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Enhancing bioremediation of oil-contaminated soils by controlling nutrient dispersion using dual characteristics of soil pore structure

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ABSTRACT

Soil structure is heterogeneous with cracks or macropores allowing bypass flow, which may lead to applied chemicals avoiding interaction with soil particles or the contaminated area. We investigated the bioremediation efficiency of oil-contaminated soils by applying suction at the bottom of soil columns during bioremediation. Unsaturated flow conditions were investigated so as to avoid bypass flow and achieve sufficient dispersion of chemicals in the soil column. The boundary conditions at the bottom of the soil columns were 0 kPa (saturated flow) and −3 kPa (unsaturated flow), and were applied to a volcanic ash soil with and without macropores. Unsaturated flow was achieved with −3 kPa and an injection rate of 1/10 of the saturated hydraulic conductivity. The resultant biological activities of the effluent increased dramatically in the unsaturated flow with macropores condition. Unsaturated conditions prevented bypass flow and allowed dispersion of the injected nutrients. Unsaturated flow achieved 60–80% of saturation, which enhanced biological activity in the soil column. Remediation results were better for unsaturated conditions because of greater biological activity. Also, unsaturated flow with macropores achieved even remediation efficiency from upper through lower positions in the column. Finally, taking the applied solution volume into consideration, unsaturated flow with −3 kPa achieved a 10 times higher efficiency when compared with conventional saturated flow application. These results suggest that effective use of nutrients or remediation chemicals is possible by avoiding bypass flow and enhancing biological activity using relatively simple and inexpensive techniques.

1. Introduction

Soils are an important environmental resource, along with air and water. They affect not only the global hydrologic cycle, but also regional water environments. Environmentally sound water and solute circulation in watersheds is desirable for conservation, protection and recovery of the natural environment in regional societies. In recent years, various regulations, such as the Soil Contamination Countermeasures Act (Ministry of the Environment, Japan, 2002) and CERCLA (U.S. EPA, 1980) were established to protect the soil environment and prevent soil contamination by environmentally toxic chemicals.

If soils are contaminated by oil or volatile organic compounds, effective remediation is sometimes challenging. When the concentration of contaminants is relatively high, *ex situ* treatment after soil excavation or soil vapor extraction can be applied (U.S. EPA, 1988). At lower concentrations, many physical/chemical civil engineering techniques are not cost-effective, because most of the contaminants remain in small micropores. In these cases, bioremediation is a promising site treatment tool (Balba et al., 1998), and its application to the removal of pollutants is typically less expensive than other civil engineering methods (Russell, 1992). Bioremediation enhances the degradation process by injecting air or nutrients, or even cultured microbes that are specific for certain degradation processes (Vidali, 2001). It has less impact on the environment and enables degradation of hazardous compounds to innocuous by-products (Vidali, 2001). *In situ* bioremediation techniques have major advantages in the remediation of contaminated soil and groundwater because they do not require site excavation (U.S. EPA, 1990).

The success of bioremediation of contaminated soils depends on how much chemical solution is conducted into the finer pores where the contaminants are usually located. However, the soil pore structure is heterogeneous, and macropores or cracks may rapidly conduct water flow below the remediation zone (Beven and Germann, 1982), in contrast to Darcy's law (Darcy, 1856)

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and Richards equation (Richards, 1931). Soil macropore networks establish a dual-domain type of transport in which water and solutes are preferentially channeled through soil macropores, while slowly diffusing into and out of the bulk soil matrix (Gerke and van Genuchten, 1993; Köhne and Mohanty, 2005). There are reports that elucidate the role of macropores in solute transport (Natsch et al., 1996; Pivetz and Steenhuis, 1995; Ronkanen and Klove, 2009). However, generally it is a challenge to control solute transport in soils so that applied chemicals are delivered into desired positions.

The motivation for this study was the need for solute transport control in soils with macropores, where bypass flow is likely to be dominant, with a consequent high risk of bioremediation failure. Engineers at remediation sites sometimes inject nutrients into the ground at high rates. However, most of the injected materials will not undergo bioremediation because of bypass flow. In this context, it is fundamental to study and utilize the natural structure of soils in order to achieve environmentally sound remediation.

Convection and dispersion are fundamental processes underpinning solute transport in porous media, which governs the distribution of solutes in soils (Ishiguro, 1992; Toride et al., 2003). If dispersion is sufficiently developed in a soil column, solutes will be well distributed and breakthrough curves will show a normal distribution. On the contrary, if solute transport in soils is governed by structure-induced bypass flow, dispersion will not be well developed in the soil column and bypass flow will deliver the solutes through the soil without interaction with most of the soil body (Kutilek and Nielsen, 1994).

In a previous report (Mori and Higashi, 2009), we introduced solutes to the soil matrix and successfully enhanced dispersion, even in soils with macropores, while avoiding bypass flow. Bimodal distribution in the breakthrough curve (bypass flow dominant) for a macroporous soil column was greatly changed to a normal distribution (matrix flow dominant), when a suction of −3 kPa and an infiltration rate of 1/10 of the saturated hydraulic conductivity (Ks) were applied. Application of this technique to contaminated soils could deliver chemicals to desired locations within the soil profile. Moreover, if control could be achieved by using a simple technique, it would be beneficial for cost effective remediation.

In this study, biostimulation was applied to oil-contaminated soils to investigate the effect of structural differences on bioremediation. Cutting oil was used as the contaminant as it is frequently used for lubrication in industrial processes (Shashidhara and Jayaram, 2010) and contamination by it is often found in the vadose zone (unsaturated zone) (Hillel, 1998). Macropore structure is more often the rule than the exception in the unsaturated vadose zone and thus the effect of soil structure needs to be carefully examined. The two objectives of this study were to examine differences in biological activity by changing the soil structure and controlling the infiltration process, and to examine how to obtain cost-effective conditions for biostimulation in structured soils where macropores are predominant.

2. Materials and methods

2.1. Soils

Bioremediation experiments were conducted using four soil columns containing a Kuroboku soil (a volcanic ash soil) from Shimane, Japan. Soils were sampled from depths of 30–50 cm. Contaminated soils were prepared by mixing the soil with cutting oil at a concentration of 5000 mg kg^{-1} to enable effective observation of the bioremediation processes. Four repacked soil columns were prepared in small stainless steel columns (diameter 5.0 cm, height 10.0 cm) to give bulk densities of 0.90 cm^3 cm^{-3} that simulated natural conditions. The columns were placed in pressure cells with membrane filters at the bottom. The experimental setup is shown in Fig. 1.

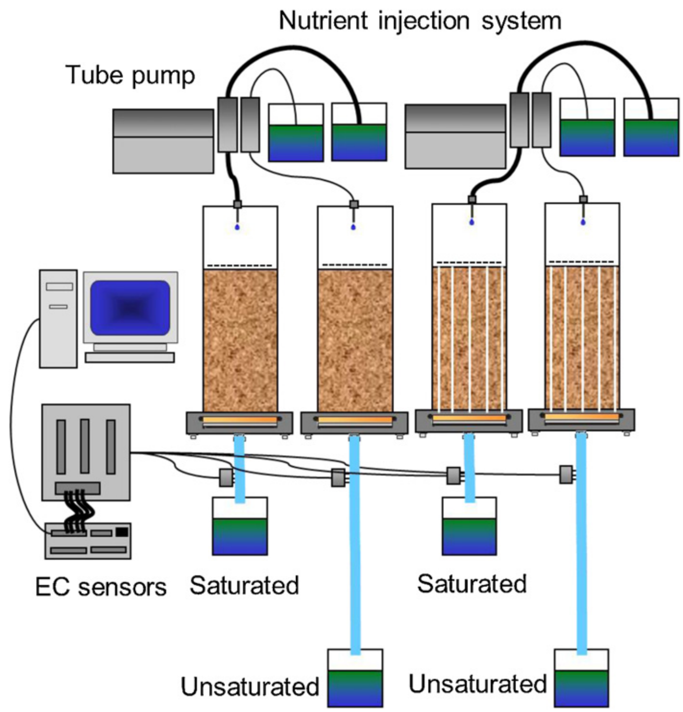

Fig. 1. Experimental setup for the biostimulation experiments.

Two columns contained only micropores with no macropores. In the other two soil columns, seven vertical macropores (simulating a natural field soil) were artificially created by gently pushing a stainless steel rod of 1 mm diameter into the soils following the technique of Mori and Higashi (2009). The macropores were sufficiently small that they were not visible from the surface. Physical properties of the soil columns with and without the macropores are shown in Table 1. Saturated hydraulic conductivities were measured using the constant head method (Klute and Dirksen, 1986).

After saturation with distilled water, samples were drained by applying gravitational pressure to the bottom of the tubes using an outlet tube. Nutrients were then applied to the top of the columns for 30 days. The applied pressure and infiltration intensity were 0 kPa and 1.4 × 10^{-4} cm s^{-1}, respectively, for the saturated infiltration conditions, and −3 kPa and 1.4 × 10^{-5} cm s^{-1}, respectively, for the unsaturated infiltration conditions. The four columns were considered to represent unsaturated or saturated flow with or without macropores.

Table 1
Physical properties of the examined soils.

Soil	Kuroboku soil (Andisol)	
Pore structure	w/o macropores	w/ macropores
Particle density (Mg m^{-3})	2.59	2.59
Porosity (m^3 m^{-3})	0.654	0.654
Macro-porosity (m^3 m^{-3})	0	0.003
K_s (cm s^{-1})	1.40 × 10^{-5}	1.43 × 10^{-4}
Bulk density (Mg m^{-3})	0.90	0.90

Macroporosity: the volume of macropores (d > 75 μm) in a soil sample divided by the bulk volume of the sample (after Soil Sci. Soc. Am., 1996).