

Groundwater "fast paths" in the Snake River Plain aquifer: Radiogenic isotope ratios as natural groundwater tracers

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ABSTRACT

Preferential flow paths are expected in many groundwater systems and must be located because they can greatly affect contaminant transport. The fundamental characteristics of radiogenic isotope ratios in chemically evolving waters make them highly effective as preferential flow path indicators. These ratios tend to be more easily interpreted than solute-concentration data because their response to water-rock interaction is less complex. We demonstrate this approach with groundwater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the Snake River Plain aquifer within and near the Idaho National Engineering and Environmental Laboratory. These data reveal slow-flow zones as lower $^{87}\text{Sr}/^{86}\text{Sr}$ areas created by prolonged interaction with the host basalts and a relatively fast flowing zone as a high $^{87}\text{Sr}/^{86}\text{Sr}$ area.

Keywords: groundwater tracers, isotope ratios, Snake River Plain, aquifer properties, aqueous solutions.

INTRODUCTION

Groundwater flow is usually focused into preferential flow pathways in fractured or highly heterogeneous aquifers (e.g., Tsang and Neretnieks, 1998). These "fast paths" pose a critical problem, because they can transmit contaminants rapidly yet may arise from subtle or hidden natural features such as zones of increased fracture density or connectivity. Detection of fast paths can be attempted through observation of anthropogenic and natural geochemical tracers. Anthropogenic tracers such as tritium and chlorofluorocarbons are effective for tracking groundwater flow when residence times are less than ~50 yr, but additional tools are needed for systems containing older groundwater. The natural chemistry of groundwater is rich in information, but new approaches to interpret the various solute concentration and isotopic data must be developed to fully exploit them.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in groundwater reflect the water-rock reaction histories and flow pathways of the waters. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of rocks and soils vary because ^{87}Sr is produced by radioactive decay of ^{87}Rb . Groundwater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are inherited from the soil or rock through which the water passes, and several studies have used $^{87}\text{Sr}/^{86}\text{Sr}$ as a natural tracer of groundwater flow

and weathering (e.g., Blum et al., 1993; Bullen et al., 1997; Johnson and DePaolo, 1994; Musgrove and Banner, 1993). As water-rock interaction progresses, $^{87}\text{Sr}/^{86}\text{Sr}$ in groundwater evolves toward the ratio of Sr acquired from the host soil or rock. In this paper we demonstrate the usefulness of $^{87}\text{Sr}/^{86}\text{Sr}$ data in imaging preferential flow

paths by using data from the Snake River Plain aquifer in and near the Idaho National Engineering and Environmental Laboratory (INEEL).

HYDROGEOLOGIC SETTING AND METHODS

The INEEL is located above the Snake River Plain aquifer of Idaho (Fig. 1). Limited, localized contamination at INEEL has motivated groundwater monitoring and protection measures (Barraclough et al., 1982; Mann and Cecil, 1990). Regional flow in the aquifer is from northeast to southwest. Groundwater enters the aquifer predominantly from the Snake River to the southeast, the Yellowstone Plateau to the northeast, and tributary groundwater systems from several valleys extending to the north. Among these are the Little Lost River and Birch Creek valleys, which contribute a large flux of water to the aquifer along the northern and northwestern

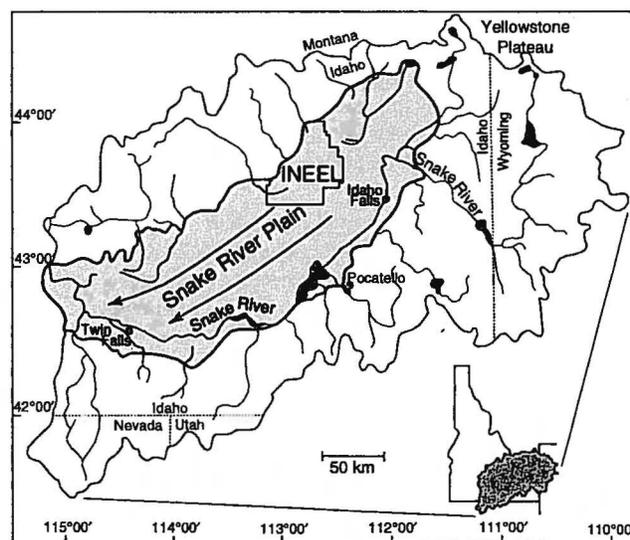


Figure 1. Snake River Plain aquifer and watersheds that recharge it. Approximate regional groundwater flow direction in aquifer is given by arrows. INEEL—Idaho National Engineering and Environmental Laboratory.

Data Repository item 200093 contains additional material related to this article.

edges of the INEEL. Recharge within the plain is minor compared to these inputs (Barraclough et al., 1982). An agricultural region that taps the aquifer for irrigation water is located southwest (down gradient) of the INEEL.

The aquifer rock is composed of tongue-shaped basaltic lavas, separated in some cases by sheets of sediments (Barraclough et al., 1982). Water flows readily through fractures, rubble zones, and other features such as collapsed lava tubes. Groundwater flow is rapid (e.g., 300 m/yr pore velocity) in the upper 100–400 m of the aquifer and is much slower in deeper layers (Blackwell et al., 1992; Mann, 1986). The network of fractures is highly heterogeneous, and lava flow trends likely lead to hydraulic anisotropy (Welhan and Reed, 1997). In fractured, heterogeneous systems such as this, preferential flow zones with relatively high flow velocities commonly occur (Tsang and Neretnieks, 1998).

Regional groundwater-flow simulations have been developed for the aquifer (Ackerman, 1995; Garabedian, 1992), but these studies sought to reveal broad regional flow patterns and did not concentrate on finding fast-flow paths at the 50 km scale as we do here. Studies tracking contaminant plumes, chlorofluorocarbon compounds, and/or anthropogenic radionuclides reveal flow directions and velocities (Barraclough et al., 1982; Beasley et al., 1993; Busenberg et al., 1993; Cecil et al., 2000). However, these relatively recent anthropogenic tracers are tracked for relatively short distances and do not reveal regional flow patterns. The natural groundwater chemistry reported in this study provides regional flow information and thus complements smaller scale studies using anthropogenic tracers.

We measured $^{87}\text{Sr}/^{86}\text{Sr}$ and concentrations of 14 elements (Li, B, Na, Mg, Al, Si, K, Ca, Mn, Fe, Rb, Sr, Ba, and U) in groundwater samples collected in and near the INEEL.¹ We acquired 55 water samples from purged wells; the samples were filtered and preserved with HNO_3 . Sr was purified from the waters via Sr-specific ion-exchange resin (Eichrom Industries). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were measured with a VG Sector 54 thermal ionization mass spectrometer and a dynamic multiple-collector routine to a precision of about $\pm 0.002\%$. Results for the NBS-987 standard during this study were 0.710275 ± 0.000013 (2σ). Dissolved concentrations were measured by inductively coupled plasma-mass spectrometry (Perkin-Elmer ELAN 6000). Calibration drift was corrected for via an internal standard containing Be, Ge, and Tl and periodic analysis of standards. Duplicate analyses of several samples indicate a 2σ uncertainty of $\pm 5\%$ or better.

¹GSA Data Repository item 200093, Groundwater chemistry data, is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301-9140, editing@geosociety.org, or at www.geosociety.org/pubs/ft2000.htm.

RESULTS AND DISCUSSION

Contour plots of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and selected elemental concentrations are given in Figure 2. The pattern exhibited by the $^{87}\text{Sr}/^{86}\text{Sr}$ data is striking. Water entering the aquifer from the Birch Creek and Little Lost River valleys to the north has high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (>0.71100) relative to waters originating east of the INEEL; the high ratios are inherited from Paleozoic sediments to the north. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios decrease along all possible southwestward flow paths, but in some areas they decrease strongly, whereas in other areas they decrease less. Particularly well defined is a relatively narrow zone of high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (the high isotope-ratio zone) that extends southward from the Little Lost River valley through the southern boundary of the INEEL. Zones with relatively low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios occur near the center of the INEEL (central low isotope-ratio zone) and in the western part of the INEEL (western low isotope-ratio zone). Visually, the $^{87}\text{Sr}/^{86}\text{Sr}$ patterns in Figure 2A suggest a channeling of the high-ratio northern groundwater through preferential flow zones between and around the western and central low isotope-ratio zones. In this paper we develop this hypothesis in light of hydrologic features of the aquifer, including flow rates and directions, mixing of chemically distinct water masses, and chemical interaction with the host rock.

One process that controls groundwater chemistry in this area is regional mixing of contrasting water masses (McLing, 1994). Mixing between the northern water masses entering from the Little Lost River valley and Birch Creek valley and waters originating east of the INEEL results in a northwest to southeast gradient in water chemistry. This gradient is apparent in all solute-concentration and isotope-ratio plots, particularly in the Li data (Fig. 2B). Mixing thus plays a role in defining the $^{87}\text{Sr}/^{86}\text{Sr}$ patterns, but the shapes and locations of the high and low isotope-ratio zones cannot be produced by this process alone. The narrowness of the high isotope-ratio zone cannot be explained by regional mixing; the water entering from the Little Lost River valley would spread out into a relatively wide zone were it not channeled by hydrologic heterogeneity.

Preferential Flow Model

Channeling of groundwater flow through the high isotope-ratio zone and relatively slow flow in the western and central low isotope-ratio zones can readily produce the observed isotope-ratio pattern. In this scenario, the high isotope-ratio zone is a fast-flow zone, where high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios originating in the Little Lost River valley and the Birch Creek valley persist far to the south because the water has relatively brief contact with the host rock. In the low $^{87}\text{Sr}/^{86}\text{Sr}$ zones, the same groundwater evolves closer to the isotopic composition of the host rock ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7070 \pm 0.0003$; Leeman and Manton, 1971;

Reed et al., 1997) because of slower groundwater flow, prolonged contact with the rock, and greater rock dissolution.

The reaction rate required to produce the observed groundwater $^{87}\text{Sr}/^{86}\text{Sr}$ evolution is not unreasonable. The groundwater pore velocity is 300 m/yr in the high isotope-ratio zone, according to contaminant migration data (Barraclough et al., 1982). The $^{87}\text{Sr}/^{86}\text{Sr}$ evolution from 0.711 to 0.705 occurs over 50 km distance there. Using an effective porosity of 20%, Sr concentrations of 300 ppm and 0.2 mg/L in the rock and water, respectively, and published mass-balance equations (Johnson and DePaolo, 1994), we calculate that 5% of the rock's Sr must be transferred to the fluid per million years. The basalt host rocks are 1 Ma or younger (Lanphere et al., 1993) and contain an unstable glass phase, so rapid reactions are expected. In addition, some of the decrease in $^{87}\text{Sr}/^{86}\text{Sr}$ within the high isotope-ratio zone is caused by mixing with lower $^{87}\text{Sr}/^{86}\text{Sr}$ water from outside the zone and thus the actual reaction rate is probably lower than this estimate.

A concurrent study has measured $^{234}\text{U}/^{238}\text{U}$ ratios in the same groundwater samples (Roback et al., 1997). The spatial pattern in the data is very similar to that of the $^{87}\text{Sr}/^{86}\text{Sr}$ data. This is especially significant because the systematics of U isotopes are different from those of Sr isotopes. Detailed examination of these data is beyond the scope of this paper, but the similarity of the U and Sr isotope results suggests that radiogenic isotope ratios in general are effective groundwater tracers.

Alternative Interpretations

Several alternative interpretations of the data were considered. The first invokes influxes of low $^{87}\text{Sr}/^{86}\text{Sr}$ water from mountains bordering the plain or below the surficial aquifer to form the western and central low isotope-ratio zones, without slower groundwater flow in these zones. However, the flux of water from these sources must be small, because the mountain-front watersheds that discharge directly to the plain are small ($<30\text{ km}^2$) and because permeability is low below the surficial aquifer (Mann, 1986). Neither source could produce a water flux of similar magnitude to that from the Little Lost River valley, which drains an area of $\sim 1000\text{ km}^2$. Thus, the fluxes of water needed to create the western and central low isotope-ratio zones in this scenario do not exist. Furthermore, rocks in the mountain-front watersheds are similar to those in watersheds feeding into the Little Lost River valley, and would likely produce similar waters ($^{87}\text{Sr}/^{86}\text{Sr} \approx 0.711$).

Small influxes of solute-rich hydrothermal water, for which there is some geochemical evidence (McLing, 1994) are another potential source of low $^{87}\text{Sr}/^{86}\text{Sr}$ solute. The Sr concentration of this water would necessarily be very high if it were to affect the $^{87}\text{Sr}/^{86}\text{Sr}$ of the fast-flowing surficial aquifer, and thus influx of this water would elevate

groundwater Sr concentrations in the western and central low isotope-ratio zones. In reality, concentrations there are lower than in the high isotope-ratio zone, so this not a viable scenario.

Water infiltrating the aquifer from the Big Lost River channel is a significant water input, mostly near the INEEL spreading areas (Fig. 2F) and near the channel's end in the northern area of the INEEL (Bennett, 1990). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of a Big Lost River water sample is 0.71056; this value is lower than the Little Lost River valley inputs (presumably because of differences in source area geology) but is higher than the observed values in the western and central low isotope-ratio zones. Thus, the Big Lost River input cannot create the low $^{87}\text{Sr}/^{86}\text{Sr}$ zones, though it may influence $^{87}\text{Sr}/^{86}\text{Sr}$ values in some areas.

Another alternative hypothesis invokes interaction of groundwater with chemically heterogeneous host rock, without fast- and slow-flow zones, as the cause of the observed pattern. However, essentially all of the aquifer rocks in this area are homogeneous basalts with a restricted $^{87}\text{Sr}/^{86}\text{Sr}$ range (Leeman and Manton, 1971; Reed et al., 1997), and lava groups are continuous between the high isotope-ratio zone and the western and central low isotope-ratio zones (Anderson, 1991).

The preceding scenarios consider the likely alternative hypotheses, but none of them appears viable. We thus conclude that the $^{87}\text{Sr}/^{86}\text{Sr}$ pattern reflects the presence of fast- and slow-flow zones. In addition, the apparent direction of flow in the high isotope-ratio zone is nearly due south; this is consistent with a more local flow direction revealed by a contaminant plume near the center of the zone (Barraclough et al., 1982). Flow is not parallel to the regional hydraulic gradient as given in Barraclough et al. (1982). This divergence implies hydraulic anisotropy, which is consistent with the dominant northwest trend of the lava flows and recent research on anisotropy (Welhan and Reed, 1997).

Radiogenic Isotopes Versus Concentration Data

Because solute concentrations are sensitive to water-rock interaction, they might be expected to provide similar imaging of fast paths. Our concentration data (Fig. 2) exhibit spatial patterns with some similarities to the $^{87}\text{Sr}/^{86}\text{Sr}$ data. A tongue of high Mg concentration and low Na and Si concentrations weakly resembles the high isotope-ratio zone. In addition, an area of high Na and Si concentrations in the western low isotope-ratio zone apparently reflects net release of Na

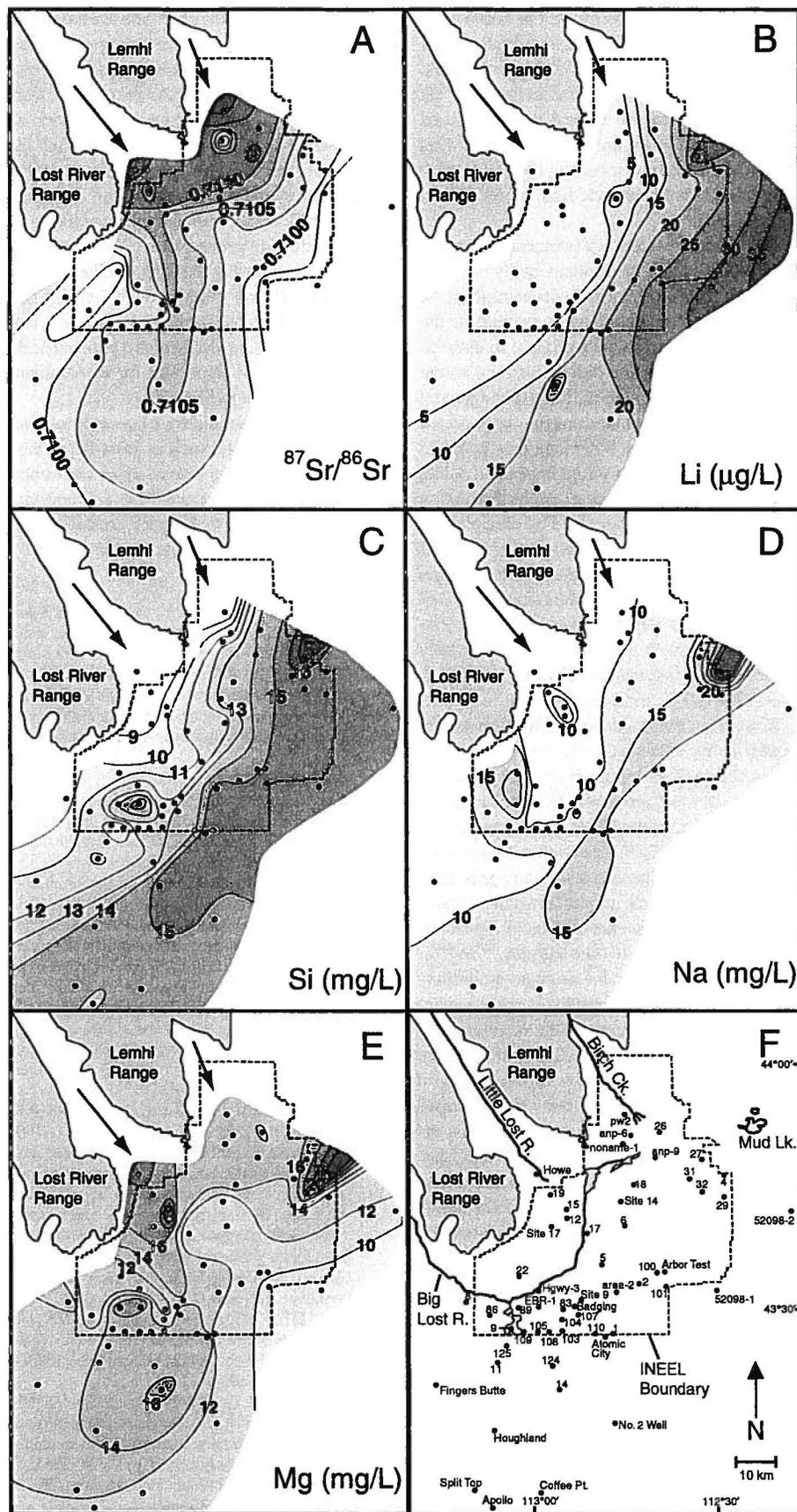


Figure 2. Contour plots of (A) $^{87}\text{Sr}/^{86}\text{Sr}$ and concentrations of (B) Li, (C) Si, (D) Na, and (E) Mg in groundwater (for tabulated data, see footnote 1). F: Sampled well locations (circles) and locations of river channels. Arrows indicate groundwater influxes from north. Spreading areas to which water is diverted during high runoff periods are shaded just south of southernmost point on Big Lost River channel. Contours were derived initially by Kriging and were modified by hand to reflect existence of aquifer boundaries and regional flow directions.

and SiO₂ by the rock as a result of water-rock interaction. However, there is an overall lack of interelement coherence in the concentration data, and the patterns are not as clear as the ⁸⁷Sr/⁸⁶Sr pattern. Other elemental concentrations measured but not plotted in Figure 2 are no better. This complexity in behavior among the solute concentrations is not unexpected, as we describe in the following.

The effects of water-rock interaction on radiogenic isotope ratios are fundamentally different from its effects on elemental concentrations. An elemental concentration changes according to the difference between solute gains due to dissolution, ion exchange, and desorption, and solute losses due to precipitation, ion exchange, and adsorption. A concentration can thus be expected to evolve in a complex way along flow paths, as the difference between solute losses and solute gains changes spatially. For example, dissolution of basalt is likely accompanied by precipitation of clay minerals, because even small amounts of Al in solution (0.1–1 μg/L) will result in supersaturation (Drever, 1988). The compositions of the precipitated clay minerals are sensitive to spatial variations in pH, temperature, and other variables, and thus the net effect of water-rock interaction is difficult to predict.

In contrast, the groundwater ⁸⁷Sr/⁸⁶Sr ratio is unaffected by solute losses due to precipitation, ion exchange, or adsorption, because the Sr transferred to the solid is isotopically identical to that in the water. Transfer of Sr from rock to groundwater drives the ⁸⁷Sr/⁸⁶Sr ratio toward the ratio of the acquired Sr. Thus, isotope ratios provide a record of water-rock interaction that is more easily interpreted than that obtained from solute concentration data. In this aquifer, ⁸⁷Sr/⁸⁶Sr evolves toward a predictable isotopic equilibrium value (0.70700) in response to water-rock interaction. In contrast, the evolution of solute concentrations is difficult to predict. Furthermore, evaporation of water increases concentrations of solutes but does not affect the ⁸⁷Sr/⁸⁶Sr ratio. Evaporative effects on the concentrations are present in the area downgradient from Mud Lake, a closed basin with highly evaporated infiltrating water (Fig. 2). Overall, ⁸⁷Sr/⁸⁶Sr and other radiogenic isotope ratios have clear advantages over concentration data. This is not to say that isotope ratios should be studied to the exclusion of concentration data, but that they should be added more commonly to the list of chemical measurements made in groundwater studies.

CONCLUSIONS

The fundamental properties of radiogenic isotope ratios make them well-behaved natural tracers and effective indicators of fast-flow paths in systems where chemical interaction between water and rock is important. Compared to concentration data, radiogenic isotopes may be more easily interpreted because the effects of water-

rock interaction, and the equilibria toward which the isotope ratios evolve, are more predictable.

Strong ⁸⁷Sr/⁸⁶Sr patterns observed in the Snake River Plain aquifer reflect the presence of fast- and slow-flow zones and arise from reaction of groundwater with the relatively low ⁸⁷Sr/⁸⁶Sr basalt host rocks. The inconsistent patterns yielded by elemental concentration data presumably reflect the more complex evolution of concentrations in groundwater. A zone of high ⁸⁷Sr/⁸⁶Sr groundwater trending southward from the Little Lost River valley through the INEEL is a high-velocity zone flanked by slower, low ⁸⁷Sr/⁸⁶Sr zones. Knowledge of this regional-scale flow channeling is important for contaminant management at the INEEL.

This approach should translate well to other groundwater settings, such as karst-dominated systems. Radiogenic isotope analyses are becoming less expensive, and now have an important role in the array of techniques used to reveal fast-flow paths in groundwater systems.

ACKNOWLEDGMENTS

This work was supported by the Office of Basic Energy Sciences, U.S. Department of Energy, under award DE-FG02-97ER14759 and contract DE-AC03-76SF00098. We thank Roy Bartholomay for help in obtaining groundwater samples. C. Bethke, C. Lundstrom, F. Phillips, and an anonymous reviewer provided suggestions that greatly improved the manuscript.

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Manuscript received March 20, 2000

Revised manuscript received June 28, 2000

Manuscript accepted July 14, 2000