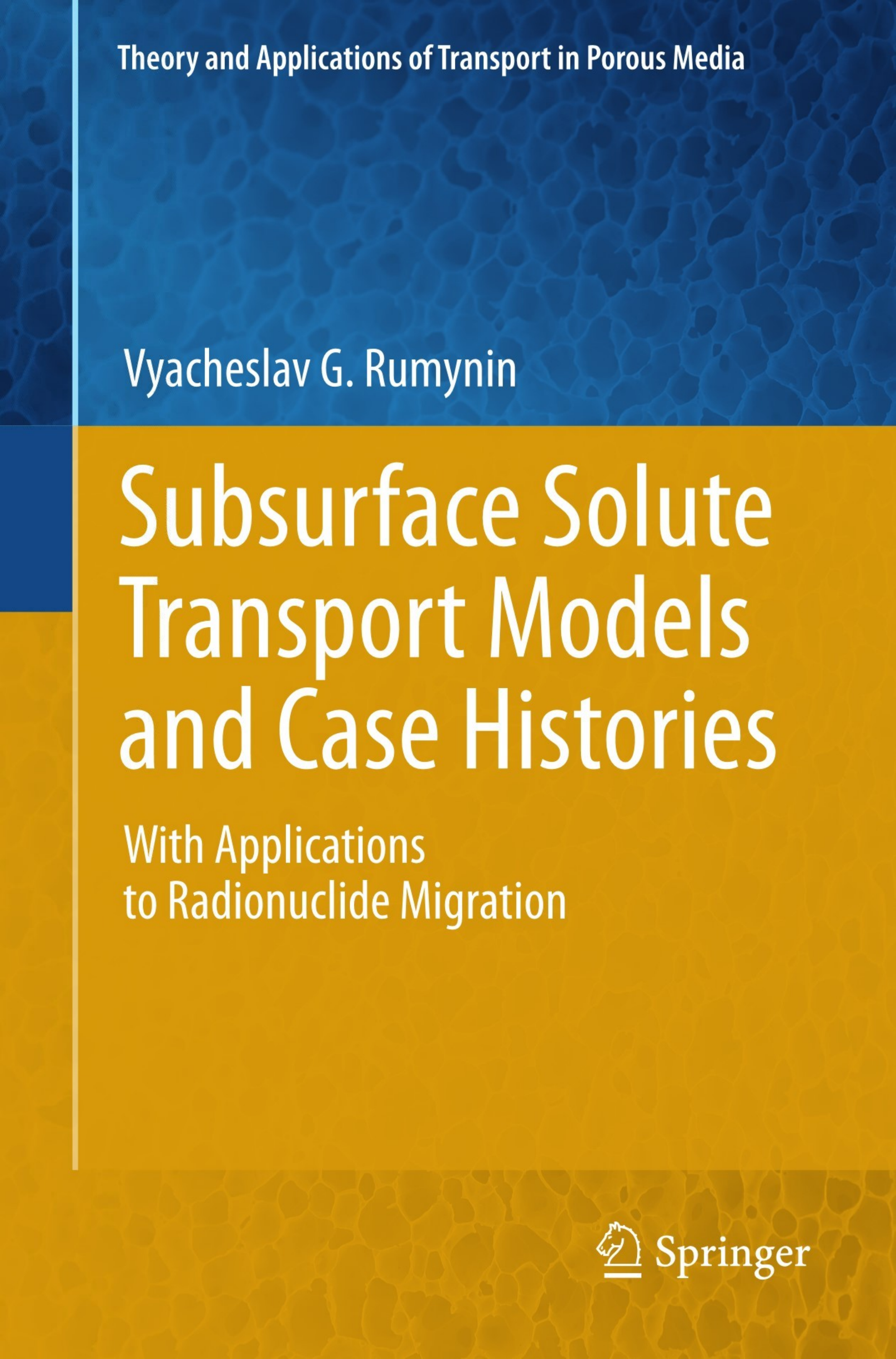

Theory and Applications of Transport in Porous Media

Vyacheslav G. Rumynin

Subsurface Solute Transport Models and Case Histories

With Applications to Radionuclide Migration

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Preface

Studies of solute fate and transport in the subsurface environment have been playing a significant role in hydrogeology over the past half century. The problem directly relates to the quality of natural water resources, which are essential to all kinds of life, and are a basic element in many sectors of human society. Most migration studies of both natural and anthropogenically derived species have considered the motion of a fluid (groundwater) accompanied by diffusion–dispersion phenomena, physicochemical interactions, as well as microbiological transformations, known to be the dominant factors providing the impact of contaminants upon groundwater supplies.

Over the last decades, essential progress in the migration process description has been achieved due to the development of mathematical background and numerical methods and laboratory and field investigations of particular transport mechanisms and physicochemical interactions. However, in many real situations, the subsurface material heterogeneity and variations in fluid properties, resulting in nonlinear contaminant plume behavior, make the prediction accuracy of the transfer processes too low to satisfy the practical needs. The lack of comprehensive field studies of solute movement is often cited as a major impediment to our understanding of solute transport in such systems.

Therefore, this work is aimed at the development of the basic knowledge of the subsurface solute transfer with a particular emphasis on field data collection and analysis coupled with modeling (analytical and numerical) tool application. The book is based mostly on field materials from author's long-standing, recent, and current experience in the study of groundwater quality related problems. The diversity of these problems is concerned with the variety of geological settings as well as the anthropogenic effects and processes caused by human activity. Some problems encountered in practice looked as challenge-like and, thus, the author was encouraged to search for new solutions and approaches. The relevant theoretical developments are concerned mainly with the formulation and solution of deterministic mass-transport equations for a wide range of engineering issues in groundwater quality assessment and forecasting that can be of some interest for bridging the gaps still existing in our knowledge of contaminant hydrogeology.

The book gives many computation examples and case studies drawn from the conducted field investigations. Those examples show the applicability of the theory and methods for solving various practical problems and making decisions in contaminant hydrology to explain the observed and to forecast the future groundwater quality. The analyzed problems are as follows:

(1) investigation and prediction of groundwater contamination by industrial contaminants and solutions (radionuclides, chloride and nitrate brine) with special focus on the effect of (a) aquifer heterogeneity, anisotropy, and dual porosity, (b) density contrast between industrial waste and groundwater, (c) physicochemical interactions that play a major role in retarding (e.g., adsorption) or enhancing (e.g., interactions between dissolved species and mobile colloids) contaminant transport;
(2) prediction of the effects of pumping on groundwater quality at wellfields: (a) the displacement of stratified initial concentration in artesian and coastal (off-shore) groundwater systems due to water pumping, (b) downward movement of mineral-weathering products in the vadoze zone (above the lowering water table) with water recharge to the producing aquifers;
(3) groundwater dating using stable and radioactive isotopes for prediction and assessment of contamination potential and the time that would be needed to displace contaminants from the groundwater system;
(4) field and laboratory tests' design and analysis, and monitoring data interpretation;
(5) partitioning of surface and subsurface flows using isotope technique;
(6) formation of evaporated salt deposits in closed surface water reservoirs having a hydraulic connection with the surrounding groundwater systems.

Several parts of the book demonstrate the potential for using numerical groundwater flow and transport models in environmental risk assessment of subsurface contamination by dense or light miscible liquid waste. Environmental isotope data were utilized for defining the groundwater systems and modeling data analysis. However, numerical modeling emerged in the book mostly as one of the primary tools used to understand the most important physical and physicochemical processes that occur in groundwater systems, as well as for getting analytical approximations for some coupled problems, which do not necessarily have exact solutions in closed analytical forms or cannot be treated with the classical methods.

One of the most essential topics addressed in the book is the migration and fate of radionuclides. Model development is motivated by field data analysis from a number of radioactively contaminated sites in the Russian Federation: near-surface radioactive waste (RW) disposal sites in northwestern Russia and the Southern Urals, and two deep-well RW injection sites in Western Siberia. These sites are part of huge nuclear industry enterprises licensed to possess radioactive materials and also involved in hazardous-waste operations, which are supervised by RosAtom, the State Nuclear Energy Corporation, Russian Federation.

The total activity of radionuclides that were released (accidentally or intentionally) in aquifers at many sites reaches hundred thousands to hundred millions Ci. Any of the three RW disposal sites out of the four mentioned here (located in Southern Urals and Western Siberia) probably contains more radioactive contamination in the subsurface than any other site in the world. Additionally, detailed information on physical, mechanical, and solute transfer properties of clay formation (which is considered as a host medium for the engineered underground RW repository in the northwestern part of the Russian Federation) is also analyzed.

Those sites play a unique role in the advancement of knowledge of the subsurface behavior and fate of many hazardous radionuclides and can be considered as field-scale laboratories. The book is focused on the modeling and analytical assessments of a range of physical and chemical processes and interactions of concern. Some of the key issues needed to be addressed included:

(1) study of the behavior of a broad spectrum of radionuclides (fission products and actinides) in waste (with low content of dissolved solids and brine) based on long-term (up to 50 years) monitoring data in shallow and deep aquifer systems;
(2) study of the spatial variability of migration properties of aquifer materials and clayey semipervious formations;
(3) assessment of the role of brine-induced advection in redistribution of radioactive components at waste disposal sites;
(4) study of adsorption hysteresis implying isotherm nonsingularity and other non-ideal sorption phenomena, as well as the assessment of their role in natural attenuation of radioactively contaminated sites;
(5) analysis of transient hydrogeochemical-barrier effects, facilitating radionuclide transport, and some other mechanisms responsible for "fast" radionuclide transport in aquifers;
(6) experimental evidence for colloid-facilitated radionuclide (actinide) transport, and mathematical description of the phenomena.

The model developments were accompanied by laboratory studies into natural attenuation, radionuclide adsorption and desorption kinetics and equilibrium (including when colloidal particles are involved). Batch tests were conducted with different radioactive solutions under different temperature and pressure conditions. Anomalous behavior of radionuclides was observed and modeled.

This study can be regarded as the continuation of a series of works started by the author in the 1970s in cooperation with the outstanding Russian scientist, hydrogeologist, V.A. Mironenko, whose contribution to the development of several lines of studies in hydrogeology and hydrogeomechanics is difficult to overestimate. At the same time, this book could not appeared were it not for the all-round support from colleagues – researchers from E.M. Sergeev Institute of Environmental Geology, St. Petersburg Division, RAS, and St. Petersburg State University, who rendered assistance in the preparation of parts of the book. In this connection, the author very much appreciates the help of Leonid Sindalovsky in implementation of many numerical algorithms considered in the book, the contribution of Pavel Konosavsky to the joint studies of adsorption hysteresis and the development of some models of solute transfer in the porous media under disturbed flow conditions. The author also appreciates Igor Tokarev's willingness to share his data on regional isotope study of a groundwater system in the area of RW disposal at Tomsk-7 site.

The study discusses experiments carried out in laboratories of A.N. Frumkin Institute of Physical Chemistry and Electrochemistry, RAS, and A.P. Alexandrov Technical Institute under supervision of Drs. Elena Zakharova, Elena Kaimin, and Elena Pankina. The author expresses his sincere gratitude to these groups for cooperation that have yielded new results.

The author appreciates the cooperation of Aretech Solutions and TIHGSA Enterprises allowing him to learn new hydrogeological aspects related to the formation of groundwater resources and quality in arid regions.

The author also much appreciates the attention to his work and fruitfull discussions with Profs. Vsevolod Shestakov and Sergey Pozdniakov, Moscow State University, and Dr. Andrei Zubkov, the head of the Environmental Protection Division (Siberian Chemical Plant), and many other brilliant experts–hydrogeologists, whose talent and enthusiasm in scientific and production work allows the author to believe in the future of the Russian hydrogeological school.

Many efforts were made by Dr. Chin-Fu Tsang and Prof. Jacob Bear to organize this work in a proper way in order to prepare the book in a format acceptable for the international publishing company, Springer. Discussions and exchange of information, ideas, and opinions with them was a great support to this work.

Finally, the author very much appreciates the help of Dr. Gennady Krichevets in professional translation of the book and many useful comments from him allowing the author to make certain improvements to the book. The author would also like to acknowledge the help of Ekaterina Kaplan for her editorial assistance and technical support of the work.

Thus, the book, along with theoretical findings, contains field information, which will facilitate the understanding of subsurface solute transport and the development of a methodology for practical application to groundwater hydrology. This book addresses scientists and engineers who are interested in the quantitative approach to studying groundwater migration processes. The book can also be profitably read by students.

December 28, 2010 Vyacheslav G. Rumynin

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Part I

The Essentials of Dissolved Species Transport in the Subsurface Environment: Basic Definitions, Fundamental Mechanisms and Mathematical Formulation

The theory of solute migration in groundwater originates from studies devoted to the description of elementary pore-scale mechanisms (processes) of the movement of dissolved species in a single fluid phase, including advection, molecular diffusion, and hydrodynamic dispersion, which are accompanied by acts of simplest sorption-type physicochemical interactions and solute decay (destruction) reactions. Those studies were mostly based on the classical theory of fluid motion in idealized porous media (Muskat 1937; Scheidegger 1957), as well as on chemical kinetics and reaction engineering. However, it has become clear that there exist some specific features in the application of conventional hydrodynamics methods to the formulation, solution, and analysis of many practically significant hydrogeological problems. In particular, the relative significance of those mechanisms and interactions in the general migration process was soon found to depend on the spatial and temporal scale of their analysis, the lithological and genetic type of geological sections, and the spatial correlation structure of their physical parameters, the structure of water flows, and the conditions on their inner and outer boundaries. This, as well as the specific features of the application of physico-mathematical apparatus to the solution of appropriate boundary problems regarding dissolved species transport in single-phase constant-density groundwater flows, will be the focus of the first part of this book.

Mathematical models used to describe solute transport in the unsaturated zone of the subsurface are also included in this part of the book. As will be shown, for accurate prediction of contaminant transport through the unsaturated zone, field equations for transport of moisture and chemicals must be coupled.

The equations given here represent a deterministic approach to describing the subsurface transport phenomena, and have been assembled from a considerable collection of previous works and investigations conducted by many recognized authorities in the field of subsurface fluid dynamics. More generalized fluid flow and transport models, accounting for the stochastic nature of aquifers and soil materials are subject of high profile, well-publicized special investigations.

The proposed material forms a bridge to the understanding of solute transport under near-natural conditions and the analysis of migration of complex-composition solutions (liquids) whose properties differ from those of formation waters. Besides, the approaches developed here will be used to assess the contributions of various physicochemical processes, which in many cases control the potential of anthropogenic impact on groundwater quality under natural conditions.

Chapter 1
Advection and Dispersion of Dissolved Species in Aquifers

The transfer of chemical components that, when in solutions, have no effect on the physical properties of aquifer materials and groundwater, is inseparable from the groundwater flow. Their advective transport involves micro- and macrodispersion processes, which control the extent of solute dispersion in homogeneous and heterogeneous aquifers. In this chapter, we will consider the migration models that describe the motion of solutions miscible with groundwater in homogeneous aquifers. The solute migration processes in heterogeneous (stratified and fractured-porous) systems will be discussed in separate chapters.

1.1 Governing Equations and Solute Transport Parameters

The traditional description of flow and solute transport in natural porous and fractured media (soils, sediments, rocks), as well as the solution of the majority of subsurface hydrology problems, are based on the continuum mechanics approach (Bear 1972; Shestakov 1995; Bear and Cheng 2010). The continuum assumption considers the fluid and solid phase as a continuous medium with flow and solute (mass) transport parameters taken to be well-defined at the REV (representative elementary volume) scale. The appropriate level of statistical averaging of medium properties is a priori attained in complexes of weakly lithified porous sediments. The construction of solute migration models in fractured rocks requires the validity of medium's continuity assumption, REV, to be analyzed more thoroughly and the possible scale effects associated with the "structuring" of groundwater flows in fracture space to be taken into account in the models (Schwartz et al. 1983; Berkowitz 2002; Kosakowski 2004; Neuman 2005; Reeves et al. 2008a, b). Effective (or equivalent) continuum models utilizing REV approach may not be applicable for real fractured rocks (see below).

The mathematical formalization of the subsurface migration is based on the groundwater flow continuity equation (which is equation of conservation of mass), Darcy's law and the solute transfer equation together with an appropriate set of boundary and/or initial conditions. This section is devoted to analysis of such models.

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1.1.1 Advection of Conservative Components in Porous and Fractured Media

Advection of particles or heat, from the classical fluid-dynamical point of view, is the transfer of matter or heat by the flow of a fluid. Advection due to the fluid's bulk motion in pores or fractures is among the major mechanisms governing solute transport in aquifers lying in the hydrodynamic zone of active water exchange. The driving force for advection is the gradient in the hydraulic head.

In unconsolidated granular porous media, this process proceeds in practically nonstructured void space; whereas liquid motion in fractured rocks, consisting of an assemblage of intact rock blocks (matrix) separated by intersecting sets of joints, proceeds in the space which generally has a distinct structure. In some cases (primarily, when the process is considered at a local scale), these distinctions require differentiation of the computation schemes (models) used to describe solute migration in two types of rock formations with different nature of void space.

1.1.1.1 Flow Field and Actual Fluid Velocity

In subsurface fluid dynamics, flow velocity field, creating potential for advection of dissolved solutes is a vector field. This field can be mathematically described by a continuity equation written in the most general form for compressible pore-fluid mixtures as follows

$$\frac{\partial \phi \rho}{\partial t} + \nabla \cdot (\rho \mathbf{q}) = 0, \tag{1.1}$$

where $\mathbf{q}$ is the specific discharge or *Darcy velocity* (a vector with 3 components) [LT^{-1}],

$$\mathbf{q} = -\frac{\mathbf{K}}{\mu}(\nabla P - \rho \mathbf{g}); \tag{1.2}$$

ρ is the liquid density [ML^{-3}]; ϕ is the porosity defined as the void space between grains (in porous-type formations) or fracture walls (in consolidated rocks) filled with water [$\mathrm{L}^3\mathrm{L}^{-3}$]; P is the hydraulic pressure; $\mathbf{K}$ is the permeability [L^2] (second-order tensor with 9 components, three of which are K_x, K_y, K_z); μ is the dynamic viscosity [$\mathrm{ML}^{-1}\mathrm{T}^{-1}$]; $\mathbf{g}$ is the gravity vector [LT^{-2}]. The term $\nabla \cdot (\rho \mathbf{q})$ is called the divergence of fluid flux, representing the net fluid influx/efflux through the element and sometimes is written as $\mathrm{div}(\rho q)$. Equation 1.1 does not include the inflow/outflow source-terms.

The first chapters of this book deal with calculating the motion of components whose concentration C has no effect on the density ($\rho = \mathrm{const}$) and viscosity ($\mu = \mathrm{const}$) of liquid in the pores (fractures). The possible initial ($t = 0$) variations of groundwater density are also neglected. The pores are assumed to be filled with water alone: no other liquids or gas phase are present. In such case, Eqs. 1.1 and 1.2 can be rewritten in terms of hydraulic head, h (Bear 1972, p. 207),

$$S_s \frac{\partial h}{\partial t} + \nabla \cdot \mathbf{q} = 0, \tag{1.3}$$

$$\mathbf{q} = -\mathbf{k}\nabla h, \tag{1.4}$$

where S_s is the specific storage of the porous medium [L^{-1}], which is the volume of water, dV_w, that a volume of an aquifer, dW_a, releases from storage under a unit decline in hydraulic head, dh; $\mathbf{k}$ is the hydraulic conductivity [LT^{-1}]. Hydraulic heads provide a measure of the total mechanical fluid potential, and Eq. 1.3, formulated on the principle of conservation of fluid volume, conserves fluid mass.

To determine the specific discharge the gradient-based Darcy law can be written in the more convenient, indicial notation, form which is valid for the general case of groundwater flow in an anisotropic medium

$$q_i = -k_{ij} \frac{\partial h}{\partial x_j}, \tag{1.4a}$$

where q_i are the components of specific discharge [LT^{-1}], h is the hydraulic head [L], $k_{ij} (i = 1,2,3, j = 1,2,3)$ are the components of symmetrical matrix (tensor) of hydraulic conductivity [LT^{-1}]; coefficient k_{ij} is connected with the permeability, K_{ij}, by the relationship $k_{ij} = K_{ij}\rho g/\mu$. Formula (1.4a) implies the assumption that summation over the same indices is carried out. In the general three-dimensional case $\partial/\partial x_j \equiv \nabla = \partial/\partial x + \partial/\partial y + \partial/\partial z$ is the *Cartesian* coordinate system, ∇ is gradient operator ($\equiv$ grad), sometimes referred to as *Hamiltonian* operator.

Now dissolved passive species will move with the same velocity as water particles (average water velocity) u_i ($\mathbf{u}$)

$$\left.\frac{\partial x_i}{\partial t}\right|_C = u_i = \frac{q_i}{\phi}, \text{ or } \left.\frac{\partial \mathbf{x}}{\partial t}\right|_C = \mathbf{u} = \frac{\mathbf{q}}{\phi}. \tag{1.5}$$

Equation (1.5) specify relationship between the specific discharge, which is used to determine the volumes of fluid passing through given surfaces, and *the actual* (*advective*) *fluid velocity* controlling the front of solute movement in the porous space. Actual fluid velocity varies over the pore space, due to the connectivity and geometric complexity of that space. This variable velocity can be characterized by its mean or average value. The average fluid velocity depends on what part of the cross-section area is made up of pores, and to what extent the pore space is connected. Therefore ϕ is the *effective porosity* (*fracture porosity* in fractured rocks) also called kinematic, advective and open porosity.

Taking C as the volumetric concentration of a chemical component (ML^{-3}, M is the amount of the species), the advective flux ($\mathrm{ML}^{-2}\mathrm{T}^{-1}$) can be expressed in terms of specific discharge (q_i):

$$J_i^a = q_i C = \phi\, u_i C, \text{ or } \mathbf{J}^a = \mathbf{q}C = \phi\,\mathbf{u}C. \tag{1.6}$$

Here, J_i^a ($\mathbf{J}^a$) is the mass of a component carried across a unit area, oriented normal to i direction, per unit time. Direction $C\mathbf{q}$ coincides with the direction of fluid motion.

In the cases where solute transport causes the appearance of density gradients or where such gradients originally exist in groundwater systems, the use of hydraulic head as only dependent variable in the analysis neglecting buoyancy component of the flow-driving force is not acceptable (Bachu and Michael 2002; Post et al. 2007). Darcy's specific discharge in such systems should be expressed in terms of a pressure function with allowance made for the space and time variations in the physical characteristics ρ and μ (1.2). Thus, a fluid pressure-based formulation is generally preferable in modeling variable density problems. Such problems, which belong to the class of coupled problems, are considered in the following parts of this book (Chaps. 12–18). The coupling of flow and transport phenomena is caused by the dependence of the water density on the salt concentration.

Strictly speaking, the use of relationship (1.5) implies that the scales and dimensions of the flow and solute migration problems are consistent. Thus, the effective value of the hydraulic conductivity, derived from pumping tests of heterogeneous aquifers, reflects the three-dimensional flow conditions. This value is always greater than the hydraulic conductivity, which governs the migration of components under constrained conditions of a one-dimensional or two-dimensional groundwater flow (Rovey and Niemann 2005). This fact follows from the analysis of basic stochastic models (Gelhar 1993; Neuman 1994), demonstrating the effect of the groundwater flow dimensions on the effective hydraulic conductivity. The ratio of calculated to actual migration velocity values can be as large as two or three, meaning that the rate of aquifer pollution will be considerably overestimated.

1.1.1.2 Effective Porosity (Fracture Porosity)

The characteristic values of active porosity for loose (not cemented, sandy) sediments commonly varies within a relatively narrow range ($\phi \approx 0.2 - 0.4$). In sand type of sediments that have not experienced cementation, the value of ϕ is commonly near the total porosity value ϕ^0. Silt, loam and clay types of sediments also feature sufficiently high ϕ values ($\phi \approx 0.3 - 0.45$). However, in argillite-like clays, where molecular diffusion dominates, a considerable portion of voids ($\phi^0 - \phi$) is inaccessible for dissolved species (Sect. 1.1.2.1). This is due to the presence of cement "walls" in the pore space. The confirmation is the radical difference between molecular diffusion coefficients obtained in experiments with undisturbed rock samples and with packing clay prepared from the same samples (García-Gutiérrez et al. 2006). Moreover, the diffusion-accessible porosity depends on the type of the migrating ion (Huysmans and Dassargues 2006).

In fractured crystalline rocks and hard sedimentary rocks such as sandstone, limestone and chalk, conceptualized as nonuniform continua with bulk properties, the scatter of parameter ϕ is much wider, while its absolute values are much less (the average interval is 0.005–0.03). Whence, it follows that the macroscopic transport in fractured rocks, all other conditions being the same, should be much faster than in porous ones.

Clearly, the errors in advection velocity estimates in porous sedimentary deposits are primarily determined by errors in the description of permeability field and the structure of groundwater flows. Variations in the porosity, ϕ, are less significant than the space variations in sediment permeability (hydraulic conductivity), so expert estimates of porosity can be used in some cases, while the hydraulic conductivity no doubt requires detailed experimental studies. Conversely, as it can be seen later, the values of ϕ in fractured type of rock formations are hardly predictable at the intuitive level. Therefore, we have to accept the fact that the results of predicting groundwater pollution in fractured rock complexes, because of their heterogeneity and anisotropism in terms of permeability and advective porosity, are often unreliable.

1.1.1.3 Anisotropy of Sediment and Rock Properties

The form of Darcy's law (1.4a) corresponds to the general case of fluid flow in an anisotropic medium. The hydraulic anisotropy of a bed is the governing factor in the advection in heavily deformed groundwater flow that form, for example, due to concentrated water withdrawal or when density advection develops in the aquifer. Of major importance in sedimentary (porous type) complexes is the anisotropy of permeability in the profile, which is due to the lithologic and facies variability. The anisotropy in fractured-rock complexes is mostly due to the presence of several systems of subvertical fractures (planar anisotropy) and the existence of bedding joints (profile anisotropy). Large tectonic fractures with distinct unidirectional orientation in a medium with primary lithogenetic jointing are most often responsible for planar anisotropy. Therefore, one of the principal anisotropy directions commonly lies in the plane of the water-bearing bed (aquifer), while the other is perpendicular to it.

1.1.1.4 On the Microstructure of Flows in Porous and Fractured Rocks

Active porosity, ϕ, for *unconsolidated or weakly consolidated* (sand–clay) *sediments* is a conventional characteristic, since, in addition to flow-through (active) zones, there always exist stagnant zones not involved in the flow but still playing a considerable role in the formation of the general mass flow (Coats and Smith 1964; van Genuchten and Wierenga 1976; Rose 1977; Golubev 1981): by accumulating the dissolved species via molecular diffusion, such dead-end zones enhance the overall "salt-related" capacity of the system as compared to the active porosity. Therefore, more appropriate characteristic for long-term forecasts would be the value of the total "connected" porosity of rocks (ϕ^0).

Taking into account the interaction between individual elements of flow-bearing media is of fundamental importance for *fractured rocks* (Tsang et al. 1991; Gelhar 1993; Berkowitz 2002; Park et al. 2003; Kosakowski 2004;