LNAPL Leakage and Resulting Contaminant Transport in the Unsaturated and Saturated Zones.

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LNAPL leakage and resulting contaminant transport in the unsaturated and saturated zones

تسرّب السوائل الخفيفة الغير قابلة للذوبان في الماء وحركة الملوثّات الناتج عنها في المناطق المشبّعة والغير مشبّعة

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ملخص البحث:

تم إعداد نموذج عددي للتدفّق المركّب وانتقال الملوثّات لدراسة تسرّب السوائل الخفيفة الغير ذائبة في الماء خــلال المنطقة غير المشبّعة من سطح الأرض والملوثّات المتعلقة بها في كل من المناطق المشبّعة والغير مشبّعة. تم اســتخدام النمـــوذج لتوصيف ودراسة حركة السوائل الخفيفة وانتشارها والتلوّث الناتج خلال المناطق الغير مشبّعة والمشبّعة. تم اـ تخدام النموذج لحساب حساسية النتائج للمتغيرات المختلفة للاستفادة منها في تصميم التجارب والقياسات الحقلية. وجد مـــن الدراســـة أن كثافة السائل الغير ذائب في الماء ومتغيرات الضغط الشعري نزيد من انتشار السائل في اتجاه على حساب الإتجاه الأخر، بيـا تؤثر باقي المتغيرات المتعلقة بالتربة والتدفق وانتقال الملوثّات على انتشار السائل في الإتجاهين نفس الكيفية. وجد أن معامل فان جنوغتن المرتبط بعلاقة درجة التشبع مع الضغط الشعري له أكبر تأثير على النتائج.

Abstract

A multiphase flow and transport numerical model was developed to study the leakage of light non-aqueous phase liquids (LNAPL) into the vadose zone and the associated contamination of both the unsaturated and saturated zones. The model was used to provide a better tool for addressing the movement and spreading the NAPL phase and the resulting contamination in both the vadose zone and the saturated zone. In order to guide experimental design and field investigations, the model was used to evaluate the sensitivity of the results to various flow and transport parameters. It was found that the NAPL density and the capillary pressure parameters (van Genuchten parameters) increased the NAPL spreading in one direction at the expense of the spreading in the other direction. The remaining soil, flow, and transport parameters affected the vertical and lateral spreading in a similar manner. Based on the results presented herein, it seems that the exponent m of the van Genuchten capillary pressure-saturation relation is the most influential parameter.

Keywords:

Introduction

Leakage of organic industrial liquids and petroleum derivatives into the subsurface is considered a major environmental problem due to the risk of long-lasting and wide-spreading contamination of both air and water. Dissolved and volatilized contaminants can be transferred from these liquids to subsurface water and air, and thus these liquids are considered as a continuous source of contaminant. Great awareness of the multiphase flow of light non-aqueous phase liquids (LNAPL), water, and air in porous media has led to considerable works during the past few decades (e.g.,

has led to considerable works during the past few decades (e.g., Forsyth and Sammon 1986; Kaluarachchi and Parker 1989; Huyakorn et al. 1994a, b; Corapcioglu et al. 1996; Wu and Karsten 1998; Liao and Aral 2000; Kim and Corapcioglu 2001; Mendicino et al. 2006; Suk and Yeh 2007; just to name a few). Spatial variability of porous media properties has been regarded as a dominant factor in characterizing flow and contaminant transport in the subsurface. This variability was also found crucial for assessing saturations and pressures of multiphase liquids in porous media. Thus it is expected that such variability would significantly impact LNAPL movement and spreading and the amount of mass lost from the NAPL body to the air phase as volatile contaminant or the water phase as dissolvable contaminant.

In this research, a two-dimensional vertical model was developed for studying multiphase flow and contaminant transport resulting from LNAPL leaking into the subsurface. We model subsurface contamination of water and air due to vertically and laterally migrating LNAPL to evaluate the amount of mass of LNAPL lost to groundwater as a dissolved contaminant and to subsurface air as a volatilized contaminant. We also studied the effects of dissolution and volatilization on LNAPL movement and spreading. In addition, the effects of soil and fluid parameters on the movement and spreading of the NAPL body and the contaminant plumes are studied. The mass transfer processes between phases and how different parameters impact these processes were evaluated. We also addressed the impact of joint variability in the porous medium properties (e.g., conductivity, porosity, van Genuchten parameters) with and without correlation on LNAPL infiltration and movement in the vadose zone and the resulting air and water contaminant plumes.

Model Formulation and Governing Equations

The leakage of LNAPL through the vadose zone requires the treatment of three phases and the description of their mass balance as well as coupled movement (i.e., three mass balance equations and the associated constitutive relations). When the resulting air and water contamination was considered, the mass of NAPL lost as a contaminant transferred to water and air was accounted for in the mass balance equations. The multiphase flow equations describe the mass balance for each of the three phases coupled with Darcy's law describing the flux of a particular constituent as a function of the pressure gradient. For both the air and

water phases, the flow equation can be written in the form:

$$\frac{\partial}{\partial t}\left[\phi \, \rho_\alpha \, S_\alpha\right] = \sigma_\alpha +$$

$$\nabla\left[\frac{k \, \rho_\alpha \, k_{r\alpha}}{\mu_\alpha}(\nabla P_\alpha + \rho_\alpha \, g \, \nabla Z)\right]$$

(1)

in which: the subscript α represents either water or air phase ("w" or "a"), ϕ is the porosity of the porous medium [dimensionless], S_α is the fluid volumetric saturation of phase α [dimensionless], σ represents a source or a sink of fluid [$ML^{-3}T^{-1}$], ρ is the fluid density [ML^{-3}], k is the intrinsic permeability [L^2], $k_{r\alpha}$ is the relative permeability for phase α [dimensionless], μ is the dynamic viscosity [$ML^{-1}T^{-1}$], P is the fluid pressure [$ML^{-1}T^{-2}$], g is the gravitational acceleration [LT^{-2}], and Z is the elevation [L].

A similar equation can be written for the NAPL, but the mass transfer terms should be added to the flux and source/sink terms. Thus for the NAPL phase (denoted by the subscript n), the flow equation is written as

$$\frac{\partial}{\partial t}\left[\phi\rho_n S_n\right] = \sigma_n + \nabla\left[\frac{k\rho_n k_{rn}}{\mu_n}(\nabla P_n + \rho_n g \nabla Z)\right]$$

$$- \phi S_w \lambda_d (C_{wm} - C_n^w) - \phi S_a \lambda_v (C_{am} - C_n^a)$$

(2)

in which: λ_d is the dissolution rate constant [T^{-1}], λ_v is the mass transfer rate for volatilization of contaminant in air [T^{-1}], C_n^w is the concentration of contaminant dissolved in water [ML^{-3}], C_n^a is the concentration of contaminant

volatilized in the air phase [ML^{-3}], and C_{wm} and C_{am} are the equilibrium concentrations of contaminant in water and air, respectively [ML^{-3}]. These equilibrium concentrations set a maximum limit to the ability of water and air to absorb dissolvable and volatile contaminants.

The flow equations of the three phases are linked by the capillary pressures. These capillary pressures are described by van Genuchten relations, which express the functional relations between saturations of the different phases and the capillary pressures (Van Genuchten, 1980). The dependence of the relative permeability of a certain phase on that phase saturation is taken similar to the formulations of Parker et al. (1987).

The transport equations for the dissolved and volatilized contaminants are written in a similar manner to the equations of Sleep and Sykes (1989). The mass balance equation for the dissolved contaminant is written as

$$\frac{\partial}{\partial t}\left[\phi S_w C_n^w + k_d \rho_b C_n^w\right] = \frac{C_n^w}{\rho_w}\sigma_w'$$

$$+ \nabla\left[\frac{kC_n^w k_{rw}}{\mu_w}(\nabla P_w + \rho_w g \nabla Z)+\right.$$

$$\left. D_n^w \nabla\left(\phi S_w C_n^w\right)\right] + \phi S_w \lambda_d (C_{wm} - C_n^w)$$

$$- \phi S_a \lambda_b (HC_n^w - C_n^w)$$

(3)

in which: k_d is the partition coefficient describing the mass

partitioning between the liquid phase (water) and the solid phase (soil particles) $[M^{-1}L^3]$, ρ_b is the bulk density of the aquifer material $[ML^{-3}]$, H is the dimensionless Henry's law constant, λ_h is mass transfer rate coefficient between water and air $[T^{-1}]$, and D_n^w is the hydrodynamic dispersion coefficient for contaminant dissolved in the water phase $[L^2T^{-1}]$.

For the volatilized contaminant, the mass balance equation is written as

$$\frac{\partial}{\partial t}\left[\phi S_a C_n^a\right] = \frac{C_n^a}{\rho_a}\sigma_a' +$$

$$\nabla\left[\frac{kC_n^a k_{ra}}{\mu_a}(\nabla P_a + \rho_a g \nabla Z)+\right.$$

$$\left. D_n^a \nabla\left(\phi S_a C_n^a\right)\right] + \phi S_a \lambda_c ((C_{an} - C_n^a)$$

$$+ \phi S_a \lambda_h (HC_n^w - C_n^a)$$

(4)

in which: D_n^a is the hydrodynamic dispersion coefficient for volatile contaminant in the air phase $[L^2T^{-1}]$ and all the other symbols are as defined earlier. Similar to the mass balance equation for the contaminant dissolved in the water phase, the equation of the volatile contaminant has two mass transfer processes: the mass gained from the NAPL body and mass transfer with the water phase. When the latter acts as a source for air contamination, the contaminant has to first dissolve in the water phase and subsequently vaporize and move to the air phase. The volatile contaminant in the air phase can also be lost within the domain to the water phase and can leave the model domain across the boundaries.

The mathematical model is composed of the five coupled partial differential equations (Eqs. 1 through 4) and a set of auxiliary equations relating capillary pressures to saturations and relative permeabilities to saturations. These equations cannot be solved analytically except for very simplified cases and under many restrictive assumptions. Here we employ a numerical solution using block-centered finite differences with a forward time central space scheme (FTCS) discretization and an implicit pressure explicit saturation (IMPES) solution procedure (Peaceman, 1977). The discretized equations and the IMPES scheme result in four equations being converted into linear systems of equations and solved using an algebraic multi-grid (AMG) solver (Stuben, 2001), and the remaining equation is solved explicitly.

Simulation Domain and Initial and Boundary Conditions

A two-dimensional vertical domain of dimensions 100 m × 15 m is selected for implementing the solution of the multiphase flow equations and contaminant transport equations (Fig. 1). Homogeneous conditions are assumed in the subsequent
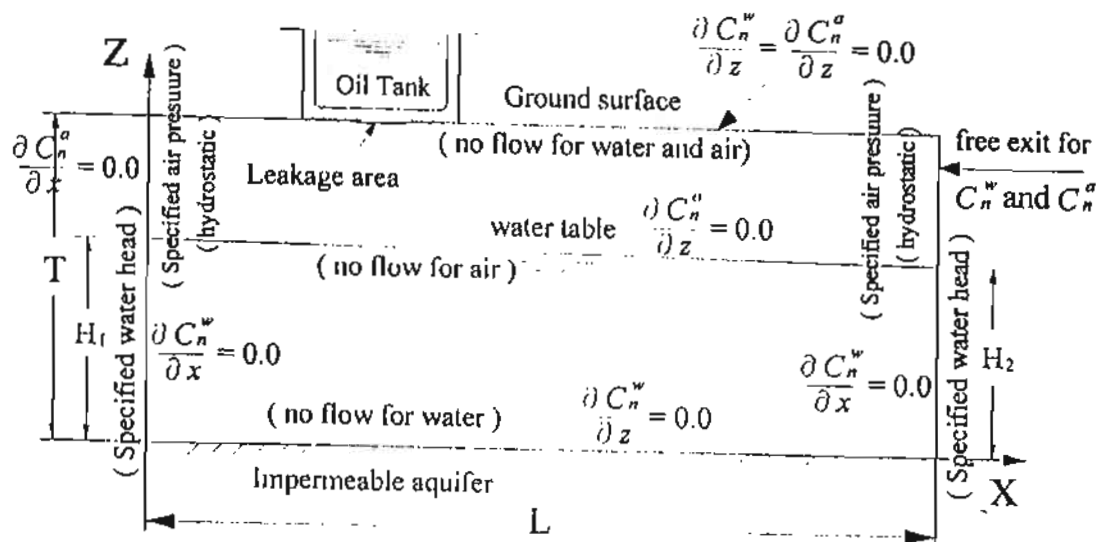
Fig. 1 Domain outline and boundary conditions for (a) the multiphase flow problem, and (b) the contaminant transport problem.

grid of square blocks of length Δx = Δz. The size of each grid cell is 0.25 m × 0.25 m with a total number of 24,000 cells (60 rows × 400 columns).

Initial and boundary conditions are identified for both the vadose zone portion and the saturated zone portion of the simulation domain. The multiphase flow problem requires specification of initial values of saturations and pressures for all phases. To get these initial values, the multiphase flow equations for water and air only are solved in a steady state condition with the assumption that NAPL does not exist in the domain initially before leakage. This gives an

initial distribution for air and water saturations and the corresponding pressures. For contaminant transport equations, it is assumed that subsurface water and air are free of contamination before leakage and thus the values of dissolved and volatilized contaminant concentrations are set equal to zero. The boundary conditions for the multiphase flow problem and the associated transport problem are shown in Fig. 1.

Results and Discussion

Sensitivity analyses were performed to investigate the impact of different parameters on multiphase flow and the influence

and dispersion in the simulation domain. The effect of these parameters on the amount of mass transferred from NAPL to water and air was also investigated. Fig. 2 displays the spatial moments for the NAPL plume after 120 days of leakage with a constant leakage rate of 500.0 kg/year with different parameter values governing the flow.

From the results shown in Fig. 2 it can be seen that overall the NAPL spreading in the vertical direction (expressed here as the second moment X_{22}) is most impacted by the multiphase flow parameters. On the contrary, the horizontal spreading, X_{11}, is the least impacted by the variability in the flow parameters. Also it can be noted that the variability in the dynamic viscosity of NAPL, μ_n, caused the largest change in the NAPL travel distance, L_{cg}, and horizontal spreading, X_{11}, whereas α_m caused the largest changes in X_{22}. It is also interesting to note that The NAPL density, ρ_n, and van Genuchten parameters, α_{na} and α_{nn} impact the vertical and horizontal NAPL spreading in an opposite manner (i.e., increasing the spreading in one direction and decreasing it in the other). All other flow parameters shown in Fig. 2 affect the spreading in both directions in a similar manner. Also, all parameters have a monotonic effect of the travel distance and second spatial moments except the m parameters as explained above.

The amount of dissolved and volatilized contaminant mass is affected by many parameters as shown in Fig. 3. The figure displays the variation of the total dissolved mass and the total volatilized mass at 120 days with the variation of the multiphase flow parameters. It is seen that increasing α_{n-a} increases vertical dispersion and decreases horizontal spreading and as a result mass of dissolved contaminants increases and mass of volatilized contaminants decreases. On the other hand, increasing α_{w-a} decreases vertical dispersion and increases horizontal dispersion and as a result mass of dissolved contaminants decreases and mass of volatilized contaminants increases. It is clear from Fig. 3 that half of the flow parameters impact the dissolved mass and the volatilized mass in the same manner (either decreasing both or increasing both), whereas the other half causes opposing trends for the two masses. The porous medium porosity impacts the masses in opposing manners at small values, but then when the porosity is increasing at the upper end of its possible range, enhanced dissolution and volatilization result. The impact of the flow parameters is more pronounced for the dissolved

mass than for the volatilized mass.

Conclusions

This research work presented two-dimensional simulations of oil infiltration (from ground storage tanks for example) into aquifers and the resulting air and water contamination. The results illustrated that the oil plume travelled faster and vertical spreading was increased with the decrease of soil porosity, NAPL viscosity, entrapped NAPL saturation, and van Genuchten parameter, α_{wa}, and with the increase of NAPL density, residual water saturation, and van Genuchten parameter α_{na}.

Spreading of the NAPL plume in the horizontal direction was increased with decreasing porosity, NAPL density, viscosity of NAPL, residual NAPL saturation, and van Genuchten parameter, α_{na}, and with increasing residual water saturation, and van Genuchten parameter, α_{wa}. The amount of mass transferred from the NAPL body to the liquid or the air phase was greatly affected by the flow parameters. The most influential parameter in this regard was the van Genuchten parameter, m.

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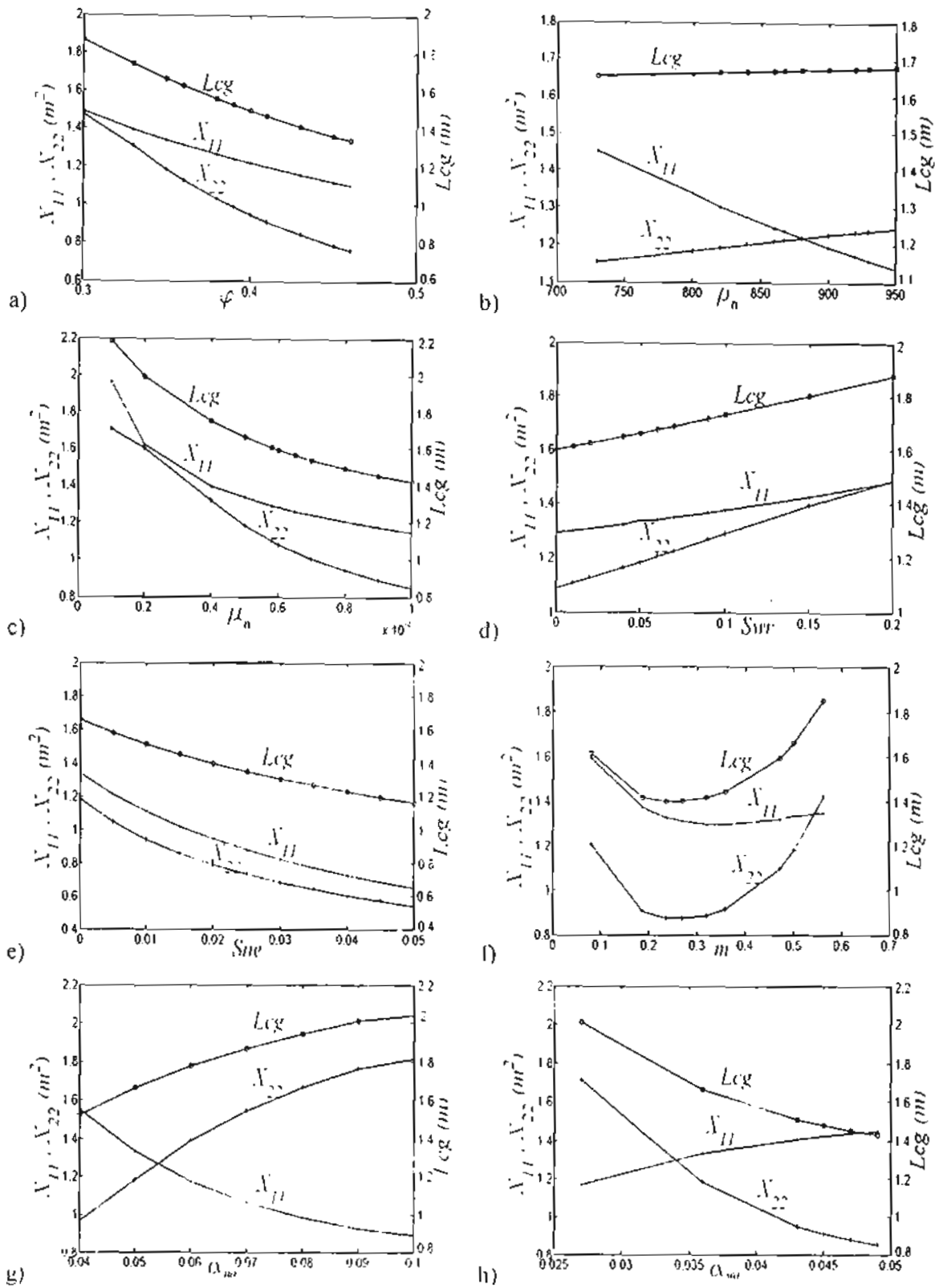
Fig. 2 Variation of NAPL plume travel distance, L_{cg}, and spatial moments, X_{11} and X_{22}, with multiphase flow parameters.
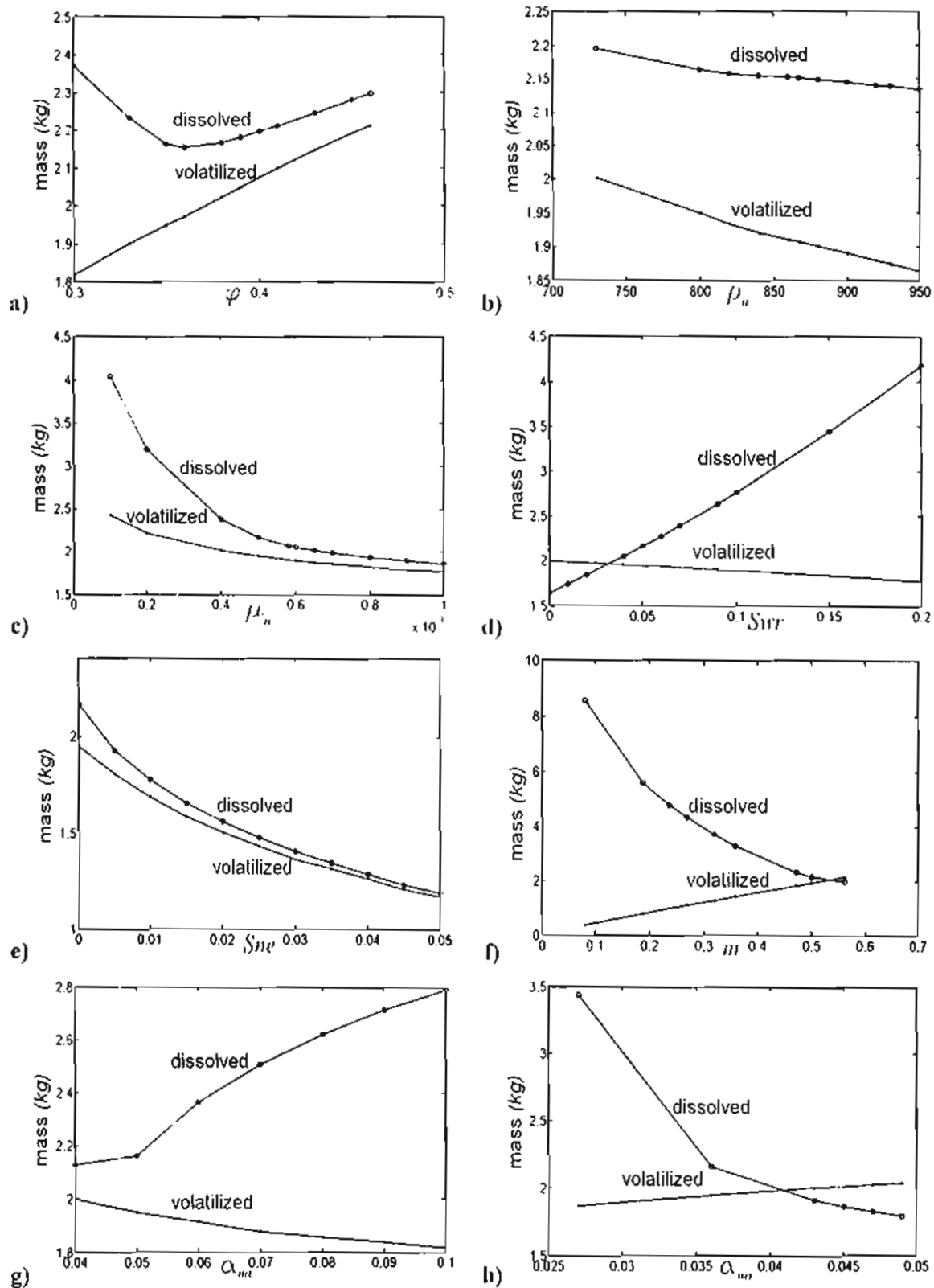
Fig. 3 Effect of the multiphase flow parameters on the total mass of dissolved and volatilized contaminants.