

A strategy for aromatic hydrocarbon bioremediation under anaerobic conditions and the impacts of ethanol: A microcosm study

Yu Dao Chen ^{a,*}, James F. Barker ^b, Lai Gui ^b

^a Department of Resource and Environmental Engineering, Guilin University of Technology, Guilin, Guangxi, 541004, China

^b Department of Earth Science, University of Waterloo, Waterloo, Ont., Canada N2L 3G1

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Abstract

Increased use of ethanol-blended gasoline (gasohol) and its potential release into the subsurface have spurred interest in studying the biodegradation of and interactions between ethanol and gasoline components such as benzene, toluene, ethylbenzene and xylene isomers (BTEX) in groundwater plumes. The preferred substrate status and the high biological oxygen demand (BOD) posed by ethanol and its biodegradation products suggests that anaerobic electron acceptors (EAs) will be required to support in situ bioremediation of BTEX. To develop a strategy for aromatic hydrocarbon bioremediation and to understand the impacts of ethanol on BTEX biodegradation under strictly anaerobic conditions, a microcosm experiment was conducted using pristine aquifer sand and groundwater obtained from Canadian Forces Base Borden, Canada. The initial electron acceptor pool included nitrate, sulfate and/or ferric iron. The microcosms typically contained 400 g of sediment, 600–800 ml of groundwater, and with differing EAs added, and were run under anaerobic conditions. Ethanol was added to some at concentrations of 500 and 5000 mg/L.

Trends for biodegradation of aromatic hydrocarbons for the Borden aquifer material were first developed in the absence of ethanol. The results showed that indigenous microorganisms could degrade all aromatic hydrocarbons (BTEX and trimethylbenzene isomers-TMB) under nitrate- and ferric iron-combined conditions, but not under sulfate-reducing conditions. Toluene, ethylbenzene and *m/p*-xylene were biodegraded under denitrifying conditions. However, the persistence of benzene indicated that enhancing denitrification alone was insufficient. Both benzene and *o*-xylene biodegraded significantly under iron-reducing conditions, but only after denitrification had removed other aromatics. For the trimethylbenzene isomers, 1,3,5-TMB biodegradation was found under denitrifying and then iron-reducing conditions. Biodegradation of 1,2,3-TMB or 1,2,4-TMB was slower under iron-reducing conditions. This study suggests that addition of excess ferric iron combined with limited nitrate has promise for in situ bioremediation of BTEX and TMB in the Borden aquifer and possibly for other sites contaminated by hydrocarbons. This study is the first to report 1,2,3-TMB biodegradation under strictly anaerobic condition.

With the addition of 500 mg/L ethanol but without EA addition, ethanol and its main intermediate, acetate, were quickly biodegraded within 41 d with methane as a major product. Ethanol initially present at 5000 mg/L without EA addition declined slowly with the persistence of unidentified volatile fatty acids, likely propionate and butyrate, but less methane. In contrast, all ethanol disappeared with repeated additions of either nitrate or ferric iron, but acetate and unidentified intermediates persisted under iron-enhanced conditions.

With the addition of 500 mg/L ethanol and nitrate, only minor toluene biodegradation was observed under denitrifying conditions and only after ethanol and acetate were utilized. The higher ethanol concentration (5000 mg/L) essentially shut down BTEX biodegradation likely due to high EA demand provided by ethanol and its intermediates. The negative findings for anaerobic BTEX biodegradation in the presence of ethanol and/or its biodegradation products are in contrast to recent research reported by Da

* Corresponding author. Fax: +86 773 5897019.

E-mail address: cyd0056@vip.sina.com (Y.D. Chen).

Silva et al. [Da Silva, M.L.B., Ruiz-Aguilar, G.M.L., Alvarez, P.J.J., 2005. Enhanced anaerobic biodegradation of BTEX-ethanol mixtures in aquifer columns amended with sulfate, chelated ferric iron or nitrate. *Biodegradation*, 16, 105–114]. Our results suggest that the apparent conservation of high residual labile carbon as biodegradation products such as acetate makes natural attenuation of aromatics less effective, and makes subsequent addition of EAs to promote in situ BTEX biodegradation problematic. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Aromatic hydrocarbons such as benzene, toluene, ethylbenzene and xylenes and trimethylbenzenes (often referred to as BTEX and TMB) are primary constituents of gasoline. For example, American Petroleum Institute (API. 91–01 gasoline contains 27 wt.% of BTEX and TMB (Barbaro et al., 1999). Aromatic hydrocarbons are of particular concern in gasoline contaminated groundwater, given their high solubility, mobility and toxicity. Biodegradation of BTEX has been demonstrated under aerobic condition (Barker et al., 1987; Alvarez and Vogel, 1995), however, the activity of aerobic hydrocarbon-degrading microorganisms can quickly exhaust limited dissolved oxygen in groundwater, which has a low solubility in water. Therefore, anaerobic conditions will often form in a plume of petroleum hydrocarbons and anaerobic biodegradation usually plays an important role in removing hydrocarbon contaminants (Cozzarelli et al., 2001).

Under anaerobic conditions, benzene biodegradation can be variable (Johnson et al., 2003). Although benzene has been shown to biodegrade under denitrifying, iron-reducing, sulfate-reducing and methanogenic conditions (Lovley et al., 1994; Kazumi et al., 1997; Cozzarelli and Baehr, 2003), many field studies reported that benzene was recalcitrant under denitrifying (Hutchins, 1991), iron-reducing (Phelps and Young, 1999), and sulfate-reducing (Edwards et al., 1992; Ball and Reinhard, 1996; Davis et al., 1999) conditions. At Canadian Forces Base (CFB) Borden, field and supporting laboratory studies have been conducted to understand the biodegradability of BTEX under aerobic and anaerobic conditions. The BTEX compounds biodegraded rapidly under aerobic conditions and oxygen availability was found to be the limiting process in plumes derived from controlled sources (Barker et al., 1987, Schirmer et al., 1999; King et al., 1999). Under denitrifying conditions, BTX were observed to degrade in microcosms (Major et al., 1988). However, in subsequent studies (Barbaro et al., 1992; Acton and Barker, 1992), only toluene was observed to degrade quickly in field experiments. Benzene was

recalcitrant and ethylbenzene and xylenes were only incompletely biodegraded. Based on these subsequent studies, addition of nitrate was considered inefficient for BTEX bioremediation due to the recalcitrance of benzene. Under sulfate-reducing conditions, Acton and Barker (1992) found only toluene biodegraded. In addition to denitrification and sulfate reduction, little evidence of other anaerobic electron acceptor (EA) processes is available under Borden conditions. For example, natural gradient field experiments (e.g., King and Barker, 1999) and in situ column experiments (Acton and Barker, 1992) demonstrate little generation of Fe^{2+} and methane even when excess hydrocarbons were available. This is in contrast to aquifer systems studied by Cozzarelli et al. (2001) and Cozzarelli et al. (1999) where iron reduction was a very significant EA process. Therefore, while the potential for anaerobic biodegradation of benzene exists, this potential is not realized at most sites for reasons that remain unclear (Kazumi et al., 1997).

In addition, a change of gasoline formulation may impact the fate of BTEX in contaminated groundwater. Ethanol is a likely replacement for methyl tertiary-butyl ether (MTBE) as a gasoline oxygenate. In fact, about half of gasoline is amended with 22% ethanol (by volume) in Brazil, and many US states use 10% ethanol blended gasoline (Powers et al., 2001). The ethanol-blended gasoline is called gasohol. A few provinces in Canada, China and Australia are or are encouraging blending 10% ethanol into gasoline. Increasingly, ethanol will be a new constituent along with BTEX in groundwater contaminated by gasohol.

Ethanol itself will not pose a risk to drinking water sources. However, ethanol and its biotransformation intermediates such as acetate (Kim et al., 1994; Wu and Hickey, 1996) can rapidly consume existing EAs that could otherwise have been used to biodegrade aromatics. A relevant field study was conducted with methanol (Barker et al., 1992) which could cause similar effects as ethanol. Two mixtures of gasoline-contacted groundwater containing BTEX only and BTEX with methanol were injected into the research aquifer at CFB Borden. Here, BTEX biodegradation was dominantly

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