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One-dimensional model for biogeochemical interactions and permeability reduction in soils during leachate permeation

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Abstract

This paper uses the findings from a column study to develop a reactive model for exploring the interactions occurring in leachate-contaminated soils. The changes occurring in the concentrations of acetic acid, sulphate, suspended and attached biomass, Fe(II), Mn (II), calcium, carbonate ions, and pH in the column are assessed. The mathematical model considers geochemical equilibrium, kinetic biodegradation, precipitation–dissolution reactions, bacterial and substrate transport, and permeability reduction arising from bacterial growth and gas production. A two-step sequential operator splitting method is used to solve the coupled transport and biogeochemical reaction equations. The model gives satisfactory fits to experimental data and the simulations show that the transport of metals in soil is controlled by multiple competing biotic and abiotic reactions. These findings suggest that bioaccumulation and gas formation, compared to chemical precipitation, have a larger influence on hydraulic conductivity reduction.

Keywords: Geochemical modeling; Leachate flow; Chemical speciation; Metal precipitation; Bacterial transport; Bioclogging; Permeability reduction

1. Introduction

The impact of leakage from landfills on the environment is governed by the transformation of inorganic and organic constituents of leachate via biogeochemical interactions. A variety of complex reactions occur between the biological (primarily microorganisms, such as bacteria), geological, and chemical components of leachate contaminated soils. These reactions not only influence chemical transport but can also affect groundwater flow by altering the soil's porosity and permeability due to bacterial growth, chemical precipitation, and anaerobic gas production. Under appropriate conditions the combined effects of biomass growth, metal precipitation and gas production on the soil's flow and transport properties can be significant (e.g., Brune et al., 1994; Rowe et al., 1997).

Reactive transport modelling has evolved as a tool for analysing the effects of complex biogeochemical interactions and reaction controls on soils. Quantification of these effects using mathematical relationships is an area of continuing research with several mathematical models having been developed to simulate the geochemical processes (Yeh and Tripathi, 1991; Engesgaard and Kipp, 1992; Walter et al., 1994) and biological transformations (Borden and Bedient, 1986; Kindred and Celia, 1989; Taylor et al., 1990; Clement et al., 1996a) in soils. The development and use of integrated biogeochemical

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models coupling transport processes to primary reactions (i.e., organic matter degradation) and secondary reactions (e.g., redox transformation, mineral dissolution and precipitation, sorption, and acid–base and homogeneous speciation) is of more recent origin (Lensing et al., 1994; McNab and Narasimhan, 1994; Zysset et al., 1994; Hunter et al., 1998; Schäfer et al., 1998a; Tebes-Stevens et al., 1998; Prommer et al., 1999; Mayer et al., 2002; Van Breukelen et al., 2004). In these models geochemical reactions are simulated assuming local equilibrium (Engesgaard and Kipp, 1992), partial redox disequilibrium (McNab and Narasimhan, 1994; Brun and Engesgaard, 2002), kinetic heterogeneous redox speciation (Tebes-Stevens et al., 1998; Mayer et al., 2002; Van Breukelen et al., 2004), or fully kinetic redox speciation (Hunter et al., 1998). The bacterial reactions have typically been simulated assuming either negligible bacterial activity (Engesgaard and Kipp, 1992), prescribed zero- or first-order microbial reactions (McNab and Narasimhan, 1994; Mayer et al., 2002; Van Breukelen et al., 2004), or bacterial growth dynamics described using Monod kinetics (Schäfer et al., 1998a; Tebes-Stevens et al., 1998).

The previous biogeochemical models have ignored the effects of biogeochemical interactions on the porosity and permeability of soil (e.g., due to biomass accumulation, chemical precipitation or gas formation) by assuming a constant biophase volume (e.g., Schäfer et al., 1998a) or instantaneous degassing (e.g., Van Breukelen et al., 2004). Recent progress has been made in the development of models to describe gas bubble formation and its effect on soil permeability (e.g., see Amos and Mayer, 2006a,b) and the patterns for the flow of gas injected into saturated porous media (Selker et al., 2007). Several models have also been developed for permeability reduction due to biomass accumulation in soil. Taylor et al. (1990) used the "spherical soil grains" approach of Kozeny–Carman (Carman, 1937) and the "cut-and-random-rejoining of tubes" approach of Childs and Collis-George (1950). Sarkar et al. (1994) used the effective medium theory to represent the soil medium as a three-dimensional network of pores and estimated the effective hydraulic conductance based on the pore throat size distribution following bacterial plugging. Clement et al. (1996a) presented analytical equations for changes in porosity, specific surface area, and permeability due to biomass accumulation in porous media. Thullner et al. (2002,2004) described the permeability reduction in pore networks from biomass growing on biofilm and in discrete colonies that occupy pores entirely. The colony-based model was later used by Seifert and Engesgaard (2007) to simulate bioclogging of porous media. While the proponents of the models successfully calibrated their models, the limited uptake and application of these models in other studies makes it difficult to identify a single model as being superior to others. Furthermore, comprehensive models that couple the effects of bioclogging and gas formation with chemical speciation have not been previously presented in the literature.

This paper presents a mathematical model for the biogeochemical interactions occurring during leachate permeation in packed columns. The model considers kinetic precipitation–dissolution and equilibrium aqueous geochemical speciation, kinetic biodegradation, chemical and bacterial transport, and biomass accumulation in soils. In particular, the model accounts for the effects of the biophase, chemical precipitate, and separate phase gas on the soil's permeability. To the authors' knowledge this is the first study to consider the above mentioned biogeochemical processes in an integrated manner, although previous studies have tackled subsets of the current system. Experimental data obtained from laboratory experiments with soil columns (Islam and Singhal, 2004) is used to assess the strengths and weaknesses of the proposed model and identify areas for further research.

2. Laboratory study

Laboratory experiments were conducted by pumping leachate collected from a local municipal landfill through packed columns to investigate the effects of leachate permeation and interactions between the soil media, bacteria and leachate constituents on soil permeability (details presented in Islam and Singhal, 2004). The columns were 50 cm long and 5 cm in diameter and were packed with 0.21–0.61 mm sized sand. The porosity of the packed media was estimated as 0.41 by gravimetric analysis. A flow of 0.23 ml/min was maintained in the columns and the head loss data was used to estimate the average saturated hydraulic conductivity as 8.8×10^{-3} cm/s. A pulse of 0.5 M sodium bromide solution was then injected into a column and an analytical solution of the advective–dispersive equation was fitted to the observed bromide breakthrough data to obtain a dispersivity of 0.12 cm. The bacterial inoculum used to seed the columns was obtained by centrifuging 24 L of landfill leachate and re-suspending the centrifugate in 2 L of raw leachate. This microbe-enriched solution was pumped into three columns at 5 ml/min, and was followed by 12 h of no flow to promote bacterial attachment to the soil. One of the columns was then dismantled to determine the initial distribution of attached biomass in soil. The remaining two columns were fed leachate, amended with acetic acid to give a final acetic acid concentration of 1750 mg/L (equivalent to 350 mg-C/L), in upflow mode at 0.23 ml/day for 58 days (Phase 1). To put