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Research papers

Numerical modeling of subsurface release and fate of benzene and toluene in coastal aquifers subjected to tides

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ABSTRACT

A numerical study was undertaken to investigate subsurface release and fate of benzene and toluene in a tidally influenced beach. The simulations were conducted by using a numerical model BIOMARUN, which coupled a multi-Monod kinetic model BIOB to a density-dependent variably saturated groundwater flow model MARUN. The fate and transport of the contaminant plume were characterized by computing its centroid trajectory, spreading area and percentage of biodegradation in beach saturated and unsaturated zone, respectively. Key factors likely affecting this process were investigated, including tide amplitude, capillarity and hydraulic conductivity. It was found that aerobic biodegradation was the major fate of the contaminant plume in the beach. Tidal action twisted the centroid of the contaminant plume during its migration in the beach, which increased the residence time of the plume in the beach. High tidal range significantly altered the spatial distribution of the contaminant biodegradation in the beach. In contrast, the capillary fringe had impacts on the percentage of benzene biodegraded in the saturated and unsaturated zone of the beach. The increase in capillary fringe enhanced the percentage of the contaminant biodegraded in the unsaturated zone, up to 40%, which is comparable to that in the saturated zone. Hydraulic conductivity seemed to have large impacts on the biodegradation rate of the contaminant in the beach. Higher hydraulic conductivity induced faster contaminant biodegradation in the beach.

1. Introduction

In the last few decades, coastal aquifers have been increasingly stressed by accidental release of a wide range of organic pollutants from various sites of petrol stations, pipelines and landfill (Essaid, 1994; Dojka et al., 1998; Atlas and Hazen, 2011; Boufadel et al., 2014; Ramakrishnan et al., 2015; Boufadel et al., 2016; Cozzarelli et al., 2016; Winegardner and Testa, 2016). Better understanding fate of the pollutants in coastal aquifer is critical in effectively assessing and remediating contaminated sites, especially for compounds (e.g., benzene, toluene and xylene) that could dissolve and disperse along groundwater flow paths, thereby resulting in a large contaminant plume formed in subsurface (Lu et al., 1999; Brovelli et al., 2007; Colombani et al., 2015; Sbarbati et al., 2015; Liu et al., 2016). In coastal aquifers, along groundwater discharge pathway, pore water flow and associated dissolved solutes (e.g., salt, nutrients and oxygen) involve very complex circulation and mixing with the seawater that infiltrates into aquifers due to multiple forcing mechanisms (e.g., tides, waves and evaporation) (Robinson et al., 2006; Li et al., 2008; Abdollahi-Nasab et al., 2010; Heiss, 2011; Geng and Boufadel, 2015b,a; Geng et al., 2016a,b). Therefore, characterizing a contaminant plume formed in subsurface such as trajectory, expansion, residence time, discharge rate and natural attenuation (i.e., biodegradation), is always a challenge for coastal aquifers.

Contamination of dissolved hydrocarbon due to petroleum spills and leaks has been widely observed in aquifers. In 1979, a buried oil pipeline near Bemidji, Minnesota, broke, causing the oil to be sprayed over approximately 6500 m^2 (Essaid and Hess, 1991; Revesz et al., 1995; Molins et al., 2010). Although significant amount of the spilled oil was removed by pumping from surface pools, trenching, burning and excavation of soil, large volumes of oil were still observed in subsurface, ranging from 0 to 8 m below the land surface, which provided a long-term, continuous source of hydrocarbon components that dissolved in, and were transported with, the flowing groundwater (Essaid et al., 1995). Between 1978 and 1998, 5262 spills due to gasoline releases were identified by U.S. Environmental Protection Agency on long island, New York, during which large plumes of benzene, toluene, ethylbenzene, and xylene (BTEX) and methyl tert-butyl ether (MTBE) were observed in subsurface, extending from source by a distance varying from

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30 m to 2000 m due to different timing and volume of the releases (Kolhatkar et al., 2001; Kelley et al., 2002; Zimmerman, 2003).

While field data were collected in numerous studies seeking to assess fate of dissolved hydrocarbon plume and its potential impacts on the surrounding environments, the data collected could only represent the overall behavior of the plume as dissolved hydrocarbon's transport, natural attenuation (i.e., biodegradation), and discharge occur simultaneously in coastal aquifers; therefore, convoluted driving dynamics and biodegradation of the contaminant plume could not be fully reflected by direct measurements. Numerical models have been recognized as important tools for improving the understanding of fate of a contaminant plume on its journey from its source zone to the ocean/river (Essaid et al., 1995; Clement, 1997; Schirmer et al., 2000; Brovelli et al., 2007; Robinson et al., 2009; Geng et al., 2014b; Lee et al., 2014; Ng et al., 2015; Li et al., 2016). Essaid et al. (1995) developed a two-dimensional, multispecies reactive solute transport model BIOMOC and used the model to investigate the field-scale dissolved hydrocarbon transport and degradation processes at the Bemidji, Minnesota, crude oil spill. The simulations included the biodegradation of volatile and nonvolatile fractions of dissolved organic carbon by aerobic processes, manganese and iron reduction, and methanogenesis. Lu et al. (1999) developed a reactive transport model to simulate the natural attenuation of a fuel-hydrocarbon plume present at Hill Air Force Base, Utah. The BTEX, oxygen, nitrate, Fe (II), sulfate and methane plumes calculated by the model agree reasonably well with field observations. Trefry et al. (2007) used groundwater model FEFLOW to simulate a dissolved hydrocarbon plume migrating within the shallow superficial aquifer to the nearby bank of the Canning River. In their study, seasonal variations in salinity and associated density effects on the subsurface flow and transport mechanism were taken into account. However, these studies were mostly focused on inland aquifers, and effect of tides on the fate of contaminant plume in coastal aquifers was not investigated. Robinson et al. (2009) used groundwater model PHWAT to simulate transport and biodegradation of a dissolved BTEX plume in an unconfined near-shore aquifer subject to tides. While tide effects were investigated, the focus of this modeling was contaminant plume migration in the saturated region of the aquifers (i.e., below the groundwater table) and thereby, the fate of dissolved hydrocarbon remained in unsaturated zone was not investigated; in addition, nutrient effects on microbial biodegradation was neglected. It has been found that transport and transformation of nutrients in coastal aquifers involved complex physical-biogeochemical processes due to tides and waves (Slomp and Van Cappellen, 2004; Spiteri et al., 2008; Anwar et al., 2014).

The objectives of this paper are (a) using a numerical model, BIOMARUN, coupling a multiple-Monod kinetic model BIOB to a 2-D (vertical slice) finite element model MARUN for variably saturated water flow and solute transport, to simulate processes of dissolved hydrocarbon (e.g., benzene and toluene) transport and biodegradation in coastal aquifers (including both saturated and unsaturated beach region), taking into account nutrient and oxygen effects on hydrocarbon biodegradation, and (b) performing sensitivity analyses to examine the key factors likely affecting the fate of dissolved hydrocarbon in coastal beaches. The key factors include tide amplitude and beach properties (i.e., permeability and capillary).

2. Numerical simulations

2.1. The BIOMARUN model

The model BIOMARUN couples a 2-D density-dependent variably saturated groundwater flow and solute transport model, MARUN, with a multiplicative Monod model BIOB for hydrocarbon biodegradation (Geng et al., 2013; Geng et al., 2014a; Geng et al., 2015). The model BIOMARUN could be used to simulate subsurface water flow, the transport and fate of dissolved salt, nutrient (e.g., nitrogen and phosphorus), oxygen (or any type of electron acceptor), and two types of substrates (e.g., hydrocarbon) and associated degraders. The governing equations of the BIOMARUN model were fully described in Geng et al. (2015). In the model, the equations of water flow and fate and transport are discretized in space by the Galerkin finite element method (Pinder and Gray, 1977) using linear triangular elements and integrated in time using backward Euler with mass lumping (Celia et al., 1990). The model considered the fate and transport processes in both saturated and unsaturated zones. As the unsaturated zone is well aerated, and the concentration of oxygen in air is ~38 times that of the solubility limit of dissolved oxygen (in water) per unit volume, we assumed that the dissolved oxygen is in equilibrium with the atmosphere (i.e., equal to 8.2 mg/L) at all the beach locations where the air content ratio is greater than 0.3 (i.e., the moisture ratio is less than 0.7). When the moisture ratio exceeded 0.7, the dissolved oxygen concentration was allowed to deplete due to microbial degradation.

2.2. Numerical implementation

The simulated domain was 200 m long with a slope of 4%, which represents a 2-D cross-shore transect through a coastal beach (Fig. 1). It was assumed that inland groundwater table was 2 m below the beach surface (i.e., H_{inland} = 23 m in the coordinate system established in this paper). At the seaward side, the tide was approximated as a cosine function, as follows:

$$H_{tide} = H_0 + A\cos\omega t, \quad (1)$$

where H_0 is the mean sea level, A is the tide amplitude and ω is the tidal angular frequency. The parameter values used for the simulation were: H = 21.0 m, A = 1.0 m and ω = 6.28 rad/d (i.e., a tidal period of a day). The beach properties (e.g., hydraulic conductivity and capillarity) and boundary conditions used to simulate the transport of salt, nutrient and oxygen were the same as that measured and used in Geng et al. (2015) for a tidally influenced beach in Gulf of Mexico. The salinity in inland groundwater and seawater was 5.0 g/L and 35.5 g/L, respectively. The measured concentration of nitrogen was 1.2 ± 0.34 mg-N/L in the beach and 0.2 mg-N/L in the sea. The oxygen concentration measured in the pore water and the sea was 8.2 mg/L, close to the solubility limit of oxygen in water in contact to the air. As the oxygen concentration of 8.2 mg/L was measured at various locations and depths of the beach, we assumed that the initial oxygen concentration was uniformly distributed in the beach. The contaminant (i.e., benzene and toluene) was assumed to be accidently released into the beach through a source point located at x = 50 m and z = 23 m, which was approximately 0.5 m above the groundwater table and 110 m landward of the shoreline. The period of the contaminant release was 15 days with a flow rate of 0.36 m^3/d. The concentration of benzene and toluene was 100 mg/L and 50 mg/L, respectively. As the concentration of benzene and toluene initially released into the beach was very low in comparison to salt concentration (5.0 g/L for inland water and 35.5 g/L for seawater), we neglected their density effects on the water flow and solute transport processes. The microbial kinetic parameter values adopted in the simulation were obtained from Chen et al. (1992), which have been widely used to simulate biodegradation of benzene and toluene (Essaid et al., 1995; El-Kadi, 2001).

For the simulations, model BIOMARUN was first run approximately 100 days without contaminant release until the hydraulic and hydrodynamic regime reached a quasi-steady state. The pressure and solute distribution then were used as initial conditions to