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DOE/ER/60663--T/

Final Project Report

**AN INVESTIGATION OF RADON RELEASE AND
MOBILITY IN THE SUBSURFACE ENVIRONMENT**

U.S. D.O.E. Award No: DE-FG03-88ER60663

JUNE 1, 1997

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1.1 Background

Processes affecting transport of volatile species in the shallow soil column have recently been recognized as having a substantial impact on a broad array of real world problems. Investigations of volatile transport have ranged from studies of probable health impacts of radon infiltration into homes (Samset, 1994; National Research Council, 1988; Pershagen et al., 1994) to pesticide and volatile organic contaminant mobility in the soil column (Cook and Solomon, 1995; Falta et al., 1989; and Jury et al., 1983 a,b). The objectives of many of these studies has been the development of numerical models of vapor phase (and solute) transport in shallow soils. An early model, LEACHM, developed by Hutson and Wagenet (1992) was recently modified enabling it to describe both solute and vapor phase transport of volatile chemicals in the soil (Chen, 1995). Subsequent tests of the latter model, named LEACHV, showed that use outside of a very restricted range of soil conditions resulted in large mass balance errors and unreasonable values for soil gas concentrations and vapor flux. The present research was undertaken in an effort to identify and correct the subroutines responsible for the problems and to allow the model to describe vapor phase transport in a much broader range of soil conditions.

Initial testing showed that the routine that solved the air transport partial differential equation was responsible for many of the model's problems. Also uncertain, was the relationship between hydraulic conductivity and air conductivity in the soil. Since LEACHV calculates air conductivity based on soil moisture and saturated hydraulic conductivity, this relationship needed to be defined. Experiments to address this issue included: measuring air conductivity with pumping tests; measuring the perturbations in soil gas differential pressure caused by barometric pressure changes; measuring the hydraulic conductivity of the soil and its response to changes in soil moisture; and conducting air conductivity tests on undisturbed soil cores. At the conclusion of the field experiments, LEACHV simulations were conducted using the field measured parameters to better identify the model's deficiencies. The identified deficiencies were then corrected and an improved air conductivity function incorporated into the model. Finally simulations were run to evaluate how well LEACHV simulations replicated radon activity measurements obtained in the field.

1.2 Overview of research

The results of field studies conducted in conjunction with this research have been consistent with previous research, in that increases in moisture content in the soil decreases the soil gas diffusion coefficient. This in turn causes an increase in soil gas radon concentration caused by the decreased air space in the soil and the decreased flux of radon from the soil. Since soil moisture is a transient condition, radon hazard-potential determinations based on spot measurements may be misleading. Yokel (1989) proposed using time invariant properties such as the soil's radium content, dry density, and dry air permeability to estimate radon hazard potential of the soil, avoiding this problem. However, if a reliable relationship between moisture content and air permeability can be determined, measurement of properties at existing moisture contents can be used in estimating the radon hazard potential of the soil. This research has attempted to refine a relationship between moisture content, air permeability, and hydraulic conductivity. To enable spot *in-situ* measurements to be applied to a full range of soil conditions, the relationships that are determined has been incorporated into an existing soil vapor transport model. This model has been calibrated and verified by comparing the results of the model simulations with field measurements of soil gas radon concentrations and surface radon flux.

In addition to studying radon transport because of its potential for causing cancer, development of a numerical model describing radon mobility can also increase understanding of soil gas phase transport in general. Radon has many advantages over VOC's in the study of gas transport processes. Included among these advantages are:

- 1) Its constant supply. The parent radionuclide, Ra^{226} , has a half life of 1.6×10^3 years and serves as a constant source of Rn^{222} to the soil gas column. In contrast VOC's or pesticides, frequently act as a limited source that constantly changes over the duration of a particular study.
- 2) Rn^{222} is easily detected *in situ* at environmental levels, analysis of VOC's is frequently limited to grab sampling and later laboratory analysis.
- 3) Radon is inert and degrades at a constant an known rate. VOC's are very reactive and degradation is extremely variable being affected by such things as soil chemistry, biota, and moisture.
- 4) Sorption of radon on the solid matrix is not significant. Although organic carbon partition coefficients for radon of up to $22.4 \text{ cm}^3 \text{ g}^{-1}$ have been measured (Wong et al., 1992), radon's relatively large air/water partition coefficient makes sorption effects small (Nazaroff, 1992).

The ratio of radon sorbed on the soil to the total radon in the soil at 20°C is typically less than 0.002 (Schery and Whittlestone, 1989). Sorption characteristics of VOC's is complex and can be a major unknown in vapor transport studies.

Through the use of radon, generalized soil gas transport properties can be determined without the uncertainties associated with sorption and degradation. After the transport properties for a site are determined, the degradation and sorption characteristics for the contaminant in question can be estimated by inverse modeling techniques, or their limits determined by sensitivity analysis. Hence, development of a model for gas transport under varying soil conditions will have a much broader applicability than just assessment of radon hazard potential.

The goals of this research were: 1) demonstrate the use of hydraulic conductivity tests to estimate air conductivity in the soil; 2) incorporate a function based on these tests into a model capable of simulating vapor and liquid phase transport in the soil; 3) investigate the use of radon as a surrogate in VOC transport studies using a numerical model. To find a relationship between the air and hydraulic conductivity of the soil, both were measured over a wide range of soil moisture contents. Concurrent with these tests, was the measurement of radon flux from the soil to the atmosphere. The field experiments were used both to develop a hydraulic conductivity to air conductivity conversion function and to evaluate LEACHV's ability to accurately simulate radon transport in the soil. Early in this project, it was found that LEACHV could not simulate vapor transport if moderate to high values of hydraulic conductivity existed. The cause of its failure needed to be identified prior to proceeding with the simulations. After the initial corrections were made, the process became an iterative one of running simulations based on field data, comparing the output with the experimental results and correcting any discrepancies that were caused by the model coding and then repeating the process until the model's performance was deemed to be satisfactory.

1.3 Summary of Work Conducted

Phase I of our research evaluated the pertinent characteristics of the soil at the experiment site. Physical characteristics of the soil were measured to develop a representative depth profile of the soil column for input into the model. Seventy four samples were collected and analyzed for bulk density and three undisturbed soil cores were analyzed to find average soil particle density. The air filled porosity could then be calculated when soil moisture was measured.

Soil moisture was monitored continuously for six months. This was a key variable since changes in soil moisture affect the air conductivity and the gas diffusion coefficient of the soil and thus determine how efficiently gas phase constituents are transported through the soil. A Time Domain Reflectometer (TDR) array measured soil moisture at eight locations every fifteen minutes over a depth interval from the soil surface down to 2.3 m.

The hydraulic conductivity was measured at saturation using a double ring infiltrometer and a disc permeameter. Hydraulic conductivity data was also desired for soil moisture contents less than saturation and to a depth that represented the effective depth from which radon could be transported to the soil surface prior to undergoing radioactive decay. This was done by wetting the soil to near saturation with oscillating sprinklers, then monitoring drainage with the TDR. Evaporation barriers were put in place so drainage was the only process for water loss in the soil. These tests showed that the soil, which is a silty clay, possessed exceptionally high saturated hydraulic conductivity, but the conductivity dropped rapidly as soil moisture decreased.

Phase II characterized the air conductivity of the soil and the effect of soil moisture changes on air conductivity. Several approaches were taken. Pressure sensing ports were placed in the soil to measure the differential pressure induced across several soil depth segments by changes in atmospheric pressure. Both soil gas differential pressure and atmospheric pressure were monitored continuously for six months - a period that included both very dry and very wet conditions. Pumping tests were done using shallow wells having a screened interval centered at 0.9 m and 1.8 m. These tests were conducted on soil that was dry, then the soil around the well was irrigated and the tests were repeated. Due to uncertainties in the field measurements, large diameter undisturbed soil cores were retrieved and air conductivity tests were done in the lab to compare with the field measurements. An air conductivity test was performed, then the soil moisture content was changed, either by adding water to increase the soil moisture content or by evaporation to decrease the soil moisture content. The water was allowed to become evenly distributed throughout the profile, then the air conductivity test was

repeated. A complete wetting and drying cycle was done on three cores, each taken from different depths. Pumping tests and the tests conducted on the soil cores revealed that, as with hydraulic conductivity, this soil was exceptionally permeable to air. Problems were encountered that made soil gas differential pressure measurements unusable for air conductivity analysis. The pressure gradients were very small, on the order of 0.2 Pa m^{-1} , and were frequently masked by disturbances induced by the wind. In addition, the depth to the lower no-flow boundary was large and a value within acceptable limits could not be determined.

In Phase III the flux of radon from the soil to the atmosphere was evaluated. Two methods were used to find the radon flux. Flux chambers, consisting of plastic cylinders capable of being sealed at the top, were inserted into the soil with a radon detector and a pressure compensating device on each chamber. The rate of increase of radon concentration in the chamber and the steady-state values were used to calculate radon flux to the atmosphere at the time the chambers were closed. Mass balance calculations were used to verify whether or not flux chambers accurately measured the radon surface flux. For these calculations, radon instruments were placed in the soil at 0.8, 1.3, and 2.3 m to measure the radon concentration in soil gas. A theoretical maximum radon content in the soil column was estimated from radium analysis of the soil and the soil's radon emanation efficiency. The flux of radon to the atmosphere was assumed to be the difference between the theoretical maximum amount of radon in the pore space and the actual amount measured.

The radon flux measured by the flux chambers varied greatly between the chambers, sometimes by almost an order of magnitude. Additional tests performed on wet soil indicated a higher flux of radon than that from dry soil. This goes counter to what was expected to happen and is attributed to preferential flow through cracks in the dry soil. Radon flux estimated by the mass balance method was consistently greater than that of the flux chambers and decreased as the soil moisture increased. The latter result was expected since the air conductivity and the soil gas diffusion coefficient decreases as soil moisture increases, decreasing transport efficiency.

Phase IV of the program determined the source of inconsistencies found in the simulations produced by the numerical model LEACHV, took corrective actions to improve the simulation output, and applied the revised model to the field radon data sets to evaluate the key site characteristics that control radon activities in the soil column. Initial simulations revealed that, with complex soil profiles or when high air conductivities were used, LEACHV produced outputs that were unstable and showed large mass balance errors. Mass balance calculations are done by the model to ensure that all of the

solute of interest in the simulation can be accounted for by basic bookkeeping: the mass of the solute at the end of a simulation should be equal to the mass at the start of the simulation plus any added and minus any lost by flux to the atmosphere, lost by degradation and transformation, or lost by drainage through the bottom of simulated soil profile. LEACHV calculates air conductivity utilizing a function that applies saturated hydraulic conductivity and the fraction of void space occupied by air. Since the performance of LEACHV appeared to be adversely influenced by the magnitude of air conductivity, it was suspected the hydraulic to air conductivity conversion function did not accurately calculate air conductivity. Consequently, developing a correct air conductivity function could correct some of the deficiencies experienced with LEACHV. A new function was developed, but problems still plagued LEACHV. The model's code was reviewed; several problems were identified and corrected. The revised model produced good results when simulations were run based on field data.

With a working multiphase transport model, and a set of successful simulations, the final phase of the research addressed the implications for methods of estimating radon flux to the atmosphere, computer simulation of radon transport in the soil, and the progress made by this research in the simulation of multiphase transport in the soil. This work showed that the computer simulations done using LEACHV bring into question the use of flux chambers in measuring soil gas flux to the atmosphere. The close correspondence between simulations and mass balance calculations indicate that flux chambers, the traditional method of measuring gas flux from the soil to the atmosphere, may significantly underestimate this flux. The final product of the research, the LEACHV model, has been field tested and proven that it can be used for simulations involving a broad range of volatile contaminants.