Journal of Applied Geophysics 80 (2012) 151–157

Contents lists available at SciVerse ScienceDirect

Journal of Applied Geophysics

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

Parameters of modeling radon transfer through soil and methods of their determination

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ARTICLE INFO

Article history:
Received 25 October 2010
Accepted 24 January 2012
Available online 16 February 2012

Keywords:
Radon
Diffusion
Convection
Emanation
Transfer
Soils

ABSTRACT

The paper considers diffusion–convection and diffusion models of radon transfer through soil and their main parameters, namely convection rate, diffusion and emanation coefficients. It is shown that physical interpretation and values of these parameters depend on the measurement method. It is proposed to consider modeling parameters as phenomenological ones and to determine them on basis of radon activity in soil pores measured under natural conditions. A simple method is proposed for the determination of parameters under specific geological and climatic conditions based on measurement of radon volumetric activity in pores at two twice different depths. The article presents the results of measuring effective diffusion and emanation coefficients of highly-dispersed clay soils of Tomsk (Russia) obtained by different methods under both laboratory and natural conditions, their analysis is conducted.

1. Introduction

The study of the process of radon transfer through soil is connected with solving a number of important tasks. The radioactive gas radon (having no taste, color or smell, 7.5 times heavier than air, a powerful natural alpha-emitter with a 3.8-day half-life) is emitted from the soil and may accumulate in buildings, which poses danger to people, especially to those who live on the ground floor (Sources, 1992). For this reason appropriate investigations are conducted in order to evaluate radon danger of building estates. Well-developed methods of radiation detection make it possible to use radon as an optimal indicator when conducting different kinds of geological and geotechnical research, for example searching for radioactive ores and forecasting seismic activity of certain areas. For solving the above mentioned problems the method of mathematical modeling of radon transfer through porous media is often used (Miklyaev et al., 2008; Novikov, 1989; Parovik and Firstov, 2009; Rogers and Nielson, 1991; Sun et al., 2004; Van der Poel, 2002). Owing to complexity and diversity of transfer processes the semi-phenomenological approach is used at modeling. It means that porous dispersion medium is substituted for continuous medium and geophysical parameters of soils and transfer mechanisms are described with the help of effective parameters.

The diffusion and diffusion–convection models are the most well known ones. Their practical application is complicated by ambiguous interpretation of the main modeling parameters and, consequently, by the problems of choosing methods of their determination. For this reason it is interesting to analyze the used models of radon transfer through soils and methods and results of modeling parameters determination.

2. Mathematical models of transfer

Two mechanisms, diffusion and convection, are used when describing the shift of radon to ground surface. The transfer equation taking both mechanisms into account (z is counted from ground surface) has the following form:

$$\partial C/\partial t = D\left(\partial^{\uparrow}2C\right)/\left(\partial z^{\uparrow}2\right) + \upsilon\partial C/\partial z - (C+S), \tag{1}$$

where C is volume concentration of radon, i.e. the number of radon particles per unit volume of porous medium, m^{-3}; D diffusion in porous medium, $m^2 \cdot s^{-1}$; υ — convection rate, $m \cdot s^{-1}$; λ is the radon decay constant, s^{-1}; S is radon source function, $m^{-3} \cdot s^{-1}$. If radon emanates only from solid soil particles, source function is called emanation intensity and is determined by the expression (Novikov, 1989; Rogers and Nielson, 1991):

$$S = K_{em} \cdot A_{Ra} \cdot \rho_d, \tag{2}$$

where K_{em} is emanation coefficient, relative units; A_{Ra} — specific activity of radium in the layer, Bq·kg-1, ρ_d — density of dry soil $kg \cdot m^{-3}$.

In the experiment it is not volume concentration but pore activity A, i.e. radon activity per unit volume of air filling the pores in the soil that is

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doi:10.1016/j.jappgeo.2012.01.010

measured. Volume concentration and pore activity are connected by the evident ratio (Novikov, 1989):

$$A = \frac{\lambda C}{\eta}, \quad (3)$$

where η is medium porosity which shows medium porosity, it determines the soil volume part for pores filled in with air. Then porous activity equation taking Eq. (3) into account has the following form:

$$\partial A/\partial t = D\left(\partial^{\uparrow}2A\right)/\left(\partial z^{\uparrow}2\right) + \upsilon\partial A/\partial z - \lambda A + \left(/\eta\cdot K_{\downarrow}em\cdot A_{\downarrow}Ra\cdot\rho_{\downarrow}d\right) \quad (4)$$

The given equation contains 3 main modeling parameters: effective diffusion coefficient D, convection rate, and emanation coefficient K_{em}.

When solving the problems of radon transfer through soils basing on Eq. (4), the values of D and K_{em} obtained by laboratory methods are often used, which is rather arguable because the given parameters describe complex physical processes dependent on atmospheric conditions. In some works effective parameters are modeled considering the transfer in each phase of the soil (liquid, solid and gas) taking into account an exchange process between phases and geophysical properties of the medium (Koarashi et al., 2000; Rogers and Nielson, 1991; Van der Poel, 2002).

Molecular diffusion in solid and liquid phases is often neglected and radon exchange between phases is considered equilibrium, which makes it possible to express radon concentration in solid and liquid phases through the concentration in gas phase. As a result, one transfer equation for gas phase is written down. Its effective parameters are determined through geophysical characteristics of the soil and its separate phases. Despite the same approach, in different works formulae for calculating effective parameters are written down in different ways.

As an example let us consider formulae for effective diffusion coefficient:

$$D = D_a\cdot\eta_a(1-\omega+\omega L)/(\eta_a(1-\omega+\omega L)+\rho_d\cdot k_a)$$

(Rogers and Nielson, 1991);

$$D = [(1-w)\cdot\tau_a\cdot D_a + w\cdot\tau_w\cdot D_w\cdot L]\cdot\eta_a$$

(Van der Poel, 2002).

In the formulae the following symbols are used: η_a is an absolute porosity of the soil which shows what part of soil volume all pores (open, dead-end and filled with water) account for; w is soil moisture, D_a, D_w are molecular coefficients of diffusion in air and water respectively; τ_a, τ_w — tortuosity of pores filled with water and air respectively; L — Henry constant; k_a — coefficient of radon adsorption on dry particles of the soil.

Analogous equations are written down for emanation coefficient.

As for convection rate υ, this case is even more complicated because in most works concerning radon transfer through soils the question of physical essence of this mechanism is not even touched upon. Traditionally convection is understood as substance shift because of the Archimedean force which occurs in the case of non-uniform heating when the more heated substance has lower density. This radon transfer mechanism is possible in solid rocks where there are deep cracks and cavities and local thermal gradients are created at great depths (tens of meters and more) due to geochemical processes and radioactive decay (Lyan Sin Joun, 1987).

From a practical point of view the most wide-spread mellow soils such as clay sands, loams, etc. are of the greatest interest.

According to different estimates, the velocity of radon shift in mellow soils is 1E−4÷1E−3 cm/s. The for its life-time $\tau = 1/\lambda \approx 5E+5$ s. radon may rise from depths that do no exceed several meters. At such depths significant temperature and pressure gradients may occur only under the influence of the atmosphere.

In the majority of works radon convection rate in solid and liquid phases is determined on the basis of Darcy's law:

$$\upsilon = -\frac{k}{\mu}\frac{dP}{dz}, \quad (5)$$

where k is medium permeability, μ is dynamic viscosity of the substance (water or air), P is pressure in the pores, (for water phase pressure gradient is determined by hydraulic gradient). The main difficulty occurring at modeling radon transfer on the basis of Darcy's law lies in determining pressure gradient.

There are several works in which relatively simple models of convection transfer based on certain physical processes are proposed. In the above-mentioned work (Lyan Sin Joun, 1987) convection rate is determined through geothermal gradient on the basis of the hydrodynamic model. In the work (Koarashi et al., 2000) convection is understood as radon transfer as a result of its displacement from the soil under water head, while in work (Pavlov, 1996/1997) convection rate is determined through moisture evaporation rate. The authors of work (Ivanova, 2001) connect the presence of pressure gradient with the expansion of temperature waves and atmospheric pressure fluctuations through soils, however, it is rather difficult to conduct calculations for real conditions within the framework of this model because of complexity of atmospheric processes. Moreover, the model does not take into account turbulent phenomena in the surface layer of the atmosphere and strong influence of atmospheric precipitation on convection displacement of radon from soils.

There is no reason for giving preference to one of the above-mentioned mechanisms. Most likely, each of them contributes to convection process of radon transfer. Complexity and diversity of physical processes responsible for radon transfer through soil make its strict mathematical description impossible. In this work (Ryzhakova and Yakovleva, 2004a) we propose to differentiate between diffusion and convection mechanisms when measuring effective diffusion coefficient under laboratory conditions, i.e. in the absence of convection. In this case convection rate can be considered a phenomenological parameter which is determined under specific geological and atmospheric conditions based on the altered values of pore activity (Ryzhakova and Yakovleva, 2004c).

In the research works of other authors (Bulashevich and Khairitdinov, 1959; Miklyaev et al., 2008; Ryzhakova, 2009; Sun et al., 2004) it is shown that a simpler diffusion model containing two effective parameters: effective coefficients of diffusion D_e and emanation K_{em} gives a good description of radon transfer through soils. In this case the parameter D_e used in modeling must take into account not only diffusion in a porous medium but also other processes responsible for radon transfer through soils including the ones connected with atmospheric phenomena. Atmosphere condition also influences the value of effective emanation coefficient because this parameter depends on the exchange processes taking place in different phases of the soil. Obviously, taking all the processes connected with atmospheric conditions into account is possible only if modeling parameters are determined under the conditions of natural deposition of soils.

Within the framework of diffusion model stationary radon transfer equation for pore activity has the following form:

$$\frac{d^2A}{dz^2} - \frac{\lambda}{D_e}A + \frac{\lambda}{D_e\eta}K_{em}A_{Ra}\rho_d = 0. \quad (6)$$

Within the limits of a uniform soil layer the equation is written down in the following way:

$$A(z) = C_1e^{-\sqrt{\lambda/D_e}\cdot z} + C_2e^{\sqrt{\lambda/D_e}\cdot z} + A_\infty, \quad (7)$$

where $A_\infty = K_{em}A_{Ra}\rho_d/\eta$ — is the stationary value of radon pore activity, it is actual for pore activity at infinitely great depths in a uniform soil.