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Simultaneous anaerobic transformation of carbon tetrachloride to carbon dioxide and tetrachloroethene to ethene in a continuous flow column

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ABSTRACT

The simultaneous anaerobic transformation of tetrachloroethene (PCE) and carbon tetrachloride (CT) was evaluated in a continuous flow column. The column was packed with quartz sand and bioaugmented with the Evanite culture (EV) that is capable of transforming PCE to ethene. Azizian and Semprini (2016) reported that PCE and CT could be simultaneously transformed in the column, with PCE (0.1 mM) transformed mainly to ethene and CT (0.015 mM) to chloroform (CF) (20%) and an unknown transformation product, likely carbon dioxide (CO_2). The fermentation of propionate, formed from lactate fermentation, was inhibited after the transformation of CT, likely from the exposure to CF. Reported here is the second phase of that study where a second bioaugmentation of the EV culture was made to reintroduce a lactate and propionate fermenting population to the column. Effective lactate and propionate fermentation were restored with a H_2 concentration of ~25 nM maintained in the column effluent. PCE (0.1 mM) was effectively transformed to ethene (~98%) and vinyl chloride (VC) (~2%). Unlabeled CT (0.015 to 0.03 mM) was completely transformed with a transient build-up of CF and chloromethane (CM), which were subsequently removed below their detection limits. A series of transient tests were initiated through the addition of carbon-13 labeled CT (^{13}CT), with concentrations gradually increased from 0.03 to 0.10 mM. GC–MS analysis of the column effluent showed that ^{13}C labeled CO_2 ($^{13}CO_2$) was formed, ranging from 82 to 93% of the ^{13}CT transformed, with the transient increases in $^{13}CO_2$ associated with the increased concentration of ^{13}CT. A modified COD analysis indicated a lesser amount of ^{13}CT (18%) was transformed to soluble products, while $^{13}CO_2$ represented 82% the ^{13}CT transformed. In a final transient test, the influent lactate concentration was decreased from 1.1 to 0.67 mM. The transformation of both CT and PCE changed dramatically. Only 59% of the ^{13}CT was transformed, primarily to CF. $^{13}CO_2$ concentrations gradually decreased to background levels, indicating CO_2 was no longer a transformation product. PCE transformation resulted in the following percentage of products formed: cDCE (60%), VC (36%), and ethene (4%). Incomplete propionate fermentation was also observed, consistent with the build-up of CF and the decrease in H_2 concentrations to approximately 2 nM. The results clearly demonstrate that high concentrations of CT were transformed to CO_2, and effective PCE dehalogenation to ethene was maintained when excess lactate was fed and propionate was effectively fermented. However, when the lactate concentration was reduced, both PCE and CT transformation and propionate fermentation were negatively impacted.

1. Introduction

Many groundwater sites are contaminated with mixtures of chlorinated aliphatic hydrocarbons (CAHs), including tetrachloroethene (PCE), trichloroethene (TCE), 1,2-dichloroethene (cDCE), vinyl chloride (VC), carbon tetrachloride (CT), and chloroform (CF) (Moran et al., 2007). Contaminant mixtures present a challenge when biological remediation processes are being considered. CT is of particular interest since a broad range of transformation products can be potentially formed under anaerobic conditions (Hashsham and Freedman, 1999), and CT and CF can inhibit organohalide respiration. CT and CF have been observed to inhibit and potentially exert toxicity on reductive dehalogenation of PCE and TCE (Maymó-Gatell et al., 2001; Bagley et al., 2000; Bagley and Gossett, 1995; He et al., 2005). Koenig et al. (2014) found CAHs with higher octanol water partition coefficients (PCE and CT) were more inhibitory to fermenting and organohalide respiring bacteria (OHRB) than CF and 1,2-dichoroethane (1,2DCA), with OHRB being more sensitive.

Abiotic transformation of CT has been reported in laboratory studies (Criddle and McCarty, 1991; Kriegman-King and Reinhard, 1992;

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Ammonette et al., 2000; Butler and Hayes, 2000) and in a field study and column study (Devlin and Mueller, 1999). CT either directly hydrolyzes to CO_2, or, under reducing conditions with electron donors such as sulfide, ferrous iron (Fe(II)), or iron sulfide (FeS), CT is transformed to carbon disulfide (CS_2) and CO_2 (Hashsham et al., 1995). In a field study and subsequent column study under sulfate reducing conditions, Devlin and Mueller (1999) found CT was primarily transformed to CF and CS_2 at a ratio of 2:1. The abiotic transformation by FeS was found to yield the 2:1 ratio of CF to CS_2. Penny et al. (2010) provide a thorough review of abiotic and biotic transformations of CT.

A *Dehalobacter* strain that obtains energy for growth via organohalide respiration of CF to dichloromethane (DCM) has been studied by Grostern et al. (2010). The reductase gene CfrA responsible for the transformation of CF to DCM has also been identified (Tang and Edwards, 2013). Dehalogenation of CF to DCM and DCM fermentation to acetate, formate, and CO_2 has been reported by Cappelletti et al. (2012); Criddle and McCarty (1991); de Best et al. (1998); Hashsham and Freedman (1999); and Lee et al. (2012). DCM and chloromethane (CM) can also be fermented to CO_2 and acetate by anaerobic acetogenic bacteria (Leisinger et al., 1994; Mägli et al., 1996; Meßmer et al., 1993). Justicia-Leon et al. (2012) also reported DCM as a sole substrate to support an enriched *Dehalobacter* culture that ferments DCM to CO_2 and acetate. Recently, an anaerobic DCM degrdading bacteria, *Candidatus dichloromethanomonas elyunquensis,* has been identified that grows on DCM and produces chloride and acetate (Kleindienst et al., 2017). Microcosm studies have been performed by Justicia-Leon et al. (2014) in which both the *Dehalobacter* strain that transforms CF to DCM (Grostern et al., 2010) and the strain that ferments DCM to CO_2 and acetate (Justicia-Leon et al., 2012) were bioaugmented and biostimulated to achieve the transformation of CF to non-chlorinated products. In a previous field study, CF was observed as a transformation product of CT and represented 30–40% of the CT transformed (Semprini et al., 1992). The ability to limit CF formation is critical for effective in-situ anaerobic treatment of groundwater containing mixtures of CT and PCE or TCE.

There are a number of abiotic and biotic pathways to convert CT to CO_2 and other products. Fig. 1 shows the major potential intermediate products that might be observed during CT transformation. The diagram was adapted from that proposed by Hashsham and Freedman (1999) and Hashsham et al. (1995) from studies with *Acetobacterium woodii*. The figure also includes the pathways for the fermentation of DCM and CM to CO_2 and acetate. An active microbial population may also play a role in the CT transformation, providing biomoelcules that initiate radical formation (Shan et al., 2010; Becker and Freedman, 1994; and Hashsham and Freedman, 1999). In addition, organohalide

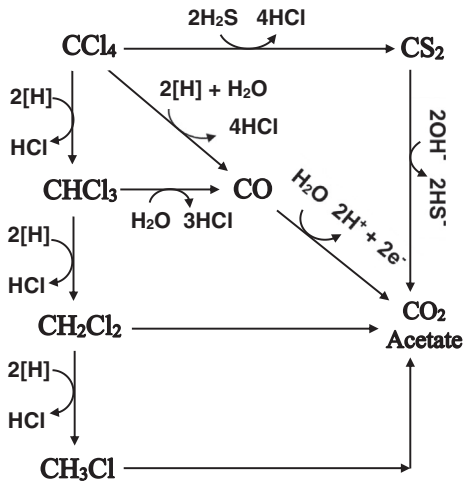

Fig. 1. Potential products from abiotic and biotic transformation of CT. Modified after Hashsham and Freedman, 1999 and Miriam et al., 1998.

respiration of CF to DCM and DCM fermentation are biotic pathways for CF to be converted to CO_2 and other non-toxic products (Lee et al., 2012).

Previous biotransformation studies have used ^{13}C to track CT and CF transformation. Lewis and Crawford (1995) used ^{13}CT to demonstrate the transformation of CT to CO_2 and nonvolatile products without the formation of CF by *Pseudomonas* sp. strain KC, later named *Pseudomonas stutzeri* strain KC. de Best et al. (1998) added ^{13}CT to an anaerobic packed bed reactor to demonstrate that a small fraction of the ^{13}CT transformed was observed as acetate. However, they were not successful in demonstrating that $^{13}CO_2$ was produced. Lee et al. (2012) used ^{13}C labeled DCM to evaluate the potential fermentation products, and observed the production of ^{13}C labeled acetate. ^{13}C labeled Polycyclic Aromatic Hydrocarbons (PAHs) have been used in soil biotransformation studies to demonstrate the mineralization to $^{13}CO_2$. Richnow et al. (1998), for example, showed that [9-^{13}C]-anthracene was mineralized to $^{13}CO_2$ in soil microbial studies. We chose to use ^{13}CT instead of ^{14}CT to avoid the need to use a radiolabeled compound and to be able to use standard GC–MS analysis to measure the $^{13}CO_2$. We also used a modified COD analysis to determine if nonvolatile products, such as acetate, were formed.

Recently Azizian and Semprini (2016) showed PCE and CT could be simultaneously transformed in a column bioaugmented with the Evanite culture (EV), a microbial consortium capable of anaerobically transforming PCE to ethene. CT was completely transformed to CF (20%), trace amounts of CM, and an unknown transformation product expected to be CO_2. PCE was effectively transformed to ethene (~98%) and VC (~2%) in the presence of CF when formate was added as an electron donor. However, fermentation of lactate, and even more significantly of propionate were negatively impacted after the exposure to CF. CF has been previously shown to inhibit propionate-consuming anaerobic systems (Rhee and Speece, 1992). CF has also been observed to inhibit the reductive dechlorination of VC (Wei et al., 2016).

The results from our previous column study raised some interesting questions that were addressed in the current study: 1) Was a large fraction of the CT transformed to CO_2? 2) Could effective lactate and propionate fermentation be established with a second bioaugmentation of the EV culture? 3) Could the transformation of high concentrations of CT be achieved while maintaining effective transformation of PCE to ethene, with limited formation of CF? and 4) How would PCE and CT transformation be affected when the influent lactate concentration was lowered? In order to answer these questions, a series of transient experiments were conducted as a continuation of the column study presented by Azizian and Semprini (2016). ^{13}CT was added to the column to quantify the transformation of CT to CO_2 and non-volatile products. The goal was to demonstrate that the simultaneous transformation of PCE and CT to non-toxic end products could be achieved under anaerobic conditions in a continuous flow column fed lactate.

2. Materials and methods

2.1. Chemicals

CAHs and ethene for the column feed and analytical standards were: PCE (99.9%), CF (99.5%), and cDCE (97%) (Acros Organics, Pittsburgh, PA, USA). VC (99.5%), CT (99.9%), ^{13}CT (99.9%), CM (99.5%), and ethene (99.5%) (Aldrich Chemical, Milwaukee, WI, USA). Hydrogen (H_2) gas (99.99%), and N_2:CO_2 (90:10) mixed gas were supplied by Airco, Inc., Albany, OR, USA.

2.2. Column material, construction, and operation

The studies were conducted in a fabricated stainless steel column (standard molybdenum-bearing grade 316, 30 cm L × 7.5 cm ID) packed with quartz Ottawa sand (20/30 mesh; Langston Company Inc.). Prior to column packing the Ottawa sand was autoclaved. The column

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