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Comparison of two numerical modelling approaches to a field experiment of unsaturated radon transport in a covered uranium mill tailings soil (Lavaugrasse, France)

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ABSTRACT

Uncertainties on the mathematical modelling of radon (^{222}Rn) transport in an unsaturated covered uranium mill tailings (UMT) soil at field scale can have a great impact on the estimation of the average measured radon exhalation rate to the atmosphere at the landfill cover. These uncertainties are usually attributed to the numerical errors from numerical schemes dealing with soil layering, and to inadequate modelling of physical processes at the soil/plant/atmosphere interface and of the soil hydraulic and transport properties, as well as their parameterization. In this work, we demonstrate how to quantify these uncertainties by comparing simulation results from two different numerical models to experimental data of radon exhalation rate and activity concentration in the soil-gas measured in a covered UMT-soil near the landfill site Lavaugrasse (France). The first approach is based on the finite volume compositional (i.e., water, radon, air) transport model TOUGH2/EOS7Rn (Transport Of Unsaturated Groundwater and Heat version 2/Equation Of State 7 for Radon; Saâdi et al., 2014), while the second one is based on the finite difference one-component (i.e., radon) transport model TRACI (Transport de RAdon dans la Couche Insaturée; Ferry et al., 2001). Transient simulations during six months of variable rainfall and atmospheric air pressure showed that the model TRACI usually overestimates both measured radon exhalation rate and concentration. However, setting effective unsaturated pore diffusivities of water, radon and air components in soil-liquid and gas to their physical values in the model EOS7Rn, allowed us to enhance significantly the modelling of these experimental data. Since soil evaporation has been neglected, none of these two models was able to simulate the high radon peaks observed during the dry periods of summer. However, on average, the radon exhalation rate calculated by EOS7Rn was 34% less than that was calculated by TRACI, and much closer to the measured one for physically-based soil radon diffusion models. Unlike TRACI, EOS7Rn was able to simulate qualitatively seasonal variations of both radon exhalation and concentration. These results show that EOS7Rn produces less numerical errors than TRACI, and can be considered as a promising model for predicting radon transport in the landfill, if soil evaporation is modelled and its numerical inversion for parameter estimation is realized.

1. Introduction

Few modelling studies have been reported in the literature for simulating in-situ experiments of transient two-phase (water–air) flow and radon transport in covered uranium mill tailings soils at landfill sites. Data-model comparison is very complicated because of the need to characterize double porosity media (e.g., macropores) and to construct the local history of climate, hydrology and bio-geo-chemistry of an UMT-landfill site.

First transient simulations of unsaturated radon transport in UMT-landfill soils began with the works of Gee et al. (1984), Mayer et al. (1981), Mayer and Gee (1983), and Simmons and Gee (1981) who showed the importance of the long-term moisture content in multidimensional numerical simulations for predicting the long-term radon activity concentration in the soil-gas phase and exhalation from the long-term climatic history of the site. In their works, however, Richards' approximation for the two-phase flow problem has been considered and no data-model comparison has been made.

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The works of Ferry (2000) and Ferry et al. (2001, 2002) are ones among seldom studies on data-model comparison at transient field conditions. They carried out during four years (from December 1997 to September 2000) experiments of radon transport in two lysimeters, and in an artificial pond filled by covered and uncovered UMT materials to study the cover material (CM) effectiveness on radon mitigation at the surface. These experiments helped them to characterize both materials and to study radon transport in the covered-UMT of the Lavaugrasse landfill site. In their experiments climatic and soil conditions were monitored every half an hour to an hour. For simulating their experimental data, they used the one-dimensional (1D) finite difference code TRACI. For the artificial pond experiment, although the shape and scale parameters of the UMT and CM hydraulic properties have been calibrated to account for CM-compaction and UMT-shrinkage (i.e., cracked UMT) during summer periods, TRACI simulations usually showed an overestimation of the transient measured radon exhalation rate at the surface of the covered and uncovered UMT materials. For the uncovered UMT-material experiment, the authors attributed the slight overestimation of measured radon exhalation rates during drainage of water after prolonged rainfall events to the hysteresis effect due to shrinkage and swelling of the UMT-material, which are not taken into account in the model. However, no sensitivity analysis has been performed to choose for the adequate mathematical model of radon diffusion in the unsaturated porous materials. It has been assumed that the well-known Rogers and Nielson (1991a) (RN) empirical formula is valid for these porous materials. However, as demonstrated by Saâdi (2014), this formula can be unphysical to represent radon transport, especially in shallow subsurface environments. Moreover, there has been no comparison between measured and simulated transient radon activity concentration in the soil-gas for the model discrimination. Additionally, no numerical verification of the model TRACI has been made to conclude about its accuracy in dealing with transient radon transport in layered unsaturated porous materials. As shown in Ferry (2000), even for homogeneous soils, the numerical verification of the transient 1D two-phase flow and radon transport problems was not very convincing, since it was limited to a comparison between TRACI and numerical data from the literature. Other mechanisms like temperature, evaporation and dewing can also be of primary importance for improving the simulation of the radon exhalation rate at the covered UMT-material, but have been neglected in the TRACI-modelling approach.

In this study, the artificial pond experiment carried out by Ferry et al. (2002) will serve as a valuable check of the accuracy of model and parameter uncertainty. We aim to demonstrate how the numerical modelling of this experiment can be enhanced when using a finite volume numerical model, EOS7Rn (Saâdi et al., 2014), with the same hydraulic and radon source properties of the CM and UMT materials and their parameterization, as implemented in TRACI, but with a physically-based three-components diffusion model.

2. Materials and methods

2.1. The experimental pond set-up

The small artificial pond (Ferry et al., 2002), with dimensions of 14 m × 15 m and height of 1.8 m, is representative of the Lavaugrasse site and was designed to reproduce a UMT disposal facility on a small scale and to study the cover material effectiveness. The radon exhalation rate at the surface was first studied at the surface of a 0.8 m layer of the UMT. A 1 m layer of the cover material, compacted every 0.6 m, was then placed on top of the first layer to study its effect on radon exhalation coming from the UMT.

Based on a textural analysis, the CM and UMT layers were characterized as loamy sand and sandy silt soils, respectively. Their hydraulic properties, i.e. their water retention curve and effective unsaturated permeability, were determined from their fine texture and calibration of the two-phase flow problem to measured soil water saturation and capillary pressure.

Characterization of the radon source properties of both materials, i.e. determination of their radium activity concentration and emanation coefficient, was carried out by laboratory methods. The first property was measured by γ-spectrometry, with a relative measurement uncertainty close to 7 and 25% for CM and UMT-soils, respectively. The radon emanation coefficient was measured by an accumulation technique in 10 dry samples of the CM-soil, whereas that of the UMT was measured by different measurement techniques in dry and wet conditions (Ferry, 2000). The relative measurement uncertainty was close to 20%.

Continuous observations of water flow and radon transport in the experimental devices were carried out in situ. Capillary pressures were measured with tensiometers (Jetfill, soilmoisture Corp., USA) at different depths in the UMT- and CM-layers, with a relative measurement error less than 1%. The moisture content in the CM-layer was deduced from permittivity measurements by dielectric probes (HMS 9000, SDEC, France). The probe calibration can result in an absolute error equal to 0.02 $m^3\ m^{-3}$. Radon concentration in the gas phase of the UMT and CM was measured by a probe using a solid state silicon detector placed in a specially designed measurement chamber (head of BARASOL; Algade, France). The relative uncertainty on the measured radon activity concentration is about 10%. It accounts for measurements errors due to count rate and probe calibration. A data logger monitored all of these measurements every 30 min. Radon flux densities were measured every three hours using an automated accumulation chamber containing an AlphaGUARD (Genitron Instruments, Germany). The detection limit of this apparatus is 10 mBq $m^{-2}\ s^{-1}$, whereas the relative error is between 20 and 40% for the range of values measured in this experiment.

Meteorological data were recorded every half an hour and included rainfall, atmospheric air pressure and temperature, relative air humidity, wind speed and sunshine.

2.2. The mathematical models

2.2.1. TRACI

This numerical model (Ferry, 2000; Ferry et al., 2002) uses a fully implicit finite difference numerical method to solve the isothermal 1D-vertical mass conserving equations of the two-phase (water–air) Darcy-flow and radon Fick-transport problems. TRACI calculates capillary pressure and moisture content profiles in a soil subject to meteorological conditions, radon concentration in the soil gas-phase and the radon flux at the surface, in relation to time.

The two-phase flow problem is formulated by the two following equations governing flow of liquid and gas phases in the soil pores in terms of their respective matric head potentials, h_l (m) and h_g (m), as dependent variables:

$$C_l\left(\frac{\partial h_g}{\partial t}-\frac{\partial h_l}{\partial t}\right)=K_s\frac{\partial}{\partial z}\left\{kr_l\left(\frac{\partial h_l}{\partial z}-1\right)\right\} \tag{1}$$

$$\left\{\phi(1-S_l)\frac{\rho_{go}}{h_o}-\rho_g C_l\right\}\frac{\partial h_g}{\partial t}+\rho_g C_l\frac{\partial h_l}{\partial t}=K_s\left(\frac{\mu_l}{\mu_g}\right)\ \frac{\partial}{\partial z}\left\{\rho_g kr_g\frac{\partial h_g}{\partial z}\right\} \tag{2}$$

with