



## Changes in air flow patterns using surfactants and thickeners during air sparging: Bench-scale experiments



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### ABSTRACT

Air injected into an aquifer during air sparging normally flows upward according to the pressure gradients and buoyancy, and the direction of air flow depends on the natural hydrogeologic setting. In this study, a new method for controlling air flow paths in the saturated zone during air sparging processes is presented. Two hydrodynamic parameters, viscosity and surface tension of the aqueous phase in the aquifer, were altered using appropriate water-soluble reagents distributed before initiating air sparging. Increased viscosity retarded the travel velocity of the air front during air sparging by modifying the viscosity ratio. Using a one-dimensional column packed with water-saturated sand, the velocity of air intrusion into the saturated region under a constant pressure gradient was inversely proportional to the viscosity of the aqueous solution. The air flow direction, and thus the air flux distribution was measured using gaseous flux meters placed at the sand surface during air sparging experiments using both two-, and three-dimensional physical models. Air flow was found to be influenced by the presence of an aqueous patch of high viscosity or suppressed surface tension in the aquifer. Air flow was selective through the low-surface tension (46.5 dyn/cm) region, whereas an aqueous patch of high viscosity (2.77 cP) was as an effective air flow barrier. Formation of a low-surface tension region in the target contaminated zone in the aquifer, before the air sparging process is inaugurated, may induce air flow through the target zone maximizing the contaminant removal efficiency of the injected air. In contrast, a region with high viscosity in the air sparging influence zone may minimize air flow through the region prohibiting the region from de-saturating.

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### 1. Introduction

Air sparging is a remediation technique for cleaning contaminated subsurface environments and has been proven effective in both lab- and field-scale studies (Bass et al., 2000; Johnston et al., 2002; USEPA, 2001). As volatilization/vaporization is the primary mechanism responsible for the contaminant mass transfer from the solution/liquid to the mobile gas phase, organic compounds with high volatilities are the major target chemicals to be removed by this process (e.g., Adams and Reddy, 2000). The rate of mass transfer between the solution/liquid and

the gas phases, which is directly related to contaminant removal efficiency, depends on many parameters including the hydrogeological settings and the physico-chemical properties of the contaminants and the contaminant-containing fluid phase (Burns and Zhang, 2001; Chao et al., 1998; Peterson et al., 2000; Tsai, 2007; Waduge et al., 2004).

The extent of the sparging influence zone generated during air sparging has been studied under various hydro-geological conditions (Hu et al., 2010, 2011; Lundegard and LaBrecque, 1995; McCray and Falta, 1996; Tomlinson et al., 2003). The shape and size of the influence zone may be delineated using appropriate models (e.g., McCray and Falta, 1997). In addition to the bulk volume of the de-saturated region formed by injected air, other parameters affecting the efficiency of the

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sparging process have also been studied (Mohamed et al., 2007; Rogers and Ong, 2000; Yoon et al., 2002). The nature of porous media (e.g., grain size and size distribution), the heterogeneity of the geologic formation, the flow rate of injected air, and the method of air injection (e.g., intermittent or continuous, spacing between air injection points), and most importantly, the chemical properties of the contaminants are parameters that affect performance of the air sparging process (e.g., Rogers et al., 2004). Organic contaminants may be in the form of a dissolved state in aqueous solution or a liquid state immiscible with the aqueous phase. Direct vaporization from the nonaqueous phase liquid (NAPL) is considered to be a more effective mass transfer process during the air sparging process than volatilization of the contaminants from the aqueous phase. The air sparging process may be effectively applied to contaminants with high vapor pressures or high volatilities (i.e., large Henry's law constants) (Johnston et al., 1998).

Studies regarding the air sparging process have primarily focused on enhancing the performance of the process without altering the physico-chemical properties of the resident aqueous solution in the aquifer. Kim et al. (2004) reported that artificial suppression of the aqueous phase surface tension prior to sparging enlarges the sparging influence zone and drains more water from the saturated zone due to reduced air entry pressure. The simple reduction of aqueous phase surface tension using an anionic surfactant (sodium dodecylbenzene sulfonate) increases the size of the sparging influence zone; hence, enhancing the removal rate of NAPLs, volatile organic compounds in water or increasing the dissolved oxygen level (Kim and Annable, 2006; Kim et al., 2009; Qin et al., 2013a, 2013b, 2014). Surfactant-enhanced air sparging (SEAS), an air sparging process with suppressed surface tension, was further tested for removing organic compounds via oxidative degradation using ozone, a gaseous oxidizing agent. Surfactant-enhanced ozone sparging (SEOS), sparging ozone-containing gas during the SEAS process, has also been proven in bench scale experiments to be effective for removing less volatile, more resistant chemicals such as *n*-decane (Kim et al., 2013).

Using air as the contaminant carrier during the air sparging process, the ratio of contaminant mass to the volume of air injected onto the subsurface may be a direct indication of the efficiency of the process. If so, it will be most effective for all of the injected air to pass through the source zone to be treated, maximizing the contact between the air and contaminant. Few studies have tried to focus the injected air on the source region to improve the removal mass ratio of contaminants to the volume of injected gas.

In this study, surface tension and viscosity of the aqueous phase were modified to control the travel path of injected air through the aquifer during the air sparging process. An aqueous solution of anionic surfactant that decreased air entry pressure in the region of application leading to enhanced air intrusion and low water saturations was applied to part of a physical (bench-scale) model to investigate whether injected air can be induced to flow preferentially through the region. An aqueous solution of a viscosity-enhancing chemical (thickener) was applied to investigate the effect of increased viscosity on the travel velocity of the air–water interface during air sparging (using one-dimensional column), and the travel path of the injected air. It is hypothesized that the aqueous region with high viscosity retards air flow thus functioning as a hydraulic barrier.

The purpose of this study was to establish a method that controls the air flow paths through porous media during air sparging by modifying surface tension and viscosity. Assuming these two different physical properties (reducing surface tension and increasing viscosity) act in opposition on air intrusion velocity at the early stage of air sparging, we investigated air flow through porous media, using bench-scale models as either focused on the target region (where the surface tension was suppressed) or diverted to the region with high viscosity. The effects of modified surface tension and viscosity on the sparging influence zone were evaluated based on the air flux magnitude and distribution which were measured using a newly developed gaseous passive flux meter technique (Kim et al., 2012).

## 2. Materials and methods

### 2.1. Materials

Quartz sand with a diameter of 300–500  $\mu\text{m}$  was used as the packing porous medium. Reagent grade sodium dodecylbenzene sulfonate (SDBS) was purchased from Tokyo Kasei Kogyo Co. Ltd. (Tokyo, Japan) and used as received. Sodium carboxymethylcellulose (SCMC), fluorescein sodium salt, and *n*-octane were supplied by Sigma-Aldrich Co. (St. Louis, MO, USA). Chemicals provided by Sigma-Aldrich were all reagent grade with >99% purity. Dichloromethane (HPLC grade) was provided by J. T. Baker Co. Ltd. (Center Valley, PA, USA). Double distilled (DI) water was used throughout the experiments.

### 2.2. Experimental setup

The schematic diagram of the experimental set up for the one-dimensional experiments is shown in Fig. 1. The length and inner diameter of the glass column were 150.0 and 1.0 cm, respectively. The sand described in the previous section was packed in the column with no head space. The column was saturated with DI water or the aqueous solution of SCMC prior to air displacement experiments. The bulk density of the sand was approximately 1.63  $\text{g}/\text{cm}^3$  estimated based on the mass of sand used for packing, and porosity was 0.40 based on the water volume added. A water reservoir and gas bottle of compressed  $\text{CO}_2$  were connected to the column at one end and water-filled tubing was connected to the other end of the column with a 30 cm head difference between the end of the tubing and the center of the column.

The set up used for the two- and three-dimensional model experiments is shown in Fig. 2. Flask 1 and flask 2 installed in the set up were filled with DI water and *n*-octane, respectively. DI water was used to saturate the incoming dry air with water vapor to avoid decreasing water saturation in the models. With constant air flow through the flask filled with *n*-octane, the air entering the models contained *n*-octane vapor at constant level, and was used to measure air flux. Gas sampling valves were used to collect samples for *n*-octane analysis before and after the sparging experiments. The flow rate of air during air sparging was controlled using a fine metering valve that was connected to a compressed air bottle. The same experimental set up was used for both the two- and three-dimensional model experiments.

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