

Large-Scale Natural Gradient Tracer Test in Sand and Gravel, Cape Cod, Massachusetts

1. Experimental Design and Observed Tracer Movement

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A large-scale natural gradient tracer experiment was conducted on Cape Cod, Massachusetts, to examine the transport and dispersion of solutes in a sand and gravel aquifer. The nonreactive tracer, bromide, and the reactive tracers, lithium and molybdate, were injected as a pulse in July 1985 and monitored in three dimensions as they moved as far as 280 m down-gradient through an array of multilevel samplers. The bromide cloud moved horizontally at a rate of 0.42 m per day. It also moved downward about 4 m because of density-induced sinking early in the test and accretion of areal recharge from precipitation. After 200 m of transport, the bromide cloud had spread more than 80 m in the direction of flow, but was only 14 m wide and 4–6 m thick. The lithium and molybdate clouds followed the same path as the bromide cloud, but their rates of movement were retarded about 50% relative to bromide movement because of sorption onto the sediments.

INTRODUCTION

During the past decade, a focus of groundwater research has been the development of quantitative models to predict contaminant transport in aquifers. These predictive tools are needed in the investigation and cleanup of contaminated subsurface systems. However, there is a growing awareness that the transport and fate of contaminants are greatly affected by the natural heterogeneity that characterizes aquifers [Anderson, 1987]. A key research question is how to incorporate this heterogeneity into quantitative analyses of flow and transport. In particular, we would like to know how to find the appropriate average physical and chemical properties that govern this transport and how to assess the reliability of predictions based on models using these effective, or average, properties [Gelhar, 1986].

Several theories account for the effects of heterogeneity of aquifer properties on flow and transport by treating heterogeneity as a stochastic process. This work has been particularly useful in understanding field-scale dispersion of solutes in groundwater [Dagan, 1982, 1984; Gelhar and Axness, 1983; Neuman *et al.*, 1987]. The stochastic theories relate macrodispersion observed in the field to variations in velocity caused largely by the spatial variability of hydraulic conductivity. Stochastic analysis has also been used to examine the effects of chemical heterogeneity of the sediments on macrodispersion of reactive solutes [Garabedian *et al.*, 1988; Valocchi, 1989].

Although these theories have changed the way in which we conceptualize flow and transport in heterogeneous aquifers, the practical application to field problems is still the subject of debate [Molz *et al.*, 1988]. The pioneering work at the Borden site in Ontario [Sudicky *et al.*, 1983; Mackay *et al.*,

1986; Freyberg, 1986; Sudicky, 1986] demonstrated the application of the stochastic approach in analyzing field-scale solute transport. But these field tests need to be extended to include other types of aquifers and different reactive species.

This paper describes a natural gradient tracer experiment that was conducted in a stratified, sand and gravel aquifer on Cape Cod, Massachusetts. The primary goal of the tracer test was to obtain a data set that was sufficiently detailed to test the application of stochastic solute transport theories in a field setting. The components of the test included injection of a solution containing both nonreactive and reactive tracers into the unconfined aquifer and monitoring of the movement and dispersion of the tracer cloud under a natural hydraulic gradient for 3 years. This paper presents an overview of the tracer test and describes the observed movement of the tracers. In a companion paper [Garabedian *et al.*, this issue], a calculation of macrodispersivity by the method of moments from the spatial distribution of the nonreactive tracer is presented. This measured dispersivity will be compared to values estimated by application of the stochastic theories to field measurements of the spatial variability of hydraulic conductivity that are presently being made [Wolf, 1988; Hess, 1989].

A second goal of the tracer test was to examine the transport of several reactive tracers that were expected to be sorbed onto the aquifer sediments and to be retarded relative to the nonreactive tracer. Laboratory tests of water and sediment collected at the site are being used to measure the chemical characteristics that are expected to control the fate of the reactive solutes in the aquifer [Stollenwerk and Kipp, 1990; Wood *et al.*, 1990]. Predictions of the behavior of these solutes using both stochastic and deterministic models will be compared to the observed behavior in order to assess the applicability of laboratory-derived chemical parameters to the field situation. Methods to determine the effective chemical parameters that are important in the field case can then be developed and tested.

SITE DESCRIPTION AND AQUIFER CHARACTERISTICS

The tracer test was conducted in an abandoned gravel pit on western Cape Cod near Otis Air Base (Figure 1). This part

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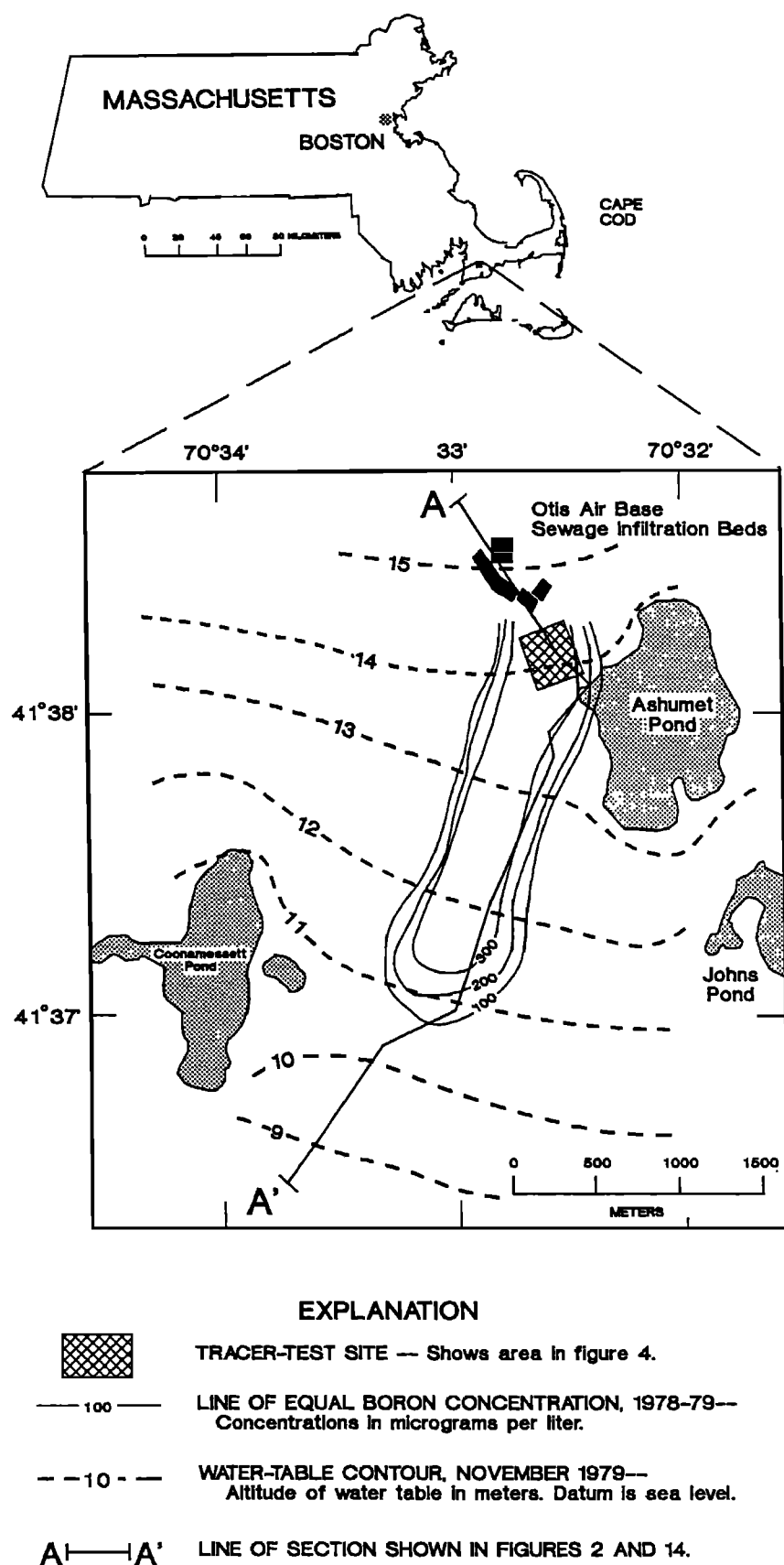


Fig. 1. Location of study area, showing sewage plume and tracer test site [from LeBlanc, 1984a, Figure 16].

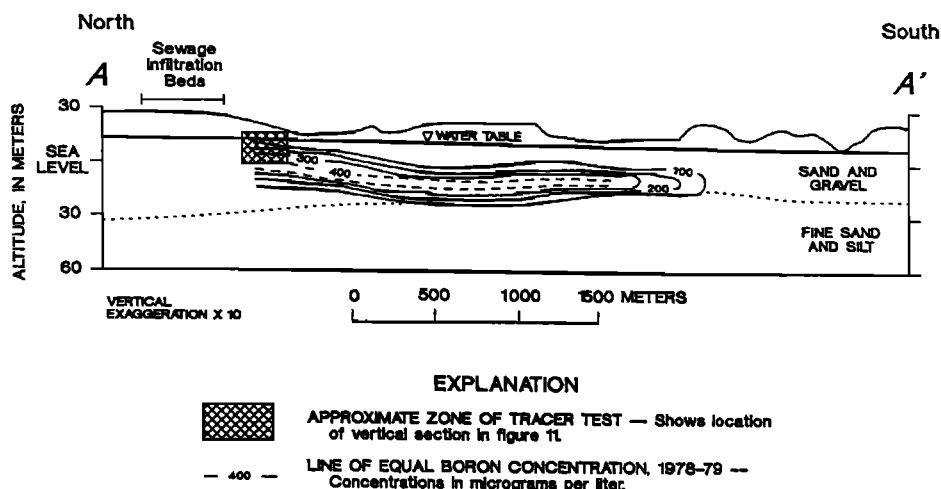


Fig. 2. Vertical location of sewage plume, geohydrologic units, and zone of tracer test. Line of section shown in Figure 1 [from LeBlanc, 1984a, Figure 9].

of Cape Cod consists of a large sand and gravel outwash plain that was deposited during the retreat of the continental ice sheets from southern New England about 12,000 years ago. The unconsolidated deposits form an unconfined aquifer which is the area's sole source of drinking water. General information on the geology and hydrology of Cape Cod is available in the works by Oldale [1982], Oldale and Barlow [1987], Guswa and LeBlanc [1985], and LeBlanc *et al.* [1986].

Hydrogeologic Characteristics

The aquifer at the tracer test site is composed of about 100 m of unconsolidated sediments that overlie a relatively impermeable, crystalline bedrock [Oldale, 1969]. The upper 30 m of the aquifer consists of a permeable, stratified, sand and gravel outwash. Beneath the outwash, the sediments consist of a fine-grained sand and silt (Figure 2). The median grain size (by weight) of the outwash is about 0.5 mm, and the outwash generally contains less than 1% silt and clay [LeBlanc, 1984c; Barber, 1987]. The sediments are composed largely of quartz and feldspar, with minor amounts of mica, glauconite, iron oxides, and other ferromagnesian aluminosilicates [Barber, 1987]. The sediment grains are commonly coated with iron oxyhydroxide and, in some zones, manganese oxide.

The estimated average hydraulic conductivity of the sand and gravel is 110 m/d. This value was obtained from an aquifer test conducted in the outwash about 2.2 km south of the site [Garabedian *et al.*, 1988]. The aquifer test also yielded a ratio of horizontal to vertical hydraulic conductivity of 2:1 to 5:1. This small vertical anisotropy is consistent with the anisotropy used in a three-dimensional groundwater flow model of the Cape Cod aquifer [Guswa and LeBlanc, 1985]. Hydraulic tests made with a borehole flowmeter [Rehfeldt *et al.*, 1989] in long-screened wells [Hess, 1989] and permeameter tests on cores collected at the tracer test site [Wolf, 1988; Wolf *et al.*, 1991] indicate that the hydraulic conductivity of the outwash varies about 1 order of magnitude. This variation results from the interbedded lenses and layers of sands and gravels that form this glaciofluvial deposit and are evident in surface exposures at the site (Figure 3).

The effective porosity of the outwash is estimated from several small-scale tracer tests [Garabedian *et al.*, 1988; Barlow, 1987] and spatial moments analysis of this test [Garabedian *et al.*, this issue] to be about 0.39. Similar values of porosity, ranging from 0.36 to 0.42, were reported by Perlmutter and Lieber [1970] and Morris and Johnson [1967] for sandy, stratified glacial deposits.

Hydrologic Characteristics

The tracer test site is located about midway between the Otis Air Base sewage disposal site and Ashumet Pond, a kettle hole pond in the outwash plain (Figure 1). The water table at the test site is generally 3–7 m below land surface and slopes to the south at about 0.15 m per 100 m (Figure 4). The source of water to the aquifer is recharge from precipitation. On Cape Cod, about 45% of the total annual precipitation, or about 50 cm/yr, recharges the groundwater system [LeBlanc *et al.*, 1986]. Most recharge occurs during late fall and winter. The groundwater ultimately discharges to the ocean about 10 km south of the study site.

The water table altitude typically fluctuates about 1 m annually because of seasonal variations in precipitation and recharge. During the first 17 months of the tracer test (July 1985 to December 1986), however, the water table fluctuated only about 0.3 m (Figure 5) because precipitation was less than normal in the fall and winter when recharge usually is greatest. During this period, therefore, the magnitude and direction of the horizontal hydraulic gradient were relatively constant; the magnitude varied from 0.0014 to 0.0018, and the direction varied from 164° to 173° east of magnetic north. Between December 1986 and May 1987, a period with above normal precipitation, the water table rose about 1.2 m, and the hydraulic gradient increased to 0.0020 and shifted to 156° east of magnetic north. The direction and magnitude of the gradient are influenced by Ashumet Pond; as water levels rise in the aquifer, the gradient tends to steepen and shift eastward toward the pond.

Vertical hydraulic gradients are too small to measure in clusters of monitoring wells at the tracer test site; the vertical gradients must be smaller than 0.3 cm (accuracy of the water level measurements) per 25 m (vertical span of the

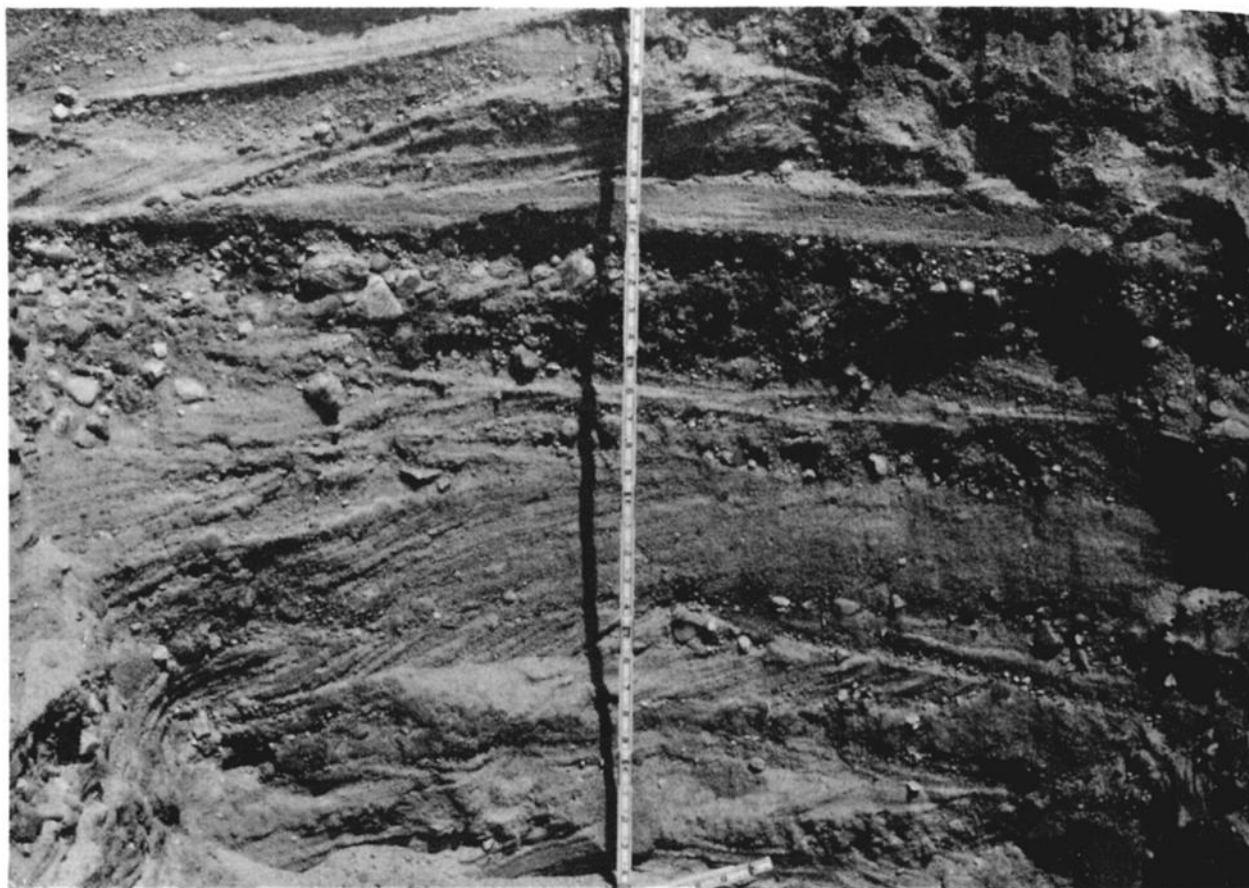


Fig. 3. Fresh exposure of sand and gravel outwash in test pit at tracer test site. Exposure in unsaturated zone about 5 m above water table. Height of section about 1 m. Location of test pit shown in Figure 4.

well clusters). Therefore, groundwater flow is nearly horizontal in the aquifer. Based on the estimates of hydraulic conductivity, hydraulic gradient, and effective porosity given above, the average velocity of groundwater in the sand and gravel is about 0.4 m/d.

Groundwater Quality

Disposal of treated sewage to the aquifer at Otis Air Base has formed a contaminant plume [LeBlanc, 1984a, b] which underlies the tracer test site. The plume, which is 23 m thick (Figure 2) and more than 3 km long (Figure 1), contains elevated concentrations of many inorganic and organic contaminants (Table 1). Water in the plume contains little or no dissolved oxygen and has a specific conductance as high as 350 $\mu\text{S}/\text{cm}$ (microsiemens per centimeter at 25°C). A zone of uncontaminated groundwater derived from precipitation overlies the plume (Figure 2). Water in this zone is aerobic and has a specific conductance of 40–80 $\mu\text{S}/\text{cm}$. The transition zone between uncontaminated and contaminated water at the site is about 3 m thick (Figure 6). The tracer test was conducted partly in the uncontaminated zone above the plume and partly in the underlying plume.

DESIGN AND OPERATION OF THE TRACER TEST

The tracer test began in July 1985 with the injection of 7.6 m^3 of tracer solution into the aquifer. Movement of the

tracer cloud was then monitored by collection of water samples from an array of 656 multilevel samplers. The test was designed and conducted so that the movement of the tracer cloud could be monitored in three dimensions as it traveled about 280 m through the aquifer. The design included a controlled injection, so that the nature of the source was well defined, and a monitoring program that minimized disturbance of the aquifer and the tracer cloud.

To develop this design, seven preliminary tracer experiments were run in 1984 and 1985 at several sites in the study area, including a small-scale natural gradient test, run at the site of the large-scale test, which used chloride as a tracer. These experiments were used to develop and test instrumentation to be used in the large-scale test and to obtain initial estimates of aquifer characteristics. These estimates were used to predict the general behavior of the tracers and, thus, to plan the sampling scheme for the large-scale test. Garabedian *et al.* [1988] describe the results of these preliminary tests.

Tracer Characteristics

The tracers used in this test were selected to meet several criteria. First, they are nontoxic at low concentrations and could be used safely in this aquifer. Second, natural concentrations of the tracers were low so that the tracers introduced for this test could be followed for a reasonable distance and

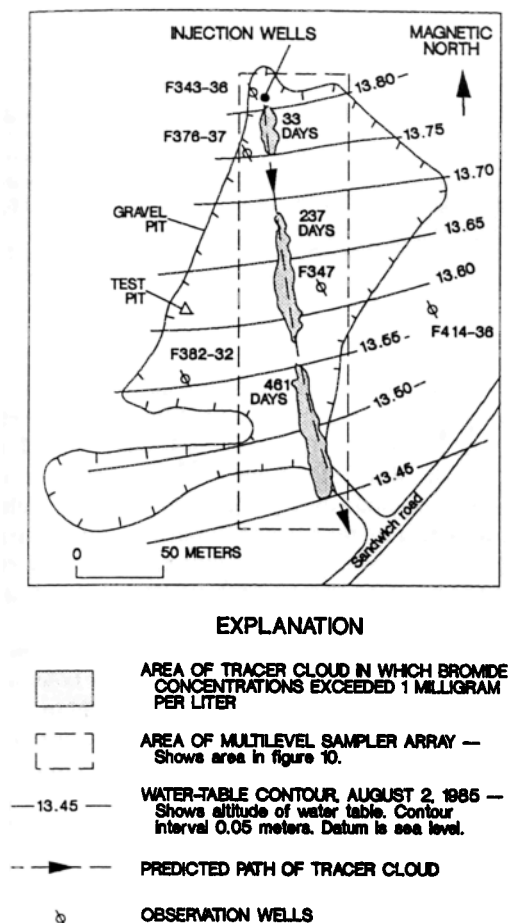


Fig. 4. Tracer test site in abandoned gravel pit, showing water table, location of selected monitoring sites, and predicted and observed path of bromide tracer cloud. Water table map from Garabedian *et al.* [1988, Figure 32]. Site of test pit in Figure 3 shown by triangle. Only observation wells referred to in this report are shown.

still be detected (Table 2). Finally, both nonreactive and reactive tracers were needed to meet the multiple objectives of the test.

Bromide (Br^-), a monovalent anion, was expected to be nonreactive [Davis *et al.*, 1980]. Its behavior is similar to that of chloride [Hem, 1985; Freyberg, 1986], but chloride is present in relatively high concentrations in this coastal aquifer (Table 1) and was not suitable for use in this test. Background bromide concentrations in the aquifer were generally less than 0.1 mg/L and averaged about 0.05 mg/L. An injection concentration of 640 mg/L bromide was used so that the tracer could be detected after as much as a 10,000-fold dilution by mixing with the ambient groundwater. This bromide concentration also kept the density difference between the tracer solution and the ambient groundwater less than one part in a thousand.

The reactive tracers were lithium (Li^+), molybdate (MoO_4^{2-}), and fluoride (F^-). Fluoride was abandoned as a tracer early in the test because fluoride concentrations were rapidly attenuated by sorption and, as a result, the fluoride cloud became masked by the significant amounts of fluoride already present in the sewage-contaminated groundwater.

The tracers were added as the salts LiBr, LiF, and

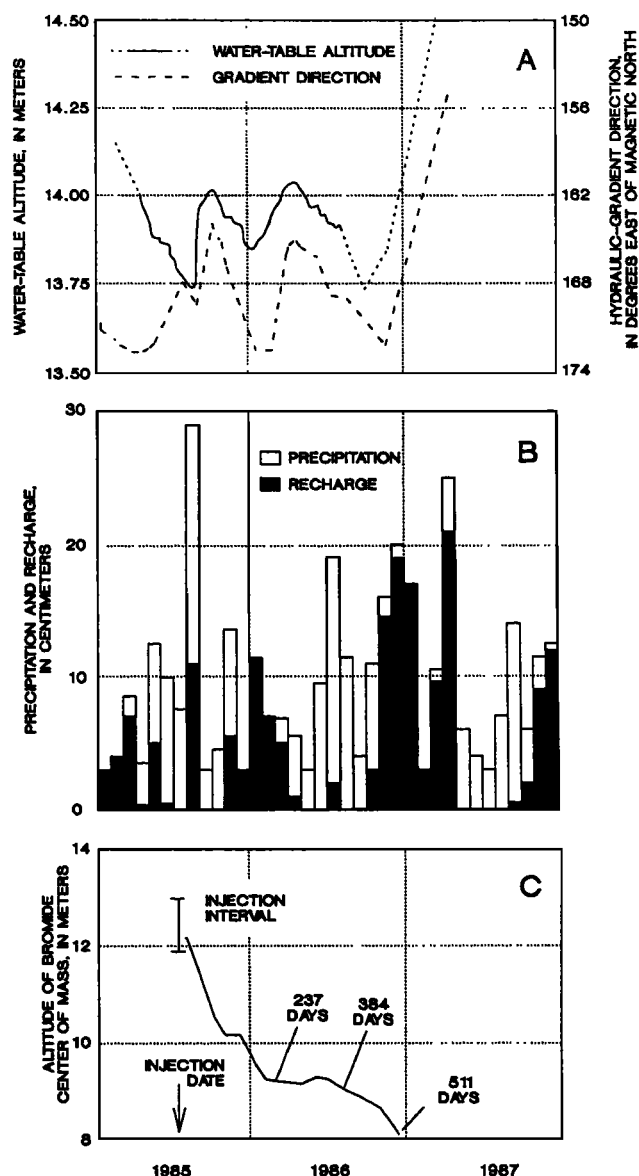


Fig. 5. (a) Altitude of the water table and direction of hydraulic gradient, (b) monthly precipitation and estimated recharge, and (c) vertical position of center of mass of the bromide cloud, 1985–1987. Water table altitude measured at well F376-37. Gradient direction determined from three wells (F343-36, F383-32, and F414-36). Location of wells shown in Figure 4.

Li_2MoO_4 to 7.6 m³ (2015 gallons) of water which was obtained from a shallow well screened in the uncontaminated zone above the sewage plume. The final solution, which was stored in two insulated tanks to maintain the water temperature near the ambient groundwater temperature, contained about 890 mg/L dissolved solids.

Injection of the Tracers

The tracers were injected into three 5.08-cm-diameter wells during a 17-hour period beginning on July 18, 1985, and ending on July 19, 1985. The tracer solution's initial temperature of 13.5°C rose gradually to 16°C during the injection period. The ambient groundwater temperature was about 13°C. Each injection well had a 1.2-m-long, slotted polyvinyl

TABLE 1. Chemical Characteristics of Groundwater at the Tracer Test Site

| Parameter | Well F347-20 (Above Sewage Plume) | Well F347-46 (in Sewage Plume) |
|---|---|--------------------------------------|
| Specific conductance, $\mu\text{S}/\text{cm}$ | 52. | 309. |
| Temperature, $^{\circ}\text{C}$ | 9.0 | 10.5 |
| pH | 5.8 | 6.4 |
| Dissolved oxygen (D.O.) | 11.2 | 0.3 |
| Total dissolved solids (TDS) | 32. | 185. |
| Alkalinity (as HCO_3^-) | 1.7 | 39. |
| Ca^{2+} | 1.1 | 8.5 |
| Mg^{2+} | .91 | 3.1 |
| Na^+ | 5.8 | 43. |
| K^+ | 0.8 | 7.7 |
| NH_4^+ (as N) | <0.01 | <0.01 |
| Cl^- | 8.1 | 27. |
| SO_4^{2-} | 7.8 | 33. |
| NO_3^- (as N) | <0.1 | 4.19 |
| PO_4^{3-} (as P) | 0.07 | 1.1 |
| SiO_2 | 6.1 | 15. |
| Fe (dissolved), $\mu\text{g}/\text{L}$ | <3. | 9. |
| Mn (dissolved), $\mu\text{g}/\text{L}$ | 35. | 850. |
| B, $\mu\text{g}/\text{L}$ | 10. | 330. |
| Dissolved organic carbon (DOC) | 0.31 | 2.23 |
| MBAS (surfactants) | 0.02 | 0.15 |

Concentrations in milligrams per liter unless otherwise noted. Samples collected on March 28–29, 1988, and analyzed by the USGS National Water Quality Laboratory. Dissolved oxygen measured with field probe. Alkalinity calculated from pH and dissolved inorganic carbon. Total dissolved solids estimated from sum of constituents. Locations of wells shown in Figure 4.

chloride (PVC) screen set at an altitude of 11.9–13.1 m, or about 1.2–2.4 m below the water table. The three wells were located 0.9 m apart along a line perpendicular to the flow of groundwater (Figure 7). The preliminary natural gradient test at the site had shown that the tracer cloud might sink below the injection zone early during the test. Therefore, the well screens were set near the water table so that the tracer

TABLE 2. Characteristics of Tracers

| Tracer | Injected Mass, g | Injected Concentration, mg/L | Background Concentration, mg/L |
|---|---------------------|------------------------------------|--------------------------------------|
| Bromide (Br^-) | 4900 | 640 | <0.10 |
| Lithium (Li^+) | 590 | 78 | <0.01 |
| Molybdate (MoO_4^{2-} as Mo) | 610 | 80 | <0.02 |
| Fluoride (F^-) | 380 | 50 | <0.20 |

cloud would remain as shallow as possible, thereby minimizing drilling costs during installation of the sampling network.

The total rate of injection was 7.6 L/min, or 2.5 L/min in each well. The slow rate of injection was chosen to reduce spreading of the cloud during injection. The initial volume of aquifer occupied by the tracer solution, assuming a porosity of 0.39 and no mixing with the ambient groundwater, was about 19.5 m^3 , equivalent to a box around the wells with dimensions of $1.2 \times 4 \times 4 \text{ m}$.

The actual solute distribution immediately after injection was not determined. Because of local aquifer heterogeneity at the injection site, the tracer distribution within the small initial volume would have been very complex. For example, concentrations during the Borden test one day after injection varied from background to injection levels over vertical intervals as small as 20 cm [Mackay *et al.*, 1986]. Despite the use of closely spaced sampling points around the injection wells at the Borden site, the estimate of chloride mass for the initial sampling was only 52% of the injected value [Freyberg, 1986], indicating a relatively poor delineation of the initial tracer cloud. Installation of many closely spaced sampling points near the injection wells during the Cape Cod test would have been impractical and would have greatly disturbed the aquifer, yet probably would not have yielded an accurate delineation of the initial tracer cloud.

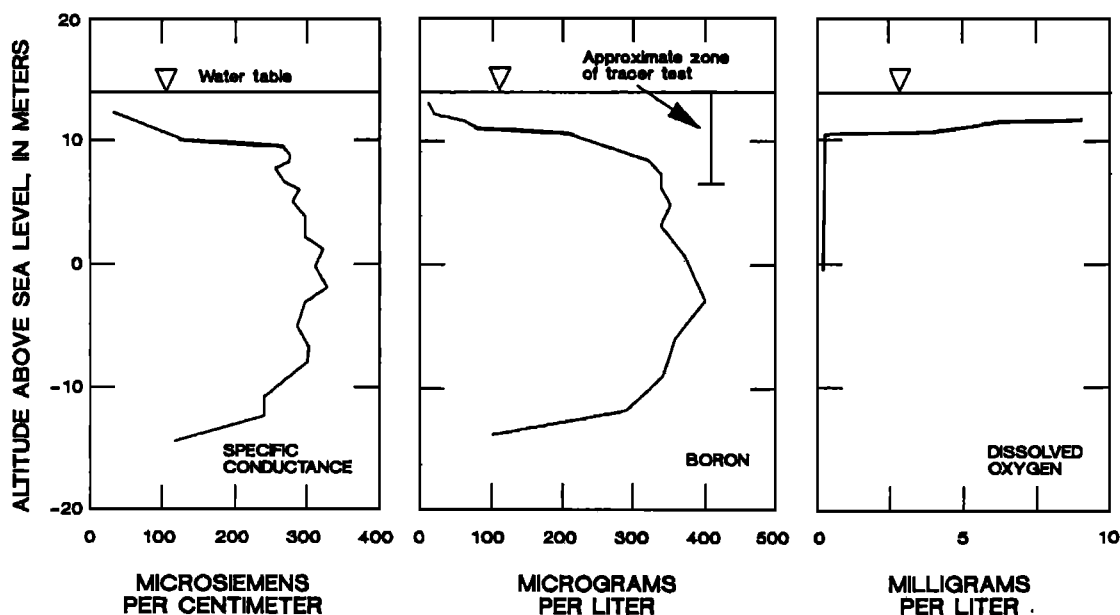


Fig. 6. Profiles of specific conductance, boron, and dissolved oxygen in a multilevel sampler at site F347, February 26, 1987. Dissolved oxygen measured in November 1986 [Smith *et al.*, 1989]. Location of site F347 shown in Figure 4.



Fig. 7. View looking south of multilevel sampler array. The three injection wells are in foreground. Distance to trees is about 225 m. Photograph taken in February 1987.

Multilevel Sampler Array

The distributions of the tracers in the aquifer were monitored by collection of water samples from the array of multilevel samplers (MLS) shown in Figure 7. The sampling array consisted of 656 MLS arranged in 71 rows and covered an area 12 to 22 m wide and 282 m long (Figure 8). The first 54 MLS were installed for the smaller-scale preliminary test that used chloride as the tracer. The remaining samplers were installed in stages throughout the test. After each round of sampling, the current location of the tracer cloud and the direction of groundwater flow inferred from the water table map were used to predict the cloud's path. Samplers were then installed ahead of the cloud to intercept its movement. Horizontal spacing between samplers increased gradually as the cloud spread. The spacing was dictated by the need to obtain sufficient data for the spatial moments analysis and by limitations on available drilling time.

The MLS, which are a modification of a design originally described by *Pickens et al.* [1978], allowed sampling of groundwater at closely spaced intervals in the vertical direction. Detailed vertical sampling was needed because limited vertical mixing and steep vertical concentration gradients were expected based on observations at the Borden site [Mackay et al., 1986], in the sewage plume [LeBlanc et al., 1987], and during the preliminary tracer tests [Garabedian et al., 1988].

Each MLS consists of 15 color-coded polyethylene tubes (0.47 cm inside diameter, 0.64 cm outside diameter). The tubes run from land surface, down the inside of a 3.17-cm-diameter PVC casing, and out holes drilled through the PVC at various depths. The open, down-hole end of each tube is screened with a fine nylon fabric. The vertical spacing between sampling ports generally is constant for a given MLS and varies with horizontal location in the array, from 25.4 cm near the injection wells to 76.2 cm near Sandwich

Road. This variation is illustrated by the representative section through the array shown in Figure 9. The total vertical span of the sampling ports, which was varied to capture the entire thickness of the cloud, ranges from 3.6 to 10.7 m. The array, when completed, included 9840 sampling points.

Most of the MLS were installed using a drive-and-wash drilling method to minimize disturbance of the unconsolidated sediments [Morin et al., 1988]. At each MLS site, a pilot hole was drilled with hollow stem augers to the water table, and flush-jointed steel casing (size NXWL, 6.03 cm inside diameter, 7.30 cm outside diameter) was set inside the augers and driven to the final depth. The sediment in the casing was washed out and the MLS was set inside the steel casing. The steel casing and augers were then pulled out and the sediments were allowed to collapse against the MLS. The annular space between the MLS and the outside of the steel casing was small (about 2.0 cm) and collapse was almost immediate. Geophysical logging of test wells installed by this method at the site indicated that voids did not remain along the casing after collapse (R. Morin, oral communication, 1985). Furthermore, field tests to detect vertical flow in the disturbed zone along the MLS [Garabedian et al., 1988] verified that short circuiting of tracer movement along the MLS, which would artificially enhance vertical spreading of tracers, did not occur. This conclusion seems reasonable because vertical hydraulic head gradients are very small at the site and because extremely steep vertical concentration gradients were observed in the tracer cloud during the test.

Although installation of the MLS disturbs the sediments, the extent of this disturbance is relatively small. A comparison of the area of disturbance around each MLS to the area of the aquifer centered around each sampler (based on horizontal spacing between samplers) shows that about 1% of the aquifer was disturbed near the injection wells where

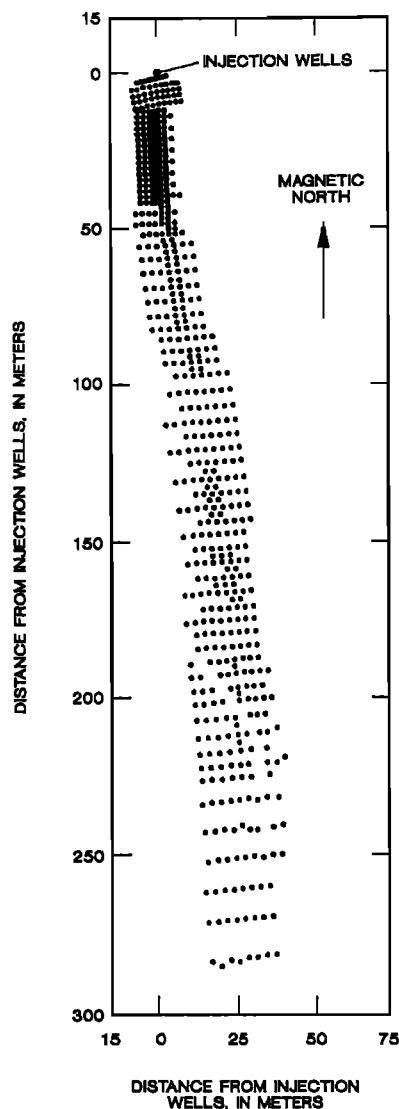


Fig. 8. Location of injection wells and multilevel samplers. Origin (0, 0) of magnetic north-oriented coordinate system is central injection well.

the MLS are close together. In most of the array, the disturbed area is only 0.07% of the total area around each MLS. When the plane perpendicular to groundwater flow along a row of MLS is considered, the disturbance ranges from 10% of the area of the vertical section near the injection wells to 4% in most of the array.

Because of time constraints, the last 15 rows of MLS (190–282 m from the injection point) were installed by augering to the final depth. In this area, the rows of MLS are spaced 4.5–9 m apart, so the increased relative disturbance caused by augering was considered to be minimal.

Collection and Analysis of Water Samples

Water samples were collected from subsets of the MLS array at about monthly intervals, beginning 13 days after injection, to obtain “snapshot” views of the three-dimensional distributions of tracer concentrations. For each round, the MLS to be sampled were chosen based on predictions of the location of the cloud from the previous

round’s data so that a complete three-dimensional view of the tracer cloud could be obtained. An attempt was made to characterize in detail the leading and trailing edges of the cloud as well as its center. As the test progressed, it was necessary to design a sampling scheme that captured the bromide cloud as well as the retarded clouds of lithium and molybdate.

A total of 19 rounds of sampling were completed between July 1985 and June 1988 (Table 3). The complete bromide distribution was captured only through December 1986. After that date, the leading edge of the bromide cloud had moved out of the array. The bromide cloud eventually traveled under Sandwich Road. Periodic sampling for the reactive tracers continued until June 1988.

Samples were collected simultaneously from the 15 ports on an MLS using sampling carts equipped with multihead peristaltic pumps. A predetermined volume of water (120–240 mL, or about three tube volumes) was pumped to waste from each tube to ensure that groundwater at the ports, not stagnant water in the tubes, was being sampled; the wastewater was removed from the site. Then, three samples were collected from each tube and stored in 60-mL polyethylene bottles at room temperature, without further preservation, until analysis. Sample preservation was determined to be unnecessary on the basis of field trials of the collection and analysis scheme.

Each sampling round generally took 2–3 days to complete with a crew of five people. As many as 13,000 water samples were collected from 40–290 MLS in a round. Although this is a large number of samples, the total mass of bromide removed from the aquifer was estimated to be between 2 and 5 g per round. Thus, only about 0.1% of the injected mass was removed from the aquifer during each sampling round.

The water samples were analyzed for bromide concentration using an ion-selective electrode, ion chromatography, and an autofluorescence technique [Orion Research, Incorporated, 1982; Fishman and Friedman, 1989]. Concentrations of bromide in all samples were determined initially with the electrode, which was accurate to concentrations as low as 1.0 mg/L. At concentrations below 1.0 mg/L, the electrode values tended to be higher than the actual bromide concentrations because of interference from other ions, particularly chloride, iodide, and ammonium, which are present at elevated concentrations in the sewage plume (Table 1). The effect of this interference was dependent on vertical location within the plume and, therefore, on depth below the water table. By comparing analyses by ion-selective electrode and ion chromatography on the same samples, Garabedian *et al.* [1988] developed an empirical correction for this depth-dependent interference which was used to correct all bromide concentrations measured with the electrode that were less than 1.2 mg/L. The correction factors also included an adjustment for the small background concentration of bromide in the aquifer. The accuracy of the empirical correction procedure was verified by comparing concentrations measured by both the ion-selective electrode and autofluorescence techniques on the set of samples collected in July 1986 [Garabedian *et al.*, this issue, Figure 3].

Lithium concentrations were measured by atomic adsorption spectrophotometry with direct aspiration [Fishman and Friedman, 1989]. Molybdate concentrations were measured

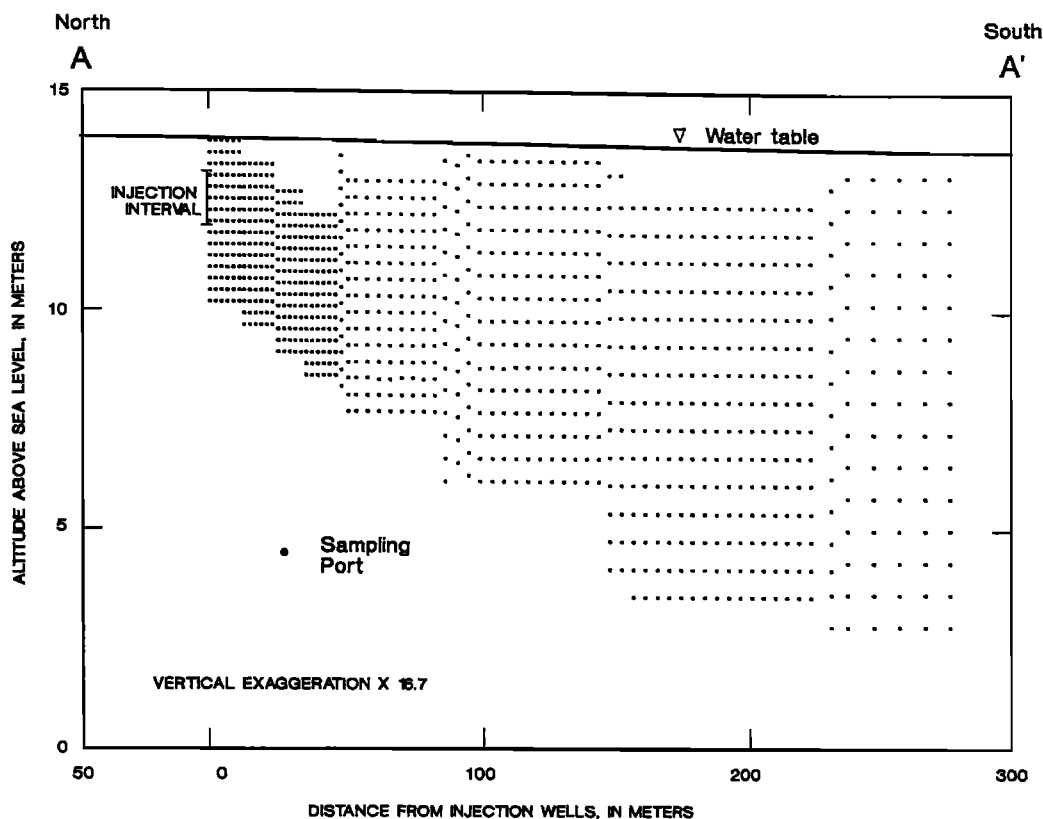


Fig. 9. Vertical location of screened interval of injection wells and ports on multilevel samplers along a representative longitudinal section through the sampler array. Water table is shown for August 2, 1985.

by a modification of the colorimetric thiocyanate technique [Hurd and Allen, 1935; Meglen and Glaze, 1973].

For each round, water samples were analyzed from the central MLS outward until the edges of the clouds were

defined. For this reason, the number of analyses shown in Table 3 is less than the total number of samples collected. The number of unanalyzed samples was small because the locations of the clouds were very predictable and few

TABLE 3. Summary of Sampling Rounds

| Nominal Date | Days Since Injection | Number of MLS Sampled | Number of Samples Analyzed | | | Maximum Observed Concentration, mg/L | | | Location of Center of Mass* of Bromide Cloud, m | | |
|----------------|----------------------|-----------------------|----------------------------|-----------------|--------------------------------|--------------------------------------|-----------------|--------------------------------|---|--------|------|
| | | | Br ⁻ | Li ⁺ | MoO ₄ ²⁻ | Br ⁻ | Li ⁺ | MoO ₄ ²⁻ | x | y | z |
| July 31, 1985 | 13 | 40 | 597 | 596 | 492 | 576 | 50.0 | 72.0 | 0.8 | -7.4 | 12.3 |
| Aug. 20, 1985 | 33 | 122 | 1700 | 1818 | 1403 | 429 | 30.0 | 50.3 | 2.7 | -16.9 | 11.7 |
| Sept. 11, 1985 | 55 | 149 | 2175 | 1800 | 1625 | 311 | 17.0 | 37.0 | 3.0 | -25.9 | 11.1 |
| Oct. 9, 1985 | 83 | 176 | 2264 | 2379 | 2188 | 124 | 12.0 | 26.0 | 5.6 | -38.9 | 10.6 |
| Nov. 6, 1985 | 111 | 174 | 1892 | 2054 | 1962 | 132 | 7.7 | 13.2 | 8.1 | -50.9 | 10.3 |
| Dec. 4, 1985 | 139 | 164 | 1916 | 1524 | 1734 | 76.6 | 6.2 | 12.8 | 10.2 | -64.7 | 10.4 |
| Jan. 8, 1986 | 174 | 160 | 1649 | 1447 | 1615 | 76.6 | 4.8 | 6.75 | 11.1 | -77.5 | 9.6 |
| Feb. 6, 1986 | 203 | 125 | 1592 | 1863 | 1314 | 61.5 | 3.0 | 8.29 | 11.4 | -88.8 | 9.4 |
| March 12, 1986 | 237 | 219 | 2147 | 1711 | 1874 | 65.2 | 2.3 | 6.77 | 11.9 | -100.1 | 9.3 |
| April 17, 1986 | 273 | 221 | 1923 | 1711 | 1767 | 46.9 | 1.0 | 4.45 | 13.3 | -114.3 | 9.2 |
| May 29, 1986 | 315 | 241 | 2270 | 1772 | 1960 | 62.3 | 1.0 | 1.84 | 16.7 | -133.9 | 9.4 |
| July 2, 1986 | 349 | 254 | 2247 | 1291 | 1455 | 64.5 | 0.80 | 1.95 | 19.0 | -147.5 | 9.4 |
| Aug. 6, 1986 | 384 | 288 | 2091 | 1885 | 2382 | 47.3 | 0.49 | 1.67 | 21.9 | -162.4 | 9.2 |
| Sept. 17, 1986 | 426 | 197 | 2002 | 994 | 1323 | 50.6 | 0.35 | 2.14 | 25.4 | -180.4 | 8.9 |
| Oct. 22, 1986 | 461 | 286 | 1656 | 2029 | 2706 | 39.0 | 0.33 | 1.76 | 28.6 | -196.3 | 8.7 |
| Dec. 11, 1986 | 511 | 290 | 1654 | 2244 | 2738 | 42.5 | 0.35 | 1.31 | 32.4 | -214.0 | 8.1 |
| May 6, 1987 | 657 | 283 | 0 | 2409 | 3160 | ... | 0.30 | 0.73 | ... | ... | ... |
| Aug. 12, 1987 | 755 | 287 | 0 | 3210 | 4247 | ... | 0.17 | 0.42 | ... | ... | ... |
| June 22, 1988 | 1070 | 142 | 0 | 0 | 2092 | ... | ... | 0.08 | ... | ... | ... |

*Location of center of mass calculated from first spatial moment of bromide distribution [Garabedian *et al.*, this issue]. The x (positive east) and y (positive north) coordinates are relative to magnetic north-oriented grid with origin at the central injection well. The z coordinate is altitude above sea level.

†Three dots indicate data were not collected for this sampling date.

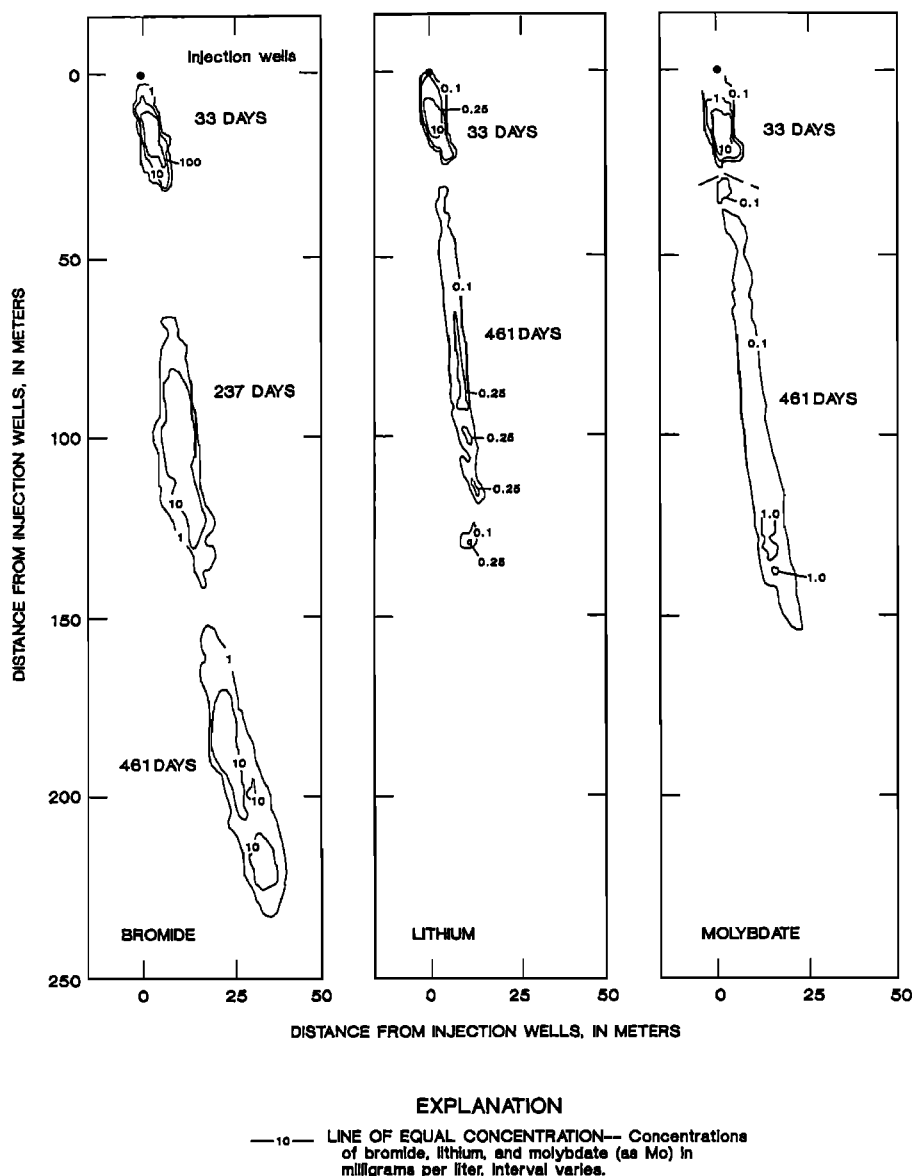


Fig. 10. Areal distribution of maximum concentrations of bromide at 33, 237, and 461 days and lithium and molybdate at 33 and 461 days after injection. Location of maps shown in Figure 4.

unnecessary samples were collected. The final data base for the period of July 1985 to June 1988 includes about 30,000 bromide analyses, 33,000 lithium analyses, and 38,000 molybdate analyses.

A measure of the success of the sampling scheme in describing the three-dimensional distributions of bromide was obtained from the spatial moments analysis. The calculated mass for each sampling round (the zeroth moment), which was obtained by spatially integrating the point values of bromide concentration over the volume of the cloud, ranged from 85% to 105% of the injected mass [Garabedian *et al.*, this issue]. For 14 of the 16 sampling rounds in which the bromide cloud was monitored, this percentage exceeded 90%. Also, there was no trend of increasing or decreasing mass with time. The excellent agreement between the calculated and injected mass indicates that the sampling scheme was successful in capturing the complete bromide cloud as it moved through the aquifer.

OBSERVED MOVEMENT OF THE TRACER CLOUD

The movement of the tracer cloud was tracked during the test by preparing maps and cross sections of the concentration data. In this paper, only selected views are shown to illustrate the cloud's movement and characteristics. The focus here is on the movement of bromide, the nonreactive tracer. Although the movement of lithium and molybdate is also discussed, the behavior of these reactive tracers will be presented in more detail in future papers.

Horizontal Movement

In Figure 10, the horizontal movement of the bromide cloud is illustrated by maps of maximum bromide concentration at 33, 237, and 461 days after injection. For each round shown, the maps were prepared by contouring values of the maximum bromide concentration at each MLS location, regardless of the depth at which the maximum oc-

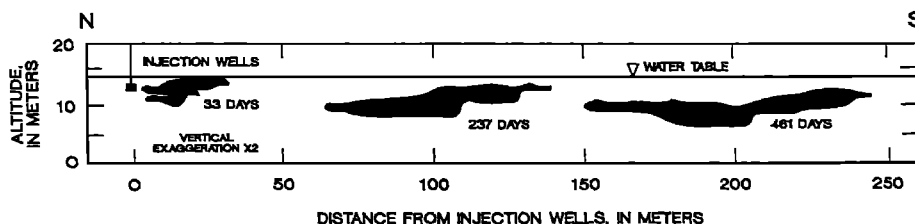


Fig. 11. Vertical location of bromide tracer cloud at 33, 237, and 461 days after injection. Cloud locations defined by zones in which bromide concentration exceeded 1 mg/L. Line of section approximately along A-A' in Figure 9.

curred. Thus, these maps delineate the maximum areal extent of the tracer cloud; similar maps were used during the test to guide the sampling strategy. It is important to note, however, that the maps of maximum concentration do not necessarily reflect the areal distribution of mass, as would maps of vertically averaged concentrations, nor do they represent horizontal slices through the tracer cloud.

During the test, the bromide cloud moved in a southerly direction along a path which matched the path predicted from the water table gradient (Figure 4). Although the direction of the water table slope varied only slightly during the test, Garabedian *et al.* [this issue] show that the trajectory of the center of mass of the bromide cloud followed these changes in gradient direction very closely.

The average rate of movement of the bromide cloud was 0.42 m/d [Garabedian *et al.*, this issue]. This observed rate matches the groundwater flow velocity estimated by Darcy's equation from the hydraulic gradient, hydraulic conductivity, and effective porosity. This average movement was accompanied by significant longitudinal spreading of the bromide cloud in the direction of flow. The cloud spread much less in the direction transverse to flow. At 461 days, the bromide cloud (as delineated by concentrations greater than 1 mg/L) was more than 80 m long but only 14 m wide. The significant longitudinal spreading was accompanied by a decrease in maximum bromide concentrations from 640 mg/L in the injection solution to 39 mg/L at 461 days (Table 3).

The lithium cloud followed a trajectory which was similar to that of the bromide cloud. However, its average rate of movement was greatly retarded. At 461 days, the lithium cloud (as delineated by concentrations greater than 0.1 mg/L) had spread to a length of almost 100 m, but its zone of maximum concentration had traveled only 90 m (Figure 10). In comparison, the bromide cloud had traveled 198 m, about twice as far, in the same time. The lithium distribution at 461 days was asymmetric, with maximum concentrations located close to the leading edge of the cloud and with a long tail of lower concentrations extending back toward the injection wells. Maximum concentrations of lithium decreased from 78 mg/L in the injection solution to 0.33 mg/L at 461 days.

Like lithium, the molybdate cloud (as delineated by concentrations greater than 0.1 mg/L) had traveled only about half as far as the bromide cloud after 461 days, but it had spread to a length of about 150 m (Figure 10). The shape of the molybdate cloud was similar to that of lithium, with highest concentrations located near the leading edge. Maximum concentrations of molybdate decreased from 80 mg/L in the injection solution to 1.76 mg/L at 461 days.

The observed retardation and attenuation of lithium and

molybdate is consistent with the expected sorption of these species onto the aquifer materials, based on geochemical considerations and laboratory experiments [Stollenwerk and Kipp, 1990; Wood *et al.*, 1990]. A spatial moments analysis [Garabedian *et al.*, 1988] indicated that the mass of lithium in solution decreased with time; at 461 days after injection, only about 10% of the injected mass remained in solution. The spatial moments also showed that the rate of movement of the lithium cloud decreased with time; the horizontal velocity of the center of mass was only 0.05 m/d during the later part of the test. The changing rate of lithium transport may reflect a nonequilibrium sorption process controlled by diffusion into pores within the sediment grains [Wood *et al.*, 1990]. A similar process has been hypothesized to explain the observed behavior of molybdenum, based on numerical simulations of laboratory column experiments [Stollenwerk and Kipp, 1990]. An analysis of the behavior of these two species during the test is currently under way.

Vertical Movement

The vertical movement of bromide is illustrated by the longitudinal sections of the bromide distribution at 33, 237, and 461 days after injection (Figures 11 and 12). During the test, the cloud moved downward, and a zone of groundwater that did not contain the tracer formed above the cloud. An analysis of the spatial moments of the bromide cloud [Garabedian *et al.*, this issue] shows that the center of mass moved downward (Figure 5 and Table 3) about 3.2 m during the first 237 days of the test (during the same period, the cloud moved about 100 m horizontally). Between 237 and 384 days, the bromide cloud moved mostly horizontally. After 384 days, the center of mass again began to move downward.

Two processes contributed to the downward movement observed during the test: (1) vertical components of flow associated with areal recharge and (2) sinking of the denser tracer cloud into the native groundwater. Both processes probably were important during the first 237 days after injection, when about 75% of the total vertical movement observed during the test occurred.

Although areal recharge contributed to the early vertical movement, it is unlikely that there was sufficient recharge to account for all the observed drop. During the first 237 days, 123 cm of precipitation were recorded at a weather station located 1.8 km from the site. Based on water balance calculations and estimates of typical seasonal losses to evaporation for Cape Cod [LeBlanc *et al.*, 1986], this precipitation resulted in about 60 cm of recharge (Figure 5). This is equivalent to about 1.5 m of water in the aquifer (given a porosity of 0.39), which is only half of the vertical movement

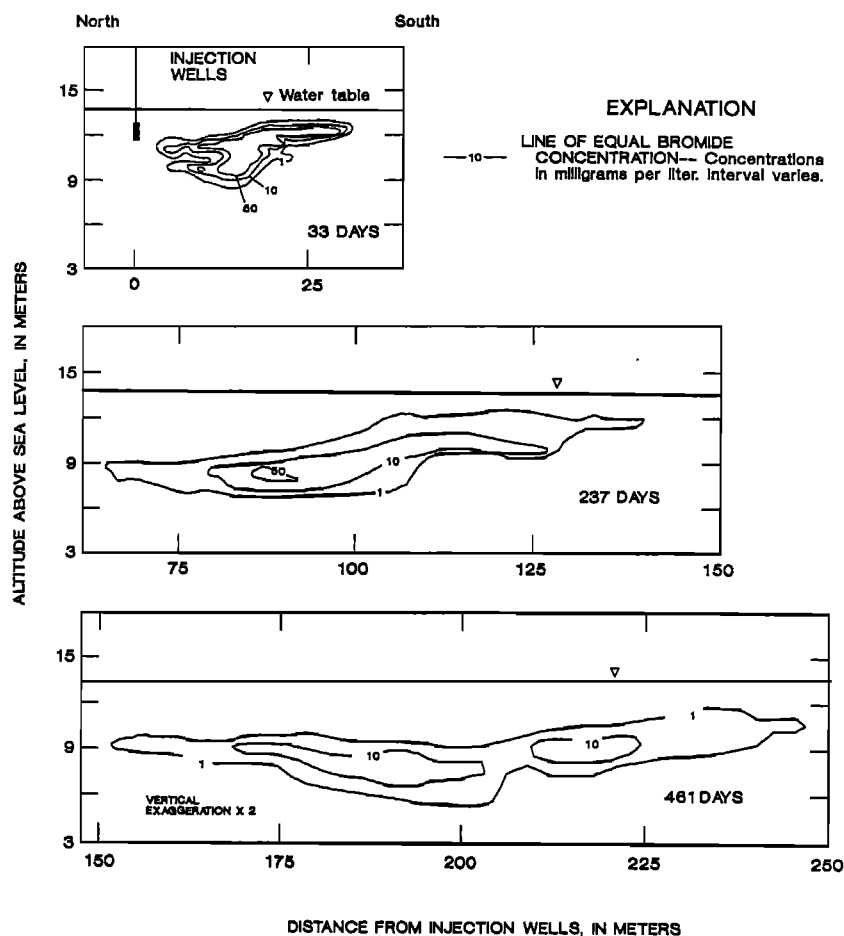


Fig. 12. Vertical distribution of bromide in tracer cloud at 33, 237, and 461 days after injection. Lines of section approximately along A-A' in Figure 9.

observed during this period. Downward flow caused by this recharge would be at an oblique angle to the water table. The angle of flow below the water table for a recharge rate of 60 cm in 237 days is about 1° , as estimated by the method of Jacob [1950]. This is about half the observed angle of 2° , based on the downward movement of the center of mass (3.2 m in 100 m of lateral transport) during this period. Thus, neither the amount of recharge nor the estimated downward component of flow at the water table caused by recharge is sufficient to explain entirely the downward movement observed early during the test.

The density contrast between the tracer solution and the ambient groundwater was sufficient to cause additional downward movement early in the test. The initial total dissolved solids concentration of the tracer solution was about 890 mg/L, which corresponds to a density of about 1.00089 g/cm^3 . The total dissolved solids concentration in the ambient groundwater ranged from 40 mg/L in the uncontaminated zone to about 150 mg/L in the sewage plume, corresponding to a density range of about 1.00004 – 1.00015 g/cm^3 . According to Hubbert's [1953] method of calculating fluid potentials, this density contrast is sufficient to cause observable sinking of the tracer cloud during the early part of the test. For the above density contrast and a ratio of horizontal to vertical hydraulic conductivity of 5:1, the predicted angle of downward movement of the tracer cloud within the ambient flow field is about 6° . The sinking of the

tracer cloud is also predicted by the method of Yih [1965], which assumes that the injected fluid body is finite in size and does not mix with the ambient fluid. For the above density contrast, without a correction for anisotropy of hydraulic conductivity, the predicted angle of downward movement of a spherical body equivalent in volume to the injected cloud is about 8° . The observed angle of downward movement of the bromide cloud during the first 83 days was about 3° .

Sinking caused by density differences most likely occurred early during the test when the density contrast between the waters was greatest. During the first 33 days of the test, for example, the center of mass moved downward 0.8 m, which is about 20% of the 4.4-m drop observed over 511 days. As concentrations in the tracer cloud decreased during the first 237 days because of dispersion (Table 3), the effect of density on the vertical movement of the cloud probably diminished and gradually became insignificant.

During the period from 237 to 384 days after injection, there was little recharge and the bromide cloud moved mostly horizontally (Figure 5). Significant recharge in late 1986 may have caused the renewed downward movement of the bromide cloud after 384 days.

The accretion of areal recharge at the water table contributed to the zone of water which formed above the bromide cloud. At 461 days after injection, this zone was 2–3 m thick (Figure 12); assuming a porosity of 0.39, this is equivalent to

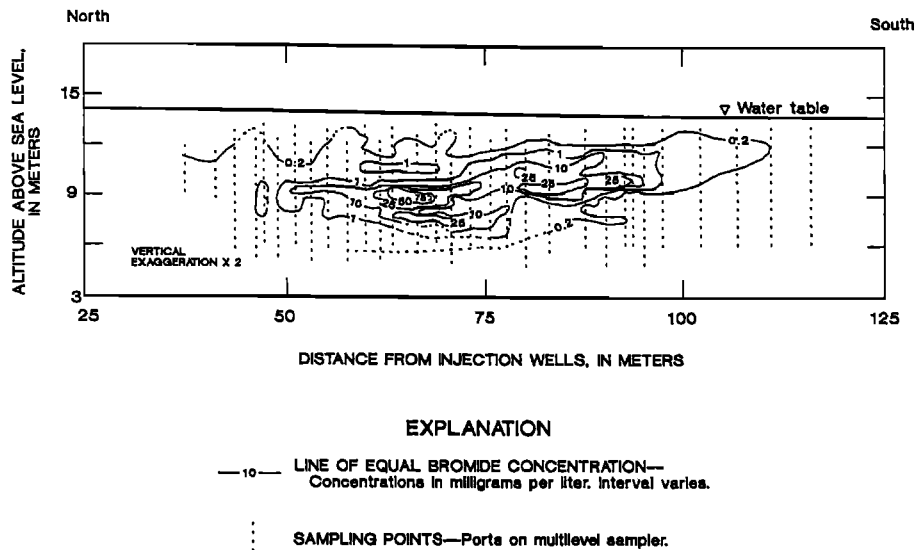


Fig. 13. Vertical distribution of bromide in tracer cloud at 174 days after injection. Line of section approximately along A-A' in Figure 9.

about 0.8–1.2 m of water in the aquifer. In comparison, the 147 cm of precipitation which fell during the 461-day period resulted in about 70 cm of recharge (Figure 5). Displacement of native groundwater around the denser, sinking tracer cloud early in the test also may have contributed to the zone.

After the initial vertical spreading of the tracer cloud during injection and the subsequent period of density-induced sinking early in the test, the bromide cloud spread only slightly in the vertical direction and remained about 4–6 m thick as it moved laterally in the aquifer. At 511 days, the last complete view of the bromide cloud, the cloud was 90 m long, 15 m wide, and about 6 m thick.

The bromide cloud had two zones of elevated concentration (Figures 12 and 13), one near the leading edge of the cloud and just below the water table and another near the center of the cloud and deeper in the aquifer, which gave it an asymmetric shape. The leading edge of the bromide cloud advanced at rates as high as 0.9 m/d during the early part of the test, whereas the trailing edge advanced at the much slower rate of about 0.2 m/d. The higher apparent velocity of the leading edge near the water table may reflect a zone of higher hydraulic conductivity. Measurements of hydraulic conductivity from cores [Wolf, 1988] and from borehole flowmeter tests [Hess, 1989] at a site 15 m west of the tracer test array detected a zone near the water table in which hydraulic conductivities are as great as 260 m/d. Because similar local variations in hydraulic conductivity may exist at the injection wells, part of the injected solution may have moved rapidly outward from the wells in a very permeable layer near the water table, while the remainder of the solution began sinking into the aquifer under the driving force of the density difference. Although both factors would have been influential only for a short distance from the wells, the resulting shape of the cloud persisted because vertical mixing was limited.

DISCUSSION

Predictability of Transport

The pattern of bromide concentration in the tracer cloud was very complex (Figure 13), particularly early in the test.

Although the complex spatial variations in concentration tended to become smoother as the cloud moved down-gradient, the general trend of these variations persisted during the entire test. This complexity, which reflects the spatial variation in hydraulic properties of the aquifer, persisted at later times because mixing, particularly transverse to flow, is limited.

Despite the complex variations in concentrations in the cloud, the general behavior of the cloud was very predictable. The path of the cloud was accurately predicted from the observed water table gradient (Figure 4), and the observed average rate of movement of the bromide cloud matched predictions made from measurements of the hydraulic gradient and estimates of hydraulic conductivity and effective porosity. The movement of the cloud also closely followed the changes in direction of the hydraulic gradient with time [Garabedian *et al.*, this issue]. These accurate predictions were possible because the groundwater flow system at the site is relatively simple and had been well defined during initial hydrogeologic studies. The predictions were based on very careful measurement of the hydraulic gradient using a network of 25 water table observation wells. This illustrates the importance of accurate definition of the water table as a first step in the study of contaminant transport in unconfined aquifers of this type. For example, the path of the 3-km-long sewage plume from the Otis Air Base sewage treatment plant also was accurately predicted from a regional water table map [LeBlanc, 1984a].

Comparison to the Borden Test and the Sewage Plume

The general movement of the tracer clouds at the Borden site [Mackay *et al.*, 1986] and the Cape Cod site was very similar despite differences in aquifer properties. The average rate of groundwater movement at the Cape Cod site was 5 times faster than at the Borden site; although the hydraulic gradient was larger at the Borden site (0.0043 compared to 0.0015), the aquifer at the Borden site has a much lower average hydraulic conductivity (6 m/d compared to 110 m/d). The Borden aquifer is a clean, well sorted, fine to medium

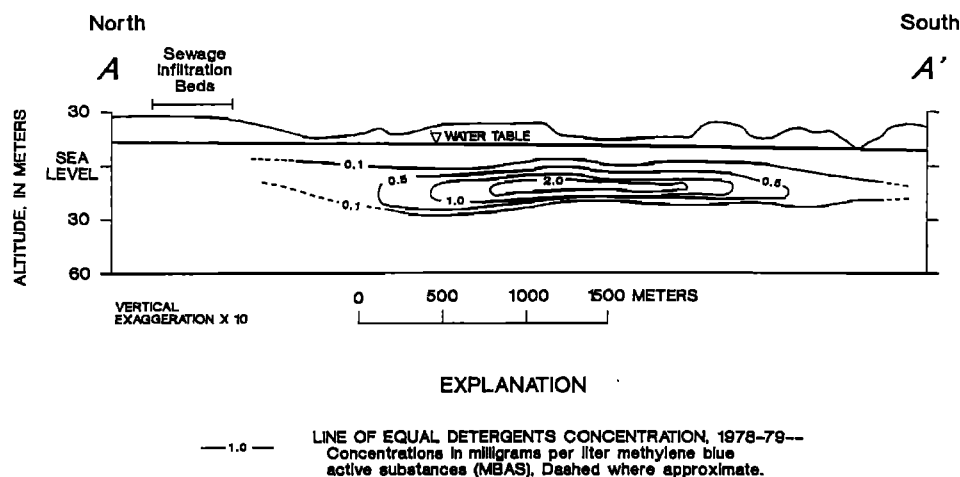


Fig. 14. Vertical distribution of detergents in sewage plume. Line of section shown in Figure 1 [from *LeBlanc*, 1984a, Figure 15].

sand, whereas the Cape Cod aquifer is a clean, medium to coarse sand and gravel. Despite these differences in hydraulic properties, the tracer clouds in both experiments spread mostly in the longitudinal direction. Spreading transverse to flow was limited in the horizontal direction and very limited in the vertical direction.

In both experiments, the tracer clouds moved downward during the early parts of the tests. The rate of vertical movement decreased with time, which suggests that the sinking was initially caused by a density contrast which diminished as concentrations in the clouds were diluted by dispersion. Additional sinking also occurred at both sites because of recharge from precipitation, which gradually formed a zone of uncontaminated groundwater above the tracer clouds. Simulations of the Borden test with a density-dependent flow and transport model [van Walsun, 1987] and the preliminary analysis of the Cape Cod test with the methods of Hubbert [1953] and Yih [1965] support this hypothesis. Additional work is under way to test this hypothesis with numerical models of the Cape Cod experiment.

These features (alignment in the direction of flow, minimal mixing in the vertical direction, and sinking below the water table) have been reported for other plumes in shallow aquifers [Perlmutter and Lieber, 1970; Kimmel and Braids, 1980; MacFarlane *et al.*, 1983; *LeBlanc*, 1984a; Ryan and Kipp, 1985]. In these cases, the source of contamination was continuous rather than a pulse injection. Because transverse dispersion is small, the plumes formed narrow tongues in which high concentrations persisted hundreds of meters from the source.

The sewage plume on Cape Cod illustrates the features commonly reported for contaminant plumes at a scale that is 10 times larger than the tracer experiment [LeBlanc, 1984a]. The similarity between the sewage plume and the tracer cloud, despite the difference in scales, can be demonstrated by comparing the cross sections of the tracer cloud (Figure 12) with a longitudinal cross section of the detergent distribution in the sewage plume (Figure 14). The detergent plume, which extends more than 3 km from its source at the Otis Air Base sewage treatment plant, is about 23 m thick and is overlain by 6 to 15 m of uncontaminated groundwater.

Detergents entered the aquifer at the treatment facility as

relatively nonbiodegradable surfactants between 1946 and 1964, and are present at concentrations exceeding 0.5 mg/L in a zone that extends from 900 to 3000 m from the infiltration beds [LeBlanc, 1984a; Thurman *et al.*, 1986]. Detergent concentrations in the wastewater decreased significantly in 1964 when biodegradable surfactants came into widespread use. The longitudinal smearing of the leading and trailing edges of the detergents plume may be caused in part by longitudinal dispersion, but the smearing may also reflect temporal variations in source concentrations.

Vertical spreading of the detergent plume occurs near the disposal site and is caused largely by vertical flow as the wastewater enters the aquifer. As the plume moves down-gradient, the contaminated water does not mix significantly with the overlying, uncontaminated water from areal recharge. The lack of vertical mixing, also observed during the tracer test, results in steep vertical concentration gradients [LeBlanc *et al.*, 1987] which persist more than 3 km from the infiltration beds. The tracer cloud and the sewage plume illustrate that this lack of vertical mixing can produce thin contaminated zones far from the source that may be difficult to locate and characterize.

SUMMARY

A large-scale natural gradient tracer test was conducted in a sand and gravel aquifer on Cape Cod, Massachusetts. The test was designed to obtain a detailed description of the movement of a nonreactive (bromide) and two reactive (lithium and molybdate) tracers as they moved through the heterogeneous, stratified glacial outwash.

The observed three-dimensional distributions of the tracers are being used to address several questions concerning the effects of heterogeneity on solute transport. The primary goal of the test was to examine macrodispersion of solutes and, in particular, to test the field applicability of stochastic theories that relate dispersion to spatial variability of hydraulic conductivity. A second goal was to examine the transport of several reactive tracers that were expected to be sorbed onto the sediments and, in particular, to assess the applicability of laboratory-derived chemical parameters to the prediction of the behavior of reactive species in heterogeneous aquifers.

In order to achieve these goals, the tracer test included the controlled injection of a pulse of tracers into the aquifer and the subsequent monitoring of movement of the tracer cloud by periodic collection of water samples from an array of multilevel samplers. This monitoring scheme provided a series of three-dimensional views of the tracer cloud as it moved down-gradient through the sampling array. The test began in July 1985 with the injection of 7.6 m³ of water containing bromide, lithium, and molybdate. Water samples were collected from the multilevel samplers during 19 synoptic sampling rounds done between July 1985 and June 1988. The analysis of these samples produced a data base containing about 30,000 bromide analyses, 33,000 lithium analyses, and 38,000 molybdate analyses.

The bromide cloud moved southward at a rate of 0.42 m/d, which corresponds closely to that predicted from the hydraulic gradient and from aquifer properties. Also, its path matched that predicted from the water table gradient. The cloud moved downward about 3.2 m during the first 237 days of the test. Part of this movement probably was caused by sinking of the denser tracer cloud in the native groundwater. Accretion of recharge from precipitation also contributed to the downward movement. The bromide cloud spread mostly in the longitudinal direction; spreading in the transverse directions was limited. At 461 days after injection, the bromide cloud had moved about 200 m, and it was 14 m wide, 4–6 m thick, and more than 80 m long.

The lithium and molybdate clouds followed the same trajectory as the bromide cloud. However, the rates of movement of these reactive tracers were retarded, as had been predicted from the results of laboratory experiments. In addition, the distributions of lithium and molybdate were asymmetric, with the highest concentrations near the leading edges of the clouds and long tails of lower concentration extending back toward the injection wells.

The general behavior of the tracer cloud was similar to that reported at the Borden site. The features of the cloud also were similar to those reported in several field studies of much larger contaminant plumes. These features included the predominant spreading in the longitudinal direction; the lack of transverse mixing, particularly in the vertical direction; the alignment of the plumes in the direction of flow; and sinking below the water table. Despite the inherent complexity of the concentration distributions in the tracer cloud, which was the result of aquifer heterogeneity, the general behavior of the cloud, seen in maps and in the spatial moments, was predictable from a careful analysis of the geohydrologic system.

This experiment met the goal of providing a detailed three-dimensional data set with which to test the effects of aquifer heterogeneity on transport. In a companion paper, Garabedian *et al.* [this issue] present a detailed analysis of the spatial moments of the bromide cloud and relate these moments to macrodispersion. Studies are under way to compare these field-measured dispersivities to values predicted by the stochastic theories. Preliminary results of this work have been reported by Wolf [1988] and Hess [1989]. The tracer test has also provided an extensive data set on the movement and retardation of two reactive species, and work is under way [Stollenwerk and Kipp, 1990; Wood *et al.*, 1990] to relate these observations to laboratory experiments and measurements of geochemical parameters at the site.

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