

Modelling of radon-222 as a proxy for vapour phase transport in the unsaturated zone

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Abstract This study uses naturally occurring ^{222}Rn as a tracer in the unsaturated zone to better understand volatile organic compounds (VOC) release, transport and interaction. Using the STELLA Research software package, a two-dimensional model of ^{222}Rn transport and generation in the unsaturated zone was produced. The model was validated using a laboratory facility comprising of a $2 \times 1 \times 0.2$ m layered chamber. In these experiments transport by diffusion is measured for a homogeneous column of material. The model described the ^{222}Rn concentrations satisfactorily for both simple and complex experiments. Sensitivity analysis and simulated results demonstrate the importance of moisture content in the transport of ^{222}Rn . The simulations show excellent comparison with experimental data and provide good estimation of ^{222}Rn activity and VOC transport in the unsaturated zone.

INTRODUCTION

There has been widespread concern on the mobility of volatile organic compounds (VOCs) and their potential to impact surrounding ecosystems and groundwater. In the USA many groundwater supplies have been threatened by contamination from VOCs and studies by numerous researchers (Abriola & Pinder, 1985; Sleep & Sykes, 1989; Mendoza & Frind, 1990; Conant *et al.*, 1996) have investigated the movement of such contaminants downwards to the water table. However, the direct transfer from underground sources to houses and the atmosphere can also pose a potential health hazard from contaminated sites. A better understanding is needed of the physical processes governing VOC release, transport in the unsaturated zone and interaction with the subsurface environment.

^{222}Rn gas, unlike other volatile pollutants, is chemically inert but it is highly mobile and responsive to many of the same processes that affect VOC movement in the unsaturated zone. ^{222}Rn is a naturally occurring radioactive gas, formed in the decay series of uranium-238, and has a half-life of 3.825 days. In a number of studies over the past decades ^{222}Rn has been used as a natural tracer to investigate gas phase transport under natural conditions in soils (Tanner, 1964; Rogers & Nielson, 1991; Tsang & Narasimhan, 1992; Chen & Thomas, 1995). Although models have been developed to describe ^{222}Rn transport in the unsaturated zone they are often restricted to simple conditions (Chen & Thomas, 1995). Models that have included barometric pressure and air flow do not include moisture content, chemical transport, decay processes or sorption, which are often retardation mechanisms for VOC transport (Massmann & Farrier, 1992; Chen & Thomas, 1995). Many investigations of ^{222}Rn transport have only considered transport in the gas phase through dry homogeneous material (Van der Spoel *et al.*, 1997) whereas, in reality, conditions in the field are inhomogeneous and unpredictable.

There has been significant research into the development of the theoretical framework for the transport of volatile organic compounds and pesticides through the unsaturated zone to groundwater supplies (Jury *et al.*, 1983; Sleeps & Sykes, 1989; Mendoza & Frind, 1990; Simunek & Suarez, 1993). Here we modify these methods to obtain an integrated model for ^{222}Rn transport in the unsaturated zone. The inverse solution of this model could then be used to constrain models of VOC transport in the unsaturated zone.

MODEL DEVELOPMENT

We assume that ^{222}Rn transport can take place in both the gas and liquid phase in the unsaturated zone. Two processes dominate ^{222}Rn transport, diffusion in both the gas and liquid phase and advection in the gas phase. Hydrodynamic dispersion can be ignored due to the short half-life of ^{222}Rn (Jury *et al.*, 1983), and surface diffusion of ^{222}Rn in the sorbed state is negligible (Rogers & Nielson, 1991). The one-dimensional (1-D) ^{222}Rn transport equation with radioactive decay and a constant source term are described by the following equation:

$$\frac{\partial C_T}{\partial t} = -\frac{\partial}{\partial x}(J_{dg} + J_{dl} + J_{ag}) - \lambda C_T + Q \quad (1)$$

where J_{dg} and J_{dl} are the diffusive flux densities in the gas and liquid phase respectively [$\text{Bq m}^{-2} \text{h}^{-1}$]; J_{ag} is the advective flux density in the gas phase [$\text{Bq m}^{-2} \text{h}^{-1}$]; Q is the ^{222}Rn production term; λC accounts for radioactive decay of ^{222}Rn ; and C_T is the total ^{222}Rn concentration in the unsaturated zone [Bq m^{-3}]. The contributions from the solid, liquid and gas phases are described as:

$$C_T = pbC_s + \theta_L C_L + \theta_g C_g \quad (2)$$

where C_s , C_L and C_g are the concentration of the contaminant in the sorbed, liquid and gaseous phases respectively [Bq m^{-3}]; pb is the bulk density of the material [kg m^{-3}]; and θ_L and θ_g are the liquid and gas porosities (dimensionless); J_{dg} , J_{dl} and J_{ag} are defined as follows:

$$J_{dg} = -\theta_g D_g \frac{\partial C_g}{\partial x} \quad (3)$$

$$J_{dl} = -\theta_L D_L \frac{\partial C_L}{\partial x} \quad (4)$$

$$J_{ag} = V_a C_a \quad (5)$$

D_g and D_L are the effective diffusion coefficients of ^{222}Rn gas in the gas and liquid phases respectively [$\text{m}^2 \text{h}^{-1}$] and V_a is the vapour phase velocity of the soil gas [$\text{m}^2 \text{h}^{-1}$]. Substitution of equations (2) to (5) into equation (1) gives the following:

$$\frac{\partial (pbC_s + \theta_L C_L + \theta_g C_g)}{\partial t} = \theta_g D_g \frac{\partial^2 C_g}{\partial x^2} + \theta_L D_L \frac{\partial^2 C_L}{\partial x^2} - V_a \frac{\partial C_a}{\partial x} - \lambda C_T + Q \quad (6)$$

Expressing C_T in terms of C_g gives:

$$\frac{\partial C_g R_g}{\partial t} = D_E \frac{\partial^2 C_g}{\partial x^2} - V_a \frac{\partial C_g}{\partial x} - \lambda C_g + Q \quad (7)$$

where R_g is the ^{222}Rn retardation factor which accounts for equilibrium between the phases, and is defined as:

$$R_g = pb \frac{K_d}{H} + \frac{\theta_L}{H} + \theta_g \quad (8)$$

where H is Henry's constant (dimensionless); K_d is the solid-aqueous partition coefficient [$\text{m}^3 \text{kg}^{-3}$]; and D_E is the effective diffusion coefficient that combines diffusion coefficient in the gas and liquid phases D_g and D_L and is given as:

$$D_E = \theta_g D_g + \theta_L \frac{D_L}{H} \quad (9)$$

D_g and D_L coefficients are $0.0396 \text{ m}^2 \text{ h}^{-1}$ and $3.6 \times 10^{-6} \text{ m}^2 \text{ h}^{-1}$ respectively; H for ^{222}Rn at 20°C is 0.245. Equation (7) is for 1-D multiphase ^{222}Rn generation decay and diffusive-advective transport. We expand equation (7) to the two-dimensional (2-D) with advection only in the vertical direction to give:

$$\frac{\partial C_g R_g}{\partial t} = D_E \frac{\partial^2 C_g}{\partial x^2} + D_E \frac{\partial^2 C_g}{\partial y^2} - V_a \frac{\partial C_g}{\partial x} - \lambda C + Q \quad (10)$$

We use equation (10) to model ^{222}Rn generation and transport in the unsaturated zone. Using the software STELLA Research, equation (10) was numerically solved in 2-D by finite difference for the following boundary conditions: when $t = 0$, concentration of ^{222}Rn is zero ($t = 0, C_g = 0$; for all x and y); at the surface (h), gas concentrations are zero for all time ($x = h, C_g = 0$; for all t); and at the bottom of the profile, an impermeable boundary with no flow exists ($\delta C_g / \delta x = 0, x = 0$; for all t).

Parameters used in modelling

A laboratory facility consisting of a perspex column of dimensions $2 \times 1 \times 0.2 \text{ m}$, with a steel framework, was set up to validate the model. There are 18 sampling points positioned on the column and a water inlet/outlet placed at the bottom. The column consists of six separate layers so modelling of ^{222}Rn transport through heterogeneous material can be simulated. ^{222}Rn concentrations are determined using a portable radiation monitor and Lucas scintillation cells. The cells were tested for reproducibility and efficiency prior to sampling (Gibbons & Kalin, 1997). The efficiency of the cells ranged from 45 to 77%.

In experiment 1, crushed granite was placed in the lower most layer. Dry homogeneous sand of grain size $780 \mu\text{m}$ and porosity 0.425 is placed on top of the granite. For experiment 2, a different sand with grain size $1400 \mu\text{m}$ and porosity 0.410 was placed on top of the granite. In experiment 3, water was rapidly introduced into the granite layer when steady state conditions in experiment 2 were achieved. ^{222}Rn

measurements were taken at 7, 24, 48, 72, 168, 312, 360, 408 and 504 h. The physical characteristics of both the sand and granite and properties of ^{222}Rn are listed in Table 1. In experiment 3, an aqueous–gaseous partition coefficient (Henry's constant) of 0.23 was used.

Table 1 Properties of ^{222}Rn , sand and granite used in experimental studies.

	Granite	Sand A (experiment 1)	Sand B (experiments 2 + 3)
Grain size(μm)	900	780	1400
θ_T	0.476	0.425	0.410
Bulk pb (kg m^{-3})	2630.0	2178.8	2570.0
^{222}Rn exhalation rate (Bq s^{-1})	8.13×10^{-5}	6.65×10^{-6}	1.73×10^{-5}
Degree of saturation, S_r	1	N/A	0

RESULTS AND DISCUSSION

For experiment 1, ^{222}Rn concentrations as a function of time at different heights are presented in Fig. 1. The solid lines in Fig. 1 represent simulated calculations from the model based on input parameters from Table 1, and the markers indicate experimental data.

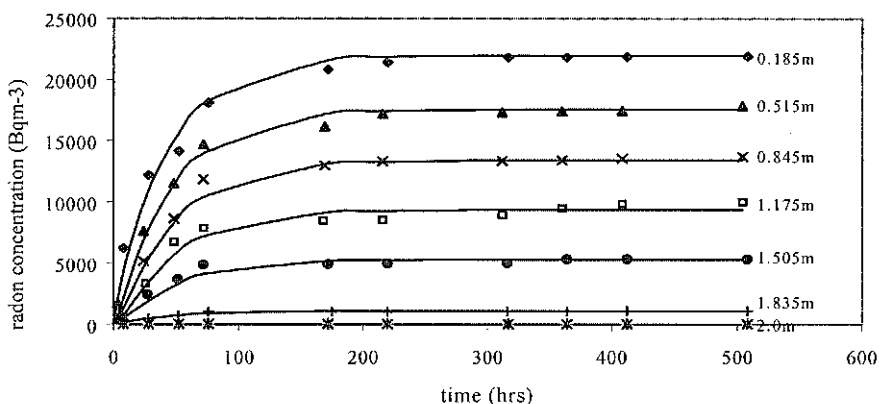


Fig. 1 ^{222}Rn concentration as a function of time for different x coordinates for experiment 1. Straight lines are simulated results and markers indicate observed data.

It is seen that the measured ^{222}Rn concentrations follow a reasonably smooth curve over the test period, with notable exceptions to this being in layer 3 (0.845 m) at day 4, where the observed measurement is higher than the simulated result, and in layer 4 (1.175 m) where the simulation underestimates concentration until day 4 of the study. From Fig. 1 it can be seen that equilibrium is reached at about 170–200 h. In experiment 2 we changed the grainsize and porosity of the sand and this resulted in a higher ^{222}Rn concentration in the column. The model predictions for all six layers were higher than what is actually observed. Such overestimation by the model for all the layers may indicate that there is a discrepancy in the input parameters as these

calculations were carried out independently. Therefore, because of the difference found with the experimental data, a sensitivity test of porosity was conducted. Simulations were carried out with porosity of the sand varying from 0.38 to 0.42 and it was found that a porosity of 0.39 for the sand gave good correlation with the observed data. The experimental and simulation results, as a function of time, are illustrated in Fig. 2 and again equilibrium is reached in the column at between 175 and 200 h.

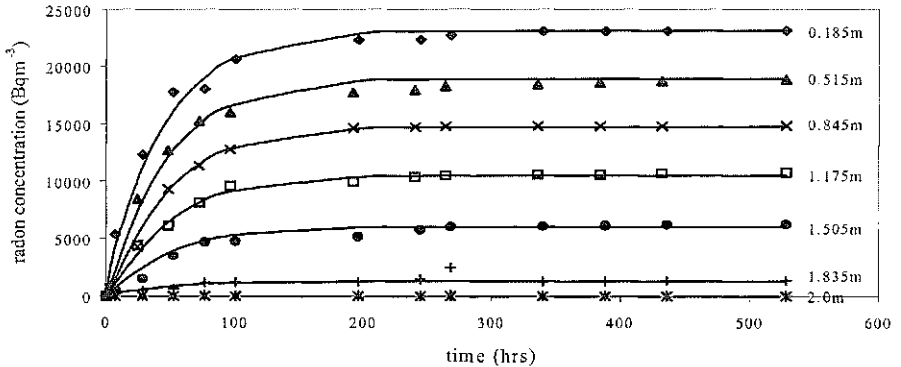


Fig. 2 ^{222}Rn concentrations as a function of time for different x coordinates for experiment 2. Markers indicate experimental results.

In the third experiment water was added to the first layer in the column after steady state conditions were achieved. The model predicted that there would be a rapid decline in ^{222}Rn activity with the introduction of water and this is what was observed. Figure 3 illustrates the results of the experimental data relative to the simulated data.

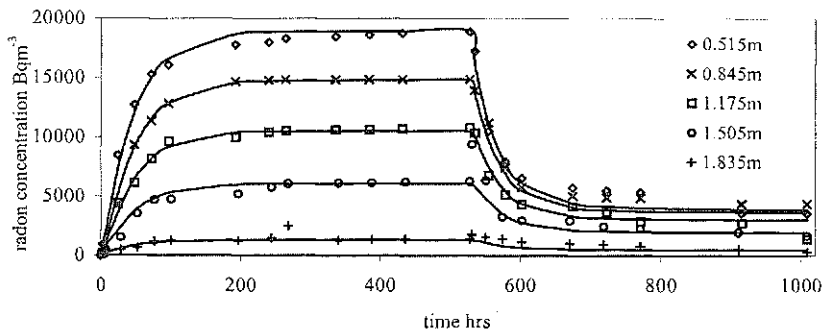


Fig. 3 ^{222}Rn concentrations as a function of time for different x coordinates; water was added to the system after 528 h. Markers indicate experimental data.

The model gives good agreement with the observed results except in layer 2. The higher concentrations observed may be a result of a capillary fringe effects. Figures 4(a) and 4(b) illustrate the correlation between the model simulations and observed data. From the graphs it is seen that the model provides excellent agreement with observed results as data points lie between 98.79% and 99.53%.

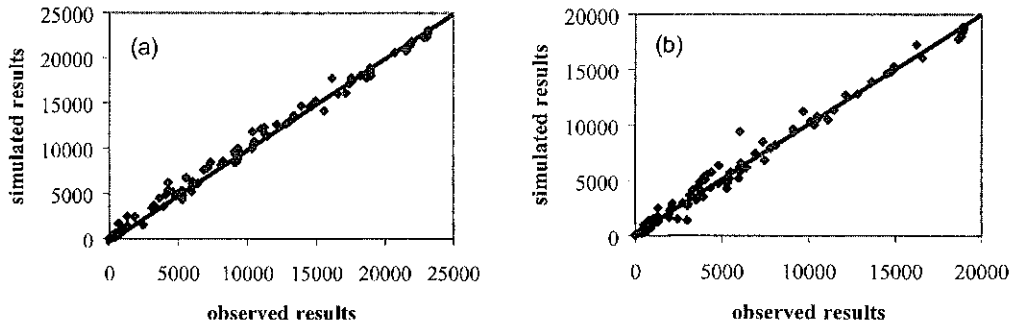


Fig. 4 Correlation between observed and simulated results: (a) experiments 1 + 2, (b) experiment 3.

CONCLUSIONS

This report describes a 2-D model for ^{222}Rn transport in the unsaturated zone. Measurements obtained from experimental studies were compared to results from simulations and these indicate that STELLA can provide an excellent representation of ^{222}Rn transport in the unsaturated zone. Model simulations and experimental studies indicate that moisture content has an important role to play in the transport of ^{222}Rn . With continuing experiments we will validate the model for ^{222}Rn transport and VOC transport so that the model may have broader applications to VOC transport in the unsaturated zone.

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