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Modeling nitrogen transport and transformation in aquifers using a particle-tracking approach

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ABSTRACT

We have integrated multispecies biodegradation and geochemical reactions into an existing particle-tracking code to simulate reactive transport in three-dimensional variably saturated media, with a focus on nitrification and denitrification processes. This new numerical model includes reactive air-phase transport so that gases such as N_2 and CO_2 can be tracked. Although nitrogen biodegradation is the primary problem addressed here, the method presented is also applicable to other reactive multispecies transport problems. We verified the model by comparison with (1) analytical solutions for saturated one- and two-dimensional cases; (2) a finite element model for a one-dimensional unsaturated case; and (3) laboratory observations for a one-dimensional saturated case. Good agreement between the new code and the verification problems is demonstrated. The new model can simulate nitrogen transport and transformation in a heterogeneous permeability field where sharp concentration gradients are present. An example application to nitrogen species biodegradation and transport of a plume emanating from a leaking sewer in a heterogeneous, variably saturated aquifer is presented to illustrate this capability. This example is a novel application of coupling unsaturated/saturated zone transport with nitrogen species biodegradation. The code has the computational advantages of particle-tracking algorithms, including local and global mass conservation and minimal numerical dispersion. We also present new methods for improving particle code efficiency by implementing the concept of tracking surplus/deficit particles and particle recycling in order to control the growth of particle numbers. The new model retains the advantages of the particle tracking approach such as allowing relatively low spatial and temporal resolutions to be used, while incorporating the robustness of grid-based Monod kinetics to simulate biogeochemical reactions.

1. Introduction

Subsurface flow can be an important pathway for transport of land-applied nitrogen to nearby surface water bodies (Bohlke and Denver, 1995; Angier et al., 2005; Green et al., 2008). Numerical simulation can be used as a tool to complement field observations to quantify nitrogen subsurface flow paths and fluxes; however, due to the complexity and heterogeneous natural of biogeochemical reactions, most existing models use simplified representations of kinetics and geology. In this paper, we advance the capability to simulate realistically complex N cycling through development of a new code for solving the equations for reactive nitrogen transport in three-dimensional variably saturated media. While many codes written to simulate nitrogen transport and transformation in the subsurface can be found in the literature, most codes use first-order decay equations (Gusman and Marino, 1999; Frind et al., 1990; Molenat and Gascuel-Odoux, 2002; Kersebaum, 1995), or Monod type kinetic reactions (Iqbal and Hiscock, 2011; Peyrard et al., 2011; Wriedt and Rode, 2006; Kinzelbach et al., 1991; Widdowson et al., 1988). Other approaches include additional geochemical reactions, normally requiring a significant number of parameters (MacQuarrie and Sudicky, 2001; Maggi et al., 2008; Grant and Pattey, 2003). Very few of these codes include multi-species biogeochemical reactions (MacQuarrie and Sudicky, 2001; Maggi et al., 2008; Grant and Pattey, 2003). The advantage of the multispecies approach is that it can account for more processes than zero- or first-order models. For example, the products of denitrification, such as CO_2 and N_2, can be tracked easily in both aqueous and gaseous phases. On the other hand, more physically

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Table 1
Features of existing reactive multispecies transport codes coupled with biodegradation and geochemical reactions.

Reference	Code name	Transport numerical or analytical method	Dimensionality	Saturated/ unsaturated	Transport reactions (single species)	Multispecies reactions
Essaid and Bekins (1997)	BIOMOC	Hybrid – particle tracking for advection, finite-difference for dispersion	2D	Saturated	Retardation, first-order decay	Monod equations
Grant and Pattey (2003)	–	Analytical solutions for gaseous transport in atmosphere, soil and aquifer	Surface water and top soil layer	Variably saturated	None	Monod equations
Gu et al. (2012)	–	Finite-element	2D	Variably saturated	None	Monod equations
Gusman and Marino (1999)	RISK-N	Analytical	1D, top soil layer	Variably saturated	First-order decay, retardation	CropSyst model (Stockle and Debaeke, 1996)
Iqbal and Hiscock (2011)	–	Finite-element	2D	Saturated	None	Monod equations
Kinzelbach et al. (1991)	–	Finite-difference	2D	Saturated	First-order decay	Monod equations
Kinzelbach (1988)	–	RWPT	2D	Saturated	Kinetic sorption	None
MacQuarrie and Sudicky (2001)	–	Finite-element	3D	Variably saturated	Retardation	Monod and kinetic reactions
Maggi et al. (2008)	TOUGH-REACT-N	Integral finite difference	3D	Variably saturated	Multiphase flow, sorption, first-order decay	Monod and kinetic reactions
Peyrard et al. (2011)	–	Finite-difference	1D	Saturated	None	Monod equations and kinetic reactions
Steefel (2009)	CrunchFlow	Integral finite difference	3D	Variably saturated	Radioactive chain reactions	Monod equations, multicomponent aqueous complexation
Tartakovsky et al. (2009)	–	Lagrangian multiscale smoothed particle hydrodynamics	2D	Saturated	Biomass attachment and detachment	Monod equation for biomass growth
Yabusaki et al. (2011)	eSTOMP	Integrated-volume finite-difference	3D	Variably saturated	First-order decay, radioactive decay, dissolution	Equilibrium, conservation, and kinetic reactions, Monod equations for biomass growth and biogeochemical reactions
Widdowson et al. (1988)	–	Hybrid – particle tracking for advection, finite-difference for dispersion	1D	Saturated	None	Monod equations
Wriedt and Rode (2006)	RT3D	Hybrid – particle tracking for advection, finite-difference for dispersion	3D	Saturated	First-order decay	Monod equations

based models require more parameters and input data to drive the model simulation. Therefore more complex models may be better suited for field scenarios where high quality and high-density field observations are available, or for hypothetical simulations where literature values of parameters can be incorporated and statistical approaches such as Monte Carlo can be used to quantify uncertainty in model inputs and outputs. For sites where data are limited, more simplified models utilizing zero- or first-order kinetics may be more appropriate (Green et al., 2008, 2010; Tesoriero and Puckett, 2011). Table 1 summarizes features of existing reactive transport models that are coupled with multispecies biogeochemical reactions or biodegradation. It should be noted that codes that utilize first-order or radioactive decay-type equations are not included in the table.

Among various numerical simulation approaches, Eulerian numerical methods have been most commonly employed (MacQuarrie and Sudicky, 2001; Maggi et al., 2008; Iqbal and Hiscock, 2011; Peyrard et al., 2011). Eulerian methods have well-known problems with numerical dispersion, artificial oscillation, and mass conservation. If Peclet and Courant numbers are chosen to be small to meet stability requirements and reduce numerical dispersion, the resulting high spatial and temporal resolutions can produce large simulation times even on modern computer hardware (Salamon et al., 2006; Navarre-Sitchler et al., 2013). Lagrangian or hybrid Lagrangian and Eulerian methods have been developed as an effective alternative to address multispecies reactive transport problems such that for large-scale applications, a coarser spatial and temporal resolution can be used and the computational time can be reduced significantly. Except for codes that incorporate sorption, first-order kinetics and radioactive decay type reactions (Maxwell and Tompson, 2006; Prickett et al., 1981; Tompson, 1993; Kinzelbach, 1988; Siirila and Maxwell, 2012), it is challenging to incorporate biodegradation and geochemical reactions in Lagrangian and hybrid codes.

The most common approach is to split the transport and reactions in two steps. The transport (advection and/or dispersion) is simulated using the Lagrangian approach, while the reactions are simulated using the Eulerian approach. The reactions can be computed directly by solving the equilibrium (Ahlstrom et al., 1977; Fabriol et al., 1993) or kinetic equations (Essaid and Bekins, 1997; Wriedt and Rode, 2006), or by using an iterative approach (Abulaban et al., 1998). However, this requires mapping of Lagrangian particles to Eulerian concentrations, which is usually computationally expensive. In order to avoid calculating Eulerian concentrations, Benson and Meerschaert (2008, 2009) developed a probabilistic approach to simulate chemical reactions without referring to local concentrations for the particle-tracking algorithm. The probability of particles A and B reacting is calculated depending