec^{-1}). A can be expressed by

$$A = \lambda N_\infty = \lambda \frac{C_{Ra}\rho\alpha}{\eta} \qquad (2)$$

Where α, the radon emanation coefficient; ρ, the dry bulk density of the soil (kg·m^{-3}); C_{Ra}, the specific activity of radium in soil (Bq·kg^{-1}); η, the effective porosity.

If convection effect is also considered, the equation can be written in the following form by Darcy's law.

$$D_1 \frac{d^2 N_1}{dx^2} + v_1 \frac{dN_1}{dx} - \lambda N_1 + A = 0 \qquad (3)$$

Where v_1 is the radon convection velocity (cm·s^{-1}).

The general solution for the differential equation **Eq. (3)** is

$$N_1 = C_1 e^{\frac{-v_1+\sqrt{v_1{}^2+4\lambda D_1}}{2D_1}x} + C_2 e^{\frac{-v_1-\sqrt{v_1{}^2+4\lambda D_1}}{2D_1}x} + A/\lambda \qquad (4)$$

To solve **Eq.(4)**, the boundary conditions are given as follow.

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1) When the thickness of the stratum increases infinitely, N_1 is not infinite;
2) The radon flux density on both sides equal to each other on the ground-air interface (z vertical axis, directed upward on the air side):

$$q_1\,|_{x=0} = q_2\,|_{z=0} = q_0 \quad (5)$$

Where q_1, the flux density on the ground side (Bq·cm^{-2}·sec^{-1}); q_2, the flux density on the air side; q_0, the radon flux density transport from the ground side to the air side, i.e. the surface radon exhalation rate.
3) When x=0, N_1 equal to zero. For the radon concentration in the air is much lower than which in the soil.

The radon concentration at the surface soil is obtained by solving the radon transport equation with the boundary conditions given above.

$$N_1 = \frac{A}{\lambda}\left(1 - e^{\frac{-v_1-\sqrt{{v_1}^2+4\lambda D_1}}{2D_1}x}\right) = \frac{C_{Ra}\rho\alpha}{\eta}\left(1 - e^{\frac{-v_1-\sqrt{{v_1}^2+4\lambda D_1}}{2D_1}x}\right) \quad (6)$$

Using the boundary conditions, q_0 can be also derived.

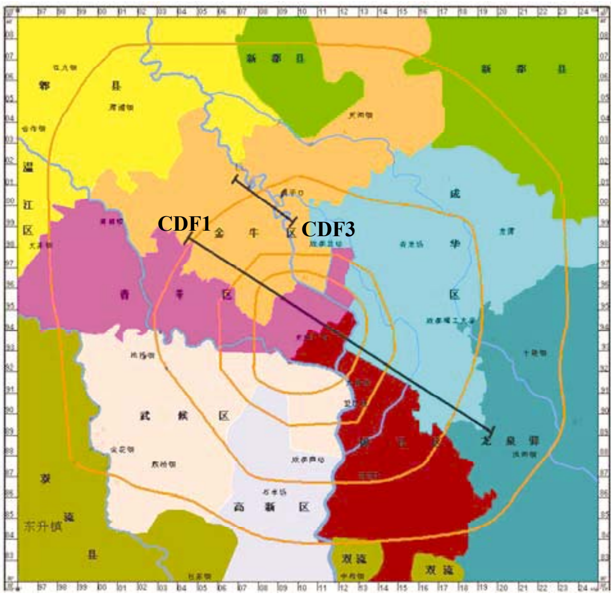

Fig. 1 Field measurement paths in Chengdu Area

III. Experiments

The field measurement paths (CDF1 to CDF3) are shown in **Fig. 1**. The paths CDF1 and CDF3 are subparallel. The total length is about 20 km. 51 measurement sites are included. The path CDF1 orients SE-NW in order to run across the different geological unit of Chengdu Area. It offers good conditions to study on how the physical properties of the soils effect on the radon concentration.

The radon concentration in soil was measured in field by a self made instantaneous radon instrument modeled IED3000 based on the electrostatic collection method. With a hole drilled on the ground, the soil gas in the depth of about 40 cm was in-situ sampled and measured. The instrument was calibrated. The detection sensitivity is 0.006 cpm·Bq^{-1} m^3.

The soil in each site was also sampled. The soil samples were collected at the depth of 40 cm or so by a Φ10 cm × 18 cm cylinder steel container. The parameters such as density, moisture and porosity were measured in the lab. The soil moisture content was determined according to the gravimetric method. Porosity data was supplied by the civil engineering lab of Chengdu University of Technology. The radium specific activity of the samples were then measured by a low-background gamma-ray spectrometer with a Germanium hyper-pure detector after drying (105°C, 24 hours) and crushing (diameter less than 0.25mm). The spectrometer has a relative efficiency of 48.3% and a resolution of 1.87 keV at 1.33 MeV. The measurement uncertainty is about 10%.

The physical parameters for several types of soil are different. After the radium concentration, the soil physical parameter difference is the secondary key factor to affect on the radon concentration in soil. In the experiment, according to the containing particle size, the soil is divided into four types, the sandy clay, the clayey sand, the clay and the sand[5)].

The average value of soil parameters for each soil type is calculated upon the data from path CDF1. The parameter results along with the theoretical equation **Eq. (6)** are used to predict the radon concentration. The measured radon concentration on CDF1 and CDF3 are then used to verify the prediction value.

IV. Results and Discussion

1. Comparison between measured and calculated value

For each site on the measurement path CDF1, the theoretical calculated value is compared to the measured value by calculating the relative deviation to each other. The theoretical values are calculated using **Eq. (6)**. The specific activity of radium C_{Ra}, the soil density and porosity were measured in the experiment. The measured C_{Ra} varied from 21.9Bq·kg^{-1} up to 70.6Bq·kg^{-1}. Other parameters, such as radon emanation coefficient and the diffusion coefficient, were assigned based on the reference[5)].

The results are list in **Table 1.** From the table, the average relative deviation between the calculated and measured concentration, including all soil types, was 97%. There is quite obvious deviation. For the clay, it is as high as 162%.

Table 1 Comparison between measured and calculated radon concentration

Soil types	Sample numbers	Measured Bq/m^3	calculated Bq/m^3	relative deviation
Sandy clay	11	2644 (700-5900)	2665 (1655-3541)	0.47 (0.03-1.36)
Clayey sand	7	2643 (700-5000)	2659 (1771-3614)	0.61 (0.2-0.76)
Clay	8	2696 (200-6496)	2851 (1882-4097)	1.62 (0.18-8.99)
Sand	5	2520 (700-7800)	2130 (1231-3001)	1.18 (0.23-3.29)
Sum/ mean	31	--	--	0.97

According to the analysis on the deviation distribution, there are only one third of the samples for which the measured and the calculated values match each other. The relative deviation is less than 30%. Furthermore, the distribution of the deviation is strongly correlated to the moisture content. The relative deviation is significant large while the moisture content is outside the range of 19% to 26%. The average value of the relative deviation can be as high as 240% if the moisture content goes beyond 26%. It is clear that moisture content is one of the key factors to cause a worse deviation. The emanation coefficient and other parameters are changed by the moisture content, so as to change the radon exhalation and transportation.

2. Moisture content influence correction

The multivariate linearity regression mathematical method is used to correct the deviation caused by the moisture content. According to **Eq. (6)**, all the parameters for difference type of soil, except C_{Ra}, are constant values. Thus, the regression equation for radon concentration in soil is

$$N = a'N_1 + b\omega + c = aC_{Ra} + b\omega + c \tag{7}$$

Where ω is the moisture content (%). a, b, and c are the variates. Based on 31 experimental sites, variates are derived by using the least square multinomial fitting and list in **Table 2.** Diffusion coefficient and emanation coefficient can be derived by the inversion fitting with **Eq. (6)**. Statistic fitting agreement test, F test and double-tail t test are performed on the regression equation. The test results show that all the variates are significant established, unless for the sandy soil samples, for the lack of sample amount, the variate a cannot pass the double-tail t test at the confidence level of 0.1.

Table 2 Values of the variables in the prediction equation for different soil types

Soil types	a	b	c	Diffusion coefficient $\times 10^{-2}\mathrm{cm^2s^{-1}}$	Emanation coefficient
Sandy clay	66.3	-210.7	3761	4	0.13
Clayey sand	135.4	234	-7431	2	0.25
Clay	124.4	-112.4	-855	1.2	0.15
Sand	67	326.7	-5715	4.6	0.14

Applying the data from **Table 2** into **Eq. (7)**, the final prediction equation was obtained. The deviations between the predicted values and the measured values are calculated and compared to the un-corrected deviations. The deviation distribution to the corrected deviation and the un-corrected ones are both drawn in **Fig. 2**. **Figure 2(c)** shows that the average deviation is improved from 97% down to 50% with the correction. In the case of deviation larger than 1, the value decreases from 305% down to 137%. Based on **Fig. 2(b)**, the frequency for which the deviation is less than 30% increases

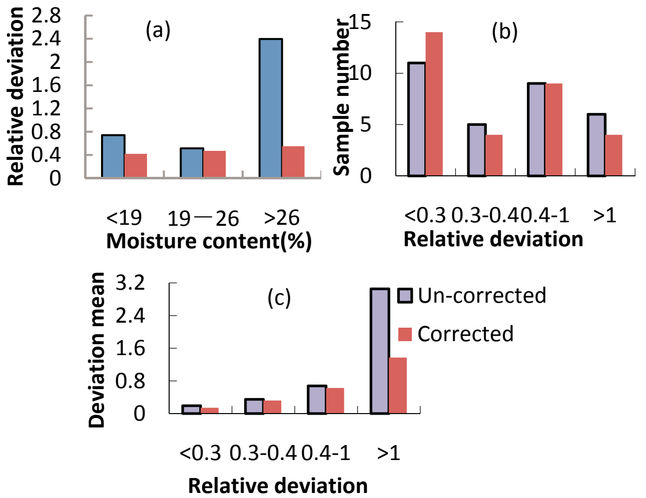

Fig. 2 Relative deviation in cases of corrected and un-corrected

clearly after the correction. The predicted value for about 60% of the samples is better agreed with the measurement value. The sample amounts for deviation larger than 1 becomes less. On **Fig. 2(a)**, the relative deviations are well distributed cover all the moisture contain section. The deviation no longer increases cause by the moisture content after the correction.

3. Verification

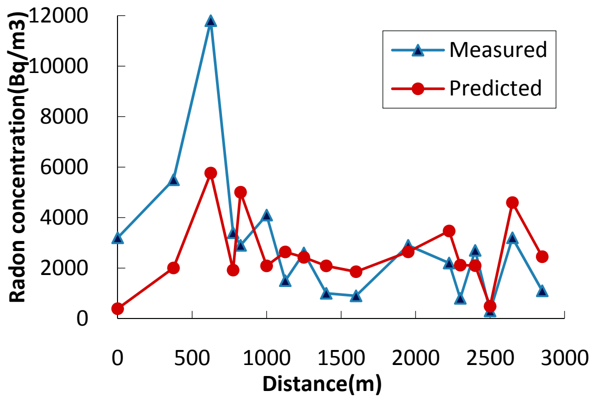

Fig. 3 The measured radon concentration V.S. predicted value in path CDF3

The measured values of radon concentration in the 17 sites in path CDF3 are used to verify the prediction equation. From **Fig. 3**, the predicted values are close to the measured ones. The average relative deviation for them is 70%. The deviation for sand soil is as high as 92%. It is much larger than the average value. The possible reason is lack of sample sites. As mentioned in section 2, lack of sample sites also causes the regression variate failure in the double-tail t test.

4. Mapping results

The superposition of a geochemical map and a geological map on the scale of 1:25 0000 is made to have the correspondence between the uranium concentration and types of soil. According to the U-Ra equilibrium coefficient from the geochemical exploration, i.e. 1.92, the radium specific activity can be calculated. Then an isoline map of the predicted radon concentration can be figured out by applying the radium information and as well as the soil parameters into **Eq. (7).** The isoline map is shown in **Fig. 4**. The 168 black dots which distribute on the 14 ×12 rectangular grid are the geochemical exploration sites. The grid size is about 4 km. Thus, it covers 56 × 48 km^2 in Chengdu Area.

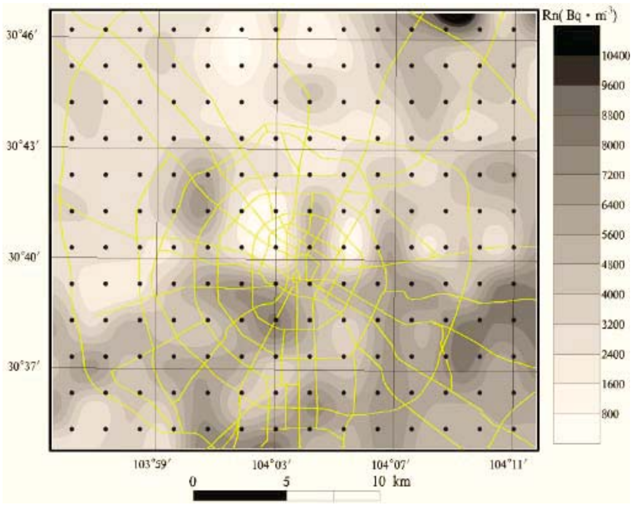

Fig. 4 Isoline map of the predicted radon concentration in surface soil in Chengdu Area

From the prediction, the average radon concentration in surface soil in Chengdu Area is 4307 Bq /m^3. It agrees with the measured value of 4047Bq/m^3 from reference 6).

V. Conclusions

A regional radon concentration in surface soil prediction method with mapping is presented. The prediction results are mostly agreed with the measured values in some sampled sites. To have more precise results, some more detail work can be done. For example, more moisture content data should be measured for the corresponding correction, and the influence from the geological structures such as faults must be studied as well.

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References

1) V.C.Rogers, K.K.Nielson, "Multiphase radon generation and transport in porous materials," *Health Phys.*, 60, 807(1991).
2) G. Ielsch, C. Ferry, G. Tymen, M.C. Robe, "Study of a predictive methodology for quantification and mapping of the radon-222 exhalation rate", *Journal of Environmental Radioactivity*, 63, 15 (2002).
3) J.C.H. Miles, J.D. Appleton," Mapping variation in radon potential both between and within geological units", *J. Radiol. Prot.*, 25, 257(2005).
4) LIU Qingcheng, YANG Yaxin, WAN Jun, "The research of fast prediction method of radon concentration in regional air", *Radiation Protection Bulletin*, 21[1], 20(2001), [in Chinese].
5) WANG Xiyuan, ZHU Li, LV Lei et al., Survey of the soil radon in China, Science Press, Beijing, 210(2008), [in Chinese].
6) ZHOU Xiaojian, ZHANG Chengjiang, SHI Zeming, "The study of radon level in Greenbelt soil in Chengdu city", *J. Environ. Health.*, 23[5], 449(2006), [in Chinese].
7) Gabriele Buttafuoco, Adalisa Tallarico, Giovanni Falcone, "Mapping Soil Gas Radon Concentration: A Comparative Study of Geostatistical Methods", *Environ Monit Assess.*, 131, 135(2007).
8) Weihai ZHUO, Takao IIDA, Masahide FURUKAWA, "Modeling Radon Flux Density from the Earth's Surface", *Journal of Nuclear Science and Technology*, 43[4] 479(2006).
9) Kainan SUN, Qiuju GUO, Weihai ZHUO, "Feasibility for Mapping Radon Exhalation Rate from Soil in China", *Journal of Nuclear Science and Technology*, 41[1], 86(2004).
10) J.D. Appleton, J.C.H. Miles, B.M.R. Green, R. Larmour, "Pilot study of the application of Tellus airborne radiometric and soil geochemical data for radon mapping", *Journal of Environmental Radioactivity*, 99, 1687 (2008).
11) Z.S. Žuni′, K. Kozak, G. Ciotoli et al., "A campaign of discrete radon concentration measurements in soil of Niška Banja town, Serbia", *Radiation Measurements*, 42, 1696 (2007).
12) Masami FUKUI, "Evaluation of Radon in Soil Gas and Natural Radioactivity in Surface Soil of Kinki District, Japan", *Journal of Nuclear Science and Technology*, 44 [8], 479 (2007).
13) V. Moreno , C. Baixeras, Ll. Font, J. Bach, "Indoor radon levels and their dynamics in relation with the geological characteristics of La Garrotxa, Spain", *Radiation Measurements*, 43, 1532 (2008).