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Modeling oil biodegradation and bioremediation within beaches

Xiaolong Geng[1], Chunjiang An[2], Kenneth Lee[3] and Michel C Boufadel[1]

Modeling oil biodegradation and remediation has become an increasingly important means to predict oil persistence and explore potential *in-situ* bioremediation strategies for oil-contaminated beaches. Beaches involve complex mixing dynamics between seawater and groundwater. Thus, numerically predicting oil biodegradation within beach systems faces major challenges in merging highly dynamic biogeochemical conditions into microbial degradation models. In this paper, we reviewed recent advances in modeling oil biodegradation from aspects of oil phases, reaction kinetics, microbial activities, environmental conditions, and beach hydrodynamics. We identified key controlling factors of oil biodegradation, highlighted the importance of fate and transport processes on nearshore oil biodegradation, and suggested some advances needed to achieve for developing a robust numerical model to predict oil biodegradation and bioremediation within beaches.

Addresses
[1] Department of Civil and Environmental Engineering, New Jersey Institute of Technology, University Heights, Newark, NJ, 07102, USA
[2] Department of Building, Civil and Environmental Engineering, Concordia University, Montreal, QC, H4B 1R6, Canada
[3] Department of Fisheries and Oceans, Dartmouth, Nova Scotia, B2Y 4A2, Canada

Corresponding author: Geng, Xiaolong (gengxiaolong@gmail.com)

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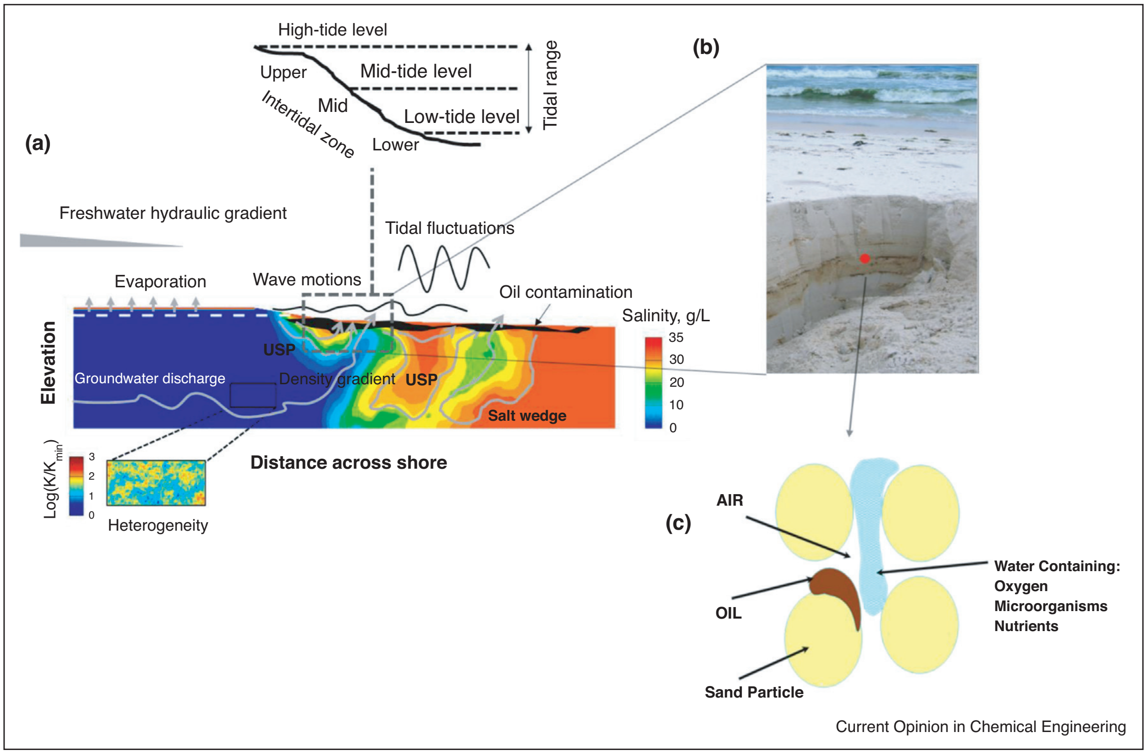
Introduction

Oil spills have been recognized as one of the most severe pollutions damaging coastal and marine environments [1,2]. Driven by winds and currents, oil spilled into the water often reaches coastlines. On April 20, 2010, an explosion of the Deepwater Horizon (DWH) offshore drilling platform resulted in the release of an estimated 3.19 million barrels of crude oil into the Gulf of Mexico, and an estimated 22 000 tons of weathered oil contaminated hundreds of kilometers of coastlines along the northern Gulf of Mexico [3]. Studies showed that indigenous microorganisms can metabolize various oil constituents as sources of carbon and energy for growth and function. Therefore, microbially mediated attenuation (i.e. biodegradation) has been expected to be an important process in the fate of the oil buried within beach sediments [4•,5,6•]. Since the 1990s, modeling oil biodegradation and remediation within beaches has become an increasingly important means to predict oil persistence and explore potential *in-situ* bioremediation strategies for oil-contaminated beaches [7,8,9•]. Although considerable efforts have been made to advance numerical understanding of oil biodegradation in marine and coastal systems, there is no review on oil biodegradation within coastal beach systems from the mathematical modeling aspects. Therefore, the goals of this paper are to summarize the currently existing components of modeling oil biodegradation within coastal beach systems, particularly for subsurface oil, and propose the current challenges and limitations with future research direction. Figure 1a illustrates multiple aspects that likely affect oil biodegradation in beaches.

Oil phases

Oil is a complex mixture of hydrocarbons and organic compounds. Oil compounds with relatively high aqueous solubilities will tend to transfer to the water phase, while the insoluble compounds will stay in oil. It has been recognized that aromatic hydrocarbon compounds are usually more soluble than alkanes of the same carbon number. In addition, within homologous series of compounds, there is a marked decrease in solubility when the carbon number increases. Compared to heavy crude oil, light crude oil has a higher percentage of light hydrocarbons. Modeling oil biodegradation can be categorized based on the oil phase that the model dealt with. Advective-diffusive-reactive (ADE) transport equations are often used to simulate biodegradation of dissolved phase of hydrocarbons in aquifer systems. The example components include dissolved petroleum hydrocarbons such as benzene, toluene, ethylbenzene, and xylene (BTEX), and petroleum-derived dissolved organic matters such as microbial-derived partially oxidized hydrocarbon degradation product and hydrocarbon oxidation products [10,11]. To model multicomponent scenarios, multiple

Figure 1



(a) Schematics of oil-contaminated beach subjected to multiple driving mechanisms, adapted from Geng *et al.* [41]. **(b)** Photograph of the oil contamination taken in a Gulf beach after Deepwater Horizon oil spill. **(c)** A sketch of residual oil trapped within Gulf beach sediments.

reactive solute transport equations with sequential aerobic and anaerobic degradation processes are often required to couple with groundwater flow model to predict fate and transport of dissolved oil compounds [12]. In beach systems, immobile phase of oil is usually presented as oil residuals trapped within sediment pores by capillary forces [4•,13] (e.g. Figure 1b). To simulate the fate of the immobile oil phase, the dissolution processes need to first be conceptualized to delineate the rate at which the oil hydrocarbon dissolves into the aqueous phase for biodegradation to occur [11,14]. For insoluble attached oil hydrocarbon, a numerical model has been developed to characterize its biodegradation rate and associated physical loss in the beach environments (e.g. tidal washout and volatilization) [15].

Reaction kinetics

The reaction kinetics of hydrocarbon biodegradation can be described by different conceptual models. These models are mainly classified into three types: first-order decay, instantaneous reaction, and Monod kinetics. The first-order decay model is one of the common expressions for representing the biodegradation of hydrocarbon. When the first-order decay model was adopted, the biodegradation of hydrocarbon was shown as an exponential decay relationship:

$$C = C_0 e^{-kt}, \tag{1}$$

Where C is the biodegraded concentration of hydrocarbon, C_0 is the initial concentration, and k is the first order biodegradation rate for hydrocarbon. Sometimes, the first-order rate is expressed in terms of a half-life, $T_{1/2}$, for hydrocarbon:

$$T_{1/2} = \frac{0.693}{k}, \tag{2}$$

Because of its simplicity, the first-order decay model has been widely used to simulate or evaluate hydrocarbon biodegradation by recent studies [16–19]. For example, Bociu *et al.* [18] conducted *in-situ* experiments to quantify degradation of sediment-oil-agglomerates (SOA) buried in the upper 50 cm of a North Florida sandy beach. The first-order decay model was adopted in their study to estimate SOA, alkane and PAH decay rates. Boufadel *et al.* [19] explored bioremediation of the Exxon Valdez oil in Prince William Sound beaches. A first order rate was estimated for total PAH at six Alaska beach sites after the proposed bioremediation strategy was implemented. The limitation of the first-order decay model is that only one parameter, decay rate, is considered to characterize the oil biodegradation, and thus, the first-order decay model cannot incorporate site-specific information such as the availability of electron acceptors or the growth of microorganism. The instantaneous reaction model describes that the microbial biodegradation kinetics are very fast in comparison with the transport of oxygen, and that the growth of microorganism and utilization of oxygen and hydrocarbon in the subsurface can be simulated as an instantaneous reaction between the hydrocarbon and oxygen [20]. Although the instantaneous model has been widely used for simulate aquifer contaminations, to authors' knowledge, it has not been applied for modeling oil contamination in aquifer systems. One of the most popular expressions for simulating the reaction kinetics of oil biodegradation is the hyperbolic saturation function, referred to as Monod or Michaelis–Menten kinetics:

$$\mu = \mu_{\max}\frac{C}{C + K_C}, \tag{3}$$

Where μ is the growth rate of microorganisms, and $\mu_{\max}$ is the maximum growth rate. The term K_C is known as the half-saturation constant or the growth-limiting substrate (i.e. hydrocarbon) concentration which allows the microorganism to grow at half the maximum specific growth rate [21]. The Monod kinetic model allows for multiple controlling factors of hydrocarbon biodegradation by superimposing and customizing the hyperbolic saturation terms. There are many extended Monod kinetics formulas used for describing the oil biodegradation kinetics. The multiplicative Monod kinetics is the most commonly used form for modeling oil biodegradation. It describes the reaction kinetics by multiplying together multiple hyperbolic saturation terms, each of which represent a factor limiting the oil biodegradation. For instance, Valsala and Govindarajan [11] adopted a multiplicative Monod model to delineate biodegradation of dissolved BTEX. In the model, the biodegradation rate is determined by a multiplicative Monod term that represents the limiting factors of oxygen availability and dissolved concentration of each hydrocarbon compounds. Other modified Monod kinetics formulas have also been used. Carvajal *et al.* [22] derived a modified Monod model by including an inhibition constant for providing a better fit to experimental data. Jeong *et al.* [7] briefly reviewed modified forms of Monod equation for considering effects of various environmental conditions on microbial growth and associated oil biodegradation rate.

Microbial activity

Microbial activity, such as the growth, decay and distribution of microorganism, plays an important role in hydrocarbon biodegradation [23,24]. Microbial secreted biosurfactants also play a vital role in oil bioavailability [25,26]. Numerical models describing microbial activity have been increasingly established over the last decade [27]. Modeling microbial growth has incorporated an explicit account of endogenous metabolism. Besides the model simply relying on population-level kinetic equations like Monod's, studies have progressively turned to individual-based or agent-based models that allow for a high degree of complexity of individuals and of interactions among individuals [28,29]. One of the conceptual frameworks, referred to as macroscopic approach, assumed that the bacteria are respond to the macroscopic bulk fluid substrate concentration. Such approach considers spatially averaged biomass concentration, and neglects the distribution of the microorganisms at pore scale [30]. In porous media, the microbial activity is strongly influenced by the extremely heterogeneous physical and biochemical conditions. In order to handle such microscale complexities, microscale models have become increasingly developed. The typical pore-scale modeling approaches include pore-scale network modeling, lattice Boltzmann method, and other direct numerical simulation [31•]. While they are computationally expensive, the pore-scale models have the advantage to resolve tempo-spatial patterns of microorganisms within porous media at relatively high resolution. However, at the current stage, macroscopic models seem to provide more adaptability for modeling oil biodegradation in coastal aquifer systems. This is because in most oil biodegradation studies involving beach aquifers, measurements of microbes are commonly reported as bulk biomass concentrations, which only conform to the macroscopic approach. Moreover, fate and transport models are developed based on Darcy-scale flow equations. They only allow to consider processes averaged over a representative elementary volume, and disregard microscopic flow in individual pores. Therefore, the pore-scale observations/simulations of microorganisms are barely used for conducting simulations of oil biodegradation with beach systems.

Environmental conditions

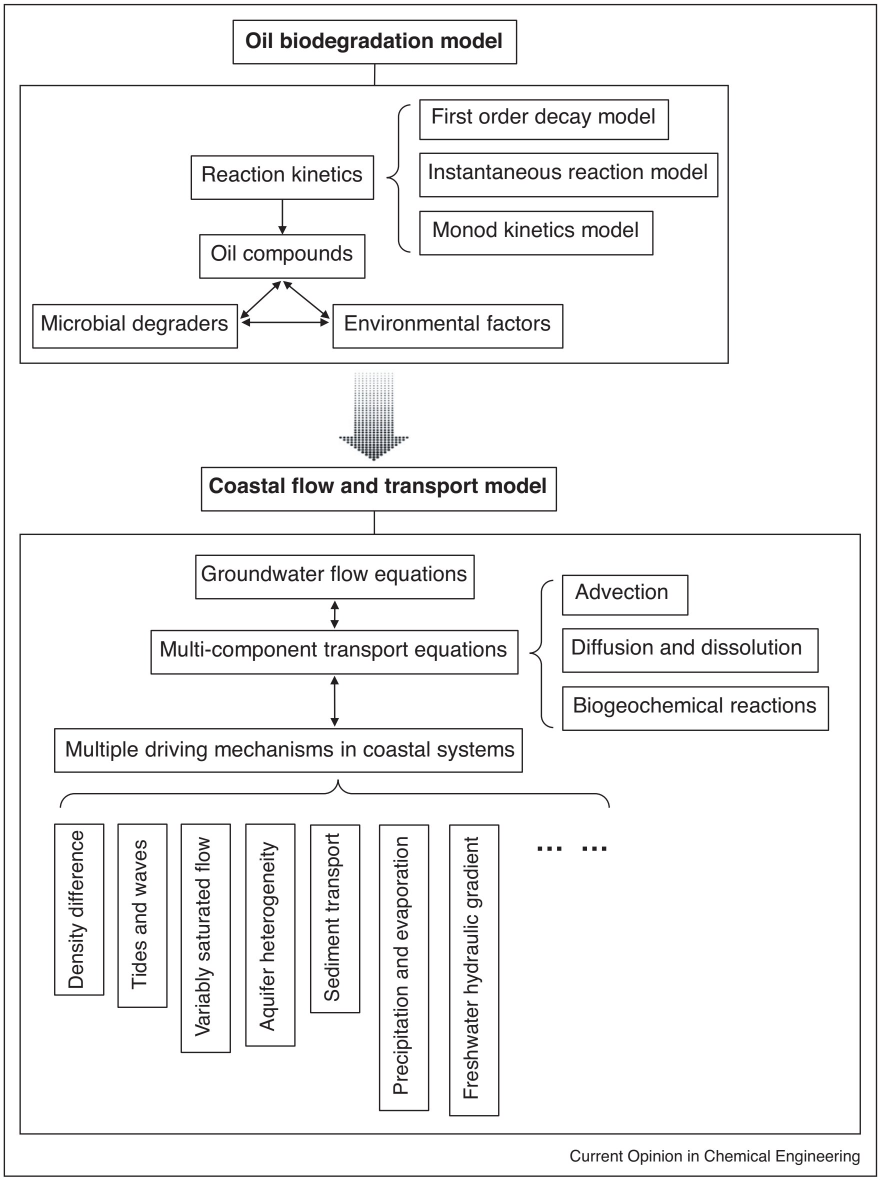
Dissolved oxygen and nutrients are key factors in oil biodegradation; limited availability of either could slow down the biodegradation rate considerably [32,33]. Therefore, parameterization of oxygen and nutrient effects is an essential step for modeling oil biodegradation. Laboratory experiments show that the threshold of the oxygen concentration for hydrocarbon biodegradation could be as low as 0.1 mg/L. However, *in-situ* studies suggest that the concentration greater than 1.5 mg/L is needed to support the efficient biodegradation of oil [4•]. Monod kinetics is commonly used to reveal the relation between dissolved oxygen and oil degrader growth. Geng *et al.* [34] proposed a modified Monod fraction, $\frac{O^4}{K_O+O^4}$, to delineate how the dissolved oxygen concentration affects aerobic microbial growth, where K_O represents the half-saturation concentration for oxygen consumption. The fourth order is used to describe a sharp switch of microbial activity from aerobic to anoxic when the concentration drops below 2.0 mg/L, and negligible limiting effects of oxygen on oil biodegradation as the concentration increases beyond 2.0–3.0 mg/L. Compared to aerobic biodegradation, anaerobic oil biodegradation within beaches is relatively slow, which can be simulated using a similar numerical scheme with a relatively small degradation rate [19,22]. During the process of oil biodegradation, microorganisms require not only electron acceptor, but also nutrients for microbial growth and function [35]. The limitation of nutrients on microbial growth can be delineated by Monod kinetics. For example, $\frac{N}{K_N+N}$ is widely used to describe impacts of nitrogen concentration on oil biodegradation, where N denotes nitrogen concentration, and K_N denotes the half-saturation concentration for nitrogen consumption. The value of K_N was estimated between 0.1 mg/L to 1.0 mg/L for different oil compounds [15].

Besides oxygen and nutrients, other environmental conditions also likely influence oil biodegradation, and therefore, need to be incorporated in modeling oil biodegradation. The biodegradation of oil can be severely reduced due to high salinity [36–38]. Studies found that the microbial community is profoundly limited by high salinity, with many hydrocarbon degraders, such as Thalassolituus, Cycloclasticus, and Oleibacter disappearing to undetectable levels, while others, such as Marinobacter increase substantially [39]. Park and Marchand [40] introduced a modified Monod equation to consider salinity inhibition effects. The salinity inhibition parameter would effectively decrease the maximum specific growth at high salinity, expressed as (μ_{max}-I_s), where the parameter I_s is the inhibition factor and equal to zero for non-saline condition. Temperature also influences the maximum specific growth rate of biomass. The Arrhenius law was originally proposed to describe the temperature effects, and later on modified forms were proposed for extended scenarios (e.g. Hougen–Watson model) [7].

Beach hydrodynamics

Beach aquifers are dynamic environments. Along coastlines, oceanic forces (e.g. waves, tides, and wind) create highly dynamics zones in which seawater and terrestrial groundwater strongly interact [41]. These groundwater-surface water interactions and the associated transport of chemicals and biota across the beach surface promote the formation of biochemically active zones that lead to dynamic biogeochemical conditions for oil biodegradation within beaches [4•,42]. Taking into account coastal groundwater flow and transport processes is critical for modeling oil biodegradation within beach systems. A robust groundwater model for characterizing coastal subsurface fate and transport processes needs to incorporate multiple driving mechanisms such as buoyancy forces caused by the density difference between fresh and saline groundwater, terrestrial hydraulic gradients, tides, waves, evaporation and precipitation [41,43] (Figure 1a). For modeling oil biodegradation within beaches, a multi-species reactive transport model would also be essential for considering oil compounds, associated microbial degraders, and relevant chemical species (e.g. nutrients and oxygen) [34,44•,45,46] (Figure 1b-c). Figure 2 illustrates a flow chart connecting between modeling oil biodegradation and modeling flow and transport processes in coastal aquifers. Robinson *et al.* [47] implemented the coupled density-dependent flow and multi-species reactive transport code PHWAT to examine the impacts of tidal actions on the aerobic biodegradation of BTEX released in coastal aquifers. Their simulation results highlighted the importance of tidal forcing on the extent of microbial degradation processes; tidal actions control the intensity of salt-freshwater mixing, period of exposure of the contaminant to the mixing zone, and rate of oxygen delivery to the aquifer. Wave-induced surface water-groundwater exchange has also been found to importantly affect fate and transport of contaminants within beaches such as nutrients, fecal bacteria, and nonaqueous phase liquids [48,49]. Besides oceanic forcing, vadose zone processes are also essential for simulating oil biodegradation within beach systems. Studies revealed that landfall of oil spills (e.g. DWH) was primarily controlled by current and waves. The oil deposition usually forms sediment-oil-aggregates (SOAs), which often get buried at shallow depths subjected to variably saturated conditions created by oceanic forcing. Geng *et al.* [45] used conducted numerical simulations of a hypothetical beach polluted with benzene and toluene, using a variably saturated model. Their simulations revealed that these contaminants occupied the unsaturated zone, where there was significant biodegradation. Prior modeling studies also revealed a dynamics role of fate and transport of oxygen and nutrients in microbial oil degradation occurred in the Gulf beaches contaminated with DWH [34,46]. Geng *et al.* [34] developed a numerical model BIOMARUN, coupling a multiple Monod kinetic model to a density-dependent variably saturated groundwater flow model, to simulate the biodegradation of low-solubility hydrocarbon in tidally influenced beaches. Their results suggested different limiting factors dominate oil biodegradation processes at different portions of the beach. In the upper intertidal zone, where the terrestrial nutrient concentration was relatively high, faster rate of oil biodegradation occurred deeper in the beach. In the mid-intertidal zone, a reversal was simulated where the oil biodegradation rate was high at shallow locations. This is due to

Figure 2



Flow chart of connecting oil biodegradation model and coastal flow and transport model.

the depletion of oxygen along the depth, which made oxygen the limiting factor for oil biodegradation. In particular, oxygen concentration in the mid-intertidal zone exhibited two peaks as a function of time, indicating a dynamic interactions between oil biodegradation and surrounding biogeochemical conditions. Geng *et al.* [46] further numerically examined potential bioremediation strategies for oil-contaminated beaches, and suggested that the replenishment of oxygen is essential for effective oil removal to occur within beach intertidal zone. Moreover, stimulation of oil biodegradation was more evident in the upper and mid-intertidal zone of the beach. In the lower intertidal zone, due to less nutrient and oxygen replenishment, bioremediation treatment has less impact there [e.g. Ref. 50].

Conclusions

Shoreline restoration and resilience after oil spills require robust numerical models to assess microbial oil degradation within beach sediments and improved knowledge of how beach groundwater flow and biogeochemical reactions affect the degradation processes. This paper reviewed modeling oil biodegradation from aspects of oil phases, reaction kinetics, microbial activities, environmental conditions, and beach hydrodynamics. We summarized recent advances in modeling schemes and technologies. We highlighted the importance of incorporating coastal fate and transport processes into modeling of nearshore oil biodegradation, and herein suggest continued research along this pathway. To improve the numerical modeling of oil biodegradation and bioremediation within beach systems, further research would be suggested from the following aspects.

- Oil landfall processes and its associated interactions with sediments need to be better characterized numerically since they can provide more precise oil fate and distribution within beach systems.
- Pore-scale modeling of microbial activities need to be considered, which allows to mimic heterogeneity and hotspots of microbial community that were ubiquitously observed in fields. High-resolution fate and transport models are also needed to comprehensively characterize heterogeneity in oil biodegradation processes within beach environments.
- A comprehensive model needs to be developed, incorporating multiple environmental controlling factors of oil biodegradation such as conditions of oxygen, nutrients, salinity, moisture, pH, and so on.
- Combined experimental and numerical approaches are recommended for oil biodegradation modeling studies. Laboratory experiments will help estimate biogeochemical parameters, and therefore reduce the uncertainty originated from model inputs. Field measurements will help a better delineation of boundary and initial conditions and later on be used for model calibration.

Conflict of interest statement

Nothing declared.

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Declaration of Competing Interest

The authors report no declarations of interest.

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