The First ANZ Young Geotechnical Professionals Conference, Mostyn, Khalili & Small (eds), Feb. 1994, Sydney, Australia

Analysis of Contaminant Transport in Porous Media

C. J. Leo
Centre for Geotechnical Research
University of Sydney

1 Introduction

All industrial manufacture and human consumption of goods and resources produce waste. By 'waste' it is meant not only the 'useless' by-product of organism or manufacture, but also any effluent, chemical, compound or substance, which if discharged into the environment may give rise to contamination problems. One of the most important problems facing society today is how we can disposed of our waste in ways which do not endanger human health and the environment.

It was not until the 1970's that environmental legislations designed to protect the environment such as the licensing of waste disposal were introduced in Australia. Since then, the state governments in Australia have continually sought to enhance environmental protection by legislation. This is exemplified by the Environmentally Hazardous Chemicals Act 1985 and the Environmental Offences and Penalties Act 1989 enacted in New South Wales and the Environment Protection (Amendment) Act, 1989 in Victoria.

In line with current expectations on environment protection, there is a move now in Australia to introduce a regulatory framework that will impose stricter control over the disposal of all forms of waste (Parker and Sadlier, 1992). If this comes into being, there will be greater emphasis on the North American and European practice of waste containment rather than conventional methods of waste disposal such as the 'dilute and disperse' of waste leachate into the groundwater system. As a result, if the natural underlying soils do not already provide sufficient barrier at the chosen disposal sites there will be increasing use of engineered systems. The long term performance of these waste facilities, often for periods of many decades, will have to be carefully assessed in relation to the effectiveness of the design of barriers. It is desirable to do this by the use of mathematical models as experimental modelling is usually both costly and limited in its ability to predict over long span of time.

This paper presents a background of contaminant transport processes of a single species in a single phase system and show some results of a boundary element method developed for assessing potential long term contaminating effects of waste storage, disposal and leakage.

2 Background of Contaminant Transport

The analysis of contaminant transport in the porous media in many cases involves the application of a suitable macroscopic mathematical model describing the movement of contaminants in the presence of hydrological, physical, chemical and sometimes biological processes. These processes include:

- groundwater advection
- hydrodynamic dispersion (incoporating molecular diffusion and mechanical dispersion
- sorption
- radioactive decay and biological degradation

Before attempting an analysis of contaminant transport, it is important to understand the controlling mechanisms of these processes. Thus, a summary is provided below.

2.1 Groundwater Advection

Groundwater advection is a subsurface hydrological process and is a major transport mechanism of contaminants in the porous media. Prior to modelling of contaminant transport, the groundwater flow conditions must be known beforehand. With computers and software packages readily available nowadays, the information on groundwater flow can be obtained by perfoming computer simulations, supplemented by data from field monitoring. In contaminant transport modelling, the time period in consideration usually span over a duration of several years or decades. For this reason, short term groundwater transients are not important in modelling the movement of contaminants, hence, it is not uncommon to adopt a steady-state groundwater model.

The fundamental equations of groundwater flow models may be formulated by considering a control volume and applying the law of mass conservation. For steady, incompressible, saturated groundwater flow, this equation is given by,

$$\nabla . \boldsymbol{Va} = 0 \tag{1}$$

where,

$\nabla = [\partial / \partial x, \partial / \partial y, \partial / \partial z]^T$

$\boldsymbol{Va} = [Va_x, Va_y, Va_z]^T$, the vector of specific discharge also commonly known as the flow rate or seepage velocities.

If, as commonly assumed, the seepage velocities are linearly related to the hydraulic gradients (Darcy-type flow), then:

$$\boldsymbol{Va} = -\boldsymbol{K} \nabla H \tag{2}$$

where,

H is the piezometric head

$\boldsymbol{K}$ is the hydraulic conductivity or permeability matrix

and where $\boldsymbol{V\ a}$ may now also be described as the vector of Darcy velocities. When equation (2) is combined with (1), the governing equation of groundwater flow is,

$$\nabla . (\boldsymbol{K} \nabla H) = 0 \tag{3}$$

This equation can be solved for given initial and boundary conditions to obtain the piezometric head H, the hydraulic gradients and the Darcy velocities. Assuming that the Darcy velocities are known, the advective mass flux, $\boldsymbol{f}_e$ of the contaminant species, is given by the expression,

$$\boldsymbol{f}_e = n \boldsymbol{V} c \tag{4}$$

where,

n is the porosity of the soil

$\boldsymbol{f}_e = [f_{e_x}, f_{e_y}, f_{e_z}]^T$ is the vector of advective fluxes

$\boldsymbol{V} = [V_x, V_y, V_z]^T$ is the vector of the average or true groundwater velocities given by $\boldsymbol{Va}/n$

c is the concentration of the contaminant defined as the mass of contaminant per unit volume of fluid

2.2 Hydrodynamic dispersion

The second transport process which needs to be considered is hydrodynamic dispersion. This is the the spreading phenomenon occuring in contaminant transport due to the combined effects of molecular diffusion and mechanical dispersion.

Spreading due to molecular diffusion is predominantly the result of Brownian motion, but it may also be driven by other forces such as osmotic forces, thermal diffusion and electro-osmosis (Luckner and Schestakow, 1991). In Brownian motion, a movement of the contaminant is induced in the direction of decreasing concentration in a way which is often described by Fick's first law. Thus, the diffusive mass flux ($\boldsymbol{f}_d$) and concentration gradients (∇c), are related by,

$$\boldsymbol{f}_d = -D_o \nabla c \tag{5}$$

where the negative sign indicates that the contaminant is moving in the direction from higher to lower concentration and D_o is the coefficient of molecular diffusion due to Brownian motion. However, the effects of other diffusive forces are also commonly assumed to be governed by analogous relationships as equation (5). Thus, for practical purposes, D_o is usually defined to represent the combined effects of all of the molecular forces.

Mechanical dispersion on the other hand is generally caused by the heterogeneity of the permeability and storage capacity in the porous media. This cause the contaminant particles to scatter longitudinally and transversely about the average paths resulting in some particles to travel ahead and some behind the average migration velocity. The coefficients of mechanical dispersion may be considered to be components of a matrix $\boldsymbol{D}_m$. These are related to the components of the true groundwater velocities by the dispersivity parameters. In an isotropic medium, the components of the dispersivity tensor are related to two parameters: α_L the dispersivity in the direction of groundwater flow, known as the longitudinal dispersivity and α_T, the dispersivity in a direction perpendicular to groundwater flow, known as the transversal dispersivity. In such soil, the coefficients of mechanical dispersion can then be written as (Bear and Buchlin, 1991),

$$D_{m_{kl}} = \alpha_T V \delta_{kl} + (\alpha_L - \alpha_T)\frac{V_k V_l}{V} \tag{6}$$

where,

k, l range over the index set (x, y, z)

δ_{kl} is Kronecker's delta

V is now the magnitude of the flow,

i.e. $V = \sqrt{V_x^2 + V_y^2 + V_z^2}$

As in the foregoing, the mass flux transported by hydrodynamic dispersion f_h, which incoporates both molecular diffusion and mechanical dispersion, is often assumed to be governed by Fick's first law, viz.

$$f_h = -n\boldsymbol{D}\nabla c \tag{7}$$

and the components of $\boldsymbol{D}$ are,

$$D_{kl} = D_o \delta_{kl} + D_{m_{kl}}$$

2.3 Advective-dispersive transport

The combined processes of advective and dispersive transport discussed in the foregoing subsections are often assumed to be additive and governed by a Fickian law (Gillham and Cherry, 1982), thus

$$\boldsymbol{f} = \boldsymbol{f}_e + \boldsymbol{f}_h \tag{8}$$

and so,

$$\boldsymbol{f} = n\boldsymbol{V}c - n\boldsymbol{D}\nabla c \tag{9}$$

The relative importance of the different transport mechanisms has been investigated, albeit mostly in one dimensional flow studies. Some of these have been summarised by Pfannkuch (1963) who demonstrated that the predominance of either mechanical dispersion or molecular diffusion in contaminant transport is a function of the Peclet number of the molecular diffusion, $Pe = Vd/D_o$ where d is the mean grain size. These data and those of other studies help to support the notion that the effect of molecular diffusion assumes a dominant role in hydrodynamic dispersion when the flow velocity is small (e.g. in clay barriers) but proportionately less so at a higher velocity. It thus follows that at very high velocities (e.g. in aquifers), the effect of molecular diffusion is negligible and hydrodyamic dispersion is likely to be overwhelmingly by mechanical dispersion.

2.4 Sorption

Besides being transported by advective-dispersive processes, the species may be removed from or added to the fluid by a number of sorptive interactions and reactions. When these occur, the species is regarded as being reactive, otherwise, it is described as being conservative.

The sorption phenomena may be broadly categorised into either adsorptive or absorptive processes. The former is generally a surface phenomenon by which the species are removed from the fluid and attached to the surfaces of the solid matrix which serve as hosts. On the other hand, if the material which is transfered from one phase to another interpenetrates the solid matrix, the process is known as absorption.

The plot of the sorbed contaminant in the solid phase against the concentration in the solution phase at constant temperature is known as an isotherm. If this relationship is known, it can be used to model the distribution of the contaminant between the solid and aqueous phases. The simpliest and most straightforward isotherm is the linear equilibrium isotherm which assumes that mass of species sorbed from solution per unit mass of solid, A, is instantaneous and reversible and is given by,

$$A = K_d c \tag{10}$$

where K_d is the partioning or distribution coefficient. This isotherm is applicable to sorption processes involving low contaminant concentrations (Travis, 1978; Luckner and Schestakow, 1991). The parameter K_d used in a linear equilibrium isotherm may be determined from the same laboratory column tests as those used for molecular diffusion D_o (see Rowe, Caers and Barone, 1988).

2.5 Other sources and sinks

In addition to sorption, contaminant sources or sinks may also result from the occurrence of other processes including injection or withdrawal of fluid containing contaminants from the porous media, radioactive decay and biological degradation.

It is convenient to model biological degradation and radioactive decay as a first order kinetic rule, thus

$$g_b = -\gamma_b c \tag{11}$$

$$g_r = -\gamma_r c \tag{12}$$

where,

the subscripts b, r denote radioactive decay and biodegradation respectively

γ is the decay constant of an exponential decay or degradation with time

If the half life of the decay or degradation is T, γ can be determined from the expression,

$$\gamma = \frac{ln2}{T} \tag{13}$$

3 Governing Equations of Contaminant Transport

The movement of a single species of contaminant in a homogeneous saturated zone of the porous media is governed by the equation of conservation of mass (Bear, 1979; Freeze and Cherry, 1979),

$$\nabla.f + n\frac{\partial c}{\partial t} + g = 0 \tag{14}$$

The quantity g can be thought of as the sum of three components, g_a (due to sorption), g_r (due to radioactive decay) and g_b (due to biodegradation) i.e.,

$$g = g_a + g_r + g_b \tag{15}$$

The quantities g_r and g_b are given by equations (11) and (12) respectively and the rate of sorption is found by using the time derivative of equation (10), thus

$$g_a = \rho K_d \frac{\partial c}{\partial t} \tag{16}$$

where,

ρ is the dry density of the soil

It then follows from combining equations (10), (11), (12), (14) and (16) that under conditions of a steady spatially uniform advective flow field occurring in an isotropic, homogeneous saturated porous medium, the equation of contaminant transport is,

$$n(D\nabla).\nabla c - nV.\nabla c = (n + \rho K_d)\frac{\partial c}{\partial t} + n\gamma c \tag{17}$$

where

$$\gamma = \gamma_{j_r} + \gamma_{j_b}$$

4 The Boundary Element Method

A number of methods have been used to solve the governing equation (17), depending on the prescribed initial distribution of c in the domain and the conditions on the boundaries of the domain. In simple cases, mostly under one-dimensional situations, several analytic and semi-analytic solutions exist. These solutions can be very rapidly implemented on a hand-held calculator or a desktop computer to produce quick preliminary estimates of potential contamination problems.

In more complicated problems of practical interest, however, it is usually necessary to resort to alternative, more powerful numerical methods, e.g. finite difference and finite element methods. One other numerical method, the boundary element method, is lesser known than the former two, nevertheless it is a technique which is rapidly gaining popularity. It is also a technique which the author has developed with his supervisor (e.g. Leo and Booker, 1992; Booker and Leo, 1993; Leo and Booker, 1993) in the course of his work to solve equation (17). The boundary element method is increasingly popular because:

- the dimensionality of the problem is reduced by one; a two dimensional problem only requires the evaluation of line integral along the boundary and as for a three dimensional problem, a surface problem
- data input preparation and problem size are greatly reduced
- it is generally more accurate than the finite element and finite difference methods
- it is suitable for solving problems with infinite boundaries such as those commonly found to occur in geotechnical problems

Of course, like all other methods, the boundary element method is not without its drawbacks, the main problem being that it can can be extremely cumbersome to use when dealing with a domain which is highly heterogeneous. It will not be the task in this paper to describe the theory relating to the development of the boundary element method due to a lack of space and since interested readers can always refer to the quoted papers which have been published. Instead, some illustrative examples of how the boundary element method can be employed to analyse contamination problems will be presented.

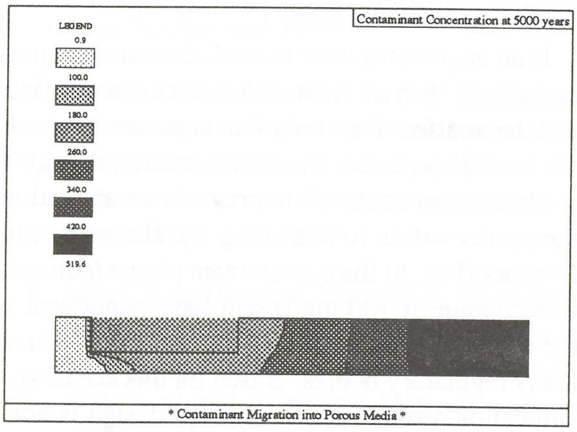

Figure 1: Scenario 1

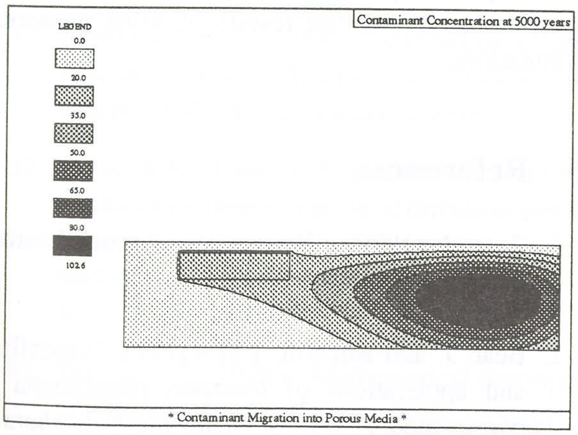

Figure 2: Scenario 2

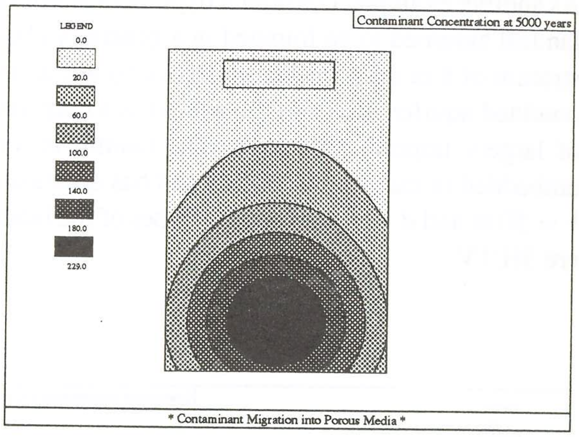

Figure 3: Scenario 3

4.0.1 Illustrative Example 1

In the following, contours of constant concentration of a conservative species at 5000 years are obtained as a result of contaminating effects from a 20m x 5m rectangular repository with an initial concentration of 1000 mg/l. The dispersion coefficient is assumed to be 0.01 m^2/a in both the x and y directions in the plane. The magnitude of the groundwater flow is 0.01m/a in either the x or the y directions. The 3 different scenarios which have been examined are:

- a repository embedded in an 8 m clay layer with x-direction advection, zero flux boundary condition at the ground surface and at the 8 m interface (i.e. the underlying layer is assumed to be much more impermeable).
- repository buried at 2 m in a homogeneous half-space with x-direction advection and zero concentration boundary condition at the ground surface.
- repository buried at 2 m in a homogeneous half-space with z-direction advection and zero concentration boundary condition at the ground surface.

Figures 1,2, and 3 are useful pictorial presentations of the extent of contaminant migration under different boundary conditions. These simulation studies help to establish the influence of boundary effects on the concentration contours.

4.0.2 Illustrative Example 2

As another example, consider a hypothetical sanitary landfill assumed to be founded in a generally clayey stratum of 8 m deep further underlain by a 2 m deep confined aquifer. Beneath the aquifer is a deep layer of largely impermeable soil. The landfill is long, embedded in the stratum of clay and has dimensions $b = 50$ m and $d = 4$ m. The sideslopes of the landfill are 3H:1V.

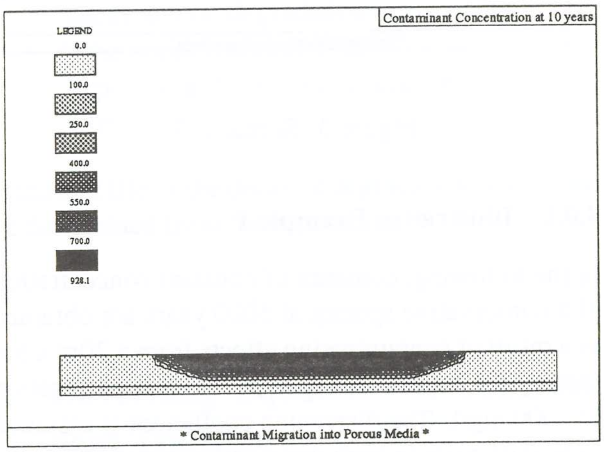

Figure 4: Contamination from landfill

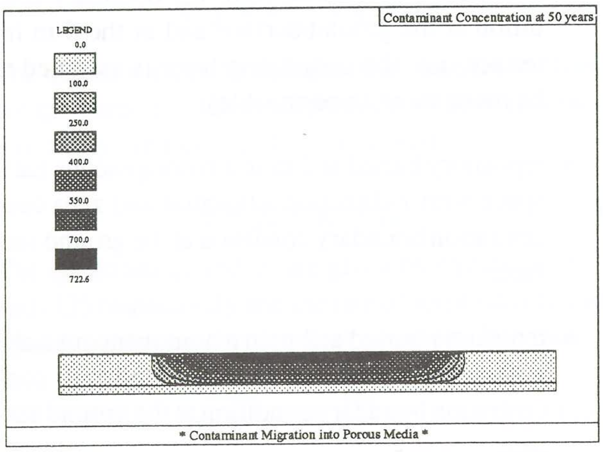

Figure 5: Contamination from landfill

The migration of leachate from the landfill into the natural groundwater regime is a major environmental concern requiring careful analysis. For the purpose of illustration, contamination contours are presented for times at 10, 50 years. The ground surface and the boundaries at infinity are specified as having zero concentration throughout the period of simulation. For simplicity, it has been assumed that a total head difference of about 2.5 m exist between the base of the landfill and the top of the aquifer, the hydraulic conductivity of the soil in the aquitard or the clay liner is of the order of 10^{-7} cm /s, thus yielding a Darcy velocity of about 0.05 m/a. This value of Darcy velocity will be assumed to occur uniformly throughout the aquitard. In more difficult situations, the steady state groundwater flow field may be determined using the program FESEP developed by Booker and Balaam (1983).

If no engineered liner is used, the natural aquitard is the only 'barrier' that stands between the landfill and the aquifer. Thus only 4 m separates the base of the landfill from the top of the aquifer, though this is of sufficient thickness to prevent contamination of the aquifer within 10 years (Fig. 4). However, within 50 years (Fig. 5) the contaminant plume (concentration contour of 100 mg/l) will have penetrated into the aquifer, violating the regulatory requirements of many regulatory bodies. Based on this scenario, the simulation results indicate that the design is not acceptable thus pointing to a need for introducing some form of liner barriers as an impediment to contaminant transport.

5 Conclusion

This paper has presented the processes of contaminant transport and the results of some numerical simulations.

6 References

1. Bear, J. (1979), 'Hydraulics of groundwater', *McGraw-Hill, New York.*

2. Bear, J. and Buchlin, J.M. (1991), 'Modelling and applications of transport phenomena in porous media', *Kluwer Academic Publishers.*

3. Booker, J.R. and Balaam, N.P. (1983), 'Program FESEP – steady state seepage analysis', *Dept. of Civil Engineering, University of Sydney.*

4. Booker, J.R. and Leo, C.J. (1993), 'A FLT boundary integral equation method for analysis of contaminant leakage from waste repositories in porous medium', *Proceedings of 2nd APCOM.*

5. Freeze A.R. and Cherry J.A. (1979) , 'Groundwater', *Englewood Cliffs: Prentice-Hall.*

6. Gillham, R.W. and Cherry, J.A. (1979), 'Predictability of solute transport in diffusion-controlled hydrogeologic regimes', *Proceedings of Symposium on Low Level Waste Disposal: Facility Design, Construction and Operating Practices, Nuclear Regulatory Commission, Washington, D.C.*

7. Leo, C.J. and Booker, J.R. (1992), 'Boundary element analysis of contaminant transport from waste repositories', *Proceedings of the International Conf. on Computational Methods in Engineering, Singapore.*

8. Leo, C.J. and Booker, J.R. (1993), 'Boundary element analysis of contaminant transport in fractured porous media', *International Journal for Numerical and Analytical Methods in Geomechanics*, V17(7), pp471-492.

9. Luckner L. and Schestakow W. M. (1991), 'Migration Processes in the Soil and Groundwater Zone', *Lewis Publishers.*

10. Parker, R.J. and Sadlier, M. A. (1992), 'Geomembrane Applications in Australia', *Australian Geomechanics No. 22.*

11. Pfannkuch H.O. (1963),'Contribution a l'etude desplacements des fluides miscibles dans un milieu poreux', *Inst. Francais du Petrole.*

12. Rowe R.K., Caers C.J. and Barone F. (1988), 'Laboratory determination of diffusion and distribution coefficients of contaminants using undisturbed soil', *Canadian Geotech. Journal, Vol. 25, pp. 108-118.*

13. Travis, C.C. (1978), 'Mathematical description of adsorption and transport of reactive solutes in soil: a review of selected literature. Oak Ridge, Tennessee: Oak Ridge National Laboratory'.