Transport of Contaminants During Groundwater Surface water Interaction

Jyoti Chaubey and Himanshu Arora

Abstract Groundwater–surface water (GW–SW) interaction plays an important role in alluvial aquifer-related studies because of the possible exchange of solutes along with water. Thus, the contamination of surface water may affect the groundwater quality as well. The present study provides a review of contaminant transport in groundwater due to GW–SW interaction. Various factors affecting the transport of contaminants during this interaction like flow of groundwater from surface water, contaminant characteristics and transport mechanisms have been studied. These transport mechanisms are formulated into a set of mathematical equations known as governing equations of groundwater flow and solute transport. The different numerical methods adopted for solving those equations have also been reviewed. The methods discussed are finite difference method, finite element method and meshfree methods.

Keywords Groundwater–surface water interaction · Contaminant transport · Numerical methods

1 Introduction

Increasing demand for groundwater as public water supply has made the susceptibility of groundwater contamination an important point of concern. There are various reasons of groundwater contamination, and interflow from surface water bodies is one of them (especially in alluvial aquifers). Various studies show the existence of interrelationship between surface water and groundwater [1].

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Contamination of any one of them will lead to the contamination of the other. Therefore, studying the transport processes of contaminant during the GW–SW interaction is of primary concern.

Factors involved in GW–SW interaction plays an important role in determining the direction of hydrologic exchange of water and contaminants. The transport of contaminants in GW–SW infiltration system can be determined by processes like advection, diffusion, dispersion, (ad)sorption/desorption and certain reactions including redox reactions, hydrolysis and biological transformations [2].

The solutions to the problems related to groundwater flow and solute transport are generally accomplished by solving the governing equations either by analytical methods or by using numerical methods. In general, the analytical methods provide an exact solution of the equations, but they are based on several assumptions (simplifying the inhomogeneity of the problem) which may not represent the actual complex field scenario. On the other hand, numerical methods account for the inhomogeneity of the nature and hence do not have such assumptions which might be severely violated in practice. Numerical methods can be broadly categorized into finite difference method (FDM), finite element method (FEM), finite volume method (FVM), method of characteristics (MOC), boundary element method and meshfree method (MM). A few of the above methods have been discussed briefly in the present study.

2 Contaminant Transport During GW–SW Interaction

The transport of contaminants during GW–SW interaction will primarily depend upon the direction of flow of water (interflow) which in turn will affect the exchange of contaminants. In the present study, the movement of contaminants from the surface water to groundwater has been studied. Hence, the contaminants from here onward will mean the contaminants present in groundwater due to the GW–SW interaction. Once the direction and amount of exchange is known, then the transport mechanisms of contaminants in groundwater need to be studied. In general, the factors responsible for the transport of contaminant during GW–SW interaction could be broadly categorized as:

- Direction of hydrologic exchange (water and contaminant) between groundwater and surface water
- Contaminants characteristics (conservative and non-conservative)
- Transport mechanisms (advection, diffusion and dispersion)
- Reactions (destructive and nondestructive)

These processes are elaborated in the proceeding sections.

2.1 Factors Affecting the Direction of Hydrologic Exchange Between GW and SW

The rate of interflow between hydraulically connected groundwater and surface water depends upon the relative levels of stream stage and the adjacent groundwater gradients, the position and geometry of the stream within the alluvial plain, groundwater aquifer geometry and parameters (such as hydraulic conductivity distribution, porosity) and boundary conditions [3].

Direction of the interflow depends upon the relative levels of water table and the free surface of the stream. If the water table level is lower than the free surface of the stream, then there will be effluent seepage from the stream, i.e., stream tends to lose surface water to the groundwater. On the other hand, stream gains water from groundwater when the adjacent water table is higher than the free surface of the stream. Flow-through streams gain groundwater through the upgradient bank and a portion of the stream bed and lose water through the downgradient bank [4] (Fig. 1).

Sophocleus [5] and Winter [6] performed analysis to investigate the groundwater coupling with the surface water and concluded that velocity, quantity and direction of exchange processes along with the spatial direction are controlled by gradient between surface water and groundwater table, leakage through riverbed material and hydraulic conductivity of soil.

Another important factor is the variable density-driven flow, where the density varies as a function of fluid pressure, suspended solid content in the fluid and

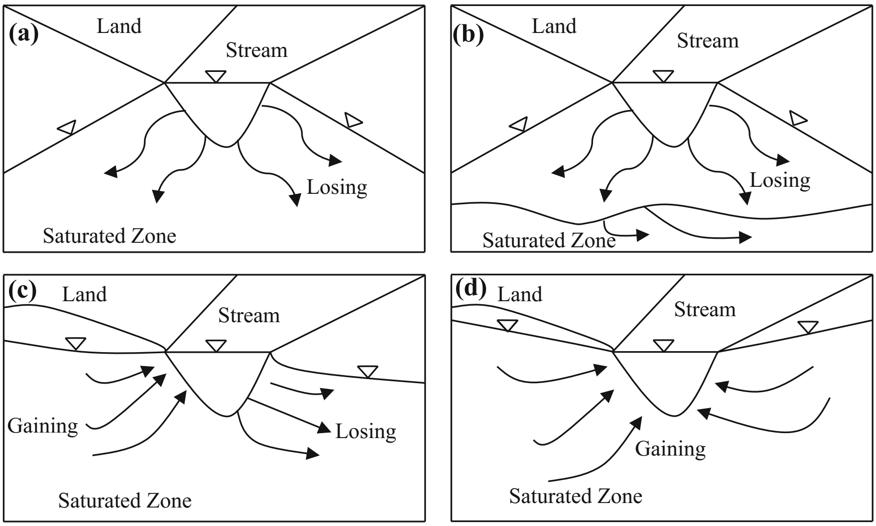

Fig. 1 Modes of exchange between surface and subsurface waters: (**a**) and (**b**) losing channel; (**c**) flow-through situation; (**d**) gaining channel

temperature of the fluid. Accounting for this flow is important in upconing below wells [7, 8], seawater intrusion in coastal regions, unstable flow phenomena where a denser fluid (seawater) overlies a less dense fluid (groundwater) [9] or wastewater disposal in deep salt formations [10].

Thus, the above studies show that the groundwater does interact with the surface water and the direction of flow depends upon the topography of terrain, the relative position of surface water bodies and groundwater flow systems, spatial distribution of flow system, i.e., hydraulic gradient and geomorphology of the region. Flow direction also depends upon the type of stream, that is whether it is ephemeral, intermittent or perennial, groundwater pumping (induced influent condition) and transpiration from groundwater by near-shore vegetation (which causes depression which in turn leads surface water to seep into the groundwater).

2.2 Contaminant Characteristics

Contaminants in groundwater are usually present as miscible solids/liquids or immiscible solids/liquids. Depending upon physicochemical properties, pollutants can broadly be classified as conservative pollutants and non-conservative pollutants. Conservative pollutants are the pollutants which do not transform physically or chemically to non-toxic substances under normal conditions (e.g., salts and metals). Pollutants which transform to non-toxic substances through physical, chemical or biological processes (e.g., organic pollutants, ammonia) are non-conservative pollutants. Conservative pollutants are long-lived and stable compounds which persist in the groundwater. Non-conservative pollutants degrade or transform into other compounds, and the transformation rate depends on the physical, chemical and biological conditions occurring within the groundwater.

2.3 Transport Mechanisms

Once contaminant (dissolved solute) enters the groundwater regime, the spreading of the contaminant is aided by several transport mechanisms which include advection, diffusion and hydrodynamic dispersion. A brief description of these mechanisms is given in the following sections.

2.3.1 Advection

The movement of the contaminant taking place due to groundwater flow is termed as advection. The amount of contaminant being transported is proportional to the contaminants concentration in flowing water and also the amount of groundwater flowing. Considering that the contaminant having an average concentration c is occupying some part of void space of porous media (let volumetric fraction = n) and groundwater is flowing with an average velocity q, then the *advective flux*, J_{adv}, of the contaminant is given as (Bear and Cheng [11])

$$J_{adv} = nqc \tag{1}$$

Thus, advection is the mass flux of the contaminant passing through a unit area of porous flow domain, normal to the flow velocity, per unit time.

2.3.2 Diffusion

The process in which the contaminant movement in groundwater is due to the concentration gradient, i.e., contaminant will move from a region of larger concentration toward a region of lesser concentration, is known as diffusion. It is independent of groundwater flow, i.e., it will occur even if the fluid is at rest. Hence, diffusion process is irreversible with time. It may occur horizontally or vertically along the layers of the aquifer medium provided the concentration gradient exists along the layers [12]. Flux due to molecular diffusion (J) is based on Fick's law and is given as

$$J = -\left(D_d^*\right)_{ij}\frac{\partial c}{\partial x_j} \tag{2}$$

where $\left(D_d^*\right)_{ij}$ = coefficient of molecular diffusion and c = concentration of contaminant.

2.3.3 Hydrodynamic Dispersion

Dispersion refers to the spreading of the contaminants along the flow path (average groundwater flow velocity direction). This happens because groundwater flow velocity is not uniform at particle level. In the void space between the particles of porous media, the magnitude of flow velocity varies from zero at the surface of the particles to the maximum value at some intermediate point in the pore space (Fig. 2a). Also the direction of the flow velocity changes due to the irregular arrangement of particles in the media (Fig. 2b). This change in magnitude and direction of the groundwater flow velocity results in mixing along the flow path. This mixing is termed as mechanical dispersion which results in a dilution of the contaminant as the flow advances [12]. Molecular diffusion and mechanical dispersion together are termed as hydrodynamic dispersion (Fig. 2) [11].

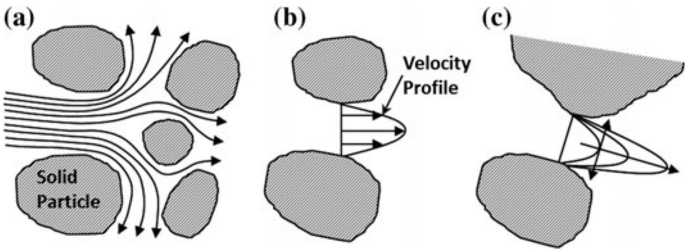

Fig. 2 Mechanical dispersion (**a**), (**b**), and molecular diffusion (**c**)

Coefficient of hydrodynamic dispersion (D_h) is given by

$$D_h = D + D_d^* \tag{3}$$

where D = coefficient of mechanical dispersion and D_d^* = coefficient of diffusion.

2.3.4 Adsorption

When contaminant enters the porous medium domain, there is a possibility that some of the contaminants may get adsorbed onto the particles (solids) of the porous medium. This adsorption results in the reduction of the contaminant concentration and retardation of the velocity of contaminant migration. Usually, the organic contaminants are more prone to adsorption and the presence of organic carbon in the aquifer material further enhances the process.

In earlier studies, contaminant transport was based on equilibrium adsorption models with two phases: sorbed immobile phase and dissolved phase. The sorbed immobile phase consisted of the solid phase and the sorbed contaminants onto it. Later on, it was concluded that this two-phase adsorption models of contaminant transport are applicable when the porous medium does not contain the mobile colloidal fines [13]. If mobile colloidal fines are present in the porous medium, then the contaminant transport models are based on equilibrium adsorption models with three phases: mobile colloidal phase, mobile liquid phase and the immobile solid phase [14].

2.4 Reactions

Reactions occurring in the saturated porous medium domain can be classified into two—destructive and nondestructive. Destructive reactions irreversibly transform or destroy the contaminant into other compounds. These reactions include abiotic reactions, biodegradation and radioactive decay. Nondestructive reactions are reversible processes which do not transform or destroy the compound, but may result in change in contaminant concentration. These reactions include precipitation and dissolution, adsorption and ion exchange.

3 Numerical Methods for Contaminant Transport Modelling

Contaminant transport during GW–SW interaction is usually studied either by field experiments or by modelling approach. Field experiments involve injecting tracers into the stream area of interest, and the quantity of the tracer in the groundwater is monitored near the reach. This tracer breakthrough curve is used to infer quantitatively the GW–SW exchange along the reach. Modelling approach involves the development of conceptual model and solving them by numerical methods to assess the GW–SW exchange during interaction. This approach is discussed in detail in the following section.

3.1 Modelling Approach

The modelling of groundwater flow and contaminant transport can be broadly categorized into following steps: (1) Representing the real physical study domain with a mathematical model, i.e., the governing equations of flow and transport. (2) Defining the problem in terms of the variables to be identified (estimated). (3) Obtaining the hydrogeological parameters of the study area and source and sink terms. (4) Prescribing the boundary conditions and initial conditions and obtaining the solution (estimating the variable) by applying the suitable solution method [15].

The three-dimensional contaminant transport equation is given by [16]

$$\frac{\partial c}{\partial t} + \frac{v_i}{R}\frac{\partial c}{\partial x_i} - \frac{\partial}{\partial x_i}\left(\frac{D_{ij}}{R}\frac{\partial}{\partial x_j}\right) + \lambda c = 0 \tag{4}$$

where i, j = 1, 2, 3 for the flow in three directions; c = concentration of contaminant; D_{ij} = dispersion coefficient along i direction due to flow in j direction; v_i = velocity of flow of water along i direction; λ = radioactive decay constant of the contaminant; and R = retardation factor related to adsorption and/or chemical reaction.

The velocity in Eq. 4 is given by,

$$v_i = \frac{\partial h}{\partial x_i}\frac{K_{ii}}{\phi} \tag{5}$$

where K_{ii} = hydraulic conductivity along i direction; h = potentiometric head; and ϕ = aquifer porosity.

The head field (h) of Eq. 5 is estimated from the three-dimensional groundwater flow equation given as [16]

$$\frac{\partial}{\partial x_i}\left(K_{ii}\frac{\partial h}{\partial x_j}\right) - W = S_s\frac{\partial h}{\partial t} \tag{6}$$

where K_{ii} = hydraulic conductivity along i coordinate axes assumed to be parallel to the principal permeability directions; W = volumetric flux per unit volume (for any abstraction from the aquifer, W = sink term, and for recharge into the aquifer, W is source term); t = time; and S_s = specific storage.

Solution of Eqs. 4 and 6 is the spatiotemporal distribution of c and h which satisfy the above equations for a given set of initial and boundary conditions, source/sink terms and flow and transport parameters. Modeling of groundwater contaminant transport involves the solution of both the equations which are either solved independently or simultaneously. Solution can be obtained by analytical method or by numerical method. Though the solutions obtained by analytical methods are exact solutions, they are based on numerous assumptions which do not hold good in complex real field scenarios. On the other hand, numerical methods, though provide approximate solution, involve lesser assumptions.

3.2 *Solution Methods for Numerical Model*

Among many, the three classical choices for the numerical solution of governing equations of groundwater flow and solute transport are FDM, FEM and FVM.

3.2.1 Finite Difference Method (FDM)

FDM is the one of the oldest numerical methods for solving the partial differential equations and is based upon the application of Taylor series expansion to approximate the differential equations. In this method, the continuous system described by Eqs. 4 and 6 is replaced by a finite set of discrete spatial and temporal points. Partial derivatives in equations are replaced by difference equation obtained from the truncated Taylor series expansion. This substitution leads to the formation of systems of simultaneous linear algebraic difference equations, the solution of which yields spatiotemporal distribution of concentration and head. A discretized flow and solute transport domain is shown in Fig. 3.

Let u be the variable, then derivative of *c* can be expressed as

$$\frac{\partial c}{\partial x} \approx \frac{c_i - c_{i-1}}{\Delta x} \tag{7}$$

$$\frac{\partial c}{\partial x} \approx \frac{c_{i+1} - c_i}{\Delta x} \tag{8}$$

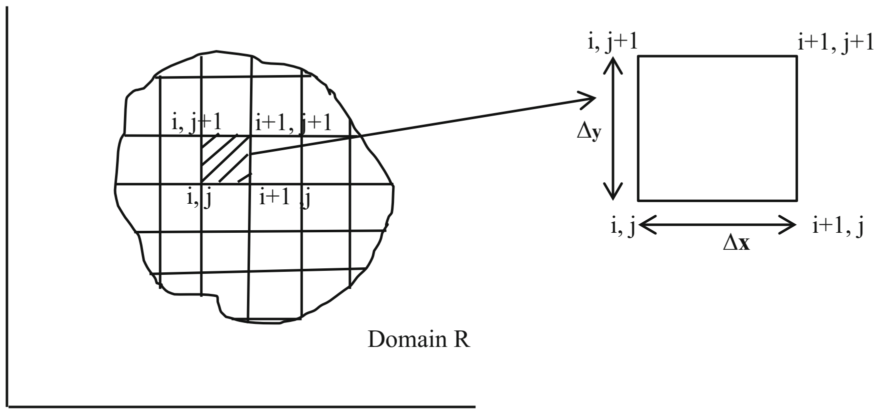

Fig. 3 Flow and solute transport domain divided into cells by a mesh of grid lines

$$\frac{\partial c}{\partial x} \approx \frac{c_{i+1} - c_{i-1}}{2\Delta x} \tag{9}$$

Equations (7)–(9) are referred to as backward, forward and centered finite difference approximations of $\partial c/\partial x$, respectively.

For example, taking a case of horizontal flow, with no induced abstraction, the governing differential equation for a confined aquifer reduces to

$$\frac{\partial^2 h}{\partial x^2} + \frac{\partial^2 h}{\partial y^2} = \frac{S}{T}\frac{\partial h}{\partial t} \tag{10}$$

Forward difference simulation for time derivative of above equation, for i, j, t = (k + 1)Δt,

$$\frac{h^k_{i,j-1} - 2h^k_{ij} + h^k_{i,j+1}}{(\Delta x)^2} + \frac{h^k_{i-1,j} - 2h^k_{ij} + h^k_{i+1,j}}{(\Delta y)^2} = \frac{S}{T}\frac{h^{k+1}_{ij} - h^k_{ij}}{\Delta t} \tag{11}$$

In above equation, there is one unknown and the equation is known as explicit scheme. Backward difference simulation for time derivative of Eq. 10, for i, j, t = (k + 1)Δt,

$$\frac{h^{k+1}_{i,j-1} - 2h^{k+1}_{ij} + h^{k+1}_{i,j+1}}{(\Delta x)^2} + \frac{h^{k+1}_{i-1,j} - 2h^{k+1}_{ij} + h^{k+1}_{i+1,j}}{(\Delta y)^2} = \frac{S}{T}\frac{h^{k+1}_{ij} - h^k_{ij}}{\Delta t} \tag{12}$$

In above equation, there are five unknowns and the equation is known as implicit scheme because the equation will have to be solved as a system of simultaneous equations for the unknowns. Although explicit scheme involves less computational time, implicit scheme is used for numerical methods because of the unconditional stability of implicit equation.

In FDM, the governing flow and transport partial differential equation (PDE) is discretized into rectangular grids, which may become cumbersome for the larger study domain with complex geometries. This led to the application of an integral form of the PDEs and subsequently led to the development of FEM.

3.2.2 Finite Element Method (FEM)

In FEM discretization process is started with the equation's integral form. Partial differential equation is transformed into an integral equation which includes derivatives of first order only. Then integration is performed numerically over elements into which considered domain is divided.

$$I = \iint F\left(x, y, U_1, U_2, \frac{\partial U_1}{\partial x}, \frac{\partial U_2}{\partial x}, \frac{\partial U_1}{\partial y}, \frac{\partial U_2}{\partial y}\right) dxdy \tag{13}$$

where x, y, U_1(x, y) and U_2(x, y) are independent variables. Objective is to minimize I or ∂I = 0.

For example, integral function for 2D steady-state flow equation in domain R can be written as

$$\text{Minimize}\, I = 0.5 \iint \left[K_{xx}\left(\frac{\partial h}{\partial x}\right)^2 + K_{yy}\left(\frac{\partial h}{\partial y}\right)^2\right] dxdy \tag{14}$$

Objective equation becomes

$$[S]\{h\} = 0 \tag{15}$$

where [S] = global matrix of coefficients which incorporates properties of porous medium and geometry and {h} = vector of unknown heads at the nodes of the element into which the entire domain is divided.

3.2.3 Meshfree Method (MM)

Above-mentioned methods are the classical methods which use the temporal and spatial discretization of domain. Latest development in the field of solving the governing groundwater flow and solute transport differential equation is the meshfree algebraic methods. In MM, predefined meshes are not required; instead, just the set of scattered nodes within the problem domain including boundaries is used. Since mesh is not created in this method, it is saves substantial time in modeling and simulation [17].

There are a number of meshfree approximation methods like moving least-squares (MLS), natural element method (NEM), reproducing kernel particle method (RKPM), smoothed particle hydrodynamics (SPH), radial basis function method (RBF), point collocation method (PCM). Features of MM include the absence of mesh, computationally inexpensive, shape functions of any desired order of continuity can be constructed, and the convergence results of MM are considerably better than the results obtained by other mesh-based shape functions [17].

The use of the type of method depends upon the requirement of the problem posed. Suppose if discontinuity has to be addressed in the solution of the governing PDE, then MM can be applied. The imposition of essential boundary conditions requires certain attention in MM and may degrade the convergence of the method. In that case, FDM or FEM can be used.

3.3 Contaminant Transport Models

Once a numerical model with a suitable solution method has been constructed, to solve the large number of set of simultaneous algebraic equations, algorithms with

Table 1 Software used for groundwater flow and solute transport modeling

S. no.	Model	Authors	Description
1	**MODFLOW** (modular finite difference groundwater flow model)	McDonald and Harbough [18]	Simulates 3D groundwater flow using cell-centered FDM
2	**MOC3D** (three-dimensional method-of-characteristics groundwater flow and transport model)	Konikow et al. [19]	Simulates 3D flow using cell-centered FDM and MOC for solute transport
3	**MT3DMS** (modular 3D multi-species transport model for simulation of advection, dispersion and chemical reactions of contaminants in groundwater systems)	Zheng et al. [20]	Simulates 3D groundwater flow using MODFLOW and solute transport by modified MOC
4	**FEMWATER** (three-dimensional finite element model of water flow through saturated–unsaturated media)	Lin et al. [21]	Simulates 3D density-dependent flow through variably saturated porous media and three-dimensional Eulerian–Lagrangian model of solute transport
5	**Random-Walk** (random-walk solute transport model for selected groundwater quality evaluations)	Prickett et al. [22]	Simulates groundwater flow using FDM and solute transport by particle-in-a-cell method (for advection) and random-walk method (for dispersion)
6	**FEFLOW** (finite element subsurface flow and transport simulation system)	Diersch [23]	Simulates 3D density-dependent saturated and unsaturated flow and solute transport using FEM
7	**STOMP** (subsurface transport over multiple phases)	Nichols et al. [24]	Coupled solution of groundwater flow and solute transport using finite difference volume approach
8	**PHAST** (program for simulating groundwater flow, solute transport and multicomponent geochemical reactions)	Parkhurst et al. [25]	Simulates multicomponent, reactive solute transport in 3D saturated groundwater flow systems using FDM

iterative procedures are required which are usually executed with the help of computer codes (or softwares) [11]. Some of the widely used computer codes are listed in Table 1.

4 Conclusion

The increasing use of groundwater for consumptive purposes has increased the risks of its vulnerability toward contamination. Among various causes of groundwater contamination, contamination from surface water sources is a major threat to the groundwater. For the effective management of groundwater sources, the understanding of transport of contaminants during GW–SW interaction is needed. For modelling the contaminant transport, along with the mechanism of transport of contaminant in porous media, a proper understanding of groundwater surface water interaction is also needed.

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