Journal of Environmental Radioactivity 135 (2014) 63–66

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

A new method for estimating the coefficients of diffusion and emanation of radon in the soil

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ARTICLE INFO

Article history:
Received 17 June 2013
Received in revised form
19 March 2014
Accepted 6 April 2014
Available online 4 May 2014

Keywords:
Radon transfer
Mathematical modeling
Diffusion coefficient
Emanation coefficient

ABSTRACT

This paper describes a new method for determining the basic parameters of soil - diffusion and emanation coefficients related to the transfer of radon in the soil matrix, which are very useful for testing models, based on diffusion and characteristics of various soil matrices regarding the dangers of radon. The method is based on the measurement of radon in soil air on two small depths, differing twice. The paper presents the results of the determination of the parameters for covering loams and clays of Tomsk (Russian Federation), obtained by this method.

1. Introduction

In solving the problems associated with radon – evaluation of radon prone region, prospecting and exploration of radioactive ores, predicting seismic areas - the method of mathematical modeling of radon transport through porous media is widely used (Der Poel, 2002; Koarashi et al., 2000; Miklyaev et al., 2008; Novikov, 1989; Rogers and Nielso, 1991; Ryzhakova, 2009; Sahoo and Mayya, 2010; Savović et al., 2011; Sun et al., 2004). Moving in the soil, radon is involved in various physical processes of molecular diffusion in air and water phases of the soil, convection and turbulent mixing of air in the surface layers of soil due to changes in atmospheric conditions (pressure, temperature, wind speed and direction), filtration with the upward water flows; moving by gas lift force generated when the soil pores are filled with water, in the process of exchange between the air, water and solid soil phases. The diversity and complexity of these processes depending also on the constantly changing weather conditions, do not allow to build a precise mathematical model of transport of radon through soils. Therefore, in simulating the semi-phenomenological approach is used, where the dispersed porous medium is replaced by a continuous medium and geophysical properties of the soil and transport mechanisms are described by means of the effective parameters.

The best known are the diffusion and diffusion-convection models, the practical use of which is hampered by the ambiguous interpretation of the physical nature of simulation parameters, and, as a consequence, by the problems with the choice of methods for their determination. It is particularly difficult to interpret and determine the parameters, describing the movement of radon in soils, diffusion coefficient D_e – in diffusion model, and diffusion coefficient D and convection velocity υ – in diffusion-convection model (Ryzhakova, 2012; Ryzhakova and Yakovleva, 2004a). In most papers on the transfer of radon, the physical meaning of these parameters (as well as the emanation coefficient K_{em}) is not disclosed, and the methods of their determination are not justified. Moreover, the values obtained under laboratory conditions, when there is no influence of the atmosphere, are often used in simulation. In addition, during the preparation of samples for measurement the physical properties of soils are changed, especially porosity and humidity that have a significant effect on the radon emanation and transport (Breitner et al., 2010; Chauhan et al., 2008; Ryzhakova and Ramenskaya, 2012; Ryzhakova and Shestak, 2009; Sakoda et al., 2010; Yu et al., 1993; Zhuo et al., 2006).

The parameters used in the semiphenomenological description of the transfer of radon are empirical in nature and must therefore be determined under conditions of natural occurrence of soils (Ryzhakova, 2012; Sahoo et al., 2010). For example, the diffusion coefficient D_e, used in the diffusion model should take into account all the processes by which radon is moved to the soil surface,

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http://dx.doi.org/10.1016/j.jenvrad.2014.04.002
0265-931X/

including processes caused by the influence of the atmosphere (Ryzhakova and Shestak, 2009). A similar situation occurs with respect to the coefficient of emanation K_{em}, which is usually defined as the proportion of radon released to loose pores of the soil. In particular, just this value is measured by γ-method (Miklyaev et al., 2005) in the measurement of emanation coefficient. However, under natural conditions not all radon, caught in the loose pores of the soil, comes to the surface. In the result of metabolic processes occurring in the air, water and solid phases of soil a part of radon is adsorbed on the surface of solid particles and is dissolved in the water contained in the pores. This is evidenced by, for example, the fact that the coefficients of emanation measured by a laboratory γ-method are 1.5 … 2 times larger than the values obtained under natural conditions (Krampit, 2004; Miklyaev et al., 2005).

The purpose of this work is to determine the average values for the diffusion and emanation coefficients using measurements of radon volumetric activity in the soil air at two small depths (less than 1 m). Measurements were carried out in loams of Tomsk (Russian Federation) on 10 different sites with exposure time (3 … 4) days that allows to smooth spatial and temporal variations of radon (Iakovleva and Ryzhakova, 2003).

2. Definition of the radon transport related parameters of the soil in natural conditions

The known methods for determining the diffusion and emanation coefficients under natural conditions are rather labor-consuming. An external source of radon and carrying out repeated (10 … 15) measurement of radon volumetric activity in soil air within a day are required in the method of "an instant source" (Bulashevich and Kartashov, 1957; Ryzhakova and Shestak, 2009) for determination of the diffusion coefficient. Other method of D_e determination is based on simultaneous measurement of radon volumetric activity at several depths (not less than 5 … 6) (Ryzhakova and Shestak, 2009). In paper (Krampit, 2004) the emanation coefficient was calculated by radon equilibrium activity measured at depth not less than 3 … 4 m.

Tomsk Polytechnic University (Russian Federation) has developed a simple, cheap and reliable method for determining the diffusion and emanation coefficients of soil associated with the transfer of radon in the soil matrix, which are very useful for testing models based on diffusion and characterization of various soil matrices in terms of the radon risks. The method is based on the measurement of radon at two small depths, differing by half. The use of two depths, differing twice, allows to get simple formula to determine the radon transport related parameters of the soil on the basis of radon transport equation for a homogeneous porous medium (Ryzhakova, 2007; Ryzhakova and Yakovleva, 2004b). In our work, for this purpose we used the diffusion equation of radon transport in porous media:

$$\frac{d^2A}{dz^2} - \frac{\lambda}{D_e}A + \frac{\lambda}{D_e\eta}K_{em}A_{Ra}\rho_d = 0, \quad (1)$$

where A – radon volumetric activity in the air of soil pores, Bq/m^3; A_{Ra} – specific activity of Radium, contained in the soil, Bq/kg; ρ_d – density of dry soil, kg/m^3, η – porosity of the medium, defining the volume share of soil, which is the share of pores filled with air; $\lambda = 2.1\cdot10^{-6}$ – radon decay constant, 1/s (coordinate z is calculated from the surface of porous medium).

For the homogeneous soil layer under zero boundary conditions $A(0) = 0$ the solution of Equation (1) is written as:

$$A(z) = A_\infty\left(1 - e^{\sqrt{\lambda/D_e}z}\right), \quad (2)$$

where $A_\infty = K_{em}A_{Ra}\rho_d/\eta$ – value of radon volumetric activity in the air of soil pores at large depths.

Using expression (2) one can write the system of equations for two depths of measurements, differing twice. And formulae to determine radon transport related parameters of soils can be obtained from this system of equations:

$$D_e = \frac{\lambda\cdot h_1^2}{\left[\ln\left(\frac{A_2}{A_1}-1\right)\right]^2}; \quad (3)$$

$$K_{em} = \frac{A_1\cdot\eta}{\left(2-\frac{A_2}{A_1}\right)A_{Ra}\rho_d}, \quad (4)$$

where A_1, A_2 – value of radon volumetric activity measured at the depths h_1 and h_2 respectively, $h_2 = 2h_1$.

The main condition of the method applicability – measured soils should be fairly uniform in depth, especially in mineralogical composition. This condition is nearly always fulfilled (except technogenic soil), since the thickness of the "active layer" in which the radon can come to the surface is relatively small and does not exceed a few meters.

To obtain the most accurate results, the depth of measurements by the order magnitude should be commensurate with the length of diffusion $L = \sqrt{D_e/\lambda}$. For example, for the loose disperse soils, loams types, the depth h_1 is about (0.4 … 0.6) m. At such depths the volumetric activity of radon varies with depth, and the dependence is nonlinear. At smaller depths, when the dependence of volumetric activity on the depth is close to linear one, formulae (3), (4) don't work, and at larger depths the radon volumetric activity varies slightly, that can cause a large error in determination of parameters.

3. Measurement methods

Measurements of radon volumetric activity in the air of soil pores were carried out simultaneously in two blast-holes by track detectors of IPRR type (individual passive radon radiometer) or by radiometer RRA-01M-03 on 10 sites of Tomsk in the Russian Federation. Recommended depths of measurements are ~(1 … 2) of the diffusion length. To provide the homogeneity of soils on chosen sites, the upper soil layer with thickness of (0.3 … 0.5) m was taken off. For observable loose dispersed soils (clay sand, loam) the recommended depths h_1 and h_2 are (0.4 … 0.6) m and (0.8 … 1.2) m correspondingly. The passive track detectors or samplers were placed into both blast-holes with diameter of 5.5 cm. Then the

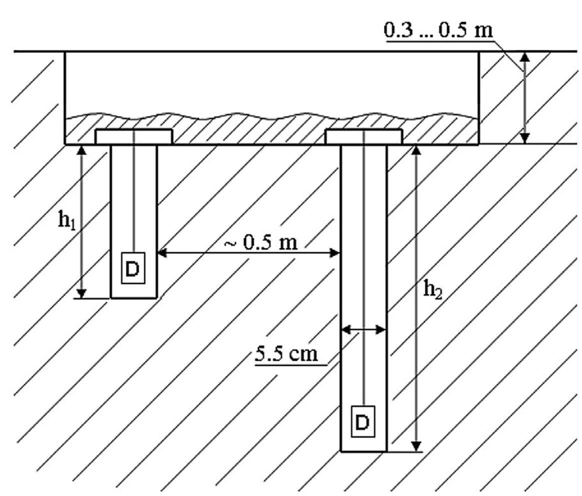

Fig. 1. Diagram of pore activity measurement (D – radon detector).