Kilometer-Scale Rapid Transport of Naphthalene Sulfonate Tracer in the Unsaturated Zone at the Idaho National Engineering and Environmental Laboratory

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ABSTRACT

To investigate possible long-range flow paths through the interbedded basalts and sediments of a 200-m-thick unsaturated zone, we applied a chemical tracer to seasonally filled infiltration ponds on the Snake River Plain in Idaho. This site is near the Subsurface Disposal Area for radioactive and other hazardous waste at the Idaho National Engineering and Environmental Laboratory. Within 4 mo, we detected tracer in one of 13 sampled aquifer wells, and in eight of 11 sampled perched-water wells as far as 1.3 km away. These detections show that (i) low-permeability layers in the unsaturated zone divert some flow horizontally, but do not prevent rapid transport to the aquifer; (ii) horizontal convective transport rates within the unsaturated zone may exceed 14 m d^{-1}, perhaps through essentially saturated basalt fractures, tension cracks, lava tubes, or rubble zones; and (iii) some perched water beneath the Subsurface Disposal Area derives from episodic surface water more than 1 km away. Such rapid and far-reaching flow may be common throughout the Snake River Plain, and possibly occurs in other locations that have a geologically complex unsaturated zone and comparable sources of infiltrating water.

Large-scale transport in the unsaturated zone is commonly assumed to be slow (a few meters per year or less) and predominantly vertical. Slow transport is likely if the flow proceeds in classic diffuse fashion, limited by low unsaturated hydraulic conductivity. Vertical transport is likely if the main transport mechanisms are dominated by gravity, as opposed to pressure or concentration gradients, and if the subsurface is effectively homogeneous within each horizontal plane.

The slow-flow generalization may not hold for geologically diverse unsaturated zones or for large, nonuniform inputs of water over the land surface. Preferential flow paths can transport water and contaminants horizontally to adjacent regions or vertically to the aquifer far sooner than might be predicted based on bulk medium properties and Richards' equation. Another important effect of preferential flow is that a relatively small fraction of the subsurface medium interacts with the contaminants, which limits adsorption and other attenuating processes.

Fluid transport that is difficult to explain by common unsaturated-flow concepts is suggested in several recent cases of contaminant detections in the unsaturated zone at substantial horizontal distances from the source of contamination. At a location about 100 m away from a waste burial site in the Amargosa Desert of Nevada, tritium has been detected in plants and in boreholes within the 108-m-thick unsaturated zone at much greater than ambient concentrations (Prudic and Striegl, 1995). Horizontal liquid movement along hypothetical preferential flow paths might plausibly cause such transport (Striegl et al., 1996; Prudic et al., 1997). In the Mojave River Basin of California, Izbicki et al. (1998) found evidence of unsaturated-zone horizontal movement of liquid water 45 m from a stream channel, on the basis of the detection of atmospherically derived tritium presumed to have infiltrated through the channel. At a nearby site, Izbicki (personal communication, 2000) noted evidence of gaseous chlorofluorocarbon transport, horizontally within the unsaturated zone to distances of 400 m from a landfill. On the Snake River Plain in southeastern Idaho near the Idaho National Engineering and Environmental Laboratory (INEEL), detection of chlorofluorocarbons, tritium, carbon-14, and chlorine-36 in the unsaturated zone suggests they may be transported vertically to the water table at about 200 m depth and horizontally as much as several kilometers in a few years (E. Busenberg and L.D. Cecil, personal communication, 1998). Contaminants in groundwater and inferred water recharge temperatures at this location suggest that recharge through the unsaturated zone can be rapid (Busenberg et al., 1993). In another INEEL study, a 1994 field experiment known as the Large-Scale Infiltration Test, water that infiltrated from a 200-m-diameter basin was found to move at a rate of 5 m d^{-1} (Burgess, 1995; Wood and Norrell, 1996; Dunnivant et al., 1998).

At waste-disposal or spill sites, tracer studies are commonly based on detection of contaminants that derive from the pollution source. Conclusions from such studies sometimes raise questions about other possible sources of contamination closer to the point of detection, other possible modes of transport (e.g., through the atmosphere or on contaminated waste-conveying vehicles), the uncertainty of timing, and the magnitude of contaminant release. Field experiments with deliberately introduced chemicals as tracers avoid some of these uncertainties and afford fairly precise knowledge of the position and temporal nature of the source.

This paper describes an artificial tracer experiment near the Subsurface Disposal Area of the Radioactive Waste Management Complex at the INEEL (Fig. 1). This facility has a variety of radioactive and hazardous chemical wastes buried at shallow depths on the eastern Snake River Plain. Some contaminants have traveled down to the large and heavily used Snake River Plain aquifer (Laney et al., 1988). At this semiarid location, seasonal streamflow, snowmelt, and local runoff episodically generate large quantities of infiltrating water for

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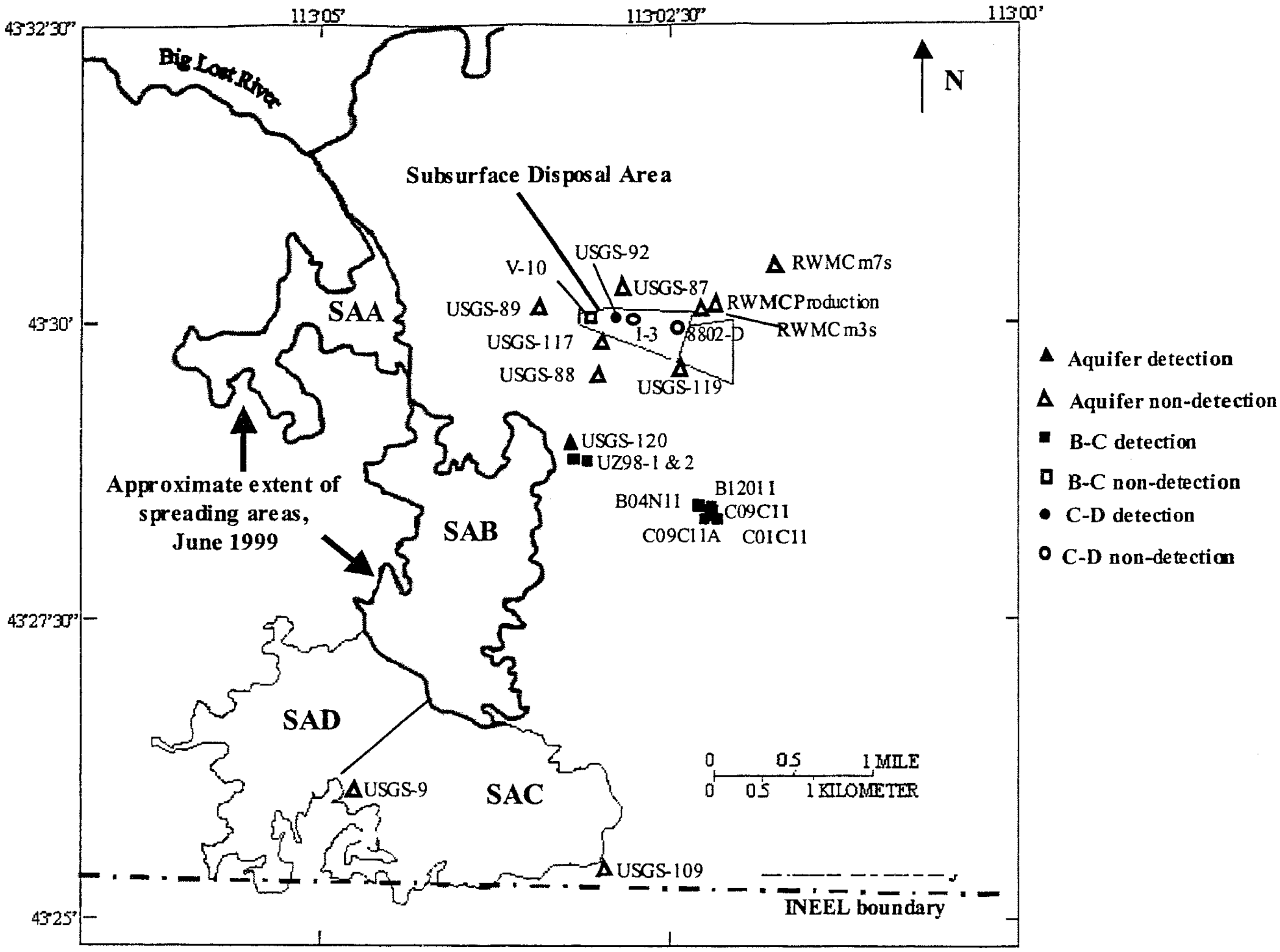


Fig. 1. The Idaho National Engineering and Environmental Laboratory Subsurface Disposal Area and vicinity, including spreading areas SAA, SAB, SAC, and SAD. The prevailing aquifer flow direction is toward the southwest. The predominant dip direction of the BC and CD interbeds is toward the east. Point symbols indicate wells sampled to determine tracer concentrations. Symbol shape indicates the stratigraphic unit associated with that well. Solid symbols represent wells where there was a significant positive detection of tracer during the experiment, and open symbols represent wells where there was no such detection.

periods of days or weeks. Particular concerns arise from seasonal artificial diversions of the Big Lost River to enhanced depressions called spreading areas, which serve as infiltration ponds. Contaminants from the Subsurface Disposal Area are known to have migrated to considerable depths in the unsaturated zone, so this additional water may enhance their further migration toward the aquifer.

Although several investigators (Robertson et al., 1974, p. 26; Hubbell, 1990; Rightmire and Lewis, 1987b) have hypothesized that water from the Big Lost River and spreading areas may affect flow in the unsaturated zone beneath the Subsurface Disposal Area, the distance (more than 1 km) and the generally expected insignificance of horizontal flow in the unsaturated zone has led to an expectation that such influence would be negligible. Some results support this assumption, for example, those of the Large-Scale Infiltration Test described above. Dunnivant et al. (1998) noted that through fractured basalt above the 55-m depth, water appeared to flow downward "within a cylinder defined by the infiltration basin." On the other hand, these investigators reported perched water and some horizontal movement just above the sediments at this level. Busenberg et al. (1993) noted that fast horizontal transport might account for previously observed horizontal spreading of ^{36}Cl and chlorofluorocarbons at the INEEL.

This evidence is inconclusive on the issue of significant horizontal flow, and on the issue of the ability of contrasting layers to inhibit vertical flow to the aquifer. Our investigation tests both of these issues, with emphasis on the hypothesis that short-term, large-volume infiltration may cause rapid, long-range transport through the unsaturated zone near the INEEL Subsurface Disposal Area.

SITE DESCRIPTION

Geology and Stratigraphy

Because the geologic character of the unsaturated zone near the INEEL Subsurface Disposal Area is critical to the transport phenomena explored in this study, we describe its major transport-related features in significant detail.

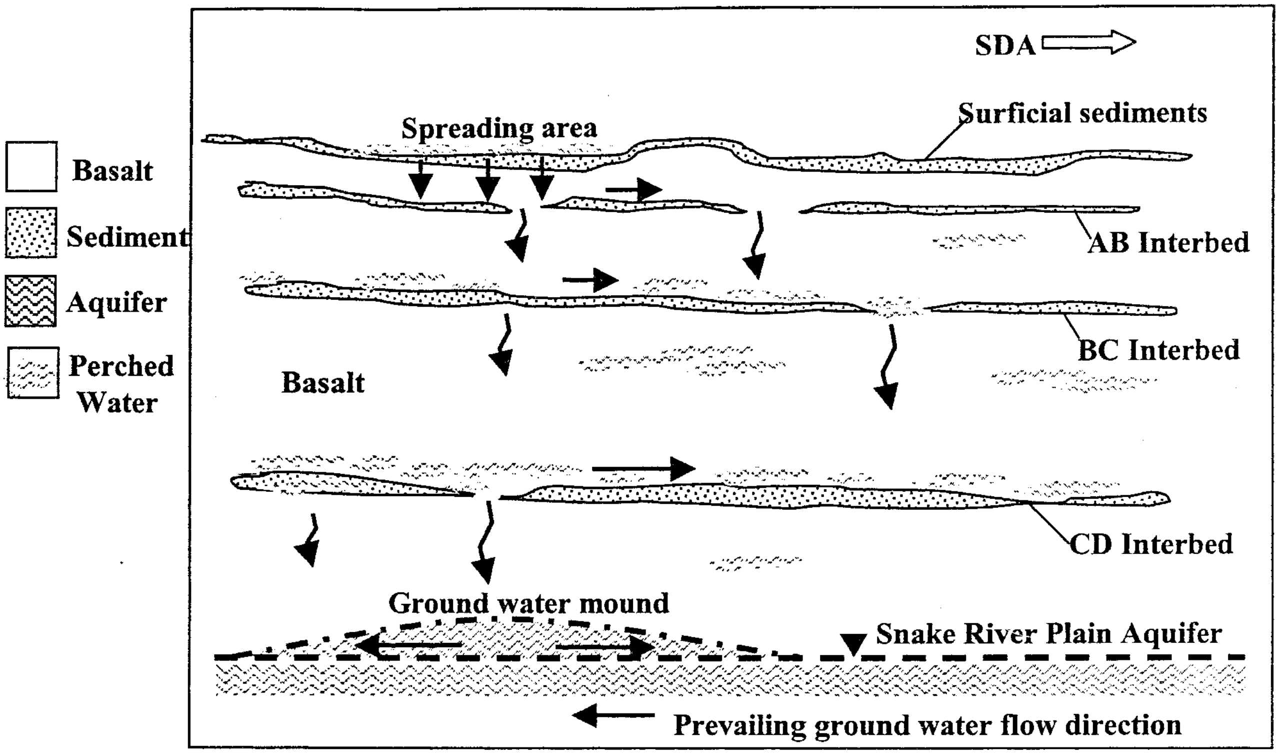


Fig. 2. Diagrammatic cross section illustrating major subsurface features, especially basalt layers, interbeds, and hypothetical perching. Arrows indicate hypothetical flow paths. This figure is not to scale; the actual depth of the BC interbed is about 35 m, of the CD interbed about 70 m, and of the water table about 200 m.

The unsaturated zone near the Subsurface Disposal Area is highly stratified and about 200 m thick, as illustrated in Fig. 2 and 3 (Anderson and Lewis, 1989; Anderson et al., 1996). Additional scale drawings and other stratigraphic information are in the works of Barraclough et al. (1976), Rightmire and Lewis (1987a,b), and Anderson and Liszewski (1997). The unsaturated zone comprises low-permeability layers of fine sediments or dense basalt, and layers of fractured or rubbly basalt that conduct water easily when wet. Depending on the magnitude and prevalence of critical hydraulic processes within these layers, they may accelerate or retard contaminant transport, concentrate or dilute contaminants, and establish the dominant movement as vertical or horizontal.

Individual basalt flows mostly are thin (3–9 m), tube-fed pahoehoe flows similar to those of Hawaii (Anderson et al., 1999). Outcrop exposures indicate that individual basalt flows

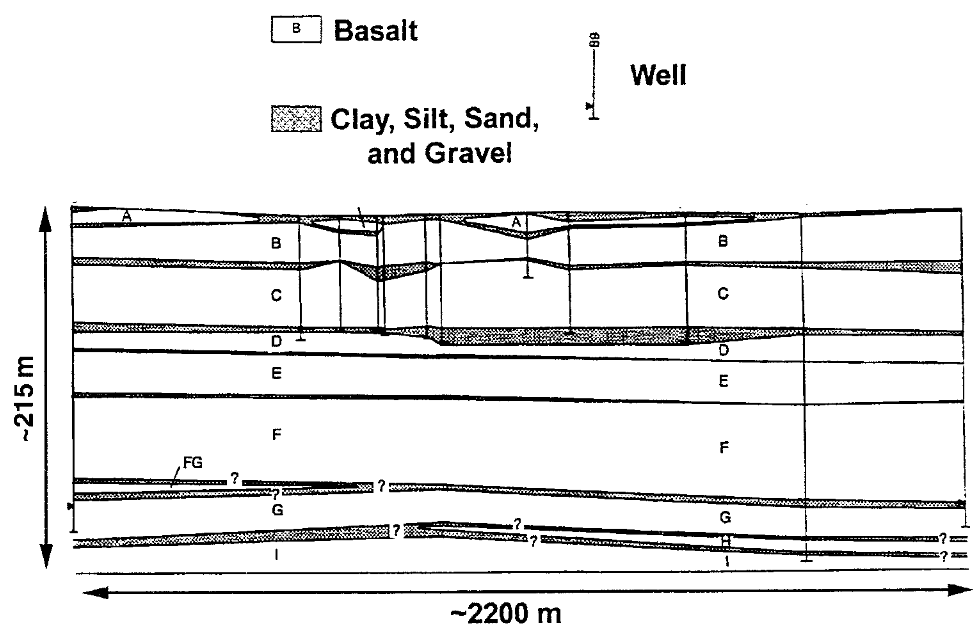


Fig. 3. West to east schematic cross section centered on the Subsurface Disposal Area (modified from Anderson and Lewis, 1989, p. 25). This diagram is based on data from 12 boreholes shown here as vertical lines. The linear interpolations between these indicate less variability in interbed thickness and topography than actually exists.

generally form long, sinuous lobes having median length/width ratios of about 3:1 (Welhan et al., 1997). Some flows are as long as several kilometers (Kuntz et al., 1994). Void spaces in these flows are greatest along their top and bottom surfaces and near volcanic vents. Many of the flows have numerous features such as lava tubes and closely spaced fractures up to a few meters wide and tens of meters long. These may be open or filled with rubble, sediment, or younger lava. In some places, basalt flows are cut by vertical fissures, dikes, and tension cracks associated with northwest-trending volcanic rift zones (Anderson et al., 1999; Hughes et al., 1999). Where they can be observed at the land surface, tension cracks cut vertically through numerous flow groups, are as wide as 2 m, and occur as closely spaced openings along the edges of eruptive fissures over distances of many kilometers.

Basalt flows and other volcanic deposits combine into a basalt flow group, a complex assemblage of overlapping flows and deposits related to a single eruption. Three important basalt flow groups, called A, B, and C, are within about 70 m of the surface at the Subsurface Disposal Area.

Rubble zones are common along the rapidly cooled margins of basalt flows. Basalt rubble and scoria (cinders) have been described in some wells near the Subsurface Disposal Area, but have not been systematically characterized. The potential for scoria is greatest in basalt-flow group C because its vent is thought to be located in this vicinity (Anderson and Liszewski, 1997).

Between some individual basalt flows, and especially between flow groups, sedimentary interbeds consist of well sorted to poorly sorted deposits of clay, silt, sand, and gravel. These interbeds are named after the basalt flow groups they lie between, for example, AB, BC, and CD (Fig. 2). Their depths below land surface vary topographically, but the two most prominent interbeds under the Subsurface Disposal Area, the BC and CD interbeds, are traditionally labeled with depths of 34 and 73 m, respectively. Near the Subsurface Disposal Area, they tend to dip in an easterly direction, the BC interbed about 3.8 m km^{-1} and the CD about 4.7 m km^{-1} on average (Anderson and Lewis, 1989). Their thicknesses vary in accordance with the topography of underlying basalt flows, and in some places they pinch out to zero thickness. Immediately below a particular basalt flow, an interbed is likely to have a baked zone of sedimentary or volcanic material whose properties may have been altered by exposure to the heat of fresh lava.

Barraclough et al. (1976) noted that at the interface between basalt and sedimentary layers, the permeable openings into the basalt are partially filled by sediment, the nature of which has been further investigated by Rightmire (1984). A layer of basalt in which the fractures are filled with fine sediments may be particularly low in effective permeability.

Hydrology

Surface Water

The Big Lost River is the principal stream within the INEEL and a major source of recharge to the Snake River Plain aquifer. To minimize the likelihood of floods at INEEL facilities, a diversion was constructed on the river in 1958 and enlarged in 1984. During high flows, water can be diverted into four spreading areas designated SAA, SAB, SAC, and SAD (Fig. 1). The estimated total capacity is 2.8×10^7 m^3. Barraclough et al. (1967) estimated characteristic ponded infiltration rates of 2.5×10^{-6} m s^{-1} (0.22 m d^{-1}) for SAA and 9.1×10^{-6} m s^{-1} (0.79 m d^{-1}) for SAB. As Fig. 4 shows, the average annual flow into the spreading areas from 1965 through 1999 was 4.9×10^7 m^3. From 1977 to 1981, the spreading areas received little water, with none at all in 1978 and 1979. There also was no inflow from 1988 to 1994. During the wet years of 1982 through 1985, approximately two-thirds of the Big Lost River flow that entered the INEEL was diverted to the spreading areas (Pittman et al., 1988).

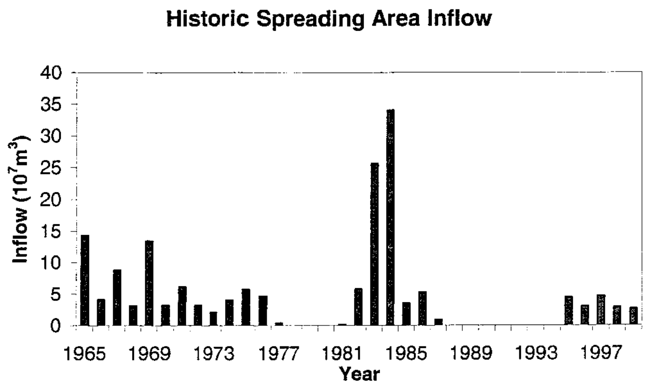


Fig. 4. Total annual inflow to spreading areas, 1965 through 1999. Data are from http://waterdata.usgs.gov/id/nwis, Station 13132513.

Flow and Perching in the Unsaturated Zone

Water movement in the unsaturated zone at this site may be predominantly vertical, but it is likely to be significantly retarded and diverted by features of the basalts and sediments. Several studies (Barraclough et al., 1976; Rightmire and Lewis, 1987b; Anderson and Lewis, 1989) have shown that water episodically accumulates in perched layers that typically persist for a few months. By analysis of water levels in wells and measured water content profiles, Cecil et al. (1991) showed that perching can take place within both sediments and basalts, and suggested that specific mechanisms for this might include (i) contrasts in vertical hydraulic conductivity between basalt flows and sedimentary interbeds; (ii) reduced hydraulic conductivity in baked zones between basalt flows; (iii) reduced vertical hydraulic conductivity in dense, unfractured basalt; and (iv) reduced vertical hydraulic conductivity from sedimentary and authigenic mineral deposits filling fractures in basalt. Additionally, the interfaces between any adjacent layers, for example sandy and silty layers within interbeds, may retard flow under unsaturated conditions (Miller and Gardner, 1962).

Perched water might enable high-permeability features such as fractures and rubble to divert flow horizontally. Such flow would probably parallel the axis of a basalt flow. Near the Subsurface Disposal Area, this direction would be generally northward because these basalt flows erupted from vents on a volcanic rift zone south of the Subsurface Disposal Area. Lateral flow within or adjacent to sedimentary interbeds is likely to move in the direction of dip of the interbeds. The horizontal continuity and dip of some of the interbeds suggests that perched water derived from the spreading areas could move eastward, toward and beyond the Subsurface Disposal Area.

Beneath the Subsurface Disposal Area, some perched water, possibly nearly all of it, may derive from the spreading areas and the Big Lost River rather than from local precipitation. Rightmire and Lewis (1987b), for example, found relatively light abundances of ^{18}O and ^{2}H in perched water beneath the Subsurface Disposal Area. This suggests a source at higher altitude than the Snake River Plain, consistent with the high-

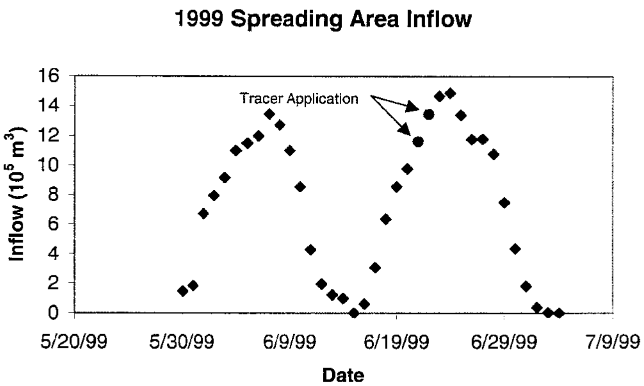


Fig. 5. Inflow to spreading areas, 30 May to 4 July 1999.

altitude streamflow present in the Big Lost River and spreading areas.

Properties and Behavior of the Aquifer

Rubble and fracture zones provide the main conduits for groundwater flow in the saturated zone at the INEEL. Anderson et al. (1999) found the effective hydraulic conductivity to range from about 3×10^{-8} to 8.4×10^{-2} m s^{-1}. They suggested that values less than about 3×10^{-4} m s^{-1} are associated with dikes and thick (>9 m) basalt flows. Greater values are associated with rubble zones and various types of fractures. The prevailing direction of regional flow in the aquifer is toward the southwest.

Fluctuating recharge rates can temporarily alter the flow rate and direction in the aquifer. Barraclough et al. (1976) suggested that periods of high infiltration cause local reversals in water-level gradients near the Subsurface Disposal Area sufficient to move water toward the northeast, against the prevailing direction. They judged infiltration from the spreading areas to be the predominant influence in this process, exceeding the influence of infiltration from local precipitation and the Big Lost River. Rightmire and Lewis (1987a,b) summarized related evidence concerning a fluctuating groundwater mound beneath the spreading areas that alters gradients in the saturated zone. By mapping the rise in water levels that occurred during the unusually wet period between July 1981 and July 1985, Pittman et al. (1988) found further support for this hypothesis.

Expected Behavior of Water from Spreading Areas

Based on available evidence from earlier investigations, and consistent with the understanding of hydrologists knowledgeable about this site, our initial conceptualization of the flow system was as follows (see also Fig. 2). We assumed that when spreading areas become filled, water infiltrates over the course of a few weeks, as long as ponded surface water remains. Initially much of the infiltrating water is taken up by unsaturated-zone pore space, including dead-end fractures. The water percolates downward, continuing for some time after surface water is gone. Vertical flow occurs mainly through highly conductive fractures, taking a few days to reach the water table. Some of the water perches in or on layers of lesser effective vertical conductivity. Most of the water goes down to create a mound, a few meters thick, on top of the aquifer. The mound spreads radially while the spreading areas have standing water, and for a few days or weeks after they empty. The mound dissipates into the aquifer and its water and solutes are carried along with the prevailing Snake River Plain aquifer flow. The perched zones may persist for a few months or longer, until horizontal and vertical flow spreads them enough to leave the porous materials unsaturated.

METHODS

The general plan of the field experiment was to work with expected spring–summer hydrologic behavior in 1999, a year of above-average precipitation. Tracer would be applied to spreading areas while they contained surface water, and samples from perched and aquifer water within several kilometers would be analyzed for tracer. Figure 5 shows the inflow to the spreading areas measured with stream gages during 1999.

Naphthalene Sulfonate Tracers

Naphthalene sulfonate tracers have been successfully tested for tracing in geothermal systems. They are environmentally benign, easily detectable by ultraviolet-fluorescence spectroscopy, and thermally stable. Figure 6 shows various naphthalene sulfonates recently tested in the laboratory and in geothermal fields (Rose et al., 2001). The third column of the figure gives the wavelengths of the peaks of the excitation and emission bands. For this study, we selected 1,5-naphthalene disulfonate as the field tracer.

The adsorption properties of the naphthalene sulfonates in groundwater other than under geothermal conditions have not been reported. Under low-temperature geothermal conditions, however, 1,5-naphthalene disulfonate was shown not to adsorb to negatively charged reservoir rocks or clays (Rose et al., 1999), since the sulfonate groups make these compounds very anionic and soluble in aqueous media. The naphthalene

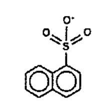
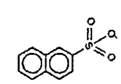
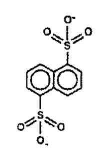
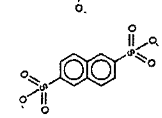
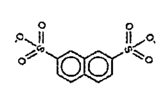
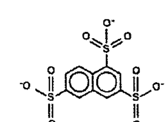
Compound	Structure	Excitation / Emission (nm)
1-naphthalene sulfonate		*217 / 333*
2-naphthalene sulfonate		*220 / 336*
1,5-naphthalene disulfonate		*218 / 334*
2,6-naphthalene disulfonate		*225 / 342*
2,7-naphthalene disulfonate		*226 / 339*
1,3,6-naphthalene trisulfonate		*228 / 342*

Fig. 6. Chemical structures and wavelengths of the excitation–emission spectral peaks of the polyaromatic sulfonates tested in the laboratory and in geothermal reservoirs. The third entry, 1,5-naphthalene disulfonate, was selected for the field experiment presented here.

sulfonates have been shown to be neither mutagenic nor carcinogenic (Greim et al., 1994).

Field Application, Sampling, and Monitoring

The tracer was applied to the spreading areas from a pontoon-type power boat. Fifty-kilogram bags of tracer were partially opened at one end and suspended over the bow of the boat, allowing the material to gradually spill out and the propeller to disperse it as the boat moved forward. The course followed with the boat during this operation was intended to broadly cover the area of ponded water, so that the concentration might be fairly uniform throughout each spreading area. On 21 June 1999, 350 kg of tracer was applied in SAA. The following day, 300 kg was applied in SAB. On 23 June, 83 surface water samples were taken at various locations and depths throughout both spreading areas to assess mixing and approximate concentrations. After the sampling on 23 June, an additional 25 kg of tracer was applied to the northeastern lobe of SAB (Fig. 1), which had filled with water overnight.

A monitoring network of existing wells, both up- and downgradient, was selected to characterize the vertical and horizontal extent of infiltrated spreading-area water. After positive detection of tracer in perched zones, additional wells near the Subsurface Disposal Area were added to the sampling scheme. In December, sampling began at additional Subsurface Disposal Area wells and at monitoring wells (B series and C series wells in Fig. 1) at the location of the Large-Scale Infiltration Test (Wood and Norrell, 1996).

Each well was sampled using a dedicated pump or bailer, or both, following U.S. Geological Survey protocol (Mann, 1996). Extreme precautions were taken in cleaning field equipment to avoid cross-contamination between wells (see Appendix). Samples were collected in clear plastic Nalgene 30-mL bottles, externally rinsed after collection, then labeled and shipped to the analysis lab at the University of Utah. Well conditions were noted in a field logbook.

Initial sampling frequency during June through September 1999 was weekly, biweekly, or monthly, depending on well location, available personnel, and anticipated travel times predicted on the basis of previous studies. After September, sampling was carried out monthly.

RESULTS

Two peaks of inflow to the spreading areas occurred in 1999, on 7 and 24 June (Fig. 5). The total inflow volume for the season was approximately 2.6×10^7 m^3, 40% of which occurred after the introduction of tracer.

Substantial amounts of perched water were found in the unsaturated-zone wells. Those at the Large-Scale Infiltration Test site (south of the Subsurface Disposal Area and east of the spreading areas) were not sampled until 183 d after tracer introduction. Water was encountered in five of the 38 wells that were probed, and water samples were recovered from three of these (C09C11, C09C11A, and B12011). On the following sampling visit, 227 d after tracer introduction, water samples were recovered from all five of the wells with standing water. These five wells were completed in, and above, the base of the BC interbed, which comprises low-permeability silty-clay loam material. Wells that did not yield water were either completed in the basalts above the BC interbed or in the basalts below the BC interbed, so for any water that might cascade in, there was nothing to prevent immediate outflow through basalt fractures. Thus, the presence of standing water in five of the wells is consistent with there being lateral flow in the basalts from the spreading areas.

Measured tracer concentrations in the spreading-area surface-water samples averaged 16.6 μg L^{-1} ($\pm$ 20.6 μg L^{-1} standard deviation, based on 8 samples) in SAA and 180.4 μg L^{-1} ($\pm$ 99.0 μg L^{-1}, based on 18 samples) in SAB. It is likely that concentrations in SAA were lower because of rapid infiltration between the time of application and the surface water sampling, and the fact that the SAA water was sampled 48 rather than 24 h after tracer introduction.

Tracer was detected in nine wells. Figure 7 shows measured concentrations vs. time. In the Snake River Plain aquifer at USGS-120, tracer was detected 9 d after introduction. The preceding sample, taken 6 d after tracer introduction, showed no tracer. The breakthrough curve for USGS-120 has a double peak, as is not uncommon for tracer studies involving preferential flow (e.g., Mohanty et al., 1998). In perched water from basalt immediately above the BC interbed at UZ98-2, tracer was detected 18 d after introduction. It also was detected in perched water from the BC interbed at the nearby well UZ98-1, 65 d after introduction, but in this well the tracer may have been accidentally introduced when the well's casing was temporarily pulled to conduct a cross-hole tomography study, and then reinstalled just before the sample was collected. In the five sampled BC-level wells at the Large-Scale Infiltration Test site, tracer was detected at relatively high concentrations (12–65 μg L^{-1}) in all water samples taken. Limitations of the sampling schedule prevent precise estimates of arrival times at these five wells, but clearly it took less than 183 d to travel this 1.3-km distance.

Directly under the Subsurface Disposal Area in USGS-92, immediately above the CD interbed, tracer was first detected 91 d after introduction. This detection was corroborated by three additional detections on samples taken 122, 147, and 182 d after introduction. Analyses of two archived water samples from this well, from 794 and 83 d before tracer introduction, indicate that 1,5-naphthalene disulfonate was not present in the perched water at this well prior to the start of this test. The pattern of declining concentration in two later detections in USGS-92 (Fig. 7) along with the fact that no samples were taken between the introduction of tracer and the 91-d sampling suggest that tracer may have arrived and peaked at this location in less than 91 d.

Measurements low enough that their difference from zero might not be significant, or where it is not obvious that their measurement uncertainty can be attributed to analytical uncertainty rather than variability in the measured population, may require more formal decision criteria before detections can be confirmed as true positives. Data from USGS-92 and USGS-120 fell into these categories and were evaluated as described in the Appendix.

DISCUSSION

The initial detection of tracer in the aquifer well USGS-120 indicates an average vertical movement of

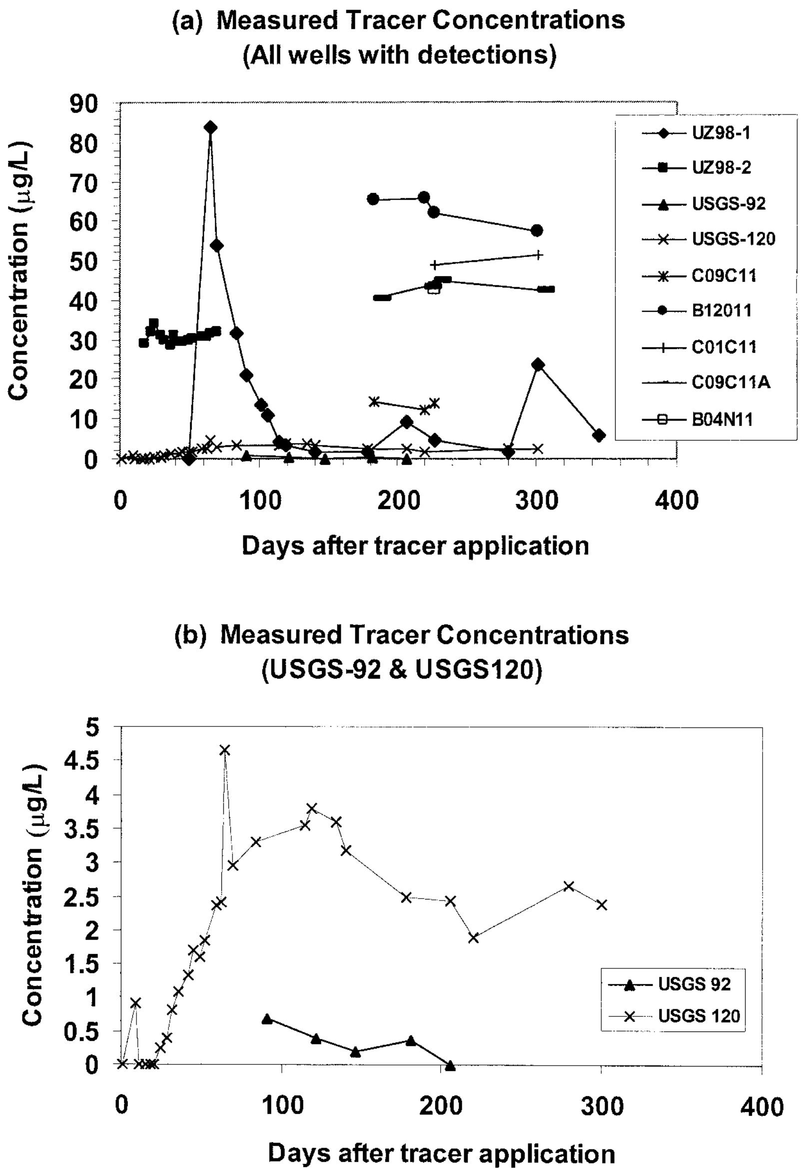


Fig. 7. Measured concentrations in selected wells with positive detections. (a) Concentration vs. time for all nine wells on the same scale. (b) Data for two of the wells on a magnified scale.

at least 22 m d^{-1} at that location. This is about a factor of 4 greater than the average rate through basalt flows A and B in the Large-Scale Infiltration Test (Dunnivant et al., 1998). The measured concentrations as great as 4.7 $\mu g\ L^{-1}$ (Fig. 7) suggest that dilution had been modest, consistent with a large amount of tracer-tagged water going all the way through the unsaturated zone and at least 0.2 km horizontally. This indicates that under ponded infiltration, the sedimentary interbeds and any other layers expected to have low hydraulic conductivity are not an effective barrier to vertical flow.

Several hypotheses may account for the tracer's apparent up-gradient (northeastward) movement over the 0.2 km or more to USGS-120 from the spreading areas. First, significant movement of tracer within the unsaturated zone may have provided an up-gradient source to migrate downward and then back down-gradient to the well. Second, highly localized mounding, as previously hypothesized, may have temporarily reversed the prevailing flow at this location. There are not enough nearby aquifer wells to adequately identify small, localized changes in gradient and flow direction.

The lack of positive detections in the other 12 sampled aquifer wells (Table 1) limits the possible generalizations concerning aquifer flow. These 12 points of nondetection are 0.7 to 2.9 km away from the nearest point of tracer introduction. Two of these, USGS-9 and USGS-109, are close to SAC and SAD, which did not contain surface water during the experiment. From SAB, USGS-9 is 1.5 km down-gradient and USGS-109 is 1.8 km across gradient. The other locations of aquifer nondetection are near the Subsurface Disposal Area, and thus are in the direction opposite the prevailing aquifer flow. Therefore, this experiment does not confirm the hypothesized spreading-area-related reversal of groundwater flow (Barraclough et al., 1976) over

Table 1. Basic location and detection information for the 24 wells sampled for measurement of 1,5-naphthalene disulfonate concentration.

Well name	Completion depth (approximate)	Distance to spreading areas (km to nearest point)	Sampling initiated (days after introduction)	Detection
V10	BC	1.3	176	no
B04N11	BC	1.2	227	yes
B12011	BC	1.2	183	yes
C01C11	BC	1.3	227	yes
C09C11	BC	1.3	183	yes
C09C11A	BC	1.3	183	yes
UZ98-1	BC	0.2	2	yes
UZ98-2	BC	0.2	18	yes
USGS-92	CD	1.3	91	yes
8802-D	CD	1.2	176	no
I-3	CD	1.5	176	no
USGS-109	Aquifer	1.8	1	no
USGS-9	Aquifer	1.5	2	no
USGS-86	Aquifer	2.9	16	no
USGS-87	Aquifer	1.7	1	no
USGS-88	Aquifer	0.7	2	no
USGS-89	Aquifer	1.1	1	no
USGS-117	Aquifer	1.1	1	no
USGS-119	Aquifer	1.3	1	no
USGS-120	Aquifer	0.2	2	yes
RWMC prod.	Aquifer	1.7	23	no
RWMC m7s	Aquifer	2.6	21	no
RWMC m1sa	Aquifer	1.0	22	no
RWMC m3s	Aquifer	1.8	22	no

distances >0.2 km. However, the possibility of considerable and rapid dilution of spreading area–derived water within the Snake River Plain aquifer makes for a significant probability that any tracer-tagged water that had traveled 0.7 km or more through the aquifer was diluted to below detection limits. Therefore the observed pattern of aquifer nondetections at horizontal distances >0.2 km does not refute the flow-reversal hypothesis.

At the BC interbed level, positive detections in seven of eight sampled wells confirm that substantial infiltrated spreading-area water is diverted horizontally by this interbed or flow paths within it or above it. Different temporal patterns of tracer concentration between the two nearby BC wells (UZ98-1 and -2, 0.2 km from SAB, Fig. 7) may be influenced in part by the casing replacement in UZ98-1 noted above. Detections in the Large-Scale Infiltration Test wells, 1.3 km east, show that the perched water can move faster than 10 m d^{-1} over considerable distances. There is relatively little dilution here. The rapid lateral flow and minimal dilution suggest a consistently low permeability of the BC interbed, and high permeability of a layer above it, between the spreading areas and the Large-Scale Infiltration Test. The easterly dip of this interbed may also be significant. Of the eight sampled BC-perched wells, the only one that has no detection is V-10. This well may be subject to much different hydrogeologic influences, since it is the only one of these wells that is distant from SAB (1.3 km) in a direction other than east. From SAA, however, its direction is eastward (2.0 km). Plausible reasons for lack of a detection here include (i) an absence of suitable flow paths in this direction, (ii) the dip of the BC interbed not having the required consistency or flow direction, (iii) a discontinuity in the BC interbed that allows perched water to percolate readily into basalt unit C between the spreading areas and the Subsurface Disposal Area, (iv) dilution to below detection limits with perched water from other sources, and (v) travel times shorter or longer than the samplings conducted. The experiment provides no confirmation that BC-perched water travels from spreading areas to the Subsurface Disposal Area. It does demonstrate that at this level in at least one direction, substantial rapid flow occurs to distances of at least 1.3 km.

At the CD interbed level, tracer was detected from one of the three sampled wells, USGS-92, directly under the Subsurface Disposal Area 1.3 km northeast of SAB and 2.2 km east of SAA. The detected concentrations (0.7 $\mu g\ L^{-1}$ and less) are low but significant (see Appendix). Because they declined after the first detection, the concentration may have peaked at this point in <91 d. They suggest an average horizontal flow rate of at least 14 m d^{-1}. This detection may indicate an isolated case of flow through a single channel. The experiment cannot rule out more widespread horizontal spreading at this depth, however, because so few CD wells were available for sampling. The other two CD wells, also under the Subsurface Disposal Area, may also have contained tracer before the sampling of these wells began or at undetectable concentrations. The results from USGS-92 demonstrate that perched water beneath the Subsurface Disposal Area comes in part from subsurface flow of spreading-area water.

Of possible driving forces for flow, if conditions are nearly saturated, gravity would be expected to dominate. Besides the average easterly dip noted above, the pattern of observed unsaturated-zone detections is consistent with the known elevation of interbed contacts at various points (Anderson et al., 1996). The average dip of the interbeds gives a modest driving force, being a component 0.004 times vertical gravity. Thus, if the component of gravity parallel to the interbeds is the chief driving force, the observed long-range horizontal transport suggests an overwhelming effective anisotropy in unsaturated-zone properties.

It is unlikely that horizontal convective transport rates >10 m d^{-1} in the unsaturated zone occurred directly through the fine-textured portions of the interbeds because of the low saturated hydraulic conductivity of these materials (ranging from 10^{-7} to 10^{-4} cm s^{-1}, Perkins and Nimmo, 2000). It is more likely that the interbeds caused perching that extended upward into some portion of the overlying basalt. Rapid transport may be possible through interconnected rubble zones, tension cracks, or similar features if they are nearly saturated. There must also be substantial impediments to vertical flow, such as interbeds or dense basalt that cause perching that extends upward into the highly conductive layer or flow path. Given the observed lengths of individual basalt flows and volcanic rift zones, such horizontal movement of water over distances of a kilometer or more is plausible.

This evidence as a whole supports several particular hydrologic phenomena.

1. There can be rapid vertical transport under the spreading areas.
2. Substantial perching of water in the unsaturated zone results directly from the percolation of water from the spreading areas.
3. Over distances of a few kilometers and durations of a few months, horizontal transport occurs in perched water in or above both the BC and CD interbeds.
4. The horizontal spreading in perched zones is not uniform with direction.

These hydrologic phenomena suggest certain subsurface features that determine the character of flow. Sedimentary interbeds and impeding features of the basalts are imperfect barriers that permit substantial vertical flow, possibly because of interbed discontinuities and basalt fractures. This is consistent with earlier evidence showing that the interbeds impede vertical flow where they are continuous, for example, the Large-Scale Infiltration Test (Wood and Norrell, 1996c; Dunnivant et al., 1998). In spite of existing vertical fast flow mechanisms, at least some of the flow-impeding features are sufficiently effective and continuous that they divert substantial flow horizontally for considerable distances. Nonuniformities in both basalt and interbed topography are likely causes of the irregular spatial patterns of tracer detection.

The long-range horizontal flow inferred from unsaturated-zone tracer detections requires alteration of previous conceptualizations of subsurface contaminant transport that did not include this sort of flow. Horizontal movement by itself does not necessarily imply a greater likelihood that water-borne contaminants will be transported to the aquifer. However, it does put more water under the Subsurface Disposal Area than would occur without spreading-area input. This water may facilitate contaminant transport to the Snake River Plain aquifer by intercepting water-borne contaminants that would otherwise percolate into and be absorbed by the underlying sedimentary interbeds; it may also transport them downward through preferential flow paths that are active because of the greater amount of water present. The horizontal flow is likely to dilute contaminants and disperse them over wider regions of the unsaturated zone. This could provide greater opportunity for sorption, especially within the sedimentary interbeds, by reducing competition for sorption sites, increasing the availability of sorption sites, and changing solution chemistry to favor sorption. Therefore some effects of the inferred horizontal and nonuniform flow are likely to enhance and others to hinder the transport of contaminants to the aquifer.

Similar behavior concerning fast, long-range flow both horizontally and vertically in the unsaturated zone may occur at other locations with the necessary site characteristics. The INEEL tracer detections were at depths of 30 m and more, suggesting emphasis on arid and semiarid areas where the unsaturated zone is deep. It is important that the unsaturated zone be highly stratified, which is common, but usually not to the obvious extreme of the Snake River Plain. The unsaturated zone must have preferential flow paths such as fractures or coarse granular material, also common at many sites. Long-range horizontal continuity of individual preferential paths and impeding layers has importance that is hard to assess; such continuity could exist at the INEEL, though if essential, the number of such paths must be strikingly large to permit the multiple BC-level tracer detections (Fig. 1). Ponded water is needed to provide a large volume of inflow to the unsaturated zone, and episodic occurrence is important so that conditions are normally unsaturated. Features that might play this role, for example, ephemeral streams and ponds, playas, and artificial recharge ponds, are fairly common on plains and valleys, though less so in mountains.

Where all the relevant features are present in some degree, this sort of rapid, long-range flow might be expected. Alluvial valleys (e.g., Mojave desert, basins of basin and range topography) have many of these characteristics, though commonly with an unsaturated zone that contains only granular material. Horizontal flow is most likely where some of the granular materials are coarse, or underlain by fractured rock. Several sites of major contaminant transport concerns are of interest. At the Hanford Site in Washington, much of the unsaturated zone has depth in the 60- to 90-m range. It has less pronounced layer contrasts than the INEEL site and no significant fractured rock in the unsaturated zone, though there are strong textural contrasts of silt to gravel size, and caliche. These differences may not be very significant for transport rates. The Nevada Test Site has an extremely deep unsaturated zone in many places, and more diversity in layers than the Hanford site. Subsurface behavior there might easily be comparable to that found at INEEL, though ponded water would be more unusual. Yucca Mountain likewise has an extremely deep unsaturated zone and clearly lacks a major source of ponded infiltration. Its stratigraphy is analogous in many ways to the Snake River Plain. For example, the layer referred to as "paintbrush tuff" is less fractured than its surrounding layers and so has been hypothesized to play a role similar to that hypothesized for the INEEL interbeds in interrupting preferential flow and inhibiting downward solute transport. In both cases, these hypotheses may be invalid; evidence for such effects in paintbrush tuff is lacking (Flint et al., 2001), while our study shows that preferential flow can move downward through or past INEEL interbeds. Our experiment suggests that even if the impeding layer can allow substantial downward movement, it does not prevent long-range horizontal flow.

At smaller scales, similar fast-flow behavior may occur in unsaturated zones a few meters thick, as commonly exist in humid as well as arid climates. Small-scale stratification and preferential paths commonly exist, so it is entirely possible to have analogous phenomena over the shorter distances. Nimmo (2002) found that field data from a wide variety of field sites do show similar behavior and have transport speeds similar to those in this study.

CONCLUSIONS

A naphthalene sulfonate tracer detectable at concentrations down to about 0.2 μg L^{-1} has proven to be stable and conservative in the subsurface to the degree necessary for large-scale saturated- and unsaturated-zone investigations. With introduction, sampling, and analysis techniques as applied in this study, it can trace water movement over a spatial scale of at least 1.3 km and a time scale of at least 1 yr.

Besides testing the chosen combination of tracer and techniques, this study was to assess long-range subsurface flow paths that might result from episodic ponded infiltration. The geologic nature, hydraulic properties, spatial distribution, source localities, and other characteristics of these flow paths remain of great interest and are poorly known. Results of the present experiment suggest several improvements for field experiments to explore these characteristics and processes further. The observed long distances and high speeds of travel suggest an extensive network of sampling wells, and a schedule that has even the most distant of these sampled early in the experiment. Multiple tracers, for example other naphthalene sulfonates (Fig. 6), are desirable for different portions of the surface water. With three tracers one could distinguish water from individual spreading areas A and B, and the Big Lost River. This would answer questions such as whether tracer in USGS 92 came from spreading area A or B. Repeat sampling of the tracer-containing surface water would give a better estimate of the initial concentration, for evaluation of attenuation and dilution. Besides offering a better understanding of subsurface preferential flow paths and their effect on solute transport, an improved study of this sort would allow the testing of important hypotheses concerning the permeability, continuity, and dip of interbeds, the role of rubble and fractures, and the partitioning of vertical and horizontal flow in the unsaturated zone.

This experiment shows that with a large input of water, the overall structure of the unsaturated zone of the Snake River Plain can generate rapid transport both vertically and horizontally. At least under ponded infiltration, fine-textured interbeds and layers of particularly dense basalt do not prevent the rapid, high-volume, vertical flow that is expected through fractured basalt. Some impediments to vertical flow, however, are effective in causing substantial perching and horizontal diversion. The observed rapid horizontal flow can persist over distances >1 km, entirely within the unsaturated zone, 100 m and more above the aquifer. Possible conduits for this horizontal flow include basalt fractures, tension cracks, lava tubes, or rubble zones positioned above low-permeability layers of basalt or interbeds. These observed behaviors are likely to enhance the vertical and horizontal spreading of contaminants in the subsurface, though by exposing the contaminants to a greater volume of sedimentary materials they may also have effects that tend to reduce aquifer contamination. Because the rapid vertical and horizontal transport in the unsaturated zone seems to be caused by natural features responding to a common sort of ponded infiltration, these results suggest that similar phenomena may occur at a variety of sites.

APPENDIX

Quality Assurance

Samples were collected and analyzed according to the guidelines of the Quality-Assurance Program at the USGS INEEL Project Office, which includes: (i) analytical methods used by the laboratories, (ii) quality-control samples, (iii) review of analytical results, (iv) audits of field performance, (v) corrective actions to resolve problems with field and laboratory methods, and (vi) reporting of data. A chain-of-custody record tracked samples from collection to delivery.

Wells equipped with dedicated submersible pumps require decontamination of detachable sampling equipment. At the wellhead, a stainless-steel discharge pipe is equipped with a sample port and a gate valve to control the flow rate. The sampling equipment, discharge pipe, and fittings were rinsed with deionized water before and after installation at the wellhead. Subsequent flushing with purged water in amounts of about 2 to 30 m^3 further reduced the possibility of cross-contamination with water from previously sampled wells. Additionally, before each sampling, at least three well-bore volumes of water were pumped from the well to remove any stagnant water. This purged water was disposed of to the ground. This is not expected to influence the subsurface movement of tracer because the amount of water was small (on the order of 10^3 m^3) and was distributed over a substantial area of vegetated land under high-evapotranspiration conditions. For wells in which a bailer was used to collect a water sample, careful decontamination procedures were followed to prevent cross-contamination between well bores by rinsing with deionized water and wiping with lab tissues. Bailed samples were poured directly from the bailer into sample bottles after three consecutive rinses of the bailer with sample water. After sampling, the bailer was rinsed thoroughly with several liters of deionized water and placed in a clean plastic bag to prevent possible contamination during transport.

The quality-assurance program includes archival retention of replicate samples for evaluation of erroneous results, occasional submission for analysis of blind replicates (duplicate samples with different sample identification numbers), blank samples (deionized water labeled as a regular sample), equipment blanks (rinsate collected during decontamination procedures), and spiked samples (of known concentration). In general, about 15 to 20% of the samples that were analyzed for tracer concentration were dedicated to quality assurance.

This study included determinations of the analytical preci-

Table 2. Results of analytical precision estimates using high performance liquid chromatography to measure concentrations of 1,5-naphthalene disulfonate. $\bar{x}$ is measured mean concentration, $\bar{s}$ is standard deviation, $\bar{s}/\bar{x}$ is relative error expressed as a percentage and *n* is the number of replicate determinations.

Nominal concentration†	0.1	0.2	0.3	0.4	0.5	1.0	5.0	10.0	50.0	100.0
$\bar{x}$	0.15	0.24	0.35	0.46	0.57	1.20	5.32	10.74	52.45	106.1
s	0.07	0.06	0.08	0.08	0.06	0.14	0.13	0.13	0.45	0.79
$\bar{s}/\bar{x}$	47	25	23	17	10	12	2.4	1.2	0.8	0.7
n	8	8	8	8	8	8	8	8	8	8

† Concentrations are in micrograms per liter.

sion or uncertainty associated with the measurement of 1,5-naphthalene disulfonate concentrations using the high performance liquid chromatograph with reverse-phase column separation. The column was a reverse phase 50 by 4.6 mm Keystone BetaBasic-18 with 3-μm particle size (Thermo Hypersil-Keystone, Bellefonte, PA).[1] The mobile phase consisted of a solution of 3.17 mM Na_2HPO_4, 6.21 mM KH_2PO_4, and 5.0 mM TBAP in 30:70% methanol/water. Test standards, consisting of nominal solution concentrations ranging from 0.1 to 100.0 μg L^{-1}, were prepared from the industrial grade tracer that was used in the field experiment.

Table 2 and Fig. 8 present the measured results for determination of the analytical precision or uncertainty. The absolute error of a measurement, taken as the standard deviation ($\bar{s}$) of eight replicate measurements of the same laboratory-grade standard, is affected primarily by the gain or sensitivity setting of the instruments. Between 0.1 and 0.5 μg L^{-1} it is essentially constant at about 0.07 μg L^{-1}. Between 1 and 10 μg L^{-1} it also is essentially constant, at about 0.13 μg L^{-1}. At higher ranges, higher gain settings result in higher absolute errors, as shown for nominal tracer concentrations of 50 and 100 μg L^{-1}. To indicate relative error, absolute error estimates are normalized ($\bar{s}/\bar{x}$) with respect to the measured means. Figure 8 shows that in the range of 0.1 to 100 μg L^{-1}, the log of the relative error is nearly linear with respect to the log of measured concentrations, and decreases as the concentration increases. At 0.15 μg L^{-1}, the lowest measured concentration, the relative error is 47% of the measured mean.

USGS-92 had the lowest of all nonzero measured concentrations in the study, and required assessment of whether the measurements were repeatable and not the result of vagaries of analytical uncertainty. Table 3 gives detailed sampling and analysis data for USGS-92, including sample splits and replicates. Five splits of an archived duplicate sample taken 122 d after introduction produced a mean value that was consistent with the value derived from a single measurement of the original sample. The repeatability, expressed as standard deviation, of the five splits (± 0.07 μg L^{-1}) is within the analytical uncertainty for measured mean concentrations near 0.4 μg L^{-1} (± 0.08 μg L^{-1}). Twenty replicate determinations of the originally analyzed sample from 122 d after introduction were run and the results (± 0.05 μg L^{-1}) indicate that the measurement uncertainty can be attributed to analytical error. Similar conclusions can be drawn for multiple independent samples taken on 13 Jan. and 16 Feb. 2000 as shown in Table 3. These results indicate that tracer detections at USGS-92 are repeatable and that most of the uncertainty in the measured concentrations can be attributed to limitations in analytical precision.

For aquifer samples from USGS-120, it needs to be established whether the positive detections are true indications of transport within the aquifer and not the result of unsaturated-zone water moving vertically downward along the annular space between the borehole casing and the surrounding wall-rock. The initial detection of tracer 9 d after introduction was followed by four sampling sessions that showed no evidence of tracer over the period of 11 to 22 d after introduction. Detections in UZ98-2, 20 m from USGS-120 and completed above the BC interbed, show that tracer was present in the unsaturated zone at this location. To test the possibility that tracer may have been carried down the side of the borehole instead of flowing through the aquifer, a 1.3 L s^{-1} pump-discharge test was conducted 84 d after introduction of tracer, with 15 independent samples taken during the 45-min duration of the test. If the tracer is from the aquifer, its concentration should be relatively constant with time, indicative of widespread dispersion before the test. If derived from annular leakage, its concentration should be highly variable, indicative of local dispersion near the borehole. Results of this test (Table 4) showed the variability of the mean (± 0.34 μg L^{-1}) to be greater than twice the analytical uncertainty (± 0.13 μg L^{-1}) for tracer concentrations between 1 and 5 μg L^{-1}. Although the multiple-sample analysis of variability was high, it was not sufficient to establish that annular leakage was occurring. A second test, lasting 24 h, was run starting 141 d after introduction. The pump discharge rate during this test was also 1.3 L s^{-1}. Nineteen independent samples were taken at various times during this test. Results indicate that the measurement uncertainty (± 0.15 μg L^{-1}) was within the analytical uncertainty (± 0.13 μg L^{-1}) for the measured concentration and was consistent with expectations for a homogeneous population. Thus, we conclude that the detection of tracer in the aquifer did not result from the transport of unsaturated-zone water in the annular space around the borehole casing, and therefore indicates a migration of tracer to the aquifer through naturally existing paths.

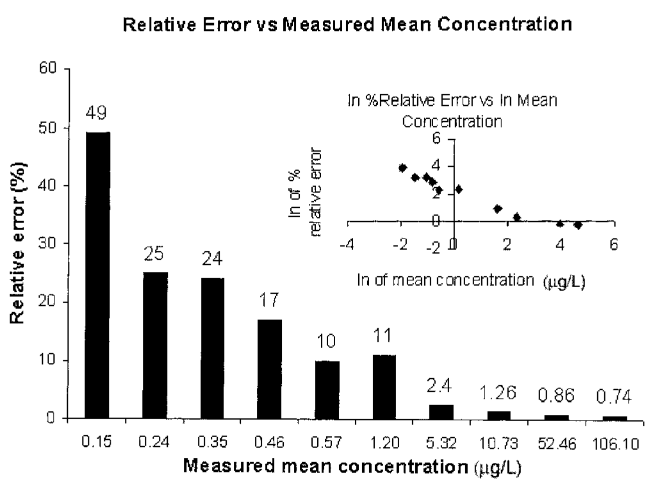


Fig. 8. Relative errors determined in measurements of analytical precision.

[1] Product names are given only to identify the equipment used, and do not imply endorsement by the USGS.

Table 3. Sampling data for USGS-92: concentrations of 1,5-nephthalene disulfonate in micrograms per liter. Day 0 is 21 June 1999, *x* is concentration from a single measurement; *n* is the number of independent samples, splits, or replicate determinations; and other symbols are as defined for Table 2.

Date	Day	x	$\bar{x}$	$\bar{s}$	n	$\bar{s}/\bar{x}$	Remarks
18 Apr. 1997	−794	0.0					Pretest archive sample
30 Mar. 1999	−83	0.0					Pretest archive sample
20 Sept. 1999	91	0.69					
21 Oct. 1999	122	0.40	0.39	0.07	5	18	5 splits of archived sample from 21 Oct. 1999
			0.40	0.05	20	12.5	20 replicate determinations of original sample on 21 Oct. 1999
15 Nov. 1999	147	0.21	0.21	0.02	5	9.5	5 replicate determinations
20 Dec. 1999	182	0.37					
13 Jan. 1999	206	0.42					3 independent samples
		0.33					
		0.30					
16 Feb. 2000	240	0.21					2 independent samples
		0.21					
17 Apr. 2000	301	0.0					

Table 4. Selected sampling data for USGS 120. Symbols are as defined for Table 3.

Date	Day	$\bar{x}$	$\bar{s}$	n	$\bar{s}/\bar{x}$	Remarks
13 Sept. 1999	84	3.29	0.34	15	10.3	45-min discharge test. 15 independent samples
		3.26	0.07	20	2.1	20 replicate determinations of Sample 13 from 13 Sept. 1999
9–10 Nov. 1999	141	3.18	0.15	19	4.7	24-h discharge test. 19 independent samples

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