Biofiltration of Volatile Pollutants: Engineering Mechanisms for Improved Design, Long-Term Operation, Prediction, and Implementation

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Progress Report

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Research Objective

Biofiltration systems can be used to treat volatile organic compounds (VOCs); however, the systems are poorly understood and are currently operated as "black boxes." Common operational problems associated with biofilters include fouling, deactivation, and overgrowth, all of which make biofilters ineffective for continuous, long-term use. The objective of this investigation is to develop generic methods for long-term stable operation, in particular by using selective limitation of supplemental nutrients while maintaining high activity and the ability to regenerate biofilter activity.

As part of this effort, we will provide a deeper fundamental understanding of the important biological and transport mechanisms in biodestruction of sparingly soluble VOCs and will extend this engineering approach and developed mathematical models to two additional systems of high-priority environmental management (EM) relevance-direct degradation and cometabolic degradation of priority pollutants such as BTEX (benzene, toluene, ethylbenzene, and xylene) and TCE (trichioroethylene), respectively.

Preliminary results indicate that we can control overgrowth of the biofilm while sustaining high degradation rates and develop basic predictive models that elucidate mass transfer and kinetic limitations in this system for alkanes. The alkanes are degraded into CO, and waterwith minimal biomass (due to the methodology proposed). This system will be used to test and model additional supplemental nutrient feeding strategies as well as methods to increase the fundamental driving forces by modification of the system. Models will be extended to non-steady-state, long-term operation. We will examine the nature of the mixed microbial community in the VOC-degrading biofilm and test for new degradative activities. We will use cosolvents with surfactant properties to enhance hydrocarbon solubility in the biofilm and evaluate their impact on 'mass transfer and reaction rate in an operating biofilter. These results will point to further potential improvements in systems of EM priority.

Research Statement

Development of general methods for long-term operation (by controlling fouling and filter deactivation) will lead to extended activity in industrial biofilters. Understanding how these methods work will make them more widely applicable and acceptable, particularly if practical mathematical models can be developed that accurately predict removal rates for multiple systems. Investigations into new biocatalyst activities will demonstrate the application of biofilters for treating a variety of EM-relevant contaminants, including alkanes, aromatics, and chloro-organics. Improvements in this technology would yield the greatest benefit as applied to contaminated U.S. Department of Energy groundwater streams that are currently being treated for low concentrations of VOCs (e.g., the Oak Ridge Y-12 Groundwater Treatment Facility). Several novel hypotheses, if proven, will yield increased rates of conversion and assist in long-term, nongrowing degradative biofilter activity.

Research Progress

Construction and Acclimation of Biofilter Units

Three biofiltration units were assembled and inoculated with the VOC-degrading microbial consortium selected for use in this project. The units are approximately 50 cm long, with 5-cm internal diameters, and constructed of glass, teflon, and stainless steel. Each unit is temperature-controlled and can be operated under a variety of continuous or recycling modes. The packing material chosen for use in this study was a commercially-available structured polyethylene woven mesh. This material provides an inert support for biomass, high surface area to reactor volume to promote mass transfer, and capillary spread of mineral nutrients to maintain moisture in the biofilm.

The three reactors were inoculated and operated under identical operating conditions for about 4 months. During this acclimation period, the columns immediately began showing varying stages of biomass development, accompanied by concomitant levels of VOC removal capacity. One column produced very low levels of biofilm during this period, such that removal levels of VOC were primarily associated with biomass levels in the trickling aqueous phase itself. The two other columns were able to remove substantially higher levels of VOCs after the acclimation period, due to well-developed biofilms.

Kinetics/Mass Transfer

Initial estimates of mass transfer/kinetics were made for these columns at the end of the acclimation period for model VOC compounds isobutane and n-pentane. First measurements indicated that kinetic limitations dominated all three systems, particularly in the one column that failed to develop a substantial biofilm coating. All rates measured were lower than those measured for one previously established column that had been in use for about 3 years (a model system for comparing results obtained in this study). Two of the columns have been allowed to continue biomass growth beyond the acclimation period so that mass-transfer limited reaction may be approached. The remaining column has been operated under steady, nutrient-limited, non-growth conditions for use as a control.

Extended Operation in Absence of Supplemental Nutrients

Extended activity of biofilter units in the absence of the supplemental nutrient ammonium has been demonstrated. This work represents a substantial breakthrough in filter operation by eliminating overgrowth of biomass in the column. Non-growth associated degradation kinetics have been measured and do not represent a substantial decrease in removal capacity as compared with growth-associated removal. We have shown that prolonged starvation via elimination of an available nitrogen source will provide at least 6 months of stable operation before eventually resulting in decreased (but restorable) capacity.

Quantification of Biomass Regeneration After Prolonged Starvation

One biofilter unit was operated under nutrient limited conditions for more than 6 months with sustained removal of both VOC gases. After this period, rates began to rapidly drop over the course of 1 month to 30% of their original values. Upon reintroduction of supplemental nitrogen, in the form of ammonium, rates began to rise rapidly as the biomass underwent regeneration. This procedure was complete in less than 14 days, after which the column operated at 100% of its previous removal efficiency. This is the first example of quantification/qualification of biomass regeneration after prolonged starvation in a trickle-bed system.

Removal Competence and Kinetics Demonstrated for C_2 to C_6 Alkanes

The microbial consortia was enriched by continuous growth using isobutane and n-pentane as the sole carbon and energy sources over a period of 5 years. At the beginning of this study, we proposed to use isobutane and n-pentane as model VOCs because the consortium was shown to degrade these two sparingly soluble gases. The removal potential of the consortium for other alkanes has since been examined; alkanes ranging from C_2 to C_6 in structure have been shown to serve as sole carbon and energy sources for the microbial consortium. Interestingly, the microbial consortium used for this study was initially developed for cometabolic TCE degradation studies. During enrichment procedures conducted for this project, the methanotrophs present in the mixture were lost (i.e., the consortium can no longer remove or degrade methane). The microbial pathways for the alkane degradations are unknown at this time. A variety of batch experiments have provided valuable kinetic parameters that have enabled us to mathematically describe uptake of these compounds in the trickle bed systems.

Removal Competence Demonstrated for Chlorinated Alkanes

The microbial consortium has demonstrated the capacity for degrading both chlorobutane and chloropentane in batch tests. In the case of chloropentane, the culture can use this difficult-to-degrade substrate as the sole carbon and energy source (non-cometabolic pathway). This result represents the first such report of noncometabolic removal of a higher-chain chlorinated alkane. The kinetic parameters of this pathway were measured and are being incorporated into our mathematical models; test runs in the trickle bed reactors will follow.

Solubility Enhancers

Two solubility enhancers have been examined thus far for their abilities to improve transport of sparingly soluble VOCs into the aqueous phase. These surface-active agents (glycerol and pluronic acid) were added at various concentrations to batch reactors containing medium only. No marked changes in VOC solubility were noted when these enhancers were used. It is possible that a critical concentration will have to be reached before any substantial effect is seen; other solubility enhancers will be investigated during year two.

Mathematical Model

We have begun development of a complex mathematical model to describe kinetics, diffusion, and mass transfer in the trickle bed systems. This is an ongoing work with several simple-case results already obtained. The model can already be used effectively and accurately for predicting a number of operation scenarios. A variety of coding methods (numerical analyses) are being investigated to solve the stiff differential equations of the two-dimensional two-pointboundary-value problem which arises from consideration of reaction and diffusion within a biofilm and along a biofilter.

Summary of Accomplishments

- Constructed and acclimated three trickling-bed biofilters.
- Measured kinetic activity and mass transfer in biofilters under study.
- Demonstrated extended activity of biofilters in absence of supplemental nutrient.
- Quantified filter regeneration after prolonged starvation.
- Demonstrated competence of microbial consortium for degrading a variety of C_2 to C_6 alkanes as sole carbon and energy sources.
- Demonstrated competence of microbial consortium for degrading chlorinated alkane as sole carbon and energy sources.
- Examined solubility enhancement agents.
- Completed mathematical modeling of biofilm diffusion, reaction, and mass transfer effects for simple cases.

Papers and Abstracts

Barton, J. W., B, H. Davison, K. T. Klasson, and C. C. Gable III. "Estimation of Mass Transfer and Kinetics in Operating Biofilters for Removal of VOCs." Presentation to be delivered at AICHE Annual Meeting, November 1997.

Barton, J. W., S. M. Hartz, K. T. Klasson, and B. H. Davison. "Microbial Removal of Alkanes from Dilute Gaseous Waste Streams: Mathematical Modeling of Advanced Bioreactor Systems." Accepted for publication in ***Journal of Chemical Technology and Biotechnology.***

Barton, J. W., K. T. Klasson, and B. H. Davison. 1997. "Extended Operation and Control of Biomass Overgrowth in Biofilters Designed for VOC Removal." Published ***in Proceedings of the Air & Waste Management Association 's 90th Annual Meeting & Exhibition,*** June 8-13, 1997, Toronto, Ontario, Canada.

Barton, J. W., K. T. Klasson, B. H. Davison. 1997. "Extended Operation and Control of Biomass Overgrowth in Biofilters Designed for VOC Removal." Presentation delivered at the Air & Waste Management Association's 90th Annual Meeting & Exhibition, June 8-13, 1997, Toronto, Ontario, Canada.

Barton, J. W., K. T. Klasson, and B. H. Davison. 1997. "Extended Performance and Evaluation of Trickle Bed Bioreactors Designed for VOC Removal." Air & Waste Management Association, Southern 'Section, Annual Meeting, August 6-8, 1997, Gatlinburg, Tenn.

Barton, J. W., K. T. Klasson, L. J. Koran, Jr., and B. H. Davison. "Microbial Removal of Alkanes from Dilute Gaseous Waste Streams: Kinetics and Mass Transfer Considerations." Accepted for publication in ***Biotechnology Progress.***

Klasson, K. T, B. H. Davison, J. W. Barton. “Chloroalkane Degradation by Enriched Microbial Consortium.” Submitted for publication.

Klasson, K. T, B. H. Davison, J. W. Barton, E.M. Just, and C. C. Gable III. “Biofiltration of Chlorinated and Non-Chlorinated Alkanes.” Submitted for publication.

Klasson, K. T, B. H. Davison, J. W. Barton, E.M. Just, and C. C. Gable IL “Biofiltration of Chlorinated and Non-Chlorinated Alkanes.” Presentation to be delivered at the American Chemical Society’s Emerging Technologies in Hazardous Waste Management IX (Enviro Expo ‘97), Pittsburgh, Penn., September 1997.