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Thermo-diffusional radon waves in soils

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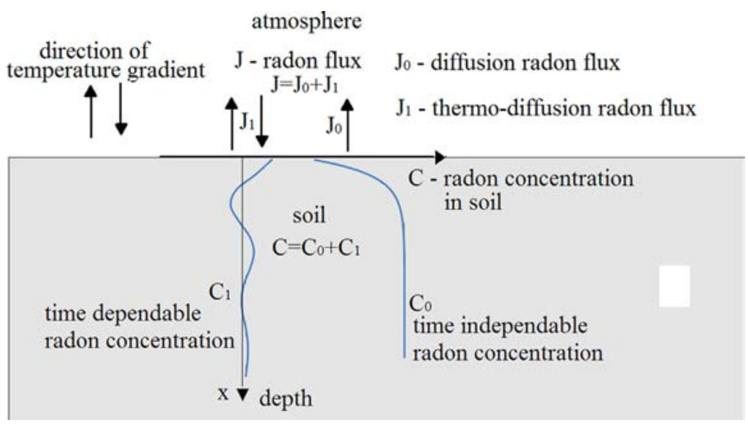
HIGHLIGHTS

- Temperature oscillations in atmosphere generate radon waves in soil.
- Radon flux in atmosphere is a harmonic function of time.
- Radon concentration waves in soil have the same frequency as the temperature waves.

GRAPHICAL ABSTRACT



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ABSTRACT

A new theoretical framework for diurnal and seasonal oscillations of the concentration of radon in soil and open air is proposed. The theory is based on the existing temperature waves in soils and thermo-diffusional gas flux in porous media. As soil is a non-isothermal porous medium, usually possessing a large fraction of microscopic pores belonging to Knudsen's free molecular field, a thermo-diffusional gas flow in soil has to arise. The radon mass transfer equation in soil for sinusoidal temperature oscillations at the soil–atmosphere boundary is solved, which reveals that radon concentration behaves as a damped harmonic wave. The amplitude of radon concentration oscillations and phase shift between radon concentration oscillations and soil temperature depend on the radon diffusion coefficient in soil, rate of radon production, soil thermal conductivity, average soil temperature, decay constant, and heat of radon transfer. Primarily numerical calculations are presented and comparisons with experimental data are shown.

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1. Introduction

The correlation between atmospheric temperature fluctuations and radon flux has been the subject of numerous investigations. Many researchers have observed annual and diurnal oscillations of ^{222}Ra (radon) and ^{220}Rn (thoron) fluxes. To our knowledge, the first paper

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which revealed correlation between outdoor radon concentration and air temperature is dated to 1956 (Okabe, 1956). Since then, monitoring of radon concentration in air is continuing all around the world. Besides the theoretical interest of this correlation, the knowledge of seasonal and diurnal radon fluctuations in soil and air has a practical application. It is of importance for the predictions of the natural disasters (radon concentration change can be a precursor indicative of earthquakes, tornados, hurricanes (Cigolini et al., 2001; Planinic et al., 2004; Tsvetkova et al., 2005; Crockett et al., 2006)), radiation monitoring over caves with tourist business (Barbosa et al., 2010), for risk assessment of radiation levels at sites of radioactive waste burial, and safety of mine workers (Kant et al., 2010).

Atmospheric diurnal radon and thoron concentrations typically reach their maxima in early morning (Schery and Grumm, 1992; Podstawczyńska et al., 2010) and seasonal radon concentrations usually have maxima in winter. However, some authors found deviations from this general tendency. Schery et al. (1984) observed the enhancement of flux density in the afternoon and Gesell (1983) gives thirteen references of measurements of radon diurnal and annual variations made in different countries where reported maxima of seasonal values occurred in different seasons (winter, fall, and summer) depending upon the location.

Reiter (1978); Grumm et al. (1990); Porstendörfer et al. (1991), and Sesana et al. (2003) accepted an eddy diffusion model to explain the periodic diurnal oscillations of radon concentration. But Schery et al. (1989) find it difficult to believe that such effects of turbulent mixing are significant. It is also not easy to explain the radon diurnal oscillations at the 0.57 m depth, recorded by Schery et al. (1984), based on the turbulent mechanism. Gesell (1983) also concluded that seasonal radon fluctuations could not be explained well by turbulent transfer model. As radon concentration oscillations correlate with temperature oscillations, a suggestion was made that thermal advection of air in the soil might be responsible. However calculations made by Schery and Petscek (1983) indicate that for real values of permeability and other soil parameters, advection is not a reasonable factor in this phenomenon. Since there is no consensus and there is no convincing analytical model, further research is needed in this topic. Merrill and Akbar-Khanzaden (1998) and Jilek et al. (2014) investigated the effect of climatic conditions (such as barometric pressure, thermal air gradient, relative air humidity, wind speed and direction and solar radiation intensity) on atmospheric radon concentration diurnal and seasonal oscillations. They collected a significant amount of experimental data, but could not discover the mechanism responsible for periodic radon concentration oscillations in the air.

There are also difficulties in explaining radon concentration fluctuations in soil and caves. Winkler et al. (2001) found that on average, at all sampling positions in the test field and nearly at all-times radon concentration at 0.5 m depth was significantly higher than at 1 m depth in contrast to theory and some field experiments (Al-Shereideh et al., 2006). Radon concentration oscillations in underground locations such as caves, tunnels, and basements of the buildings are often opposite to that above the ground level i.e., in winter, they were lower than in summer (Li et al., 2006; Perrier et al., 2007; Dueñas et al., 2011).

Although extensive research efforts have been made to develop an adequate model of radon transport and numerous papers were devoted to diurnal and seasonal radon concentration oscillations in the atmosphere, the models' predictions are still very often in conflict with the experimental data. Clarification of radon movement in soils is needed to predict radon fluctuations, and new ideas have to be brought forth to explain the mechanism of seasonal and diurnal radon oscillations.

Goldman et al. (1987, 1992) first proposed the idea that thermo-diffusion (thermo-transpiration) is a dominant mechanism of air exchange in soil. Minkin (2001, 2002, 2003) developed this idea and theoretically and experimentally showed that thermogradient is a driving force of radon indoor entry. The review (Minkin and Shapovalov, 2008) of published papers related to radon transport revealed that there are numerous misconceptions about the mechanisms of radon infiltrations into homes and thermo-diffusion should be considered as a competitive (Zafrir et al., 2013) and in some cases dominant mechanism of radon transport in soil (Dong et al., 2004; Minkin and Shapovalov, 2007). This fact makes it reasonable to apply the theory of thermo-diffusion radon transport to explain diurnal and seasonal outdoor radon oscillations. This research article presents a novel theory of these fluctuations based on thermo-diffusion. All calculations below are made in the international system of units.

2. Transport equations

Natural soils are generally not homogeneous. Mineral composition, structure, and water content vary with depth and location. However, the assumption of a homogeneous soil may still be reasonable and is widely used in soil physics and in solving problems of radon transport (Van Wijk and de Vries, 1963; Clements and Walkening, 1974; Marshall and Holmes, 1979; Sposito, 1976; Van Der Spoel et al., 1997). To facilitate the mathematical treatment some approximations are made:

1. The soil is homogeneous.
2. All variables change only in vertical direction (x is depth positive downward; at the soil surface $x = 0$).
3. No heat is generated in soil or converted into other form of energy.
4. Soil is a porous medium.
5. Diurnal and seasonal temperature oscillations on the soil–atmosphere border are periodic.
6. Soil's intrinsic properties are not functions of time (often used assumption: Clements and Walkening, 1974; Schery et al., 1984, 1989; Ota and Yamazawa, 2010).

Using these assumptions, the temperature distribution in soil can be described as a damping harmonic wave function (Van Wijk, 1963; Van Wijk and de Vries, 1963; Marshall and Holmes, 1979; Hillel, 1982)

$$T(x,t) = T_0 + T_1(x,t) = T_0 + T_{10}\exp(-\alpha x)\cos(\omega t - \alpha x)$$

where T_0 is the average soil temperature, T_{10} is the amplitude of temperature oscillations at the earth surface, $\alpha = \sqrt{\frac{C_{soil}\omega}{2\kappa}}$ is the damping constant (angular wave number), κ is thermal conductivity, C_{soil} is volumetric heat capacity of soil, t is time, ω is angular seasonal or diurnal frequency ($\omega_d = 2\pi\ \mathrm{day}^{-1} = 2\pi\,/\,86{,}400\ \mathrm{s}^{-1} = 7.27 \times 10^{-5}\ \mathrm{s}^{-1}$ for diurnal variations and $\omega_s = 2\pi\ \mathrm{year}^{-1} = 1.99 \times 10^{-7}\ \mathrm{s}^{-1}$ for seasonal variations). The variable part of the last equation can be written in a complex form

$$T_1(x,t) = T_{10}\exp[-\alpha x(1+i)]\exp(i\omega t) \tag{1}$$

where $i = \sqrt{-1}$ and $T_{10}(x,t)\exp[-\alpha x(1+i)]$ is a complex amplitude of temperature oscillations at the depth x. This equation shows that at the depth x the amplitude of temperature oscillations is smaller than T_{10} by a factor $\exp(-\alpha x)$ and that there is a phase shift, $-\alpha x$, between temperature oscillations on the soil surface and at depth x. Thermal conductivity, κ, and volumetric heat capacity, C_{soil}, depend on the type of soil, porosity, humidity and have a large range. For typical soil $\kappa = 1.2\ \mathrm{J\ m^{-1}\ s^{-1}}$ and $C_{soil} = 2.0 \times 10^6\ \mathrm{J\ m^{-3}\ °C^{-1}}$ (Van Wijk, 1963) and therefore one can calculate typical diurnal and seasonal damping lengths $l = 1/\alpha$ (at the depth $x = l$ the amplitude of oscillations is $1/e \approx 0.37$ times of the amplitude at the surface); they are $l_d = 0.13$ m and $l_s = 2.5$ m. For real soils, parameters κ and C_{soil} have very large ranges and therefore damping length for the diurnal variation has approximately the range from 0.03 m to 0.16 m; the damping length for seasonal variations is $\sqrt{365} \cong 19$ times greater than the diurnal one.

Real soils have pore radius distributions that depend on many factors. In general, a large fraction of these pores have radius smaller