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BUOYANT ADVECTION OF GASES IN UNSATURATED SOIL

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Abstract

In unsaturated soil, methane and volatile organic compounds can significantly alter the density of soil gas and induce buoyant gas flow. A series of laboratory experiments was conducted in a two-dimensional, homogeneous sand pack with gas permeabilities ranging from 110 to 3,000 darcy. Pure methane gas was injected horizontally into the sand and steady-state methane profiles were measured. Experimental results are in close agreement with a numerical model that represents the advective and diffusive components of methane transport. Comparison of simulations with and without gravitational acceleration permits identification of conditions where buoyancy dominates methane transport. Significant buoyant flow requires a Rayleigh number greater than 10 and an injected gas velocity sufficient to overcome dilution by molecular diffusion near the source. These criteria allow the extension of laboratory results to idealized field conditions for methane as well as denser-than-air vapors produced by volatilizing nonaqueous phase liquids trapped in unsaturated soil.

INTRODUCTION

The subsurface environment in many locations has been contaminated by petroleum hydrocarbons and volatile organic solvents. These contaminants are in general, poorly soluble in water and relatively volatile, suggesting that gas-phase transport might be a more significant transport pathway for human exposure than ground water. Certainly there is less current engineering in the control of gaseous contaminants than efforts devoted to monitoring and treating contaminated ground water. Besides contamination issues associated with volatile liquid contaminants, there has been a long-term concern over the subsurface migration of methane gas from municipal landfills. Subsurface methane transport through unsaturated soils can cause explosive hazards in buildings and can carry with it volatile hazardous compounds such as vinyl chloride (Wood and Porter 1987).

Atmospheric air that is sufficiently enriched in methane or a volatile organic compound has a density significantly different from ambient air. Methane-enriched air is lighter than atmospheric air and would tend to rise up through the soil. Volatile organic liquids produce vapors that are denser than air and can cause the spread of these contaminants down to the water table and influence the efficiency of contaminant removal by soil vapor extraction. For these reasons, this research quantified the importance of buoyancy-induced vapor

migration in unsaturated porous media through controlled experiments, comparison with a numerical model, and extrapolation of the results through order of magnitude reasoning.

There are a number of transport processes that can be important in unsaturated soil. Molecular diffusion is of major importance in the gas phase, particularly when compared to aqueous systems, since the molecular diffusivity is 10^4 times larger. However, molecular diffusion is hindered in soils by the tortuous path and contaminant partitioning into soil water and soil organic matter (Jury et al. 1983). Pressure gradients induce gas advection in soils, and these pressure gradients can arise through a number of processes. Soil vapor extraction causes forced advection towards extraction wells (Crow et al. 1987; Massmann 1989; Johnson et al. 1990; Shan et al. 1992). Forced advection in soils also arises from barometric pressure fluctuations at the soil-atmosphere interface (Massmann and Farrier 1992). Finally, advection can be induced by differences in gas density caused by temperature or composition.

Limited experimental data documents the importance of buoyancy-induced gas migration in unsaturated soil. The qualitative work of Schwille (1988) showed that vapors formed by liquid dichloromethane with a vapor pressure of 58 kPa were transported more rapidly downward through a 450-darcy (4.5×10^{-10} -m²) porous medium than upward. Unpublished data by R. L. Johnson cited by Mendoza and Frind (1990) show that a liquid spill of trichloroethylene (TCE) into moist sand with a permeability of 50 darcy produced some evidence of enhanced contaminant transport by density-induced vapor migration.

In contrast to the limited experimental data, there is extensive literature on predictive models for gas migration in unsaturated soils. A number of these models have simulated forced advection of dilute gases in soils, including Mohsen et al. (1980) and Metcalfe and Farquhar (1987), who address methane release from landfills; Loureiro et al. (1990), who model radon transport into the basement of buildings, and a number of models related to the transport of volatile organic solvents in unsaturated soil (Abriola and Pinder 1985; Baehr and Corapcioglu 1987; Gierke et al. 1990). With the recognition that volatile organic liquids become trapped in the subsurface following their release, gas transport models have included the effects of volatilization and buoyancy-induced advection (Sleep and Sykes 1989; Falta et al. 1989; Mendoza and Frind 1990; Mendoza and McAlary 1990). These models differ in their representation of the source term, and in the two-dimensional geometry and boundary conditions. In general, they agree that buoyant advection of gases requires gas permeabilities greater than 10 darcy and contaminant partial pressures greater than 10 kPa at the source. While such modeling efforts have included many of the fundamental transport processes, they have not led to easily applied, quantitative generalizations.

TRANSPORT PROCESSES

Prior to discussing the experimental approach to studying buoyancy-induced gas migration, this section presents a quantitative treatment of isothermal gas transport in porous media. Density differences are caused by gas molecules that are either lighter or heavier than air molecules.

Buoyancy-induced flow is a subset of advective processes within porous media described by Darcy's law. For a horizontal direction x and vertical direction z , positive upward, the velocity components are given by

$$u = - \frac{k}{\mu} \frac{dP_T}{dx} \quad (1)$$

and

$$v = -\frac{k}{\mu} \left(\frac{dP_T}{dz} + \rho g \right) \quad (2)$$

where u and v = Darcy or approach velocities in the x and z -directions, respectively; k = the gas-phase permeability of the medium; μ = gas viscosity; P_T = the total pressure; ρ = fluid density; and g = gravitational acceleration (Bear 1972). When density differences caused by temperature or fluid composition are sufficient to generate vertical flows, then the maximum expected velocity $v_{\max}$ can be estimated from

$$v_{\max} = -\frac{kg}{\mu} (\rho_s - \rho_\infty) \quad (3)$$

where ρ_s = density of the gas at the source; and ρ_∞ = gas density far from the source. This relationship was derived by a number of researchers using related approaches (Bear 1972; Turner 1973; Cheng and Minkowycz 1977; Schery and Petschek 1983; Wilson et al. 1987). Falta et al. (1989) found that (3) was in close agreement with numerically simulated gas velocities within porous media partially saturated by a volatile organic liquid. The maximum vertical velocity may not be realized elsewhere because of contaminant diffusion, constraining boundaries and forced advection.

The source density ρ_s is calculated from the application of the ideal gas law for a contaminant at a partial pressure of P_c having a molecular weight of M_c , and a total pressure of P_T

$$\rho_s = \frac{P_c}{RT} M_c + \frac{P_T - P_c}{RT} M_a \quad (4)$$

where R = gas constant; T = absolute temperature; and M_a = average molecular weight of air, which is 29 g/mol. For a source that is 100% methane, $P_c = P_T$ and the gas density at 20°C is 0.665 kg/m³, which is far lighter than air with a density of 1.206 kg/m³. For a pure liquid solvent, P_c is taken as the vapor pressure, and for TCE with a vapor pressure of 8.0 kPa and a molecular weight of 131.4 g/mol, the source gas density is 1.543 kg/m³. Significant density differences are possible in gases compared to aqueous systems.

Molecular diffusion will dissipate the high contaminant concentrations near sources and diminish the density difference. For dilute contaminants, diffusion within porous media is modeled by Fick's law with a modification by Millington and Quirk (1961) to account for reduced pore space and a longer, more tortuous path. The effective diffusivity D_{eff} for a gas saturated (dry) porous medium is given by

$$D_{\text{eff}} = n^{4/3} D_m \quad (5)$$

where n = porosity; and D_m = molecular diffusivity in air. When the contaminant is not dilute in the gas mixture, Fick's law requires modification to account for bulk gas motion using the Stefan-Maxwell equations (Bird et al., page 570, 1960). Other diffusive processes have some importance in porous media, but for atmospheric pressures and gas-phase permeabilities above 0.1 darcy, inclusion of Knudsen diffusion and the dusty gas model are not necessary (Massmann 1989; Thorstenson and Pollock 1989; Baehr and Bruell 1990).

The importance of buoyant advection is diminished by gas diffusion, and a criterion for the significance of buoyant advection is based on a dimensionless ratio of buoyant to diffusive transport, called the Rayleigh number for mass transport, Ra_m

$$Ra_m = \frac{gkL|\rho_s - \rho_\infty|}{\mu D_{\text{eff}}} \quad (6)$$

where L = vertical height of the source in the system. Absolute values are used for the density difference because both light and dense source gases induce density-driven flows. There is considerably more information on the equivalent heat transfer problem with a Rayleigh number defined as

$$Ra_h = \frac{gkL|\rho_s - \rho_\infty|}{\mu \alpha} \quad (7)$$

where α = thermal diffusivity for the porous medium. An approximate minimum Rayleigh number for buoyancy-driven heat transfer is available for a square cross section of porous media with a temperature difference across the sides and insulating boundaries at the top and bottom. At a Rayleigh number of 10, the heat transfer rate in such a system is about twice the pure conductive rate (Nield and Bejan, page 220, 1992). Thus buoyancy-induced flow should dominate mass transport at Rayleigh numbers greater than 10 by analogy.

There is a real difference between thermally induced flows and composition induced density differences. Typical boundary conditions for heat transfer studies impose either a constant temperature or a constant heat flux at a boundary. The analogous mass-transfer problem also requires the injection of fluid volume that sets up advective flows, and can alter the density difference. For a constant mass-injection rate of methane gas, decreased permeability causes a buildup in the total gas pressure at the source. For sufficiently low permeabilities, the gas density at the source can be comparable to ambient density, and there would be no buoyancy contribution to the flow. Furthermore, heat transfer occurs through the fluid and solid phases in the porous medium while mass transfer occurs only through the gas filled porous region.

METHODS

To quantify the importance of buoyancy-induced flow in gas-saturated porous media, experiments were conducted in the laboratory and compared with a numerical model developed by Falta et al. (1992a,b). To simplify the experimental procedures and allow comparisons of data with model predictions, pure methane gas was injected horizontally into a homogeneous, dry sand pack. Methane could be injected at a known flow rate, while volatile liquid hydrocarbons could not be as easily emplaced as a known source, nor would the local evaporation rate be known. Properties of 100% methane and dry air at one atmosphere pressure are summarized in Table 1.

Fig. 1 diagrams the two-dimensional apparatus used in the experiments. The stainless-steel vessel had a width of 150 cm, a height of 70 cm, and a thickness of 5 cm. The bottom boundary was impermeable and the right-hand boundary was open to the top channel. Sweep air was introduced into the top channel at the right-hand side and vented to a hood from the left-hand side. Along the left-hand boundary, the lower 35 cm was impermeable while a 30-cm-long manifold was placed 5 cm below the top of the sand surface and provided a vertical plane source of methane. The methane injection rate into the sand pack was distributed uniformly over the manifold by flow through a 0.03- μm polycarbonate

membrane filter to create a small pressure drop. The membrane was protected from the sand grains by coarser filters and stainless-steel mesh. The source gas was greater than 99% pure methane and the sweep air was laboratory air that underwent filtration, desiccation, and passage through activated carbon (Seely 1991). Methane flow rates were set by a flowmeter calibrated with a soap bubble flowmeter and corrected to standard temperature and pressure.

The vessel was packed with three different sizes of Monterey silica sand as indicated in Table 2. The sand was placed in the vessel by continuously pouring through a series of screens and vibrating the vessel. Table 2 also includes the calculated porosities and the calculated permeabilities from measured pressure drops across columns packed with the same media at similar porosities. Fig. 1 also indicates the location of gas-sampling ports within the vessel. These ports extended into the middle of the sand and had a stainless-steel screen to keep the porous medium from entering the tube. Each tube had a mini-inert valve for repeated syringe sampling without disruption of flow in the vessel. For sampling, first a 250 μL volume was withdrawn and wasted to clear the sampling point, second, a 100 μL volume was withdrawn by the sampling syringe, wasting 50 μL and injecting 50 μL directly into a gas chromatograph. A flame-ionization detector was used for methane analysis and output was recorded as peak heights on a strip chart recorder. Peak heights were calibrated daily with Scott Specialty Gas methane standards. The data were reproducible within 10%. Small sampling volumes, as outlined previously, removed gas from less than 1 cm^3 of the porous media.

An experiment was initiated by injecting sweep air into the top channel and manifold system for 24–48 h to remove any residual methane left from a previous experiment. Methane gas was flushed through the injection manifold to establish 100% methane in the manifold. Then the methane injection rate was set and the manifold exit was closed so all methane was forced into the vessel. Steady-state conditions within the vessel were established in 24 h, and gases were then sequentially sampled starting at the lowest expected concentration and working toward the methane source. This procedure eliminated syringe cross-contamination problems and the possible effect of removing gas upgradient from unsampled locations that could affect later samples. Identical methane concentration profiles were recorded after 48 h, and effluent methane concentrations in the sweep air revealed that the methane injected was recovered in the effluent to within 10%.

The experimental data are compared with results from an integrated finite-difference numerical simulation model developed by Falta et al. (1992a, b). The model solves the complete mass-balance equations for four components (air, water, organic chemical, heat) existing in three phases (gas, water, nonaqueous-phase liquid). The code has been used to simulate steam injection into porous media for the recovery of nonaqueous phase liquids. In this application, the model was simplified to represent only isothermal gas flow and diffusion. The model internally calculates viscosity and gas density from temperature, pressure, and the mole fraction of methane in the gas phase. Simulations with and without gravitational acceleration can be used to identify the contributions of buoyancy effects on methane transport. For these numerical simulations, the vessel was divided into 190 elements. The vertical element spacing was a uniform 5 cm and horizontally the element spacing varied. The first horizontal element was 3 cm followed by the element spacing of 4.5 cm, 5 cm (four elements) 12.5 cm, 15 cm, 20 cm (two elements), 22.5 cm, and 32.5 cm furthest from the vertical plane source. Methane was injected into one large element by combining six elements in the first column. Boundary conditions for the simulations matched the experimental setup with atmospheric pressure at the top, static pressure on the right side, and no flow on all other boundaries except for the injection location. The methane concentration was assumed to be zero at the top and along the right boundary. Model inputs are specified in Tables 1 and 2 and did not include any adjustable parameters.

RESULTS

Steady-state methane profiles within the vessel were measured for three different medium permeabilities and various methane injection rates, as indicated in Table 3. Concentration data are available for all runs in Seely (1991), and this presentation will emphasize a few selected results to illustrate when buoyancy-induced flow is significant. Later criteria are developed based on the Rayleigh number and a dimensionless ratio of injection velocity to upward buoyant velocity that determine when buoyancy will dominate the mass transfer.

For the lowest permeability material, 110 darcy, the experimental measurements and simulation results are shown in Fig. 2 for an injection velocity of 1.1×10^{-4} m/s. The injection velocity is the flow rate divided by the total cross-sectional area and is equivalent to a Darcy velocity. Fig. 2(a) shows the measured contours of methane concentration in terms of percent by volume. Contours were constructed using a commercial contouring program. The region near the injection manifold exceeds 80% methane and the right-most boundary is at a low enough value to not influence methane transport near the injection region. Simulated steady-state methane contours with gravity are shown in Fig. 2(b), and simulations without gravity are in Fig. 2(c). The two numerical simulations are similar and closely match the experimental data indicating that buoyancy induced transport is not very significant in this case, and that pressure-driven flow and methane diffusion adequately describe methane transport in the model.

On increasing the methane injection velocity to 4.4×10^{-4} m/s the importance of advective transport is seen in Fig. 3. Again the experimental measurements in 3(a) are in close agreement with simulations including gravity [Fig. 3(b)], and the simulation without gravity [Fig. 3(c)] shows a small change in the concentration profile. At this higher injection rate, the methane concentration contours are pushed out further to the right with advection dominated flow out to 60 cm in the lower 40 cm of the vessel. Elsewhere, the influence of the near-zero concentration boundaries are felt along the top and right-hand side as advection and diffusion both contribute to the profiles.

For sand at a permeability of 750 darcy, the influence of buoyancy induced flow is noted, but only at higher methane injection rates. Fig. 4 shows measured and simulated contours of methane concentrations when the injection velocity is 4.4×10^{-4} m/s. The experimental data in Fig. 4(a) and the results of a simulation with gravity in Fig. 4(b) are very close with the simulated contours slightly further out than the measurements. Near the methane injection manifold there is a large region containing in excess of 80% methane. The contours indicate buoyant transport of gases upward toward the top channel, which is at a near-zero methane concentration resulting in steep concentration gradients near the top channel. Upward-buoyant flow entrains surrounding soil gas leading to advective flow towards the methane source. Steady-state profiles of methane result from a balance between entrained flow inward and diffusive flow outward from the methane source. Comparison of Fig. 4(a and b) with simulation results in Fig. 4(c) that did not include gravity indicates that buoyant effects are dominating the actual flow field.

On increasing the permeability to 3,000 darcy the effect of buoyancy-induced flow is dominant at all but the lowest injection rate. In Fig. 5(a and b) experimental contours are compared with simulated concentration contours when the injection velocity was only 1.1×10^{-5} m/s. Experimentally, only a few sample locations recorded methane concentrations greater than 20% and simulated concentrations were a little less. At this low injection rate, methane transport is dominated by diffusion from the plane source towards the top channel. There is no influence of gravity on the simulation due to the low methane concentrations resulting from the low injection rate. At an injection velocity of 4.4×10^{-4} m/s, the contours

of methane measurements in Fig. 6(a) show a region very close to the injection manifold that is high in methane with little methane further out in the vessel. High methane concentrations and high permeability result in rapid upward transport to the top channel and entrainment of soil air that limits outward methane diffusion into the vessel. Computer simulations that include gravitational effects are shown in Fig. 6(b) and closely agree with experimental measurements. In contrast, simulation with no gravitational acceleration in Fig. 6(c) is extremely poor in predicting methane contours. This clearly illustrates the importance of buoyancy flow in this case.

ANALYSIS

The experimental results demonstrate the tradeoffs in permeability and injection rate for a lighter-than-air gas introduced horizontally into a completely unsaturated porous medium. As expected from previous results in studies of heat transfer in porous media, the Rayleigh number is one of the critical parameters in determining if buoyant advection can be important. As summarized in Table 3, the 110-darcy medium with a Rayleigh number of 2.6 had methane concentration profiles that were more or less independent of gravitational acceleration. The 750-darcy medium with a Rayleigh number of 18 had significant buoyant advection only at higher methane injection rates. The highest permeability medium, 3,000 darcy, was at a Rayleigh number of 72, and methane profiles were nearly always dominated by buoyant advection except at the lowest methane injection rate of 1.1×10^{-5} m/s. Thus, having a Rayleigh number, as defined in (6), greater than 10 does not uniquely determine the importance of buoyant advection. Since the Rayleigh number is defined in terms of the density of the injected gas, this density may not be the density a short distance from the plane of injection due to diffusion. This condition is illustrated in Fig. 7, where experiments are characterized by their Rayleigh number and the ratio of methane injection rate to maximum buoyant velocity predicted by (3). The experimental results were classified as either having significant buoyant advection (filled squares), limited buoyant advection (filled triangles), or no buoyant advection (open squares). From this sparse plot, it is possible to generalize that buoyant advection requires a Rayleigh number exceeding about 10 and gas injection rates that exceed at least 1/10 the maximum vertical velocity induced by buoyancy. At lower Rayleigh numbers the methane either diffuses away at low injection rates or advects away by pressure-driven flow at higher injection rates without developing buoyant flow. When the Rayleigh number is larger than 10, low injection rates allow methane to diffuse away from the source, and high methane concentrations cannot be developed to provide the density difference necessary to drive the buoyant flow. With increasing methane injection rates, the methane concentration at the source can approach pure methane, and density effects result.

An order-of-magnitude boundary layer analysis supports the aforementioned reasoning. A buoyancy-dominated boundary layer forms when a light fluid is horizontally injected into a porous medium containing a denser fluid. The boundary layer is a consequence of the upward flow of fluid at the buoyant velocity $v_{\max}$ given in (3) and the advective and diffusive transport of the gas horizontally outward. For fluid injection over a vertical length of L , the time available for horizontal advection and diffusion τ is

$$\tau = \frac{L}{v_{\max}} \quad (8)$$

During this time, the boundary layer is advected outward a distance $U_w \tau$ where U_w is the horizontal injection velocity at the wall. The compound also diffuses outward a distance

$(D_{\text{eff}}\tau)^{1/2}$. The approximate condition for which the horizontal thickness of the boundary layer has equal contribution from advection and diffusion is

$$U_w\tau \approx (D_{\text{eff}}\tau)^{1/2} \quad (9)$$

which can be rearranged using (3), (6), and (8) to give

$$Ra_m \approx \left(\frac{U_w}{v_{\text{max}}} \right)^{-2} \quad (10)$$

This relationship is plotted in Fig. 7, along with the criterion that Rayleigh numbers exceed 10. These conditions are reasonable for delimiting the necessary methane injection rate required for buoyancy-induced flow to dominate. At lower injection velocities, diffusion dissipates the methane and there is insufficient density difference to drive buoyant flow.

An alternative analysis of buoyancy-induced flow can be based on analytical solutions to the flow field near the source. Injection of heated fluid horizontally into porous medium has been modeled by similarity and boundary layer approaches (Sparrow and Cess 1961; Cheng 1977; Merkin 1978). This approach was applied to gas injection into porous media, but the assumptions that were required could not be duplicated in the laboratory apparatus due to its finite size and the high gas phase molecular diffusivity (Seely 1991).

Table 3 includes the maximum vertical buoyant velocity predicted by the numerical model. To obtain the buoyant contribution to the vertical velocity, the component of the vertical velocity associated with volumetric injection was obtained from simulation results without gravitational acceleration. These nonbuoyant velocities were subtracted from the simulated velocities predicted when buoyant advection was included to isolate the vertical velocity due to buoyancy. These maximum buoyant velocities predicted by the simulation model are lower than the maximum velocities predicted by (3) even at high injection rates because the simulation model velocities are averaged over the first 4.5 cm from the source and possibly by the presence of the no-flow bottom boundary. At the highest methane injection rates into the 750- and 3,000-darcy media, the numerical predictions are about half the maximum expected from (3).

Application of these results to idealized field conditions is possible using the dimensionless scaling developed in Fig. 7. This research suggests that buoyancy-driven methane transport requires

$$Ra_m > 10 \quad (11)$$

and from (10)

$$U_w > \frac{v_{\text{max}}}{\sqrt{Ra_m}} = \sqrt{Ra_m} \frac{D_{\text{eff}}}{L} \quad (12)$$

For a source term that is 5 m in vertical height, the Rayleigh number criterion in (11) for methane requires that the permeability exceed 25 darcy. At this permeability, the required methane injection velocity is only 3.8×10^{-6} m/s. If the source-term vertical height increases to 50 m, then the required permeability drops to 2.5 darcy and the methane

injection rate can be an order of magnitude less, 3.8×10^{-7} m/s, and still have buoyancy-dominated flow.

While the experimental system and analysis are based on the injection of lighter-than-air methane, the results offer some insight into the volatilization of nonaqueous phase liquids. Table 4 lists seven common subsurface contaminants and summarizes their chemical properties (Falta et al. 1989). The minimum permeability to achieve a Rayleigh number of 10 was calculated for each compound assuming a contaminated region 5 m in height. The minimum permeabilities range from 330 darcy for low volatility xylene to only 10 darcy for highly volatile methylene chloride. The previous modeling efforts that suggested buoyancy-induced flow would become significant at permeabilities greater than 10 darcy and vapor pressures greater than 10 kPa were for organic-liquid source terms up to 5 m in vertical extent and these conditions correspond to Rayleigh numbers on the order of 4. The simulations also assumed a saturated vapor next to the trapped organic liquid. The criterion in (11) agrees with these modeling results, but also allows extrapolation to less volatile compounds where buoyancy-driven flow can be significant when the formation is more permeable or there is a greater vertical extent of separate phase contamination.

Further refinements in predictive ability for trapped nonaqueous phase liquids must be based on experimentally measured liquid volatilization rates and consideration of subsurface heterogeneities. The criterion in (12) does not have direct application to the volatilization of trapped liquids because the injection velocity in this case is a function of soil gas movement that is determined by the liquid's vapor pressure and the surrounding gas phase permeability. Since liquid contaminants are expected to be the wetting fluid in unsaturated soil, they tend to imbibe into less permeable zones. This has been demonstrated by Ho and Udell (1992) in their laboratory study of soil vapor extraction, and subsurface heterogeneities will likely constrain the importance of buoyancy dominated flow due to mass-transfer limitations. Volatile organic chemicals would also tend to partition into soil organic matter and soil water. However, with a sufficiently large source term of volatile liquid, the soil organic matter and the soil water will equilibrate with the soil gas, and then steady-state vapor migration is independent of partitioning.

CONCLUSIONS

Subsurface contamination by methane gas and volatile organic compounds is common and presents a number of exposure pathways for humans and ecosystems. When methane or other organic compounds are at high partial pressures, the density of the vapor is significantly different from that of air and density-induced vapor migration is expected. A series of experiments involving horizontal methane injection into an unsaturated sand vessel has delimited conditions when buoyancy-induced flow dominates. Two criteria were developed based on achieving a minimum value of a Rayleigh number and a minimum injection velocity. The observed methane concentrations within the experimental apparatus were successfully modeled by a two-dimensional finite-difference numerical model with no calibration of parameters. The dimensionless criteria for the dominance of buoyancy-induced flow can be extrapolated to field conditions and offers insight into the conditions necessary for organic liquids to induce dense vapor migration when they volatilize.

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APPENDIX II. NOTATION

The following symbols are used in this paper:

D_{eff}	effective gas-phase molecular diffusivity in porous media (L^2/T)
D_m	gas-phase molecular diffusivity (L^2/T)
g	gravitational acceleration (L/T^2)
k	gas-phase permeability (L^2)
k_{min}	minimum gas-phase permeability for significant buoyancy-driven flow (L^2)
L	vertical height of source gas (L)
M_a	molecular weight of air (M/mol)
M_c	molecular weight of contaminant (M/mol)
n	porosity
P_c	partial pressure of contaminant ($M/L T^2$)
P_T	total gas pressure ($M/L T^2$)
R	gas constant ($M L^2/T^2 \text{ mol K}$)
Ra_h	Rayleigh number for heat transfer
Ra_m	Rayleigh number for mass transfer
T	absolute temperature (K)
U_w	gas injection velocity at wall (L/T)
u	horizontal velocity (L/T)

v	vertical velocity (L/T)
$v_{\max}$	maximum vertical velocity from (3) (L/T)
x	horizontal distance (L)
z	vertical distance (L)
α	porous-media thermal diffusivity (L^2/T)
μ	gas viscosity ($M/L\ T$)
ρ	gas density (M/L^3)
ρ_s	source gas density (M/L^3)
ρ_α	gas density far from the source (M/L^3)
τ	characteristic time that gas molecules are in boundary layer (T)

FIG. 1.
Vertical Cross Section of Experimental Apparatus

FIG. 2. Methane Concentration Profiles in Percent Methane for $k = 110$ darcy and $U_w = 1.1 \times 10^{-4}$ m/s. Axis Dimensions Are in Centimeters: (a) Experimental Values; (b) Numerical Predictions with Gravity; and (c) Numerical Predictions with-out Gravity

FIG. 3. Methane Concentration Profiles in Percent Methane for $k = 110$ darcy and $U_w = 4.4 \times 10^{-4}$ m/s. Axis Dimensions Are in Centimeters: (a) Experimental Values; (b) Numerical Predictions with Gravity; and (c) Numerical Predictions without Gravity

FIG. 4.

Methane Concentration Profiles in Percent Methane for $k = 750$ darcy and $U_w = 4.4 \times 10^{-4}$ m/s. Axis Dimensions Are in Centimeters: (a) Experimental Values; (b) Numerical Predictions with Gravity; and (c) Numerical Predictions without Gravity

FIG. 5. Methane Concentration Profiles in Percent Methane for $k = 3,000$ darcy and $U_w = 1.1 \times 10^{-5}$ m/s. Axis Dimensions Are in Centimeters: (a) Experimental Values; and (b) Numerical Predictions with Gravity

FIG. 6. Methane Concentration Profiles in Percent Methane for $k = 3,000$ darcy and $U_w = 4.4 \times 10^{-4}$ m/s. Axis Dimensions Are in Centimeters: (a) Experimental Values; (b) Numerical Predictions with Gravity; and (c) Numerical Predictions without Gravity

FIG. 7.

Regions of Significant and Insignificant Buoyant Flow (■: Significant Buoyant Advection; ▲: Limited Buoyant Advection; and □: No Buoyant Advection)

TABLE 1

Properties of Methane Gas and Dry Air at 20°C (Katz et al. 1959)

Property (1)	Methane (2)	Air (3)
Molecular weight (g/mol)	16	29
Density (kg/m ³)	0.665	1.206
Viscosity (kg/m s)	1.09×10^{-5}	1.82×10^{-5}
Molecular diffusivity (m ² /s)	2.18×10^{-5}	—

TABLE 2

Properties of Porous Media

Size interval of sand (mm) (1)	Porosity, n (2)	Permeability, k (darcy) (3)
0.42–0.6	0.388	110
1.18–1.7	0.374	750
1.7–4.75	0.377	3,000

TABLE 3

Summary of Experimental Conditions and Simulation Results

Permeability (darcy) (1)	Ra_m (2)	$v_{\max}^a$ (m/s $\times 10^4$) (3)	U_w (m/s $\times 10^4$) (4)	$v_{\max}^b$ (m/s $\times 10^4$) (5)
110	2.6	0.51	0.11	0.04
110	2.6	0.51	1.1	0.20
110	2.6	0.51	4.4	0.17
750	18	3.5	0.11	0.27
750	18	3.5	0.55	0.95
750	18	3.5	1.1	1.4
750	18	3.5	2.2	—
750	18	3.5	4.4	1.7
3,000	72	14	0.11	1.0
3,000	72	14	0.55	2.9
3,000	72	14	1.1	4.2
3,000	72	14	2.2	—
3,000	72	14	4.4	6.8

^aFrom (3).^bPredicted by numerical model.

TABLE 4

Chemical Properties of Liquid Contaminants, and Predicted Minimum Permeabilities for Buoyancy-Induced Flow

Chemical (1)	M_c (g/mol) (2)	Vapor pressure (kPa) (3)	ρ_s (kg/m ³) (4)	k_{min}^a (darcy) (5)
Xylene	106.2	1.2	1.21	330
Benzene	78.1	12.7	1.42	53
Trichloroethylene (TCE)	131.4	9.9	1.58	32
1,1,1 trichloroethane	133.4	16.5	1.87	19
Carbon tetrachloride	153.8	15.1	1.93	17
Chloroform	119.4	25.6	2.11	14
Methylene chloride (dichloro-methane)	84.9	58.4	2.50	10

^a Assuming $D_{eff} = 4.1 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$; $\rho\alpha = 1.19 \text{ kg m}^{-3}$; and $\mu = 1.8 \times 10^{-5} \text{ Pa s}$.