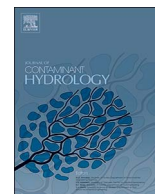




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Density-driven transport of gas phase chemicals in unsaturated soils

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ABSTRACT

Variations of gas phase density are responsible for advective and diffusive transports of organic vapors in unsaturated soils. Laboratory experiments were conducted to explore dense gas transport (sulfur hexafluoride, SF₆) from different source densities through a nitrogen gas-dry soil column. Gas pressures and SF₆ densities at transient state were measured along the soil column for three transport configurations (horizontal, vertically upward and vertically downward transport). These measurements and others reported in the literature were compared with simulation results obtained from two models based on different diffusion approaches: the dusty gas model (DGM) equations and a Fickian-type molar fraction-based diffusion expression. The results show that the DGM and Fickian-based models predicted similar dense gas density profiles which matched the measured data well for horizontal transport of dense gas at low to high source densities, despite the pressure variations predicted in the soil column were opposite to the measurements. The pressure evolutions predicted by both models were in trend similar to the measured ones for vertical transport of dense gas. However, differences between the dense gas densities predicted by the DGM and Fickian-based models were discernible for vertically upward transport of dense gas even at low source densities, as the DGM-based predictions matched the measured data better than the Fickian results did. For vertically downward transport, the dense gas densities predicted by both models were not greatly different from our experimental measurements, but substantially greater than the observations obtained from the literature, especially at high source densities. Further research will be necessary for exploring factors affecting downward transport of dense gas in soil columns. Use of the measured data to compute flux components of SF₆ showed that the magnitudes of diffusive flux component based on the Fickian-type diffusion expressions in terms of molar concentration, molar fraction and mass density fraction gradient were almost the same. However, they were greater than the result computed with the mass fraction gradient for > 24% and the DGM-based result for more than one time. As a consequence, the DGM-based total flux of SF₆ was in magnitude greatly less than the Fickian result not only for horizontal transport (diffusion-dominating) but also for vertical transport (advection and diffusion) of dense gas. Particularly, the Fickian-based total flux was more than two times in magnitude as much as the DGM result for vertically upward transport of dense gas.

1. Introduction

Soil and groundwater contamination by volatile organic compounds (VOCs) is a great environmental risk due to high toxicity. In particular, VOC vapors from nonaqueous phase liquid (NAPL) residing in soils or dissolved forms in soil moisture and groundwater migrate and spread more rapidly in unsaturated soils than dissolved forms of VOCs in saturated zones, since gas diffusion coefficients of organic vapors are several order of magnitude larger than aqueous diffusion coefficients (Bedient et al., 1999). As the popularity of monitored natural attenuation as a subsurface remediation strategy increases, a complete understanding and reliable prediction of gas phase transport of VOCs under natural conditions are necessary (Rivett et al., 2011). Vapor transport in porous systems can be driven by concentration differences

and total pressure variations, denoted as diffusive (dispersive) and advective transport, respectively. These transport mechanisms are usually described with Fick's law of diffusion and Darcy's law, respectively. Additionally, the gravitational force may enhance or suppress the vertically advective transport of organic vapors (Sleep and Sykes, 1989; Mendoza and Frind, 1990; Lenhard et al., 1995; Jang and Aral, 2007). In the past, dusty gas model (DGM) equations were also discussed and applied to relevant problems for the deficit of Fick's law of diffusion in representing multicomponent gas transport in unsaturated soils, see for example, Webb (1996), Sleep (1998), Fen and Abriola (2004) and Molins and Mayer (2007).

Organic vapors mixed with soil air form a gas mixture with a total density different than the air density. Based on Fick's law of diffusion, gas diffusion is driven by gas density variations. Fen and Abriola (2004)

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showed that mathematical models developed in the past for gas phase transport in unsaturated soils employed different forms of Fick's law of diffusion. The diffusive (dispersive) flux component based on Fick's law of diffusion, hereafter called Fickian-type diffusion, is in a form either in terms of a molar (or mass) fraction or molar (or mass) concentration gradient. For multicomponent gas transport in porous systems (soils), nonequimolar effects due to different molecular masses between the air gases and organic vapors are addressed in the DGM equations. The Fickian-type diffusion may not be sufficient to describe diffusive transport of a gas mixture with very different molecular masses in porous systems. Altevogt et al. (2003) presented an alternative way to address the nonequimolar effects. They additionally include a slip velocity, adopted from Jackson (1977), in a standard, mass-based advective-diffusive transport equation and solved the equation for chemical gas density. The slip velocity caused by slip flow (or called diffusive slip in Kramers and Kistemaker, 1943) accounts for nonequimolar counter diffusion of gases of different molecular masses. Jackson (1977) showed that the flux component based on the slip velocity is included in the 2-component DGM equations under limited conditions including: (1) gas diffusion in the molecular regime, i.e., the mean free path of gas molecules in the order of mean pore size or larger; (2) pressure diffusion, i.e., diffusion due to total density variations in space, is neglected. Further, an additional diffusive flux component driven by gravitational force was first presented in Thorstenson and Pollock (1989) in which the DGM theory was also adopted. The significance of this component has not yet been delineated in modeling or experimental work.

Advection is mainly driven by total density (gas pressure) variation in space and gravitational force if gas transport is not horizontal. Advection driven by gas density is sometimes presumed to be negligibly small in some modeling and/or experimental work in cases of no external pressure gradient, for example, Corapcioglu and Baehr (1987), Jury et al. (1990), Lenhard et al. (1995), Hamamoto et al. (2008) and Cotel et al. (2011). Thus, the modeling work may be only based on the gas phase transport equation for the solution of component molar (or mass) concentration. However, gas pressure variations may be induced during gas phase transport due to nonequimolar weights of gas molecules (Lenhard et al., 1995; Altevogt et al., 2003). Such modeling work needs to solve the gas transport equation coupled with flow equation for molar (or mass) concentration and total pressure (or total density or equivalent pressure head) as those presented in, for example, Metcalfe and Farquhar (1987), Rathfelder et al. (2000), Sleep and Sykes (1989), Mendoza and Frind (1990) and Jang and Aral (2007). For the solution of the equivalent pressure head, it is presumed the dependence of total density of gas mixture on total gas pressure is negligibly small. Mendoza and Frind (1990) pointed out the equivalent pressure head formulation may increase numerical accuracy for scaling the pressure potential to the same magnitude as the elevation potential. Fen and Abriola (2004) found the mass-based formulations of these modeling approaches may predict different gas pressure and chemical concentration variations for gas phase chemical transport in one-dimensional (1-D) porous systems. Thus, it is necessary to investigate the gas pressure and chemical concentration variations in the laboratory to assess the validity of various modeling approaches on predicting gas phase chemical migration in unsaturated soils.

Dense gas transport in porous systems has been investigated in the experimental work of many researchers, including Lenhard et al. (1995), Conant et al. (1996), Altevogt et al. (2003), Jellali et al. (2003), Hamamoto et al. (2008) and Cotel et al. (2011). Lenhard et al. (1995) and Jellali et al. (2003) measured organic vapor concentration distributions in sand tanks and Conant et al. (1996) in field. Lenhard et al. (1995) obtained small gas pressure variations in space in their sand tank experiment and did not consider advection in the analysis. Altevogt et al. (2003), Hamamoto et al. (2008) and Cotel et al. (2011) carried out transport experiments in soil columns and observed the spatial and temporal variations of chemical vapor/gas concentration

along the columns. Altevogt et al. (2003) and Cotel et al. (2011) also observed small gas pressure variations ($>$ or $<$ 1 Pa) in soil columns when chemical gas migrates from a VOC source. Lenhard et al. (1995), Altevogt et al. (2003) and Cotel et al. (2011) showed gravitational force plays an important role in vertical transport. The modeling approaches for the interpretation of the experimental results in their work were all based on Fickian-type diffusion on a mass or molar basis. Altevogt et al. (2003) also considered the slip component.

The DGM equations represent a framework for describing multicomponent gas transport processes in porous media and have not fully been verified experimentally in soil systems. In addition to measuring gas phase chemical concentration and pressure variations for assessing the magnitudes of diffusive and advective flux components along a soil column, the related transport parameters, such as effective molecular, Knudsen diffusion coefficient and soil permeability, should be determined independently. Relevant experiments have been conducted in Altevogt et al. (2003) and Hamamoto et al. (2008) for soil air permeability and effective gas diffusion coefficient and in Cotel et al. (2011) for soil permeability only. Cotel et al. (2011) used the relationship of Millington and Quirk (1961) to estimate the effective gas diffusion coefficient and considered dispersion. These studies estimated the effective gas diffusion coefficient with a Fickian-type diffusion expression rather than the DGM equations.

The objective of this study is to assess the DGM and Fickian-based modeling approaches on predicting dense gas transport. A series of transport experiments were conducted to investigate the migration of chemical gas-nitrogen mixture along a soil column placed either horizontally or vertically. The results of other similar gas phase transport experiments presented in the literature were also compared with the model predictions. We also used the measured chemical gas concentrations, gas pressure and transport parameters to compute various flux components of dense gas to assess different diffusion approaches on determining the migration rate of gas phase chemical from a source zone.

2. Materials and methods

2.1. Mathematical formulations of gas phase transport model

For simulating gas phase transport in soil columns, 1-D, component molar (or mass) balance equation (transport equation) is generally utilized. Assuming a rigid porous matrix, single gas flow and no reactions or external sources/sinks, the transport equation of a gas component i on different bases may be written as (Cunningham and Williams, 1980):

$$\theta_a \frac{\partial C_i}{\partial t} + \frac{\partial N_i^T}{\partial z} = 0 \quad (1a)$$

$$\theta_a \frac{\partial \rho_i}{\partial t} + \frac{\partial G_i^T}{\partial z} = 0 \quad (1b)$$

where θ_a is gas-filled porosity; C_i and ρ_i are the molar and mass concentration of species i , respectively; t is time coordinate; z is vertical coordinate which is directed downward; N_i^T and G_i^T represent the total molar and mass flux of species i , respectively. N_i^T is the sum of total diffusive, N_i^D , and viscous fluxes, N_i^V , of species i on a molar basis; and G_i^T is the sum of G_i^D and G_i^V on a mass basis.

Different theoretical bases for the total diffusive flux component were presented in past modeling work. The first is DGM theory. Here, we present the flux component based on the two-component DGM equations as

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