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A review of model applications for structured soils: a) Water flow and tracer transport

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A B S T R A C T

Although it has many positive effects, soil structure may adversely affect the filtering function of the vadose zone that protects natural water resources from various sources of pollution. Physically based models have been developed to analyze the impacts of preferential water flow (PF) and physical non-equilibrium (PNE) solute transport on soil and water resources. This review compiles results published over the past decade on the application of such models for simulating PF and PNE non-reactive tracer transport for scales ranging from the soil column to the catchment area. Recent progress has been made in characterizing the hydraulically relevant soil structures, dynamic flow conditions, inverse parameter and uncertainty estimations, independent model parameterizations, stochastic descriptions of soil heterogeneity, and 2D or 3D extensions of PNE models. Two-region models are most widely used across all scales; as a stand-alone approach to be used up to the field scale, or as a component of distributed, larger scale models. Studies at all scales suggest that inverse identification of parameters related to PF is generally not possible based on a hydrograph alone. Information on flux-averaged and spatially distributed local resident concentrations is jointly required for quantifying PNE transport. At the column and soil profile scale, model predictions of PF are becoming increasingly realistic through the implementation of the 3D soil structure as derived from hydrogeophysical and tracer techniques. At the field scale, integrating effects of the soil structure and its spatial variability has been attempted by combining 1D PNE approaches with stochastic parameter sampling. At the catchment area scale, the scarcity of data makes validation of PF related model components a task yet to be accomplished. The quest for easily measurable proxy variables, as 'the missing link' between soil structure and model parameters, continues in order to improve the practical predictive capability of PF–PNE models. A follow-up paper complementing this manuscript reviews model applications involving non-equilibrium transport of pesticides, as representatives of reactive solutes.

Abbreviations: 1D, one-dimensional; 2D, two-dimensional; 3D, three-dimensional; BC, boundary conditions; BTC, break through curve; CD, convection–dispersion; CDE, convection–dispersion equation; CLT, convective lognormal transfer function; CT, computer tomography; CV, coefficient of variation; DEM, digital elevation model; DFM, discrete fracture model; DPM, dual-permeability model; DP-MIM, dual permeability model with mobile and immobile zones; GIS, geographic information system; IC, initial conditions; KDW, kinematic dispersive wave; KW, kinematic wave; MIM, mobile–immobile model; MRI, magnetic resonance tomography and imaging; PC, personal computer; PF, preferential flow; PNE, physical non-equilibrium; SPM, single-permeability model; STM, stream tube model.

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1. Introduction

The phenomenon of non-equilibrium preferential flow (PF) and transport in fractures, macropores or other local high-permeability zones, is encountered in various environmental systems. PF has received increasing attention in different fields, such as hydrogeology, petroleum engineering, (soil) hydrology, and waste disposal sites construction. For instance, aquifer systems in heavily fractured rock, or in limestone with dissolution conduits, are often vulnerable to contaminants and need special protection when used as sources for drinking water (Birk et al., 2006). The remediation of heterogeneous sedimentary aquifer systems contaminated by organic chemicals at former military or industrial sites is an intricate, long-lasting, and costly task if the contaminants are distributed across layers with contrasting hydraulic and chemical properties. Prolonged tailing of concentrations has been encountered due to the slow release of residual contaminants from fine textured and organic (e.g., lignite) layers to coarse textured layers (Grathwohl et al., 2004). For similar reasons, a considerable effort is required to extract residual amounts of oil from subsurface petroleum reservoirs in fractured rock or heterogeneous sediment systems (Al-Huthali and Datta-Gupta, 2004). PF processes in the vadose zone of fractured rocks have also been intensely studied (Dasgupta et al., 2006a), e.g., in the framework of assessing the suitability of potential nuclear waste repositories (Wu and Pruess, 2000). In the agricultural environment, PF and agrochemical transport in structured soils may significantly affect the growth of crops (Kramers et al., 2005) and water resources (Sinkevich et al., 2005).

A review on PNE processes, including model applications, was presented a decade ago by Jarvis (1998). Overviews of the relevant processes and conditions triggering PF in soils are discussed in the classic paper by Beven and Germann (1982) and in the recent review by Jarvis (2007) which covers the state-of-the-art in our understanding of PF and (agro-) chemical transport, and the effects of soil and crop management practices. The latter is also treated in another recent review (Ma et al., 2007). Finally, the value of PF to ecosystem services was assessed with a global perspective by Clothier et al. (2008).

Researchers have attempted to translate process information gained from experimental observations into conceptual theories and mathematical models that are 'physically-based' or 'mechanistic'. The governing equations are implemented in computer simulation tools, which can then be tested, modified, and/or (in)validated against new experimental data. Increasingly sophisticated models have been developed to analyze PF in various environmental systems, where there exist considerable differences in spatial and temporal scales, type of flowing liquids (e.g., water, brine, oil) and transported agents (e.g., dissolved chemicals, NAPLs, colloids, particles, microbes). Given the limited scope of this review, the focus is on vadose zone models developed to assess the impact of human activities on water and solute balances in the agrosphere.

The objective of this paper is to give an overview of recent (past decade) developments in the application of physically-based models describing preferential water flow and solute transport through structured agricultural soils. The PF model concepts are only briefly explained here. For details, the reader is referred to recent review articles by Šimůnek et al. (2003), Gerke (2006), Jarvis (2007), and Šimůnek and van Genuchten (2008). Here, the emphasis is on the *applications* of such models in describing *experimental* observations. This paper reviews model applications to conditions involving PF and non-equilibrium transport of conservative solutes at different spatial scales. First (Section 2), an overview of model concepts and corresponding computer models is given. Then, model applications concerning preferential water flow (Section 3) and solute transport (Section 4) are reported, followed by (Section 5) a discussion of two-region approaches and (Section 6) methods of independent parameter determination. Subsections within Sections 2, 3, 4, and 6 are organized with regard to increasing spatial scale.

Modelling of fingering flow (e.g., De Rooij, 2000; Wang et al., 2004), funneling flow (e.g., Ju and Kung, 1997), and reactive, colloid or particle-facilitated solute transport are excluded from this contribution. Model applications involving transport of pesticides, as representatives for reactive solutes, will be reported in a companion review article.

2. Overview of model approaches

2.1. Conceptual models

Classical physically based model concepts for water flow and solute transport in structured soils can be broadly classified into continuum, bi- or multi-continuum, and network approaches (Fig. 1).

The traditional conceptualization of soil is that of a porous medium with continuous properties ('Continuum', Fig. 1a). Variably saturated water flow through such a porous system is uniform and at local equilibrium, and is often described using the Richards equation. Bimodal or multimodal hydraulic functions (e.g., Othmer et al., 1991; Wilson et al., 1992; Ross and Smettem, 1993; Durner, 1994; Mohanty et al., 1997) are an indirect way of representing soil structure in such a uniform flow approach. A one-domain PNE concept of infiltration (Fig. 1b) was realized by assuming a rate-limited equilibration between the soil water content and the pressure head in the Richards equation (Ross and Smettem, 2000).

Other conceptualizations of PNE regard the soil as being composed of two or more regions (domains) (Fig. 1c–f). The stream tube model (STM), or stochastic convective model, assumes displacement of tracer at (usually) steady-state flow through independent flow tubes (Fig. 1c); it can be used with a convective lognormal transfer function (CLT) (Jury and Roth, 1990).

Two-region models of soil conceptualize structural pores as the fast flow region, whereas the soil matrix has a lower or zero saturated hydraulic conductivity. Multi-region approaches consider additional regions — up to three so far in model applications. Domains are not geometrically separated, but are conceptually superimposed over the same soil volume. Common to these approaches are zero-dimensional, usually first-order, transfer terms for lateral exchange of water and/or solute between the regions.

The mobile–immobile model (MIM) (Fig. 1d) assumes that the soil micropore network is poorly connected and of such a low permeability that water is immobile in the vertical direction. The MIM approach was originally developed for simulation of fluid displacement in fractured petroleum reservoirs (Warren and Root, 1963; Coats and Smith, 1964).

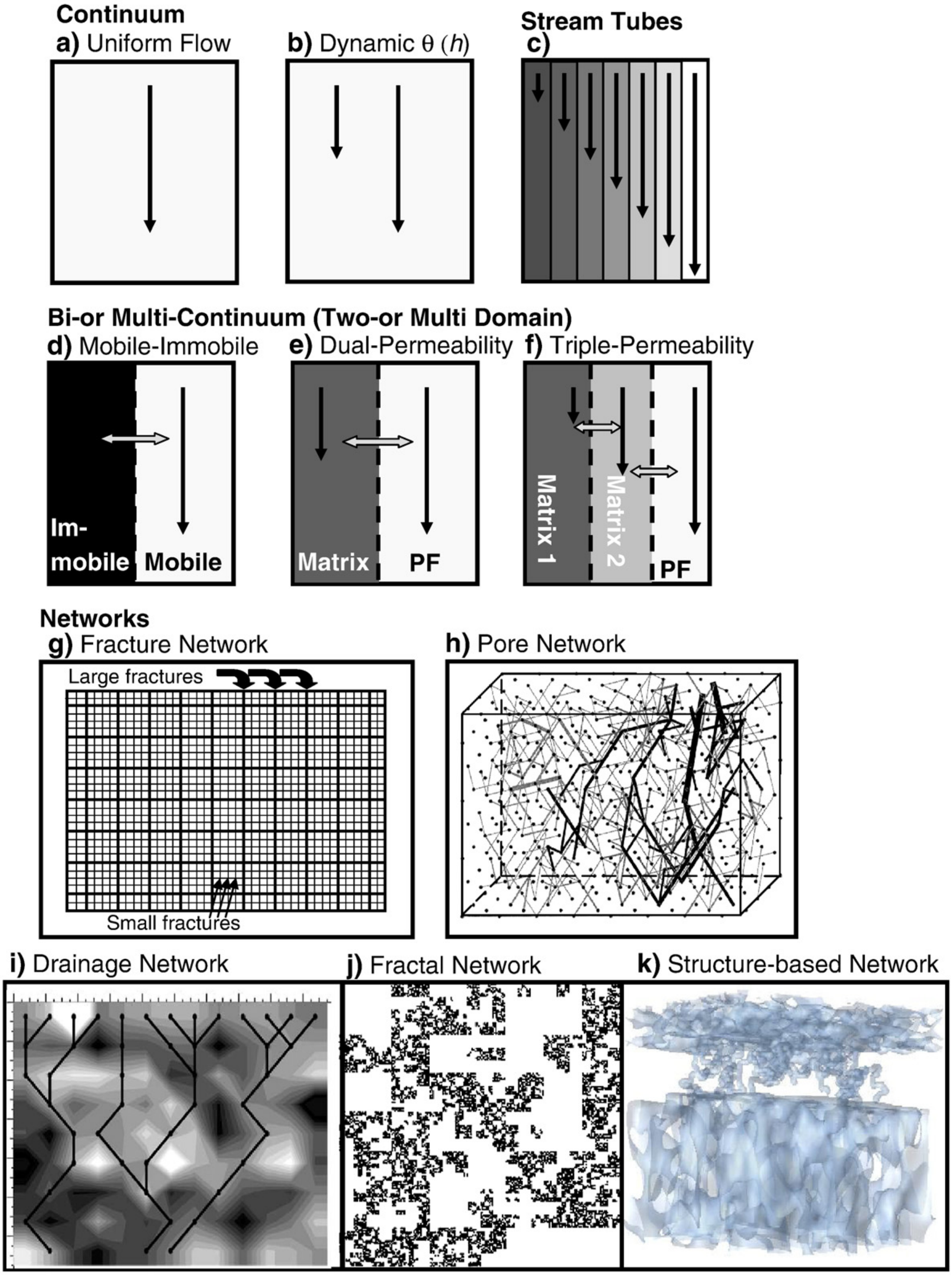

Fig. 1. Conceptual models of water flow and solute transport in structured soils. Model dimensions may vary, e.g. 1D, 2D or 3D (a.–f.), 2D or 3D (g.–j.), 3D (k.). Sources: g. modified after Graf and Therrien (2006), h. modified after Vogel (2002), i. modified after Deurer et al. (2003), j. Anderson et al. (2000), reprinted with permission from the Soil Science Society of America, k. Vogel et al. (2006), reprinted with permission of the authors.

The approach was later adapted for structured soils (van Genuchten and Wierenga, 1976; Gaudet et al., 1977).

The dual-permeability model (DPM) (Fig. 1e) assumes that the porous medium consists of two overlapping pore domains, with water flowing relatively fast in one domain (often called the macropore, fracture, or interporosity domain) and slow in the other domain (often referred to as the micropore, matrix, or intra-porosity domain). The DPM approach was proposed initially for studying fractured rock reservoirs in hydrogeology and petroleum engineering (Barenblatt et al., 1960; Duguid and Lee, 1977).

The multi-region model (Fig. 1f) comprises three flow regions (e.g., matrix, primary and secondary fractures) with two transfer terms (Gwo et al., 1995; Hutson and Wagenet, 1995). The same concepts shown in Fig. 1a–e were applied, or were combined in different pairs, to describe water and solute movement (see e.g. Šimůnek et al., 2003).

The above approaches can be further classified regarding their scope. The first group comprises continuum and bi-continuum models which consider flow in the entire soil as being controlled by both capillary and gravity forces ('capillary PF models'). Bi-continuum models assume that the Richards equation can be used also for the network of structural pores. Flow in the PF system can be directed upwards during periods of evapotranspiration.

The second group includes two-domain models which assume that flow in the PF domain is controlled by gravity only and is always directed downwards ('gravity-driven PF models'). PF is often described by the Kinematic Wave (KW) equation. Models of this group are tailored to soils with macropores or heavy clay soils with distinct shrinkage cracks.

A third group comprises simplified descriptions of macropore flow, such as capacity ("tipping bucket") or routing approaches ('empirical PF models'). The capacity approach assumes zero downward flow from each layer until filled to field capacity. The routing approach assumes instantaneous bypass flow.

While the above concepts were often implemented in agricultural system models, other concepts were more rarely applied, and for research only (Fig. 1c, g–k).

The 'network' conceptual models (Fig. 1g–k) here comprise three entirely different approaches. Discrete fracture models (DFM, Fig. 1g) were developed for describing flow and transport in discrete fractures and in the matrix of fractured rock (e.g., Wang and Narasimhan, 1985). Exchange of water and solute between the fracture and matrix domains is calculated as advection–diffusion process without a need to resort to the first-order approximation. However, a dense spacing of nodes in the numerical grid is required near fractures, which triggered the development of advanced numerical techniques for adaptive grid refinement (Slough et al., 1999; Li et al., 2005) and time discretization (Park et al., 2008). A fundamentally different, since microscopic, approach is the pore network model (Fig. 1h), which calculates Poiseulle flow through the void soil pores arranged in a 2D or 3D geometric network. Pore network models have demonstrated potential for prediction of hydraulic properties of structured soil (e.g., Vogel and Roth, 2001, 2003). Solute transport calculations are based on either particle tracking or local advection and diffusion in the pore network. However, calculations are still limited to steady-state flow conditions at sample dimensions of 10^{-1} m. The drainage network (Fig. 1i) concept (Deurer et al., 2003) identifies flow path networks by spatially linking the scaling factors of water retention curves. Fractal (Fig. 1j) and structure-based (Fig. 1k) concepts are briefly described later with their applications (Sections 3.2.1 and 4.2.3).

For applications at larger scales, distributed models (i.e., models that account for spatial variations in variables and parameters) were developed (Fig. 2). Those models can be further subdivided into (here so-called) hydrological unit models (Fig. 2a), which are discretized into sub-catchments, or smaller units, based on similar physical, geomorphological and hydraulics characteristics. In these units, the vadose zone is represented by 1D or 2D continuum or two-domain submodels. Other distributed approaches considered here are the 3D surface/vadose zone/groundwater models (Fig. 2b) with 3D subsurface discretization and a PF description for the vadose zone. Ideally, all submodels are fully coupled by appropriate boundary conditions. Examples of computer models (Table 1) based on the above concepts are described in the following section, with focus on the PF components.

2.2. Computer models

2.2.1. Capillary PF models

The PF representations in the models below are closely related to the concept and equations of the DPM of Gerke and van Genuchten (1993). Thus, the following model descriptions can focus on deviations and modifications.

DUAL (Gerke and van Genuchten, 1993) is a 1D-DPM based on two Richards and convection dispersion equations for matrix and fracture pore systems, coupled by first-order terms for bi-directional exchange of water and solute. Water transfer is driven by the pressure head difference between the flow domains, while solute transfer proceeds by advection and by diffusion driven by the inter-domain concentration difference. The original research model was adapted for application to field conditions (Gerke and Köhne, 2004). DPOR-1D (Lewandowska et al., 2004) employs a single integrodifferential mass balance equation that includes an integral term for water exchange between matrix and PF subdomains. Despite an increased accuracy, fewer BCs and parameters need to be defined than in DUAL. In MURF and MURT (Gwo et al., 1995, 1996), (vertical) MUlti-Region Flow and Transport models, three overlapping pore regions with different soil hydraulic properties are considered. First-order water and solute exchange occurs between regions arranged 'in series'. TRANSMIT (Hutson and Wagenet, 1995) is another similar three-region 1D model. HYDRUS-1D (Šimůnek et al., 1998, 2003, 2005, 2008a,b,c; Šimůnek and van Genuchten, 2008) describes water, heat, and solute movement in the vadose zone. The latest version includes a variety of SPM, MIM, and DPM approaches; the more frequently used MIM and DPM are again based on DUAL (Gerke and van Genuchten, 1993). Inverse parameter optimization is implemented using the Marquardt–Levenberg method which can accommodate different types of data in the objective function.

DPOR-2D (Szymkiewicz et al., 2008) is the 2D equivalent of DPOR-1D. 2DCHEM (Nieber and Misra, 1995) is a 2D DPM for application in drained soils. S_2D_DUAL (Vogel et al., 2000) may additionally consider spatially distributed

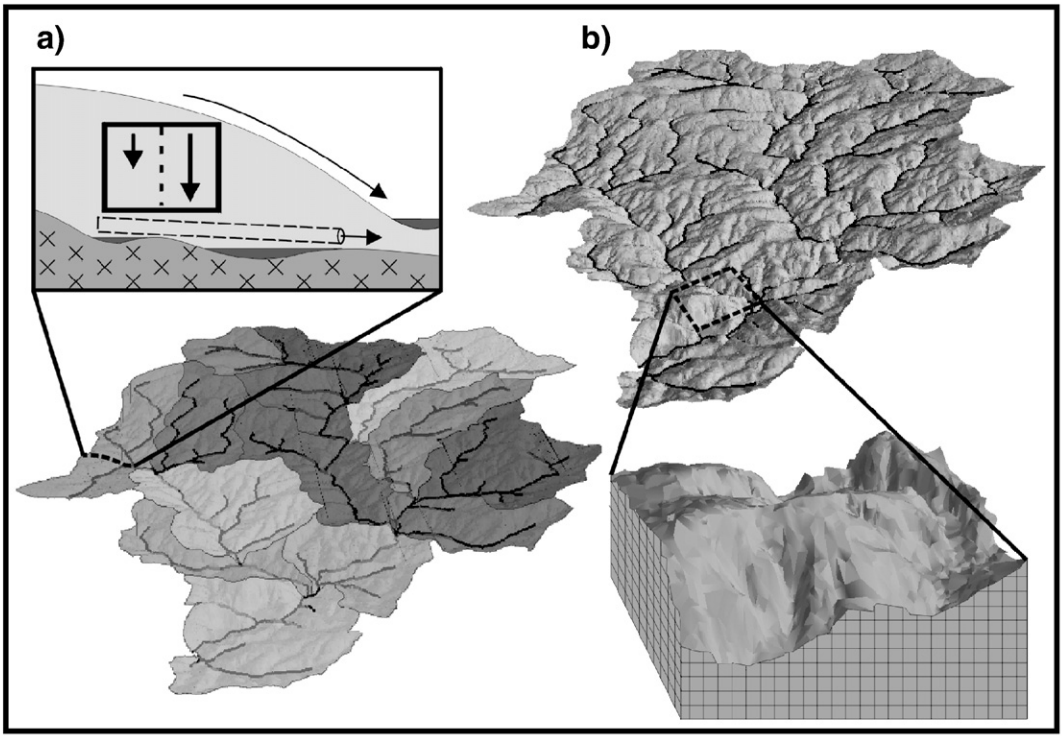

Fig. 2. Distributed models. a) Hydrological unit approach, b) 3D surface/vadose zone/groundwater models.

parameters in the 2D grid. HYDRUS-2D (Šimůnek et al., 1999), and its latest update HYDRUS (2D/3D) (Šimůnek et al., 2006), are the two- and two/three-dimensional equivalents of HYDRUS-1D, respectively. The origin of HYDRUS models can be traced back to the early 1970s (Neuman, 1972; see Šimůnek et al., 2008b). Additional features in HYDRUS-2D concern boundary conditions (e.g., tile drainage) and tools for calculating spatially distributed model parameters. The TOUGH model family is mentioned for completeness. The integral finite differences method allows TOUGH2 to be used as a DPM with multi-phase (e.g., water/air) flow equations in one to three dimensions. TOUGH codes have been extensively used for problems of flow and chemical transport in fractured rock (Pruess, 2004).

2.2.2. Gravity-driven PF models

A kinematic wave (KW) one-region model (Beven and Germann, 1982; Germann, 1985) is based on the boundary layer flow theory and was used for describing PF (e.g., Germann and Di Pietro, 1996, 1999). The KW model assumes that the wetting front proceeds by convective film flow in the mobile region and does not exchange water with the immobile region. The kinematic dispersive wave (KDW) model (Di Pietro et al., 2003) was developed by applying a second-order correction to the KW model. MACRO (Jarvis, 1994; Larsbo and Jarvis, 2003; Larsbo et al., 2005) is a 1D DPM that combines a KW description of water flow and solute convection for the macropore region with Richards water flow and solute convection dispersion in the matrix. Water transfer into the matrix is treated as a first-order approximation to the water diffusion equation and is proportional to the difference between actual and saturated matrix water contents. The latter is defined at a particular boundary pressure head that partitions the total porosity into matrix pores and macropores. Reverse transfer of water in excess of matrix saturation is instantaneously routed from the matrix into macropores. In an analogue fashion, solute advection and diffusion are calculated as first-order rate processes. MACRO considers swell–shrink dynamics of clay soils. The KW model and MACRO theoretically calculate identical hydrographs for saturated matrix conditions without lateral water transfer. The version MACRO 5.0 (Larsbo and Jarvis, 2003; Larsbo et al., 2005) is Windows-based and provides options for Monte-Carlo uncertainty estimation and for parameter estimation based on the SUFI program (Abbaspour et al., 1997).

SIMULAT (Diekkrüger, 1996) is a 1D DPM that considers gravitational flow and solute convection in the macropores, advective solute exchange with the matrix, and vertical Richards flow and solute transport by convection and dispersion in the matrix. The Root Zone Water Quality Model, RZWQM (Ahuja et al., 2000) utilizes a DP-MIM description of 1D vertical soil water flow and chemical movement. Three transport regions are assumed to exist in the soil: cylindrical macropores, the mobile soil matrix, and the immobile soil matrix. In the macropores, water flow is calculated using the Poiseulle equation and solutes are displaced by convection. In the mobile matrix region, water flow is described using the Green-Ampt approach during infiltration and the Richards equation during redistribution, while solute moves by convection. Water transfer is assumed to proceed uni-laterally from the macropores into the matrix by (Green-Ampt) imbibition, which can be restricted by a sorptivity (impedance) factor. Solute exchange

Table 1
Characteristics of common 1D models for preferential flow and tracer transport [a]

	RZWQM	MACRO	HYDRUS-1D [b]	CRACK-NP	SIMULAT
Documentation	(Ahuja et al., 2000; Malone et al., 2004)	Larsbo and Jarvis, (2003, 2005)	(Šimůnek et al., 2005; 2008a, Šimůnek and van Genuchten, 2008)	Armstrong et al. (2000)	Aden and Diekkrüger (2000)
Model type(s)	DPM, DP-MIM	SPM, DPM	SPM, MIM, DPM, DP-MIM	MIM	DPM
Water flow, matrix	Green-Ampt (downward), Richards (redistribution)	Richards	Richards	Infiltration: Philips (but immobile water in peds within soil)	Richards
Water flow, PF paths	Poiseulle (gravity flow in cylindrical macropores)	Kinematic wave (gravity flow in macropores)	Richards	Poiseuille (gravity flow in cracks)	Kinematic wave (gravity flow in macropores)
Potential ET	Penman Monteith or prescribed input	Penman Monteith or prescribed input	Penman Monteith, Hargreaves, or prescribed input	Prescribed input data	Penman Monteith or prescribed input
Real ET	Reduction functions for ET	Reduction functions for ET	Reduction functions for ET	No	Reduction functions for ET
Solute transport, matrix	Convection	Convection–dispersion	Convection–dispersion	Immobile	Convection–dispersion
Solute transport, PF paths	Convection	Convection	Convection–dispersion	Convection	Convection
Water transfer	Green-Ampt, one-way (macro-matrix)	First-order	First-order	Philip infiltration	Darcy
Solute transfer	matrix-macro: instant mixing; mo-im: first-order (diffusion)	First-order (convection–diffusion)	First-order (convection–diffusion)	Diffusion (Ficks law)	Convection
Heat flux	Yes	Yes	Yes	No	Yes
Root growth	Yes	Yes	Yes	Yes	Yes
Root uptake	Water, solute	Water, solute	Water, solute	Water	No
Volatilization	Yes	(Yes) — lumped dissipation rate	Yes	No	No
Surface runoff	Yes	(Yes)	(Yes)	(Yes)	No
Solute in runoff	Yes	No	No	No	No
Management options	Crop rotations, tillage, fertilizer, manure, pestic. application mode	Tillage	No	No	No
Tile drainage	Yes	Yes	Yes	(Yes)	Yes
Inverse method	No	SUFI	Levenberg–Marquardt	No	No
Graphical User Interface	Yes	Yes	Yes	No	No
Selected further features	crop yield, nutrient cycle, equilibrium chemistry	Swelling–shrinking, consolidation, surface sealing, canopy intersection and washoff, Monte Carlo uncertainty estimation, Metabolite transport	Particle transport, CO_2 transport, chemistry of carbonate system, major ion chemistry, metabolite fate	Swelling–shrinking, canopy interception	

(ET — evapotranspiration, T — temperature, mo — mobile region, im — immobile region, macro — macropores, PF — preferential flow, θ — soil moisture content, (Yes) means: is considered in a simplified way, SPM — single porosity model, MIM — mobile–immobile water model, DMP — dual-permeability model, DP-MIM — dual-permeability model with immobile water).

[a] Model features relevant to transport of reactive solutes such as pesticides and management are shown in the companion paper.

[b] Similar features are included in HYDRUS-2D and HYDRUS (2D/3D).

proceeds by instant mixing between the macropore solution and a thin matrix layer (e.g., 0.6 cm), and by first-order diffusion between mobile and immobile matrix regions (Malone et al., 2001).

VIMAC. (Greco, 2002) is a three flow domain model which considers flow in swelling and shrinking clay soils. Flow in the matrix is modelled by means of Darcy equation. Water film flow in shrinkage cracks of variable aperture depending on the matrix saturation is described using the KW equation, as is flow in permanent macropores, a fraction of which may be defined as dead-end pores with internal catchment. Horizontal water exchange between the three regions is described by first-order equations taking into account partial wetting of macropore and cracks, and other effects (Greco, 2002).

CRACK-NP. (Armstrong et al., 2000) is a MIM that describes 1D vertical water flow and solute transport in clay soils with planar shrinkage cracks and peds (soil aggregates) of cubic shape. The crack and matrix porosities are dynamic functions of the soil moisture. Water flow in the cracks is calculated using Poiseuille's law modified to account for the cracks' tortuosity and connectivity (Germann and Beven, 1981). Lateral water uptake of peds is calculated using the Philips equation modified for the fractional wetted aggregate surface area. No vertical flow is considered in peds. Solute transport in the cracks proceeds by convection, with lateral advective and diffusive losses of solute to peds, and potential reverse diffusive transfer from peds into cracks.

M-2D was developed by Kohler et al. (2001) by combining the 2D Richards-CDE based SWMS-2D model (Šimůnek et al., 1994) with a 1D DPM adopted from MACRO (Jarvis, 1994). First-order water transfer is described similarly as in Gerke and van Genuchten (1993), whereas solute transfer is by advection only.

2.2.3. Empirical PF models

Physically-based two-region approaches with parsimonious parameter demand were developed. Two types can be distinguished with respect to treatment of PF: instantaneous routing from the surface to the bottom (PF routing approach), and a layer-wise cascade of macropore water in excess of a certain saturation threshold (capacity approach).

2.2.3.1. PF routing approaches. The FRACTURE submodel of the HYDRUS-1D (Novák et al., 2000) formally divides infiltration into the soil matrix into two components: vertical infiltration through the soil surface (1D-Richards equation), and lateral infiltration via soil cracks (Green-Ampt approach). Excess water that does not infiltrate the soil surface is routed into soil cracks. The Soil Water Atmosphere and Plant (SWAP) model (van Dam et al., 1997; van Dam, 2000) comprises 1D Richards and CD equations for the matrix and a routine for routing water and solute in dynamic shrinkage cracks, coupled by first-order water and diffusive solute transfer. Lateral infiltration from the cracks into the soil matrix is calculated as a first-order process and depends on the active crack area. An express fraction of solute may be routed directly to the drains.

2.2.3.2. Capacity approaches. The COUP model (Jansson and Karlberg, 2004) describes matrix flow using the Darcy and the continuum equations, while infiltration excess of the soil matrix is cascaded as bypass flow to lower soil compartments. Water infiltration into frozen soil and heat transport are also considered. COUP combines the SOIL (Jansson, 1991) and SOILN (Eckersten et al., 1998) models. The LEACHW model based on the Richards equation (Hutson and Wagenet, 1992) was extended with a description of water moving downward through fractures in a tipping bucket fashion (Booltink, 1994). The Solute Leaching Intermediate Model, SLIM, is a MIM of capacity type. SLIM has two main parameters: the amount of immobile water and the permeability (Addiscott et al., 1986; Addiscott and Whitmore, 1991). The 1D-DPM IN^{3}M (Weiler, 2005) calculates infiltration into the matrix and horizontal water transfer using Green-Ampt equations, and assumes the Darcian flow for redistribution. Infiltration exceeding the soil matrix capacity triggers surface initiation of PF. The excess water is distributed into the macropores according to the stochastic Weibull frequency distributions of observed macropore drainage areas. Additionally, subsurface PF initiation may occur at layers boundaries. Macropore flow is routed downwards by layer-wise mass balance (Weiler, 2005). Based on concepts of Bouma (1990), Kohler et al. (2003) developed two simple capacity DPM of water flow: the surface trigger model assumes that gravitational PF is generated at the soil surface, while the subsoil trigger model assumes that PF is created at the topsoil-subsoil boundary when the water storage capacity of the topsoil is exceeded.

2.2.4. Catchment scale models

2.2.4.1. 3D surface/vadose zone/groundwater models. MODHMS is a 3D water flow model based on the 3D Richards equation (Panday and Huyakorn, 2004). Areal overland flow and flow through a network of streams and channels is described by diffusion wave equations. A bypass routing approach accounts for PF to tile drains that depends on weekly rainfall, irrigation, and an empirical bypass fraction parameter. FRAC3DVS is a 3D DFM (3D matrix, 2D parallel fractures) for variably saturated ('VS') flow and solute transport in fractured porous formations (Therrien and Sudicky, 1996). Vertical, horizontal and inclined 2D fractures are incorporated into the 3D grid by superimposing 2D parallel plate elements onto 3D matrix block elements. Fracture faces and matrix blocks share common nodes along the fracture walls, such that continuity of hydraulic head and concentration is ensured in the calculation of mass exchanges. Water flow is calculated using Richards equation in the matrix and an adapted form of Richards equation for the fractures. Transport is by advection and dispersion in both the fractures and the matrix. FRAC3DVS can be regarded as an extension of the 2D FRACTRAN groundwater model (Sudicky and McLaren, 1992). A recent successor of FRAC3DVS is the 3D fully integrated subsurface and overland flow and solute transport model HydroGeoSphere (Therrien et al., 2005). The Integrated Hydrology Model, InHM (VanderKwaak, 1999; VanderKwaak and Loague, 2001), also gives a fully coupled description of the surface and subsurface water flow and solute transport over the 2D land surface and in the 3D dual continuum subsurface under variably saturated conditions. Overland flow is calculated using the 2D nonlinear diffusion-wave equation together with Manning's equation. In the subsurface 3D Richards equation is assumed; optionally in continuum or dual-continuum form.

2.2.4.2. Hydrological unit models. WEC-C (Croton and Barry, 2001) is a distributed water and solute displacement model in a GIS-like environment. Vertical PF can be alternatively considered as a 1D DPM (Gerke and van Genuchten, 1993) or 3D DPM. Groundwater flow is modelled as a 2D Darcy system and overland flow using 2D Manning and kinematic wave equations. Boundary conditions account for interception, canopy storage, transpiration, evaporation, stream routing and groundwater discharge. WEC-C was validated against synthetic data examples (Croton and Barry, 2001). CATFLOW (Zehe et al., 2001), based on the original model HILLFLOW (Bronstert, 1994), subdivides the catchment area into a number of hillslopes connected to a drainage network. Each hillslope is conceptualized as 2D vertical cross-sections through its main slope. Subsurface flow and transport in each hillslope is described by 2D Richards-CDE equations. The effect of macropores on water flow is represented by a linear increase of the bimodal hydraulic conductivity function above a saturation threshold of 0.8. Surface runoff along hillslope elements, subsurface drainage, and runoff in the channel network is calculated based on the advection diffusion approximation of the 1D Saint Venant equations. The Distributed Hydrology Soil Vegetation Model, DHSVM (Wigmosta et al., 1994), was extended with PF descriptions

(Beckers and Alila, 2004). DHSVM uses the 1D Green-Ampt equation to calculate vertical unsaturated matrix water movement. Macropore flow is initiated by surface ponding and is instantaneously routed to the bedrock surface. Lateral groundwater flow in the matrix is calculated using a quasi 3D Darcian approach, while lateral PF in the groundwater is computed using a Darcy-based routing approach. Preferential and matrix flows are disconnected. Overland flow is calculated using a linear reservoir (capacity) scheme. The 1D Distributed Subsurface-Surface Flow Dynamics Model, DSFDM (Mulungu et al., 2005) adds a PF component to the Famiglietti and Wood (1994) catchment scale model. Three soil layers are considered in DSFDM: in the top 1-m soil layer, macropore flow and lateral interflow may occur; the middle 'transmission' zone assumes vertical flow in the soil matrix, and the bottom layer with Darcy (lateral) flow of groundwater in the soil matrix. Preferential infiltration through macropores and interflow in pipes, as well as overland flow, are described using KW equations.

3. Modelling of preferential flow systems

A primary reason for the interest in PF is its impact on contaminant transport. However, a number of studies have utilized water flow observations only, without solute observations, to evaluate PF models and to study PF processes at different scales. The review of such model applications (Table 2) should indicate the model complexity and data required to describe PF under the given conditions.

3.1. Scale I (soil column, lysimeter)

3.1.1. Constructed soil columns

The advantage of using soil columns (length approx. 0.1 m to 1 m) for a basic flow process analysis is that IC and BC can be optimally controlled, and the heterogeneity of soil properties is negligible. Water flow experiments were conducted with dual-porosity media with known geometry to eliminate unknowns that might otherwise obscure the PF process under study. The water transfer term and domain specific hydraulic functions in capillary PF models (DPM) were evaluated. Continuum models were also tested regarding their capability of simulating PF. Examples are discussed below.

3.1.1.1. Water transfer term. Ahuja et al. (1995) used RZWQM to describe the effect of macropore flow under rainfall in a soil column with a 3-mm artificial macropore. In order to match the macropore bottom outflow, the lateral absorption of water from macropore to soil matrix had to be adjusted for compaction of the macropore wall (Ahuja et al., 1995).

Table 2
Model applications for preferential WATER flow

Scale	Model type; name	Dimension, equation name or description	Authors of model application
Scale I			
AC	SPM, MIM, DPM; HYDRUS-1D	1D; Richards (2× for DPM), f.o. (or s.o.) water transfer	(Castiglione et al., 2003; Köhne and Mohanty, 2005; Köhne et al., 2006a)
AC	DPM; DPOR_1D	1D; single integro-differential equation	Lewandowska et al. (2005)
AC[a]	MIM; KDW model	1D; Convective dispersive kinematic wave, no water transfer	Di Pietro et al. (2003)
UC	SPM; n.a.	1D; Richards, kinetic θ–h relation	Ross and Smettem (2000)
UC	TPM; VIMAC	1D; Richards (matrix), KW (water film in cracks), KW (macropores)	Greco (2002)
UC	SPM, MIM, DPM; HYDRUS-1D	1D; Richards (2× for DPM), f.o. water transfer	Šimůnek et al. (2001)
L	SPM, DPM; MACRO	1D; Richards (matrix), KW (macropore), f.o. water transfer	(Jabro et al., 1998 (L); Ludwig et al., 1999 (F); Alaoui et al., 2003 (UC); Akhand et al. 2006 (P))
Scale II			
S	DPM/S_1D_Dual	1D; Richards (2×)	Zumr et al. (2006)
S	DPM (stochastic)/IN^{3}M	1D; Green-Ampt, Darcy	Weiler (2005)
P	SPM/MIN3P + Rosetta	1D; Richards, modified $K(h)$	Gérard et al. (2004)
P	MIM/Fractal active region model	1D; Richards, θ-h is a function of active (i.e., mobile) region, no water transfer	Liu et al. (2005)
P		0D-distributed, initiation of macropore flow at surface or subsurface according to geostatistic microtopography and macropore distribution	Weiler and Naef (2003)
Scale III			
H	DPM/Hillslope PF model	3D-Richards (matrix), 1D-Manning (pipe flow)	Tsutsumi et al. (2005)
W, H	DPM/DHSVM	(Pseudo) 3D; Green Ampt, routing (no transfer), Darcy	Beckers and Alila (2004)
W, S	SPM (distributed)/CATFLOW	2D-vertical Richards (DEM and GIS based) with bimodal $K(h)$ for hillslope elements, and 1D simplified Saint Venant equations for surface runoff and drainage	(Zehe and Blöschl, 2004; Zehe et al. 2005)
W	DPM/MODHMS + PF	3D-Richards (matrix), 1D-bypass routing (macropore)	Vrugt et al. (2004)
W	SPM, DPM/TOPLATS&SIMULAT	3D (quasi); 2D-(TOPLATS)& 1D (SIMULAT)	Bormann et al. (2005)
R	DPM/DSFDM	1D-distributed, KW	Mulungu et al. (2005)

Scale I: artificial or repacked soil column (AC), undisturbed column (UC), lysimeter (L); scale II: soil profile (S), plot (P), hillslope (H), field (F); scale III: watershed (W), river basin (R); PF — preferential flow, DEM — digital elevation model, SPM — single porosity model, DPM — dual permeability model, TPM — triple permeability model, MIM — mobile immobile model, KW — kinematic wave, KDW — kinematic dispersive wave, f — fast flow region, m — matrix, f.o. — first-order, s.o. — second-order, SCM — stochastic convective model, STM — stream tube model, DFMD — discrete fracture matrix diffusion model, SOTS — second-order two-site model, MRM — multireaction model, BIM — Lagrangian transport model with bimodal lognormal probability density functions (pdfs), TFM — lognormal transfer function model.

[a] Repacked soil columns inoculated with earthworms.

Köhne and Mohanty (2005) tested the effect of having a first- or second-order water transfer term (Köhne et al., 2004a) in the DPM in HYDRUS-1D (Šimůnek et al., 2003). DPM simulation results were compared with those obtained using the 2D Richards equation based HYDRUS-2D (Šimůnek et al., 1999) as a reference flow model. A column setup (80 cm height, 24 cm diam.) provided outflows separately from the matrix and the central cylindrical PF region, as well as pressure heads and water contents in the PF and matrix regions at various positions to enable PF model inversion. The DPM could describe the observed hydraulic nonequilibrium between the matrix and the PF domain. Region-specific results came closer to those of the 2D-reference model when using the second-order mass transfer term (Köhne and Mohanty, 2005).

In a comparable experimental study, the same DPM with first-order term could model water transfer accurately only when used with a scaling factor that varied with the macropore hydraulic conductivity (Castiglione et al., 2003).

3.1.1.2. Domain specific hydraulic functions. Lewandowska et al. (2005) conducted infiltration experiments with a double-porosity medium composed of a mixture of sand and sintered clayey spheres arranged in a periodic manner. The resulting inflow and outflow observations were used to verify the DPM approach 'DPOR-1D' (Lewandowska et al., 2004). With domain specific hydraulic parameters (van Genuchten, 1980) identified independently for the sand material and clayey spheres using HYDRUS-1D (Šimůnek et al., 1998), the macroscopic prediction satisfactorily approximated observed inflow and outflow for the composite dual-permeability medium (Lewandowska et al., 2005). A similar procedure for parameterizing a DPM resulted in a satisfactory simulation of water drainage from a repacked soil column with cylindrical PF region (Köhne et al., 2006a). The 2D counterpart of DPOR-1D, named DPOR-2D, was successfully tested using data obtained from an axi-symmetrical experimental infiltration setup with a similar mixture of sand and sintered clay spheres (Szymkiewicz et al., 2008).

3.1.1.3. Continuum models. Different (single-) continuum models were also evaluated for their performance in describing preferential water drainage. For example, the KDW model (Di Pietro et al., 2003) was tested using water flow observations obtained in laboratory experiments conducted with repacked soil columns (28 cm height, 15 cm diam.) inoculated with earthworms. With model parameters estimated from the observed drainage hydrograph, the KDW model described the outflow hydrographs accurately (Di Pietro et al., 2003). In another example, the Richards model with bimodal functions (Durner, 1994) performed similarly well as the DPM in describing water outflow from the bulk soil (Köhne and Mohanty, 2005).

The success of the continuum models prompted the question of whether water flow measurements alone are sufficient to obtain reliable values of hydraulic DPM parameters by inverse identification. Using the experimental setup of Köhne and Mohanty (2005), Köhne et al. (2006a) found that the outflows from either the matrix or the PF region could only be described when the DPM was fitted to region-specific outflow data. It was concluded that for natural soils where experimental data do not come in 'separated form' for fast and slow flow regions, domain-related hydraulic parameters of the DPM may be difficult to identify from water flow observations alone (Köhne et al., 2006a).

3.1.2. Intact soil columns and lysimeters

Undisturbed, naturally structured soil cores and lysimeters more closely represent natural field conditions than repacked soil columns with artificial macropores. Both capillary and gravity-driven PF models (see Section 2.1) were evaluated based on experimental data from such conditions. The following examples compare equilibrium and physical nonequilibrium models. Different types of data were used in the analyses, including outflow, water contents, and pressure heads.

3.1.2.1. Capillary PF models. Ross and Smettem (2000) applied their one-region nonequilibrium model (Fig. 1b) and a corresponding uniform flow model to describe TDR measured water contents and outflow responses from large (0.67 m height, 0.3 m diam.) undisturbed soil columns subject to constant rainfall. The surface 0.2 m was a fine sandy loam and the subsurface a kaolinitic clay. The presence of PF was inferred from commencement of outflow prior to complete wetting of the soil. The uniform flow model failed to describe this behaviour. By contrast, the PF model could describe both cumulative outflow and water contents for five out of six other soil cores, when only the macropore hydraulic conductivity was individually calibrated (Ross and Smettem, 2000).

Šimůnek et al. (2001) implemented the Marquardt–Levenberg parameter optimization in HYDRUS-1D for inverse DPM, MIM and SPM simulations of upward infiltration experiments performed on undisturbed sandy loam soil cores (0.10 m height and diam.), without macropores, sampled from a tilled soil horizon. The multi-objective function was defined in terms of the cumulative upward infiltration rate, pressure head readings, and the final total water volume in the soil core. The observations could be similarly well characterized using MIM and DPM, as opposed to using the Richards equation based SPM, but the data were insufficient to achieve uniqueness for many (up to 9) estimated parameters required for two-domain models (Šimůnek et al., 2001).

3.1.2.2. Gravity-driven PF models. Alaoui et al. (2003) compared MACRO (Jarvis, 1994) and a corresponding one-domain KW approach to assess the extent of PF in a structured soil column (0.45 m height, 0.39 m diam.). The soil column contained aggregates, roots, and earthworm channels and texture changed from loamy sand (top) to sand (bottom). Measured water contents using TDR and outflow were studied during infiltration into unsaturated soil. Outflow did not start earlier than the TDR response. The model results suggested intense exchange between macropores and the permeable sandy matrix. For these hydraulic equilibrium conditions, with only one calibrated parameter, KW results came fairly close to those using MACRO (with six calibrated parameters). However, the KW simulation showed step-increases of water contents and outflow that were not apparent in the data (Alaoui et al., 2003). This convective flow effect was removed by second-order correction to the KW model (Di Pietro et al., 2003).

VIMAC was applied to model infiltration experiments carried out on a large undisturbed clay soil column (55 cm high, 30 cm diam.) and lasting from 6 h to 8 days. Hydraulic model parameters were derived from independent measurements of hydraulic conductivity and water retention curves, shrinkage characteristics, aggregate water diffusivity, and soil morphology. The permanent macropores' cross-section areas were calibrated. VIMAC was capable of reproducing outflow and TDR-measured water content profiles. Results improved when the internal catchment process in dead-end macropores was included (Greco, 2002).

Jabro et al. (1998) compared the gravity-driven PF model MACRO (Jarvis, 1994) with the empirical two-domain PF models SLIM (Addiscott et al., 1986; Addiscott and Whitmore, 1991) and SOIL (Jansson, 1991), and with continuum models LEACHW (Hutson and Wagenet, 1992) and NCSWAP (Molina and Richards, 1984), to evaluate the significance of considering PF for simulating water drainage. The silt loam soil contained worm holes, decayed root channels and cracks. The models were compared with regard to their accuracy in predicting drainage fluxes below the root zone of corn as measured with 18 pan lysimeters (area 0.465 m^2, depth 1.2 m) during 5 years. The first of these years was taken for model calibration. Simulated drainage fluxes for all five models were not significantly different, and were all within 95% confidence intervals of the measured values (Jabro et al., 1998).

3.1.3. Summary for preferential water flow modelling at scale I (column, lysimeter)

Most PF models applied at scale I are one-dimensional. The typical time scale of the experiments is between hours and days. Both continuum models and non-equilibrium approaches with one or two domains were used. For artificial dual-porosity media, PF processes as represented by region specific observations of flow, water contents, and pressure heads could be successfully described with various DPM. A second-order water transfer term could further enhance the accuracy for certain geometries of the flow system.

If the simulation objective was to describe the hydrograph only, simpler one-domain equilibrium flow models often performed equally well. Thus, structured soils may often be regarded as a continuum for the purpose of modelling water flow. Consequently, inverse identification of two-domain model parameters generally yielded poor or uncertain results when using data of water outflow or infiltration. Hence, a hydrograph must be considered insufficient to make meaningful statements about PF. Conversely, combined data on flow and soil hydraulic state variables include PF information that can only be described by a non-equilibrium model.

3.2. Scale II (plot, field)

Compared to scale I, additional uncertainties for modelling at scale II (plot, field) arise due to the spatial heterogeneity and temporal dynamics of the system with regard to soils, crops, meteorological variables, boundaries, and the lateral surface or subsurface runoff.

The plot and field scales are not exactly defined terms. From an agronomic perspective, the 'field scale' is the smallest management unit that would represent the entire agricultural field and its crop. In line with more common terminology in soil hydrology, the field scale is defined here as covering a considerable part of a field (0.1 to 100 ha) under the prevailing boundary conditions, while the plot scale refers to a small part of a field (1 m^2 [soil profile] to 1000 m^2) often subject to experimentally controlled boundary conditions (irrigation). Depth is in the meter range. Several studies involved tile-drained fields, because of the straightforward delineation of domain boundaries, the integral function of the drain outflow, and the potential environmental effect of drains on the diffuse pollution by agrochemicals.

The main conceptualizations of soil (texture and structure) heterogeneity for water flow studies involved (i) deterministic application of a 1D PF-model with a single set of parameters, and (ii) stochastic application of a 1D PF-model with several sets of parameters sampled from their respective frequency distributions. Approach (i) was frequently adopted due to its relative simplicity. However, calibrated 'effective' parameter values are lumped with heterogeneities and other scale effects, and thus may lose their physical meaning. Approach (ii) allows for uncertainty analysis of simulation results. Uncertainty may have various sources, including uncertainty of parameters, model structure, initial and boundary conditions, and user subjectivity. An alternative to approach (ii) is the distributed model.

3.2.1. Plot scale

At the plot scale, typically the 1D deterministic approach (i) was applied. The same problems of identifying PF parameters were encountered as on the column scale, so only brief examples will be given. Ludwig et al. (1999) modified and calibrated MACRO (Jarvis, 1994) for simulation of groundwater levels and matric potentials observed during 2 weeks in tile-drained aggregated clay and sandy loam soil plots (15 m by 100 m). DPM simulation results improved only slightly compared to those using a one-domain (Richards) model, and the water transfer rate parameters in the DPM could not be identified (Ludwig et al., 1999).

Zumr et al. (2006) applied the 1D counterpart of the DPM S_2D_Dual (Vogel et al., 2000) for the Levenberg–Marquardt estimation of soil hydraulic parameters using measured pressure heads below a plot with pseudogley soil containing desiccated cracks, biopores and interaggregate fractures. Problems with reproducing observations were attributed to the uncertainty of allocating tensiometer values to the matrix or macropores (Zumr et al., 2006).

Gérard et al. (2004) tried to predict 4 years of daily measurements of soil water content in a forest soil plot with PF in a capillary pore network. They utilized pedotransfer functions (ROSETTA, Schaap et al., 2001) and a bimodal hydraulic conductivity function (Mohanty et al., 1997) with the Richards' flow submodel of MIN3P (Mayer et al., 2002). However, soil water contents were under-predicted in the wet range. The match improved after calibration, but calibrated PF parameters appeared to be of limited physical meaning (Gérard et al., 2004).

The seasonal dynamics of structured soil hydraulic properties (Messing and Jarvis, 1990, 1993; Dasgupta et al., 2006b) pose an additional challenge. Akhand et al. (2006) applied MACRO to simulate 60 days of hourly readings of water contents, matric potentials, and tile drain discharge from a silt–loam soil plot (7 m by 65 m) with cracks and worm burrows. MACRO was calibrated for relatively wet autumn conditions. In the drier validation period of spring and summer when soil cracks developed, the model underestimated bypass flow and tile discharge (Akhand et al., 2006).

There is increasing evidence that PF patterns in soil can be characterized by fractal or multi-fractal geometries (Rieu and Sposito, 1991; Hatano and Booltink, 1992; Baveye et al., 1998; Boufadel et al., 2000; Olsson et al., 2002, 2007). Liu et al. (2005) incorporated fractal PF patterns into a MIM. Fractal information was included in terms of a constitutive relationship for the fraction of mobile water as a function of the saturation. A preliminary validation of the model was performed based on field observations (van Dam et al., 1990) of the variation of mobile water content with soil depth (Liu et al., 2005).

3.2.2. Field scale

Examples of the 1D stochastic approach (ii) and hydrological unit approach are presented as field scale applications. PF models traditionally do not consider a population of macropores, but assume a single set of properties representing some 'equivalent' PF path at the field scale. Booltink (1994) was among the first to represent heterogeneity in bypass flow modelling stochastically. The LEACHW model (Hutson and Wagenet, 1992) was extended with a tipping-bucket description of fracture flow in a clay soil at a tile drained site of 100 m × 300 m. The Monte Carlo analysis was performed for soil hydraulic parameters related to PF. Their frequency distributions were partly derived from measurements including the contact areas of soil aggregates adjacent to PF paths, stained macropores, water contents, and hydraulic conductivities. Monte Carlo parameters from the final set of acceptable model simulations were used as input for model validation. The model reproduced the sharp groundwater level rise and an instant drain discharge, after storm events and due to PF through shrinkage cracks. An accurate characterization of bypass flow after short-duration, high-intensity rain events required a short time interval (< 15 min) for rainfall measurement (Booltink, 1994).

In a more recent example, the effect of rain intensity on the fraction of different macropores involved in PF was evaluated in a stochastic application of the DPM IN^{3}M (Weiler, 2005). A sensitivity analysis showed that flow variability in macropores at higher rainfall intensities markedly reduces water transfer and increases bypass flow. This might give an additional, field-scale explanation for the frequently observed phenomenon of the small transfer rate coefficient calibrated in DPM to simulate non-equilibrium PF and transport (see discussion in Jarvis, 2007). IN^{3}M predicted water balance, water content variation, and dye coverage with depth as observed in irrigation and dye tracer experiments at three field sites reasonably well (Weiler, 2005).

Weiler and Naef (2003) simulated macropore flow initiation at the surface or at a saturated subsurface layer. Simulations were based on spatial distributions of earthworm burrows and surface micro-topographies surveyed at four grassland sites. A simple routing model of PF treated as matrix saturation excess was applied. The results showed that macropore density primarily controlled the total macropore flow, while surface micro-topography controlled the probability distribution; only a few macropores contributed significantly to the total macropore flow. Qualitative agreement was found between results from model simulations and from several published laboratory experiments (Weiler and Naef, 2003).

Tiemeyer et al. (2007) developed and tested the spatially distributed model MHYDAS-DRAIN to assess the effects of tile drainage and PF on the hydrology of lowland catchments. The PF component was modelled using the transfer function approach. The model was applied to two fields (4.2 and 4.7 ha) in the Pleistocene lowland of North-Eastern Germany. Modelled discharge rates and groundwater levels agreed reasonably well with measured data, while parameter values were found to depend on the calibration criteria as well as on the spatial and temporal resolution of the modelling domain (Tiemeyer et al., 2007).

3.2.3. Summary for scale II (plot, field)

Applied PF models simulating PF at the plot and field scale do not differ substantially from those used at the column scale; they are still 1D and usually based on one- or two-domain concepts. The application time scale usually ranges from weeks to months, whereas time intervals required for accurate calculation of bypass storm flow are still in the same range as for scale I. Non-uniqueness problems were limiting inverse PF parameter identification at this scale. Some of these problems may be caused by the uncertainty in relating local scale (microscopic) measurements to an effective (macroscopic) region of a two-domain model. Other limitations and problems observed already at scale I (see Section 3.1.3) are also transmitted to scale II. Stochastic PF model studies have shown potential to integrate effects of heterogeneity, parameter uncertainty, and PF triggering processes into field scale modelling. Macropore flow variability was found to reduce water transfer, which may serve as a new explanation for the small transfer rate coefficients in DPM required to simulate PF and transport (see Section 4).

3.3. Scale III (hillslope, catchment area)

A multitude of hydrological processes operating at different spatial and temporal sub-scales control the dynamic catchment response. Catchment hydrology is a vast and dynamically developing research area far beyond the scope of this review. Importantly, large catchment hydrological models address problems at a scale increasingly relevant to global climate models. Interestingly, due to uncertainties involved in studying catchment hydrology, a paradigm shift appears to be underway where traditional approaches are challenged, new philosophical perspectives emerge, and innovative measurement and modelling approaches are proposed. The relations between form and function, patterns and processes, genesis and dynamics of catchments are moving into focus. The reader is referred to an excellent overview of the state-of-the-art in catchment hydrology research as covered in three invited commentaries (Tetzlaff et al., 2008; Soulsby et al., 2008; Dunn et al., 2008). Macropore flow in the vadose zone is but one out of several rapid flow processes that may affect the overall hydraulic response and water quality of a catchment. A yet small but developing research area is on utilizing models to analyze rapid flow processes at the hillslope and catchment scales.

3.3.1. Hillslope scale

Many recent experimental investigations of hillslope hydrology have considered the role of PF (e.g., Noguchi et al., 1999; Buttle and McDonald, 2002; Tromp-van Meerveld and McDonnell, 2006a,b; Lin et al., 2006).

3.3.1.1. Lateral preferential flow: a threshold process. Lateral PF seems to be a hillslope characteristic responsible for peak rapid runoff generation in humid catchment areas (e.g., Lin and Zhou, 2008). Lateral PF is a complex process that can take a variety of forms including flow in 'pipes' (i.e., large macropores oriented parallel to the soil surface) along the base of the soil profile or in the topsoil (e.g., Uchida et al., 2001), or flow through a dynamic network of PF paths embedded in the soil matrix, movement in a thin saturated layer or along micro-channels above bedrock, and transport in exfoliation fractures in bedrock (Buttle and McDonald, 2002). Evidence from hillslope studies suggests that although individual macropore segments are often smaller than approximately 0.5 m in length, they have a tendency to self-organize into larger PF systems as sites become wetter (Noguchi et al., 1999; Sidle et al., 2001). There is evidence that a threshold of total storm event precipitation has to be surpassed in order to activate the lateral PF system and trigger subsurface pipe flow (e.g., Tromp-van Meerveld and McDonnell, 2006a,b).

3.3.1.2. Hillslope PF models. Different conceptual models were proposed, including interconnected, self-organizing PF systems in forested hillslopes (Sidle et al., 2001), hillslope pipe flow (Uchida et al., 2001), and flow through a subsoil macropore network as one of four main flow pathways (Lin et al., 2006).

Computational models include a 3D DPM that describes lateral PF in a hillslope containing dry, partially filled, or fully filled pipes in various arrangements. Matrix and macropore flows were computed using the 3D Richards and 1D Manning's equations, respectively, coupled with an integral expression for water transfer flux. The model results compared well with bench scale experiments of Sidle et al. (1995) (Tsutsumi et al., 2005).

Lehmann et al. (2007) applied a 2D percolation model that represents the hillslope as a grid of sites forming a lattice connected by bonds. Water-occupied sites connected by a bond form a cluster. A cluster may grow until, at a certain occupation probability (called the 'percolation threshold'), it spans the whole system and becomes a percolation cluster. Model validation was attempted based on published hillslope data of 123 events with measured rainstorm amount, outflow and initial water content at 70 cm (Tromp-van Meerveld and McDonnell, 2006a). Percolation theory appeared to be a valid approach to quantify threshold processes controlling the hillslope outflow after rainstorm events (Lehmann et al., 2007).

A threshold of rainfall depth halting the rapid water table rise and initiating lateral PF in hillslopes is also assumed in modified DHSVM (Beckers and Alila, 2004).

3.3.2. Catchment area scale

3.3.2.1. Catchment area ≈ 4 km^2. Zehe and Blöschl (2004) used CATFLOW (Zehe et al., 2001) to study how uncertainty of initial soil moisture values is propagated into model prediction uncertainty of the hydraulic responses in the agricultural Weiherbach catchment (3.6 km^2), Germany. The model parameterization was based on laboratory and catchment data. The results showed that the uncertainty in model predictions of discharge hydrographs was highest for initial soil moistures around the threshold value separating matrix and macropore flows. The predictive uncertainty decreased with increasing rainfall intensity. Moreover, satisfactory discharge predictions were only obtained when using spatially distributed precipitation values, not just an average value. Finally, calculations were repeated for a plot within the catchment. Perhaps surprisingly, the model uncertainty was reduced from plot (CV = 0.21) to catchment (CV = 0.08). This net reduction was explained by averaging of small-scale variability; this countervailing effect exceeded additional sources of variability at the catchment scale (Zehe and Blöschl, 2004).

Zehe et al. (2005) found that the simulated Weiherbach runoff pattern depended on average initial soil moisture. By contrast, spatial variability of initial soil moisture had a moderate effect (approximately +/−10% difference in runoff peaks) only for dry or intermediate saturation, where different realizations caused the saturation threshold to be surpassed at different catchment locations, thus triggering the two key processes of PF and overland flow. Spatial patterns of precipitation and macroporosity represented additional sources of runoff prediction uncertainty (Zehe et al., 2005).

3.3.2.2. Catchment area ≈ 10 km^2. Beckers and Alila (2004) modified (see Section 2.2.4) and applied DHSVM (Wigmosta et al., 1994) to analyze the contributions of preferential hillslope runoff to peak flow generation in a temperate rain forest catchment (10 km^2) on Vancouver Island, British Columbia, Canada. Hydrometeorological data from 1972 to 1990 were utilized. DHSVM was applied to evaluate hillslope contribution to peak flow generation in the 10-km^2 catchment area. Downslope subsurface flow rates during PF network formation were prescribed based on three tracer tests. Soil depths between 0.5 m and 1.25 m were defined assuming Gaussian random fields with a correlation length of 50 m. Soil hydraulic properties were fitted by comparing observed and simulated streamflows at four weirs in the catchment. The model was able to simulate peak flows and groundwater responses for both low and high intensity rains. While the differences between the 'matrix-flow-only' and the 'matrix-PF' model approaches for simulating low and medium streamflow were subtle, higher values of model efficiency were achieved for the 'matrix-PF' model of storm runoff. The model evaluation suggested that a unit area discharge threshold of 2.8 mm/h separates matrix flow dominated runoff from PF dominated runoff. Matrix flow dominated basin streamflow 67% of the year, while storm event flows were almost entirely derived from PF (Beckers and Alila, 2004).

3.3.2.3. Catchment area ≈ 40 km². MODHMS (Panday and Huyakorn, 2004) was applied with global inverse parameter optimization to simulate spatially distributed tile drainage for a 2-year period in the Broadview Water District (38.8 km^2), San Joaquin Valley, California (Vrugt et al., 2004). The domain was horizontally discretized into 200 m × 200 m square cells, which were vertically subdivided into 14 layers of 0.3 m (top) to 0.9 m (bottom). Inverse estimation of 7 parameters, including initial and saturated water contents, bypass fraction, crop coefficient, drain depth and conductance, and a Miller–Miller scaling factor for hydraulic conductivity, was carried out. The simulation results of MODHMS were compared with those obtained using a simple storage-based bucket model and a spatially averaged 1D flow model. The study concluded that the drain conductance and the bypass coefficient were determined well (while soil hydraulic properties were not) and that these parameters dominated the hydrology of the Broadview Water District (Vrugt et al., 2004). However, the conclusions about PF remain uncertain, since the simple bucket model gave similar results.

3.3.2.4. Catchment area ≈ 75 km². Jones et al. (2008) presented an application of the InhM model (VanderKwaak and Loague, 2001) to model hydrological processes in the 75 km^2 Laurel Creek Watershed in Southern Ontario, Canada. Subsurface flow was approximated as a steady-state process. Model calibration yielded good agreement between the observed and computed river baseflow discharge and hydraulic head values in 50 wells. Subsequent transient overland flow simulations based on rainfall time series were utilized to predict the observed discharge hydrographs. Moderate agreement was obtained. The model was deemed to be feasible to analyze the spatially heterogeneous and temporally dynamic response of a watershed to rainfall events (Jones et al., 2008).

3.3.2.5. Catchment area ≈ 350 km². Modified DSFDM was applied to simulate river flow in the temperate forested Hikimi river basin (352 km^2) in a mountain region in Japan (Mulungu et al., 2005). Ten model parameters were calibrated against the river flow in 1991 using hourly rainfall data. Of these optimized parameters, the saturated hydraulic macropore conductivity was the most sensitive. By contrast, the water transfer rate between the matrix and macropores was not a sensitive parameter. After model calibration, river flow in three subbasins was predicted between 1992 and 1996. Simulation results appeared satisfactory, except that peak flows were underestimated (Mulungu et al., 2005).

3.3.3. Summary for scale III (hillslope, catchment area)

Models operating at the catchment scale require a physically sound integration of the surface, vadose zone, and underlying aquifer systems. Preferable, although difficult to parameterize, is a fully coupled model scheme where the three layers interact with each other. An additional requirement posed here was to include PF. Most of the above models fulfil these requirements, including fully 3D model systems (MODHMS, InHM, HydroGeoSphere), and distributed models with some empirical simplifications in their equations (DSFDM, DHSVM). CATFLOW was applied with free drainage at the lower boundary of individual hillslope segments, and thus without groundwater system (Zehe and Blöschl, 2004). This approach may still give valuable insights for relatively small catchments where the focus is on fast runoff components, or for studying first-order controls. Still other approaches, such as percolation network models, are rare.

Compared to the predominantly vertical PF at smaller scales, overland flow and lateral PF (e.g., pipe flow) appear as important rapid runoff components in hillslopes. A consensus is emerging about a strongly nonlinear PF response to rainfall depth and intensity, with a threshold value triggering rapid runoff response in a hillslope. The identification of such thresholds for certain types of hillslopes or catchments might be one way towards simplifying the prediction of their hydrological behaviour.

Problems encountered in the identification of bypass or transfer rate parameters are not surprising, given that those problems were already encountered at smaller scales. However, models could still be used to assess how uncertainty of antecedent soil moisture and precipitation, and their spatial variability, is propagated into model prediction uncertainty.

It seems to be an unresolved question if solving overland or groundwater flow equations for horizontal cell sizes of 100 m × 100 m, with average properties, can be considered a physically-based approach. Likewise, the vertical discretization limit for solving Richards equation in the vadose zone can be in the cm range (Vogel and Ippisch, 2008) and is also difficult to fulfil at catchment scale. Advanced numerical techniques (Li et al., 2005; Park et al., 2008) may eventually overcome these problems.

Nevertheless, considerable advances have been achieved. Water flow studies in highly instrumented hillslopes were instructive for conceptual model development and evaluation. Sophisticated catchment scale PF models were developed and first applications for the catchment scale have been reported. The validation of rapid runoff model components appears challenging in catchment hydrology studies (i.e. studies that solely look at water flow not using solute or other additional information).

4. Modelling of tracer transport in preferential flow systems

A major concern with PF is its effect on contaminant solute transport. Studying conservative solute transport is often regarded as a prerequisite for understanding reactive solute displacement. Moreover, some agrochemicals, such as nitrate, behave relatively conservatively under certain conditions. Thus, preferential tracer transport was investigated in numerous model applications at different scales (Table 3).

The flow mode, i.e., steady state vs. transient, may have a distinct effect on PNE solute transport (Meyer-Windel et al., 1999; Cote et al., 1999). Traditional column studies involve transport under steady-state flow conditions and are modelled using closed-form analytical solutions. However, water flow in the vadose zone involves dynamic infiltration, redistribution, and capillary rise processes. Thus, models of solute displacement in the vadose zone generally consider transient water flow and require numerical solutions.

4.1. Scale I (soil column, lysimeter)

4.1.1. Constructed soil columns, steady state flow

Similar to investigations of water flow, studies of preferential solute transport involved repacked soil columns with artificial PF paths. Due to the impact of lateral concentration

Table 3
Model applications for preferential solute transport

Scale	Model type; name	Model dimension; flow state; description	Authors of model application
Scale I			
AC	MIM; n.a.	1D; steady state, MIM for macropore with radial (Fickian or first-order) diffusion into matrix	Young and Ball (1997, 1998)
AC	MIM; n.a.	1D, steady state, MIM, first-order (vs. Fickian diffusion) transfer term	Griffioen (1998), Griffioen et al. (1998)
AC[c]	MIM; BIM (Bimodal advection model)	1D, steady state, Langrangian mobile–immobile advective transport with bimodal lognormal pdf	Rosqvist et al. (2005)
AC	SPM, DPM; VARSAT2D, 2DCHEM	2D, Richards (2× for DPM), CDE (2× for DPM), f.-o. transfer of water and solute	Allaire et al. (2002a,b)
AC	MIM, DPM, SPM; HYDRUS-1D	1D, Darcy (2× for DPM), CDE (2× for DPM), f.-o. transfer of solute	Zhang et al. (2004) (AC[4])
AC, UC	MIM; Single Spherical Aggregate Model (SSA)	1D, steady state, MIM, Fickian diffusion transfer, analytical solution	Cote et al. (1999)
UC	MIM; CXTFIT	1D, steady state, MIM, f.-o. transfer, analytical solution. Also: CDE, method of moments	Vanderborght et al. (1997) (UC[2]); Langner et al. (1999), Kamra et al. (2001), Ersahin et al. (2002), Seo and Lee (2005), Stagnitti et al. (2000, 2001)
UC	MIM; CXTFIT and MODFLOW/MT3D, DFM; FRACTRAN	1D, steady-state, saturated, MIM (CXTFIT), 2D-Darcy, CDE-matrix, cubic law, CDE-fracture (FRACTRAN); 3D-Darcy, CDE (MODFLOW/MT3D)	Jørgensen et al. (2004)
UC	SPM, MIM, STM; n.a.	1D, steady-state, model comparison	Vanderborght et al. (2002, 2006)
UC	DPM; n.a.	1D, steady-state, Darcy (2×), CDE (2×)	Schwartz et al. (2000)
UC	DP-MIM; 2-MIM	1D, steady-state, CDE (2×) mobile–mobile–immobile model, transfer	Kjaergaard et al. (2004)
UC	TPM; CXTFIT + PF	1D, steady-state, saturated, CDE (matrix), Poiseuille (macropore, laminar), Manning (macropore, turbulent)	Perret et al. (2000)
UC	DFM (MIM); boundary layer (BL) model	1D, steady-state, kinematic wave advective transport in macropore, diffusion equation for solute transfer	Wallach and Parlange (1998, 2000)
UC	MIM, DPM (TPM); MURT	1D, CDE (2×), first-order solute transfer with embedded small-scale structure	Gwo et al. (1998)
UC	Temporal moments analysis (TMA)	1D, steady-state, statistical integral functions of dimensionless BTC moments	Stagnitti et al. (2000), Kamra et al. (2001)
UC	DPM, SPM; MACRO	1D, Richards, CDE (matrix); kinematic wave, convection (macropores)	Kätterer et al. (2001), Logsdon et al. (2002), Larsbo et al. (2005), Larsbo and Jarvis (2006)[b]
UC	MIM, DPM, SPM; HYDRUS-1D	1D, Richards (1–2×), CDE (1–2×), f.-o. transfer of water and solute	Köhne et al. (2004a,b)
Scale II			
S	SPM; n.a.	3D, Richards, CDE, stochastic or deterministic description of spatially distributed hydraulic properties	Vogel et al. 2006
S	DPM, SPM; MACRO	1D, Richards, CDE (matrix); kinematic wave, convection (macropores)	Merdun and Quisenberry (2004)
S	Multiscale random cascade; Universal Multifractal Model (UM)	1D, no flow model, random cascade with multifractal spatial distribution of hydraulic conductivity, e.g. to analyze spatial dye distribution pattern	Olsson et al. (2002, 2007)
S,P,F	MIM, DPM, SPM; HYDRUS-2D	2D, Richards (2×), CDE (2×), f.-o. transfer of water and solute	Abbasi et al. (2003) (P), (2004) (F); Haws et al. (2004) (S2), (2005) (F); Köhne and Gerke (2005) (P); Köhne et al. (2006a) (F)
P	Drainage network construction; n.a.	2D, no flow model, reconstruction of paths connecting similar scaling factors of water retention curves.	Deurer et al. (2003)
F	SPM, STM, MIM; n.a.	3D, Richards or Darcy-Buckingham, CDE, stochastic or deterministic spatially distributed properties	Vanderborght et al. (2006) (UC[2])
F	MIM, DPM, SPM; HYDRUS-1D	1D, Richards (1–2×), CDE (1–2×), first-order transfer of water and solute	Gerke and Köhne (2004), Köhne et al. (2006a,b)
F	DPM; SWAP + PF component	1D, Richards, CDE (matrix); empirical mass balance and routing of water and solute (cracks), first-order water and diffusive solute transfer; dynamic shrinkage cracks	Van Dam (2000)
F	DPM; (sub)surface trigger model	0D, steady-state, two-domain capacity model for preferential flow and solute advection	Kohler et al. (2003)
F	Convective lognormal transfer function; n.a.	1D and 2D, no flow, convective lognormal transfer function	Gaur et al. (2006)
F	DPM; M-2D	2D Richards and CDE (matrix); 1D kinematic wave and convection (macropores)	Abbaspour et al. (2001)
Scale III			
H	SPM (distributed, DEM based); Hill-Vi	i) 1D Vertical unsaturated (preferential) flow. Mixing within cells, advection between unsaturated and saturated zones and between cells; ii) quasi-3D, saturated lateral flow routing between grid cells	Weiler and McDonnell (2006) (H[b]), (2007); McGuire et al. (2007)

(continued on next page)

Table 3 (*continued*)

Scale	Model type; name	Model dimension; flow state; description	Authors of model application
Scale III			
W, P	DFM; FRAC3DVS	3D-Richards, CDE (matrix), modified 2D-Richards and CDE (parallel fractures)	Van Der Hoven et al. (2002) (W[d])
W	DPM[a] (distributed); WEC-C	3D Darcy, 2D overland, 1D vertical PF: Richards (2×) + advection (2×)	Croton and Barry, 2001 (W[b])
W	SPM (distributed, DEM-and GIS-based); CATFLOW	2D Richards' and CDE; bimodal $K(h)$ with linear increase above threshold represents macropore fraction; 1D Saint Venant for surface runoff and drainage.	Zehe et al. (2001)
W	DPM[a] (distributed); Mike She/Daisy + MACRO	3D Darcy, 2D overland , 1D channels, 1D PF: Richards, CDE (matrix); kinematic wave, advection (macropores)	Christiansen et al. (2004)

Scale I: artificial or repacked soil column (AC), undisturbed column (UC), lysimeter (L); scale II: soil profile (S), plot (P), hillslope (H), field (F); scale III: watershed (W), river basin (R); PF — preferential flow, SPM (DPM, TPM) — single (dual, triple) permeability model, MIM — mobile immobile model, KW (KDW) — kinematic (dispersive) wave model, f — fast flow region, m — matrix, f.-o. (s.-o.) — first- (second-) order, SCM — stochastic convective model, STM — stream tube model, DFM — discrete fracture model, pdf — probability density function.

[a]In the vadose zone part of the model, [b]"synthetic, virtual soil"; assumed (or statistically generated) model parameters, [c]municipal waste sample, [d]foamed zeolite/iron pellet material.

equilibration on solute transport in the vadose zone (Flühler et al., 1996), a common theme was the effect of the solute mass transfer term on PNE transport simulations. Steady-state flow conditions are considered below. First, findings of some classical studies are reported.

4.1.1.1. Solute transfer term. For well-defined, uniformly sized and shaped matrix spheres and rectangular blocks or hollow cylindrical macropores, exact solutions of solute diffusion based on Fick's second law can be employed as their transfer term in the MIM. For such geometries, equivalent shape factors were derived for use in a first-order rate model of mass transfer as a zero-dimensional approximation of the diffusion equation (e.g., Rao et al., 1980a,b; >van Genuchten and Dalton, 1986). The MIM with first-order transfer term is less accurate for transport in spherically aggregated systems than for transport through cylindrical macropores (van Genuchten and Dalton, 1986), and for conditions of severe PNE in spherically aggregated systems may be even less accurate than the CDE with an equivalent dispersivity that lumps the effects of intra-aggregate diffusion (Parker and Valocchi, 1986). Moreover, analytical solutions for the first-order mass transfer term and Fick's second law were compared to the cyclic solute transfer into and out of an immobile region. Limitations of the first-order concept were found for short time periods with insufficient equilibration between phases of inward and outward diffusion (Griffioen, 1998).

Young and Ball (1997) compared the effects of input boundaries and other experimental conditions for soil columns with cylindrical macropores vs. spherical aggregates. The fitted first-order rate solute mass transfer coefficient, α, in the MIM systematically varied with column conditions and asymptotically approached a constant value only for large dimensionless time. The concept of a non-dimensional residence time scale was useful in characterizing these variations, particularly for cylindrical macropore geometry (Young and Ball, 1997).

In accordance with this result, the examination of published MIM analyses of laboratory column data sets revealed a dominant trend of linear variation of α with residence time and solute advective velocity (Griffioen et al., 1998; Maraqa, 2001; Haggerty et al., 2004).

Alternatively to the first-order model, exact diffusion equations were utilized in MIM for studying lateral solute diffusion. A spherical aggregate model based on analytical solutions of the radial form of Fick's second diffusion law was used to estimate the effects of intra-aggregate concentration gradients on solute transport through aggregated soil columns. The model could be used to explain the transfer mechanisms that increased leaching by 20% in an intermittent (as compared to continuous) flow regime (Cote et al., 1999). In a cylindrical macropore system, a MIM with diffusion equation could be used to determine the effective diffusion coefficient of soil surrounding a sand filled macropore (Young and Ball, 1998).

4.1.2. Constructed soil columns, transient flow

4.1.2.1. Solute transfer. Ahuja et al. (1995) used RZWQM to study transport of a surface-applied Br^- tracer under rainfall in a soil column with a 3-mm artificial macropore. A good match of the rapid Br^- breakthrough resulted when the macropore water was assumed to mix with only 0.5 mm of matrix solution, revealing restricted solute transfer through the compacted macropore walls (Ahuja et al., 1995).

Cey et al. (2006) applied 3D DFM HydroGeoSphere (Therrien et al., 2005) to simulate the effect of lateral mass transfer on infiltration and tracer transport into a hypothetical soil column containing a vertical fracture. Rapid water transfer restricted PF and model results were more sensitive to the matrix hydraulic conductivity than to fracture properties (Cey et al., 2006). The above examples show two extremes; highly restricted transfer with rapid breakthrough (Ahuja et al., 1995) vs. fast transfer resulting in restricted PF and high sensitivity of model results to soil matrix properties (Cey et al., 2006).

Since lateral solute transfer is now governed by the interplay of advective (water transfer driven) and diffusive components, scaling rules derived for steady-state conditions will not generally apply for transient flow conditions. Mass transfer under dynamic conditions, with changing directions to and from matrix, might even compensate some of the bias of the first-order term introduced for steady-state conditions, although this potential effect has not been analyzed yet. At least in view of other uncertainties, the inherent inaccuracy of the first-order term may be tolerable from a practical point of view (Jarvis, 2007). However, improved shape factors (see below, Section 4.1.3) and second-order terms (Köhne et al., 2004a) were also tested.

4.1.2.2. Macropore arrangement and its conceptualization. The effect of varying the continuity and tortuosity of (artificial) macropores on measured tracer transport and transient flow through soil columns was analyzed by Allaire et al. (2002a) using the Richards flow model VARSAT2D (Nieber et al., 1994) and CDE based 2DCHEM (Nieber and Misra, 1995). Both models were coupled. The PF region in 2DCHEM was deactivated; instead, the known macropore structure and matrix were directly implemented in the 2D model grid. Simulations showed that surface connected macropores favoured the rapid transport of solutes applied in solution, but delayed solute transport if the solute was initially incorporated in the soil. The latter result was confirmed repeatedly also for field conditions (e.g., Larsson and Jarvis, 1999a,b; Jarvis, 2007). Furthermore, several tortuous macropores could be lumped into a single straight effective macropore for simulations of the effluent tracer BTC. The importance of macropore continuity and tortuosity increased with solute retardation (Allaire et al., 2002a).

In a companion paper, Allaire et al. (2002b) further compared the 2D model explicitly describing the macropore geometry with models considering various simplifications, such as a single straight 'effective' macropore, an SPM with bimodal hydraulic functions, and a DPM with the first-order transfer term. The DPM was the best of the three simplified approaches for predicting breakthrough curves during steady-state flow. For transient flow (infiltration), differences between the BTCs predicted by the different approaches strongly decreased due to physical equilibrium caused by adevctive lateral solute transfer (Allaire et al., 2002b).

The acceleration of solute transport by macropores reported above and in many other studies is environmentally critical only when surface applied agrochemicals are exposed to a rain storm following shortly after their application to the soil surface. For other conditions, PF bypassing the matrix may even slow down vertical leaching of solutes residing (and being 'protected') in the matrix (Larsson et al., 1999b; Köhne et al., 2006b; Jarvis, 2007).

4.1.3. Intact soil columns, steady-state flow

Studies discussed in this section addressed the questions how natural soil structure can be represented in a two-region model, what are the effects of different saturations on PNE tracer transport at steady-state flow, what controls parameter identifiability in two-region models, and which alternative (other than two-region) model approaches can be used.

4.1.3.1. Embedding soil structure into shape factors. As mentioned above, the first-order transfer rate coefficient usually contains a shape factor representing some idealized geometry (e.g. spheres, cylinders). However, natural soils have a more complex structure. Gwo et al. (1998) embedded the aggregate scale structure and pore scale hydrodynamics into the first-order mass transfer term by integrating solute fluxes over mixed-size parallel slab matrix blocks using closed form solutions of diffusion equations. Their time-independent shape factor was calculated by summing up solute fluxes over matrix blocks of different categories (shape, size, water content and effective diffusion coefficient) with different probabilities. The transfer term was implemented into a DPM version of MURT (Gwo et al., 1995). Forward simulations of tracer BTCs from an aggregated saprolite soil column were conducted, assuming aggregates grouped into 6 size categories of 0.3 to 18 mm radii, with different observed frequencies. The BTCs could be predicted for different transport velocities. Intra-region dispersivities were an order of magnitude smaller than those obtained by fitting a DFM or MIM to the BTC (Gwo et al., 1998).

A multi-rate solute transfer model was developed by Haggerty and Gorelick (1995). A MIM with this transfer model yielded significantly improved simulations of measured BTCs in unsaturated soil columns as compared to a MIM with first-order transfer. The assumed lognormal distribution of transfer rates represented different time scales of diffusion and required one additional parameter (Haggerty and Gorelick, 1998).

The surface-area-to-volume ratio of the matrix can be closely linked to shape factors (Gerke and van Genuchten, 1996). Diffusion into the matrix of a dual-porosity soil was described by an equivalent cylinder system which preserves the surface-area-to-volume ratio as derived from image analysis of soil thin sections (Rappoldt and Verhagen, 1999).

The software package CXTFIT (Toride et al., 1999) with analytical solutions for MIM and inverse parameter estimation procedures was frequently used to describe tracer miscible displacement experiments, as for example in the following studies also described below (Vanderborght et al., 1997, 2000; Langner et al., 1999; Wallach and Parlange, 2000; Kamra et al., 2001; Ersahin et al., 2002; Seo and Lee, 2005).

4.1.3.2. Parameter identifiability in two-region models. The MIM mathematically degenerates to the CDE when concentrations in mobile and immobile zones are at equilibrium (Parker and Valocchi, 1986). For such conditions BTCs described by CDE and MIM become identical (Langner et al., 1999). The success of parameter identification may depend on different factors. The extent of PNE directly affects the identifiability of MIM parameters, as Vanderborght et al. (1997) demonstrated by analyzing asymptotic parameter confidence intervals derived from sensitivity matrices. The MIM parameter identification depends on the solute application mode (pulse input is better than continuous input), soil structure type (macropores are better than aggregates), and types of data errors (constant coefficient of variation is better than constant variance) (Vanderborght et al., 1997).

Similarly, the extent of PNE also affects the identifiability of DPM parameters. Physically unrealistic fitted values of the solute transfer coefficient and the fraction of sorption sites were obtained for Br effluent BTCs from structured clay soil columns. The almost similar performance of fitted CDE and DPM suggested that the transport regime was close to equilibrium. Response surface analysis was used to analyze the problems in inverse parameter identification (Schwartz et al., 2000).

Comegna et al. (2001) conducted seventeen leaching experiments with three intact soil columns under both saturated and unsaturated conditions. A CDE, MIM, and CLT (convective lognormal transfer function) models all gave indistinguishable results when applied to fit the effluent BTCs, even those with an asymmetrical shape. However, fitted values of the retardation factor $R<1$ in the CDE might suggest the presence of PNE, in particular since the MIM could be used with $R=1$ without decrease in goodness-of-fit (Comegna et al., 2001).

Moreover, non-local techniques such as TDR for measuring resident solute concentrations usually fail to reveal PNE. Therefore, flux concentrations of effluent BTCs were mostly used to estimate PNE related model parameters (Vanderborght et al., 2000). On the other hand, the MIM parameters for the nitrate BTC measured with a vertical TDR probe were purportedly similar to those obtained from the effluent BTC (Seo and Lee, 2005). This could indicate that vertical TDR probes may be a suitable tool to sense a mixture of resident and flux concentrations in vertical PF paths and matrix.

4.1.3.3. Saturation dependence of PNE. Ersahin et al. (2002) analyzed bromide effluent BTCs obtained from columns collected from different horizons of a silt loam soil. For saturated flow conditions, columns with larger macroporosity had larger values of pore water velocity, dispersivity, and diffusive mass exchange rate coefficient, but smaller values of the mobile water fraction than more homogeneous columns. These differences were reduced with decreasing matric heads between 0 and −5 cm, with fairly constant values between −5 cm and −10 cm. Thus, macropore flow and PNE, as reflected by asymmetric BTCs, disappeared with decreasing saturation (Ersahin et al., 2002). Similarly, the PNE of structured soil columns disappeared below the −5 cm matric head, where CDE and MIM simulations of tracer (tritiated water and pentafluorobenzoic acid) BTCs became alike (Langner et al., 1999).

Similar observations were made also by Pot et al. (2005), who needed increasingly complex models (from CDE, MIM, to DP-MIM; all as part of HYDRUS-1D) to describe tracer displacement column experiments for increasing fluxes and saturations, and by Schwartz et al. (2000), who could describe tracer breakthrough at −10 cm pressure head using the CDE, while at −5 and 0 cm the fit improved when using a DPM.

Evidence from these and numerous other studies suggests that PNE in soils is generated at pressure heads larger than −10 to −6 cm in macropores of 'equivalent cylindrical diameter' larger than 0.3–0.5 mm (Jarvis, 2007).

4.1.3.4. Alternative model approaches. Different other approaches, such as temporal moments, DFM, CLT (see above, Comegna et al., 2001), and three-region approaches were compared with MIM to assess their individual weaknesses and strengths. Stagnitti et al. (2000) found the statistical temporal moments method to be as suitable as MIM for describing tracer BTCs, while in another study, the moment method failed to capture preferential peak concentrations of BTCs (Kamra et al., 2001).

A DFM that uses a KW equation to describe advective transport in the fracture, i.e., a first-order process to describe dissolved chemical transfer through a boundary layer and a diffusion equation to describe the solute diffusion into the matrix, was successfully fitted to BTCs measured for fractured clay–till soils. By contrast, a corresponding model with local concentration equilibrium at the fracture–matrix interface (without boundary layer) underestimated chemical breakthrough during early stages (Wallach and Parlange, 2000).

Jørgensen et al. (2004) compared the suitability of SPM, MIM and DFM approaches for assessing Br transport in saturated columns (0.5 m × 0.5 m) of clayey till with vertical fractures and root channel biopores. Simulations using FRACTRAN (Sudicky and McLaren, 1992) in the SPM mode and MIM (Toride et al., 1999) required fitted parameter values that varied several orders of magnitude for different flow rates. Only the FRACTRAN in DFM mode could be applied with constant calibrated values of fracture spacing and aperture to approximate the observed BTCs during alternating flow rates (Jørgensen et al., 2004).

DFM can simulate PNE transport at steady-state flow more accurately than MIM with first-order transfer. For fractured soils with aggregate coatings or organic linings, DFM with boundary layer further enhances the accuracy.

For some soils, assuming two regions is not sufficient for transport characterization. A MIM and a DP-MIM (Fig. 1) were compared for analyzing BTCs of tritiated water and colloids, measured in 54 soil columns with different clay contents under steady-state flow conditions with a 5-cm applied suction (Kjaergaard et al., 2004). Only the three-region model could fit all BTCs and supported the hypothesis that there were two mobile and a diffusion domain in the clayey soils (Kjaergaard et al., 2004). The physical reality of postulated flow regions could be supported by combining X-ray CT scans of macropores with measured tracer breakthrough to parameterize a three-mobile-region model (Perret et al., 2000).

4.1.4. Intact soil columns and lysimeters, transient flow

Modelling preferential solute transport under transient flow conditions requires a large number of model parameters that usually cannot be measured. Some model packages including MACRO (e.g., Jarvis, 1994; Larsbo et al., 2005) and HYDRUS-1D (Šimůnek et al., 2003, 2008a,b) have increasingly come to be used with inverse and uncertainty estimation techniques. Applications of these two models are reported below as examples of other related approaches.

MACRO was coupled with the inverse parameter estimation program 'Sequential Uncertainty Fitting', SUFI (Abbaspour et al., 1997), for analyzing Br and Cl BTCs observed on intact soil columns. The SUFI procedure starts with user-defined prior uncertainty domains for parameters to be fitted. Each uncertainty domain is divided into equidistant strata and parameter values are defined by the first moment of each stratum (Roulier and Jarvis, 2003). MACRO-SUFI was applied to study the effects of the initial saturation and the mode of tracer application (in soil vs. in irrigation water). A single fitted parameter set could be applied to simulate almost all elution curves, irrespective of initial saturation and application mode, reasonably well (Kätterer et al., 2001).

The 'Generalized Likelihood Uncertainty Estimation' (GLUE) (Beven and Binley, 1992; Beven and Freer, 2001) was also coupled with MACRO (Larsbo et al., 2005). The motivating theory behind GLUE assumes that all models and measurements are to some extent wrong, so that many parameter sets may represent the observations equally well. The GLUE procedure runs multiple MACRO simulations with sets of parameters sampled from predefined distributions within specified uncertainty limits. Simulations with model efficiencies larger than the predefined threshold value are considered acceptable. For a Cl displacement experiment, water percolation and Cl concentrations in both the effluent and final resident profiles of intact soil columns were accurately reproduced by MACRO with GLUE. Model efficiencies for acceptable parameter sets can be used to recognize, which parameters can be conditioned (or inversely identified) in a particular transport experiment (Larsbo et al., 2005).

Yet another inverse approach, called 'parameter identification method using the localisation of information' (PIMLI) (Vrugt et al., 2001), was applied with MACRO to test the effect of the column design on parameter identification (Larsbo and Jarvis, 2006). PIMLI identifies the subsets of data that contain the most information for the identification of each parameter. These subsets are then used iteratively to reduce parameter uncertainty. The synthetic data representing transport experiments on clay and loam soil columns contained sufficient information to reduce uncertainty in the mass transfer coefficient, saturated matrix hydraulic conductivity and dispersivity. The information content was largest in the early stages of the experiments, such that high time-resolution measurements were suggested for the first irrigation(s) following solute application (Larsbo and Jarvis, 2006).

HYDRUS-1D (Šimůnek et al., 2005) includes the Levenberg–Marquardt method for multi-objective inverse parameter estimation. Köhne et al. (2004b) used the MIM in HYDRUS-1D for inverse simulation of pressure heads, water contents, water outflow, and Br breakthrough in the effluent of six aggregated soil columns with different initial water contents subject to intermittent irrigations. Inverse identification of the required MIM parameters was fairly successful, except for the saturated water contents in mobile and immobile regions, which were highly correlated (Köhne et al., 2004b). Using DPM and DP-MIM in HYDRUS-1D could further improve model performance (Köhne et al., 2006a).

4.1.5. Summary for scale I (column, lysimeter)

For soil columns with artificial PF paths, and for natural undisturbed columns, the effects of experimental conditions, including the tracer application mode, flow mode (steady-state and transient), saturation, flow velocity, and residence time, on PNE, solute transfer, and parameter identifiability, were investigated. Steady-state and transient flow conditions required analytical and numerical solutions of PNE models, respectively. Steady-state flow conditions were frequently used for investigations at scale I (less so at larger scales). Model approaches used for both steady-state and transient conditions involved SPM, MIM, DPM, three-domain models, DFM, and statistical method of moments. DFM (with boundary layer) was the most accurate approach for steady-state conditions. SPM, even if provided with bimodal hydraulic functions, generally failed to simulate preferential solute transport. The success of the other approaches often increased with the level of model complexity, such as adding another flow domain, but this approach was rarely independently confirmed using CT. However, model success was also dependent on the parameter identifiability, as related to experimental conditions, types of data available, and the extent of PNE. Significant PNE was found for pressure heads above −10 cm (see also Jarvis, 2007). For transient flow, different procedures were applied for inverse estimation of parameters and uncertainty (Levenberg–Marquardt, GLUE, SUFI, PIMLI). Fitted parameters that are assumed to reflect constant soil properties in the conceptual model, such as mobile region, dispersivity, and transfer rate coefficient, may vary with flow rate and saturation if the model does not accurately describe involved processes. However, it is not clear to which extent these effects are relevant for transient vadose zone conditions. Structure related shape factors were suggested as a means of including complex natural soil structure in the solute transfer term.

4.2. Scale II (plot, field)

Deterministic 1D and 2D two-region models with single parameter sets were evaluated (compare Section 3.2.1.) and compared with corresponding one-region continuum approaches at the plot and field scales to assess the relevance of considering PF. Similar as for soil water flow studies at this scale (Section 3.2.), tile-drained fields were often involved in Sections 4.2.1–4.2.2. A few alternative model applications are described in Section 4.2.3.

4.2.1. 1D two-region approaches

4.2.1.1. Capillary PF models

Gerke and Köhne (2004) compared the DPM of Gerke and van Genuchten (1993) with a corresponding Richards-CDE based SPM (Vogel et al., 1996) for simulating tile drain discharge and Br concentrations in a 0.5-ha field with a structured loamy soil. The SPM and DPM described the discharge hydrograph similarly well, but only the DPM reproduced the Br breakthrough pattern. Hence, the DPM was deemed to be capable of capturing relevant PNE transport mechanisms during transient PF (Gerke and Köhne, 2004).

Köhne et al. (2006a) studied the feasibility of the inverse (Levenberg–Marquardt) identification of DPM parameters from a drainage hydrograph. The DPM implemented in HYDRUS-1D was used to fit hydraulic and transport parameters, either sequentially or simultaneously, using observed tile-drainage hydrographs and Br concentrations. Only the simultaneous fitting procedure was successful in describing Br breakthrough. From these and lab-scale results it was inferred that a hydrograph alone is insufficient for the inverse identification of soil hydraulic DPM parameters (Köhne et al., 2006a).

4.2.1.2. Gravity-driven PF models

Merdun and Quisenberry (2004) applied MACRO for simulating drainage flow and Cl transport in structured loamy sand soil profiles at 6 plots. MACRO outperformed a one-domain model that over-predicted water content and Cl concentrations with depth and time (Merdun and Quisenberry, 2004).

Similarly, MACRO gave a satisfactory description of soil water contents, drainflow, and resident and flux concentrations of Br observed in the 0.4-ha Lanna field site (Sweden) with structured clay soil, when all available data were used in the inverse parameter estimation (Larsbo and Jarvis, 2005).

4.2.1.3. Empirical PF models

Kohler et al. (2003) successfully applied their Surface and Subsurface Trigger DPM to explain tile discharge and Br flux BTCs observed in a 2-year tile-drained field experiment near Zurich, Switzerland. Results were used to assess matrix and PF components. Results suggested that 73% of Br was exported in PF paths within 2 years after the applications, and that the storage volume of the top soil had to be filled first to trigger PF in the subsoil (Kohler et al., 2003).

The empirical DPM SWAP was applied to field observations of soil water contents and Br concentrations in cracked clay soil, groundwater levels, and fluxes of tile-drainage water and Br during 572 days. A single domain model was not able to mimic the field-average water flow and solute transport. Routing of PF through cracks considerably improved the SWAP simulation of water contents and Br leaching to groundwater. However, the observed retardation and high dispersion of Br could still not be reproduced (van Dam, 2000).

4.2.2. 2D two-region approaches

While lateral water flow, e.g., in tile drained soils, can be captured as a sink term to a 1D model, lateral solute transport is problematic to model in a 1D framework. Hence, 2D two-region models were developed and evaluated to model field scale preferential solute transport.

Kohler et al. (2001) evaluated their combined 2D/1D DPM named M-2D. For a test data set based on a column experiment with preferential tracer transport, M-2D performed much better than SWMS-2D (its 2D component), and similarly to MACRO (its 1D component). A field tracer test was conducted in a tile-drained municipal waste site in Switzerland with measurements of tile discharge and electric conductivity. M-2D approximated experimental observations during both calibration and validation time periods (Kohler et al., 2001).

Modified HYDRUS-2D model was used in several studies comparing 2D-approaches based on SPM, MIM, or DPM for simulating PNE tracer transport in agricultural fields (e.g., Abbasi et al., 2003, 2004; Köhne and Gerke, 2005; Haws and Rao, 2004; Haws et al., 2005; Köhne et al., 2006b).

Abbasi et al. (2003) compared the 2D MIM and SPM using inverse simulations of water flow and tracer movement at a sandy loam field plot with furrows (3 m×3 m) in Phoenix, Arizona. The topography of the furrow cross-sections was considered explicitly in the numerical grid. Water contents, infiltration rates, and resident solute concentrations were used in the objective function. The similarity of the inverse 2D MIM and SPM results led to the conclusion that equilibrium transport prevailed at this field site (Abbasi et al., 2003). Regarding this conclusion one must caution that the spatial resolution of concentration data sampled at discrete positions is usually insufficient to detect PF (e.g., Kasteel et al., 2005). Abbasi et al. (2004) extended investigations to a field (115 m×20 m) covering several sets of three furrows each. Fitted hydraulic and transport parameters at the field scale were more variable, yet relatively similar on average, to the plot-scale study (Abbasi et al., 2004).

At the Bokhorst loamy soil site in Northern Germany, the final Br distribution derived from multiple soil cores in a soil profile was described almost similarly by the 2D MIM and 2D SPM, and thus reflected matrix transport. Br concentration peaks in tile effluent were only reproduced by the 2D MIM and thus revealed preferential displacement. Nonequilibrium was highly dynamic; it was limited to periods of high intensity rainfall suggesting a threshold process (Köhne and Gerke, 2005). Simulations were further refined using a 2D DPM (S_2D_DUAL, Vogel et al., 2000) and improved boundary conditions (Gerke et al., 2007).

At the Infeld site in North-West Germany, the observed rapid Br effluent breakthrough at low concentrations could also only be simulated using the 2D MIM approach. Simulation results suggested that over 60% of the surface applied Br was immobilized by transfer into the stagnant soil water region, and that the 2D flow field induced by tile drains enhanced Br dispersion (Köhne et al., 2006b).

Haws et al. (2005) simulated water and solute fluxes from subsurface drains of two macroporous silty clay loam plots (48.5 m×60 m) in agricultural fields in West Lafayette, Indiana. The 2D SPM and 2D MIM simulations matched the drainage hydrographs, but not solute breakthrough. They concluded that a hydrograph fit cannot be used as evidence of the physical meaning of model parameters (Haws et al., 2005). Similar conclusions were drawn in other studies (Gerke and Köhne, 2004; Köhne et al., 2006a; McGuire et al., 2007).

Another advantage of a 2D (vs. 1D) model is that soil spatial heterogeneity can be considered in addition to PF (Vogel et al., 2000).

4.2.3. Alternative 2D or 3D approaches: fractals, networks, scaleway

Olsson et al. (2002) used a multiscaling approach to describe the spatial distribution pattern of PF as visualized by dye tracer tests. The Universal Multifractal Model was based on the random cascade process with a multifractal spatial distribution of hydraulic conductivity. The model reproduced hydraulic conductivity functions and key features in the observed dye distribution, and was suitable for describing PF pathways (Olsson et al., 2002).

The drainage network approach by Deurer et al. (2003) was conceived for PF in hydrophobic sandy soils, but might apply to structured soils as well. In three profiles (10 m length, 1.4 m depth) at a gleyic sandy Podsol in Northern Germany, flow path networks were identified by spatially linking the similar linear scaling factors of water retention curves. The network fraction decreased exponentially with depth, corresponding to a strengthening of PF. Assuming 1D piston-flow through the effective transport volume of the network, the arrival times of the tracer peak concentration observed in a Br tracer experiment could be predicted (Deurer et al., 2003).

This concept of an exponential decrease of the macropore fraction with depth was incorporated by Haws and Rao (2004) in the MIM within HYDRUS-2D (Šimůnek et al., 1999) to investigate the effect on solute transport. A strong effect on the BTC peak position in the soil profile was found, but the effluent BTC could be approximated by a depth-averaged macropore fraction (Haws and Rao, 2004). However, a more general approach would be to relate transport parameters in models to the texture and observed morphological features of soil horizons, instead of making a priori assumptions (see discussion in Jarvis, 2007).

Vogel and Roth (2003) proposed the ‘scaleway’ approach, which is a conceptual framework for the explicit consideration of the structure (heterogeneity) in modelling flow and transport at the scale of interest, where microscopic heterogeneities are averaged and replaced by effective descriptions. The scaleway needs a representation of the structure, a process model at the scale of interest, and corresponding effective material properties. In an application of the scaleway, the solute BTC in an undisturbed soil (Orthic Luvisol) column could be accurately predicted based on computer tomography (CT) and network modelling. This approach seems to be more physically based than, e.g., statistical multi-fractal approaches.

Vogel et al. (2006) explored the use of three different approaches to incorporating the relevant 3D-structure of a given soil for modelling flow and transport. They performed a Brilliant Blue dye tracer experiment in a 2.5 m^2 plot. A 3D hydraulic structure model (Fig. 1k) was constructed by combining (i) direct measurement of soil layers, (ii) correlated anisotropic random fields to describe heterogeneity within soil layers, and (iii) a genetic earthworm burrow model based on percolation theory. This hydraulic structure model was implemented into a 3D Richards-CDE model. The direct simulation of dye distribution in the soil compared reasonably well with observations. Hence, a rough representation of the soil structure and hydraulic properties may be sufficient to approximate tracer transport (Vogel et al., 2006).

Gaur et al. (2006) applied 1D and 2D convective lognormal transfer functions (CLT) to evaluate whether concentrations in tile drain outflow can be predicted from spatially variable soil surface transport properties determined by multiple (45) TDR probes that recorded the electrical conductivity (EC) in a field plot (14 m × 14 m) above 1.1-m deep tile drains. Additionally, the EC of the tile drainage flow was measured. Only predictions made using 2D (not 1D) CLT reproduced the observations. It was concluded that the surface solute transport properties determined by TDR can be combined with a 2D CLT transport model to make reasonable predictions of tile flux concentrations for the soil that is relatively uniform with depth (Gaur et al., 2006).

4.2.4. Summary for scale II (plot, field)

The predominant flow mode at scale II was transient both in the experimental and model analyses. Contrasting approaches were applied to conceptualize the effects of soil structure on tracer transport. Two-region (MIM, DPM) models are the most widely used approaches at scale II for characterizing preferential flow and transport. Increasingly, these PF models also consider transient variably-saturated flow in 2D, and are complimented with options for inverse parameter identification.

Model comparisons again demonstrated better performance of the two-region as compared to continuum models for describing conservative solute transport, while relatively similar results were obtained for the hydrograph. Furthermore, two-region approaches were used for better understanding of PF effects on solute transport. For example, preferential flow and transport can be triggered in the subsoil, and bypass flow may under certain conditions reduce tracer leaching. Alternative models included stochastic multifractal description, drainage network reconstruction, a 3D hydraulic structure model, drainage network reconstruction, and 1D or 2D convective lognormal transfer function models (CLT).

Non-ideal transport may be caused by the soil structure (micro-heterogeneity), the spatial variability (macro-heterogeneity) of soil properties, or a combination of both. A decade ago Feyen et al. (1998) suggested that modelling concepts for micro- and macro-heterogeneity should be coupled in one overall mathematical framework, since both types of heterogeneity are relevant for agricultural and environmental management. A few recent approaches have presented possible avenues in that direction (e.g., Vogel and Roth, 2003; Vogel et al., 2006).

4.3. Scale III (hillslope, catchment area)

Weiler and McDonnell (2004) proposed virtual (numerical) experiments to explore first-order controls on complex hillslope hydrology. They applied their quasi-3D, distributed hillslope model HillVi. Results suggested that high drainable porosity increased channeling of lateral flow at the soil–bedrock interface. Weiler and McDonnell (2007) additionally provided HillVi with lateral PF pipes. The model was applied to predict runoff, and transport of Br applied as a line source tracer, at a hillslope in the Maimai catchment area (New Zealand). Pipe network configuration had a much higher influence on Br movement than on runoff. Although some tracer was rapidly displaced through the pipe system, the bulk of tracer remained in the soil. This explained the (frequently) observed dominance of pre-event water in runoff events (Weiler and McDonnell, 2007).

Van Der Hoven et al. (2002) also investigated how to separate storm hydrographs into pre-event water and storm-event water, but based on the $\delta^{18}O$ natural isotope content. While this is a problem relevant to catchment scale, the experimental analysis of $\delta^{18}O$ values in shallow groundwater focussed on one multi-port well in fractured saprolite. The DPM FRAC3DVS (Therrien and Sudicky, 1996) was applied to analyze the data. Results suggested that the $\delta^{18}O$ content was not constant over the course of a storm event, and that most of the matrix water was isolated from flow in fractures. It was concluded that quantitative hydrograph separation using isotope techniques is not possible in most situations (Van Der Hoven et al., 2002).

Christiansen et al. (2004) coupled the 1D vadose zone — 3D groundwater hydrology model MIKE SHE operating at the catchment scale (Refsgaard and Storm, 1995), the 1D agro-ecosystem model DAISY (Hansen et al., 1991; Abrahamsen and Hansen, 2000), and a 1D DPM based on MACRO (Jarvis, 1994). The resulting model was used to study the effects of macropore flow on stream discharge and groundwater levels in a small catchment (1.5 km^2) in Zealand, Denmark. Simulation results indicated an erratic and spatially variable nature of macropore flow depending in a complex manner on soil characteristics and hydrological regime. Three years of tracer applications were not sufficient to estimate average leaching to groundwater, or to predict which rainfall events generated the highest macropore flows and leaching of tracer or pesticide. Model validation for macropore flow was hampered by the lack of experimental data at this scale (Christiansen et al., 2004).

Zehe et al. (2001) assembled the CATFLOW model based on experimental studies carried out in the Weiherbach catchment (6.3 km^2) in S-W Germany. CATFLOW integrates three different spatial scales (plot, field, catchment) and three different time scales for BCs and parameters (constant, season, event). On different space-time scales, water and solute dynamics are dominated by different processes: On the *plot scale*, flow and transport are approximated as 1D vertical processes; infiltration (PF) and solute adsorption are relevant on the event time scale and evapotranspiration, interception, and degradation dominate seasonal scale processes. On the *hillslope (field) scale*, runoff generation and infiltration (PF) govern water balance on the event time scale, while on the seasonal scale, precipitation variation, land use changes and soil fauna activity affect water and solute displacement. On the *catchment area scale*, water dynamics depend on topography, land use, and geometry of the drainage channels. The CATFLOW simulation results were compared to observations of water contents, infiltration rates, and flow patterns from the Weiherbach catchment. Again, model validation was limited by the non-representativeness of point data for larger areas (Zehe et al., 2001).

According to Weiler and McDonnell (2007), conceptualization and parameterization of the effects of lateral PF on hillslope hydrology currently represent the greatest challenge in modelling PF processes at the catchment scale.

4.3.1. Summary for scale III (catchment area)

Model analysis of PF effects on solute transport is a formidable task at the catchment area scale, where PF is one of the many processes governing solute delivery from the catchment to the stream. Although several models have been developed in the past decade, only a few of them were applied to realistic situations, and consequently the effect of PF on catchment hydrology and water quality is still not well understood. A principal problem at the catchment area scale is to assess and validate models based on experimental data: at the lower scale end, point measurements (e.g., soil water contents) are measured at the model sub-grid scale and are usually scarce, while at the upper scale end, observations of the integrated catchment response (e.g., river hydrograph) are above the process scale represented in the model. The validity of quantitative hydrograph separation using mass balance techniques for natural isotopes (e.g., $\delta^{18}O$) was questioned.

Among approaches that may be worth further exploration are nested experimental tracer tests and models (plot-field-catchment area), advanced measurement methods including geophysical and remote sensing techniques, and stochastic simulations and virtual experiments.

5. Discussion

5.1. Comparing strengths and weaknesses of two-region models

While several other promising PF approaches are being developed and tested as described in previous sections, the two-region concept is now by far the most widely applied approach to model PF in agricultural structured soils and thus our discussion here will be focused on them.

Capillary, gravity driven, and empirical two-region approaches differ in their complexity and model assumptions. For model applications the question arises if any one of these approaches can be considered superior to others, if only for a particular purpose. It would be desirable to base a model ranking on an evaluation of reported model applications. However,, this is difficult for three reasons: (i) most studies, with few exceptions (e.g., Logsdon et al., 2002; Aden and Diekkrüger, 2000; van Dam, 2000), only report overall success of a model after calibration, (ii) calibration methods and criteria for the reported success differ between the studies, and (iii) there are too few studies on model comparison based on the same data set and furthermore, of those studies appear to be of limited significance due to the uncertainty introduced by different model users. Thus in the following we discuss models' strengths and weaknesses on the basis of underlying model conceptualizations.

The following general features should be included in a DPM to be applicable to a range of conditions: 1) physically based process descriptions with parameters (at least in principle) linked to measurable quantities, 2) options to consider soil heterogeneity; at least soil layering, 3) transient flow in both domains at a high time resolution, 4) program-controlled, domain-separated, dynamic upper BCs to conceptualize the dynamics of water and solute infiltration into matrix and macropores at different rainfall rates, and other surface processes such as surface runoff and evaporation, 5) lower BCs realistically representing different situations including variable groundwater table, gravity flow, or drainage, 6) bi-directional, rate-limited transfer of water and solute, 7) capability of initiating PF at both surface and subsurface layers, 8) inverse and/or uncertainty estimation procedures, and 9) a range of options required to describe agricultural management options (e.g., regarding tillage, drainage, crops, mechanical compaction, different modes of chemical application) and different agrochemicals.

Even recent empirical capacity PF models meet several of these requirements, e.g., the relatively comprehensive COUP model (Jansson and Karlberg, 2004). However, capacity and routing approaches cannot describe PF for moderate rainfall rates below the saturated soil hydraulic conductivity. Furthermore, they usually do not consider subsurface initiation of PF at layer boundaries. Exceptions are the Subsurface Trigger model (Kohler et al., 2003) and the 1D stochastic IN^{3}M model (Weiler, 2005). Lateral transfer between the macropores and matrix is usually considered by layer wise mass balance without physically based equation. Thus, empirical PF approaches are suited for specific conditions only.

Currently, DPM approaches which fulfil most or all of the above requirements, are either based on gravity-driven macropore flow, such as MACRO (Larsbo et al., 2005), or assume that flow through the soil structure network can be described similarly as flow in a porous-medium, which is controlled by capillary and gravity forces, such as HYDRUS-1D (Šimůnek et al., 2008a,b) (Section 2.).

The most obvious conceptual difference between PF models concerns the question of an adequate description of bypass flow. A recent detailed discussion (Jarvis, 2007) of flow processes in macropores based on findings from various experimental and modelling studies concludes that PNE in soils is usually generated at pressure heads larger than −10 to −6 cm in macropores of a minimum equivalent diameter of 0.3–0.5 mm, and that gravity is the main driving force for PF. This suggests the choice of a pressure head of −10 cm separating matrix from macropore regions. While DARCY's law might also be accepted as a reasonable quasi-empirical model of macropore flow, the gravity-driven KW approach should give a (quasi-) physically based description of PF in structured soils (Jarvis, 2007). However, the advantage of using a gravity-driven over a capillary approach might not be universally given for all structured soils, conditions, and scales, as discussed below.

5.1.1. Microscopic scale

Current conceptual ideas about pore scale fracture flow processes appear to be largely based on laboratory studies performed with low permeability fracture surfaces (e.g., Tokunaga et al., 1997; Or and Tuller, 2000; Ghezzehei, 2004; Dragila and Weisbrod, 2004). These processes may not be representative of flow in many macroporous soils, where the larger matrix hydraulic conductivity causes faster water exchange and slows down macroporous flow. Even conclusions based on laboratory studies of macropore flow velocities in soils might not tell the whole story. For technical reasons, these studies were mostly performed for macropore diameters of several mm, where film flow was observed for unsaturated conditions and full flow for saturated conditions (e.g., Logsdon, 1995; Ghodrati et al., 1999), compliant with the gravity-driven KW approach. Indeed, Jarvis (2007) alerted against the possible bias introduced by the high irrigation intensities of larger than 10 mm h^{-1} used in the majority (60%) of the reviewed studies. By contrast, the lower end of PF, i.e., flow in smaller macropores or at lower flux rates induced by natural rainfall rates, is less well known. Significant preferential movement of NO_3 occurred in soil columns with earthworm burrows and root channels even at fluxes as low as 1.5% and 5% of the saturated hydraulic conductivity of the column, respectively (Li and Ghodrati, 1995, 1997). A CT based analysis of flow in macropores in three different soils yielded, although uncertain, pressure head values of −40, −14, and −10 cm for the transition between matrix and macropore flow (Mori et al., 1999). Measurements of diffusion into soil aggregates, and of BTCs in soil columns at different pressure heads (0 to −15 cm), suggested that with decreasing saturation, transport through smaller macropores continues at reduced degree of nonequilibrium (Haws et al., 2004). In conclusion, limited experimental evidence does not seem to prove that capillary forces cannot affect PF in soils.

5.1.2. Macroscopic scale

Two-region models are not exactly based on pore scale physics, but are macroscopic approaches (e.g., Jarvis, 2007). Thus, the question is not how accurately the model represents pore-scale processes, but how the model, supplied with 'efficient' parameters, can describe some average processes and phenomena. This question has been evaluated for STM (stream tube models) and CDE models (Vanderborght et al., 2006), but not for DPM. At least some qualitative considerations are attempted here. A characteristic of large soil columns, and in particular of an entire field, is the heterogeneity of soil texture and structure, where the latter includes fractures, root channels, and earthworm burrows of differing size, directions and continuity, causing different transport patterns in different soil horizons (e.g., Kulli et al., 2003). The effect of structure variability on the hydraulic conductivity function of the fast flow region can be represented in the gravity-driven KW approach by the kinematic exponent, and in the capillary PF approach, e.g., by the tortuosity factor in van Genuchten's (1980) model. Additionally, solute transport within such a heterogeneous PF network will be highly dispersive due to different local flow velocities (Vogel et al., 2006). Dispersion within the structure network can be represented in the capillary PF approach by using the CDE, while in the gravity driven approach entirely by mass exchange with the matrix. Moreover, a 2D capillary PF approach can be used to simulate lateral PF.

5.1.3. Conclusion

In summary, the capillary PF approach appears to be a useful concept for the purpose of describing bypass flow and PNE transport in soils with hierarchical structure, e.g., structured loamy soils. For clay soils with open cracks, or soils with large continuous macropores at the surface, the gravity-driven approach is better suited. Both gravity-driven and capillary PF approaches are considered to be largely physically-based for applications at the soil column scale, but semi-physical when used at the field scale, where the structure of many soils is too complex to be classified into exclusively non-capillary or capillary pores.

2D approaches were also used for both, the gravity-driven approach (M-2D; Kohler et al., 2001) and the capillary PF approach (e.g., HYDRUS (2D/3D) (Šimůnek et al., 2006b). Adding a description of spatial variability of soil properties to 1D (Booltink, 1994; Weiler, 2005), or 2D (Vogel et al., 2000) approaches would strengthen the physical basis for field scale applications. However, model applications involve striking a balance between parameterization effort and the required accuracy. In that practical sense, both approaches have been demonstrated to be useful in applications for different soils at scales from the column to the field.

5.2. Benefits and limitations of inverse methods

In the past decade, the use of inverse techniques has become a standard procedure to facilitate DPM applications (e.g., Section 4.1.4). Roulier and Jarvis (2003) argued that inverse estimation procedures provide objective, reproducible, and unambiguous results, providing the problem is well posed. This statement is certainly accurate, but, unfortunately, in DPM field applications the problem is often not well posed. This was shown in simulations of preferential water flow based on scarce data (Section 3). When modelling solute transport, several parameters related to flow, transport, and lateral transfer have to be estimated. To better constrain the inverse solution, many studies considered different types of flow and transport data simultaneously in the objective function. Two problems arise in this approach. (i) Data obtained at a local scale, such as tensiometer or TDR readings and concentration profiles, can only be represented in a spatially resolved (2D or 3D) model. Additionally, in a two-region approach which data type corresponds to which domain must be defined. (ii) Automated weighting procedures cannot fully compensate the effects associated with different data sets (with divergent mean and variance, and variable data 'noise') in the objective function. Some manual fine tuning of weights or other factors is usually needed for optimal results (e.g., Scorza jr. et al. 2007, Köhne et al., 2004b). The uncertainty involved in inverse procedures has given incentive to develop uncertainty estimation procedures such as GLUE and SUFI (Section 4.1.4.) and multi-objective optimization algorithms (Wöhling et al., 2008). Although inverse techniques have become very helpful tools, their further improvements are needed for their objective use. This may include focussing on subsets of data which are most important (Larsbo and Jarvis, 2006), or recognizing significant patterns.

6. Independent parameter determination

6.1. Scale I (soil column, lysimeter)

6.1.1. CT, MRI, and tracer techniques

The X-ray CT scan technique was used to analyze the large pore structure, while MRI was used to assess both the 3D pore space arrangement and flow velocity. Progress was achieved by combining techniques including CT, MRI and solute or dye tracer breakthrough to describe relations between porosity distribution, flow paths, and soil hydraulic and transport properties (Table 4).

Several studies evaluated the usefulness of hydrogeophysical methods for independent model parameterization. For example, Císlerová and Votrubová (2002) studied CT identification of local high porosity zones. The CT scans identified macropores in the undisturbed sandy loam. However, CT was insufficient to separate fast and slow flow domains in the fine sand columns, where this supplementary information could be provided by MRI and dye tracer distribution.

Perret et al. (1999) used CT for establishing the 3D arrangement of the geometry and topology of macropores. Perret et al. (2000) further applied CT for real-time examination of Potassium iodide movement through the macropores and matrix of a large undisturbed soil column. Based on the CT results, distribution density functions of macropore size and hydraulic radius were assigned to the two (laminar and turbulent flow) macropore subdomains of a three-mobile-region model. The breakthrough curves measured in the matrix domain were fitted using the CDE (Toride et al., 1999). Measured potassium iodide breakthrough in the macropores could then be predicted using the three-region model (Perret et al., 2000).

Vanderborght et al. (2002) explained concentration patterns of two adsorbing fluorescent dye tracers in a soil column with the large 3D pore structure derived from CT imaging. Additionally, breakthrough of Cl and dyes in a column, and dye adsorption in batch tests, were measured. The data were used together to explain the success of 1D MIM, and the failure of SPM and STM, in describing observed breakthrough of Cl and dyes. In MIM, the fraction of sorption sites in the mobile region could only be inversely identified when the mobile region fraction was independently determined from dye concentration maps. It was concluded that the combination of effluent breakthrough data and patterns of local concentrations in the soil provided the information to identify governing transport processes and model parameters (Vanderborght et al., 2002).

Kasteel et al. (2000) used the CT derived X-ray absorption coefficients, which are related to local bulk densities, as a proxy for hydraulic properties for two different flow regions. The relative hydraulic conductivity, water retention, and connectivity functions of the fast flow region were derived using a network model based on 3D CT images of local densities in a small undisturbed soil block (resolution 0.04 mm) (Vogel and Roth, 1998, 2001). These relative local hydraulic functions were scaled using measured absolute values, and were then implemented as spatially distributed functions in the 3D Richards-CDE model SWMS_3D (Šimůnek et al. 1995) for direct simulation of observed tracer breakthrough. Tracer breakthrough was predicted nearly as accurately as when MIM was fitted to the data (Kasteel et al., 2002). Javaux et al. (2006) also considered the CT-derived 3D microscale spatial distribution of hydraulic parameters in SWMS_3D to reproduce tracer BTCs in a sandy monolith.

The MRI technique was applied for assessing the 3D pore space arrangement in a repacked soil column with sand and gravel layers (Baumann et al., 2000). Flow velocities within PF paths and the diffusion coefficient were measured using MRI at a spatial resolution of $1.3 \times 1.3 \times 5$ mm^3 and at time intervals of 10 s for different applied flow velocities between 0.9 and 40 cm/min (Baumann et al., 2000).

A 3D hydraulic tomography method, i.e., inversion of travel times of transient pressure propagation measured in hydraulic and pneumatic tests in an unsaturated fractured sandstone core, was used to estimate the local hydraulic property distribution and diffusivity (Brauchler et al., 2003). Other hydrogeophysical techniques include self potential, ground penetrating radar, induced polarization, electrical resistivity tomography, electromagnetic induction, microwave radiometry, and time domain reflectometry (TDR) (Vereecken et al., 2004).

6.1.2. Imbibition and diffusion in soil aggregates to estimate mass transfer

Gerke and Köhne (2002) and Köhne et al. (2002a) investigated the effect of aggregate coatings as an approximate estimator for water and solute mass transfer parameters. Aggregate coatings were found to reduce water imbibition (Gerke and Köhne, 2002) and solute diffusion (Köhne et al., 2002a) into soil aggregates by approximately an order of magnitude. Results were only a rough approximation of transfer mechanisms in the field, since the effects of PF in natural soil position (partial wetting, hydraulic inactivity) could not be considered.

6.1.3. Soil hydraulic measurements to derive PF-related soil hydraulic functions

Hydraulic measurements on soil cores with PF paths were fitted with bimodal (Coppola, 2000), dual-permeability (Köhne et al., 2002b), and multi-modal (Priesack and Durner, 2006) hydraulic functions. Physically-based dual-domain hydraulic functions were developed by upscaling modelled liquid configurations in partially saturated fractured porous media, and were fitted to hydraulic data (Tuller and Or, 2002; Or and Tuller, 2003). However, uncertainty will be introduced for all such models by purely hydraulic fitting (e.g., Köhne et al., 2006a).

6.1.4. Column experiments and pedotransfer functions

Methods for estimating parameters of the steady-state MIM involve parameter fitting under optimized experimental conditions. For example, the immobile water content, the solute transfer coefficient, and the dispersion coefficient were estimated using a vertical TDR method that was used to analyze resident tracer breakthrough (Lee et al., 2000, 2001, 2002). A flow interruption technique for intermittent solute leaching through soil columns was applied to estimate the mobile and immobile water contents, and the mass transfer coefficient (Ilsemann et al., 2002). Several other studies have reported similar analyses for the MIM (Shaw et al., 2000; Ersahin et al., 2002).

Table 4
Independent parameter determination for preferential flow and transport models

Scale	Parameters (model)	Determination approach	Authors
Scale I			
Ped	Diffusion coefficients of 'interface PF-matrix'	Diffusion measurement with intact and surface-removed aggregates	Köhne et al. (2002a,b)
Ped	Hydraulic conductivity of 'interface PF-matrix'	Flow measurement with intact and surface-removed aggregates	Gerke and Köhne (2002)
AC	3D pore space arrangement (of repacked soil), diffusion coefficient, flow velocity in preferential flow paths	3D estimation of pore space ($0.5 \times 0.5 \times 0.5$ mm^3 resolution), flow velocity ($1.3 \times 1.3 \times 5$ mm^3, time resolution 10 s), and diffusion coefficient in soil column using magnetic resonance tomography (MRT).	Baumann et al. (2000)
AC	3D equivalent preferential flow paths in fractured porous media (flow-transport DFM)	Genetic-based computer search algorithm to invert scale-representative equivalent fracture networks using BTCs and structural information	Gwo, (2001)
UC	3D pore space arrangement and its relation to flow paths	3D visualization of pore space (resolution 0.5–1 mm) of dry undisturbed soil samples using X-ray computer tomography (CT) images	Císlerová and Votrubová et al. (2002)
UC	3D pore network and water retention and permeability functions of fractured porous media	3D scan (resolution 0.04 mm) of a small undisturbed soil block using X-ray CT images, reconstruction of 3D pore network connectivity defined by the 3D Euler number.	Vogel and Roth (2001)
UC	3D structure of local-scale (1-mm) hydraulic properties	3D scan (resolution 0.5–1 mm) of an undisturbed soil column using X-ray computer tomography (CT) images. X-ray absorption coefficients related to local ρ and $K(\theta)$ were used in a 3D flow-transport model to predict observed tracer breakthrough	Kasteel et al. (2000)
UC	3D geometry and topology[a] of macropore networks (conceptual model of four flow domains)	CAT scans and image analysis of four large (800 mm length, 77 mm diam.) intact dry soil columns, 3D reconstruction of macropore networks, solute breakthrough analysis in space and time	(Perret et al., 1999, 2000)
UC	3D hydraulic property distribution and diffusivity	3D hydraulic tomography by inversion of travel times of transient pressure propagation and diffusivity measured in hydraulic and pneumatic tests in unsaturated fractured sandstone core.	Brauchler et al. (2003)
UC	3D-CT scan: model parameters, e.g., θ_m, α, λ_m, f (MIM), soil properties that control transport of adsorbing solutes	3D X-ray CT scans combined with solute breakthrough analysis for Cl and two fluorescent tracers in three intact soil columns: 3D pore structure, spatial dye distribution after leaching, modelling (CDE, MIM, STM)	Vanderborght et al. (2002)
UC	$\theta(h)$ dependency on micromorphology	Micromorphology of soil pore structure was studied on thin sections. Hydraulic properties were studied on undisturbed soil samples using constant head and multi-step outflow methods	Kodešová et al. (2006)
UC	θ_{im}, α, Dm (MIM)	TDR method analyzing resident concentration breakthrough of salt solution applied at step input.	(Lee et al., 2000, 2001, 2002)
UC	θ_{im}, α (MIM)	Intermittent solute leaching and flow interruption for soil columns	Ilsemann et al. (2002)
UC	$\theta(h)$ and $K(h)$ parameters (bimodal, multimodal, or DPM)	Parameter fitting of retention and hydraulic conductivity measurements on soil cores	(Othmer et al., 1991; Ross and Smettem, 1993; Durner, 1994; Coppola, 2000; Köhne et al., 2002b, Priesack and Durner, 2006)
UC	$\theta(h)$ and $K(h)$ of fractured porous media	Upscaling of modelled liquid configurations of cross-sections of partially saturated fractured porous soil or rock	(Tuller and Or, 2002; Or and Tuller, 2003)
UC	K_s, K_b h_b, n^*, D_v, d, mixing depth (MACRO DPM)	Statistical variation in MACRO parameters calibrated in simulations of water flow and solute transport between 3 soils with different textures	Merdun and Quisenberry (2004)
UC	K_s, θ_{im}, α (MIM)	Statistical variation in MIM parameters with tracer BTCs, dye stained soil fractions, and soil properties	(Vervoort et al., 1999; Shaw et al., 2000)
UC	d (MACRO DPM)	Statistical variation in the mass transfer coefficient for BTCs in 33 intact soil columns representing a range of soil textures and organic matter contents	Jarvis et al. (2007)
Scale II			
S	K_s, θ_{im}, α (MIM)	Infiltration of tracers with a tension infiltrometer; infiltration rates and tracer concentrations in soil samples below infiltration area	Alletto et al. (2006)
S	K, θ, θ_{im}, α (MIM)	Tracer infiltration with disk infiltrometer for a series of applied tensions; infiltration rates and tracer concentrations in soil samples.	Casey et al. (1998)
S	K_s, K_b h_b, n^*, θ_{sma}, d (DPM; MACRO)	Measurements of wet-end K–θ–h relations and results from image analysis for Mollisols (sandy loam to silty clay).	Logsdon (2002)

(continued on next page)

Table 4 (*continued*)

Scale	Parameters (model)	Determination approach	Authors
Scale II			
S	Pore spectrum parameters of a model of flow in equivalent capillary tubes	Tracer mass flux breakthrough patterns under transient unsaturated flow conditions were used to quantify equivalent pore spectra of macropore-type preferential flow pathways.	Kung et al. (2006)
S	3D distribution and correlation structure of BB concentration	Brilliant blue dye (BB) and bromide (Br) infiltration, image analysis (BB), soil sampling (Br).	Kasteel et al. (2005)
S	Statistical categorization of soil layers with flow regimes ranging from homogeneous to PF	cluster analysis is used to derive different transport mechanisms from multiple dye staining patterns in vertical soil profiles from 25 plots	Kulli et al. (2003)
P	K_s, λ_c, θ_{im}/θ, α (MIM)	Dripper-TDR field method measuring point source chemical movement	Al-Jabri et al. (2002, 2006)
F (UC)	A (MIM), k (two-site model)	empirical relation between α and residence time, and between k and pore water velocity	Maraqa (2001)
F-UC	Correlation between α and residence time (and v)	Comparison of results from 316 solute transport experiments in 35 articles, plus own experiments.	Haggerty et al., 2004
F-UC	Relation between λ_L, v, $K(\theta)$ and soil properties controlling lateral mixing regime	Review of multiple leaching experiments in a range of soils, linking λ_L to soil morphological features, and analysis of depth dependence of $\lambda_L\lambda_L$.	(Vanderborght et al., 1997; Vanderborght et al., 2002; Vanderborght and Vereecken, 2007)

Scale I: soil aggregate (Ped), artificial or repacked soil column (AC), undisturbed column (UC), lysimeter (L); scale II: soil profile (S), plot (P), field (F); acronyms: computer tomography (CT), saturated hydraulic conductivity (K_s), macroscopic capillary length (lambda(c)), immobile water fraction (θ_{im}/θ), mass exchange coefficient (α) and dispersion coefficient (D_m) or dispersivity (λ), saturated and boundary hydraulic conductivities (K_s and K_b), boundary head between macropore and micropore domains (h_b), exponent (n*) of relation between K and water content (θ), macropore fraction (θ_{sma}), and half spacing (d) between equivalent parallel fractures, dispersivity (D_v), ρ bulk density, $K(\theta)$ hydraulic conductivity function.
[a]density, position, length, volume, wall area, hydraulic radius, tortuosity, inclination, number of branches.

Moreover, it was suggested to direct more effort to derive pedotransfer functions accounting for soil structure effects (Merdun and Quisenberry, 2004; Pachepsky et al., 2006). The relation between lateral dispersivity, unsaturated hydraulic conductivity, and soil morphological properties, was studied in tracer experiments carried out in columns and field plots with key Belgian soil types (Vanderborght et al., 2001). Procedures based on combined analyses of soil hydraulic measurements, tracer BTCs, and dye stained soil fractions have been proposed to relate MIM parameters to soil clay content (Shaw et al., 2000), or to soil structure formation (Vervoort et al., 1999). The latter authors conceptually linked soil structure development to the predominant state of saturation of a soil horizon as depending on its location in pedon and landscape position. Jarvis et al. (2007) analyzed the variation of the mass transfer coefficient, d, in the DPM MACRO (Larsbo et al., 2005) as a function of the geometric mean particle size and the organic matter content. First, SUFI (Abbaspour et al., 1997) was utilized to identify d for chloride BTCs observed for 33 intact topsoil columns sampled from 3 different field sites. Multiple linear regression suggested that approximately 50–60% of the variation in d can be explained by two easily measured fundamental soil properties (Jarvis et al., 2007).

6.1.5. Summary on independent parameter determination, scale I (soil column, lysimeter)

Techniques using CT, MRI and tracers were applied either independently or simultaneously, and their results were implemented in 3D models to predict PF and transport as functions of the soil structure. Additionally, laboratory procedures were tested to determine two-region model parameters, involving region-related water contents, hydraulic functions, dispersivities, and parameters related to water and solute transfer. The attempt was also made to statistically relate such model parameters to basic soil properties. More research on the links between soil structure and solute transport is needed to support the use of DPM for management purposes. Despite the progress that has been made, the value of these measurement techniques for field conditions has yet to be proven.

6.2. Scale II (plot, field)

6.2.1. Field techniques to estimate two-region model parameters

Jaynes et al. (1995) and Clothier et al. (1995) suggested field methods for estimating MIM parameters. Using disk infiltrometers, several tracers were infiltrated into the soil and their distributions below the disk were then measured in soil samples taken after infiltration. A single applied tension yielded one set of values for the hydraulic conductivity, K, the total water content, θ, the water content of the immobile zone, θ_{im}, and the mass transfer coefficient, α (Alletto et al., 2006), while several tracer infiltrations conducted at different tensions produced K, θ, θ_{im}, and α, as near-saturated functions of the matric potential (Casey et al., 1998). Al-Jabri et al. (2002) suggested a variant of this method for simultaneous measurement at multiple locations based on point-source dripper lines. They used the Wooding's (1968) analytical solution for estimating K and the capillary length, λ_c, while MIM transport parameters were estimated according to Jaynes et al. (1995). In an application with several dripping rates, the saturated hydraulic conductivity, K_s, the ratio of the immobile and the saturated water content, θ_{im}/θ_s, and λ_c were similar, while α was different compared to corresponding values derived using the disk infiltrometer (Al-Jabri et al., 2002). The dripper line method was further extended by the installation of a TDR probe beneath each infiltration spot to also measure solute breakthrough to yield the dispersion coefficient in the mobile region (Al-Jabri et al., 2006). The accuracy of these methods depends on the validity of the underlying assumptions, including piston movement, a constant mobile water concentration equal to the infiltration concentration, and negligible solute exchange with the immobile region (e.g., Alletto et al., 2006).

Different lab and field methods, including disk infiltrometer techniques, image analysis, and lab water desorption tests, were suggested for independent estimation of hydraulic parameters for the MACRO model (Logsdon, 2002).

Kung et al. (2006) analyzed the field-scale flux breakthrough patterns of four conservative tracers sequentially applied during a 10-h infiltration. They used a six-parameter capillary tube model to quantify equivalent macropore spectra of PF pathways. Results indicated that pathways with increasingly larger equivalent pore radii became hydraulically active during soil wetting in the course of infiltration (Kung et al., 2006).

6.2.2. Dye tracer experiments

Dye tracer experiments were often used to infer solute transport processes, test model hypotheses, and estimate model parameters. Absolute dye concentrations can now be calculated with reasonable accuracy (Forrer et al., 2000). The spatial resolution (1 mm) of dye concentration maps of horizontal soil cross-sections can be interpolated to infer the 3D distribution of the absolute dye concentrations, sufficient for characterizing soil structure effects on transport (Kasteel et al., 2005). An important step towards more realistic transport simulations will be to better understand and describe flow at interfaces or transitions between different soil layers. Kulli et al. (2003) analyzed dye staining patterns in vertical soil profiles of 25 plots at 8 sites. Zones of concentrating flow (attractor zone), spreading flow (dispersion zone), and no alteration of flow regime (transmission zone) were identified. A cluster analysis yielded classification of seven different transport regimes ranging from homogeneous to preferential. Soils with a similar sequence of soil horizons did not always display a similar sequence of certain transport features. Other factors, such as gradual or sharp textural layer boundaries, may invoke different flow patterns in similar underlying horizons (Kulli et al., 2003).

6.2.3. Combinations of techniques: CT, dye tracing, and breakthrough curves

Hydrogeophysical, dye tracing, and breakthrough analyses alone have their conceptual limitations. Using CT allows accurate macropore network reconstruction, but may not show which part of the network will be hydraulically active. A dye tracing profile reveals the macropore network engaged in dye transport, but adsorption may restrict the dye from part of the water flow network. A leaching BTC provides a temporal pattern of the system response, but does not allow conclusions about the internal spatial transport dynamics. However, complementary information on transport processes may be gained when using multiple techniques simultaneously (e.g., Vanderborght et al., 2002). Hydrogeophysical field monitoring techniques (e.g. Gaur et al., 2006) should be further developed for integrating both spatial and temporal flow-transport observations. The processes linking ‘form’ (soil structure, layering, interfaces) and ‘functions’ (solute transport patterns) should be further explored utilizing tracer experiments (Kulli et al., 2003; Kasteel et al., 2005) for structure based prediction of solute transport (Vervoort et al., 1999; Vogel et al., 2006).

7. Conclusions

7.1. Water flow

When only simulating water flow (without solute transport) through structured soils, equilibrium single-domain models often yield results similar to two-domain (MIM, DPM) approaches, unless dynamic shrinkage cracks are present. An inverse parameter identification of two-domain PF parameters based on water outflow information suffers from non-uniqueness problems. At the field scale, stochastic methods were utilized to analyze the effects of heterogeneity, parameter uncertainty, and PF triggering processes, such as precipitation rate and depth thresholds. The effects of domain-separated boundary conditions in two flow-domain approaches need to be considered. Complex catchment area hydrology was described with distributed hydrological unit models or 3D coupled numerical bypass flow models, some of which considered lateral PF as important hillslope flow process. Threshold values of rainfall intensities apparently also trigger PF at the catchment area scale. However, scarce data and model dicretization beyond the process scale make flow model validation at the catchment area scale complicated.

7.2. Solute transport

Model approaches applied at the field, and smaller, scales often involved SPM, MIM, and DPM or three-domain models that can consider natural transient flow conditions. Alternative approaches included DFM, 1D stream tube models (STM), method of moments, a stochastic multifractal approach, a drainage network reconstruction, a structure-based 3D flow transport model and hillslope models with lateral pipe flow. Several inverse, uncertainty, and parameter sensitivity quantification concepts were tested. Inverse schemes need to make better use of localized important information or patterns. Parameter uncertainty is a useful tool, e.g., for including effects of spatial variability in field-scale predictions. A systematic correlation between parameter sensitivities and soil properties (for experiments conducted at a particular scale) might further help to identify important parameters for certain conditions. The effects of experimental conditions on PNE solute transport were investigated at the column scale. 2D, inverse, and transient two-region models were applied at the field scale. At the catchment area scale, evaluating PF effects on solute transport has only just begun and will require innovative approaches, such as combining tracer tests and modelling at nested scales (plot-field-catchment area), geophysical and remote sensing techniques, stochastic methods and scenario analysis (‘virtual experiments’).

7.3. Parameter determination

Progress includes both, independent determination of parameters of two-region models, and the development of other predictive model approaches. At the column scale, the combined use of CT, MRI and tracer techniques showed promise in predicting PF and transport as a function of the soil structure. Pedotransfer functions that were statistically related to fundamental soil properties, as proxies of soil structure, may permit a rough prediction of two-region model parameters. Despite the progress made, an independent determination of hydraulic and transport parameters is still very limited because of the simplifying assumptions underlying various methods, and often requires separation of analyzed soil samples from their natural setting. Methods that showed potential for parameter estimation and predictive model capabilities at the field scale included (i) experimental methods, e.g., a combination of the tension infiltration method with tracer applications, an analysis of flux breakthrough patterns of sequentially applied tracers, or combined dye tracer and hydraulic measurements for setting up a 3D soil hydraulic structure model, and (ii) statistical evaluations, e.g., of relations between solute transfer coefficients and the residence time, or classification of transport regimes using cluster analysis of dye concentration spatial patterns in different sequences of soil horizons. Soil structure and its spatial variability need to be jointly considered in modelling at the field and catchment area scales relevant for agricultural and environmental management.

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