Vadose Zone Transport Field Studies:
Isotope Tracer Results
FY00 Annual Report (9/1/00)

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Introduction

During the third of the five simulated leaks at the Sisson and Lu site, a mixture of non-radioactive isotopic tracers was added to the leak water. These tracers were chosen to represent specific contaminants of concern at Hanford and included 2 kg of D_2O, 3 g $NaH^{13}CO_3$, 25 mg ^{87}Sr, 1 mg ^{145}Nd, 1 mg ^{179}Hf and 3 mg ^{207}Pb. In addition to the isotopic tracers, approximately 6 kg of KBr was added to the same solution to provide a non-reactive chemical tracer (Br^-). 22 days after the tracers were released, borehole (S2) was drilled approximately 4 m from the injection well and 4 days later another borehole (S3) was drilled at about the same distance from the injection well. Core samples from these boreholes were sealed and shipped to the Center for Isotope Geochemistry (CIG) at the E.O. Lawrence Berkeley National Laboratory for processing. In addition, four samples of the tracer mixture taken during injection were also sent to CIG.

The porewaters in the core samples were vacuum distilled from ~200 g of core material. In addition, the CO_2 evolved during distillation of the water (representing the dissolved inorganic carbon compounds or DIC in the porewater) was collected. 1:1 mixtures of the sediments and de-ionized water were then prepared for isotopic analyses of the dissolved solids in the porewaters. To date, the δD and $\delta^{18}O$ values of the porewaters distilled from the samples and the $\delta^{13}C$ values of the CO_2 have been analyzed. A limited number of the $^{87/86}Sr$ ratios of the 1:1 extracts have also been measured. The bromide results presented with the deuterium results were analyzed by researchers from PNNL for a separate set of core samples.

Results and Discussion

The hydrogen isotope and bromide results for borehole S2 are plotted together on Figure 1. The water contents of the samples determined for the two types of analyses are shown on the left side of the figure and are in reasonable agreement. The relative concentrations of both deuterium and bromide plotted on the left side of Figure 1 also show similar trends. There are several interesting features of these data. The relative concentrations of the tracers are as high as 50% of the initial concentration of the leak solution. This is very high and allows for very little mixing with either the porewaters in the sediments prior to the test or the other solutions leaked into the ground during the test (the tracer solution represented only 20% of the total water injected). It is also significant that the highest concentrations of tracers are above the perched water horizons rather than within them. This suggests that the tracer solution spread along the top of the perched water horizons without significant mixing.

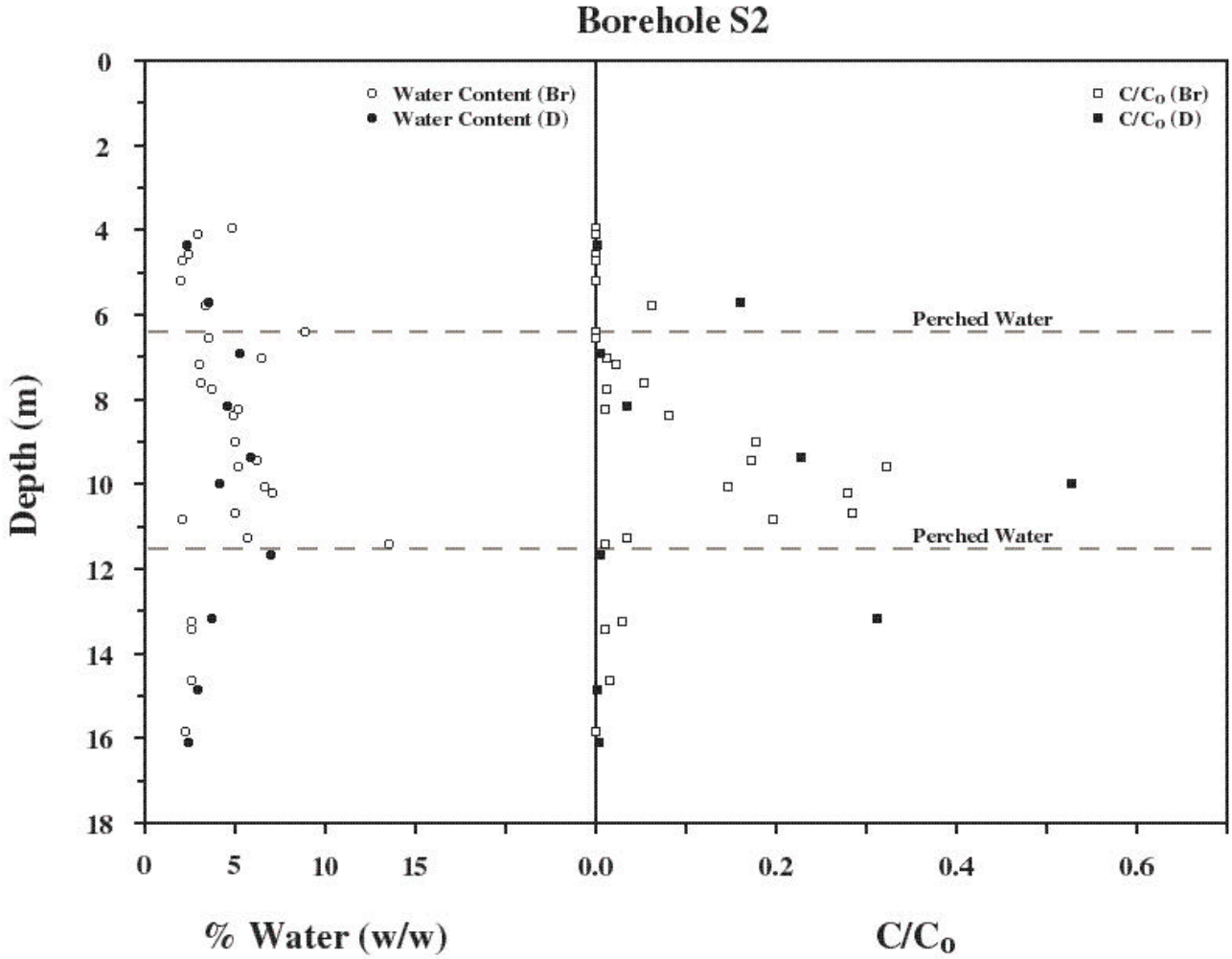

Figure 1. The water contents determined for both the bromide and deuterium samples from borehole S2 are plotted on the right. Plotted on the left are the concentrations of deuterium and bromide relative to their initial concentrations (C_o) in the tracer solution. Also shown are the approximate depths of perched water horizons in the hole (determined from the measured water contents and the results of geophysical studies at the site).

The $^{87/86}Sr$ ratios of four of the samples from this hole have been analyzed. Three of those samples had highly elevated deuterium concentrations and one was near background. The strontium isotope ratios of all of the samples, however, were within the range of background. This is significant because it indicates that the strontium added to the tracer solution was strongly retarded during subsurface transport.

Despite addition of $NaH^{13}CO_3$, the $\delta^{13}C$ values of the DIC in both the tracer solution and the porewaters extracted from the cores were not significantly elevated. This suggests that the DIC in the tracer solution exchanged with atmospheric CO_2, despite efforts to prevent this from happening.

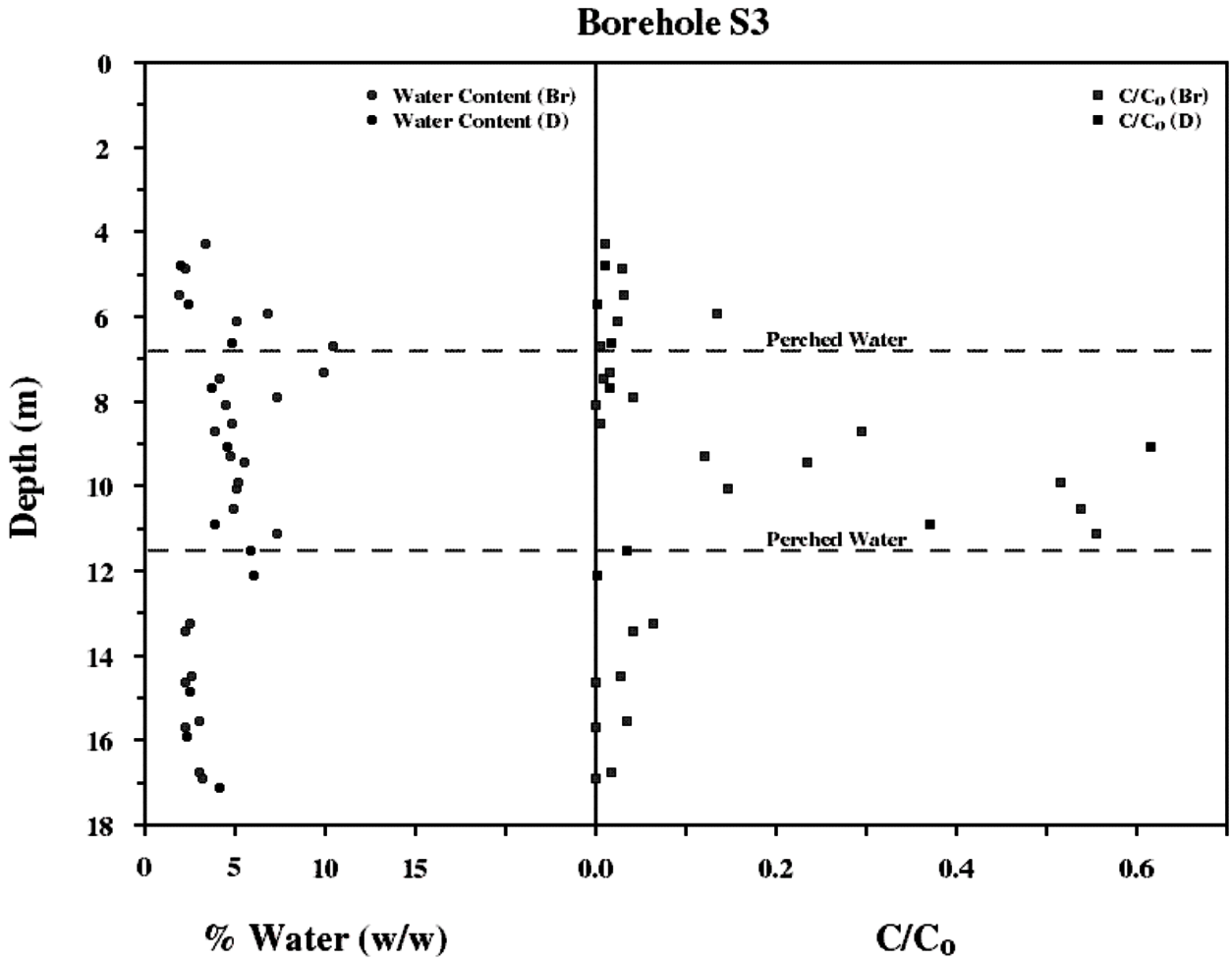

Figure 2. The water contents determined for both the bromide and deuterium samples from borehole S3 are plotted on the right. Plotted on the left are the concentrations of deuterium and bromide relative to their initial concentrations (C_o) in the tracer solution. Also shown are the approximate depths of perched water horizons in the hole (determined from the measured water contents and the results of geophysical studies at the site).

The deuterium and bromide results from borehole S3 are plotted on Figure 2. These data are similar to those for borehole S2. The relative concentrations of both deuterium and bromide are very high (reaching greater than 50% of the concentration in the tracer solution). In addition, the peak concentrations of the tracers are also above the perched water horizons, rather than mixed into them. This supports the hypothesis that the tracer solution (and the other leaks?) spread out laterally on top of the saturated zones in the perched water horizons. To date, data for the other tracers are not available for this borehole.

Conclusions

The preliminary results presented here suggest several interesting conclusions about infiltration of contaminants through the vadose zone at Hanford:

- The tracer solutions move laterally through coarser sediments above perched water horizons with relatively little mixing. The extent of this lateral spreading is not clear. A smaller Geoprobe core was drilled about six weeks after the tracer solution was introduced into the ground at a distance of approximately 12 m from the injection well and no bromide was detected in the samples.
- Initial results indicate that the sediments significantly retard strontium.

Shown in Figure 3 is a schematic representation of these findings. This model is highly speculative and only outlines what happens during the initial stages of infiltration. Future sampling will help significantly delineate the spread of the tracers and quantify the degree of retardation of contaminants such as strontium.

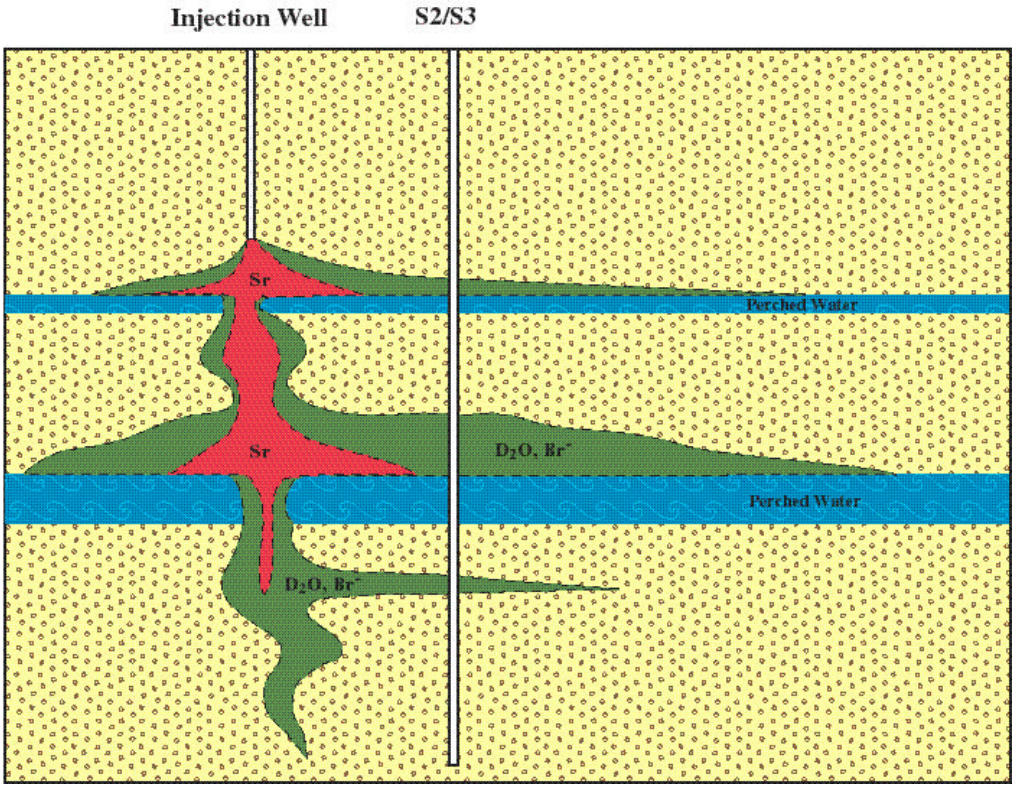

Figure 3. Schematic model of the spread of D2O, bromide and strontium in the Hanford sediments during the initial stages of a leak from a point source.

Table 1. Isotopic compositions of different water sources at Hanford

Source	δD (‰)	$\delta^{18}O$ (‰)	$\delta^{13}C$ (‰)[3]	$^{87/86}Sr$	$^{145/144}Nd$
Columbia River	-133 to -123	-17.8 to -16.6	-9.6 to -1.6	0.7135 to 0.7147	
Groundwater	-146 to -104	-19.6 to -13.1	-35.6 to -6.6	0.7068 to 0.7155	
Vadose Zone Porewaters	-145 to -100	-17.3 to -9.1	-17.9 to -4.1	0.7145 to 0.7210	
Tracer Solution	2574 to 2723			1.3	7.4 to 1.6

Table 2. Isotope data for S2 core.

Depth (ft)[1]	Depth (m)	$\%H_2O$[2]	δD (‰)	C/C_o D	$\delta^{18}O$ (‰)	$\delta^{13}C$ (‰)[3]	$^{87/86}Sr$[4]	$^{145/144}Nd$[4]
14.25	4.34	2.3	-123	0.001	-14.7	-11.1		
18.75	5.71	3.5	323	0.160	-15.4	-10.5	0.7147	0.3533
22.75	6.93	5.3	-112	0.005	-15.8	-11.6		
26.75	8.15	4.6	-28	0.035	-16.3	-9.1	0.7123	0.3498
30.75	9.37	5.9	513	0.228	-16.4	-8.4		
32.75	9.98	4.1	1356	0.528	-16.3	-7.8	0.7139	0.3508
38.25	11.66	7.0	-112	0.005	-14.9	-10.7	0.7146	0.3495
43.25	13.18	3.7	749	0.312	-15.7	-14.9	0.7153	0.3517
48.75	14.86	2.9	-124	0.001	-15.2	-9.9		
52.75	16.08	2.4	-119	0.003	-15.2	-10.0	0.7118	0.3515

[1] Depth at midpoint of interval sampled.

[2] % weight of water per total weight of sample.

[3] Carbon isotope ratio of dissolved inorganic carbon extracted from porewater in cores.

[4] Isotope composition in 1:1 mix of sample and de-ionized water .

Table 3. Isotope data for S3 core.

Depth (ft)[1]	Depth (m)	$\%H_2O$[2]	δD (‰)	C/C_o D	$\delta^{18}O$ (‰)
15.75	4.80	2.0	-119	0.002	-14.8
18.75	5.71	2.4	-123	0.001	-15.2
21.75	6.63	4.8	-75	0.018	-16.4
25.25	7.70	3.7	-85	0.015	-16.7
29.75	9.07	4.6	1602	0.616	-16.5
35.75	10.90	3.9	887	0.361	-16.5
37.75	11.51	5.9	-30	0.034	-16.4
39.75	12.12	6.0	-123	0.001	-14.8
48.75	14.86	2.5	-122	0.001	-15.5
52.25	15.93	2.3	-124	0.001	-15.2
56.25	17.14	4.1	-125	0.001	-15.0

[1] Depth at midpoint of interval sampled.

[2] % weight of water per total weight of sample.

[3] Carbon isotope ratio of dissolved inorganic carbon extracted from porewater in cores.

[4] Isotope composition in 1:1 mix of sample and de-ionized water .