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1,1,2,2-Tetrachloroethane aerobic cometabolic biodegradation in slurry and soil-free bioreactors: A kinetic study

Dario Frascari[a,*], Martina Cappelletti[b], Stefano Fedi[b], Davide Zannoni[b], Massimo Nocentini[a], Davide Pinelli[a]

[a] Department of Chemical, Mining and Environmental Engineering – University of Bologna, Via Terracini 28, 40131 Bologna, Italy
[b] Department of Evolutionary and Experimental Biology – University of Bologna, Via Irnerio 42, 40126 Bologna, Italy

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ABSTRACT

In this work the aerobic cometabolic biodegradation of 1,1,2,2-tetrachloroethane (TeCA) by propane-utilizing bacteria was studied in slurry bioreactors containing soil and groundwater from 5 aquifers as well as in soil-free bioreactors. The main goals were: (a) to identify and calibrate a kinetic model of TeCA cometabolism; (b) to select and characterize a TeCA-degrading bacterial consortium; (c) to compare the results obtained in slurry and in soil-free bioreactors. The results showed that 4 of the 5 tested aquifers contain TeCA-degrading bacteria, indicating that aerobic cometabolism is a potentially effective approach for TeCA-contaminated aquifers. In bioaugmentation tests, a TeCA-cometabolizing consortium developed in the slurry bioreactors induced a strong reduction of the lag-time for the onset of TeCA cometabolism. The soil-free tests yielded a satisfactory TeCA degradation performance, indicating that on-site soil-free bioreactors represent an interesting technical solution for the aerobic cometabolic bioremediation of CAH-contaminated groundwaters. The mineralization of the organic Cl was equal to about 97%. The prolonged TeCA biodegradation determined a progressive selection of the bacterial strains more effective in TeCA degradation and less affected by degradation product toxicity. The tested Michaelis–Menten-based kinetic model proved an effective tool to interpret the experimental data of TeCA aerobic cometabolism.
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1. Introduction

Chlorinated Aliphatic Hydrocarbons (CAHs) are widespread wastewater and groundwater contaminants [1,2]. Research on aerobic CAH cometabolism showed that methane [1,3,4], toluene, phenol [5–8], ammonia [2,9], propane [10–21] and butane [22–24] can be effectively utilized as growth substrates. In particular, methane, propane and butane present the advantages of the absence of toxicity and of the possibility to inject hydrocarbon/air gaseous mixtures directly in the groundwater or in the vadose zone, a technology known as cometabolic air sparging [14,25]. Although some applications of aerobic cometabolism for the in situ bioremediation of CAH-contaminated sites have recently been reported [14,25,26], the environmental industry is giving a slow and skeptical response to the encouraging research results in this field. The main reasons for this attitude are: (i) in situ aerobic cometabolism implies the risk of clogging the aquifer portion surrounding the injection wells as a result of an excessive biomass growth [7,8]; (ii) the lag-times for the onset of CAH cometabolism can be very long [16,17]; (iii) in case of bioaugmentation, the in situ transport of the injected bacteria is a complex and uncertain process [27].

This study focuses on the aerobic cometabolism of 1,1,2,2-tetrachloroethane (TeCA). TeCA was a very common solvent prior to World War II, but substantial releases have also occurred in the recent past [28]. For example, the total TeCA release in the United States (including atmospheric emissions) was equal to 30 t in 1991 and to about 1 t in 2007 [29]. While several studies report the anaerobic biodegradation of TeCA, this compound has been generally regarded in the literature as non-biodegradable under aerobic conditions [30–34]. Its aerobic cometabolic biodegradation was evidenced for the first time in 1996 [1], and subsequently documented at low concentrations by two works of this research group [16,20]. The main goals of this study were: (i) to perform an in-depth investigation of the long-term aerobic cometabolic TeCA biodegradation over a wide concentration range (0–708 μM), evaluating the toxic effects of the degradation products and the degree of mineralization of the organic Cl; (ii) to identify and calibrate a complete kinetic model of TeCA cometabolism, representing a fundamental block for the design of a full-scale treatment; (iii) to select, characterize and stock a high-performing TeCA-degrading consortium, potentially utilizable for bioaugmentation treatments; (iv) to compare the TeCA degradation performances and the consortium composition obtained in slurry and soil-free bioreactors. The latter were introduced as a first step towards the implementation of CAH cometabolism in an on-site bioreactor within the framework of a pump-and-treat process. The aerobic

* Corresponding author. Tel.: +39 051 2090416; fax: +39 051 6347788.
E-mail address: dario.frascari@unibo.it (D. Frascari).

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Table 1
Experimental scheme of the microcosm study.

Group n.	Type of microcosms	Type of aquifer material	Growth substrate	CAHs	Maximum TeCA concentration tested (μM)	Inoculum
1	Slurry	A	Methane, propane	TeCA + 5-CAH mixture[a]	0.15	No
2	Slurry	A	Propane	TeCA	16	I0[b]
3	Slurry	A, B, C, D or E	Propane	TeCA	5	I1[c]
4	Soil-free	No aquifer material	Propane	TeCA	708	I2[d]

[a] Vinyl chloride, cis- and trans-dichloroethylene, trichloroethylene, 1,1,2-trichloroethane.
[b] Soil/biomass/water suspension sampled from the propane-fed microcosms of group 1.
[c] Soil/biomass/water suspension sampled from the propane-fed microcosms of group 2 (consortium C1).
[d] Inoculum I1, progressively deprived of soil as explained in Section 2.1 (consortium $C2_0$).

cometabolism of CAH-contaminated groundwater in a bioreactor represents an interesting alternative to in situ bioremediation, thanks to the elimination of the risk of aquifer clogging and to the possibility of effectively colonizing the bioreactor with a previously selected consortium, thus eliminating any lag-time. However, as the CAH-degrading consortia are typically obtained from soil-containing assays, it is important to evaluate possible changes in biodegradation performance deriving from the loss of the soil-attached biomass fraction.

2. Materials and methods

2.1. Experimental scheme and operational details relative to the batch bioreactors

The experimental work was divided into 4 groups of 119 mL batch bioreactors, referred to in the following as microcosms. The experimental scheme is summarized in Table 1. The first 3 groups consisted of slurry bioreactors, each containing 17.4 g of dry soil, 53 mL of groundwater and 59 mL of headspace air. The materials used for these slurries were taken from five different sites, as explained in the following. The 4th group consisted of soil-free bioreactors, each containing 44 mL of headspace air and 75 mL of a synthetic mineral medium (composition in μM: $(NH_4)_2SO_4$ 797, $MgSO_4{\cdot}7H_2O$ 244, $CaSO_4{\cdot}0.5H_2O$ 150, K_2HPO_4 8900, $NaH_2PO_4{\cdot}H_2O$ 5355, $FeSO_4{\cdot}7H_2O$ 22.6, $NaNO_3$ 9000, $MnCl_2{\cdot}4H_2O$ 1.52, $ZnSO_4{\cdot}7H_2O$ 0.510, H_3BO_3 1.00, $Na_2MO_4{\cdot}2H_2O$ 0.450, $NiCl_2{\cdot}2H_2O$ 0.144, $CuCl_2{\cdot}2H_2O$ 0.10, $CoCl_2{\cdot}6H_2O$ 0.10). In both the slurry and the soil-free tests, the gaseous substrate (methane or propane) was re-supplied each time the previous pulse was completely consumed. The resulting substrate average feed rate was equal to 330 μmol_C/week. Prior to each substrate addition, oxygen was introduced, so as to maintain aerobic conditions. Ammonium (NH_4Cl) and phosphate (KH_2PO_4 and K_2HPO_4) were periodically added. Before each new CAH addition, the microcosms were stripped with 0.22 μM-filtered air for 20 min to remove dissolved carbon dioxide and possible volatile degradation products.

The first group of tests consisted of 4 slurry microcosms, representing a part of a larger study [16] aimed at investigating the feasibility of the aerobic cometabolic bioremediation of an aquifer, labelled A, historically contaminated by a mixture of vinyl chloride (VC), cis- and trans-dichloroethylene, trichloroethylene, 1,1,2-trichloroethane and TeCA. Each microcosm, set-up with soil (sand 48%, silt 46%, clay 6%) and brackish groundwater from site A, contained the above-mentioned 6-CAH mixture at concentrations varying between 0.15 μM (TeCA) and 25 μM (VC). Two duplicate microcosms were supplied with methane (125 μM in the aqueous phase), and 2 with propane (46 μM). Each time all the 6 CAHs were completely degraded, they were re-introduced. The CAH amount introduced in a given microcosm was varied from pulse to pulse in order to study the effect of concentration on the transformation rates. Further experimental details on these tests are described by Frascari et al. [16]. The microcosms of group 1, as well as all the other slurry tests, were maintained in agitation in a roller at 25 °C. This temperature was chosen as compromise between the goals to work at the average temperature of site A (17.5 °C) and the need to stay above the lab room temperature, so as to ensure a constant temperature in the heated chamber.

The attainment of a stable process of aerobic cometabolic degradation of TeCA, usually considered as non-degradable under aerobic conditions, was considered the most interesting result of the first part of the study. Therefore the second group of slurry microcosms, set-up like the first one with aquifer material from site A, was aimed at investigating in more detail the aerobic TeCA cometabolism using only propane (46 μM) as the primary substrate, given the more promising results obtained with this compound in comparison with methane. To avoid the inhibiting effects exerted by the other 5 CAHs, these tests were first air-stripped to remove all the CAHs, then spiked with only TeCA using a saturated solution of TeCA in water (0.0238 M at 4 °C). Three initial TeCA concentrations were tested (0.24, 0.54 and 2.50 μM), with 3 replicate microcosms for each TeCA level. To reduce the lag-times for the onset of TeCA biodegradation observed in the 1st group, each microcosm of group 2 was initially inoculated with 2 mL of a soil/biomass/water suspension sampled from the propane-fed microcosms of group 1 after several pulses of CAH degradation. The TeCA concentration in the subsequent pulses was progressively raised, up to a final value of 16 μM.

The 3rd group consisted of 15 propane-fed slurry microcosms, set-up with aquifer material from 5 sites (site A + 4 more sites labelled B, C, D, E). A and B were historically contaminated by CAH mixtures including TeCA and were constituted primarily by sandy/silty soils. C, D and E were not contaminated: C contained a sandy soil, whereas D and E contained humic soils containing 1–1.2% of organic carbon. For each aquifer material, 2 microcosms were not inoculated, and 1 was bioaugmented with 2 mL of a soil/biomass/water suspension sampled under sterile conditions from the 9 microcosms of group 2 after several months of TeCA cometabolism. The microbial consortium contained in this sample was named C1. The goal of group 3 was on the one hand to evaluate the presence of TeCA-degrading strains in contaminated and pristine aquifers, and on the other to evaluate the capacity of the bacterial consortium developed in group 2 (C1) to induce a rapid onset of TeCA cometabolism in the presence of different indigenous consortia. After air-stripping of the CAHs, where present, the TeCA concentration was initially set to 0.95 μM, and progressively raised in the subsequent pulses up to 5 μM. The propane concentration was equal to 46 μM in each pulse.

In the 4th group of tests (soil-free microcosms), TeCA cometabolism by propane-growing bacteria was studied in the absence of soil in order to evaluate the feasibility of an on-site biological treatment of a TeCA-contaminated groundwater. To obtain a nearly soil-free TeCA-degrading inoculum from the slurry microcosms, the soil/biomass/water suspension sampled from group

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