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Numerical Calculation of Mass Transfer from Elliptical Pools in Uniform Flow Using the Boundary Element Method

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Abstract. The three-dimensional problem of advection-dispersion associated with an elliptical non-aqueous-phase liquid (NAPL) pool is addressed using the boundary element method. The boundary condition on the plane of the pool is such that over the pool the concentration is equal to the saturation concentration while a no flux boundary condition is imposed in the region not covered by the pool. The numerical results are verified by asymptotic analytical solutions obtained in the limits of diffusion-dominated and convection-dominated mass transport. For cases of practical interest an empirical expression is obtained for the Sherwood number that matches the numerical results over a wide range of the relevant parameters. Comparison with experimental results suggests that the corresponding numerical results predict a higher overall mass transfer coefficient.

Key words: NAPL pools, contaminant transport, boundary element method.

1. Introduction

Underground water constitutes an important proportion of the world water reserves. Its over-exploitation and the increase in subsurface contamination renders this resource very vulnerable. Leaking underground storage tanks, ruptured pipelines, surface spills, hazardous waste landfills and disposal sites can release contaminants to soil and ground water. When released to the soil contaminants may (i) dissolve in the ground water (aqueous-phase transport), (ii) spread through the pore spaces as vapor (vapor-phase transport), (iii) sorb to colloidal particles and be transported with these particles (facilitated transport), or (iv) transported as a separate non-aqueous-phase liquid (known as a NAPL) that is immiscible in water and therefore travel separately from water (non-aqueous-phase liquid transport).

Most of the NAPLs are organic and chlorinated solvents, and petroleum hydrocarbons. When a NAPL spill, which is more dense than water, infiltrates the

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subsurface environment, it will continue to migrate downwards leaving behind trapped ganglia until it encounters an impermeable layer. There, it will form a flat source zone or pool with relatively limited spatial extent (Hunt *et al.*, 1988). On the other hand, NAPL pools with densities lower than that of water will, as they approach the saturated region, spread laterally and float on the water table in the form of a pool. Due to their low solubility in water, NAPL pools may lead to long-lasting sources of groundwater contamination (Bradford *et al.*, 1998).

The dissolution of a NAPL pool in a porous medium is modeled as an advection–dispersion equation where the convection velocity is assumed to be uniform and equal to the interstitial velocity of the water (Bear, 1972). The problem is addressed in its fundamental form by considering all physical processes in their macroscopic level. Hence, all the significant mechanisms of mass transfer are captured, that is, convection–diffusion–reaction. Expressions for the dispersion coefficients were obtained by Bear and Verruijt (1987) where it was shown that, to first order, they depend linearly on the advection velocity. It should be noted however, that in the case of small particles in porous media the dependence becomes parabolic for asymptotically small Péclet numbers scaled by the particle size. At the NAPL–water interface, the solubility concentration is often adopted as an appropriate concentration boundary condition. This implies that thermodynamic equilibrium is instantaneously approached at the interface and the process is dominated by the advective–dispersive transport of the dissolved NAPL away from the interface. This assumption was justified by residual NAPL dissolution experiments by Borden and Kao (1992) and Borden and Piwoni (1992).

There is a relatively large body of available literature on the mathematical development of NAPL pool dissolution (Prakash, 1984; Abriola and Pinder, 1985; Leij *et al.*, 1993; Toride *et al.*, 1993; Seagren *et al.*, 1994; Chrysikopoulos *et al.*, 2002). However, there is limited literature on the development of theoretical and experimental overall mass transfer relationships. Furthermore, for simplicity, mathematical models that address the mass transport problem assume a homogeneous (Neumann, Dirichlet or a linear combination) boundary condition on the plane of the pool, by employing an average and time invariant mass transfer coefficient that represents the entire pool (Hunt *et al.*, 1988; Chrysikopoulos *et al.*, 1994; Holman and Javandel, 1996; Chrysikopoulos *et al.*, 2003).

The two-dimensional problem of NAPL transport which takes into consideration both the presence of the pool and of the impermeable layer has been addressed using the boundary element method in Fyrillas (2000). Similar techniques were first used by Stone (1989) to address the problem of heat/mass transfer from an elliptical film to a fluid undergoing a simple shear flow and by Bender and Stone (1993) to consider the steady-state current at surface micro-electrodes. In agreement to the classic boundary solution of Lévêque (1928) and the asymptotic analysis by Phillips (1990), Stone (1989) has found that at high Péclet (Pe) numbers the total transport is of $\mathcal{O}(Pe^{1/3})$; in the case of uniform flow, which is the topic of this work, the total transport is of $\mathcal{O}(Pe^{1/2})$ for high values of Péclet number.

The boundary element method offers the natural choice for this problem as it combines numerical simplicity and accuracy. Regarding the former, the reduction of the differential equation to an integral equation over the boundary reduces the dimensionality, hence the complexity, of the problem. Furthermore, the integral representation allows the consideration of an infinite-domain, direct calculation of the concentration gradient, and de-singularization of the singular points which translate into a high degree of accuracy. In general however, the numerical solution of integral equations of the first kind is susceptible to oscillations due to the ill-conditioning of the influence matrix (Pozrikidis, 1992, 1997). However, the integral equations considered in the above references exhibit regular behavior and the major difficulty is related to the presence of a singularity in the concentration gradient along the edge of the pool. For the two-dimensional case Belward (1969) has obtained an analytical expression for the concentration gradient by expanding the kernel in terms of the modified Mathieu and Mathieu equations. He has shown analytically the persistence of the square-root singularity at the leading and trailing edges of the pool for all values of the Péclet number; he has also obtained the form of the singularity for more general two-dimensional axisymmetric cases (Belward, 1972, 1974).

In this paper the work in Fyrillas (2000) is extended to three-dimensions considering the mass transport from elliptical pools in a fluid of uniform velocity. In the next section we formulate the problem and present a Fredholm integral equation of the first kind for the concentration gradient over the pool location. In Section 3 analytical asymptotic expressions are obtained in the limit of small and large Péclet numbers, which correspond to diffusion-dominated and convection-dominated mass transport, respectively. A collocation boundary-element method using constant functions over each element is used to solve the integral equation numerically and the results are presented in Section 4. The numerical results are verified with the asymptotic analytical solutions. Finally, in Section 5, we compare the results predicted by this analysis with available experimentally determined mass transfer coefficients.

2. Formulation

Consider an elliptical pool that is formed on top of an impermeable layer within a three-dimensional, semi-infinite medium (Figure 1). This is related to the problem of NAPL transport in a saturated, homogeneous and isotropic porous medium. It is also related to the conductive–convective transport from a surface film on a planar insulated boundary to a fluid in uniform flow.

It is assumed that the flow and the transport process are steady as we are concerned with the long-time behavior of the process. In particular, the present results offer a lower-bound on the total mass transfer. The steady-state transport of the

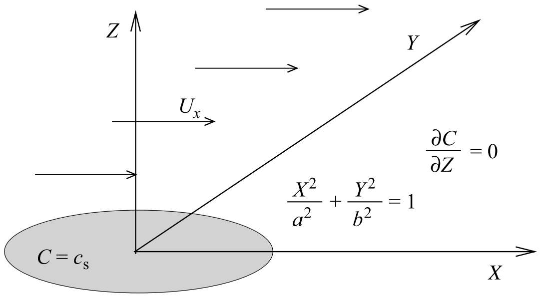

Figure 1. Schematic representation of the conceptual physical model along with boundary conditions. Uniform flow with unidirectional velocity U_x is flowing over an elliptical pool. The concentration over the pool is the saturation concentration c_s, while a no flux boundary condition is imposed in the area not covered by the pool.

dissolving contaminant into the ambience under uniform flow conditions is governed by

$$\begin{aligned} U_x \frac{\partial C(X,Y,Z)}{\partial X} &= D_x \frac{\partial^2 C(X,Y,Z)}{\partial X^2} + D_y \frac{\partial^2 C(X,Y,Z)}{\partial Y^2} + \\ &\quad + D_z \frac{\partial^2 C(X,Y,Z)}{\partial Z^2} - \lambda C(X,Y,Z), \end{aligned}$$

where λ is the first-order decay coefficient, D_x, D_y and D_z are the dispersion coefficients, and U_x is the average fluid velocity. The appropriate (inhomogeneous) boundary condition on the planar boundary is:

$$\frac{\partial C}{\partial Z}\left(\frac{X^2}{a^2} + \frac{Y^2}{b^2} > 1, Z = 0\right) = 0, \qquad C\left(\frac{X^2}{a^2} + \frac{Y^2}{b^2} \leqslant 1, Z = 0\right) = c_s,$$

while the far field conditions are

$$C(\pm\infty, \infty, Z) = C(X, Y, \infty) = 0.$$

The problem is non-dimensionalized by scaling the X and Z coordinates with the semi-axis a, the Y coordinate with the semi-axis b and the concentration by the saturation concentration c_s to obtain the dimensionless parameters:

$$Pe_x = \frac{U_x a}{D_x}, \qquad Pe_y = \frac{U_x b^2}{a D_y}, \qquad Pe_z = \frac{U_x a}{D_z} \quad \text{and} \quad \Lambda = \frac{\lambda a}{U_x}.$$

The problem takes the form:

$$\begin{aligned} \frac{\partial c(x,y,z)}{\partial x} &= \frac{1}{Pe_x}\frac{\partial^2 c(x,y,z)}{\partial x^2} + \frac{1}{Pe_y}\frac{\partial^2 c(x,y,z)}{\partial y^2} + \frac{1}{Pe_z}\frac{\partial^2 c(x,y,z)}{\partial z^2} - \\ &\quad - \Lambda c(x,y,z) \end{aligned} \tag{1}$$

with boundary conditions

$$\frac{\partial c}{\partial z}(x^2+y^2>1, z=0)=0, \qquad c(x^2+y^2\leqslant 1, z=0)=1,$$

and

$$c(\pm\infty, \infty, z)=c(x, y, \infty)=0.$$

The boundary element formulation can be easily derived as outlined by Stone (1989), Bender and Stone (1993) and Fyrillas (2000). When evaluated on the plane of the pool ($z=0$) we obtain a Fredholm integral equation of the first kind for the concentration gradient over the pool location, $x^2+y^2\leqslant 1$,

$$\begin{aligned} 1 \;=\; & -\frac{\sqrt{Pe_x\,Pe_y}}{2\pi\sqrt{Pe_z}}\int_A \frac{\partial c(x', y', z'=0)}{\partial z'}\exp\left[\frac{Pe_x(x-x')}{2}\right]\times \\ & \times\frac{\exp\left[-\sqrt{(Pe_x/4+\Lambda)}\sqrt{(x-x')^2\,Pe_x+(y-y')^2\,Pe_y}\right]}{\sqrt{(x-x')^2\,Pe_x+(y-y')^2\,Pe_y}}\,\mathrm{d}S(x', y'), \end{aligned} \tag{2}$$

where A defines integration over the pool (unit disk).

3. Analysis

In this section we obtain asymptotic solutions in the cases of diffusion-dominated and convection-dominated mass transport. For simplicity it is assumed that there is no decay, that is, $\Lambda=0$.

3.1. DIFFUSION-DOMINATED MASS TRANSPORT

In the limit of small Pe_x, and Pe_y ($Pe_x\approx Pe_y\ll 1$), the convection term can be neglected and the problem is equivalent to mass transport from an elliptical disk. The mass flux is given by (Landau and Lifshitz, 1960; Stone, 1989)

$$\frac{\partial c}{\partial z}=-\frac{\sqrt{Pe_z}}{(Pe_x\,Pe_y)^{1/4}}\frac{1}{\sqrt{\beta}K(1-\beta^2)}\frac{1}{\sqrt{1-r^2}}, \tag{3}$$

where $\beta=\sqrt{Pe_y/Pe_x}$ and K is the complete elliptic integral of the first kind. Here and in what follows r and θ are the polar coordinates in the x–y plane.

The boundary element formulation (2) suggests that the applicability of the above expression is less restrictive and is also valid for any value of Pe_y, provided that $Pe_x\ll 1/Pe_y$. Physically it implies that the convection effect has to be small enough such that to suppress any coupling between convection in the x-direction and diffusion in the y-direction.

3.2. CONVECTION-DOMINATED MASS TRANSPORT

In the limit of large Pe_y the axes of the ellipse in the Y-direction becomes infinite and the pool resembles an infinite strip. Hence, we assume a 'quasi-steady' approximation in the y-direction (Stone, 1989); that is the profile of the gradient $(\partial c/\partial z)$ on a constant y line is approximated by the classical boundary layer result $-\sqrt{Pe_z/(\pi x)}$ to obtain the corresponding result for the three-dimensional case

$$\frac{\partial c}{\partial z}(z=0) = -\sqrt{\frac{Pe_z}{\pi}}\frac{1}{\sqrt{r\cos(\theta)+\sqrt{1-r^2\sin(\theta)^2}}}. \tag{4}$$

4. Numerical Results

A numerical evaluation of the concentration gradient by applying the boundary element method to the integral (2) is performed. The numerical approach employed is the collocation boundary element method (Pozrikidis, 1992, 1997), where the local basis functions are step functions. Boundary element offers the natural choice for this problem taking into consideration the linearity of the problem. It allows the reduction of the dimensionality of the problem, that is, although the problem is associated with an infinite 3D domain, the problem is solved over the unit disc; furthermore, the high degree of accuracy permits the numerical solution of asymptotic cases with respect to the Péclet numbers, that is, we were able to consider both small and large Pe_x and Pe_y numbers.

The analysis of this section closely follows previous analyzes on similar problems (Stone, 1989; Fyrillas, 2000). The pool is subdivided in circular segments and the integral equation (2) is 'de-singularized' by defining the function f:

$$f(x,y) = -\frac{1}{\sqrt{1-r^2}}\frac{\partial c(x,y,z=0)}{\partial z}.$$

The coefficients of the influence matrix are computed by Gauss quadratures (Press *et al.*, 1989), Gauss–Legendre for the non-singular elements and Gauss–Chebyshev for the singular elements.

Experimentation has shown that a correct evaluation of the diagonal terms of the influence matrix is very important for stability and accuracy. It involves integration of a singular kernel and in addition, when the element is next to the boundary of the pool, it involves a square-root singularity along the boundary. The former is related to the Green's function of the problem while the latter is an artifact of the de-singularization of the boundary element equation. The presence of the two singularities renders an analytical integration too tedious. Instead, we numerically integrate over the unit disk using Gauss–Chebyshev quadrature. This takes into account the square-root singularity along the boundary. The origin of the polar coordinate system is taken to be at the origin of the singular element which removes the singularity of the kernel. The diagonal term can be evaluated by subtracting the sum of the integrals of the rest of the elements.

4.1. AVERAGE (OVERALL) MASS TRANSFER COEFFICIENTS

The average mass transfer coefficient is defined as (Chrysikopoulos *et al.*, 1994; Incropera and DeWitt, 1990):

$$\begin{aligned}\bar{h}_\mathrm{m} &= -\frac{D_\mathrm{e}}{A_\mathrm{s}c_\mathrm{s}}\int_A \frac{\partial C(X,Y,Z=0)}{\partial Z}\,\mathrm{d}X\,\mathrm{d}Y \\ &= -\frac{D_\mathrm{e}b}{A_\mathrm{s}}\int_0^1\int_0^{2\pi}\frac{\partial c(x,y,z=0)}{\partial z}r\,\mathrm{d}r\,\mathrm{d}\theta, \end{aligned} \tag{5}$$

where A_s is the area of the pool and A defines integration over the pool; $D_\mathrm{e} = D/\tau^*$ is the effective molecular diffusion coefficient; D is the molecular diffusion coefficient and τ^* is the tortuosity.

We also define the dimensionless Sherwood number (Incropera and DeWitt, 1990)

$$\overline{Sh} = \frac{A_\mathrm{s}/b}{D_\mathrm{e}}\sqrt{\frac{Pe_x}{Pe_z}}\bar{h}_\mathrm{m},$$

which for the asymptotic expressions (3) and (4) is calculated to be

$$\overline{Sh} = \frac{2\pi}{\beta K(1-\beta^2)} \quad \text{for diffusion-dominated,} \tag{6}$$

and

$$\overline{Sh} \approx 4.9442\sqrt{\frac{Pe_x}{\pi}} \quad \text{for convection-dominated.} \tag{7}$$

As a benchmark, the code was tested for the case that there is no convection, that is, the Laplace equation, and the results were compared to the analytical results (3) and (6). As shown in Figure 2 there is an excellent agreement between the numerical and the analytical results.

In Figures (3)–(5) we compare the Sherwood number obtained from the numerical results with that calculated from the asymptotic results (6 and 7). For small Pe_x (Figure 3), the numerical results suggest that (6) adequately predicts the overall mass transfer coefficient as justified in Section 3 (diffusion-dominated mass transport).

For moderate and large values of Pe_x (Figures 4 and 5), comparison between (6) and the numerical results suggest that the agreement between the two is restricted for small values of Pe_y while the asymptotic result (7) is readily attained. In the same figures, we also plot an approximate expression which was calculated by curve-fitting a large number of numerical data. The expression lies within 5% of

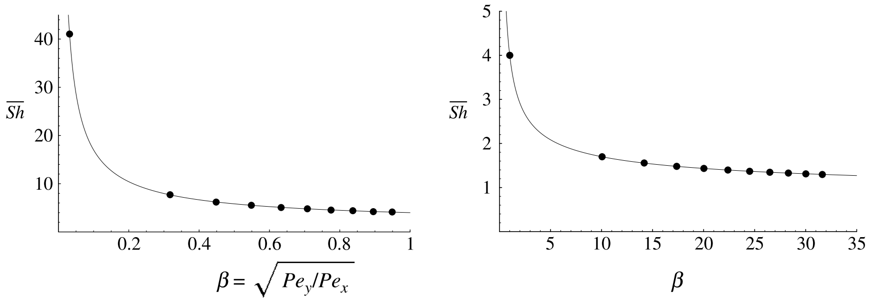

Figure 2. Comparison between the analytical result (6) (solid curves) and numerical results (solid points) for the case of no convection (Laplace equation).

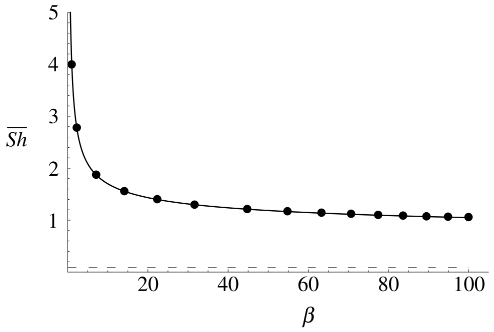

Figure 3. Comparison between the asymptotic result (6) (solid curve) and the numerical results (solid points) for $Pe_x = 0.001$. The dashed line corresponds to the asymptotic result (7).

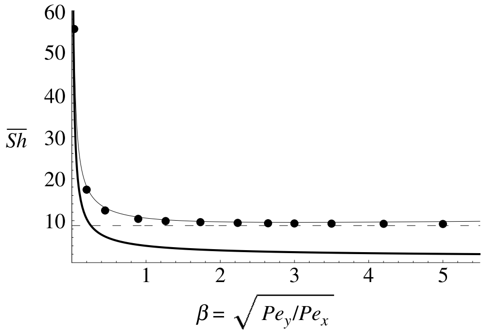

Figure 4. Comparison between the asymptotic result (6) (thick-solid curve) and the numerical results (solid points) for $Pe_x = 10$. The dashed line corresponds to the asymptotic result (7) while the fine-solid curve to the empirical equation (8).

the numerical data for Péclet numbers less than 100:

$$\overline{Sh} = \frac{2\pi}{\beta K(1-\beta^2)}\left(1 + \frac{0.3038\, Pe_x^{0.8094}}{\exp(0.0323\log^2(Pe_x))}\sqrt{\beta}\right). \tag{8}$$

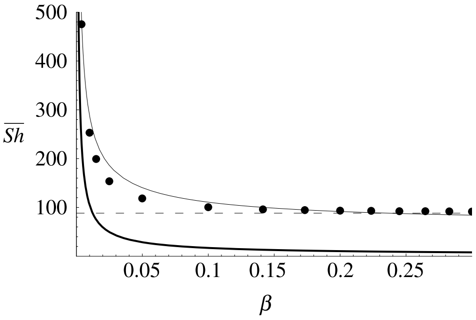

Figure 5. Comparison between the asymptotic result (6) (thick-solid curve) and the numerical results (solid points) for $Pe_x = 1000$. The dashed line corresponds to the asymptotic result (7) while the fine-solid curve to the empirical equation (8).

5. Comparison with Experimental Results

Experimental results for mass transfer coefficients are quite limited. In Figure 6, results for $\bar{h}_m$ predicted by this work are compared with those obtained experimentally by Lee (1999) and Lee and Chrysikopoulos (2002) in a circular ($a = b = 3.8\,\text{cm}$) trichloroethylene (TCE) pool dissolution experiment in a homogeneous, fully saturated bench scale aquifer under various interstitial velocities. The dispersion coefficients are evaluated using (Bear and Verruijt, 1987)

$$D_x = \alpha_L U_x + D_e \quad \text{and} \quad D_z = D_y = \alpha_V U_x + D_e, \tag{9}$$

where α_L and α_V are the longitudinal and vertical dispersivities respectively. Lee (1999) and Lee and Chrysikopoulos (2002) has obtained $D_e = 2.11 \times 10^{-2}\,\text{cm}^2/\text{h}$, $\alpha_L = 0.259\,\text{cm}$, and $\alpha_V = 0.019\,\text{cm}$.

In Figure 6, we show a comparison between the theoretical and the experimental results for the mass transfer coefficient. The numerically obtained overall mass transfer coefficient $\bar{h}_m$ (solid line in Figure 6) is higher than the one obtained in the

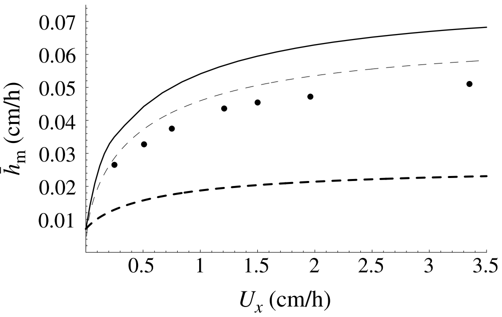

Figure 6. Comparison between numerical results (solid curve) and experimental data (solid points) in a circular TCE pool dissolution experiment in a homogeneous, fully saturated bench scale aquifer under various interstitial velocities. The fine-dashed curve corresponds to the two-dimensional results obtained in Fyrillas (2000). The thick-dashed curve corresponds to the three-dimensional results without convection (Eq. (6)).

experiments (solid points). A possible explanation might be that the experiments were performed in a rectangular glass tank with finite dimensions while the numerical domain is assumed to be infinite. Another factor for the disagreement is that in the presence of small particles, micro-scale and adsorption-desorption effects play a significant role on the dependence of the dispersion coefficients on the advection velocity; in such cases the dependence becomes asymptotically parabolic for small Péclet numbers.

6. Conclusions

We have presented a boundary element formulation and a numerical solution of the problem of advection–dispersion mass transport associated with an elliptical NAPL pool in uniform flow. The problem is modeled by defining the concentration over the pool, which is assumed to be equal to the solubility concentration, and a no-flux boundary condition in the area not covered by the pool. We derive a Fredholm integral equation of the first kind for the concentration gradient which is de-singularized and solved numerically using a collocation boundary-element method. The agreement between the numerical results and the asymptotic results is quite satisfactory. Comparison with experimental results suggests that the corresponding numerical results predict a higher overall mass transfer coefficient. Possible explanations are that the experimental domain is finite while the numerical domain is infinite and also the assumed linear relationship between the advection velocity and the dispersion coefficients. The latter is not valid in the case of small particles in porous media because in such cases the dependence becomes asymptotically parabolic for small Péclet numbers.

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