

DEPENDENCE OF BAROMETRIC PRESSURE, WIND VELOCITY AND TEMPERATURE ON THE VARIATION OF RADON EXHALATION

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ABSTRACT

An exhalation of radon depends on soil condition and weather condition strongly. A lot of researchers have emphasized importance of air pressure and soil moisture content as main factors of the variation. We have lasted a continuous measurements of radon exhalation rate together with soil conditions and meteorological elements for a long term in the Kanto Loam Layer that permeability was low relatively (10^{-12}m^2). In addition, we have applied a multi-phase, one dimension time dependent transportation model that described the transportation of radon in the soil to the experimental results. Based on both sides of the experiment and theory, the following results have provided about main factors influencing the variation of radon exhalation in the soil of low permeability.

An important factor making the pressure difference is the time variation of surface pressure. According to the calculation, the relation of a pressure difference and a pressure change depends on permeability remarkably. In very low permeability soil, a barometric pressure change gives small contribution to pressure difference. This makes pressure effect on an exhalation rate unclear.

Comparing the pressure differences obtained experimentally and theoretically, it was found that wind velocity has remarkable contribution to generation of a pressure difference. Accordingly contribution to an exhalation rate of wind velocity was estimated to be significant.

By introducing temperature characteristics of radon partition coefficient between soil air and water into a time dependent transport equation, temperature effect on radon exhalation was evaluated. This result showed that the effect of temperature is typically minor.

1. INTRODUCTION

Exhalation of radon(^{222}Rn) from soil has already been extensively studied (Wilkening 1974, Schery 1983, 1989, Dorr 1990, Kojima 1997). The influence of meteorological parameters is less well known although there are certain effects such as those due to changes in barometric pressure and water content in the soil (Owczarski, 1990). Although some researchers have pointed out the effects due to other parameters such as wind and temperature, it is not clear whether meteorological elements are major or minor factors affecting radon exhalation.

In the present work, the long-term continuous measurements of the radon exhalation were conducted at one site in combination with measurements of radon concentration in soil gas, soil temperature, pressure difference between the surface and 1 m depth, soil moistures content and meteorological parameters. Based on this large number of data, quantitative relations between the exhalation of radon and wind or surface temperature were found. To understand an underlying physical processes of wind and temperature effects, a comparison of

measurement with a multiphase radon transport model has been attempted.

2. EXPERIMENTA METHOD

The experimental arrangement (Fig.1) for measurement of radon exhalation consists of a closed circuit system with the following components, a) a flow-through accumulator, b) an air pump to circulate the air through the system, c) an ionization chamber of 1.5 l to measure of radon concentration, d) a membrane filter, which traps radon and thoron daughters and e) thoron decay chamber (Kojima 1997).

The flow-through accumulator is designed unattended repetitive measurements and is made of stainless steel with diameter of 40 cm and height of 29 cm. The opening of bottom 5 cm diameter of the accumulator was buried in the soil. The lid of the accumulator was automatically opened and closed by a controlled air compressor. At first, the lid of the accumulator was opened for introducing open air into the chamber and after 1 hour it was closed for accumulating the radon from the ground surface for 3 hours. The radon concentration of the sucked air from the closed accumulator was always monitored by an ionization chamber with counting intervals of 10 minutes. To eliminate the fluctuation of data, the averages of 60 readings of ionization current per one datum were adopted for calculation. The radon exhalation rate was determined by the linear increase of the counting rate up to three hours; within the accumulation time, back diffusion did not lead to a deviation from the straight increase of radon concentration. The data from the automated monitor were cross-checked by comparing with grab sampling data.

Soil temperatures at 50,100,150 cm depth and soil moisture contents (water fraction by volume) at 0.2 m and 0.5 m depth were measured with platinum sounding probes and soil moisture probes respectively. The moisture probes measure the total volume ratio of water in the soil by measuring the effective capacitance of the surrounding soil. Also, the pressure difference between the surface and 1 m depth was monitored with an accurate pressure gauge. At the surface, the meteorological data (wind direction, velocity, temperature, relative humidity, rain) were always monitored.

Measurements were carried out at a campus of the Science University of Tokyo, located in the suburbs of Tokyo. The soil is Kanto Loam Layer consisting of a fine loam. The soil air permeability was of the order of 10^{-12} m² under the ordinary water content that is more than 0.35 in volume fraction. The climate has a rainy season during June to July, a humid and hot summer monsoon during July to September and a cold and windy winter monsoon during December to February. To eliminate the influence of soil disturbance by the installation of the equipment, data for 6 months after the landfill were not used.

3. RADON TRANSPORT EQUATIONS

Radon is emanated from solid grains and the emanated radon further is distributed between the water and gas phases in the soil pores. Also, the radon transports primarily by diffusion and advection mechanisms in soil pores. So, multiphase radon generation and transport model is required to simulate radon transport under realistic conditions (Rogers 1991a).

The radon (C_r) balance equation for the pore air is given by:

$$\frac{\partial \varepsilon_a C_a}{\partial t} = \frac{\partial}{\partial z} \left(D_a \frac{\partial C_a}{\partial z} \right) + \frac{\partial}{\partial z} \left(\frac{\kappa}{\mu} \frac{\partial P}{\partial z} C_a \right) - \lambda \varepsilon_a C_a + \lambda R \varepsilon_s \rho_s E_a + T_{wa} - T_{as} \quad (1)$$

where

$\varepsilon_a, \varepsilon_w, \varepsilon_s$ = Volume ratios of pore air, water and solid respectively.

D_a = Radon bulk diffusion coefficient ($m^2 s^{-1}$)

κ = Air permeability (m^2)

μ = Dynamic viscosity (Pa s)

$\frac{\partial P}{\partial z}$ = Pressure difference between surface and depth z

λ = Decay constant (s^{-1})

R = radium concentration in soil material ($Bq kg^{-1}$)

ρ_s = Bulk dry density ($kg m^{-3}$)

E_a and E_w = Coefficients of radon emanation into the pore air and water

T_{as} = Transfer factor of radon from pore air to solid surface ($Bq m^{-3} s^{-1}$)

T_{wa} = Transfer factor of radon from pore water to pore air ($Bq m^{-3} s^{-1}$)

The first term on the right side represents diffusion in the pore air, the second term, advection, the third term, radioactive decay and the fourth, the radon source. The five and six terms represent transfer factors of radon from water to pore-air and from pore air to solid surface.

In similar manner, the equations for radon in the soil water and for the radon adsorbed on the soil surface. The three equations can be transformed an equivalent equation in single-phase (Rogers 1991).

$$\frac{\partial f_s C_a}{\partial t} = \frac{\partial}{\partial z} \left(D \varepsilon_p \frac{\partial C_a}{\partial z} \right) + \frac{\partial}{\partial z} \left(\frac{\kappa}{\mu} \frac{\partial P}{\partial z} C_a \right) - \lambda f_s C_a + \lambda R \varepsilon_s \rho_s E \quad (2)$$

where

$f_s = \varepsilon_a + \varepsilon_w k + \varepsilon_s \rho_s k_a$

$E = E_a + E_w$

The boundary and initial conditions are

$C(z=0, t) = 0$, $C(z=\infty, t) = \phi / \lambda$, $C(z, t=0) = f(x)$,

where we use a solution in steady state as $f(x)$.

The above equations can not easily be solved analytically even in case of simplified problems. Therefore, numerical solutions for these equations are obtained by mean of the finite difference method and the Crank-Nicholson technique.

The radon exhalation (F) can be obtained from the following equation.

$$F = -(D_a + D_w \kappa) \frac{\partial C_a}{\partial z} - \frac{\kappa}{\mu} \frac{\partial P}{\partial z} C_a \quad (3)$$

Since the exhalation rate is the total radon flux at the surface ($z=0$), the second term on the right side vanishes due to the boundary condition.

4. RESULTS

4.1 Barometric pressure and wind effect

Figure 2 shows relation between radon exhalation and barometric pressure (above) and pressure difference (below) between surface and 1 m depth, measured during the Mar. 1998. We can not find a statistically significant correlation between radon exhalation and barometric pressure. Measurements in different months show a similar tendency. Many experiments (Schery 1982,1984, Duence 1987) have shown the existence of a significant pressure effect and mathematical models (Clement 1974, Cripps 1996) have predicted such an effect. A main reason for the apparent no pressure effect in our experiments may be due to very low air permeability of soil at this site. Although a pressure change contributes more or less to exhalation rates, an extent of the effect closely depends on the soil conditions. This fact can easily be shown by a model calculation.

Meanwhile, our experiment shows on the other hand that the pressure difference, induced by a pressure change, is related to exhalation with correlation coefficient of -0.56 . An existence of the pressure difference effect is not surprising because the pressure difference between surface and underground has direct effect on a transport of radon in soil air.

Figure 3 shows the statistical aspect of the effect of wind on radon exhalation in February and March 1998. A significant correlation between exhalation and wind velocity is evident. This may suggest that wind velocity is one of contributors to develop the pressure difference together with pressure change. The increase of wind velocity induces negative pressure difference by lowering surface pressure; the effect results in the increasing upward flow of soil air. This is consistent with the results of a positive correlation between radon exhalation rate and wind velocity. However, the data include the other effects produced by parameters such as a barometric pressure change, which is often associated with the wind velocity. Decreasing barometric pressure as passing a cold front usually accompanies a severe wind.

In order to divide the effects of wind and barometric pressure, we calculate the induced pressure field in soil due to the change of surface pressure. The pressure field in the soil can be obtained by solving the following a pressure equation (Clement et al. 1974,Cripps 1996).

$$\frac{\partial^2 p(z,t)}{\partial z^2} = \frac{\varepsilon \eta}{\kappa(t)P_0} \frac{\partial p(z,t)}{\partial t} \quad (4)$$

In the above equation, atmospheric pressure, P is divided into the mean pressure, P_0 and small pressure perturbation, p . The above equation is numerically solved.

Figure 4 shows time dependent measurements for radon exhalation, wind velocity, barometric pressure, and the measured pressure difference between ground surface and 1 m depth, during a period of February 16 to 19, 1998. We easily see, from the above trace in Figure4, that some sharp peaks of radon exhalation just correspond to those of wind velocity. Wind velocity seems a major factor affecting exhalation in view of time series of data. In below trace in Figure 4 , the simulated pressure differences calculated by using equation (4) and the measured barometric pressures are plotted with measured data. The simulated pressure differences are relative values and are normalized to the levels of experimental pressure differences. Experimental pressure differences can consider to be induced by pressure difference, wind velocity and other factors. On the other hand, simulated pressure differences, of course, depend on only pressure difference. It is therefore considered that discrepancy between both pressure differences is mainly due to wind velocity. From comparison of experimental pressure differences and theoretical values, it is found that a large discrepancy between both values appears when increasing wind velocity like those at noon on Feb.17 and 18. The remarkable discrepancy ($1\sim 2\text{Pa/m}$ in the range of wind velocity to 6 m/sec) is comparable with the variation range (about 2Pa/m) generated by pressure change

during 4 days. We can consequently concluded that wind is a major factor affecting radon exhalation at our measurement site with soil air permeability of the order of 10^{-12}m^2 . Unfortunately, we have not succeeded to incorporate the wind effect into a radon transport equation in soil.

4.2 Temperature effect

Figure 5 shows the scatter plot of radon exhalation rates versus surface temperature during the period of February and March 1998. A trend of increased exhalation with increased surface temperature is seen with the present data. There are several possible physical explanations for the presence of the temperature effect. Based on the kinetic theory and thermal expansion id soil air, Shery(1983) concluded that for typical diurnal temperature variation this effect is small. A thermally induced convection has been proposed as a factor affecting diurnal variation in exhalation (Malakohv 1966). Shery (1983) also has showed that such an effect would not occur or would be too small to detect.

To explain large seasonal variations of radon in soil air, radon partitioning between soil air and water has been proposed (Washington and Rose 1990). The partition coefficient (Dstwarld Cefficient) of radon between water and air is temperature sensitive (Battino 1979). In order to the relative importance of radon partitioning, radon exhalation rates are calculated by introducing temperature dependence of partition coefficient into a time-dependent transport equation (Equation (4)).

Figure 6 shows the time varying oscillatory radon exhalation rate when the surface temperature has sinusoidal time variation with a period of one day and the amplitude of 20°C . The soil temperature is given to be constant value of 15°C at 1 m depth and the soil temperature profile between the surface and this constant temperature depth is linearly interporated. In this calculation, we use the values of 10^{-12}m^2 for soil air permeability and 0.35 for water content respectively. For 20°C surface temperature variation, the exhalation rate is varied by about 10 % under our soil condition. Taking into consideration of the given temperature profile in soil, our result will be evaluated overestimatly. The effect of temperature seems to be typically minor.

Of meteorological and soil factors, barometric pressure, wind and water content in soil can have major effects. We attempt to simulate the time varying radon exhalation by using the measured data of major factors affecting exhalation. The top trace of Figure 7 shows the predicted and measured radon exhalation rates for a 4-day period. The middle traces show water contents at 0.2 m depth and 0.5 m depth, and pressure difference between surface and 1 m depth, used in this simulation; the bottom is surface temperature. As the pressure difference is induced by barometric pressure change and wind, the predicted exhalation reflects the effect of barometric pressure, wind and water content. Meanwhile temperature effect is ignored. It is found, from the results, that the time variation in the exhalation rate predicted by the transport model and data of major factors affecting exhalation fairly agree with those observed experimentally.

5. CONCLUSIONS

This paper has presented the experimental and theoretical results on the effects of barometric pressure, wind and temperature affecting radon exhalation. Wind has a major effect and contribution is often the same extend as barometric pressure change, whereas the

effect of temperature is typically minor.

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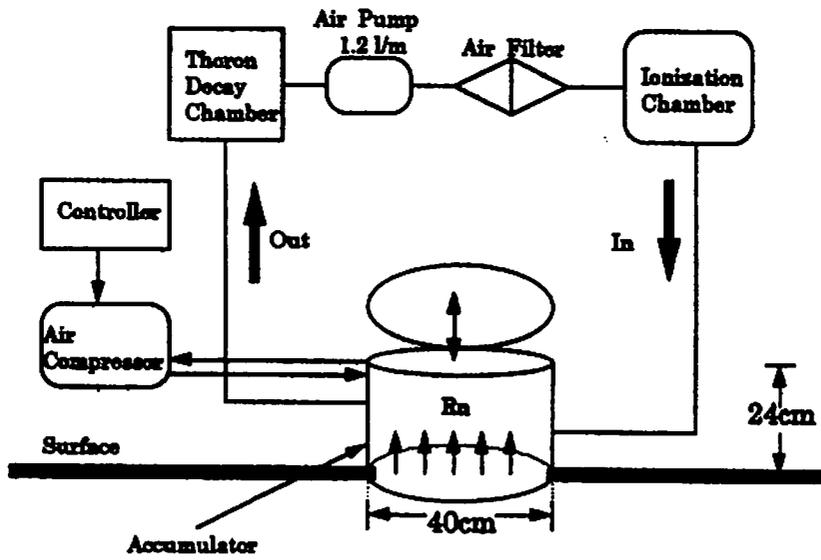


Fig.1. Experimental arrangement of radon exhalation.

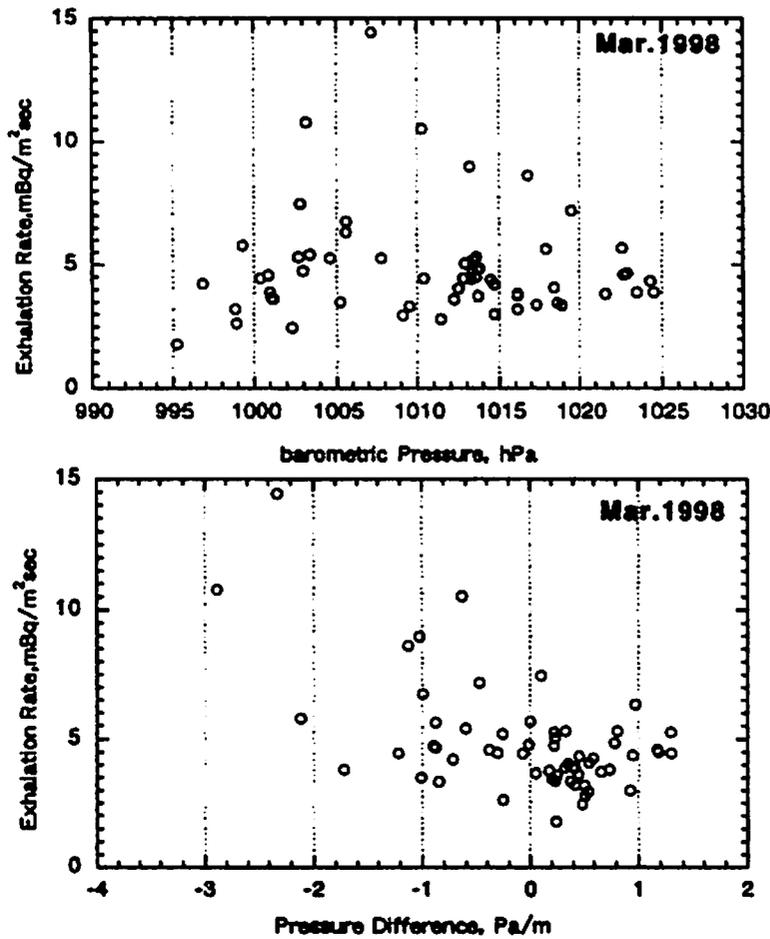


Fig.2. Relation between exhalation rates and barometric pressure (above) and pressure difference(below), during Mar.1998

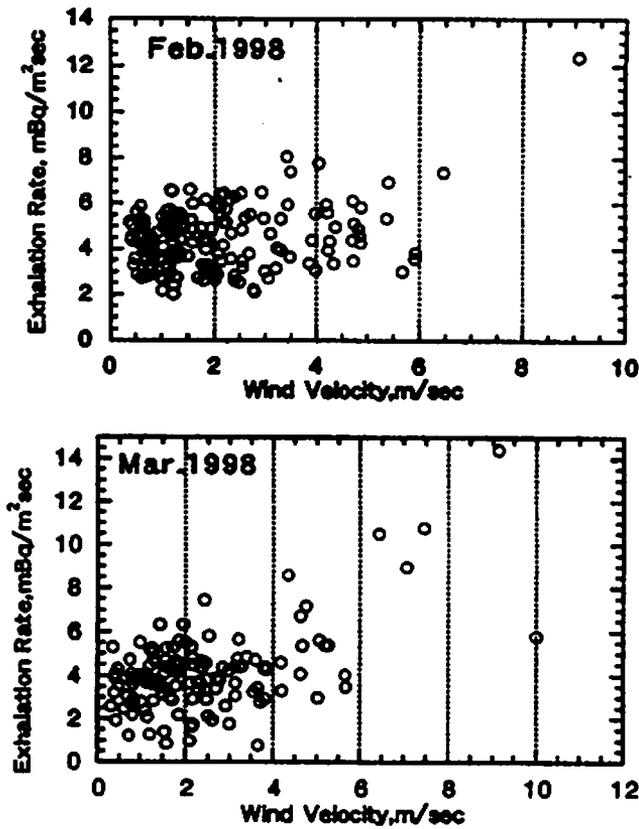


Fig. 3. Scatter plots of radon exhalation versus wind velocity.

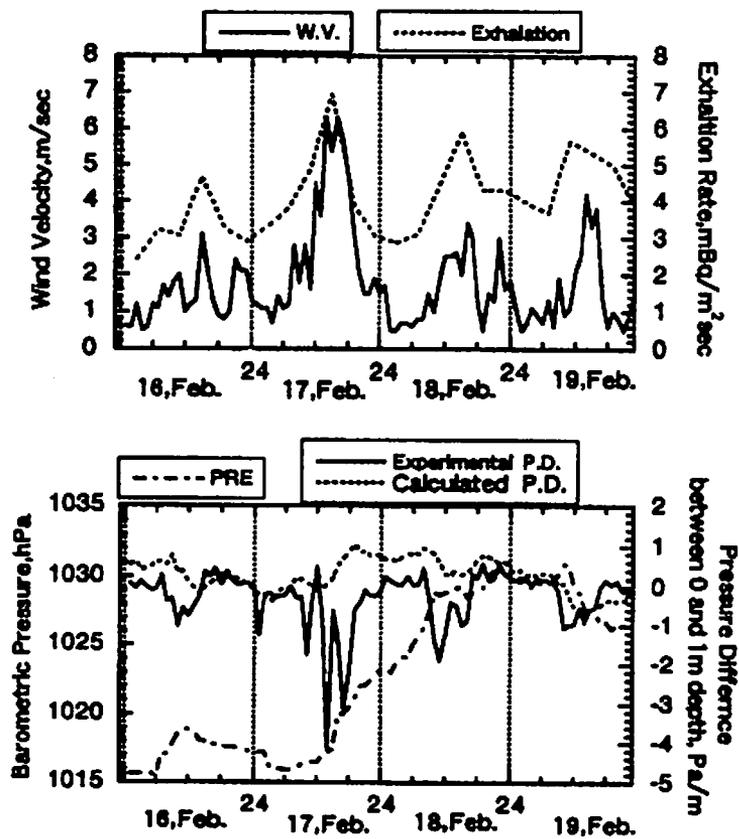


Fig. 4. Time dependent measurements for exhalation, wind velocity (W.V.), barometric pressure (PRE) and pressure difference (P.D.) between surface and 1 m depth.

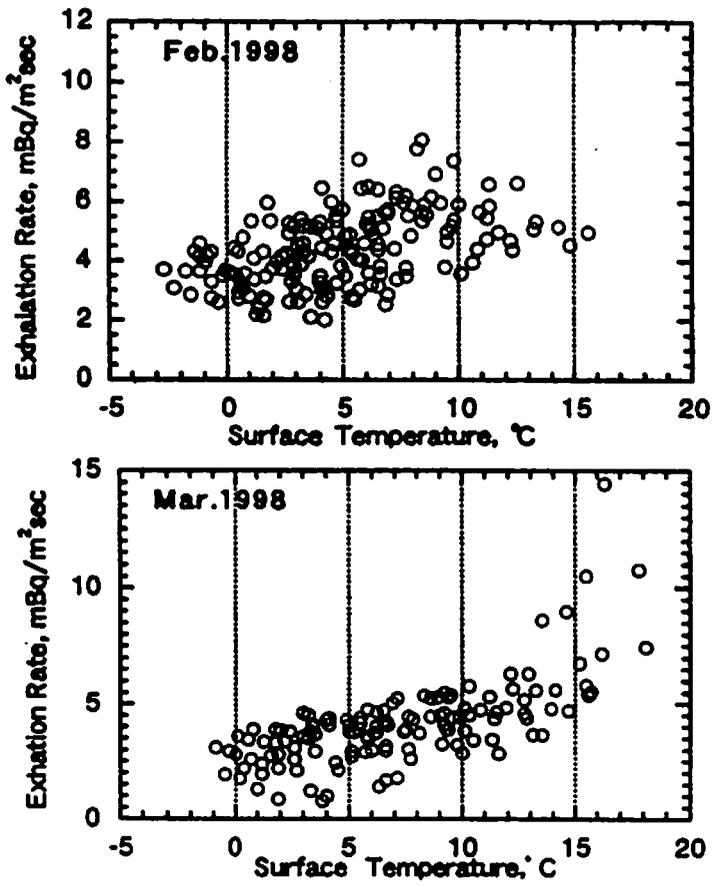


Fig.5. Scatter plots of exhalation and surface temperature.

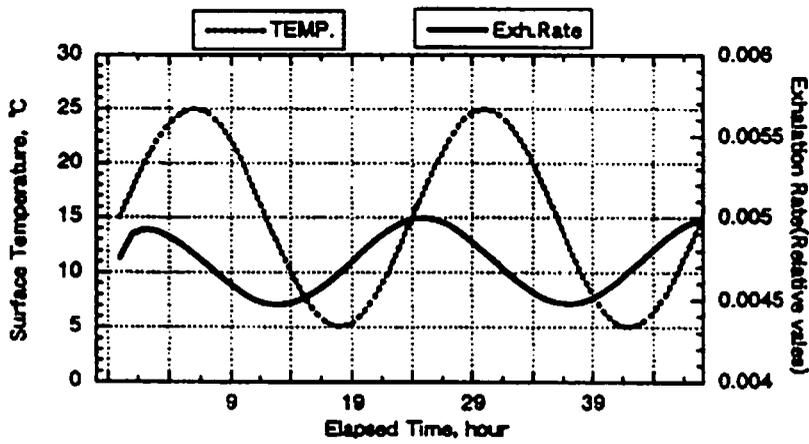


Fig.6. calculated radon exhalation rate when the surface temperature has sinusoidal variation.

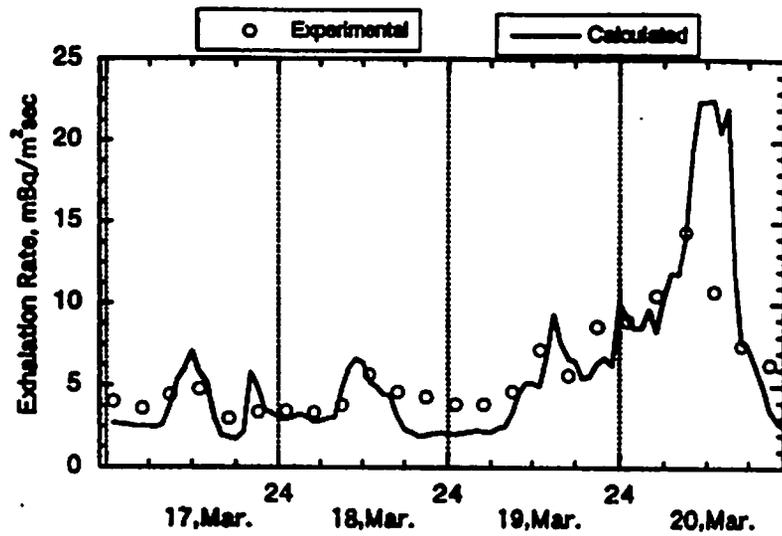


Fig.7(a). Time variation of experimental and simulated radon exhalation rates.

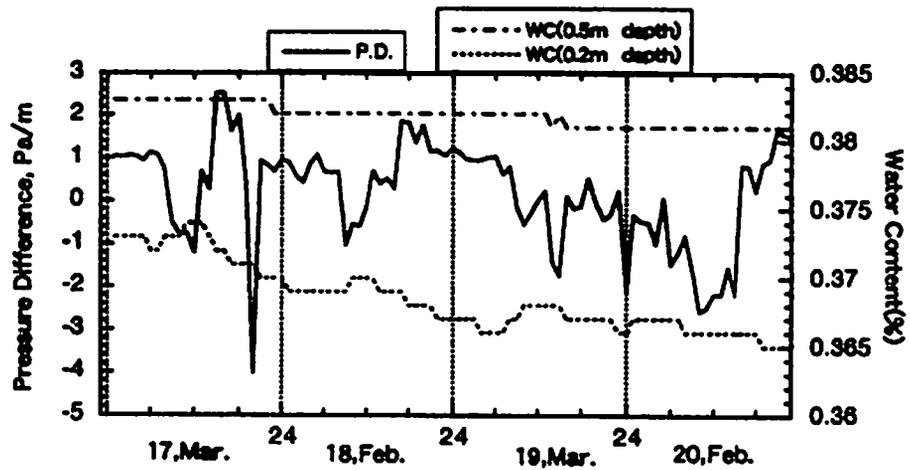


Fig.7(b). Time variations of hourly data of pressure difference and water content at 0.2 and 0.5 m depths, used for this simulation.