0273-2289/94/4546—0775$03.20

Alternating Extraction/Injection Well Interactions for *In Situ* Bioremediation

M. S. SHOUCHE,[1] J. N. PETERSEN,*[,1]
R. S. SKEEN,[2] AND B. S. HOOKER[3]

[1]Chemical Engineering Department, Washington State University, Pullman, WA 99164-2710; [2]Waste Treatment Technology Center, Battelle Pacific Northwest Laboratories, Richland, WA; and [3]Chemical Engineering Department, Tri-State University, Angola, IN

ABSTRACT

The success of *in situ* biological contaminant destruction depends on the distribution of microorganisms through a flow field. Using numerical tools, a strategy is developed which causes the biomass to grow in a more uniform fashion throughout the flow field. The results obtained using this strategy are compared with a conventional strategy. Here, we demonstrate that by properly alternating the direction of flow through a flow field and by internally recycling the nutrients and contaminants, the biomass can be distributed more uniformly and significantly more contaminant destruction can be attained than would be possible if a unidirectional strategy were employed.

Index Entries: *In situ* bioremediation; mathematical model; carbon tetrachloride; alternating direction.

INTRODUCTION

Cost effective methods must be developed to destroy subsurface contaminants such as carbon tetrachloride (CCl_4). Such compounds have been extensively used as industrial solvents, and contamination of the subsurface is extensive (*1*). For a number of years, bioremediation has been considered as a potential method for treating groundwaters contaminated with halogenated aliphatic compounds such as carbon tetrachloride (*2*).

*Author to whom all correspondence and reprint requests should be addressed.

Such contaminants are often present in different layers of the subsurface. Many are present in the surface soils; others have penetrated into the vadose zone. Frequently contaminants penetrate farther into the saturated zones where the pores are completely filled with water, and where groundwater supplies occur.

Because the contaminants sorb to soil surfaces, remediation of such saturated zones may be problematic. For example, Valocchi (3) showed that when desorption of the contaminants from the soil surface becomes the rate limiting step in the remediation process, the time required to reduce the contaminant concentration to acceptable levels using a pump and treat method may be unacceptably long. In such situations, the treatment of many pore volumes of water may be required to remove the contaminants that have sorbed to the aquifer solids. *In situ* bioremediation can significantly reduce the time required for cleanup by decreasing the contaminant concentration in the aqueous phase and thereby increasing the driving force for its desorption from the soil surface (3). For these reasons, *in situ* bioremediation is considered an attractive alternative to conventional aboveground treatment methods.

Past studies (*1,4*) have shown that improper addition of the nutrients to the subsurface may induce significant biomass growth near the point where nutrients are injected into the flow field. This accumulation will eventually lead to plugging of the soil's porous matrix. Through the use of predictive tools, Shouche et al. (4) demonstrated that extensive growth occurs near the inlet of the flow field because of the high concentrations of the nutrients. This biofouling will limit the uesful life of remediation wells, and thus may determine the success of *in situ* bioremediation. These numerical predictions are in qualitative agreement with field test data. For example, in the field demonstration of CCl_4 destruction reported by Semprini et al. (*1*), the injection well had to be abandoned after 66 d of operation. They speculated that the injection well was plugged by biomass at that time.

Semprini et al. also surmised that by adding the nutrients in a pulse, rather than continuous, fashion, the rate of biofouling could be reduced. Shouche et al. (4) have further demonstrated that the characteristics of the pulse plays a crucial role in microbial growth and contaminant degradation. The pulse characteristics that they examined were the concentration of the nutrients in the pulse, the duration of time for which the pulse was applied, and the time between two consecutive pulses. They found that the amount of biomass accumulation within a flow field could be minimized by proper choices of these pulse characteristics for the addition of the electron donor. Other researchers have shown (*5–7*) that optimal control theory may also be used to determine nutrient addition strategies that provide enhanced bioremediation.

Although previous nutrient injection strategies minimized the growth of biomass in the flow field, the growth was limited only at one end, the inlet of the flow field. More uniform growth of biomass in the flow field

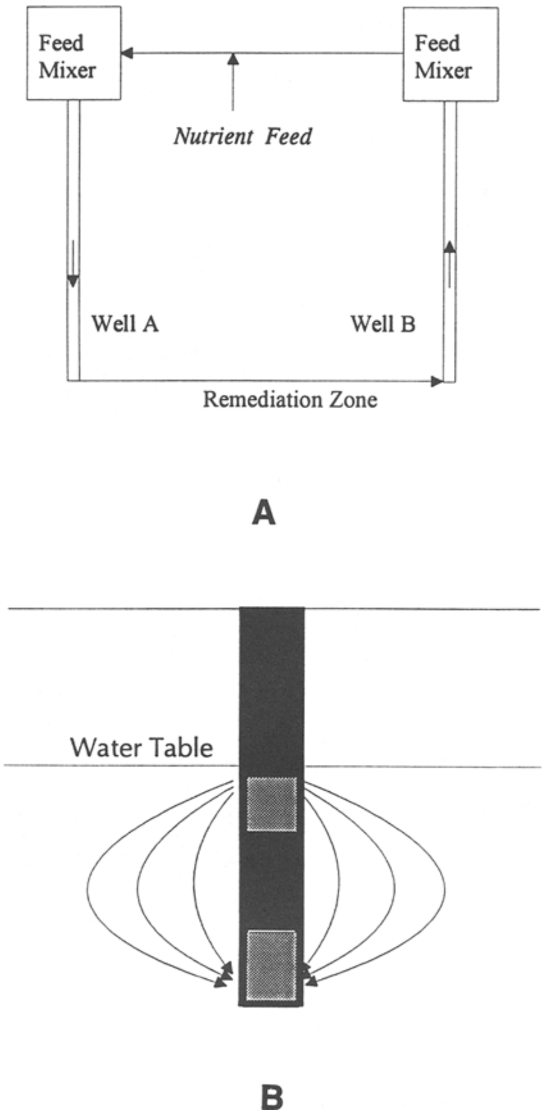

Fig. 1. Potential well configurations which would lend themselves to an alternating direction approach. (**A**) uses two wells, both of which contain feed mixers and the necessary pumps to serve as either injection or extraction wells. (**B**) uses a single well with two screened intervals and the necessary packings and pumps to allow either to be used as injection or extraction points. Thus, the flow direction can be reversed in either (**A**) or (**B**) from that shown.

could enhance the destruction of the biological contaminants. This growth pattern could be achieved by alternating the direction of flow through the site at regular intervals. Several potential well designs could be employed to accomplish this scheme, two of which are shown schematically in Fig. 1. Figure 1A shows a possible configuration if two wells are used. In this case, both wells would be used as injection and as extraction wells in an alternating fashion. The flow direction would be dependent on which of these wells is used as an injection well at any particular time. For example, at the beginning of the operation, the first well (A) may be responsible for nutrient addition to the aquifer, along with groundwater recirculation. In this case, the second well (B) would serve as the extraction site. Fluids that are extracted from well B would be pumped back to A, forming a closed loop. After some time interval, the two wells would switch function, well B would become the injection well and A would be the extraction well, reversing the flow direction.

Alternatively, a single well with multiple screened intervals, as shown schematically in Fig. 1B, could be used to accomplish this scheme. In this case, the direction of flow could either be from the lower screened interval to the upper, or vice versa. Additional packings would have to be installed in such wells as compared to a well that is capable of unidirectional flow only, increasing the cost of the well. However, the additional cost may be offset by the enhanced remediation achieved when using the alternating direction strategy.

In this article we develop this alternating direction nutrient injection strategy using a mathematical description of an *in situ* bioremediation process for the destruction of CCL_4. A comparison is made between the alternating extraction/injection well and the fixed extraction/injection well for *in situ* bioremediation of CCl_4.

Mathematical Model

The mathematical model used in this study is based on the kinetics proposed by Semprini et al. (*1*). These kinetics were developed to describe the field demonstration of *in situ* CCl_4 degradation conducted at Moffett Naval Air Station, located in Mountain View, CA. A 1-D flow field, 2 m in length is assumed, based upon the physical system in use at the Moffett test site. Within the flow field, transport processes of advection, dispersion, and sorption, biological destruction of the contaminants, consumption of the nutrients, and the microbial growth are accounted for. The microbial kinetics assume that two populations of microorganisms are responsible for the remediation of CCl_4. The first population is made up of denitrifiers that use acetate as the primary substrate. The second population grows on the decay products of the denitrifiers, and is inhibited by nitrate. The microbial populations are assumed to be attached to the soil surface. Both of the microbial populations are assumed to be capable of transforming CCl_4, but the rate of transformation for the second population is much higher. Both the transformations are assumed to be governed by Monod kinetics.

With these assumptions, the model for CCl_4 degradation is described by a set of 5 partial and 4 ordinary differential equations. A detail description of the model along with the development of the governing equations, initial conditions, boundary conditions, and porosity calculations is described elsewhere (*4*). In this work, this set of ordinary and partial differential equations was solved using DSS/2 (*8*), which employes the numerical method of lines to solve the set of simultaneous partial differential equations. In the DSS/2 system, the Runge-Kutta-Fehlberg formula was used to integrate with respect to time, using a time step of 0.001 d. This system of equations was then integrated in time until the biomass reached a concentration of 9800 mg/L at some point in the flow field. Using the methods of Shouche et al. (*4*), a flow field having an initial porosity of 0.23 is predicted to be plugged at this biomass concentration.

The nitrate was fed to the flow field in a continuous fashion at a concentration of 26 mg/L in all cases tested. Conversely, acetate was fed in a pulse fashion with the width of the acetate pulse set at 32 min and the concentration of the acetate in each pulse set at 605 mg/L. The time between the end of one pulse and the start of the next was 11.9 h, so that the total time between the start of two consecutive pulses was 0.518 d. Additionally, in all cases, the initial nitrate concentration within the aquifer was set at 26 mg/L, whereas the acetate concentration was set at zero. Finally, the CCl_4 was assumed to be initially distributed throughout the flow field at a concentration of 37 ppb. No additional CCl_4 was fed to the flow field. These conditions were chosen as a basis for comparison since the flow field is predicted to plug at an operating time close to 50 d, under unidirectional flow conditions.

RESULTS AND DISCUSSION

The objective of this study was to use numerical tools to ascertain the effect that alternating the direction of flow would have on the biomass growth and CCl_4 degradation in a flow field.

Figure 2A shows the time at which the flow field is predicted to become plugged as a function of the number of consecutive pulses introduced to the system before changing the flow direction. In addition, on Fig. 2B the maximum, minimum, and average CCl_4 concentration in the flow field at the time of plugging are also shown. Examination of the time required to plug the flow field reveals three distinct regions of behavior. In the region where less than 15 consecutive pulses are introduced before the direction of flow is reversed, the time required to plug the field is relatively constant at approximately 60 d. The second distinct region extends from 16 to 81 consecutive pulses before the direction of flow is reversed. In this region, the time that the site can be remediated before the field is predicted to plug oscillates between a low of about 48 d and a high of about 60 d. Finally, in the region in which more than 81 consecutive pulses are introduced

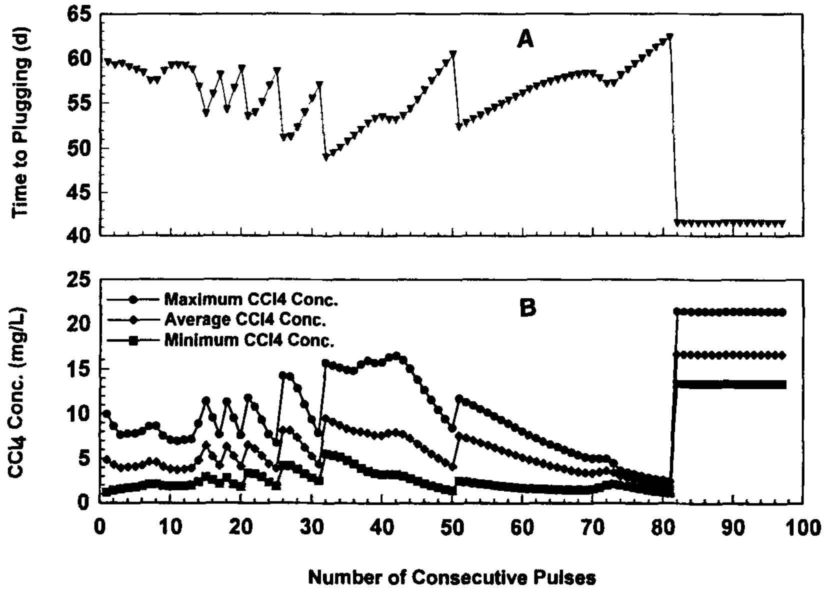

Fig. 2. The time required to plug the flow field, and the maximum, average and minimum CCl_4 concentrations in the flow field at this time as a function of the number of consecutive pulses introduced into the flow field between changes in the direction of flow.

before the flow direction is reversed, the flow field plugs after 42 d. Note that this third region corresponds to a situation in which the direction of flow is never changed, since the time at which the flow field plugs is less than the time at which the direction of flow is to be changed.

It is apparent from the data shown in Fig. 2 that the total time that a remediation system can be operated is maximized when using either less than 15 consecutive pulses before the direction of flow is switched, or between 70 and 81 pulses in a single direction before changing the direction of flow. Under these conditions, the time to plug the flow field is near the predicted maximum and is not a strong function of the number of pulses introduced before changing the direction of flow. Moreover, more complete CCl_4 destruction is observed when the system is operated for as much time as possible before switching the direction of flow.

Looking specifically at the second region in Fig. 2, the time required to plug the flow field as function of the number of consecutive pulses introduced before the direction of flow is changed has a saw tooth shape. The phenomena that cause this shape can be understood by examining the cases illustrated in Fig. 3. This figure shows the biomass concentrations in the flow field as functions of time and position for the cases where 50 (3A) and 51 (3B) consecutive pulses are used. These two conditions correspond to a peak and subsequent low of a saw tooth on Fig. 2. In Fig. 3A, for the first 25 d of operation, nutrients are fed from the end of the flow

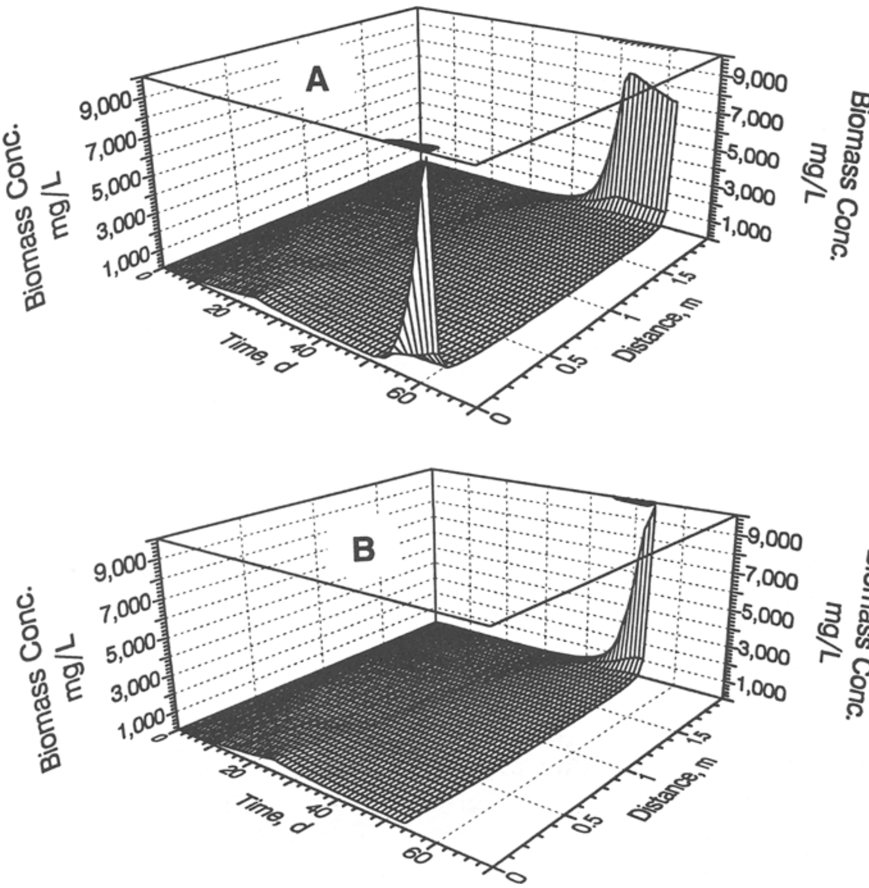

Fig. 3. The biomass concentration within the flow field as a function of time and position for two cases. In (**A**), 50 consecutive pulses are introduced to the flow field between changes in the direction of flow. In (**B**), 51 consecutive pulses are introduced between changes in the direction of flow.

field at 0 m. During this time, the biomass grows slightly, but does not reach the critical level where exponential growth will produce high levels of biomass. When the flow direction is reversed, the biomass at the 2 m end does reach the stage where exponential growth produces significant quantities of biomass. However, immediately before the flow field is plugged, the flow direction is again reversed. At this time, since the nutrients are being introduced at the 0 m end of the flow field, the biomass at this point (0 m) grows until the flow field plugs. In contrast to this case, when 51 consecutive pulses are used, the 2 m end of the flow field plugs before the flow direction is reversed for the second time (Fig. 3B).

This concept is further illustrated on Fig. 4, which shows the biomass concentration at the 0 and 2 m end of the flow field as a function of time

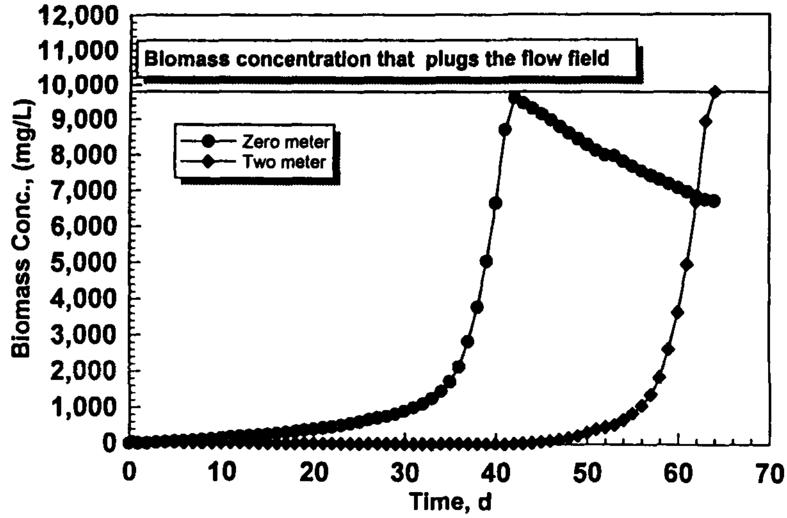

Fig. 4. The biomass concentration at the entrance and exit of the flow field as a function of time when 81 consecutive pulses are introduced to the flow field before the direction of flow is changed.

for the case in which 81 consecutive pulses are introduced to the flow field before the direction is reversed. Here, the biomass on the 0 m end of the flow field rises to a value that is very near that at which the flow field is assumed to become plugged. However, just before this level is reached the direction of flow is reversed and the biomass concentration at the 0 m end of the flow field begins to diminish and growth begins at the 2 m end of the flow field. Now, since this end of the flow field has never received high nutrient concentrations, the biomass level is low at this 2 m point. Thus, considerable time is necessary before the biomass grows to the levels at which the flow field will plug. This switch allows nearly 20 more d of operation, during which time significant quantities of the CCl_4 are destroyed. If, however, one more nutrient addition pulse were to be added to the 0 m end of the flow field before the direction of flow were changed, complete plugging is predicted to occur.

The effect of using a larger number of consecutive pulses before changing flow directions on the CCl_4 concentration in the flow field is shown on Fig. 5. This figure shows the predicted CCl_4 concentration in the flow field at the time of plugging under two different conditions. In the first case, 50 consecutive pulses are added before the direction of flow is switched. In the second case, 81 consecutive pulses are introduced to the flow field before the direction of flow is changed. Note from Fig. 2 that both of these will allow the bioremediation system to be operated for approximately 60 d before the biomass would grow to the level where the flow field would be plugged. It is apparent from Fig. 5, however, that, for the case where 50 consecutive pulses are added before the direction of flow is switched,

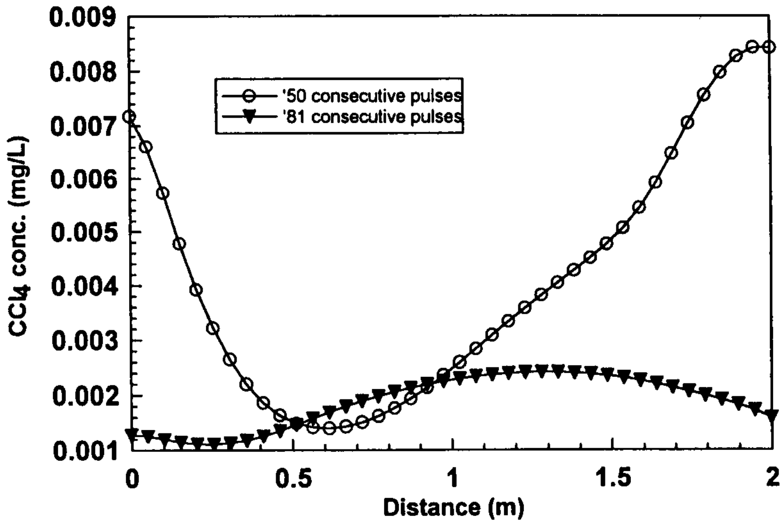

Fig. 5. The CCl_4 concentration as a function of distance at the time when the flow field plugs for two different cases. If the direction of flow is changed after 50 consecutive pulses have been introduced, then a nonuniform distribution is obtained. However, if the direction of flow is changed after 81 consecutive pulses, then a relatively uniform distribution is obtained. In this latter case, the predicted CCl_4 concentration would be below the drinking water standard of 5 ppb throughout the flow field.

the CCl_4 degradation is achieved predominantly in the center of the flow field. Conversely, a more uniform amount of destruction is achieved when 81 consecutive pulses are added before changing the direction of flow.

Let us now attempt to understand the higher biological destruction activity that is predicted to occur in the center of the flow field in the case in which 50 consecutive pulses are introduced between changes in the direction of flow. This phenomena can be understood by examining the assumptions made by Semprini et al. in the development of the kinetic expressions that were employed to describe the biological processes. These authors assumed that two microbial populations were involved. The first population is assumed to grow on acetate and nitrate, but the second population is assumed to use soluble decay products from the first population as its primary carbon source. Further, Semprini et al. assumed that the second population is primarily responsible for CCl_4 destruction. Hence, when a pulse of nutrients is introduced into the flow field, the first population will grow in the region where the nutrient concentration is high. After the pulse has passed, the first population will begin to die and produce the soluble decay products that are required for the second population to grow and degrade the CCl_4. These soluble decay products are swept along with the flow, to regions between the two wells, and stimulate growth and contaminant destruction by the second population.

In the case in which 81 consecutive pulses are introduced before the flow direction is reversed, the CCl_4 concentration is relatively uniform throughout the flow field and the marked decrease in the center of the field that was observed in the 50-pulse case is not present. To understand why this CCl_4 profile develops, we must first recall that the time which elapses between the start of each consecutive pulse is 0.518 d. Thus, 41.4 d will elapse during the course of 81 nutrient injection pulses. Now, recall that the flow field will plug if unidirectional flow is carried out for 42 d. Clearly, by providing 81 consecutive injections, at the same end of the field, the flow field is on the verge of plugging when the direction of flow is reversed. Additionally, the amount of biomass that has accumulated at the other end of the flow field at the time that the direction is changed will be minimal. Thus, when the direction of flow is changed, the time required to accumulate sufficient biomass to plug the flow field from the 2 m end is then at a maximum.

From a practical standpoint, maximizing the time between changes in flow direction would be the simplest to implement in the field since this strategy would minimize the number of times that the pumps would need to be cycled. In addition, considerable expense may be associated with manipulating the down well equipment to change the direction of flow.

Collectively, these results suggest that the most practical operating strategy would be to direct the flow in a unidirectional fashion for as long as possible without completely plugging the subsurface formation with biomass. When this point is reached, the direction of flow should be reversed. By operating the system in this way, the amount of contaminant that can be destroyed is maximized, a uniform distribution of contaminant through the flow field is obtained, and the amount of required operator intervention is minimized.

CONCLUSIONS

A mathematical description of *in situ* bioremediation has been used to examine the effects of a nutrient injection strategy that alternates the direction of flow through a subsurface formation will have upon the biomass growth and CCl_4 destruction that would be attained in the flow field. This mathematical model predicts that by changing the direction of flow often, the life of the nutrient injection well can be significantly extended past the time when the formation would have been plugged with biomass had a unidirectional strategy been employed. Further, these predictions suggest that the best strategy would be to operate the nutrient injection well in a unidirectional fashion until the flow field begins to become biofouled. At this time, the direction of flow should be reversed. These results suggest that in so doing, a uniformly low contaminant concentration can be obtained through out the flow field.

In an actual field site, one would never operate the nutrient injection wells until plugging occurs. However, these results indicate that the best way to operate such a site might be to feed the nutrients in a unidirectional fashion until biomass accumulation causes the flow to be reduced to a value near an unacceptable level. At this time, the direction of flow should be reversed such that the injection well then becomes an extraction well. In so doing, the life of the site would be extended significantly, the amount of contaminant degraded would be increased, and a more uniform contaminant distribution will be obtained throughout the site.

ACKNOWLEDGMENTS

The portion of this work conducted at Washington State University was supported by a contract from the Pacific Northwest Laboratory. Drs. Petersen and Hooker were supported by the Northwest College and University Association of Science (Washington State University) under Grant DE-FG06-89ER-7522 with the US Department of Energy. This work was supported by the US Department of Energy Office of Technology Department, VOC-Arid Integrated Demonstration. Pacific Northwest Laboratory is operated by Battelle Memorial Institute for the US Department of Energy under contract DE-AC06-76RLO 1830.

REFERENCES

1. Semprini, L., Hopkins, G. D., Janssen, D. B., Lang, M., Roberts, P. V., and McCarty, P. L. (1991), *In-situ Biotransformation of Carbon Tetrachloride Under Anoxic Conditions,* EPA Report No. EPA 2-90/060, U.S. EPA, Ada, OK.
2. McCarty, P. L. (1988), in *Environmental Biotechnology,* Omenn, G. S., ed., Plenum, New York, pp. 143–162.
3. Valocchi, A. J. (1986), *Water Resource Res.* **22(12)**, 1693–1701.
4. Shouche, M. S., Petersen, J. N., and Skeen, R. S. (1993), *Appl. Biochem. Biotechnol.* **39/40**, 763–779.
5. Culver, T. B. and Shoemaker, C. A. (1993), *Water Resources Research* **28**, 823–831.
6. Heinricher, A., Lenhart, S., and Solomon, A., submitted to *Natural Resource Modeling J.* 1993.
7. Shouche, M. S., Petersen, J. N., and Skeen, R. S. (1993), *Environ. Progress,* in press.
8. Schiesser, W. E. (1987), *DSS/2 Introductory Programming Manual,* Lehigh University, Bethlehem, PA.