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Contaminant transport through porous media: An overview of experimental and numerical studies

S.B. Patil [*] and H.S. Chore [a]

Department of Civil Engineering, Datta Meghe College of Engineering, Sector-3, Airoli, Navi Mumbai-400 708, India

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Abstract. The groundwater has been a major source of water supply throughout the ages. Around 50% of the rural as well as urban population in the developing countries like India depends on groundwater for drinking. The groundwater is also an important source in the agriculture and industrial sector. In many parts of the world, groundwater resources are under increasing threat from growing demands, wasteful use and contamination. A good planning and management practices are needed to face this challenge. A key to the management of groundwater is the ability to model the movement of fluids and contaminants in the subsurface environment. It is obvious that the contaminant source activities cannot be completely eliminated and perhaps our water bodies will continue to serve as receptors of vast quantities of waste. In such a scenario, the goal of water quality protection efforts must necessarily be the control and management of these sources to ensure that released pollutants will be sufficiently attenuated within the region of interest and the quality of water at points of withdrawal is not impaired. In order to understand the behaviour of contaminant transport through different types of media, several researchers are carrying out experimental investigations through laboratory and field studies. Many of them are working on the analytical and numerical studies to simulate the movement of contaminants in soil and groundwater of the contaminant transport. With the advent of high power computers especially, a numerical modelling has gained popularity and is indeed of particular relevance in this regard. This paper provides the state of the art of contaminant transport and reviews the allied research works carried out through experimental investigation or using the analytical solution and numerical method. The review involves the investigation in respect of both, saturated and unsaturated, porous media.

Keywords: contaminant transport; porous media; saturated; unsaturated; experimental investigation; analytical studies; numerical modelling; finite element methods (FEM); meshfree methods

1. Introduction

Groundwater is a valuable natural resource. Its contamination is one of the most typical hydro-geological and environmental problems. In many parts of the world, groundwater resources are under increasing threat from growing demands, wasteful use and contamination. The fate and transport of solute in soils and groundwater has long been a focus of experimental and theoretical

∗Corresponding author, Assistant Professor, E-mail: sbpatil_2009@rediffmail.com
[a] Professor and Head, E-mail: hschore@rediffmail.com

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research in subsurface hydrology. Solute transport in the soil and the groundwater is affected by a large number of physical, chemical and microbial processes; and the properties of the media. In many practical situations, one needs to predict the time behaviour of a contaminated groundwater layer. Most of the groundwater contaminants are reactive in nature and they infiltrate through the vadoze zone, reach the water-table; and continue to migrate in the direction of groundwater flow. Therefore, it is essential to understand the transport process of contaminants through the subsurface porous media. Several mathematical models have been developed in view of this based on the several numerical studies. Besides, there have been some experimental investigations conducted for understanding the behaviour of contaminant transport through different types of media. This paper reviews some of the works related to experimental and numerical modelling of contaminant transport through saturated and unsaturated porous media published in the literature. It further provides the information of various approaches that can be applied for modelling of contaminant transport.

1.1 Causes and sources of contamination

A groundwater contaminant is defined by most regulatory agencies as any physical, chemical, biological or radiological substance or matter in groundwater. The contamination can occur by natural processes, agricultural operations urban run offs, waste disposal practices, spills and leaks etc.

The natural process includes leaching of chemical deposits which further results in increased concentrations of chlorides, sulphates, nitrates and other inorganic chemicals. Besides leaching, the other most significant source is runoff. The water carries metals, pesticides, microorganisms and other organic chemicals. The third general source of groundwater contamination is waste disposal. It includes disposal of liquid and solid wastes. The liquid wastes include disposals from septic tanks, cesspools, sewage effluent, sludge in regard with the industrial wastes, surface impoundments and injection wells are probably the largest contributors to groundwater contamination.

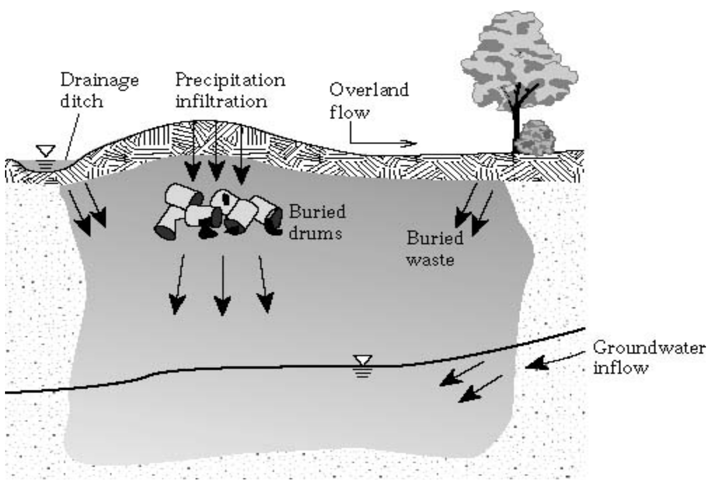

Fig. 1 Sources of fluid for the generation of landfill leachate

Moreover, the solid wastes include buried waste that is subjected to leaching by percolating rain water and surface water or by groundwater contact with the fill. This generated leachate (Fig. 1) can contain high levels of BOD, COD, nitrate, chloride, alkalinity, trace elements and even toxic constituents. On the contrary to this, the transient sources of groundwater and soil contamination include agricultural infiltration (Nitrogenous fertilizer, pesticides), leachate from chemicals stored above ground, infiltration from pits, ponds and lagoons, landfill leachate (municipal, industrial); and infiltration, spills (Jet fuels, industrial chemicals, waste oils, etc.).

The physiochemical reactions that can alter the concentration of an organic contaminant in groundwater can be grouped into five categories such as: hydrolysis of the contaminant in water, oxidation-reduction, biodegradation of the contaminant by microorganisms, adsorption of the contaminant by the soil; and volatilization of the contaminant to the air present in the unsaturated zone. The relative importance of each of these reactions depends on the physical and chemical characteristics of the contaminant and on specific conditions.

1.2 Transport process

To predict the environmental consequences of groundwater contamination one ought to know as to where the contaminant would interfere, when it would arrive; and what are the potential concentrations. The contaminant is introduced in groundwater by: (i) advection which is caused by flow of groundwater; (ii) dispersion which is caused by mechanical mixing and molecular diffusion; and (iii) retardation which is caused by adsorption.

The mathematical relationship between these processes can be written as follows

$$\frac{\partial}{\partial_{x_i}}\left[D_{ij}\frac{\partial C}{\partial_{x_i}}\right]-\frac{\partial}{\partial_{x_i}}\left(C\upsilon_i\right)-\frac{C'W'}{n}=R\frac{\partial C}{\partial t} \tag{1}$$

$$V_i=\frac{-K_{ij}}{n}\frac{\partial h}{\partial_{x_j}} \tag{2}$$

$$R=\left[1+\frac{\rho_b K_d}{n}\right] \tag{3}$$

where

C = contaminant transport
V_i = seepage or average pore water velocity in the direction x_i
D_{ij} = dispersion coefficient
K_{ij} = hydraulic conductivity
C' = solute concentration in the source or sink fluid
W' = volume flow rate per unit volume of the source or sink
n = effective porosity
h = hydraulic head
R = retardation factor
x_i = Cartesian coordinate

The following discussion uses a simplified two-dimensional representation to describe the transport of contaminants in groundwater. In a homogeneous, isotropic medium having a unidirectional steady state flow with seepage velocity V

$$D_L \frac{\partial^2 C}{\partial x^2} + D_T \frac{\partial^2 C}{\partial y^2} - V \frac{\partial C}{\partial x} = R \frac{\partial C}{\partial t} \tag{4}$$

where
C = contaminant concentration
V = seepage or average pore water velocity
D_L = longitudinal dispersion coefficient
D_T = transversal dispersion coefficient
R = retardation factor

1.2.1 Advection

The advection is the movement of dissolved solute with flowing groundwater at the seepage velocity in porous media. Advection and hydrodynamic dispersion are the physical properties that control the solute flux. The advection is governed by the Darcy's law as it is the transport of the solute with respect to flowing groundwater and hydrodynamic dispersion results from mechanical mixing and molecular diffusion. Darcy's law states that the flow rate of water through soil from point 1 to point 2 is proportional to the head loss and inversely proportional to the length of flow path

$$Q = -K.A \frac{h_2 - h_1}{L} \tag{5}$$

where
Q = groundwater flow rate
A = cross sectional area of flow
h_2-h_1 = head loss between point 1 and point 2
L = distance between point 1 and point 2
K = hydraulic conductivity

The actual seepage or average pore water velocity can be calculated as

$$V = \frac{Q}{n, A} - \frac{K}{n} \frac{h_2 - h_1}{L} \tag{6}$$

Where n is effective porosity or percent of interconnected pore spaces that actually contributes to the flow.

The Eq. (2) is a conservative estimate of the migration velocity of the contaminant transport. When only an advection is considered, a contaminant moves with the groundwater flow at the same rate as water, but in reality it is also affected by dispersion and retardation.

1.2.2 Dispersion

It is the result of two processes- molecular diffusion and mechanical mixing. The mechanical dispersion (Fig. 2) or mechanical mixing occurs when contaminated groundwater mixes with non-contaminated groundwater resulting in a dilution of the contaminate, which is called dispersion.

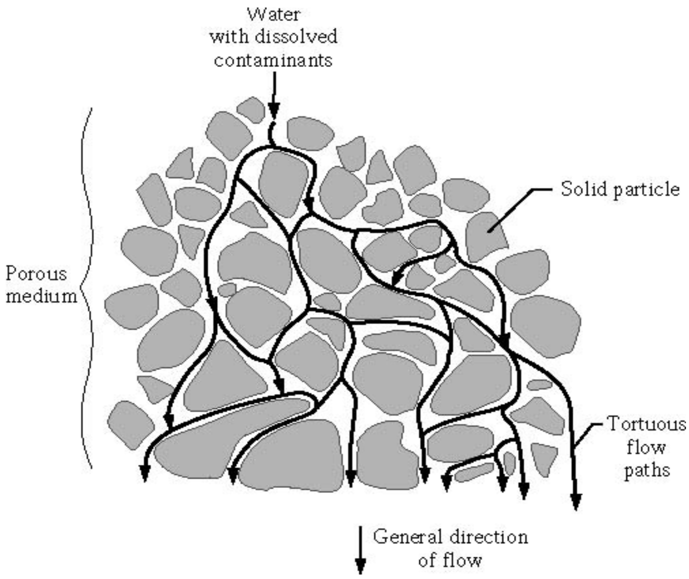

Fig. 2 Schematic of mechanical dispersion

The molecular diffusion is process where ionic or molecular constituents move in the direction of their concentration gradients. In this, the constituents move from regions of higher concentrations to regions of lower concentrations, the greater the difference greater the diffusion rate.

Molecular diffusion can be expressed by Fick's law as

$$F = -D_f \frac{dC}{dx} \tag{7}$$

where

F = mass flux per unit area per unit time
D_f = diffusion coefficient
C = contaminant concentration
dC/dx = concentration gradient

The Fick's law was derived for chemicals in unobstructed water solutions. When this law is applied to porous media, the diffusion coefficient should be smaller since the ions follow longer paths caused by the presence of solid particles in the solid matrix and because of adsorption on solids. This application yields an apparent diffusion coefficient D^* represented by

$$D^* = w.D_f \tag{8}$$

Where 'w' is empirical coefficient less than 1 which takes into account the effect of the solid phase of the porous media on the diffusion. Freeze and Cherry (1979) suggested the range of 'w' being 0.5 to 0.01 in order to account for tortousity of the flow path. Since these two processes cannot be separated in groundwater flow, the coefficient of hydrodynamic dispersion is taken into account.

$$D_L = a_L.V + D^* \tag{9}$$

$$D_T = a_T.V + D^* \tag{10}$$

where

D_L = longitudinal mechanical mixing component of dispersion
D_T = transversal mechanical mixing component of dispersion
a_L = longitudinal dispersivity
a_T = transversal dispersivity
a rough approximation of a_L by Gelhar *et al*. (1992) is
a_L = 0.1 L

where L is the length of the flow path.

For lengths less than 3500 m, given the a_L = 0.0175 $L^{1.46}$ for transverse dispersivity. The value of a_T is typically 1/10 to 1/100 of the longitudinal dispersivity a_L. These coefficients can be determined from breakthrough column tests in the laboratory or tracer tests in the field.

Advection-dispersion transport

Advection and Dispersion is predominant in saturated or nearly saturated clay barriers.

For one dimensional flow

$$D_x \frac{\partial^2 C}{\partial x^2} - \tilde{\upsilon} \frac{\partial C}{\partial x} = \frac{\partial C}{\partial t} \tag{11}$$

This can also be expressed along a flow line by using L for x where L is the co-ordinate direction along the flow line. D_L is the longitudinal coefficient of hydrodynamic dispersion and $\tilde{v}_L$ is the average linear velocity along the flow line.

The analytical solution to the Eq. (11) is given below

Initial condition $C(x, 0) = 0 \quad x \geq 0$
Boundary condition $C(0, t) = C_0 \quad t \geq 0$
Boundary condition $C(\infty, t) = 0 \quad t \geq 0$

$$C(x,t) = \frac{C_0}{2}\left[erfc\left(\frac{x - vt}{2\sqrt{D_L t}} \right) + \exp\left(\frac{vx}{D_L} \right) erfc\left(\frac{x + vt}{2\sqrt{D_L t}} \right) \right] \tag{12}$$

where x = distance from injection point

Argument of $\exp(vx / D_L)$ is the Peclet number $P_e = \tilde{v}_L x / D_L$

i.e., measure ratio of the rate of transport by advection to the rate of transport by diffusion.

The large Peclet numbers (P_e > 100) indicate that advection dominates. When advection dominates, the second term on the right hand side becomes negligible.

1.2.3 Sorption

Sorption is the exchange of molecules and ions between solid phase and liquid phase, including adsorption and desorption. Adsorption is attachment of molecules and ions from the solute to the solid phase causing a decrease of concentration of the solute this is called Retardation. Desorption is the release of molecules and ions from the solid phase to the solute.

The retardation coefficient can be calculated on the distribution or adsorption coefficients of the contaminant and the characteristics of the porous medium as

$$R = \left[1 + K_d \frac{\rho_d}{n}\right] \tag{13}$$

where

K_d is distribution or adsorption coefficient described previously. The values ρ_d and n are the bulk density and porosity of the soil. The velocity of the contaminant in groundwater can be calculated as follows

$$V_c = \frac{V}{R} \tag{14}$$

where V_c is the velocity of the contaminant movement in groundwater, V is the groundwater velocity, and R us the retardation factor. A high retardation factor, i.e., high adsorption coefficient significantly retards the movement of the contaminant in groundwater.

1.3 Contaminants in the sub-surface and modelling

Migration of contaminants in soil and groundwater has become an area of increasing research and interest in the recent decades. The transport process of the contaminant in the sub-surface and atmospheric environment is illustrated in Fig. 3. Once released into the subsurface system, contaminants will interact hydrologically, physically and chemically with both- the native water and the granulated solid matrix. The major hydrological and physical processes of interaction include advection, dispersion, diffusion, decay; and chemical reactions. The physical processes determine the way mass is moved from one point to another − the system of groundwater conveyance and mixing. The chemical and biological processes redistribute the mass among different chemical forms, or into and out of the aqueous system. To assess the risk of groundwater contamination by contaminants, their mobility and persistence in the soil need to be determined.

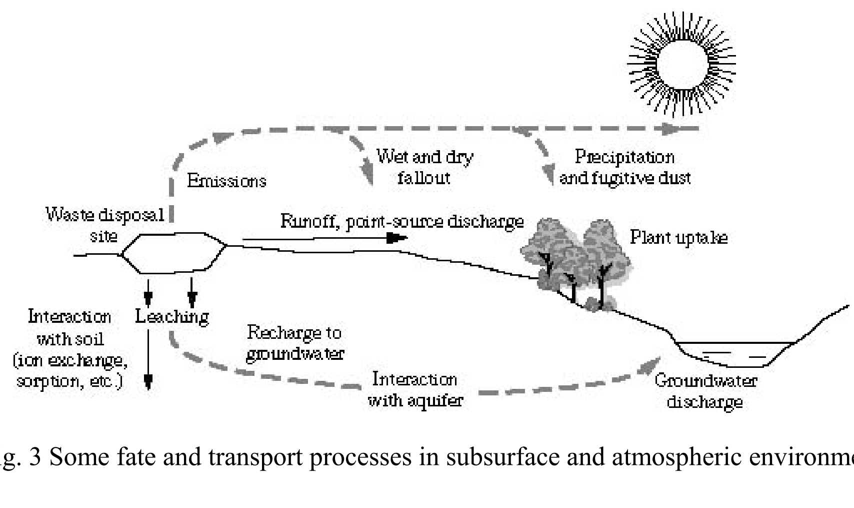

Fig. 3 Some fate and transport processes in subsurface and atmospheric environment

The numerical studies leading to the development of the mathematical modelling, laboratory and field experiments have been conducted by the several researchers with the ultimate goal of understanding the behaviour of contaminants in porous media and for predicting the future contamination level. The successful engineering solutions to these problems entail a firm understanding of the principles of solute transport through porous media. The complex factors that control the movements of contaminants in the porous media and the resulting behaviour of contaminant plume are usually intricate to assess because of the interactions of many factors, i.e., dispersion, advection, sorption degradation, etc., that affect the degree and rate of contaminant movement.

For solving the transport problems having simplified boundary conditions and geometries, exact analytical solutions are available in the literature (Bear 1979). While the historical response of the system could be inferred from the available field records, there is no way of computing the future behaviour of the full-scale system unless it is actually subject to conditions. A full-scale experiment would be prohibitively costly and time consuming. The only feasible recourse therefore is to construct a model, which reasonably portrays the behaviour of full-scale system and simulate the relevant physical parameters, and describes the overall significant characteristics of the transport phenomena.

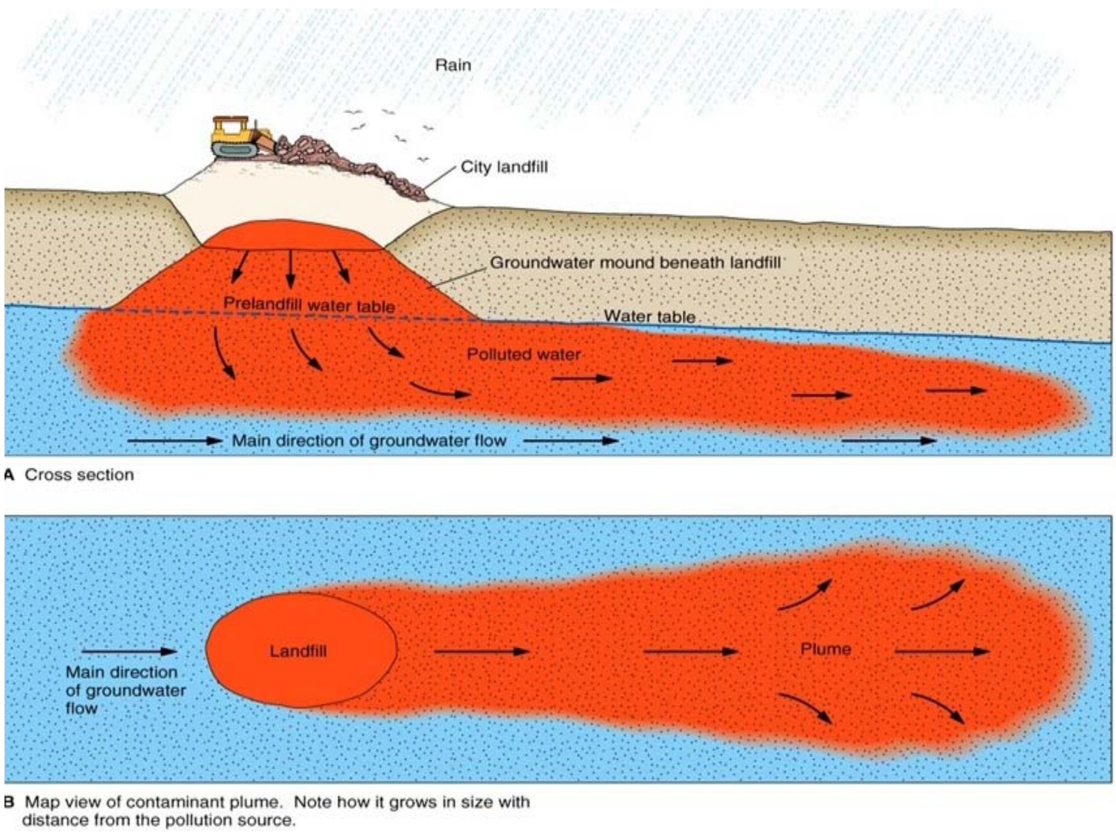

Fig. 4 Ground water contamination

In order to protect the environment from contamination by pollutants, waste materials are usually placed in engineered landfills (Fig. 4). The modelling of contaminant migration through landfill liners and natural soil deposits in geo environmental engineering is an important task since the landfills have to contain the waste till their design life.

To demonstrate the likelihood of compliance with regulations, environmental engineers now make routine recourse to the mathematical models of the proposed landfill. One outstanding feature of the modelling approach is the capacity for predictions to be made well into the future, thereby demonstrating the likely impacts of current practice on the environment and future generations. The validity of the predictions based on mathematical modelling of contaminant transport has been investigated and shown to be good for time periods measured in decades by Quigley and Rowe (1986).

Impressive arrays of analytical and numerical models are available, both- for instantaneous pluses and for continuous sources in the literature. Whatever model is contemplated, one of the more formidable problems in contaminant transport is the difficulty in assessing the important parameters and coefficients, including source concentration and dimensions, seepage velocity, time since the contaminant first entered the groundwater and up to three dispersivities for a three dimensional problem (Bear 1979).

The use of numerical models in problems of contaminant transport is rapidly increasing in response to the need to measure, monitor and apply predictive approaches to contaminant plumes of various size and shape. The available analytical solutions of the transport equation consider one or two governing processes, usually in a simple flow domain with uniform transport parameters. In contrast, the numerical simulation of contaminant transport offers a vehicle that can integrate, in some approximate way, the effects of several controlling processes; and it is the only method of calculation which can accommodate geometries and parameter distributions.

2. Review of literature

Deterioration of the environmental indicators of many ecosystems has led, over the last two decades; to stringent environmental control and an increase in research into the fate of contaminants in soil, air and water. The ability of practitioners and regulators to predict the extent and rate of dispersion of pollution plumes can help them develop better pre-emptive or remedial strategies. The contamination of soil and groundwater by chemicals has become an increasing concern in the recent past. These chemicals enter the groundwater system by a wide variety of mechanisms, including accidental spills, land disposal of domestic and industrial wastes and application of agricultural fertilizers. Once introduced into an aquifer, these contaminants will be transported by flowing groundwater and may degrade water quality at nearby wells and streams. For improving the management and protection of groundwater resources, it is important to first understand the various processes that control the transport of contaminants in groundwater. Predictions of the fate of groundwater contaminants can be made to assess the effect of these chemicals on local water resources and to evaluate the effectiveness of remedial actions.

There is need for the study in the area of contaminant transport modelling for the prevention of unacceptable long-term environmental impact of the contaminants on environment. Consequently, the review intends to focus on the subject related to transport of contaminants in the subsurface environment; through expatiation on the mathematical modelling and experimental studies, asserting predominantly on the transport processes affecting the growth of contaminant plume in the soil layer, and the pertinent contaminant transport models developed.

This section provides a critical review of the various approaches available for modelling of contaminant transport through the different types porous media such as saturated and unsaturated media. The review based on the previous research is further classified under the sub-heads of analytical, experimental and numerical studies in respect of saturated and unsaturated porous media. A brief review of the available literature on contaminant transport through landfill liners is also provided.

2.1 Preferential path and transport processes in sub-soil

The residues in the soil are subjected to various processes, viz., adsorption, movement and degradation. Advection carries the contaminant at an average rate as a plug flow. However, in reality, the solute is seen to spread out from the flow path. This spreading or mixing phenomenon is called dispersion. At the microscopic level, the fluid flow within a porous medium is actually a movement along the tortuous three dimensional passages in voids.

The local velocities in the passages are different from their macroscopic average values, both in magnitude and in direction. Due to the complexity of the micro-geometry of porous media, one has to describe the flow phenomena in porous media on a macroscopic basis. The spatial average method is a way to transfer properties of porous media from the microscopic level to the macroscopic level. The dispersion consists of mechanical hydrodynamic dispersion and molecular diffusion. Mechanical dispersion refers to the spreading and mixing caused by variation in velocity with which water moves and the fluid mixing due to the effects of unresolved heterogeneities in the permeability distribution.

The molecular diffusion is caused by the non-homogeneous distribution of contaminant in a fluid. The contaminant molecules in high concentration will move to the low concentration areas to form a uniform concentration distribution. Several mechanisms causing macroscopic mixing are generally accounted for in the dispersion coefficient; viz, mixing due to tortuosity, inaccessibility of pore water, recirculation due to flow restriction, macroscopic and hydrodynamic dispersion and turbulence in flow path. The formation of a dispersion coefficient tensor for an anisotropic medium requires five dispersivities by Bear (1979). However, Burnett and Frind (1987) suggested an approximate formulation which requires specification of only three dispersivities. Their approach is based on the recognition that transverse dispersivity is generally much smaller in the vertical direction than in the horizontal direction.

Klotz *et al*. (1980) conducted a large number of laboratory and field experiments which dealt with the relationship between longitudinal and transversal dispersion coefficients in loose soil and the mean flow velocity. Gelhar *et al*. (1992) reviewed 59 different field studies and denoted that the longitudinal dispersivities range from 10^{-2} to 10^{4} m for scales ranging from 10^{-1} to 10^{5} m, but the largest value for higher reliability data was only 250 m. Transverse dispersivity is 6 to 20 times smaller than longitudinal dispersivity.

2.2 Work on saturated porous media

Some of the published works on the various studies in regard with the contaminant transport through saturated porous media are reviewed in this section.

2.2.1 Analytical studies

The analytical models are the valuable tools for investigating solute transport in porous media and for estimating potential for contaminant transport in groundwater. Analytical solutions are usually derived from the basic physical principles and are free from numerical dispersions and other truncation errors that often occurred in numerical simulations. They provide a useful approach for making screening-level calculations and are also ideal for addressing uncertainty under conditions of limited information. They also provide computationally efficient tools for modelling the fate and transport of groundwater contaminant plumes (Clement 2001).

In general, the analytical models can be evaluated much more quickly than numerical solutions, making it feasible to compute a large number of results for *Monte Carlo* analysis, for example. For a given transport equation, analytical solutions would differ according to the assumed domain geometry, the source geometry and boundary conditions. The ease of use makes analytical transport models the obvious foremost step in any mass transport modelling.

Ogata and Banks (1961) presented a solution of the differential equation governing the process of diffusion, which gave an asymmetrical concentration distribution similar to the experimental behaviour. Numerical experiments demonstrated that 'this solution approaches that is given by symmetrical boundary conditions, provided the dispersion coefficient to be small and the region near the source is ignored'.

Several analytical solutions for the movement of chemicals in a one dimensional semi-infinite system using Laplace transform techniques has been developed by van Genuchten (1981). The governing transport equation includes terms accounting for linear equilibrium adsorption, zero-order production and first-order decay. An analytical solution for contaminant transport from a finite and continuous source in a continuous flow regime has been developed by Domenico and Robbins (1985). The significance of the approach is that it provides a closed form solution without involving numerical integration procedures. Domenico (1987) developed a mathematical model for a finite source that incorporates one dimensional groundwater velocity, longitudinal and transverse dispersion, and some form of decay for either radionuclide's or biodegradable organics. This solution is one of the most widely used analytical solutions for three dimensional solute transport in the field of contaminant hydrogeology and has been implemented in several risk assessment codes.

Wexler (1992) presented analytical solutions to the advection-dispersion solute equation for a variety of boundary condition types and solute-source configurations in one, two and three dimensional systems having uniform groundwater flow. Solutions were presented in a simplified format, together with information on important assumptions in derivation and limitations to their use. Runkel (1996) developed an exact analytical solution to the advection-dispersion equation subjected to a continuous load of finite duration. The results of the analysis were compared with the approximate analytical solution. The necessity of the exact solution to be used for verification of numerical solutions wherein higher level of accuracy was underscored. Three dimensional analytical solutions for solute transport in saturated homogeneous porous media were developed by Sim and Chrysikopoulos (1999). The models accounted for three dimensional dispersion in a uniform flow field, first-order decay of aqueous phase and sorbed solutes with different decay rates and non-equilibrium solute sorption onto the solid matrix of the porous formation.

Park and Zhan (2001) generated analytical solutions of multi-dimensional concentration fields originated from one, two and three dimensional, finite sources within finite thickness aquifers using the Green's function method. Guyonnet and Neville (2004) evaluated the analytical solution, for calculating three dimensional solute transports with decay for a vertical plane source at a constant concentration. Analytical solutions for two primary diffusive flux scenarios, i.e. outward and inward diffusive flux scenarios, for constant boundary solute concentrations and a variety of initial solute concentration distributions were presented by Shackelford and Lee (2005). Further, Srinivasan *et al*. (2007) performed a rigorous mathematical analysis on the origin and development of the three dimensional *Domenico solution* (1987). It was demonstrated that the Domenico solution is a true analytical solution when the value of longitudinal dispersivity is zero.

2.2.2 Numerical studies

Numerical modelling of pollutant migration in porous media has recently received a great deal of attention due to an increased interest in the preservation of the quality of the environment and particularly the protection of groundwater from various pollutants. Numerical models are able to account for the complexity of the subsurface and accommodate complicated boundary conditions. The finite difference methods have traditionally been applied to solve flow and transport equations. One of the most important implementations of the finite difference approach is in the powerful code SWIFT (Dillon *et al*. 1978) and its succeeding works.

The finite element method has been applied to numerous problems of flow through porous media, groundwater flow, multiphase flow, flow with a phreatic surface, hydrodynamic dispersion, consolidation; and heat and mass flow through porous media. It can handle any shape of boundary and any combination of boundary condition, inhomogeneous and anisotropic media, moving boundaries, free surfaces and interfaces, deformable media, etc. Numerical solutions have been presented in the literature to the problems of the saturated and unsaturated zones treated as a single flow domain, usually using pressure as the dependent variable. This section gives a brief review of some of the published works related to numerical modelling of contaminant transport through saturated porous media.

van Genuchten and Alves (1982) published a list of mathematical models and several computer programs for solution of the one dimensional convective-dispersive solute transport equations. Laplace transformation technique was used to derive the solutions that are applicable to semi-infinite system. Rowe and Booker (1985) presented a technique for analysing two dimensional transport of contaminant from a landfill into a homogeneous clayey layer. Rowe and Booker (1986) proposed a technique for the analysis of two and three dimensional pollutant migration through a layered soil medium. It was an extension of their earlier solution for plane diffusion in a single homogeneous layer of soil using the finite layer method. The parametric investigations were carried out to study the characteristics of contaminant migration in a layered soil system. Rowe (1987) reviewed the methods of predicting contaminant transport through saturated and unsaturated clayey barriers.

Frind (1988) investigated the influence of exit boundary condition on the *Galerkin* finite element solution of advection-dispersion equation. The study revealed that the numerical solution in a finite domain with free exit boundary behaves like an infinite domain solution. Rowe (1989) reviewed various simpler techniques that are used to make an assessment of potential contaminant impact of a waste disposal facility. The diffusion was found to be an important (often critical) mechanism controlling contaminant transport in well designed modern landfills with clayey barriers.

Tompson and Gelhar (1990) developed a three dimensional model based on random walk particle method to study detailed contaminant movement through large heterogeneous porous media. Harari and Hughes (1994) developed and analysed finite element method for solving problems of steady advection-diffusion with production. Rowe and Nadarajah (1996) discussed the importance of verifying contaminant transport codes and the techniques that are commonly used. Several finite element models for solving the advection diffusion- reaction equation were compared by Codina (1998) and the similarities and differences between the models were further emphasized. Zairi and Rouis (2000) proposed a two dimensional finite element model for contaminant transport through a landfill liner. The model was validated by comparing the results with the Ogata analytical solution and *Rowe* semi-analytical solution, together with the experimental results.

A numerical model for the two-dimensional simulation of passive pollutant transport in a porous medium using particle strength exchange method was proposed by Zimmermann *et al.* (2001). Chao *et al.* (2003) proposed a composite modelling approach for simulating the three dimensional subsurface transport of dissolved contaminants with transformation products through an unsaturated and saturated porous media. The errors of the general form of finite difference method for two dimensional advection–dispersion equations with linear sorption were examined by Ataie-Ashtiani and Hosseini (2005).

Ataie-Ashtiani and Hosseini (2005) examined the errors of the general form of finite difference method for two dimensional advection–dispersion equation with linear sorption. The variation of truncation errors were illustrated as function of Peclet, Courant, and sink/source numbers for explicit, implicit, Crank–Nicolson and alternating direction implicit upstream methods. Li *et al.* (2005) presented a combined approach combining multigrid methods and adaptive local grid refinement, in conjunction with the Lagrangian–Eulerian finite element method to simulate contaminant transport in the 3D subsurface.

A new class of algorithms for solving one and two dimensional transient advection-dispersion equation has been presented by Rao and Medina (2005, 2006). The multiple domain algorithm (integrated with a finite difference formulation) was used in these studies to solve the contaminant transport equations. Carlier *et al.* (2006) presented a probabilistic model to reproduce laboratory flow column experiments for granular materials with a single intergranular or a dual porosity. The probabilistic computations are in good agreement with the experimental data.

Craig and Rabideau (2006) modelled two dimensional contaminant transport using an *Eulerian* finite difference method based on the analytical element method derived parameters. El-Zein *et al.* (2006) proposed three dimensional finite element method for solving a wide variety of soil contamination problems. A *Galerkin* weighted-residual statement for the three dimensional form of the equations in the Laplace domain was formulated. Remesikova (2007) proposed a numerical model based on operator splitting approach for solving convection–dispersion–adsorption problems with both equilibrium and non – equilibrium adsorption.

2.2.3 Experimental investigations

Scientific investigations of transport involving repacked and/or homogenized soil cores often bear little resemblance to the physical reality. Nevertheless, experimental studies are the essential tools in the geo-environmental engineering for understanding the transport of adsorbed and non-adsorbed solutes through soil. Experiments can be used to obtain the properties necessary to model the movement of contaminant in porous media in a realistic situation. In nutshell, the experiments provide valuable insight about the porous medium, the behaviour of chemicals, and associated processes such as diffusion, dispersion, anion exchange and sorption during transport. The brief review of some of the significant experimental investigations reported in the past is given below.

Barone *et al.* (1992) described a laboratory diffusion-test for estimation of the diffusion coefficient (D) and the adsorption coefficient (K_d) for several volatile organic species in a clayey soil. Rowe and Badv (1996a) conducted a series of chloride diffusion tests on a clayey silt, silt and sand- both for single layer and two layer systems and suggested that the existing solute transport theory can adequately predict the chloride migration through landfill liners at near saturated conditions. Wang *et al.* (1998) conducted the two types of column and well simulation tests to obtain the properties necessary to model the movement of contaminants in porous media in a realistic way and compared the numerical predictions made by the Laplace transform finite element method (LTFEM). The study revealed that the LTFEM is able to deal with a contaminant transport problem under non-uniform flow conditions.

Rosqvist and Destouni (2000) modelled lithium transport through an undisturbed solid waste sample and a pilot-scale experimental landfill by use of probabilistic *Lagrangian* approach. The relevant conceptualisation and quantification of the water and solute movement through preferential pathways is found to be critical for meaningful extrapolation of observations in environmental assessments of long-term landfill performance. Rowe *et al*. (2000) performed the several inorganic diffusion tests on *Geosynthetic Clay Liners* (GCL) and bentonite specimens; and stated that 'there is a linear correlation between the diffusion coefficient and the final bentonite void ratio for both sodium and chloride'.

Likos and Lu (2004) described a new lecture module and laboratory experiment for demonstrating chemical transport phenomena in soils. Simple coloured dye was used to simulate a contaminant in a flowing soil-water system, thus precluding the requirement for expensive and complex analytical equipment involved in traditional chemical transport testing and creating a highly visually oriented learning environment.

Fox *et al*. (2011) presented an experimental and numerical investigation of coupled consolidation and contaminant transport in porous media. Diffusion and large strain consolidation-induced transport tests were conducted on composite specimens of kaolinite slurry consisting of an upper uncontaminated layer and a lower layer contaminated with potassium bromide. Effluent concentrations and mass outflows were higher for the boundary nearest to the contaminated layer in the double-drained case. Simulations also indicated that, for the conditions of this investigation, a reduction in specimen height yielded earlier breakthrough and higher levels of contaminant mass outflow.

Ballarini *et al*. (2012) described the detailed numerical simulation of highly controlled laboratory experiments using uranine, bromide and oxygen depleted water as conservative tracers for the quantification of transverse mixing in porous media. Synthetic numerical experiments reproducing an existing laboratory experimental set-up of quasi two dimensional flow through tank were performed to assess the applicability of an analytical solution of the 2D advection-dispersion equation for the estimation of transverse dispersivity as fitting parameter. From the results, an improved experimental set-up as well as a numerical evaluation procedure could be developed, which allow for a precise and reliable determination of dispersivities.

Massimo *et al*. (2012) performed multi-tracer laboratory bench-scale experiments and pore-scale simulations in different homogeneous saturated porous media. The results show that a non-linear compound-dependent parameterization of transverse hydrodynamic dispersion is required to capture the observed lateral displacement over a wide range of seepage velocities.

Recently, Sharma *et al*. (2013) reported the experimental investigation of contaminant transport through saturated layered soil using the soil column experiment. The flow of the solute transport through such system was also simulated numerically. A pulse type boundary condition was used during the experiment. An implicit finite difference numerical analysis was also carried out to get the numerical solution of advective dispersive transport including equilibrium sorption and first order degradation constant for the multi-layered soil.

2.2.4 Case studies

The design of waste disposal facilities typically involves a barrier that separates the waste from the general groundwater system. The impact of a disposal facility on groundwater quality would depend on the nature of the site, the climate, the type of waste, the local hydro-geology and the presence of a dominant flow path; and perhaps most importantly, on the nature of the barrier which is intended to limit and control contaminant migration (Rowe *et al*. 2004).

The bottom barrier is indented to minimise the migration of contaminants from the facility and hence the environmental impact of the facility is intimately related to its design and long-term performance. Natural clayey deposits, recompacted clay liners or geosynthetic clay liner frequently represent a key component of the barriers. The migrations of contaminants from solid waste landfills through both natural and compacted clay were examined through several well documented case histories (Barone *et al*. 1991, King *et al*. 1993). Diffusion, the movement of contaminants from areas of high concentration to areas of low concentration, has been researched extensively for low hydraulic conductivity barriers such as compacted clay liners (CCLs), geosynthetic clay liners (GCLs) and geomembranes (Lake and Rowe 2005).

Quigley and Rowe (1986) presented field and laboratory chemical profiles for a variety of dissolved chemical species migrating primarily by diffusion through 30 m thick natural clay deposits below a 15 year old domestic waste landfill. Birdsell *et al*. (2000) presented a case study that predicts the groundwater pathway dose for the performance assessment of the active, low-level, solid radioactive waste site located at Los Alamos National Laboratory, New Mexico. The finite element code FEHM (Finite Element Heat and Mass Transfer Code) was used for modelling the contaminant transport through porous and fractured media.

Cantrell *et al*. (2003) compiled distribution coefficient values measured for radio-active nuclides and toxic compounds at the Hanford site. Crowe *et al*. (2004) developed a two dimensional contaminant transport model for the transport of septic-system-derived contaminants to the marsh at Point Pelee, Ontario, Canada. The continuous source located at the free surface, wetland or within the saturated domain was considered while developing the afore-mentioned model. Lake and Rowe (2005) examined the performance of a geomembrane/compacted clay composite liner of 2.9 m thick, at the end of its 14 year operational lifespan of a landfill located in Ontario, Canada. This case study highlighted the importance of the compacted clay liner as part of the composite liner system in acting as a diffusion barrier during the lifetime of the lagoon as well as using relatively non-conservative contaminants such as chloride and sodium to estimate geomembrane failure times.

2.3 Works on unsaturated porous media

The prominent literature pertaining to the analytical, numerical and experimental studies germane to the unsaturated media is reviewed briefly in the sub-sequent sections.

2.3.1 Analytical studies

Mualem (1976) proposed a simple analytical model for predicting the unsaturated hydraulic conductivity curves by using the moisture content-capillary head curves and the measured values of the hydraulic conductivity at saturation. The model was based on a reasonable approximate evaluation of the hydraulic conductivity of a pore domain with varying shape. van Genuchten (1980) while proposing a model based on the theory postulated by Mualem (1976), formulated a new and relatively simple equation for soil-water content-pressure head curve for deriving closed form analytical expression for the relative hydraulic conductivity.

Nachabe *et al*. (1995) extended the Broadbridge and White nonlinear model to derive a closed form analytical solution for a one dimensional conservative solute transport in the unsaturated zone. It is to be noted that this analytical model would provide a means for evaluating the accuracy of existing numerical models. Prakash (2000) presented quasi-analytical model to analyse the transport of contaminants through the unsaturated and saturated soil zones. Guyonnet *et al*. (2001) proposed an analytical solution to the problem of steady-state solute transport by advection and diffusion-dispersion through a multi-layered mineral barrier, as well as an approximate transient solution. The hydrodynamic properties of the barrier are described by the functions of van Genuchten model (1980).

A range of analytical solutions to the one dimensional solute transport for horizontal flow by applying the concept of scale and time dependent dispersivity to unsaturated flow has recently been presented by Sander and Braddock (2005). Priesack and Durner (2006) derived an analytical expression for the conductivity of soils with heterogeneous pore systems, by combining the multi-modal representation of the retention function of Durner (1994) with the conductivity representation model of Mualem (1976).

2.3.2 Numerical studies

van Genuchten (1982) reviewed the accuracy and computational efficiency of numerical schemes for solution of the governing one dimensional flow and mass transport equation through unsaturated porous medium. Numerical schemes considered in the study included finite difference (FD), linear finite element method (LFE), mass-lumped linear finite element method (MFE), *Hermitian* finite element (HFE) with different Gauss and Lobatto points. It was concluded that the FD and MFE schemes generate the most stable results for steep concentration (or moisture) fronts and HFE scheme is superior in locating the correct spatial location of the moisture (or concentration) front.

Faust (1985) developed a finite difference model for modelling the transport of immiscible fluid under saturated and unsaturated conditions in the porous media. The formulation in terms of volumetric water saturation and fluid pressure in the immiscible fluid was presented and direct matrix technique and Newton-Raphson iteration was used for solving the equations. The numerical results illustrated the applicability of the model for the problems associated with immiscible contaminants in groundwater.

Piver and Lindstrom (1991) epitomised several of the main sources of errors that are introduced by the numerical approximations of the finite difference and finite element methods used to solve the transport equations for the unsaturated zone. The study noted that both the methods ‘give good predictive approximations for examining the transport behaviour of different types of chemicals for the same set of hydrodynamic conditions in the unsaturated zone of the subsurface’.

Panday *et al*. (1993) proposed a three dimensional finite element model for solving the unsaturated flow and transport equations. The Galerkin and upstream weighted residual procedures were employed for solving the flow and transport equations. Application of the model was illustrated by simulation examples involving assessment of moisture movement and contaminant migration from a shallow waste disposal design above multilayer unconfined aquifer system. Kool *et al*. (1994) developed the transport model for simulating three dimensional (3D) contaminant transport from land waste disposal sites with transformation products. Laplace transform – Galerkin technique was used for solving the transport equation. The model was verified by comparison against a fully 3D, variably saturated flow and transport code for a hypothetical landfill problem; and it was found to yield reasonable predictions of field scale solute transport.

van Genuchten and Simunek (1996) reviewed the developments in numerical techniques used for solving the unsaturated flow and transport equations, including methods for solving large sparse matrices resulting from spatial and temporal numerical discretisation. Meyer *et al*. (1997) presented a method to combine the generic probability distributions for unsaturated and saturated zone soil hydraulic parameters with site-specific water retention data using a Bayesian analysis for Site Decommissioning Management Plan (SDMP) sites.

Fityus *et al*. (1999) presented an approach for analysing one dimensional contaminant transport through an unsaturated landfill liner. A finite layer formulation was used to simplify the contaminant mass transport equation and account for heterogeneous soil profiles. Various assumptions concerning the flow regime beneath the landfill and the functional relation between volumetric water content and the diffusion coefficient in the transport equation were made to highlight differences between contaminant transport through saturated and unsaturated soils. Li *et al*. (1999) developed a numerical model to simulate miscible contaminant transport through unsaturated soils. The purpose of the model was to simulate the six physical and chemical phenomena governing miscible contaminant transport (i.e., advection, mechanical dispersion, molecular diffusion, adsorption, degradation and immobile water effect) in the soils. A finite element procedure, based on the characteristic Galerkin method with an explicit algorithm to solve the model equations was developed.

Diaw *et al*. (2001) proposed a one dimensional transport model (Wamos-T) for modelling non-reactive solute transport in saturated-unsaturated porous media. The transport equation is solved using operator splitting with discontinuous finite element method combined with a slope limiting procedure for discretisation of advection term. An implicit finite difference scheme was used for solving the dispersion and reaction terms. El-Fadel *et al*. (2002) assessed leachate migration away from the landfill (former quarry) for controlling the associated environmental impacts. A three dimensional, multi-phase, variably saturated model (PORFLOW) was used in the investigation to simulate subsurface flow and contaminant transport in a fractured porous medium. The results indicated the significant potential adverse impacts in the immediate vicinity of the landfill, and also pointed out the importance of point-of-compliance specifications in the landfill performance criteria.

An approximation scheme for solving a coupled system of flow and contaminant transport with adsorption in unsaturated–saturated porous media was developed by Kacur and Van Keer (2003). The approximation was based on time stepping, operator splitting and the method of characteristics. Finsterle (2004) reviewed the inverse modelling approaches for unsaturated and multiphase flow models. Based on the study, it was suggested that ‘a comprehensive inverse modelling package is an essential tool to improve test design and data analysis of complex multiphase flow system’.

Rood (2004) developed a one dimensional model for contaminant transport in the unsaturated zone under steady-state and transient flow conditions, from the principles of mixing-cell model. The solute transport processes included explicit treatment of advective processes, first-order decay, linear sorption and dispersion through implicit and explicit schemes. It was inferred that ‘though there are differences between the output produced by the mixing-cell model and those produced by the analytical and finite element methods, the differences cannot be considered meaningful in light of the rather large uncertainties associated with unsaturated transport modelling’.

Chu and Marino (2006) reviewed the compartmental systems and compartmental modelling methodologies, which are used for simulating contaminant transport in porous media and surface waters. Freiboth *et al*. (2007) developed a non-isothermal multiphase multi-component flow and transport model. The model accounts for the effects of swelling and shrinking of the media on the flow and transport, by adopting the hydraulic properties using constitutive relationships.

Javadi *et al*. (2008) presented a numerical model for predicting two dimensional contaminant transport and flow of water and air through unsaturated soils. In the model, the nonlinear system of governing differential equations was solved using a finite element method in the space domain and a finite difference scheme in the time domain. The model was validated using standard experimental results of contaminant transport in unsaturated soils.

2.3.3 Experimental investigations

The experimental study was carried out by Maciejewski (1993) in the context of the hydrodynamic dispersion of ionic solutes in unsaturated porous media. The results indicated linear relationship between the coefficient of dispersion and pore water velocity for given water content and a linear dependence of the dispersion coefficient on the water content for a constant pore water velocity. Porro and Wierenga (1993) performed a large column experiment to model the transport of relatively non-reactive contaminants through homogenous unsaturated soil under both transient and steady-state conditions.

Rowe and Badv (1996b) proposed a methodology for estimating the diffusion coefficients of chloride and sodium in unsaturated coarse sand and fine gravel. Methodology was based on the parameters obtained from the saturated diffusion tests conducted for similar material that was found to perform well in predicting the advective-diffusive transport of chloride and sodium through a two-layer soil system consisting of compacted clayey silt underlain by unsaturated fine gravel.

Awadallah and Abu-Ghararah (1997) demonstrated the horizontal contaminant transport through unsaturated sandy clay soil both analytically and experimentally. The contaminant transport equation with constant concentration boundary condition and constant dispersion coefficient was solved using the Boltzmann transformation. The predicted solute concentration profile was compared with that obtained experimentally and a fair agreement was observed in the analytical and experimental solutions. This ensured augmenting the applicability of the analytical method to simulate horizontal transport of non-reactive contaminants through initially dry soils.

2.4 Meshfree methods

The meshfree method, which is an extension of the conventional finite element method, is a very new area of research in computational solid and fluid mechanics. The mesh-free methods are in a rapidly developing and growing stage. The meshfree methods are being currently used in solving most of the problems in computational solid problems. Nowadays, these methods are finding increased applications in the area of geotechnical and geo-environmental engineering. In this section, some of the available publications relevant to the present study are briefly reviewed.

The diffuse element method (DEM), a meshfree method, was developed by Nayroles *et al*. (1992). In this, the moving least squares (MLS) approximation, proposed by Lancaster and Salkauskas (1981) for surface fitting, was used to develop the shape functions. Further, the DEM was extended to more solid foundation and the element free Galerkin method (EFGM) was proposed (Belytschko *et al*. 1994b, 1996), in which the MLS approximation was used in the Galerkin weak form to establish a set of algebraic equations. The EFGM was found to be very accurate and the rate of convergence of the EFGM obtained from numerical tests was found to be higher than that of the FEM. In addition, the irregularity of nodes did not affect the performance of the EFGM. The EFGM has been successfully applied to a large variety of problems including two and three dimensional linear and non-linear elastic problems.

Rao and Rahman (2000) presented an efficient meshfree method for analysing linear-elastic cracked structures subjected to single- or mixed-mode loading conditions. The method involves an element free Galerkin formulation in conjunction with an exact implementation of essential boundary conditions and a new weight function (student's *t* distribution weight function). Wang and Liu (2002) proposed a meshfree method called radial point interpolation method (RPIM) based on radial and polynomial basis functions for the solution of partial differential equations. The point interpolation method and Gaussian weak form was combined to establish a set of algebraic equations. A general point interpolation method was proposed with the combination of radial and polynomial basis functions. Boztosun and Charafi (2002) and Boztosun *et al*. (2002) proposed a meshfree method based on radial basis functions (RBF) for the solution of advection-diffusion equation. It was demonstrated that for a given accuracy, the proposed method 'is much faster than the traditional methods'.

Boztosun *et al*. (2003) presented a meshfree collocation method that uses compacted supported thin plate radial basis functions (CS-RBF) proposed by Wendland (1995) for obtaining a stable and accurate solution of the linear advection-diffusion equation. Authors conducted a comparative study of the numerical solution using TPS-RBF, CSRBF and finite difference. An algorithm based on the meshfree method for modelling the groundwater contaminant transport was developed by Li *et al*. (2003). The Kansa's method (1990) was used as a tool for solving the partial differential equations using collocation and Hardy multi-quadrics (MQ) radial basis functions. Numerical results presented for both the one and two dimensional contaminant transport illustrated the simplicity and accuracy of the method in comparison with the analytical solutions.

Vrankar *et al*. (2004, 2005) presented the approach for modelling the radionuclide migration through the geosphere using Kansa's method with geostatistics. The Hardy multi-quadrics (MQ) radial basis functions were used for solving the advection-dispersion equation. Results obtained from the radial basis schemes for the porous media were similar to the results obtained by the finite different methods. It may be noted that for solving advection-dispersion equation, radial basis function method is an appropriate alternative to the traditional method like finite difference method.

Coetzee *et al*. (2005) analysed the ultimate capacity of vertically loaded anchor and one loaded at 45° using a meshfree method called material point method. The material point method can model anchor pull out successfully and no special interface elements are needed to model the anchor–soil interface. Chinchapatnam *et al*. (2006) investigated the application of unsymmetric and symmetric meshfree collocation techniques with radial basis functions for solving the unsteady convection–diffusion equation. The method of lines approach was employed to discretise the governing operator equation and compared the performance of various globally supported radial basis functions. The collocation techniques required a very dense set of collocation points in order to achieve accurate results for high Peclet numbers.

Kumar and Dodagoudar (2008) proposed a methodology for modelling the one dimensional advection-dispersion equation involving first-order degradation through a saturated porous medium using EFGM. Student's *t*-distribution function was used as a weight function in the meshfree analysis. It was found that EFGM results agreed well with those obtained from the experiments, this ensures the accurate formulation of the EFGM.

Mategaonkar and Eldho (2011, 2012) have developed Meshfree models for isotropic cases only and the same were applied for hypothetical problem and further on a field problem to compute head distribution. The PCM models which are based on radial basis function were compared with FEM simulations while that of field problems are compared with boundary element based model results.

3. Conclusions

An effort has been made in this paper to provide the state-of-the-art of the concepts of contaminant transport through saturated and unsaturated porous media. Contaminant transport in the subsurface has been one of the most important research topics in the hydrological sciences and engineering involving the knowledge of environmental, geotechnical and hydraulics engineering subjects in civil engineering.

Several researchers conducted experimental investigations either in laboratory or in field besides carrying out the analytical studies and numerical modelling for understanding the behaviour of contaminants in porous media and for predicting the future contamination level. The experimental studies are regarded as complimentary to the analytical and numerical studies

With the advents of high power computers in the decade of 1970s, various methods such as finite difference, finite element, finite layer making use of the numerical techniques. The finite difference method (FDM) has been in use to solve differential equation systems for centuries.

The FDM works well for problems of simple geometry and was widely used before the invention of the much more efficient, robust finite element method (FEM). The FEM is now widely used in handling problems with complex geometry. The FEM is more suitable for modelling aims at predicting the contaminant transport in cases where clay is used as a barrier below landfills as it ensures accurate formulation of FEM. Recently, the extension of finite element method such as 'meshfree method' has become popular.

Still there is a need for the more reliable and efficient numerical method/s for modelling the contaminant transport which should account for advection-dominant transport processes without loss of accuracy and must take into account uncertainties associated with various flow and transport parameters such as saturated hydraulic conductivity, saturated soil – water moisture content, residual soil moisture content, dispersion and sorption coefficients and any other factor which contains some degree of uncertainty. These uncertainties must be explicitly accounted for, if scientifically and practically based estimates of contaminant concentration are to be achieved.

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