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A bio-reactive barrier sequence for petroleum hydrocarbon capture and degradation in low nutrient environments

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ABSTRACT

Sequenced permeable reactive barriers, containing materials for nutrient release and petroleum hydrocarbon adsorption, can promote biofilm formation and enhance biodegradation in low nutrient, cold environments. This study characterised the degree of biofilm formation and biodegradation of Antarctic diesel within laboratory scale permeable reactive barriers, containing ammonium exchanged zeolite for ammonium release onto a granular activated carbon bed. The adsorption of Antarctic diesel on the granular activated carbon bed resulted in low cell growth on the surface of the carbon, with biodegradation indices revealing limited access to and degradation of compounds adsorbed on the granular activated carbon bed. Under Antarctic soil water conditions, where nutrient supply is low, increased granular activated carbon porosity was not shown to enhance biofilm formation. Where exhaustion of the adsorptive material was observed, desorption of naphthalene and dodecane followed, leading to biofilm formation and petroleum hydrocarbon degradation on the surface of the material. This study has important implications for bio-reactive material selection, and the sequencing of materials within these barriers and other water treatment applications.

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1. Introduction

The migration of petroleum hydrocarbon contaminants in soil water can be effectively intercepted through the installation of permeable reactive barriers (PRB) (Mumford et al., 2013). In a PRB, soil water flows through a single material, or sequence, of reactive materials, wherein the contaminants are physically adsorbed and chemically or biologically degraded (Yeh et al., 2010). The finite operational capacity of many reactive materials, such as granular activated carbon (GAC), requires regeneration or the replacement of materials and subsequent costly disposal in appropriate landfills (Prasse et al., 2015). In the context of petroleum hydrocarbon remediation in remote and cold regions, such as the Antarctic, high transport and disposal costs challenge the feasibility of remediation technologies often employed in temperate environments (Camenzuli and Freidman, 2015).

One strategy to overcome these limitations is the use of biofilms in so called bio-reactive barriers. The interaction between petroleum hydrocarbons and reactive materials provides a carbon supply for the attachment and growth of microorganisms transported in the soil water. Where this flow is sufficient for growth, this can result in biofilm formation generating a bio-reactive barrier (Mumford et al., 2015). Growth can be promoted in the bio-reactive barrier if nutrient supply is controlled and the media composition is uniform (Careghini et al., 2013). As a result, biodegradation can occur in a more controlled manner than in soil, enhancing the performance of the reactive material and the longevity of petroleum hydrocarbon treatment within a PRB. This can facilitate a lower frequency of material replacement and disposal, leading to reduced costs (Snape et al., 2001).

Sequenced PRBs have previously been installed in the Antarctic to intercept and degrade petroleum hydrocarbon spills (Mumford et al., 2013; Statham et al., 2016). These PRBs contained materials for controlled nutrient release, as well as fuel capture and

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biodegradation (Mumford et al., 2013). Mumford et al. (2015) reports that the use of nutrient amended zeolites promoted large populations of hydrocarbon degrading bacteria. More recent studies have also supported the increased biodegradation of petroleum hydrocarbons in the presence of nutrient amended zeolites (Freidman et al., 2016a). Yet, despite the high retention of petroleum hydrocarbons on GAC and high cell counts observed, low degradation was recorded (Mumford et al., 2015). This suggests that microbial cells attached to the GAC may be limited in their ability to desorb and degrade petroleum hydrocarbon contaminants adsorbed to the GAC. The heterogeneous pore structure of GACs, in which micropores (<2 nm Ø), mesopores (2–50 nm Ø) and macropores (>50 nm Ø) are present, may be a contributing factor (Aktas & Cecen, 2007). GAC is primarily comprised of micropores, however the relative proportion of pore types can vary between carbonaceous materials and activation methods (Aktas & Cecen, 2007).

Two mechanisms have been reported to contribute to the biological desorption of petroleum hydrocarbons and regeneration of GAC (Aktas & Cecen, 2007). These are 1.) desorption due to a concentration gradient induced by biofilm activity, and 2.) desorption due to exo-enzymatic reactions (Aktas & Cecen, 2007). In the first mechanism the concentration of petroleum hydrocarbons in the liquid phase is reduced due to microbial activity. Desorption then occurs from the GAC and petroleum hydrocarbons become available for further biofilm formation (Ng et al., 2010). In the second mechanism, enzymes secreted by microbial cells diffuse into the pores of the GAC and react with the adsorbed petroleum hydrocarbon compounds causing hydrolytic transformation (Ng et al., 2010). Xiaojian et al. (1991) suggest that many exo-enzymes are too large to facilitate desorption across much of the GAC pore network. It is estimated that for enzyme catalysed desorption to occur, the diameter of the GAC pore must be at least three times greater than the size of the enzyme. As the average molecular diameter of exo-enzymes is greater than 3–4 nm, Xiaojian et al. concluded that the pore diameter must be larger than 10 nm. This excludes the micropores as sites available for desorption but still leaves the mesopores and macropores as possibilities for enzyme activity.

The importance of pore size has also been shown in studies where mesopores within GAC were more efficiently regenerated than micropores, due to the small pore diameter of micropores and overlapping of adsorption forces from the opposite walls within these pores (Bansal & Goyal, 2005; Putz et al., 2005). Other factors that can also influence the accessibility of petroleum hydrocarbons on GAC include the carbon activation process, which has been found to be highly irreversible on thermally activated carbons (Aktas & Cecen, 2007). Thermal activation procedures in an inert atmosphere can remove surface oxide groups from GAC, resulting in a highly reactive pore network that strongly binds organics when in contact with water (Aktas & Cecen, 2006).

A comprehensive investigation into the biological performance of GAC in a sequenced PRB is challenging at bulk scale for several reasons. Firstly, the contaminant flux and hydraulic conductivity of PRBs in environments subject to freeze-thaw cycling is variable (Mumford et al., 2014). Bulk scale mixing of PRB materials and the presence of multiple petroleum hydrocarbon products may also conceal the contribution of individual materials to biodegradation (Mumford et al., 2013). While previous works have shown that nutrient amended zeolites can enhance the biodegradation of petroleum hydrocarbons in soil water (Freidman et al., 2016a), there are further challenges to be overcome. Lag phases have been observed in biofilm formation and degradation of petroleum hydrocarbons is incomplete, suggesting that adsorption materials are required to manage risk and prevent migration.

This study builds upon the previous research of Mumford et al. (2015) and Freidman et al. (2016a) by investigating the biodegradation of special Antarctic blend (SAB) diesel within a laboratory scale sequenced PRB (Fig. 1a–f). SAB diesel is primarily comprised of n-C_9-C_{14} alkanes and is the most commonly used fuel by Australia in Antarctica (Snape et al., 2005). The laboratory design was chosen to investigate the influence of nitrogen delivery, achieved through soil cation interaction with ammonium exchanged zeolite, for the promotion of biofilm formation and biodegradation of petroleum hydrocarbons on GAC (Fig. 1a–c). Ammonium exchanged zeolite was replaced with inert sand in control flow cells (Fig. 1b). Furthermore, the influence of low temperature (Fig. 1d), porosity (Fig. 1e) and SAB diesel concentration (Fig. 1f) were investigated alongside the initial sequence (Fig. 1a) to expand the application of this study to potential temperate water treatment applications.

2. Materials and methods

2.1. Sequenced PRB materials

The conditioning of ammonium exchanged zeolite and the physicochemical properties of Antarctic soil collected from Australia's Casey Station are reported in previous works (Freidman et al., 2016a). The surface area of GAC and zeolites was determined by nitrogen (N_2) adsorption at 77 K (−196 °C) using the Micrometrics Accelerated Surface Area and Porosimetry System (ASAP 2010, Micrometrics) and the Brunauer-Emmett-Teller method recommended by the manufacturer (Table 1). The limitations of N_2 adsorption are that microporous silica plugs and high cation loading of ammonium exchanged zeolite can restrict the diffusion of N_2, potentially resulting in low surface area measurements (Table 1) (Groen et al., 2003). Similarly the large N_2 molecule can interact with impurities and be prevented from entering the micropores on GAC, affecting surface area and pore size distribution measurements (Groen et al., 2003). The pore size distribution was determined using density functional theory (Table 1). The laboratory PRB sequences used in this study are reported in Fig. 1.

Two commercial GAC materials, derived from renewable resources, were examined for the assessment of cell attachment and biofilm formation (Table 1). The wood based GAC (PICABIOL) has an open honeycomb pore structure, large enough to accommodate microbial cells (Freidman et al., 2016b). The high pore volume is generated by contacting the raw material with phosphoric acid to swell the wood and open up the cellulose structure (Bansal & Goyal, 2005). This honeycomb morphology allows SAB diesel compounds to pass rapidly within mesopores and micropores situated deeper within the PICABIOL GAC particle (Bansal & Goyal, 2005). The second material was a coconut based product, GC1200 GAC, which has a uniform microporous structure and high hardness rating. A high hardness rating makes the material suitable for applications under conditions of freeze-thaw cycling (Mumford et al., 2014). The GC1200 GAC is currently used for water treatment applications in Antarctica (Statham et al., 2016), while PICABIOL GAC has been reported to retain a large population of cells in pilot tests assessing material performance, suggesting its potential promise for Antarctic remediation (Snape et al., 2001).

The SAB diesel adsorption capacity on PICABIOL GAC and GC1200 GAC materials was achieved under mixed batch conditions at 23 °C (Table 1). Duplicate samples of 4 g of carbon were each mixed with a 100 ml solution of 10,000 mg l^{-1} SAB diesel in deionised water at 50 rpm for 24 h. The SAB diesel solution was replaced every 24 h across a 72 h period. Adsorption on GAC may have occurred through contact with SAB diesel in the aqueous phase, although the solubility is low (~7 mg l^{-1}), or by contact with free phase SAB diesel. Both mechanisms have been observed

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