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Research article

Ethanol content in different gasohol blend spills influences the decision-making on remediation technologies

Leonardo Vilela Steiner [a], Débora Toledo Ramos [a], Ana Maria Rubini Liedke [a], Maria Pilar Serbent [b], Henry Xavier Corseuil [a, *]

[a] Federal University of Santa Catarina, Department of Sanitary and Environmental Engineering, Florianópolis, Santa Catarina, Brazil
[b] State University of Santa Catarina, Department of Sanitary Engineering, Ibirama, Santa Catarina, Brazil

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ABSTRACT

Gasohol blend spills with variable ethanol content exert different electron acceptor demands in groundwater and the distinct dynamics undergone by these blends underscores the need for field-based information to aid decision-making on suitable remediation technologies for each gasohol blend spill. In this study, a comparison of two gasohol releases (E10 (10:90 ethanol and gasoline, v/v) and E25 (25:75 ethanol and gasoline, v/v) under monitored natural attenuation (MNA) and nitrate biostimulation, respectively) was conducted to assess the most effective remediation strategy for each gasohol release. Microbial communities were assessed to support geochemical data as well as to enable the characterization of important population shifts that evolve during biodegradation processes in E25 and E10 field experiments. Results revealed that natural attenuation processes sufficiently supported ethanol and BTEX compounds biodegradation in E10 release, due to the lower biochemical oxygen demand they exert relative to E25 blend. In E25 release, nitrate reduction was largely responsible for BTEX and ethanol biodegradation, as intended. First-order decay constants demonstrated that ethanol degradation rates were similar ($p < 0.05$) for both remediation technologies (2.05 ± 0.15 and 2.22 ± 0.23, for E25 and E10, respectively) whilst BTEX compounds exhibited different degradation rates ($p > 0.05$) that were higher for the experiment under MNA (0.33 ± 0.06 and 0.43 ± 0.03, for E25 and E10, respectively). Therefore, ethanol content in different gasohol blends can influence the decision-making on the most suitable remediation technology, as MNA processes can be applied for the remediation of gasohol blends with lower ethanol content (i.e., 10% v/v), once the aquifer geochemical conditions provide a sufficient electron acceptor pool. To the best of our knowledge, this is the first field study to monitor two long-term gasohol releases over various time scales in order to assess feasible remediation technologies for each scenario.

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1. Introduction

The dependence of fossil fuels and the potential threats they can pose to the environment have boosted the development and use of alternative renewable fuels (Schnoor, 2006). Ethanol has been increasingly added to the worldwide energy matrix, typically through gasoline-blended formulations to alleviate dependence on fossil fuels and reduce the environmental issues associated with fossil fuels (Goldemberg, 2007). In Brazil, commercial gasoline has an ethanol mandatory blending percentage of 27% (Brazil, 2015), while in the United States 10% of ethanol is blended into gasoline formulations (US EPA, 2015). In EU member states, the current blending percentage of ethanol to gasoline is up to 10% (European Parliament, 2009) but countries such as Spain, Germany, Italy and the United Kingdom opted for a 5% ethanol percentage to the commercial gasoline (European Environmental Agency, 2015). In the Asian continent, China primarily uses pure gasoline and diesel as commercial fuels (USDA, 2007) followed by E10 blends (10:90 ethanol and gasoline, %) that are used in 9 of their 22 provinces (Pang et al., 2008). In India, 5% of ethanol is blended into gasoline in 11 states (Sukumaran et al., 2010). As fuel leaks and spills are commonly observed during storage and transport (Das and Chandran, 2011), this can lead to increasing contaminated sites by the widely used ethanol-blended gasoline fuel. Since these

* Corresponding author.
E-mail address: henry.corseuil@ufsc.br (H.X. Corseuil).

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formulations contain priority contaminants such as BTEX (benzene, toluene, ethylbenzene and xylenes), they require remedial actions when released to the environment.

Monitored natural attenuation (MNA) is a well-established strategy to remediate contaminated sites that relies on natural attenuation processes to achieve remediation goals within a reasonable time frame. MNA is minimally invasive and the cost of implementation and monitoring is relative low (Adriano et al., 2004; Blum et al., 2009; Corseuil et al., 2011; Kao et al., 2006; Khan and Husain, 2003; Mackay et al., 2006; Naidu et al., 2012). The efficiency and applicability of MNA depends primarily on the site characteristics, the time needed to remove contaminants and potential risks to human health (Khan et al., 2004). When natural attenuation processes are insufficient to reduce contaminants concentration or when the time required or risk involved are not compatible with natural attenuation processes, active remediation technologies (i.e., biostimulation) can be applied to speed up contaminants attenuation and meet remediation goals.

The high biochemical oxygen demand (BOD) commonly exerted by ethanol leads to the exhaustion of the available electron acceptors (Da Silva and Alvarez, 2002) and thereby gasohol blends with higher ethanol content would require a greater stoichiometric electron acceptor demand. To exemplify, for a 100L-spill of E25 and E10 blends, the theoretical BOD for ethanol biodegradation (reaction (1)), according to McCarty (1969) model, would be 2.5 times higher for E25 as compared to E10 blend (calculations provided in SI). Therefore, the enhanced consumption of electron acceptors can make natural attenuation processes unfeasible to deal with gasohol blends spills with high ethanol content. In this case, engineering interventions (i.e., active remediation technologies) may be required to avoid persistent contaminants concentrations.

$$CH_3CH_2OH + 1.1O_2 + 0.4HCO_3^- + 0.4NH_4^+ \rightarrow 0.4C_5H_7O_2N + 0.5CO_2 + 2.8H_2O \quad (1)$$

Remediation technologies can be either aerobic or anaerobic and the decision-making is dependent on the scenario of the contaminated site. Although aerobic strategies generally exhibit faster degradation rates (Corseuil et al., 1998; Ruiz-Aguilar et al., 2003), they are not universally applicable as hydrocarbons contaminated sites are invariably anaerobic due to the rapid oxygen consumption by indigenous microorganisms. Therefore, the majority of hydrocarbon contaminants are degraded by anaerobic microorganisms, which makes anaerobic technologies more suitable to deal with gasohol releases.

Among the existing anaerobic strategies, nitrate biostimulation that refers to the use of nitrate as terminal electron acceptor to enhance the conversion of organic compounds into carbon dioxide and water (Wilson and Bouwer, 1997), is widely applied for the remediation of aromatic compounds (Cunningham et al., 2001; Da Silva et al., 2005; Hutchins et al., 1991; Schreiber and Bahr, 2002; Wilson and Bouwer, 1997). The broad use of nitrate biostimulation can be explained by (1) the higher oxidation potential provided (0.25–0.85 V) as compared to other anaerobic processes such as iron reduction (0.10 to −0.50 V), sulfate reduction (−0.20 to −0.70 V) or methanogenesis (−0.25 to −0.75 V) (Christensen et al., 2000; Stumm and Morgan, 1996), (2) the high solubility of nitrate salts that facilitates the injection through the site and (3) the relatively low cost (Hutchins et al., 1998; Korda et al., 1997).

Nitrate biostimulation usually involves the continuous injection of nitrate salts into the groundwater. Nevertheless, some aquifers may already have considerable background concentrations of nitrate, as is the case of areas under agricultural activities that usually exhibit significant amounts of fertilizer-derived nitrate (Galloway et al., 2004; Sebilo et al., 2013). Thus, depending on the scenario of the contaminated site, nitrate reduction or other redox processes (such as iron or sulfate reduction) could occur naturally and this must be taken into account before deciding whether to apply active remediation technologies or to rely on monitored natural attenuation processes.

Given the several gasohol blends that are currently used worldwide and the associated risk of spills that require remedial actions, these field studies will advance the current understanding on the complex dynamics undergone by different gasohol releases in groundwater and overall site management. Furthermore, the information obtained can be potentially used for the development of risk assessment models to confidently predict the behavior and biodegradation of different gasohol blends in real environments, thus underscoring the need for field-based information. This can aid decision-making process on the most suitable remediation strategy by enabling a more cost-effective and targeted response to different gasohol blends spills, which correspond to the main concerns of cleanup decisions.

This study presents two long-term field experiments (monitored over 11 and 6 years) of different gasoline-ethanol blends (E25 (25:75 ethanol and gasoline v/v) and E10 (10:90 ethanol and gasoline v/v)) under nitrate biostimulation and natural attenuation that were conducted to assess the most effective remediation strategy. To the best of our knowledge, this is the first field study to monitor two different gasohol releases over various time scales in order to assess feasible remediation technologies for each scenario.

2. Materials and methods

2.1. Field experiments

Two field experiments were conducted in neighboring areas (located at a distance of 23 m) at Ressacada Experimental Farm in Florianópolis, SC, Brazil. The experiments were established by the release of 100 L of E25 (25:75 ethanol and gasoline, v/v) and E10 (10:90 ethanol and gasoline, v/v) into source-zone areas of 1.0 m × 1.0 m for E25 and 1.5 m × 1.0 m for E10, at the water table level (Fig. 1). Geological characterization of the sites were previously described (Da Silva and Corseuil, 2012). Multilevel wells were installed in E25 (6 injection wells and 64 sampling wells) and E10 (58 sampling wells). A peristaltic pump and Teflon tubing were used to collect samples at different depths (2, 3, 4, 5 and 6 m below ground surface [bgs] for E10 and 2.3, 2.8, 3.8, 4.8 and 5.8 m bgs for E25) to capped sterile vials without headspace. The levels that exhibited the most significant concentration of ethanol and BTEX were presented in the results.

Ressacada Experimental Farm has a natural availability of electron acceptors (Table 1). The background nitrate concentrations are possibly present due to previous cattle farming activities in the area, while sulfate is likely related to minerals (i.e. pyrite) that infiltrate from soil and are dissolved into the groundwater. These background concentration of electron acceptors were already presented in other Ressacada field studies (Corseuil et al., 2011; Müller et al., 2017; Ramos et al., 2013). Therefore, in E10 site, monitored natural attenuation was conducted to evaluate whether the natural availability of electron acceptors (i.e., nitrate and sulfate) could be sufficient to support ethanol and BTEX biodegradation. In E25 site, nitrate was injected as a supplementary source of electron acceptor to stimulate nitrate reduction processes and enhance organic contaminants biodegradation. Injections initiated 2 months after E25 was released and were performed by the release of 5 L of $NaNO_3$ (4 g L^{-1}) into the injection wells three times a week

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