Selected HYDRUS modules for modeling subsurface flow and contaminant transport as influenced by biological processes at various scales

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Abstract: A large number of modifications or special modules of the HYDRUS software packages have been developed during the past several years to evaluate the effects of a range of biohydrological processes on subsurface water flow and the transport of various chemicals and contaminants. The objective of this manuscript is to briefly review the different modules that were included, and to present various applications illustrating the effects of biological processes on water flow and solute transport and reactions in variably-saturated media.

Key words: biological processes; contaminant transport; CO_2 production and transport; reactive transport modeling; unsaturated

Introduction

There is increasing evidence that biological processes can have major effects on water flow and the fate and transport of contaminants in the subsurface at various temporal and spatial scales (Seuntjens et al. 2005; Rockwood et al. 2005; amongst others). Like many other models, the HYDRUS software packages (Šimůnek et al. 2008) were originally developed to simulate only water flow and heat and solute transport in soils, and as such ignored many important biological processes. During recent years a large number of modifications or special modules have been included in the HYDRUS models to allow evaluation of possible effects of biological processes on fluid flow and contaminant transport in the subsurface. The objective of this manuscript is to briefly review these modules and to present selected applications of the HYDRUS codes illustrating the effects of biological processes on water flow and solute transport and reactions in soils. However, it is beyond the scope of this paper to provide many details about the modules. Readers are invited to find these details in referenced literature.

HP1 – Coupled HYDRUS-1D and PHREEQC codes

A comprehensive modeling tool in terms of available chemical and biological reactions was recently developed by coupling HYDRUS-1D with the PHREEQC geochemical code of Parkhurst & Appelo (1999). This coupling resulted in the very flexible simulator, HP1, which is an acronym for HYDRUS1D-PHREEQC (Jacques & Šimůnek, 2005; Jacques et al. 2006). The combined code contains modules simulating (1) transient water flow in variably-saturated media, (2) the transport of multiple components, (3) mixed equilibrium/kinetic biogeochemical reactions, and (4) heat transport. HP1 is a significant expansion of the individual HYDRUS-1D and PHREEQC programs by combining and preserving most or all of the features and capabilities of the two codes into a single numerical model. The code uses the Richards equation for variably-saturated flow and advection-dispersion type equations for heat and solute transport. However, the program can now simulate also a broad range of low-temperature biogeochemical reactions in water, the vadose zone and in ground water systems, including interactions with minerals, gases, exchangers, and sorption surfaces, based on thermodynamic equilibrium, kinetics, or mixed equilibrium-kinetic reactions (Šimůnek et al. 2008). Various applications of HP1 were presented by Jacques & Šimůnek (2005) and Jacques et al. (2006, 2008a,b).

There is much evidence in the literature that microorganisms, present throughout the subsurface, can
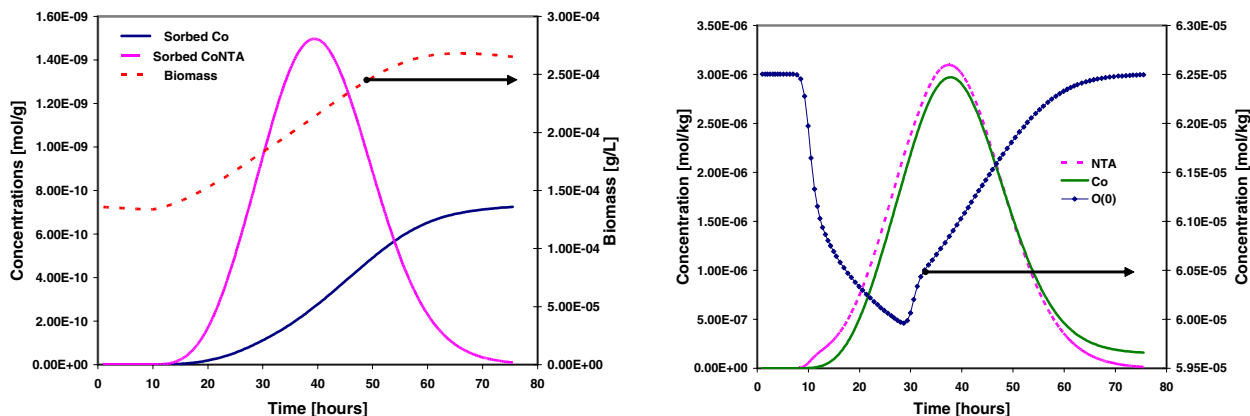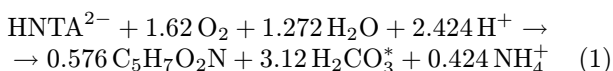
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Fig. 1. Selected output of the HP1 program: a) sorbed concentrations and biomass, and b) total concentration of main components and oxygen.

catalyze many chemical reactions and thus may affect the fate of both organic and inorganic contaminants. Many bacteria can beneficially degrade or immobilize environmental contaminants (e.g., Ginn et al. 2002; Gargiulo et al. 2007b). As an illustration of the use of HP1, we present here its application to a problem first presented by Tebes-Stevens & Valocchi (1998), and also used by Parkhurst & Appelo (1999), involving bacterially mediated degradation of an organic contaminant. This example involves the transport of a pulse of cobalt and nitrilotriacetate (NTA), an organic chelating agent that has been detected along with metals and radionuclides in some contaminated groundwater, into a 10–m long soil column. The example accounts for several interacting chemical processes that are common to many environmental problems: bacterially-mediated degradation of an organic substrate (NTA), bacterial cell growth and decay, kinetic metal sorption (Co^{2+}), and aqueous speciation including metal-ligand complexation ($CoNTA^-$). For this application we assumed that only a single protonated form is biodegradable, which leads to the following overall stoichiometry under aerobic conditions (Tebes-Stevens & Valocchi, 1998):

$$HNTA^{2-} + 1.62\,O_2 + 1.272\,H_2O + 2.424\,H^+ \rightarrow$$
$$\rightarrow 0.576\,C_5H_7O_2N + 3.12\,H_2CO_3^* + 0.424\,NH_4^+ \quad (1)$$

where $C_5H_7O_2N$ is the simple elemental formula for biomass (Rittmann & VanBriesen, 1996) and $H_2CO_3^*$ represents both aqueous CO_2 and H_2CO_3. Equation (1) assumes that electrons are transferred from the donor substrate, $HNTA^{2-}$, to the acceptor, O_2, in order to produce the energy required for synthesis of new biomass. The overall rate of the above biological reaction is described using a Monod-type kinetic model as follows:

$$r = r_r X_m\ \frac{S}{K_S + S}\ \frac{A}{K_A + A} \quad (2)$$

where r [mol/sec/L] and r_r [mol/sec/g biomass] are the actual and maximum reaction rates, respectively, X_m is the biomass [g cells/L], S and A are the concentrations of the electron donor substrate ($HNTA^{2-}$) and the electron acceptor (O_2), respectively, and K_S and K_A are the half-saturation constants for the substrate and the electron acceptor, respectively. The change in biomass is related to $HNTA^{2-}$ degradation rate by a yield coefficient and to the actual biomass by a biomass decay constant.

This example demonstrates several unique capabilities of PHREEQC, and hence also of HP1 which preserved most of the PHREEQC geochemical features. For example, the HP1 code allows users to define species that may not appear in existing geochemical databases (e.g., NTA), as well as include additional kinetic equations, which may be specified using short Basic programs that are read as input (such as in this case the kinetic degradation of NTA, the corresponding growth of biomass, and kinetic sorption of metals and metal-ligand complexes). These features offer users of the program unprecedented flexibility to define their own chemical problems and reactions. Figure 1 presents selected output from HP1 for the final node of the 10m long soil column. Only dissolved oxygen enters the soil column in this saturated flow problem. During the breakthrough of the NTA pulse, oxygen is consumed by the NTA degradation reaction. Although this example involves some sophisticated coupling between chemical and biological processes, it is still limited to one oxygen consuming reaction (NTA degradation), one electron donor (NTA) and one bacterial population.

The particle (bacteria or viruses) transport module

Bioremediation and/or bio-augmentation strategies to clean up recalcitrant chemicals in subsurface environments could be greatly enhanced by the efficient delivery of specialized bacteria to targeted locations in the subsurface (Gargiulo et al. 2007b). At the same time, bacterial contamination of groundwater can be a serious problem that may lead to large outbreaks of waterborne diseases (e.g., Ginn et al. 2002; Reynolds et al. 2004). An ability to accurately predict the fate and transport of pathogenic microorganisms in unsaturated soil environments is therefore needed in efforts to protect surface and ground-
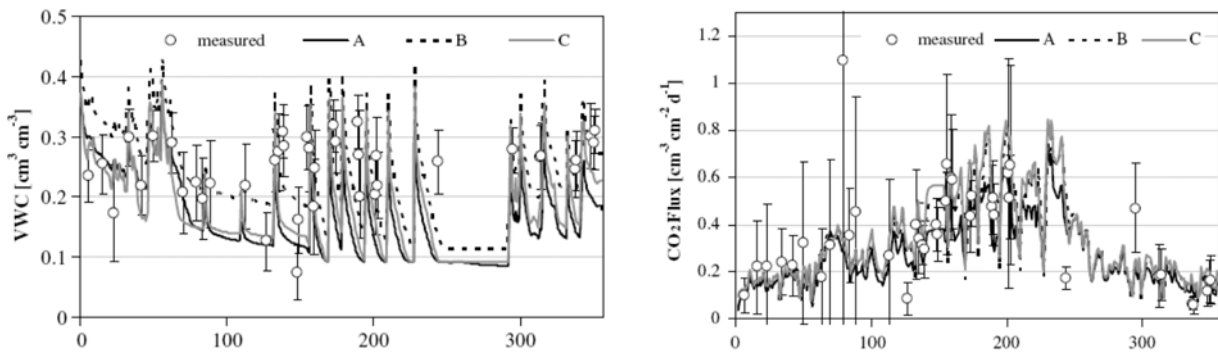
Fig. 2. Measured and simulated volumetric water contents (VWC) and CO_2 fluxes at the experimental field in Davis, California (Buchner et al. 2008).

water resources from contamination (Gargiulo et al. 2007b).

The fate and transport of bacteria in porous media is more complex than that of solutes since bacteria transport is affected by several additional processes, such as filtration, straining, sedimentation, adsorption, and desorption, growth, and decay/death of bacteria. Various modifications of the HYDRUS models have been made to allow the code to simulate these processes involved in the transport of viruses and bacteria. The governing convection-dispersion equation for virus or bacteria transport is similar as the general equation for colloid transport assuming applicability of the attachment/detachment concept:

$$\frac{\partial \theta c}{\partial t} + \rho \frac{\partial s_1}{\partial t} + \rho \frac{\partial s_2}{\partial t} =$$
$$= \frac{\partial}{\partial x}\left(\theta D \frac{\partial c}{\partial x}\right) - \frac{\partial q c}{\partial x} - \mu_w \theta c - \mu_s \rho (s_1 + s_2) \quad (3)$$

where c is the (virus or bacteria) concentration in the aqueous phase, s is the solid phase (virus or bacteria) concentration, the subscripts 1 and 2 represent two kinetic sorption sites, respectively, θ is the water content, ρ is the bulk density, D is the hydrodynamic dispersion coefficient, t is time, x is the distance, and μ_w and μ_s represent inactivation and degradation coefficients in the liquid and solid phases, respectively. The latter two coefficients, when assigned negative values, can also be used to account for bacteria growth (Gargiulo et al. 2007a). Two kinetic sorption sites can be used in Equation (3) to represent various chemical or physical processes. One sorption site, for example, may be used to represent the chemical processes of attachment/detachment, while the other sorption site may represent the physical process of straining. Alternatively, one of the sorption sites could be used to represent attachment/detachment to/from air-water interfaces.

The transport model described above has been successfully used by, among many others, Schijven & Šimůnek (2002) to simulate the transport of viruses at the field scale, and Gargiulo et al. (2007a,b; 2008) to simulate the column-scale transport of hydrophobic (*Rhodococcus rhodochrous*) and hydrophilic (*Deinococcus radiodurans*) bacteria strains in sands of different

matrix grain sizes, at different water saturations, and at different growth stages (metabolically active or stationary phases). With further modifications to account for bacteria aggregation, the model was additionally used by Bradford et al. (2006) to simulate the transport and straining of *E. coli* bacteria at the column scale. Yet further modifications of this module allow consideration of the bacteria- (or colloid-) facilitated solute transport (Šimůnek et al. 2006; Pang & Šimůnek 2007).

The SOILCO$_2$ module of HYDRUS-1D

Carbon dioxide production and oxygen consumption represent important processes resulting from microbiological activity in the soil environment and in the unsaturated zone. Microbiological respiration, combined with plant root respiration and high water contents, can change CO_2 concentrations several orders of magnitude from values that are at equilibrium with the CO_2 content of the atmosphere (about 0.036% at present), up to extreme values of about 20% (Šimůnek & Suarez, 1993). The solubility of many solid phases, such as carbonates and oxihydroxides, significantly changes within this range of CO_2, primarily because changes in soil CO_2 lead to changes in the soil pH for all but acid soils (Šimůnek & Suarez 1994).

The SOILCO2 and UNSATCHEM modules (Šimůnek & Suarez 1993, 1994) of HYDRUS-1D consider the effects of CO_2 producing microbiological activity and CO_2 transport in soils on geochemical transport. We refer to the original codes for detailed descriptions of the many processes that were considered. Suarez & Šimůnek (1993) discussed several examples of seasonal simulations of soil CO_2 concentrations and CO_2 fluxes to the atmosphere for different crops, and compared their results with published field data. The coupled program was obtained by combining various features from SOILCO2 (i.e., water flow, heat transport, and CO_2 transport) and UNSATCHEM (major ion chemistry) within a new module within HYDRUS-1D. The resulting module of HYDRUS-1D has seen several applications recently to lysimeter and field data (e.g., Bauer et al. 2008; Herbst et al. 2008; Buchner et al. 2008). Results from one application by Buchner et al. (2008) are shown in Fig. 2.
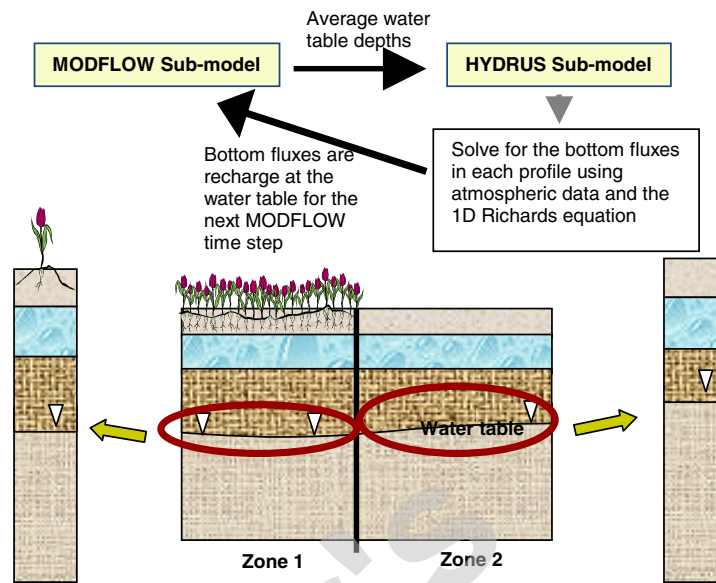
Fig. 3. Schematic description of the coupling procedure for water flow in HYDRUS-MODFLOW.

The HYDRUS module for MODFLOW

Water fluxes in the subsurface of forested and agricultural ecosystems at the field or catchment scale can be highly variable spatially because of several interconnected processes. In addition to natural subsurface heterogeneity, infiltrating water is often distributed unevenly due to above-ground interception and redistribution of rainfall by plant canopies (Sansoulet et al. 2008). Transpiration and evaporation often also vary widely over the landscape as a function of geographical location, slope exposure, plant species, and many other factors. These various processes can have important ecohydrological consequences since they significantly affect groundwater recharge and nutrient leaching.

To be able to address spatially variable saturated-unsaturated hydrological processes at the larger scale, Seo et al. (2007) and Twarakawi et al. (2008) recently developed the HYDRUS package for Modflow 2000 (Harbaugh et al. 2000). By fully incorporating HYDRUS-1D within the widely used MODFLOW program, the resulting HYDRUS package provides MODFLOW with recharge fluxes at the water table, while MODFLOW provides HYDRUS with the position of the groundwater table that is used as its bottom boundary. While the HYDRUS package considers the main processes and factors affecting fluxes in the vadose zone, such as precipitation, infiltration, evaporation, redistribution, capillary rise, root-water uptake, water accumulation at the ground surface, surface runoff, and soil moisture storage, MODFLOW considers all of the groundwater flow processes. The combined software thus dramatically expands capabilities of the two separate modules for application to a wide range of spatial and temporal scales.

Figure 3 schematically shows the invoked coupling of the HYDRUS and MODFLOW models. The ground-water modeling domain for MODFLOW was discretized into grids/blocks using finite-differences as described by Harbaugh et al. (2000). Based on similarities in soil hydrology and topographical characteristics, the discretized MODFLOW domain can be divided into zones that comprise one or more grids of the MODFLOW model. A separate HYDRUS soil profile is then assigned to each of these zones. The approach assumes that the HYDRUS soil profile adequately represents vadose zone flow for the entire zone. The HYDRUS vertical soil profiles are subsequently discretized vertically into finite elements. Since the numerical solution of the Richards equation requires much finer temporal (often as low as few seconds) as well as spatial discretizations than traditional groundwater models, the two models (HYDRUS and MODFLOW) interact (i.e., exchange information about the groundwater recharge rate and the groundwater level) only at the end of each MODFLOW time step, with HYDRUS generally requiring multiple time steps during each MODFLOW time step (Seo et al. 2007). MODFLOW receives the recharge flux from HYDRUS and calculates a new water table depth for a particular time step. A new water table depth is then assigned as the pressure head bottom boundary condition in the HYDRUS package for the next MODFLOW time step (Fig. 3). Twarakavi et al. (2008) recently compared the HYDRUS package to other contemporary modeling approaches and evaluated its performance for three case studies of increasing complexity and spatial and temporal scales. They concluded that the HYDRUS package provided a much more efficient alternative and could account better for vadose zone processes than other existing MODFLOW packages. For large-scale ground water flow problems, the HYDRUS package provides an optimal trade-off between computational effort and accuracy of model simulations of coupled vadose zone – groundwater problems.

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