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Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1993

Annual Report

Prepared by
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Idaho National Engineering Laboratory
EG&G Idaho, Inc.

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ABSTRACT

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program, funded by the U.S. Nuclear Regulatory Commission, is (a) studying the degradation effects in EPICOR-II organic ion-exchange resins caused by radiation, (b) examining the adequacy of test procedures recommended in the Branch Technical Position on Waste Form to meet the requirements of 10 CFR 61 using solidified EPICOR-II resins, (c) obtaining performance information on solidified EPICOR-II ion-exchange resins in a disposal environment, and (d) determining the condition of EPICOR-II liners.

Results of the eighth year of data acquisition from the field testing are presented and discussed. During the continuing field testing, both Portland type I–II cement and Dow vinyl ester-styrene waste forms are being tested in lysimeter arrays located at Argonne National Laboratory-East in Illinois and at Oak Ridge National Laboratory. The study is designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over a 20-year period.

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EXECUTIVE SUMMARY

The March 28, 1979 accident at Three Mile Island Unit 2 released approximately 560,000 gal of contaminated water to the auxiliary and fuel handling buildings. The water was decontaminated using a three-stage demineralization system called EPICOR-II containing organic and inorganic ion-exchange media. The first stage of the system was designated the prefilter, and the second and third stages were called demineralizers. Research is being conducted at the Idaho National Engineering Laboratory on materials from four of those EPICOR-II prefilters under three tasks of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program.

Field testing consists of examining the effect of disposal environments on solidified resin wastes from EPICOR-II prefilters. The purpose of this task, using lysimeter arrays at Oak Ridge National Laboratory in Tennessee and Argonne National Laboratory-East in Illinois, is to expose samples of solidified ion-exchange resin to the actual physical, chemical, and microbiological conditions of a disposal environment. The study is designed so that continuous data on chemical species and nuclide release and movement, as well as environmental conditions, will be obtained over a 20-year period. Each month, data are retrieved from the data acquisition system. At least quarterly, water is drawn from the porous cup soil-water samplers and from the lysimeter leachate collection compartment. Those water samples are analyzed for chemical species and beta- and gamma-producing nuclides.

Results show that radionuclides are continuing to move from the waste forms and through the soil column. VES is comparable to cement in retaining Sr-90, unlike findings from Savannah River Laboratory, which found cement to be a better retainer than VES.

A source term computer code is used to model the release of radionuclides from the lysimeter waste forms. Also, comparisons of code prediction to actual lysimeter data have been made.

ACRONYMS AND ABBREVIATIONS

ANL-E	Argonne National Laboratory-East
ASTM	American Society for Testing and Materials
BC	boundary condition
CV	coefficient of variation
DAS	data acquisition system
DUST	Disposal Unit Source Term
INEL	Idaho National Engineering Laboratory
NRC	U.S. Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
VES	vinyl ester-styrene

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INTRODUCTION

The March 28, 1979 accident at Three Mile Island Unit 2 released approximately 560,000 gal of contaminated water to the auxiliary and fuel handling buildings. The water was decontaminated using a demineralization system called EPICOR-II developed by Epicor, Inc.[a] The contaminated water was cycled through three stages of organic and inorganic ion-exchange media. The first stage of the system was designated the prefilter, and the second and third stages were called demineralizers. After the filtration process, the ion-exchange media in 50 of the prefilters contained radionuclides in concentrations greater than the limits for low-level wastes. These prefilters were transported to the Idaho National Engineering Laboratory (INEL) for interim storage before final disposal. A special overpack (high-integrity) container was developed during that storage period to dispose of the prefilters at a commercial disposal facility in the State of Washington. As part of the EPICOR and Waste Research and Disposition Program funded by the U.S. Department of Energy, 46 prefilters were disposed. Four prefilters used in U.S. Nuclear Regulatory Commission (NRC) studies were stored in temporary storage casks outside the Hot Shop of Test Area North Building 607 at the INEL. Those four prefilters were disposed during this reporting year at the Radioactive Waste Management Complex on the INEL Site.

Under the EPICOR and Waste Research and Disposition Program, continuing research has been conducted by EG&G Idaho, Inc. on materials from those EPICOR-II prefilters.[1,2,3] That work is now directed by the NRC as part of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program. Studies are being conducted on organic ion-exchange resins from selected prefilters. The resins were examined to measure degradation, and tests are being performed to characterize solidified ion-exchange media.

The results of resin degradation from studies of the first and second sampling, as described in References 4 and 5,were compared with those of the third sampling described in Reference 6. The degradation studies determined the acceptability of EPICOR-II prefilters for disposal in high-integrity containers at the commercial disposal site at Hanford, Washington by identifying (a) degradation effects on the ion-exchange resins caused by contained radiation and (b) the possible release of contained radionuclides from ion-exchange resins. Those studies are complete and are not reported here.

Another aspect of this program was investigated—the solidification of EPICOR-II wastes from prefilters PF-7 and PF-24 using Portland type I-II cement and vinyl ester-styrene (VES), a proprietary solidification agent developed and supplied by the Dow Chemical Company.

The formulations used for the immobilization of EPICOR-II wastes were developed to produce waste forms meeting the regulatory requirements of 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste." The NRC, in its "Technical Position on Waste Form, Rev. 1,"[7] provides guidance to waste generators on waste form test methods and acceptable results for compliance with the waste form requirements of 10 CFR 61. In this study, EPICOR-II waste forms are annually subjected to the specified

a. Mention of specific products or manufacturers in this document implies neither endorsement or preference nor disapproval by the U.S. Government, any of its agencies, or EG&G Idaho, Inc., of the use of a specific product for any purpose.

compression-test procedures to ensure compliance with stability requirements. The data indicate that the waste form strength is increasing with age.

Solidified waste forms containing EPICOR-II ion-exchange resin waste are currently being field-tested using lysimeters. The intent of the testing is to expose waste-form samples to the physical, chemical, and microbiological environment of typical disposal sites in the eastern United States (References 1, 2, and 3). The lysimeters are expected to monitor the release of nuclides from buried waste forms and provide data that accurately determine movement as a function of time and environmental conditions. Emphasis is placed on investigating the requirements of 10 CFR 61. The study is designed so that continuous data on nuclide release and movement, as well as environmental conditions, will be obtained over a 20-year period.

This report contains data from the eight years of lysimeter operation, as well as cumulative data on water balance and chemical species and nuclide content of water samples. Data for this report were retrieved from the data acquisition system (DAS), from chemical speciation of water samples, and from beta and gamma analyses of lysimeter water samples.

RESIN SOLIDIFICATION

In this task, EPICOR-II waste forms solidified with Portland type I–II cement and VES are annually subjected to compression-testing per ASTM C39. One specimen of each type of waste form (all organic and organic with zeolite) in each solidification agent (cement and VES) are normally tested. No testing was performed this fiscal year under this task.

FIELD TESTING

Materials and Methods

Experiment Description. Solidified waste forms containing EPICOR-II ion-exchange resin waste are currently being field-tested using lysimeters. Lysimeter sites have been established at Oak Ridge National Laboratory (ORNL) and Argonne National Laboratory-East (ANL-E). Instrumentation within each of the five lysimeters at each site includes porous cup soil-water samplers and soil moisture/temperature probes. The probes are connected to an onsite DAS, which also collects data from a field meteorological station located at each site. A detailed description of the lysimeters and their installation is presented in Reference 8 while data from the first eight years of operation are contained in earlier reports.[9,10,11,12,13,14,15]

Description of Waste Forms. Waste forms used in the field test are composed of solidified EPICOR-II prefilter resin wastes. Two waste formulations are used in the solidification project (Table 1). Type I is a mixture of synthetic organic ion-exchange resins from PF-7 (phenolic cation, strong acid cation, and strong base anion resins), while type II is a mixture of synthetic organic ion-exchange resins from PF-20 (strong acid cation and strong base anion resins) with an inorganic zeolite. Waste type I contains 5% Sr-90, while type II contains about 1% Sr-90. Of the other radionuclides in those wastes, Cs-137 and Cs-134 are the major constituents, with Sb-125 found in trace amounts.

Portland type I–II cement and VES were used to solidify both types of resin wastes. Individual waste-form specimens were manufactured by allowing a mixture of solidification agent and resin waste to solidify in polyethylene molds that were 4.8 cm in diameter by 10.2 cm high. Enough of the mixture was added to each vial to produce specimens with an average diameter of 4.8 cm and a height of 7.6 cm (137.5 cm^3). Each lysimeter contains seven of these 4.8 × 7.6-cm waste-form specimens stacked end-to-end to form a 1-L waste volume. Table 1 shows the types of specimens placed in the lysimeters. A complete description of waste form manufacture is given in Reference 16. Bench testing of similar waste forms, per the requirement of the Branch Technical Position on Waste Form, is described in Reference 17.

Description of Lysimeters. The lysimeters are designed as self-contained units that can be easily disposed at the termination of the field test experiment. A total of ten lysimeters are used, with five placed at each field site. Each lysimeter is a right circular cylinder (0.91 m in diameter by 3.12 m in height), constructed of 12-gauge, 316 L stainless steel (Figure 1). Internally, the lysimeter is divided into two sections, the upper volume being 1,532 L and the lower volume being 396 L. A 3.8-cm, Schedule 40 stainless steel pipe serves as an access to the lower compartment. The upper compartment of each lysimeter contains the soil column with waste forms, three temperature/moisture probes, and five soil moisture cups as shown in Figure 1. The cups are numbered 1 through 5 as noted in Figure 1. The lower compartment serves as a leachate collector, which is emptied and sampled through the 3.8-cm pipe.

Table 1. Lysimeter waste form composition.

Lysimeter number	Fill material	Waste form description	Prefilter number
1	Soil	Cement with type I waste	PF-7
2	Soil	Cement with type II waste	PF-24
3	Soil	VES with type I waste	PF-7
4	Soil	VES with type II waste	PF-24
5 ANL-E	Silica oxide	Cement with type I waste	PF-7
5 ORNL	Silica oxide	Cement with type II waste	PF-24

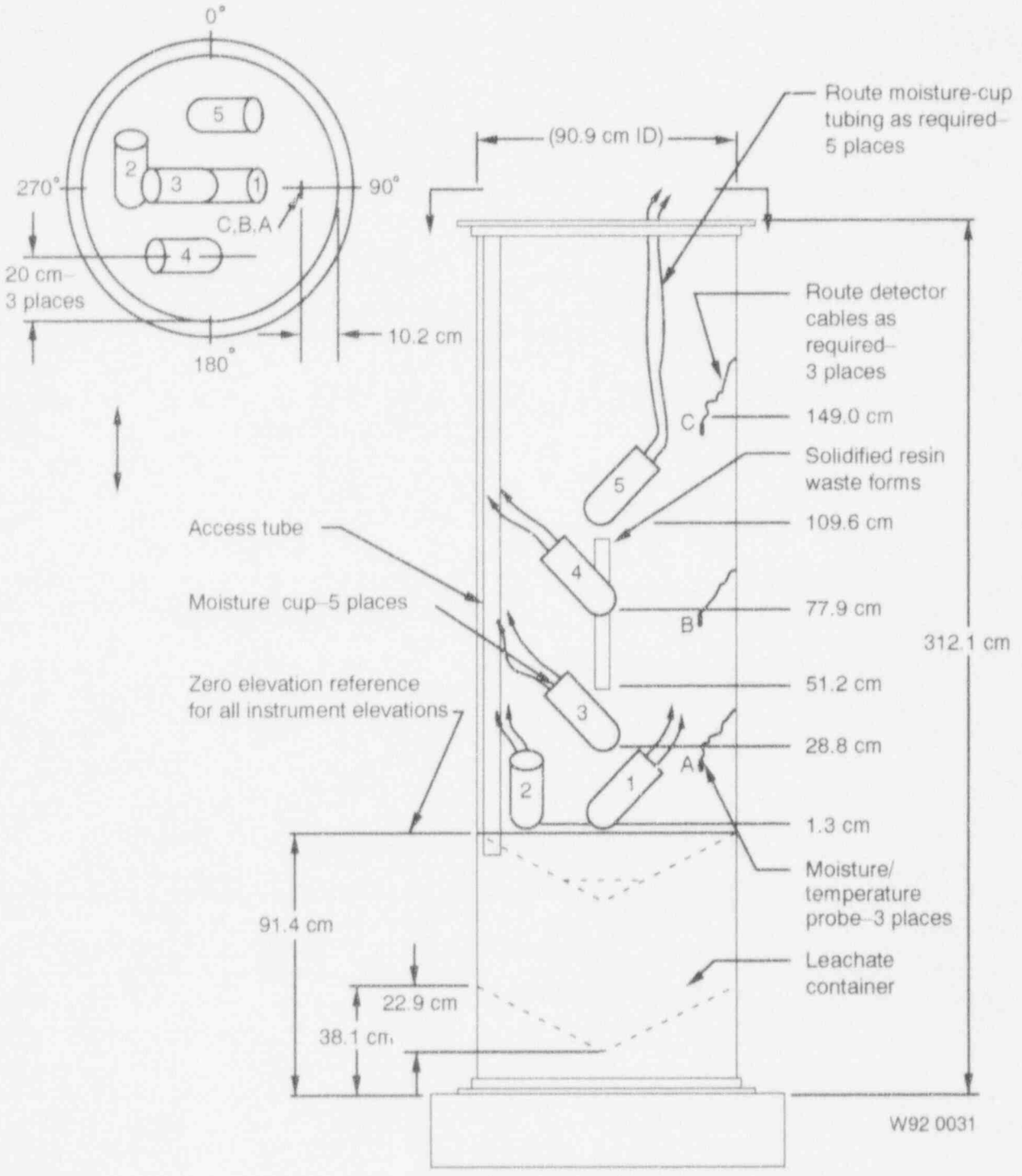

Figure 1. EPICOR-II lysimeter vessel component locations.

Four lysimeters at each field site (numbered 1 through 4) are filled with soil; the remaining one (number 5) is a control lysimeter filled with an inert silica sand (Reference 8). Two different soils were used. One was representative of Midwestern soils; the other was intended to approximate soil found at Barnwell, South Carolina. ANL-E used local indigenous soil that fits the NRC criteria for Midwestern soil. It is a Morley silt loam with the surface layer removed. The resulting subsurface soil is a clay loam. Soil at ORNL was not found to be a suitable substitute for Barnwell soil; therefore, acceptable soil was transported to ORNL from the Savannah River Plant adjacent to the Barnwell facility in South Carolina.

Soil temperature and moisture sensors are physically located within a common housing or probe. These probes are located at three elevations (149, 77.9, and 28.8 cm, as measured from the bottom of the soil column) within each lysimeter. The function of these probes is to provide data on whether or not the buried waste forms experience freezing temperatures and if the surrounding soil is moist. Because all of the soil lysimeters at each site are exposed to the same environment, the current placement of probes provides a planned redundancy in data collection. Therefore, as long as there are functioning probes in any of the soil lysimeters at each site, sufficient data to satisfy reporting criteria will be available.

Data Retrieval and Analysis. Electrical impulses from the environmental instruments are collected by, processed in, and stored by the DAS for periodic retrieval. The DAS processes input into recognizable data using programmable steps. Output from the soil moisture probes, for example, is processed by a polynomial equation that was derived from laboratory calibration of the probes (Reference 8).

Data output from the DAS is stored on a cassette tape and is then retrieved and translated to an IBM PC-compatible disk file. These files are printed either as graphs or in an alphanumerical format. Graphs present data over an extended time period and were used for this report.

Water from each lysimeter is drawn from porous cup soil-water samplers and lysimeter leachate collection compartments at least quarterly. These water samples are analyzed routinely for gamma-producing nuclides and, as required, for the beta-producing nuclide Sr-90. Water analyses are performed at ANL-E by the Environmental Services Laboratory and at ORNL by the Environmental Radio Analysis Laboratory. Both of these laboratories have a traceable quality assurance program and use accepted analytical procedures for nuclide determination.

Results and Discussion

This report contains DAS data from ANL-E and ORNL obtained from July 1992 through June 1993. In addition, information on water balance and on nuclide chemical species content in soil water and leachate is compiled from the initiation of the project (ANL-E—August 1, 1985; ORNL—June 1, 1985) through June 1993. Many of the data are displayed in graphic format so that information can be correlated easily with time. There were two periods of time when the DAS was not in operation at ORNL. They were from July through August 17, 1992, and again in mid September of 1992. Both were equipment failures requiring repair.

Weather Data. Precipitation, air temperature, and relative humidity, as recorded by the ANL-E and ORNL data acquisition systems during the 12-month reporting period, are presented in Figures 2 through 6. In October 1990, the anemometer at ANL-E ceased normal operation. During this reporting period, the anemometer at ORNL appears to have failed at times due to mechanical wear of bearings. Also, relative humidity readings at both sites have been questionable. Because of these failures, windspeed and relative humidity data are not included in this report. Total official precipitation for the period (measured by reference rain gauges near each site) was 129.8 cm at ANL-E and 137.8 cm at ORNL. This year, ANL-E was above the normal annual rainfall while ORNL was nearly equal to the norm (ANL-E—85.2 cm; ORNL—138.8 cm).[18,19] This is the sixth time in the past seven years that ORNL has equalled or exceeded the normal amount of yearly precipitation. The monthly precipitation pattern for each

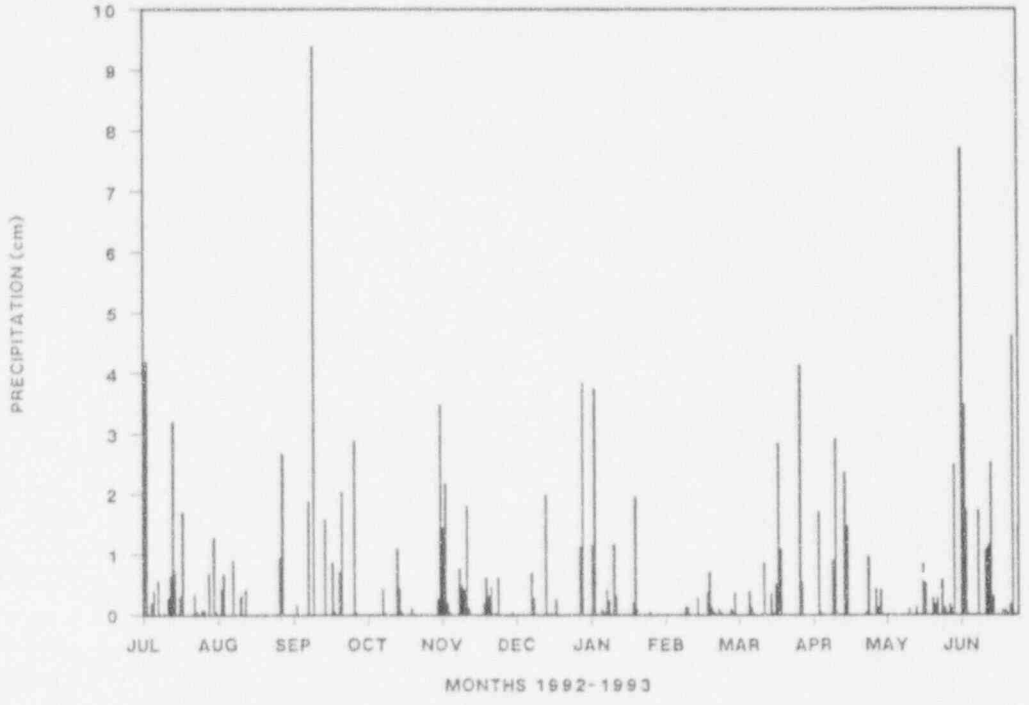

Figure 2. ANL-E weather data—precipitation.

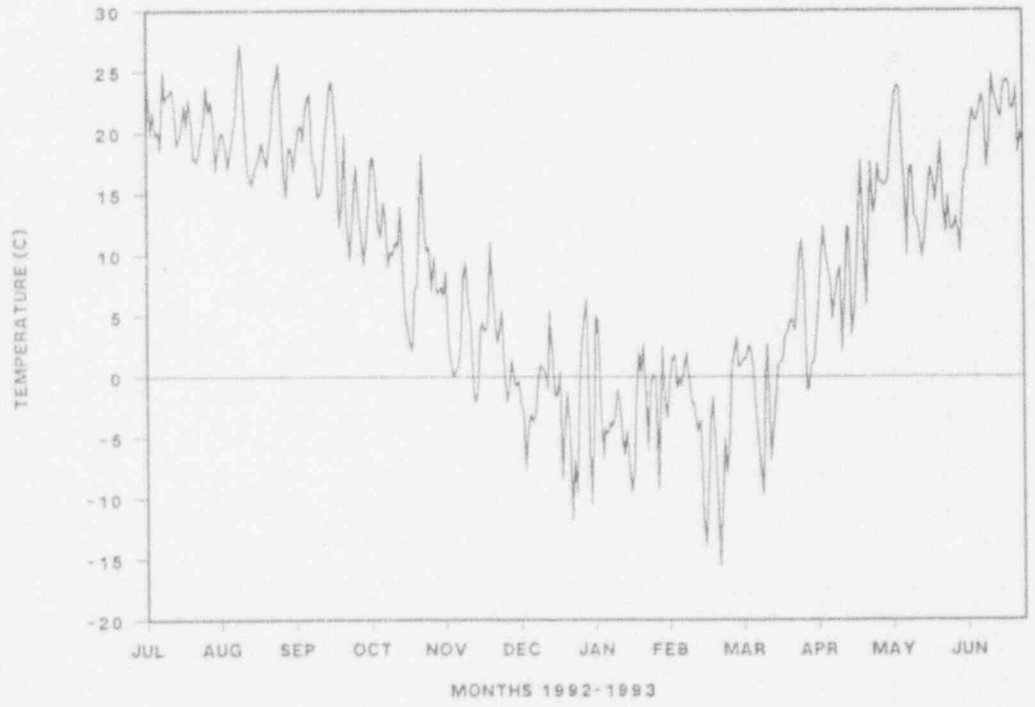

Figure 3. ANL-E weather data—air temperature.

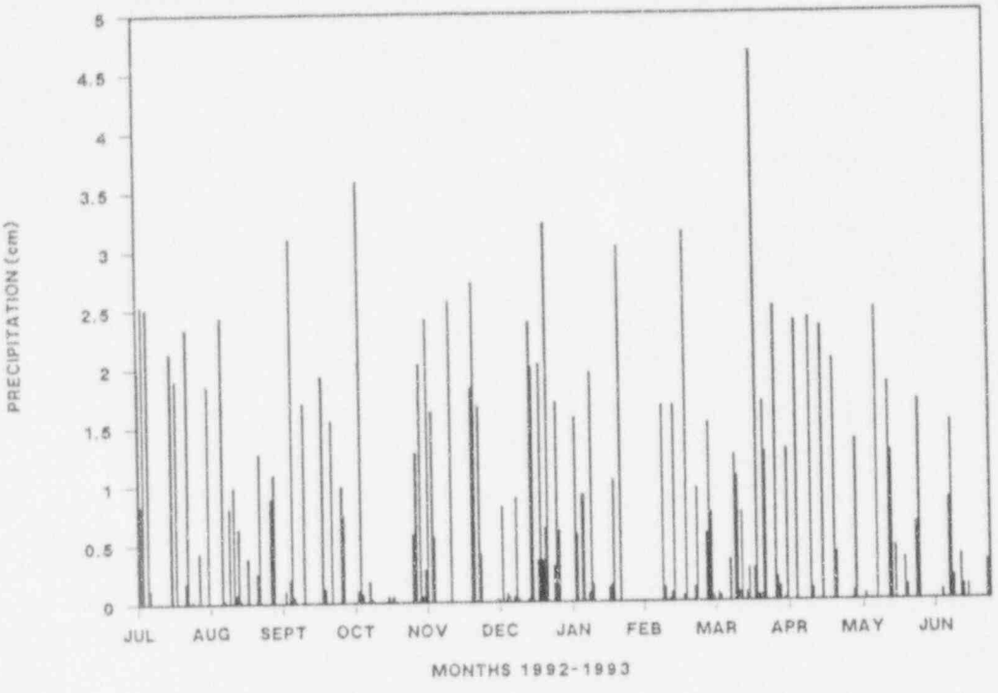

Figure 4. ORNL weather data—precipitation.

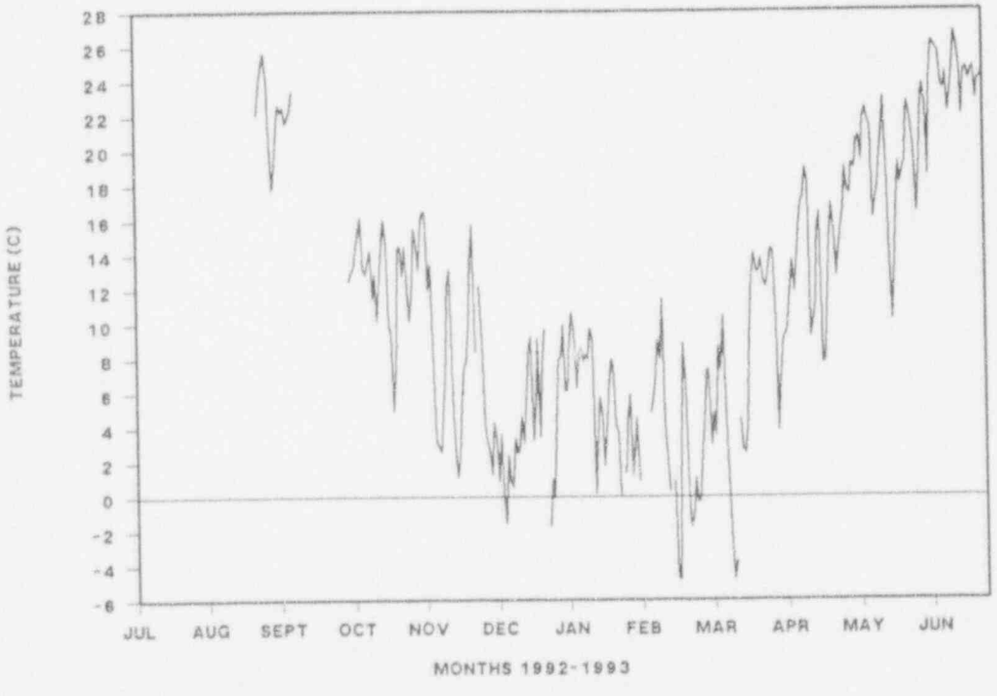

Figure 5. ORNL weather data—air temperature.

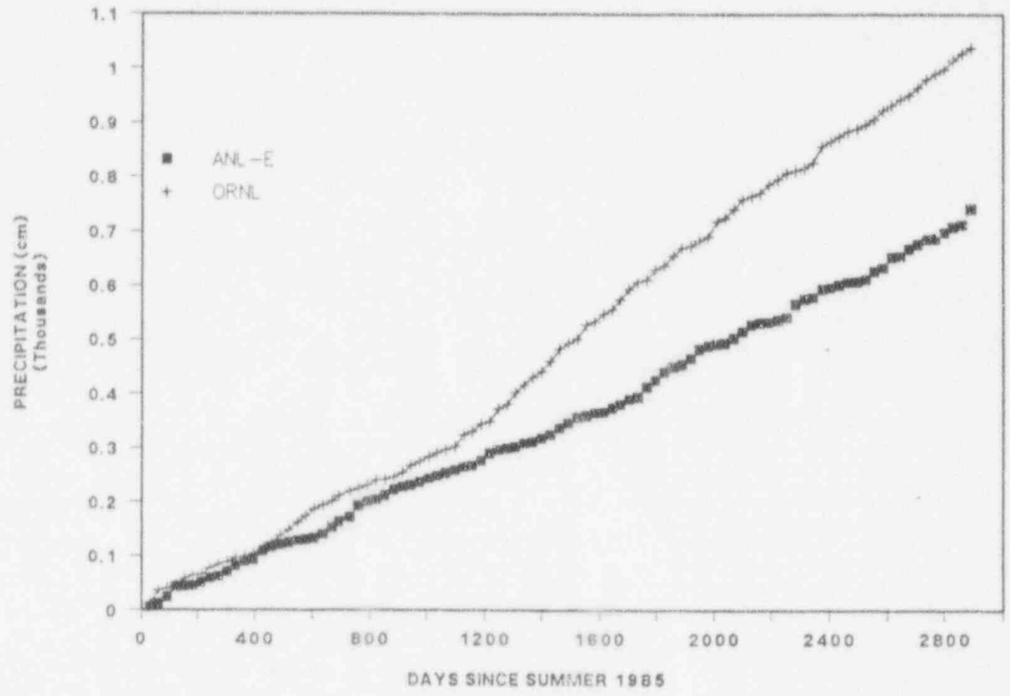

Figure 6. ANL-E and ORNL weather data—cumulative precipitation.

site can be seen from the histograms in Figures 2 and 4. Figure 6 shows the cumulative pattern of precipitation for both sites since the initiation of field work. By the end of this reporting period, there was a cumulative total of 742.1 cm at ANL-E, while ORNL received a total of 1,038.6 cm.

Air temperature data from ANL-E (Figure 3) show that periods of freezing temperatures occurred from late October 1992 until the first part of April 1993. ORNL experienced periods of freezing temperatures from early November until early March (Figure 5).

Lysimeter Soil Temperature Data. The lysimeter soil temperature data recorded at ANL-E and ORNL during the reporting period are shown in Figures 7 through 15. At no time during the reporting period was a freezing temperature recorded by a functioning temperature probe at the depth of the buried waste forms within a lysimeter. A direct correspondence can be seen between air temperature and soil temperatures at both locations.

As stated in past reports, a number of temperature probes at ANL-E have failed during the last five reporting periods, all the temperature probes in ANL-4 and one in ANL-2 had failed to function; therefore, data from these probes were not included in the report. During the 1991–1992 reporting period, it appeared that two of the probes in ANL-3 as well as one in ANL-5 were not functioning properly. Partial deterioration of the remaining ANL-3 probe was seen during that reporting period. The probes have probably been damaged by corrosion of the metal parts (Reference 10). At the present time, a more damage-resistant replacement for these probes has not been found. Occasional erratic behavior of some ORNL probes seen during the last reporting period has been reduced to a single spike on several outputs. The ORNL-5 probe, at a level of 28.8 cm, is suspect.

Lysimeter Soil Moisture Data. Data from the moisture probes at both ANL-E and ORNL,

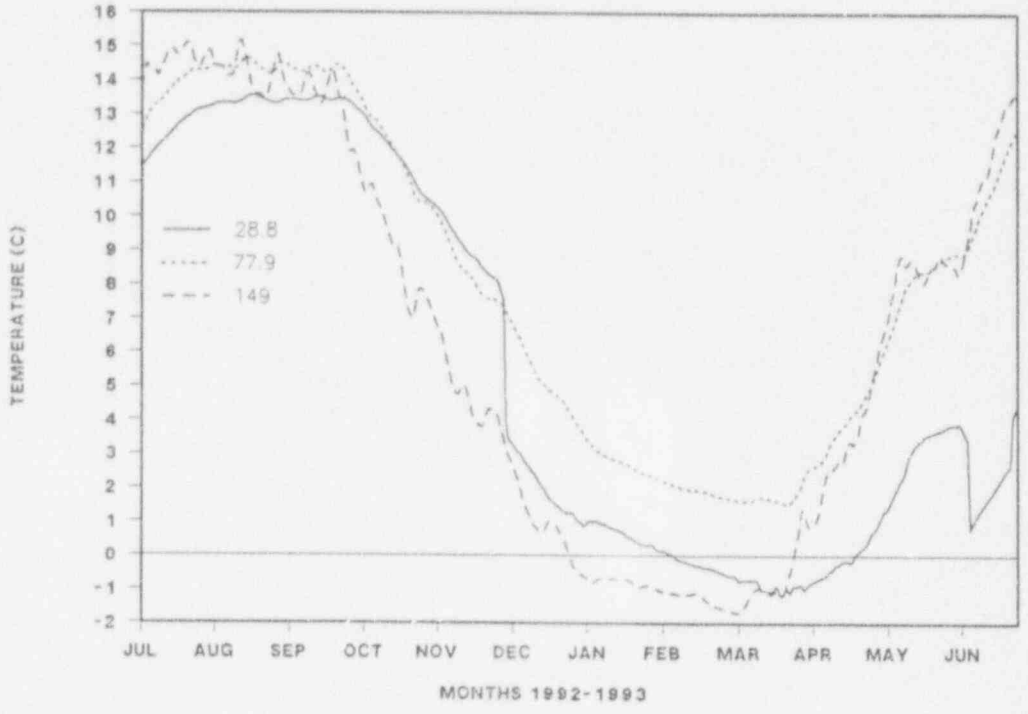

Figure 7. ANL-E lysimeter 1 soil temperature.

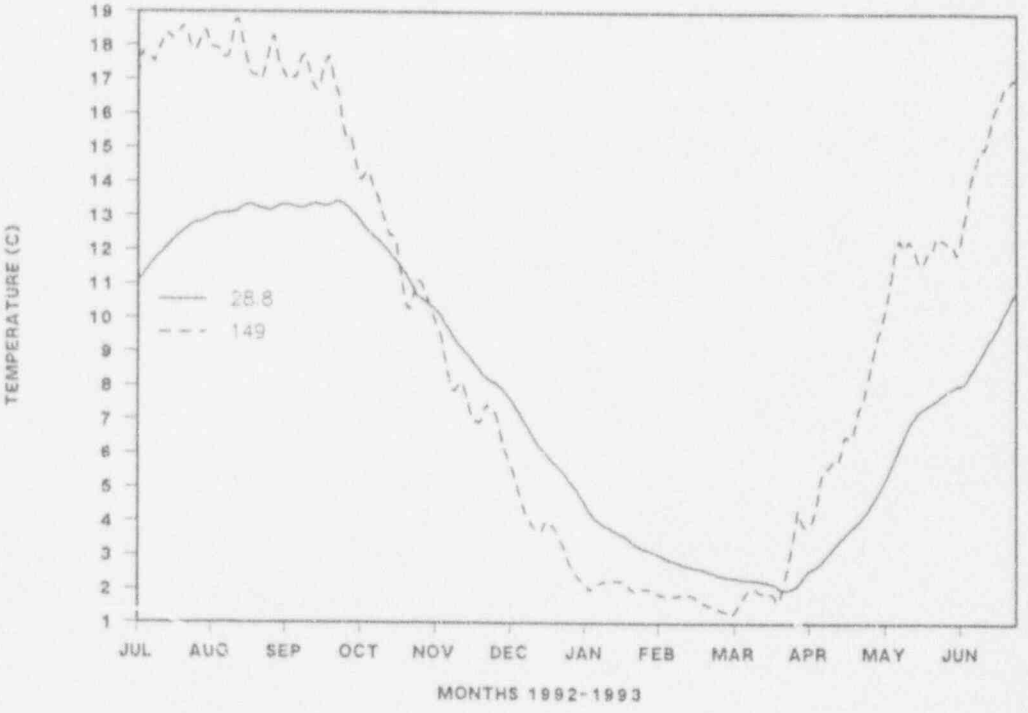

Figure 8. ANL-E lysimeter 2 soil temperature.

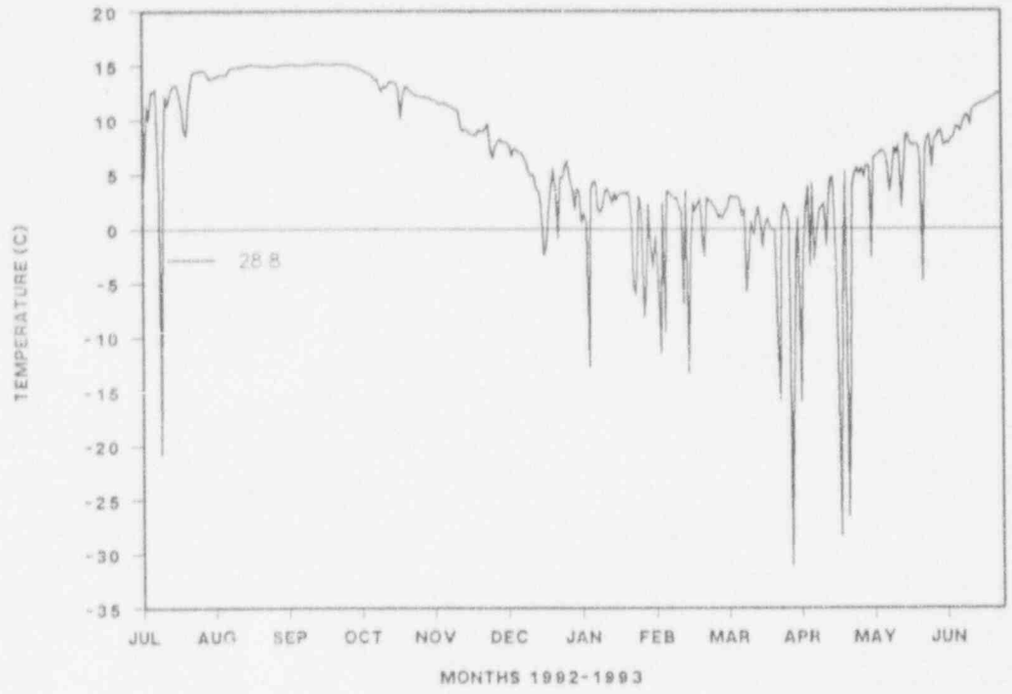

Figure 9. ANL-E lysimeter 3 soil temperature.

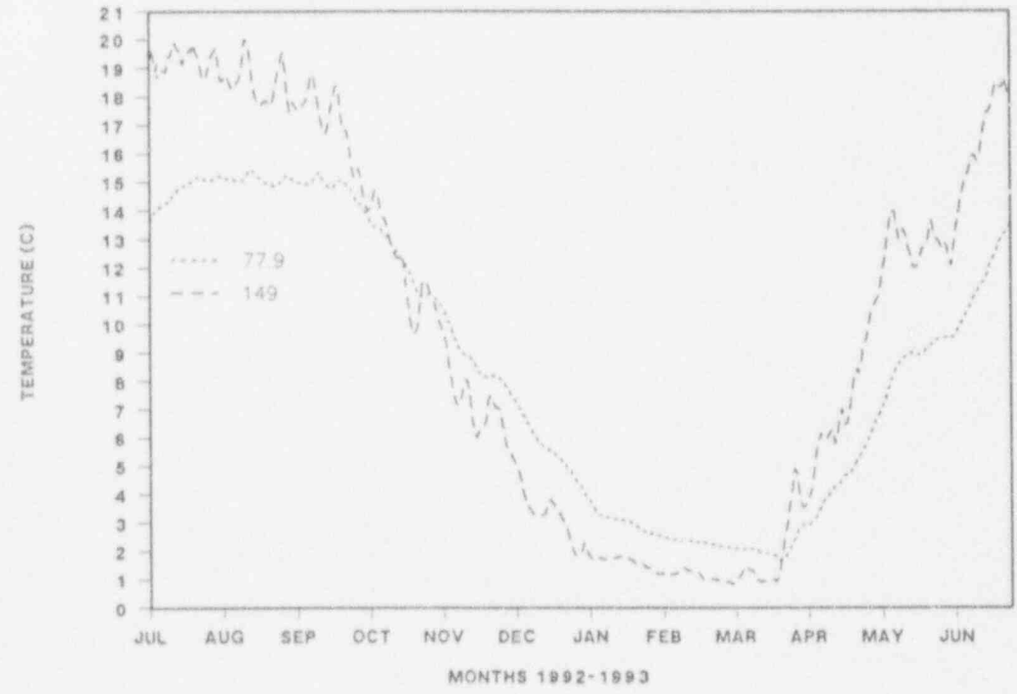

Figure 10. ANL-E lysimeter 5 soil temperature.

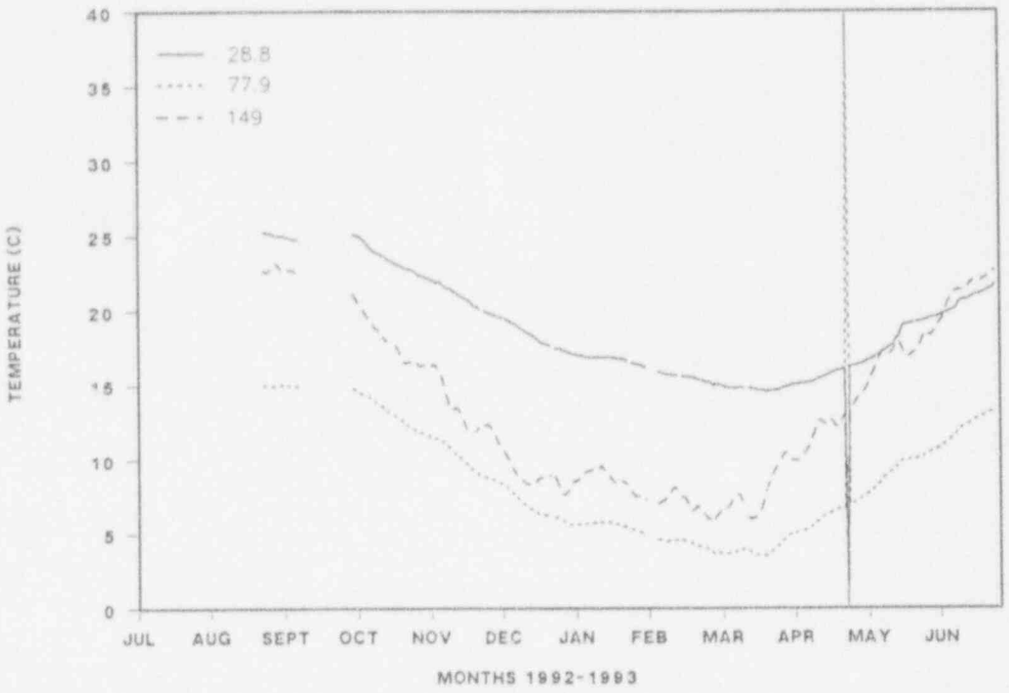

Figure 11. ORNL lysimeter 1 soil temperature.

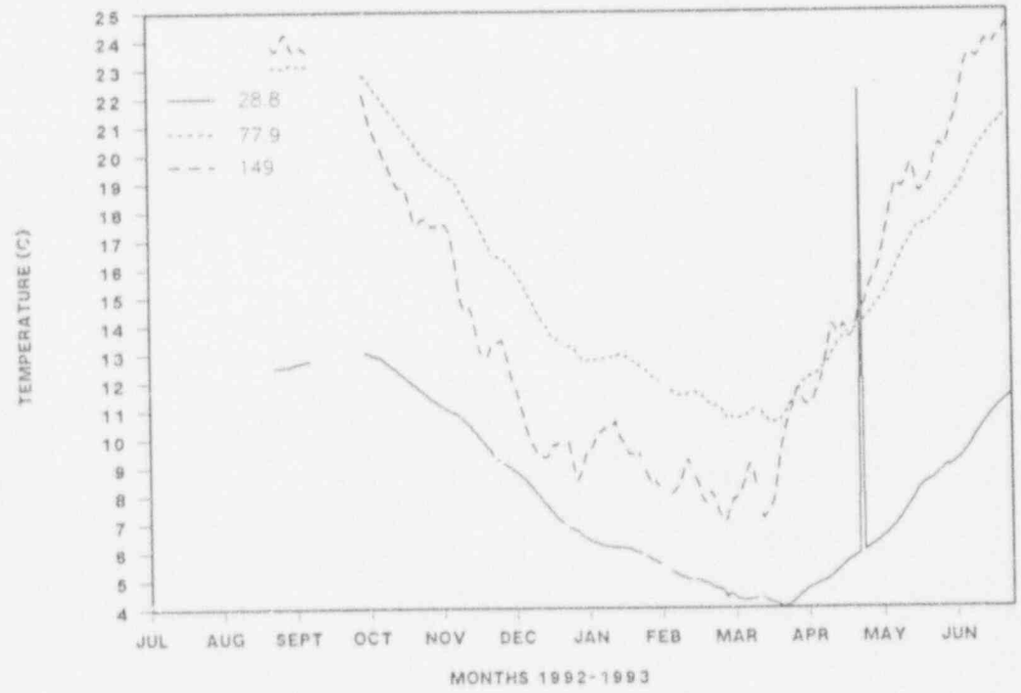

Figure 12. ORNL lysimeter 2 soil temperature.

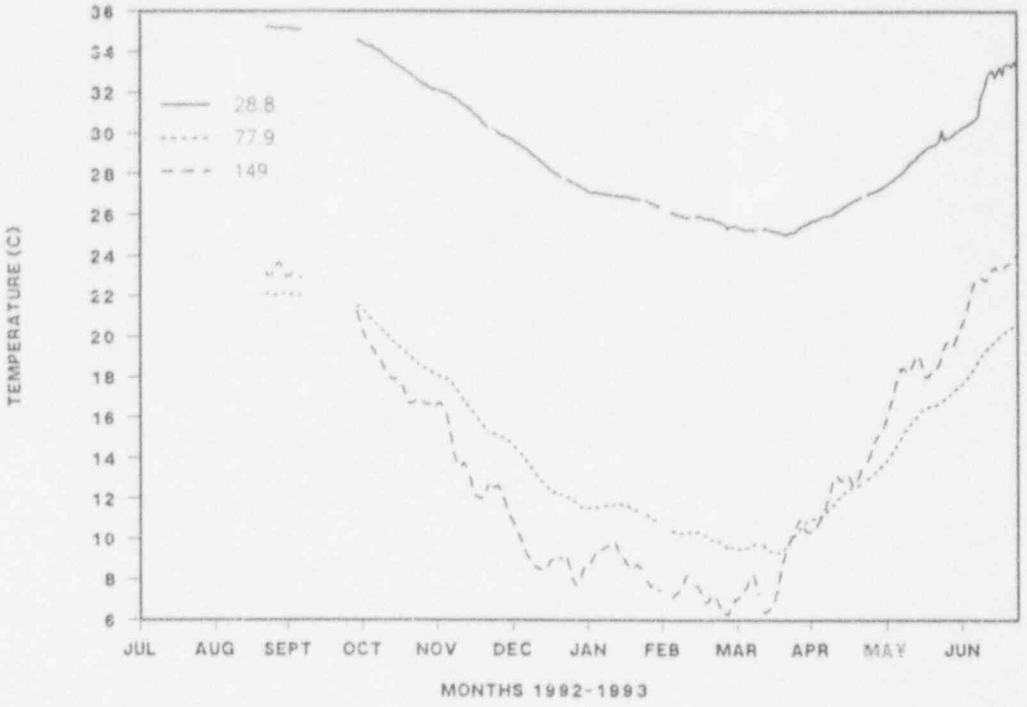

Figure 13. ORNL lysimeter 3 soil temperature.

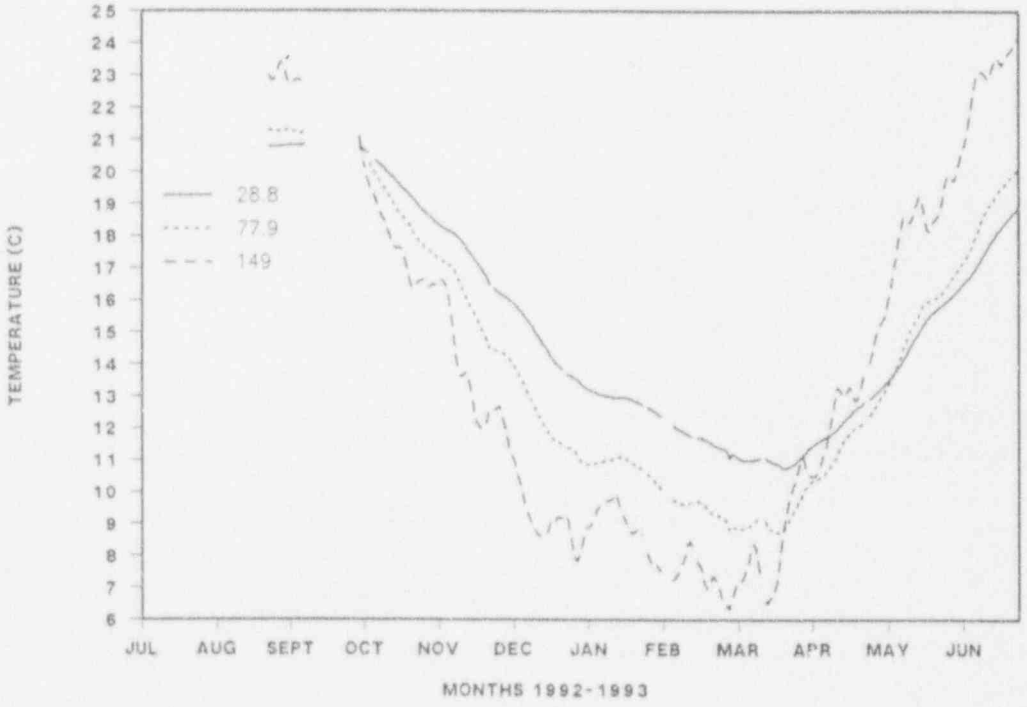

Figure 14. ORNL lysimeter 4 soil temperature.

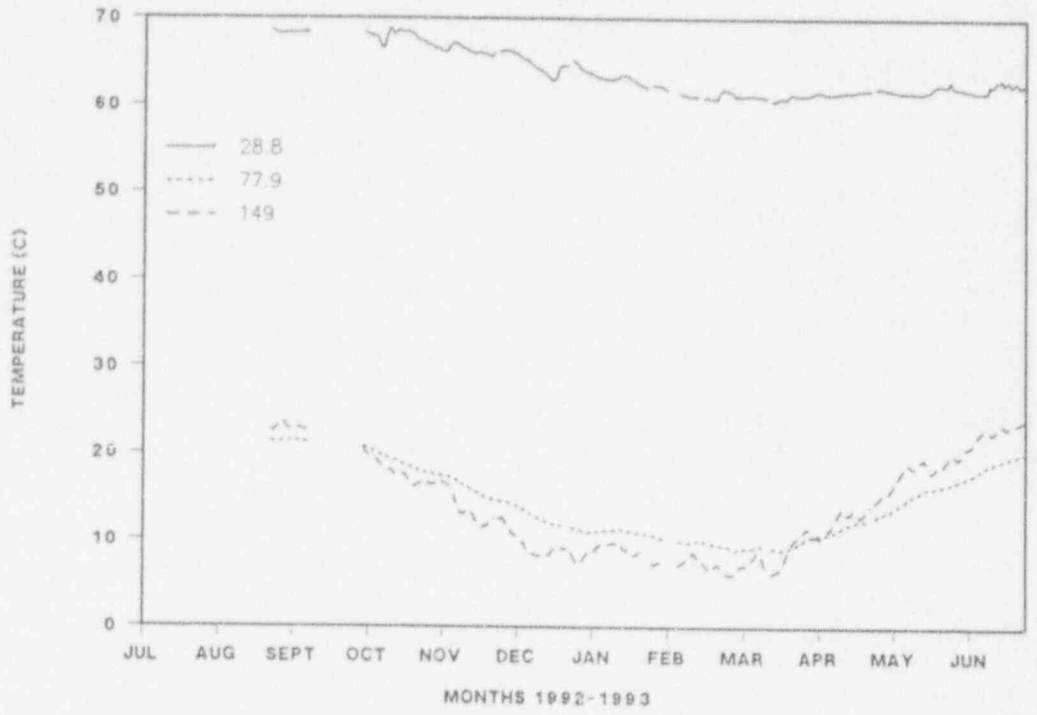

Figure 15. ORNL lysimeter 5 soil temperature.

shown in Figures 16 through 25, indicate that the lysimeter soil columns at both sites have remained moist during the reporting period. The probe output from the soil column of each lysimeter over time (as determined by averaging the outputs of the three probes in each lysimeter) showed that the variation in detected moisture among the lysimeters at each site was relatively similar and not excessive. There was a coefficient of variation (CV) of 37.3% at ANL-E and 20.8% at ORNL. The probes continue to serve their original purpose of providing some indication of lysimeter soil moisture. As was mentioned in the section on soil temperature, some of the probes at ANL-E are no longer functioning. This condition was discussed in a previous report (Reference 10).

Soil moisture in the soil column of the lysimeters at each site is quantified gravimetrically once each year (see Tables 2 and 3). Some idea of the accuracy of the soil moisture probes can be calculated by comparing the once-a-year gravimetric soil moisture data of each soil lysimeter to yearly averaged probe data (Table 4). Percent differences between the gravimetric data and moisture probe data for ANL-E lysimeters range between 6.5 and 31.1%. These values have decreased significantly during this reporting period, and are well within a reasonable range given the use of the information. As in the past, data from the ORNL probes continue to overestimate the actual percent soil moisture.

In addition to using the moisture probe and gravimetric data to calculate soil moisture starting the summer of 1991, a neutron moisture detecting probe was used at ANL-E. Operation of the neutron probe, using 1991 calibration curves, produced data that were comparable to gravimetric overall average values within 4.8%, but underestimated those values (see Table 2). A new calibration curve using 1992 data did not decrease these variations. The variability between actual and measured moisture may be caused by the neutron probe integrating moisture data that were simultaneously measured both inside and outside the lysimeter. It appears that these soils vary in moisture content, with the outside soil being drier. Neutron probe measurements were first made at

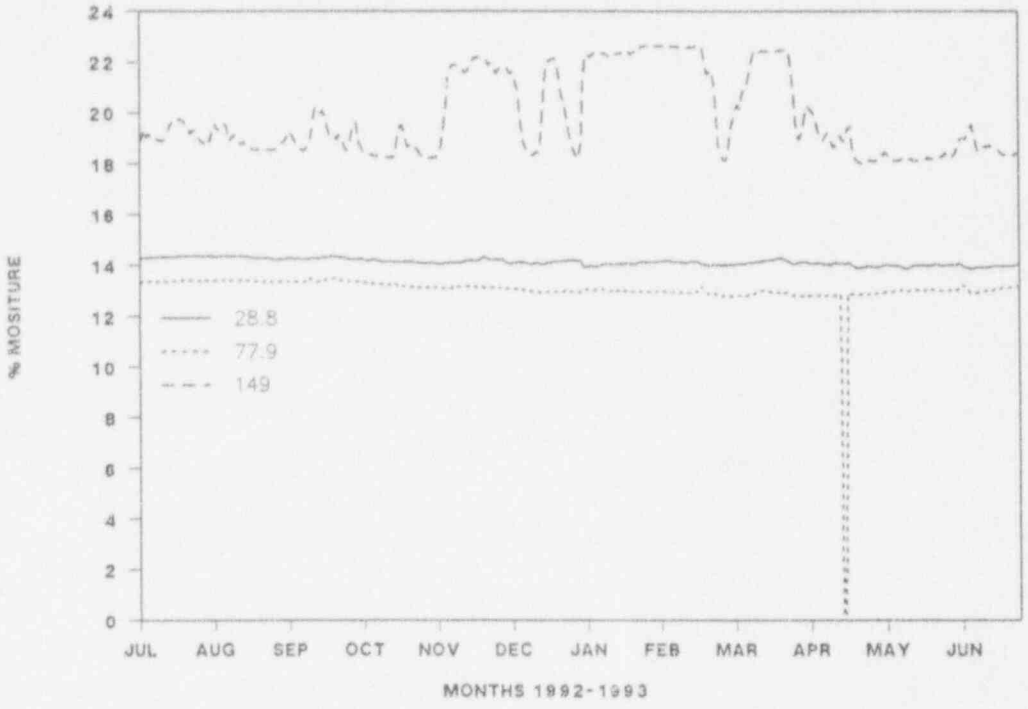

Figure 16. ANL-E lysimeter 1 soil moisture.

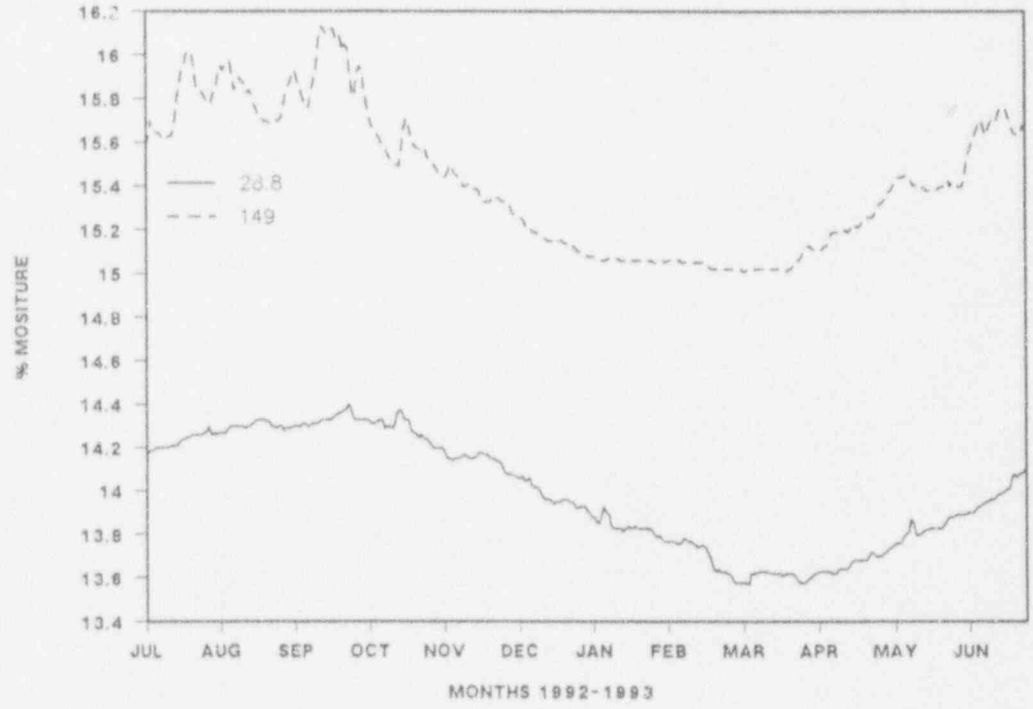

Figure 17. ANL-E lysimeter 2 soil moisture.

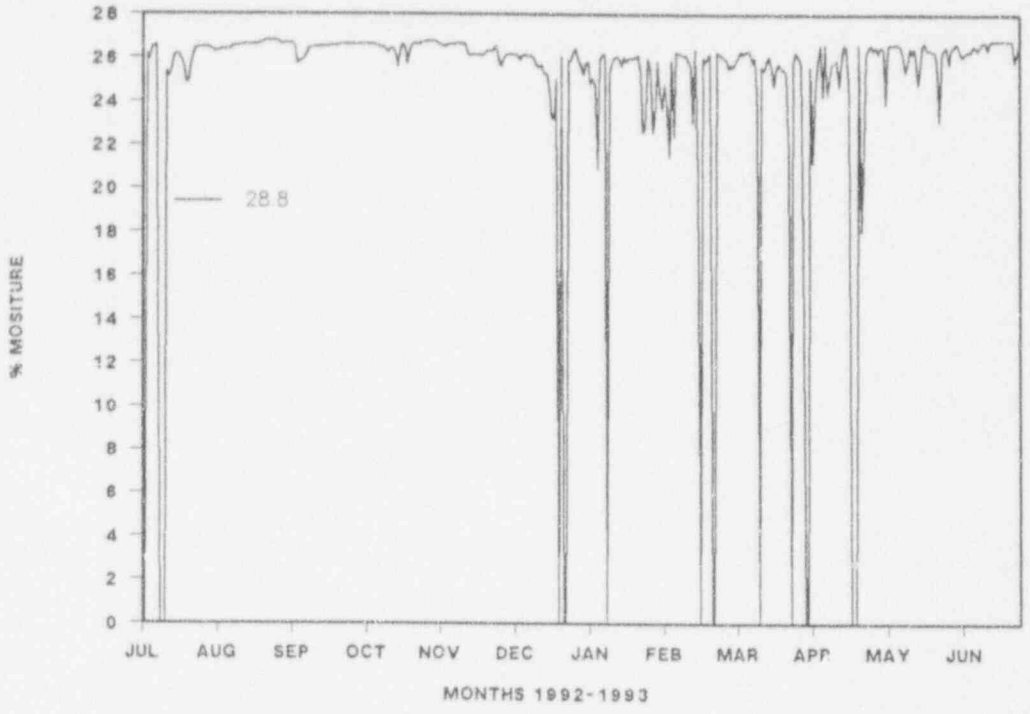

Figure 18. ANL-E lysimeter 3 soil moisture.

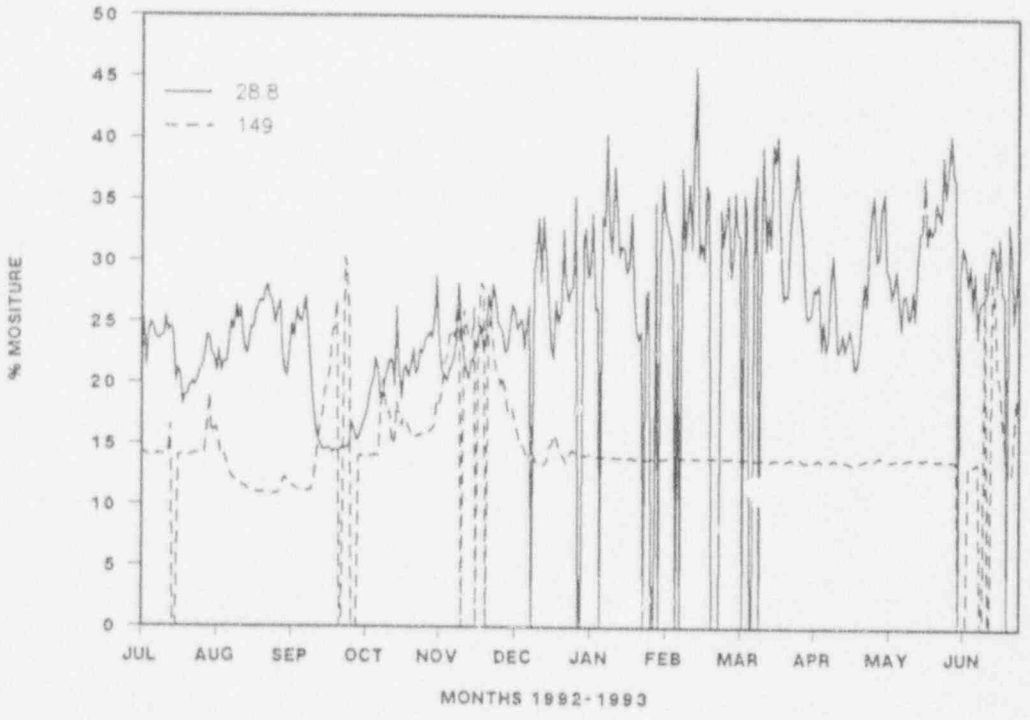

Figure 19. ANL-E lysimeter 4 soil moisture.

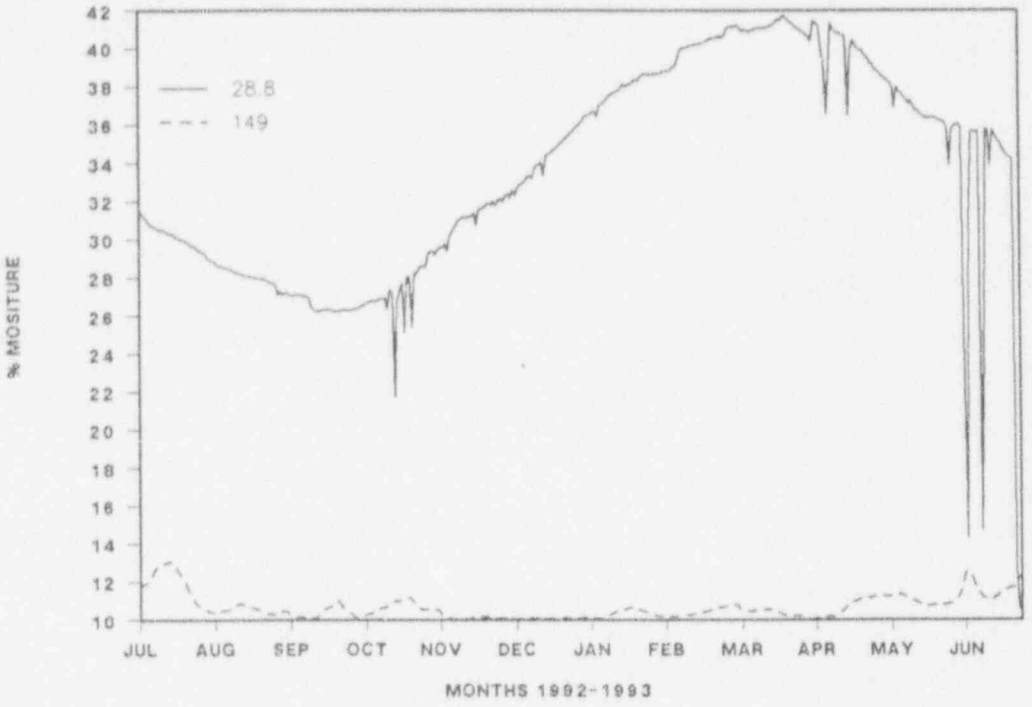

Figure 20. ANL-E lysimeter 5 soil moisture.

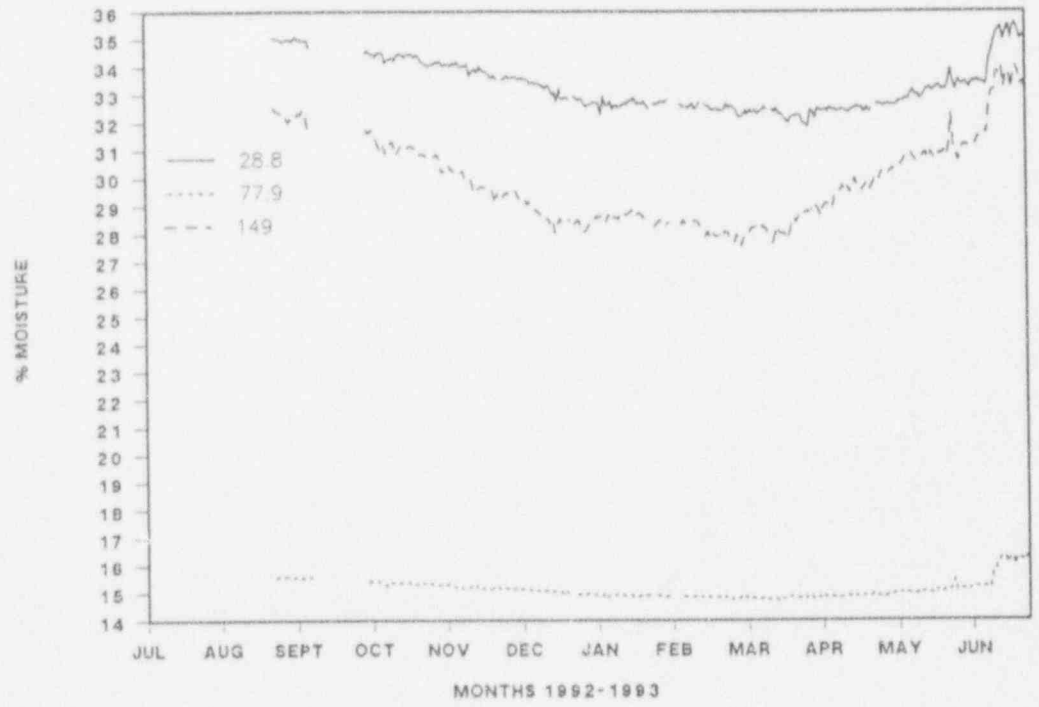

Figure 21. ORNL lysimeter 1 soil moisture.

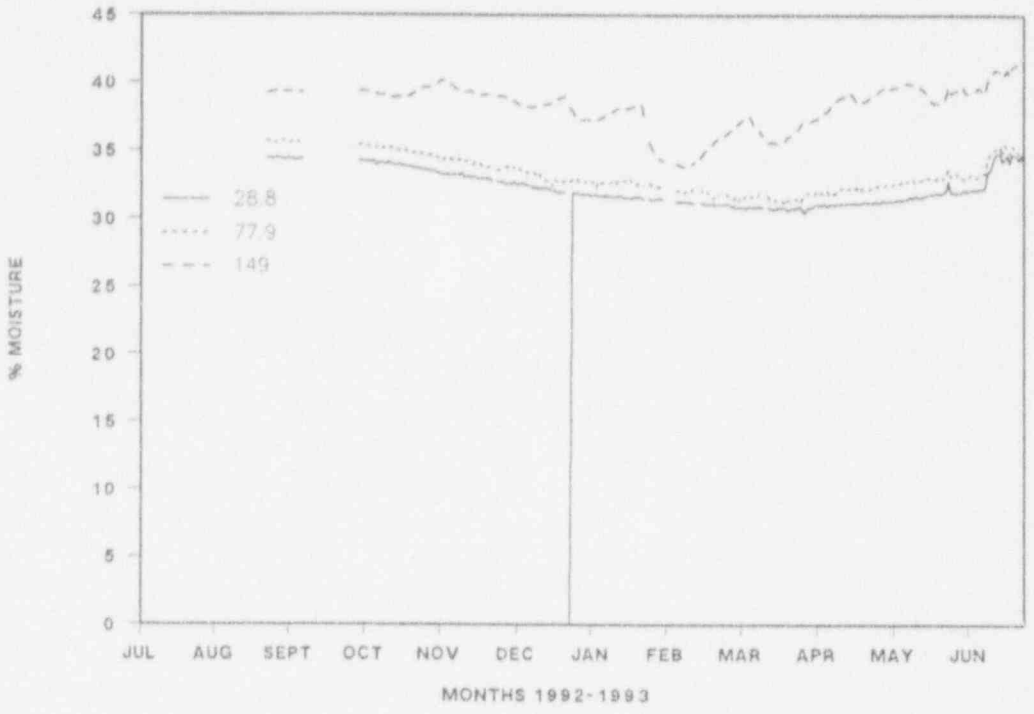

Figure 22. ORNL lysimeter 2 soil moisture.

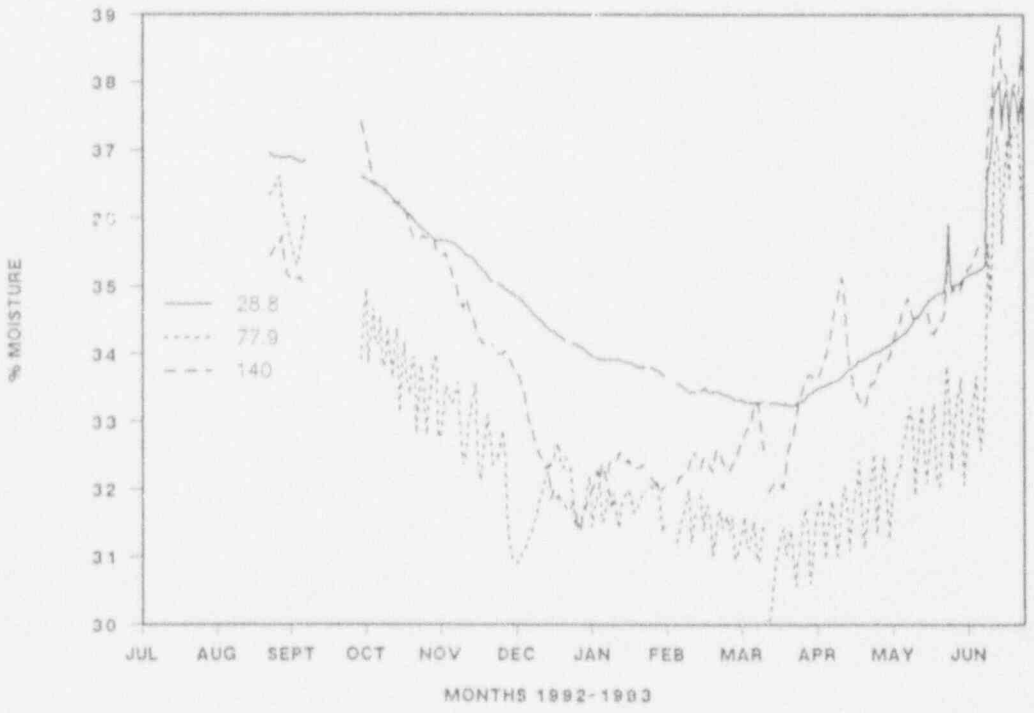

Figure 23. ORNL lysimeter 3 soil moisture.

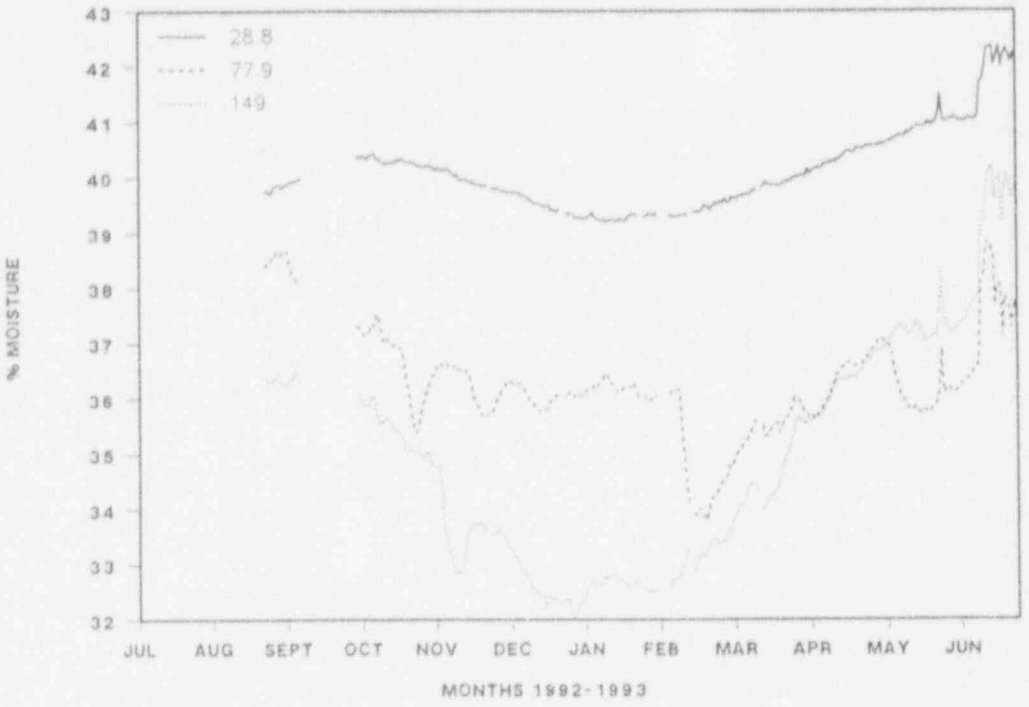

Figure 24. ORNL lysimeter 4 soil moisture.

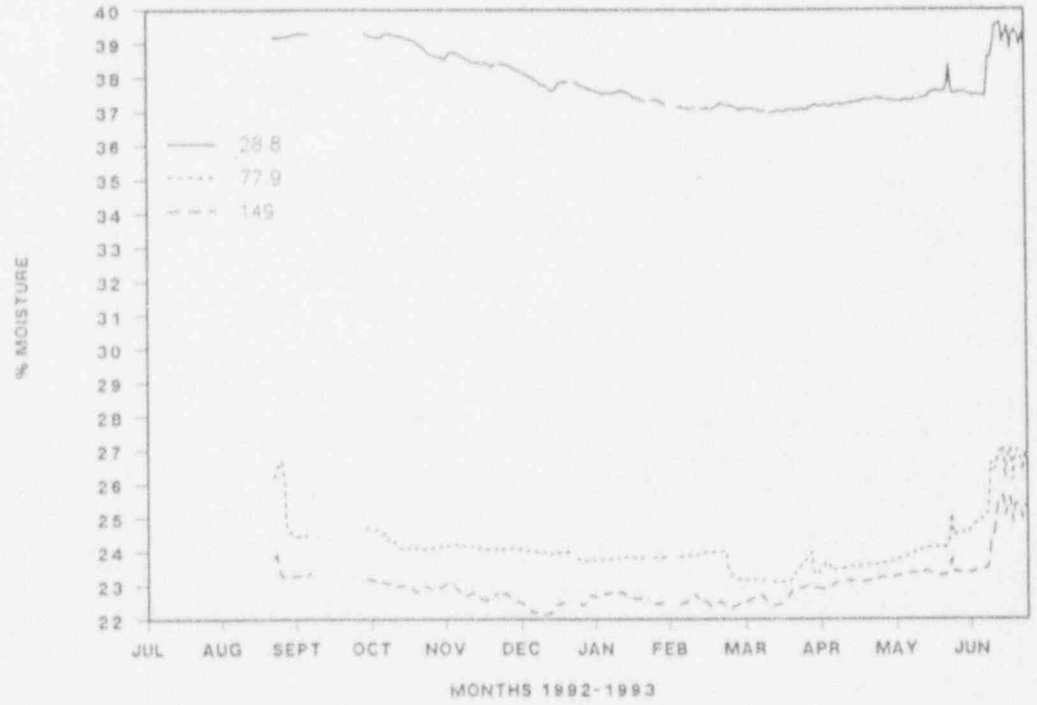

Figure 25. ORNL lysimeter 5 soil moisture.

Table 2. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	% moisture (dry weight) Gravimetric	Neutron probe
1	0–41	15.5	—
1	41–62	22.6	19.9
1	62–85	26.5	20.8
1	82–107	24.0	—
1	107–133	24.2	—
1	133–153	23.6	22.0
1	153–182	22.9	—
1	182–202	22.9	23.3
2	0–41	16.2	—
2	41–62	19.9	19.0
2	62–82	20.1	20.1
2	82–107	21.6	—
2	107–133	22.0	—
2	133–153	22.1	21.8
2	153–182	22.2	—
2	182–202	23.0	24.2
3	0–41	18.2	—
3	41–62	21.9	18.9
3	62–82	24.2	22.3
3	82–107	24.2	—
3	107–133	23.2	—
3	133–153	23.9	22.6
3	153–182	24.6	—
3	182–202	23.6	23.7
4	0–41	20.3	—
4	41–62	25.6	21.2
4	62–82	28.6	22.5
4	82–107	25.0	—
4	107–133	27.0	—
4	133–153	22.9	23.4
4	153–182	24.3	—
4	182–202	24.7	23.8

a. Samples were collected on July 22, 1993.

Table 3. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	% moisture (dry weight) Gravimetric	Neutron probe
1	0–41	12.3	13.0
1	41–62	14.8	15.7
1	62–85	16.3	15.8
1	82–107	16.6	16.3
1	107–133	17.7	15.2
1	133–153	18.0	16.4
1	153–182	18.7	16.8
1	182–202	19.5	17.5
2	0–41	10.0	13.4
2	41–62	13.8	16.2
2	62–82	14.1	16.2
2	82–107	13.5	15.8
2	107–133	14.6	14.2
2	133–153	17.0	17.0
2	153–182	17.4	17.0
2	182–202	16.7	17.6
3	0–41	12.3	17.5
3	41–62	15.0	18.5
3	62–82	16.0	18.7
3	82–107	15.9	18.7
3	107–133	16.9	17.8
3	133–153	17.6	18.6
3	153–182	19.0	18.9
3	182–202	18.0	19.4
4	0–41	12.2	15.5
4	41–62	14.3	19.1
4	62–82	15.4	20.2
4	82–107	15.9	19.8
4	107–133	16.6	17.9
4	133–153	17.8	19.4
4	153–182	18.9	19.9
4	182–202	19.0	20.9

a. Samples were collected on June 16, 1993.

Table 4. Comparison of the average percent moisture values in lysimeter soil column as determined from probe and gravimetric data.

Lysimeter number	Average percent moisture for soil column probes for preceding 12-month period[a]	Average percent moisture for soil column determined gravimetrically for summer 1992	Percent difference between actual and probe
ANL-1	15.7 ± 3.7	22.8 ± 3.2	31.1
ANL-2	15.9 ± 2.1	21.0 ± 2.2	24.3
ANL-3	24.6[b]	23.0 ± 2.1	6.5
ANL-4	17.1 ± 7.0	24.8 ± 2.5	31.0
ORNL-1	26.1 ± 9.6	16.7 ± 2.3	56.3
ORNL-2	34.6 ± 3.2	15.9 ± 1.4	117.6
ORNL-3	33.8 ± 1.0	18.5 ± 0.6	82.7
ORNL-4	37.0 ± 2.6	19.1 ± 1.7	93.7

a. July 1992 through June 1993.

b. Average from one probe.

ORNL in 1992. This year's data are given in Table 3. Comparison of the neutron probe results to gravimetric results, in overall average values, shows that the probe underestimated by 1.7%. In spite of the difference between actual and measured soil moisture at ANL-E, the accuracy appears very good at ORNL. Therefore, it can be said that the use of the neutron probe provides a rapid, accurate estimate of moisture in the soil column.

Soil moisture (as gravimetrically determined) at each sampling depth has remained uniformly consistent between intrasite lysimeters during the past several years (Figures 26 and 27). The uniformity of soil moisture in the ANL-E lysimeters (Figure 26) continues to be of interest given the long-term, nonuniform decrease in water infiltration into the ANL-E soil lysimeters. Lysimeters 1 and 2 appear to have less stored water than 3 and 4 (Table 4). While action to improve drainage of the ANL-E lysimeters was taken early in the experiment, initial drainage rates cannot be restored. Observations of surrounding indigenous soils have confirmed that this soil has a low permeability after being disturbed. Therefore, the present conditions within the lysimeters are indicative of what would be found if a disposal trench were constructed in the same soil. Since FY 1989, no efforts have been made to improve drainage of these lysimeters. Instead, water is no longer allowed to pond on the soil surface. Water in excess of 2–3 cm in depth is now removed from the lysimeter surfaces. Because of less intense periods of precipitation and the resulting drier surface soils, less water ponded on the surface of the lysimeters. During the previous reporting period (Reference 15), all of the soil-filled lysimeters had standing water removed. Water accumulation at ANL-E during the last 12 months occurred in lysimeters 1, 2, 3, and 4. From ANL-1, 495 L of water were removed, 452 L from ANL-2, 76 L from ANL-3, and 393 L from ANL-4.

As shown in Figures 26 and 27, the amount of moisture within the deeper horizons of the lysimeter soil columns at each of the sites appears to have remained fairly constant (see Tables 2 and 3 and References 9, 10, 11, 12, 13, 14, 15). At the time of the 1993 sampling, the average soil moisture of ANL-E soils had increased from 54.7% to 56.3% of the soil moisture holding capacity, while at ORNL, this value remained approximately the same: 38.9% for 1992 and 39.4% for 1993. These values have remained fairly constant from year to year.

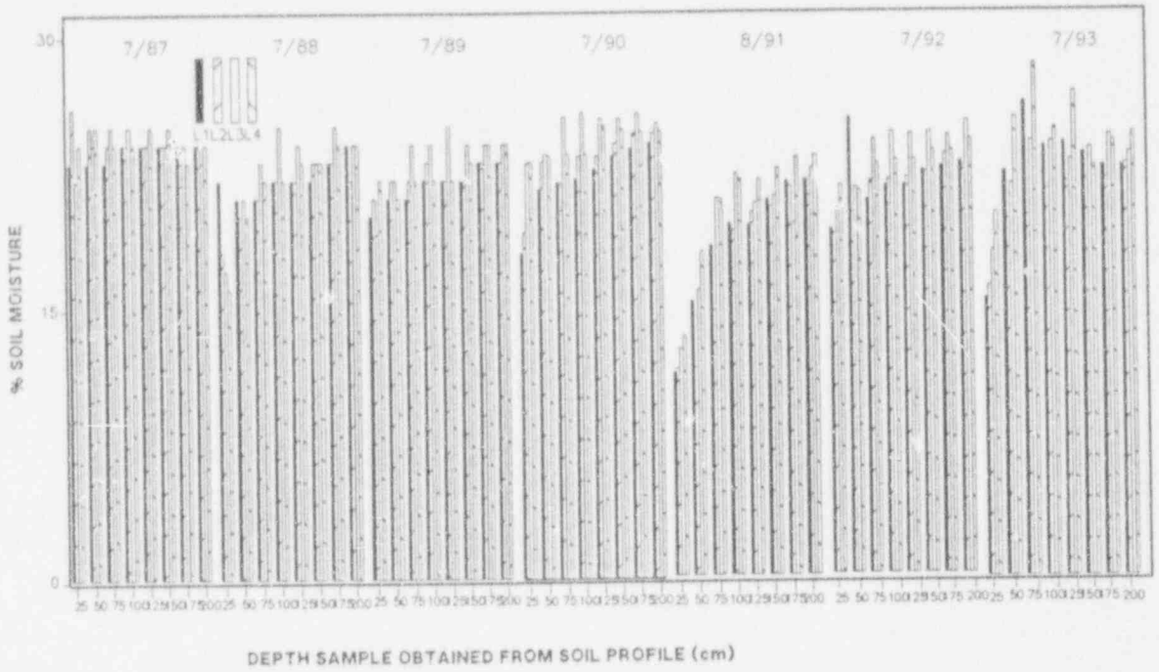

Figure 26. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.

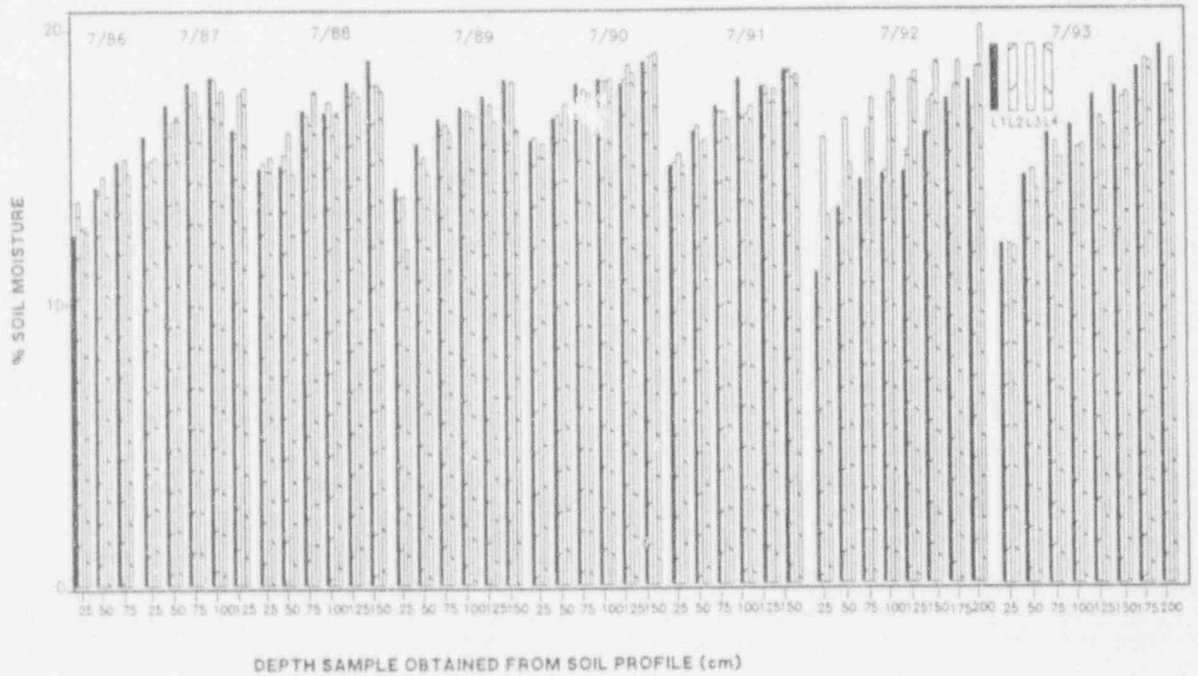

Figure 27. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.

By using the cumulative rainfall data from each site since the lysimeters were placed in operation (Figure 6), it is possible to calculate the approximate volume of water that has been received by the exposed lysimeter surfaces (6,489.5 cm^2). The cumulative volume of precipitation received by each ANL-E lysimeter was 4,815.8 L; at ORNL, this value was 6,739.9 L. Figures 28 and 29 show the volume of precipitation that has passed through the lysimeters. The throughput of precipitation is dependent on site conditions and lysimeter fill material. At ANL-E, an average of 1,929.2 ± 872.8 L, with a range of 29.8 to 65.2% of total precipitation received, has passed through the soil lysimeters, while for the control, this value was 4,829 L or 100.3% of the calculated available precipitation. For ORNL, the values were 6,050.5 ± 45.6 L (89.8%) for the soil-filled lysimeters and 6,910 L (102.5%) for the control. These data are comparable to the previous year's data (Reference 15). Soil in the ORNL lysimeter is more permeable than the ANL-E soils (an observation made by comparing the control lysimeter at each site with that site's soil lysimeters, which are shown in Figures 28 and 29). Also, the small deviation in total yearly leachate throughput with the ORNL soil lysimeters (0.8%) continues to demonstrate that these lysimeters perform as a unit as compared to the individual drainage activity of the ANL-E lysimeters.

The data for ANL-E indicate that there is an increasing disparity in water balances for the ANL-E soil lysimeters. However, a comparison of the total amount of water associated with each of these lysimeters (water removed from the surface plus the quantity of leachate) shows that each of the lysimeters is exposed to equal volumes of water. During the past year, each lysimeter had a total of 612.8 ± 30.2 L (CV 4.9%) of water that was removed as a combination of leachate and standing water. During the previous year, this volume was 283.2 ± 12.8 L (CV 4.5%).

The total volumes of precipitation that have moved through the lysimeters represent an average of 2.74 pore volumes for the ANL-E soil lysimeters and 8.52 pore volumes for soil lysimeters at ORNL, while the controls at ANL-E and ORNL were 10.50 and 10.72 pore volumes, respectively. These data show that the ORNL soil lysimeters have had an average of three times more water pass through them as those at ANL-E.

Radionuclide Analysis. Water samples are normally collected on a quarterly basis from leachate collectors and moisture cups from each of the lysimeters during the 12-month period. At each sampling, only water from the leachate collectors (1 L of collected quantity) and those cups (0.1 L or the noted collected quantity) closest to the waste forms (cups 3) are generally analyzed for gamma-producing nuclides and the beta-producing nuclide Sr-90. The analysis protocol, however, triggers the analysis of water from additional cups in a sequential manner if nuclides are found in a cup 3 sample. For example, when nuclides are found in a cup 3 of a lysimeter, water should be analyzed from cup 1 (directly below cup 3), then cup 4, followed by cup 2 (see Figure 1 for cup placement). Because of funding levels, however, it has not been possible to follow this protocol. During the first five years of operation, water samples from only cups 3 were routinely analyzed at the sites. However, for the past three years, water from cups 1 has also been analyzed and reported. In addition, this year, water from cups 2 was collected during the third quarter for analysis at ANL-E, and cups 5 water was collected during the fourth quarter for analysis at ORNL (Tables 5 and 6, respectively).

Tabulated results of beta and gamma analysis for the liquid samples taken during the period are found in Tables 5 and 6. Four samples were taken at each site during the 12-month period. The cumulative amounts of nuclides as determined in water samples obtained from lysimeter cups 3 and leachate collectors for all sampling periods are displayed graphically in Figures 30 through 36.

As has been reported in the past (References 9, 10, 11, 12, 13, 14, 15), not all nuclides are appearing consistently in either the water obtained from the cups or the leachate collectors. The nuclide that appears with the most regularity at both sites continues to be Sr-90. Consistent, significant, increasing occurrences of this nuclide

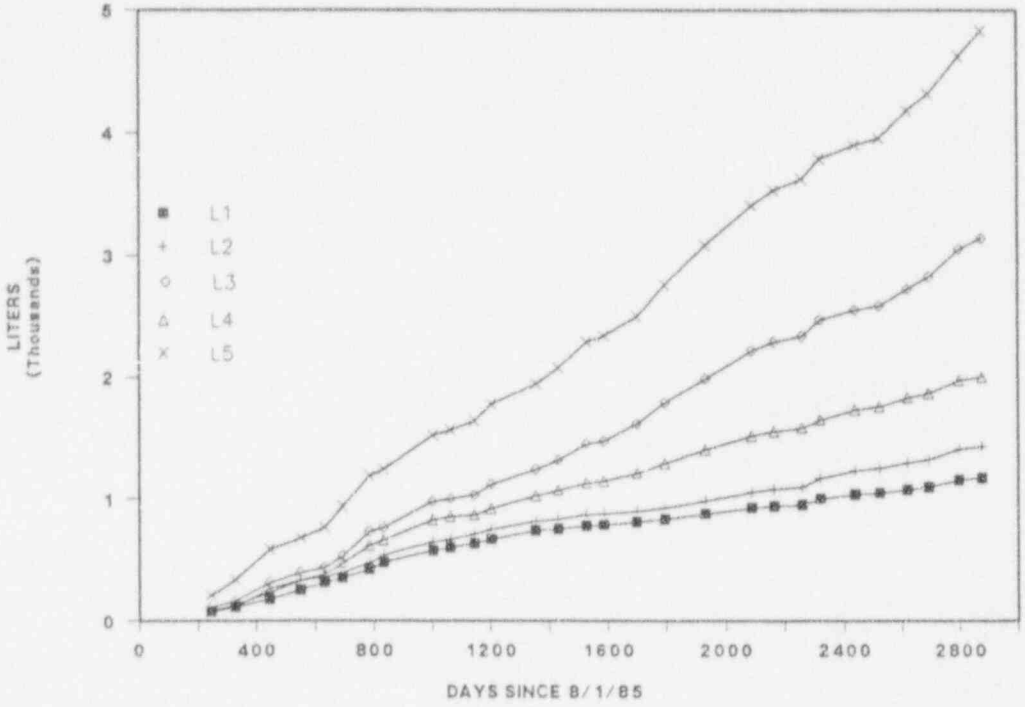

Figure 28. ANL-E cumulative volume of leachate from lysimeters.

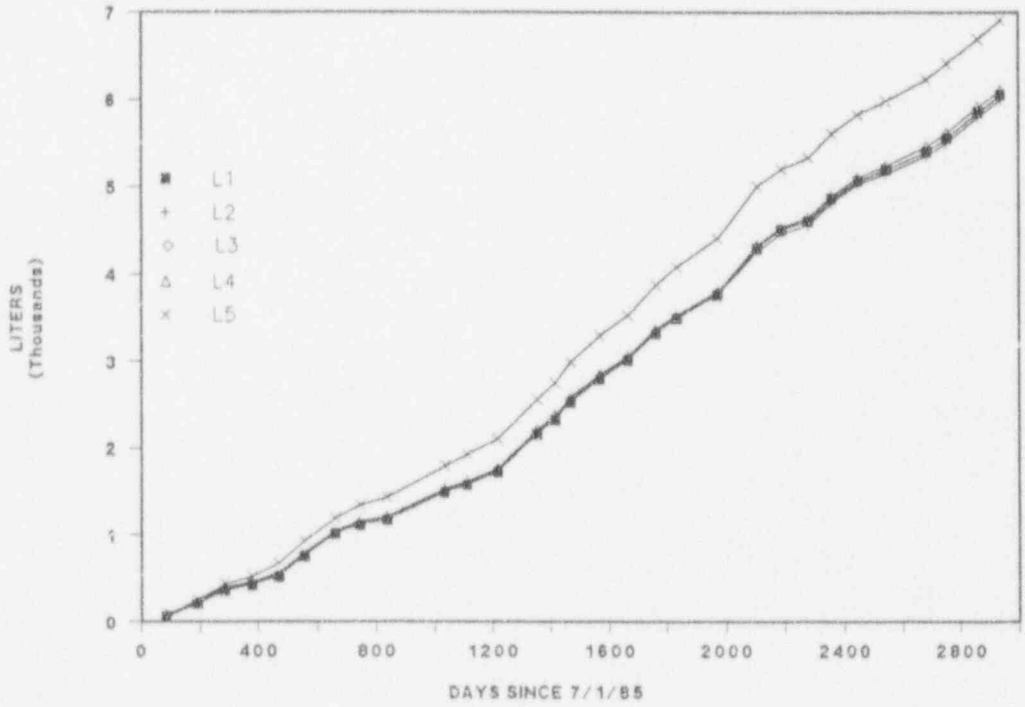

Figure 29. ORNL cumulative volume of leachate from lysimeters.

Table 5. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 8 (1992–1993).

	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
Sample identification	Oct 92	Dec 92	Apr 93	Jun 93	Oct 92	Dec 92	Apr 93	Jun 93	Oct 92	Dec 92	April 93	June 93
Lys 1[b]	0.3 ± 5.7	<5	<5	<5	6.5 ± 2.7	2.7 ± 1.2	<5	<5	3.5 ± 3.5	89 ± 2	1.1 ± 0.4	0.3 ± 0.4
Lys 2	-0.3 ± 3.2	<5	<5	<5	1.1 ± 3.2	<5	<5	<5	3.8 ± 3.5	1.5 ± 0.3	1.3 ± 0.4	0.3 ± 0.3
Lys 3	0.3 ± 6	<5	<5	<5	4.3 ± 5.7	<5	<5	<5	297 ± 27	228 ± 8	232 ± 10	224 ± 3
Lys 4	3 ± 4	<5	<5	<5	4.1 ± 2.4	<5	<5	<5	7 ± 4	5.7 ± 0.3	8 ± 0.5	3.4 ± 0.5
Lys 5	-2 ± 6	<5	<5	<5	3.8 ± 4.1	<5	<5	12 ± 2	1,162 ± 54	1,270 ± 20	910 ± 14	930 ± 18
Lys 1-3[c]	51 ± 114	<5	<5	<5	116 ± 92	<5	113 ± 17	<5	5,946 ± 270	1.7E+4 ± 166	1.4E+4 ± 127	3.0E+4 ± 232
Lys 2-3	8 ± 70	<5	<5	<5	378 ± 81	514 ± 77	338 ± 51	430 ± 20	8,378 ± 270	5,970 ± 85	184 ± 19	4,905 ± 91
Lys 3-3	-24 ± 15	<5	<5	<5	60 ± 146	<5	255 ± 38	32 ± 6	1.30E+4 ± 2.7E+4	1.40E+4 ± 2.3E+4	1.60E+4 ± 2E+4	1.40E+4 ± 2.3E+4
Lys 4-3	-16 ± 76	<5	<5	<5	-16 ± 62	<5	<5	<5	4.9E+4 ± 2,703	3.5E+4 ± 258	3.6E+4 ± 228	4.8E+4 ± 332
Lys 5-3	5 ± 127	<5	<5	6 ± 1	12.0E+4 ± 2,703	12.9E+4 ± 1.8E+4	10.1E+4 ± 1.5E+4	<5	3.2E+4 ± 2703	2.4E+4 ± 260	2.5E+4 ± 239	2.8E+4 ± 370
Lys 1-1[c]	8 ± 32	<5	<6	<5	38 ± 24	<5	<5	15 ± 2	32 ± 16	27 ± 3	14 ± 2	16 ± 2.2
Lys 2-1	-11 ± 30	<5	<5	—[d]	7 ± 17	123 ± 18	<5	—[d]	15 ± 15	88 ± 12	2.6 ± 2.2	—[d]
Lys 3-1	-19 ± 62	<5	<5	<5	5 ± 65	<5	50 ± 7	18 ± 2	3,784 ± 270	3,292 ± 36	6,359 ± 46	4,739 ± 45
Lys 4-1	135 ± 18	<5	<5	—[d]	-35 ± 241	<5	<5	—[d]	186 ± 76	456 ± 29	176 ± 12	—[d]
Lys 5-1	8 ± 70	<5	<5	<5	-3 ± 68	11 ± 2	34 ± 5	43 ± 7	1,459 ± 81	1,619 ± 21	1,158 ± 21	1,045 ± 20
Lys 1-2	—	—	<5	—	—	—	37.3 ± 5.6	—	—	—	6.5 ± 3.3	—
Lys 2-2	—	—	<5	—	—	—	<5	—	—	—	2.6 ± 2.2	—
Lys 3-2	—	—	<5	—	—	—	<5	—	—	—	26.3 ± 3.7	—
Lys 4-2	—	—	<5	—	—	—	15.9 ± 2.4	—	—	—	5.2 ± 3.3	—
Lys 5-2	—	—	<5	—	—	—	223.7 ± 33.6	—	—	—	1,246 ± 19	—

a. Concentration ± 2 sigma.

b. One-L subsample from leachate collector.

c. Total moisture cup sample size is approximately 0.1 L.

d. None detected.

Table 6. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 8 (1992–1993).

	Concentration (pCi/L)[a]							
	Co-60				Cs-137			
Sample identification	Oct 92	Dec 92	Mar 93	Jun 93	Oct 92	Dec 92	Mar 93	Jun 93
Lys 1[b]	0.8 ± 3.5	64.9 ± 5.4	1.6 ± 5.4	0.3 ± 6.8	1.6 ± 3.2	38 ± 5.4	1.4 ± 4.3	6.8 ± 4.6
Lys 2	0.3 ± 4.3	3.2 ± 3.0	3.2 ± 8.4	3.8 ± 7.3	4.1 ± 3.0	6.2 ± 2.7	1.1 ± 9.7	49 ± 11
Lys 3	4.3 ± 3.5	3.2 ± 2.4	4.6 ± 1.6	3.2 ± 13.2	2.7 ± 5.7	13 ± 2.7	–0.3 ± 5.7	16 ± 8
Lys 4	2.4 ± 3.8	5.7 ± 4.9	–7.3 ± 15.1	–5.4 ± 16	0.5 ± 4.3	3.2 ± 5.9	2.2 ± 9.5	22 ± 99
Lys 5	0.3 ± 3.8	4.3 ± 3.5	0.5 ± 11	0.5 ± 12	28 ± 2.7	30 ± 5.4	26 ± 9.5	12 ± 8.4
Lys 1-3[c]	–19 ± 81	–22 ± 70	35 ± 73	14 ± 54	135 ± 60	11 ± 54	43 ± 70	38 ± 62
Lys 2-3	8.1 ± 68	–13 ± 108	49 ± 65	–11 ± 68	–2.7 ± 76	49 ± 50	32 ± 76	2.7 ± 68
Lys 3-3	32 ± 54	19 ± 173	8.4 ± 30	30 ± 68	30 ± 65	–49 ± 208	6.8 ± 23	19 ± 76
Lys 4-3	32 ± 54	57 ± 38	5.4 ± 70	2.7 ± 43	–2.7 ± 70	8.1 ± 100	–30 ± 78	2.7 ± 38
Lys 5-3	22 ± 38	5.4 ± 108	62 ± 62	–19 ± 70	6,757 ± 270	108 ± 62	4,324 ± 270	8,649 ± 270
Lys 1-1[c]	32 ± 54	2.7 ± 70	–38 ± 97	8.1 ± 60	–2.7 ± 70	30 ± 65	22 ± 90	19 ± 60
Lys 2-1	–16 ± 76	19 ± 116	8.1 ± 84	24 ± 57	16 ± 70	2.7 ± 103	78 ± 43	–38 ± 76
Lys 3-1	11 ± 60	13 ± 103	30 ± 97	46 ± 54	35 ± 68	30 ± 78	–16 ± 89	–11 ± 154
Lys 4-1	–8.1 ± 73	38 ± 116	24 ± 87	49 ± 60	–14 ± 78	–2.7 ± 235	16 ± 70	11 ± 73
Lys 5-1	2.7 ± 60	11 ± 84	22 ± 70	8.1 ± 70	78 ± 68	95 ± 57	–30 ± 176	5.4 ± 73
Lys 1-2	—	—	—	21.6 ± 62.2	—	—	—	–5.4 ± 75.7
Lys 2-2	—	—	—	–16.2 ± 73.0	—	—	—	10.8 ± 70.3
Lys 3-2	—	—	—	–29.7 ± 75.7	—	—	—	27.0 ± 62.2
Lys 4-2	—	—	—	37.8 ± 29.7	—	—	—	8.1 ± 29.7
Lys 5-2	—	—	—	8.1 ± 62.2	—	—	—	324.3 ± 54.1

Table 6. (continued).

Sample identification	Concentration (pCi/L)[a] Sb-125 Oct 92	Sb-125 Dec 92	Sb-125 Mar 93	Sb-125 Jun 93	Sr-90 Oct 92	Sr-90 Dec 92	Sr-90 Mar 93	Sr-90 Jun 93
Lys 1[b]	–2.7 ± 7.8	0.3 ± 12	–3.2 ± 11	–5.9 ± 14	324 ± 27	486 ± 27	432 ± 27	486 ± 27
Lys 2	0.3 ± 6.5	1.9 ± 8.4	0.3 ± 18	–3.0 ± 8.6	9.2 ± 3.8	20 ± 6.5	15 ± 3.0	22 ± 6.5
Lys 3	–1.6 ± 12	–0.8 ± 7.6	0.5 ± 17	0.3 ± 8.4	3.8 ± 3.2	10 ± 4.9	3.5 ± 1.9	7.8 ± 4.9
Lys 4	2.2 ± 9.5	–1.1 ± 14	–5.4 ± 26	5.4 ± 12	–0.3 ± 2.3	2.7 ± 3.8	–0.1 ± 1.4	1.6 ± 3.5
Lys 5	–2.2 ± 8.9	0.8 ± 9.2	1.6 ± 21	3.2 ± 15	1,838 ± 54	1,919 ± 81	811 ± 27	865 ± 54
Lys 1-3[c]	—[d]	—[d]	—[d]	—[d]	9.4E+4 ± 2,703	7.5E+4 ± 2,703	6.5E+4 ± 2,703	8.4E+4 ± 2,703
Lys 2-3	—[d]	—[d]	—[d]	—[d]	1.2E+4 ± 207	0.84E+4 ± 270	1.0E+4 ± 270	1.1E+4 ± 270
Lys 3-3	—[d]	—[d]	—[d]	—[d]	17.3E+4 ± 2,703	14.3E+4 ± 2,703	17.8E+4 ± 8,108	23.7E+4 ± 8,108
Lys 4-3	—[d]	—[d]	—[d]	—[d]	6,757 ± 270	5,946 ± 270	7,838 ± 270	7,838 ± 270
Lys 5-3	—[d]	—[d]	—[d]	—[d]	3.2E+4 ± 2,703	60 ± 22	1.0E+4 ± 541	1.4E+4 ± 541
Lys 1-1[c]	—[d]	—[d]	—[d]	—[d]	7,297 ± 270	6,757 ± 270	8,649 ± 270	1,027 ± 270
Lys 2-1	—[d]	—[d]	—[d]	—[d]	214 ± 24	222 ± 35	324 ± 27	351 ± 54
Lys 3-1	—[d]	—[d]	—[d]	—[d]	70 ± 11	103 ± 24	105 ± 16	138 ± 30
Lys 4-1	—[d]	—[d]	—[d]	—[d]	38 ± 11	76 ± 22	30 ± 19	76 ± 30
Lys 5-1	—[d]	—[d]	—[d]	—[d]	70 ± 14	62 ± 22	51 ± 24	108 ± 36
Lys 1-2	—	—	—	–54.1 ± 167.6	—	—	—	37.8 ± 10.8
Lys 2-2	—	—	—	32.4 ± 151.4	—	—	—	29.7 ± 18.9
Lys 3-2	—	—	—	–2.7 ± 162.2	—	—	—	7.0 ± 13.2
Lys 4-2	—	—	—	2.7 ± 73.0	—	—	—	13.0 ± 8.1
Lys 5-2	—	—	—	2.7 ± 170.3	—	—	—	23.8 ± 9.5

a. Concentration ± 2 sigma.

b. One-L subsample from leachate collector.

c. Total moisture cup sample size is approximately 0.1 L.

d. None detected.

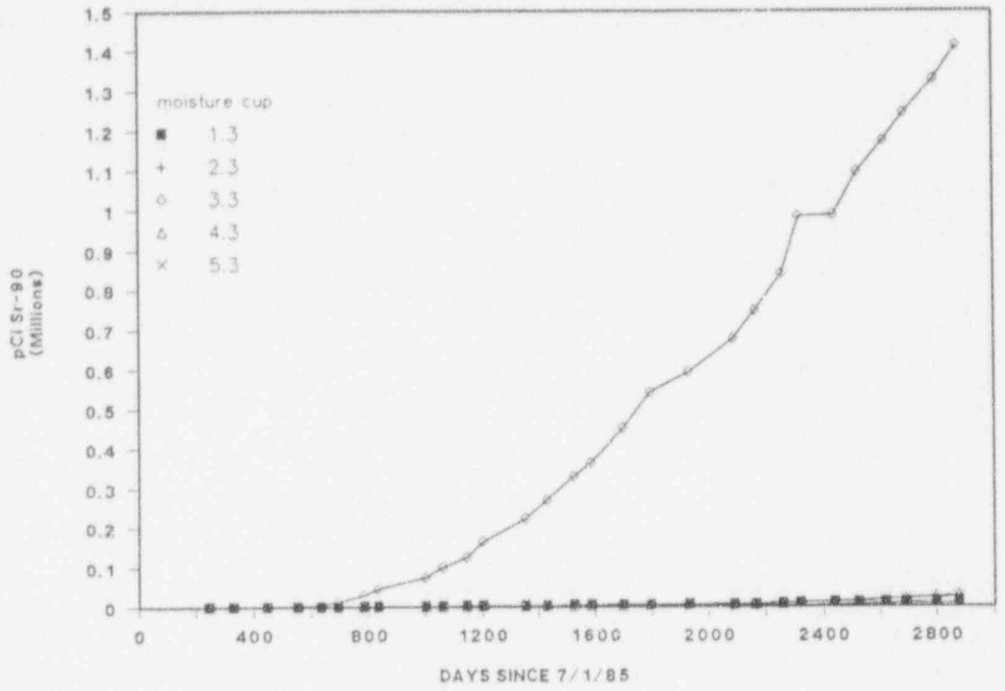

Figure 30. ANL-E cumulative Sr-90 collected in moisture cups number 3.

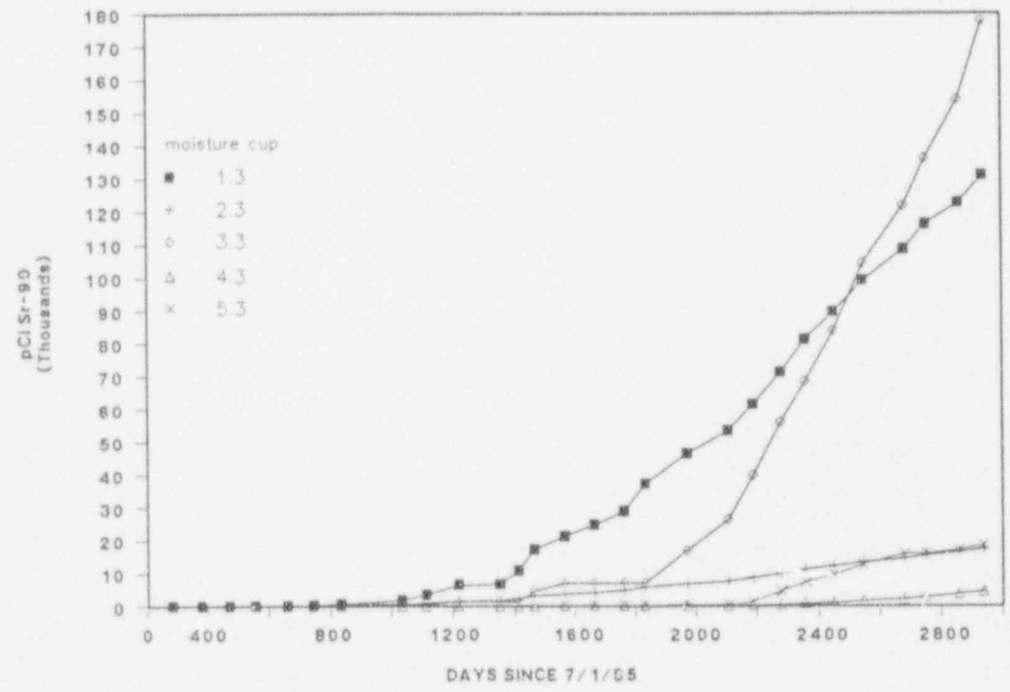

Figure 31. ORNL cumulative Sr-90 collected in moisture cups number 3.

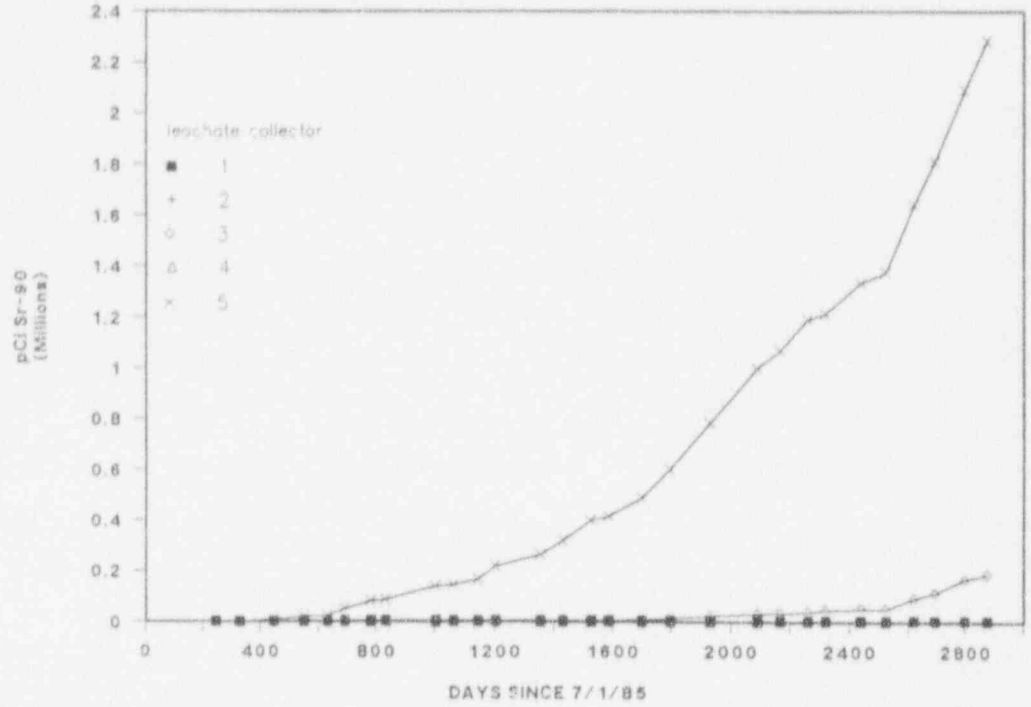

Figure 32. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

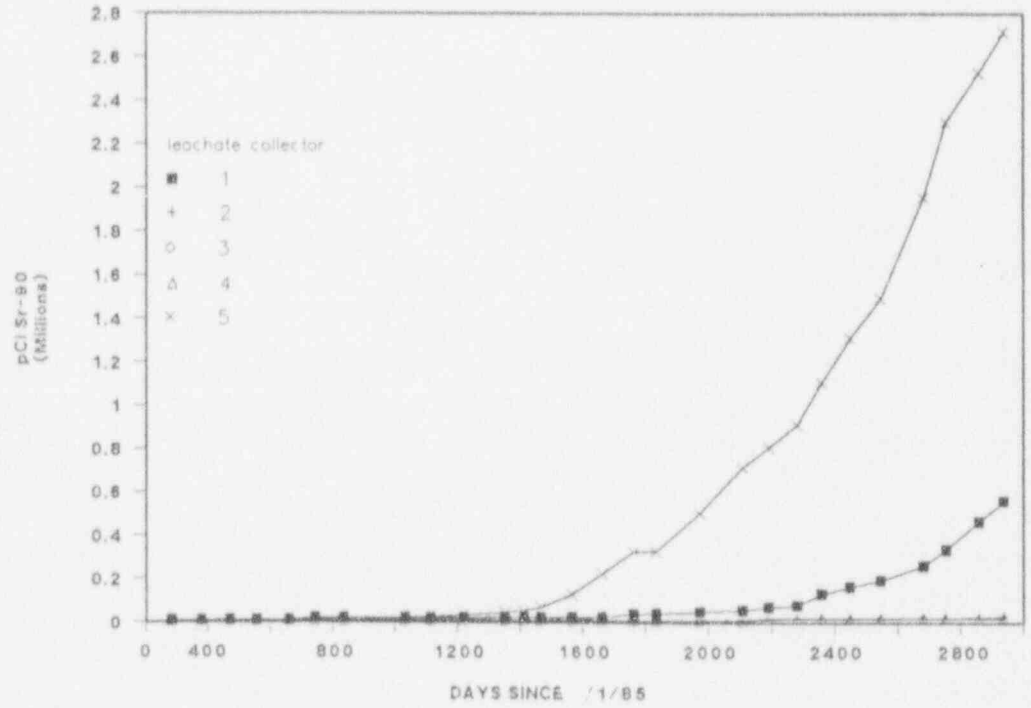

Figure 33. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

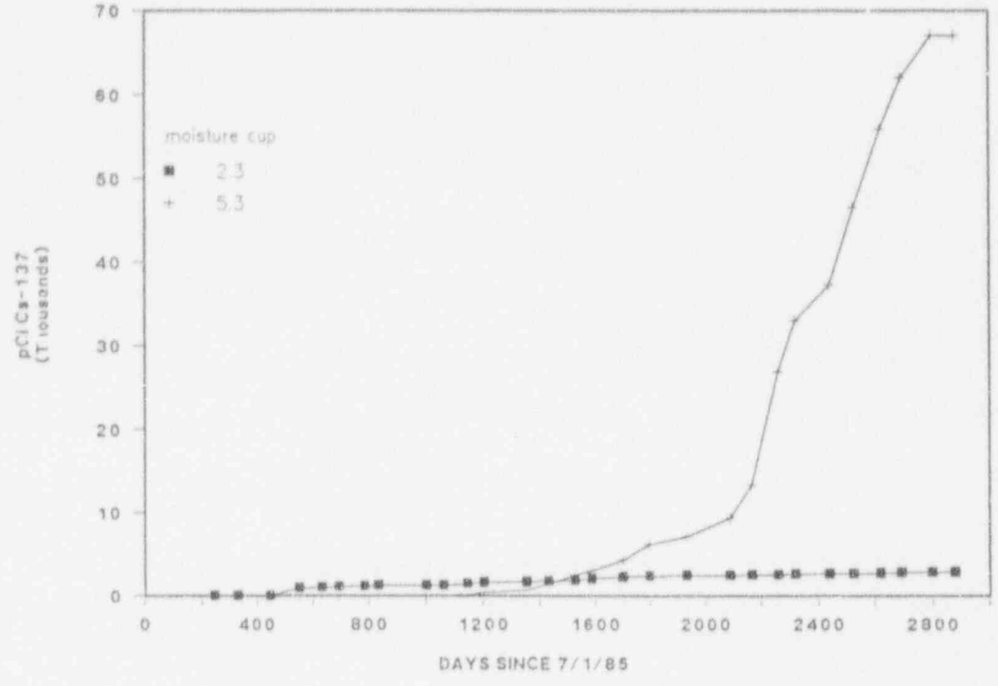

Figure 34. ANL-E cumulative Cs-137 collected in moisture cups number 3.

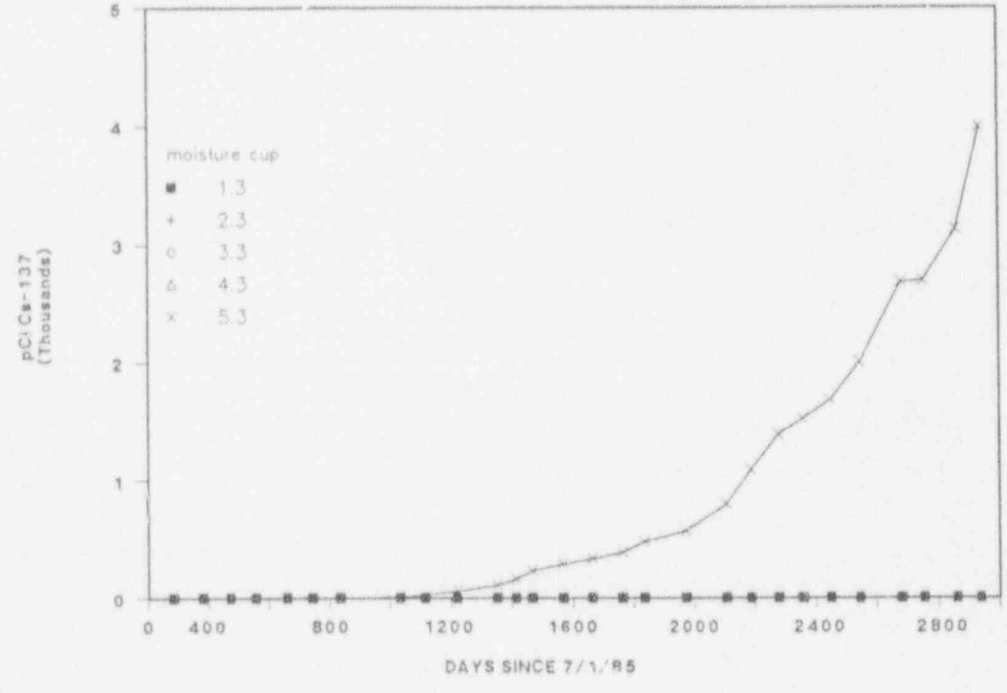

Figure 35. ORNL cumulative Cs-137 collected in moisture cups number 3.

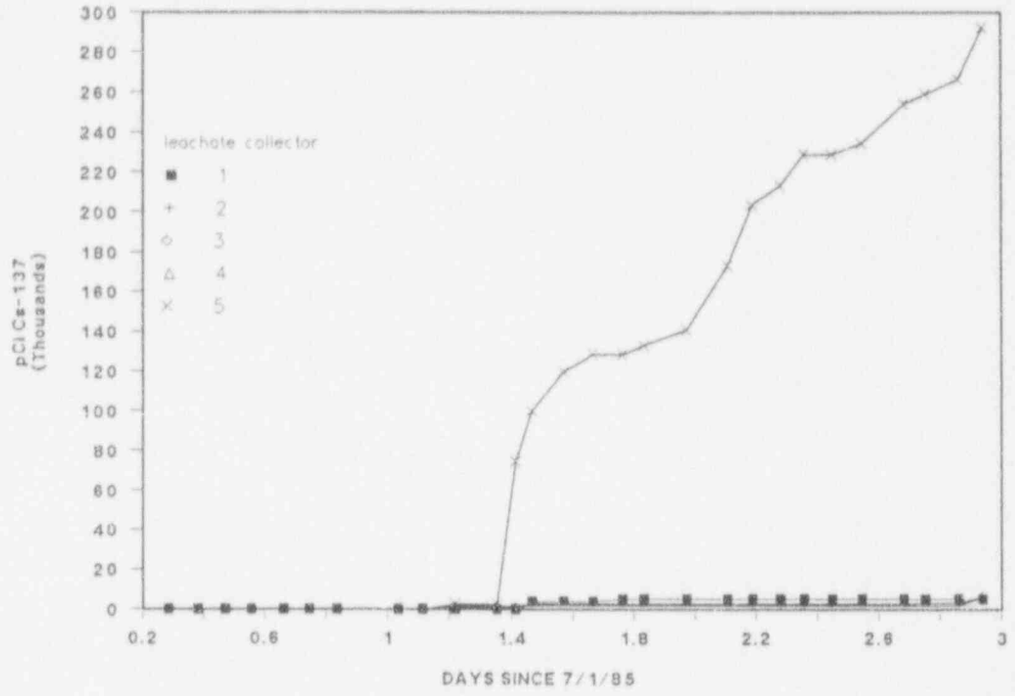

Figure 36. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

continue in all cups 3 at both ANL-E (range of 28 to 129% increase) and ORNL (range of 31 to 179% increase) (Tables 5 and 6; Figures 30 and 31). There continues to be standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a cumulative total of 1,411,575 pCi from 3-3 at ANL-E (an increase of 40% over last year) (Figure 30) and 177,677 pCi from 3-3 at ORNL (70% increase over last year), which is now well above ORNL 1-3 (32%) (Figure 31). The releases into ANL 3-3, ORNL 1-3, and ORNL 3-3 are almost linear, indicating a continuance of an established rate of release. In addition, the increase in Sr-90 release (45% increase) continues in ORNL 5-3 as well as a 179% increase into ORNL 4-3 (Figure 31). The above data show that significant quantities of Sr-90 continue to be transported from the waste forms.

During the past 12 months, amounts of Sr-90 in leachate water from the control (sand-filled) lysimeters at each site have remained similar and at least one order of magnitude larger than the largest cumulative release from a soil lysimeter (Figures 32 and 33). This is comparable to the previous year's findings (References 9, 10, 11, 12, 13, 14, 15). For leachates from soil lysimeters, intersite-comparable percentages of total inventory of Sr-90 were found in ANL-E 1, 2, 3, and 4 and ORNL 2, 3, and 4 (Table 7). There was an increase in the total cumulative quantity of Sr-90 released in the leachate water in all lysimeters at both sites this year (Tables 5 and 6). For ORNL lysimeters 1, 2, and 4, the percent of total inventory of the nuclide released in leachate water was comparable to or greater than that in the cups. These data follow a trend seen over the past 30 months and make it appear that a pulse of Sr-90 could be moving through the soil columns of the ORNL lysimeters. For the control lysimeters at both sites, there was substantially more Sr-90 in the leachate than in cups 3 (two orders of magnitude for ANL-E and ORNL).

The percent of total Sr-90 being measured in the leachate water and cups 3 continues to be somewhat inconsistent between the two sites (Table 7). Perhaps this represents a difference in how the environment at the two sites affects the movement of Sr-90 being released from the waste forms. This difference is also seen when the

Table 7. Percent of total Sr-90 and Cs-137 inventory per lysimeter extracted from moisture cups and leachate water through July 1993.

Lysimeter number	Solidification agent	Percent total inventory Sr-90 × 10^6				Percent total inventory Cs-137 × 10^6			
		Moisture cups		Leachate water		Moisture cups		Leachate water	
		ANL-E	ORNL	ANL-E	ORNL	ANL-E	ORNL	ANL-E	ORNL
1	Cement	79	720	37	3,096	—	—	—	2.0
2	Cement	240	534	62	902	0.9	—	—	0.1
3	VES	5,152	648	691	87	—	—	—	1.5
4	VES	662	98	107	469	—	—	—	0.3
5	Cement	135	557	12,550	82,313	22	0.2	—	107

percent of total Sr-90 found in the leachate water from the two control lysimeters is examined. The percent passing through the ORNL control has increased to 6.5 times that of ANL-E (Table 7).

Gamma-producing nuclides continue to occur with regularity at both sites. ANL 2-3, below a cement waste form containing large amounts of Cs-137, continues to receive significant quantities of Cs-137 (Table 5; Figure 34). Since Cs-137 began appearing in ANL 5-3, the quantity of this nuclide has dramatically increased in each of the sampling periods with significant increases (45% this year) during the last four years (Figure 34). However, no cesium was recovered from the water of this cup during the last sampling. Leachate water from ANL-5 has received sporadic releases this year. There continues to be no sustained occurrence of Cs-137 in any ANL-E leachate water.

Measurable amounts of Cs-137 began to occur in ORNL 5-3 during the May 1988 sample (Figure 35) and have continued in subsequent samplings for a total of 3,994 pCi (100% increase over last year). Detectable amounts of Cs-137 have been consistently found in leachate water from ORNL-5 and sporadically in the other ORNL waters, though none have been found during the past three years (Figure 36 and Table 6). Breakthrough of Cs-137 into the ORNL-5 leachate collector occurred in November 1988, some seven months after its occurrence in moisture cup ORNL 5-3 (Figures 35 and 36). Thus far, a total of 292,324 pCi have passed through to the collector (23% increase this year).

During previous samplings, the presence of both Cs-137 and Sr-90 were discovered at the surface of lysimeter ORNL-5, which is the sand-filled control. Radionuclide activity was first detected during a routine gamma survey of the lysimeter's surface in 1991. At that time, more activity was found near the center than at the edges. Core samples were obtained from the center of the lysimeter at depths from 0 to 2.5 cm and from 2.5 to 5 cm for analysis of Cs and Sr-90. Analysis detected 1,760 pCi Cs-137, 10 pCi Cs-134, and 0.5 pCi Sr-90 per gram of sand in the 0 to 2.5-cm core, and 306 pCi Cs-137, 3 pCi Cs-134, and 0.1 pCi Sr-90 in the 2.5 to 5-cm core material. These data showed that more nuclides were at the surface, suggesting some type of an active deposition mechanism. There remained a question, however, concerning the source of the nuclides. In August of 1992, samples were again taken from the lysimeter and analyzed for Cs-137 and Cs-134. The results were similar to the previous sampling, with 1,533 pCi Cs-137 and 6 pCi Cs-134 being found per gram in the surface, and 574 pCi Cs-137 and 2.4 pCi Cs-134 per gram in the 2.5 to 5.0-cm sample. Last year, a comparison was made between the ratio of Cs-137 and Cs-134 in the surface material and the ratio in the buried waste form. The ratio of the two types of cesium at the surface was 264, and the ratio at 5 cm was 242. Within the analysis uncertainty, the similarity of the two ratios suggests that the source of the nuclides was the same. To determine if the waste form was the source of the nuclides, the present ratio of these nuclides in the waste was calculated by using the standard radioactive decay equation. Based on waste history, the calculated cesium ratio in the waste form was 252. The ratio of cesium in the waste form (which would change only due to time or if there were an alternate source of cesium) is for all practical purposes the same as that of the cesium detected on the surface material. Therefore, it was concluded that the surface contamination of cesium came from the waste form. Measurement of Cs-137 in cup 5, the upper cup (Figure 1), shows a presence at that location in June (Table 6).

If the cesium at the surface migrated from the waste form, and it appears that it did, then it is important to find out how this nuclide migrated more than 1 m upward. Cesium tends to be sorbed much like potassium to clays or other sorptive material. Therefore, it would be expected that both the free unassociated cesium ions and the particles to which they could sorb would be washed downward away from the waste form during periods of water infiltration. Data on the occurrence of cesium in the leachate from lysimeter ORNL-5 seems to confirm that assumption (Table 6; Figure 36). However, since the fill material in the lysimeter is a fine-to-medium-grained silica sand

with a very low cation exchange capacity, a case can be made for cesium migrating as a solute in the pore water, which could move upward due to a wicking effect caused by evaporation. It is not likely that extensive evaporation is a regular occurrence, since the quantity of water moving through this lysimeter accounts for ~90% of the amount of precipitation that falls on the lysimeter surface. However, ORNL has experienced extended periods (three or more weeks) of hot weather with no rainfall during the summer months. Evaporation from the surface, enhanced by increased temperature, could result in an upward flux of water. Of course, any solute carried by this water would be left behind as a residue on the surface. The presence of wind-accumulated clays and organic matter on the sand surface could then fix the cesium and prevent its reentry. Planning to determine the mechanism of this unexpected cesium movement is underway. A sand core will be extracted and examined in FY-94.

For the fourth year in a row, Sb-125 has not been found in ORNL-5 leachate water. Also, this is the fifth year of its absence in ORNL cup 5-3.

By using a matrix (as in Table 7), several comparisons can be made based on the intra- and intersite data. Overall, of the nuclides contained in the waste forms (Reference 8), a greater recovery of Sr-90 has occurred in terms of quantity and percent of inventory than of other nuclides. Next is Cs-137, followed by Sb-125 and Co-60 (not listed in Table 7). Compared to Sr-90, the recovery of Cs-137 appears insignificant. There have been significant occurrences of Cs-137 in cups 3 of the ORNL soil lysimeters during past years, and there was evidence of its reoccurrence in ORNL 1-3 (Table 6). On the other hand, this nuclide has been consistently occurring in ORNL 5-3 (Figure 35) and in the leachate collector of the ORNL-5 lysimeter (Figure 36). Cesium-137 has also occurred in the moisture cups of ANL-E lysimeters 2 and 5 but not in the leachate water. More Cs-137 has passed through the ORNL lysimeters than those at ANL-E.

At ANL-E, a comparison of Sr-90 occurrence in cups 3 and the leachate collectors (Table 7) contrasts the difference between movement of the nuclide away from the waste form into the bulk water solution versus its transport with the water through the soil column. This behavior might be influenced by the amount of water passing through the ANL-E lysimeters (Figure 28). However, a lack in uniformity is also seen with the ORNL data (Table 7), and these lysimeters have larger quantities of water (up to five times as much), with more uniform unit-to-unit movement (Figure 29).

As seen from Table 1, the lysimeters at both sites have been loaded with waste forms based on solidification agent and total nuclide content. Numbers 1, 2, and 5 were solidified with cement; numbers 3 and 4 with VES. ANL-1, -3, and -5, and ORNL-1 and -3 contain 5% of activity as Sr-90; the others contain 1% of activity as Sr-90 (Reference 8). This provides a total of five matched sets for the sites (ANL-1 and -2, ANL-3 and -4, ORNL-1 and -2, ORNL-3 and -4, and ANL-5 and ORNL-5). It could be assumed that nuclide leaching from these waste forms would be proportional to content, i.e., those with the higher loading would have proportionally larger Sr-90 releases, but the total percent of release should be close to the same. The first part of this assumption appears to be correct in the case of Sr-90 movement into cups 3 for both sites when compared to other cups at that site (Table 7). Figures 30 and 31 show that cumulative total quantities of Sr-90 in water retrieved from cups 3 are higher from the lysimeters with the higher loaded waste forms (range of 34 to 4,637% more) (Figures 30 and 31). The same was also true for the four soil lysimeters when the quantity of Sr-90 in leachate water is compared (13 to 3,825%). So it appears that there is a general trend for more Sr-90 to be removed from the higher loaded waste forms with a subsequent movement through the soil column. The assumption of a uniform percent release of Sr-90 from the waste forms, however, is not supported by the data (Table 7). For the moisture cup soil water collection, three of the five sets have a higher total percent released to the cup water from those lysimeters containing the higher loaded waste forms (35 to 678%), while two of the five have the higher Sr-90 released to the leachate water (243 and 546%). However, the other sets were the reverse of this.

A greater percentage of Sr-90 continues to be found in ANL 3-3 and ANL 4-3 (which both contain VES waste forms) than in the other ANL-E cups 3 (Table 7). As has been noted, the length of the soil column appears to moderate the quantity of the nuclide that travels from the waste form to the leachate collector. The leachate collectors in those same ANL-3 and -4 lysimeters also receive a higher percentage of Sr-90 than the other ANL-E collectors but a significant amount less than the cups 3 (13 and 16%). The percent of available nuclide that continues to move into the leachate of ANL-5 is much greater than that of the other ANL-E lysimeters (1,716 to 33,819%), thus providing further evidence of the moderating effect of soil.

Greater quantities of Sr-90 are moving through the ORNL lysimeters in comparison to the ANL-E lysimeters. Once again, there appears to be no correlation between the type of waste form and the amount of nuclide recovered in the leachate collector. About 0.082% of the Sr-90 contained in ORNL-5 has now been recovered in leachate from that lysimeter. The percent of available Sr-90 that has moved into the ORNL-5 leachate collector remains significantly higher than the other ORNL collectors (2,559 to 94,510%).

Recovery of Sr-90 in the ORNL cups is comparable for those lysimeters containing the cement waste forms and one of the two containing VES waste forms. These data together with those from ANL-E continue to indicate that cement and VES have comparable releases.

On an intersite comparison, it can be seen that larger quantities of Sr-90 and Cs-137 are moving in the ORNL lysimeters (Table 7). Soil type and precipitation (environmental factors) appear to be the controlling factors.

Use of Lysimeter Data for Performance Assessment and Source Term Calculations. It is becoming apparent, through operational experience and cumulative data provided by the NRC lysimeter array during the past eight years, that lysimeters are a valuable source of data used in the development of site-specific performance assessments. The operational lysimeters are providing continuous data from the near-field (that area comprised of the waste form and surrounding soil). These data directly relate to waste form stability. Information that can be obtained from the data includes the mass balance of released constituents, the solubility of radionuclides in a site-specific geochemical system, as well as the retardation or dispersion of released constituents during transport to the far-field. Also, soil-pore water chemistry (inorganic and radioactive constituents), soil mineralogy, soil water/mineral mass ratio, net infiltration rate, soil profile moisture and temperature, porosity, hydraulic conductivity, and dispersiveness are being or could be extracted from lysimeter output. Such data are invaluable as input into source term and performance assessment codes since they represent a field data set, which contains complete information that characterizes environmental, hydrogeological, geochemical, and waste form effects.

The relationship between input parameters for codes and data derived from lysimeter operation is compared in Table 8. The data could be used in such codes as PATHRAE,[20] PRESTO,[21] and others to predict the stability of waste forms for a 300-year period of time.

Source term code studies were performed using the data produced through FY-93 by the ANL-E and ORNL field experiments. A brief summary of the pertinent characteristics of the lysimeters is in order. At each site, four of the lysimeters are filled with soil while the fifth control lysimeter is filled with Unimin silica oxide sand. At ORNL, the soil used is from the C horizon of a Fuquay sandy loam from the Savannah River Plant adjacent to the Barnwell facility in South Carolina. ANL-E lysimeters are filled with a local soil that represents a typical Midwestern type. It is a morley silt loam with the surface layer removed. Each lysimeter is filled with seven cylindrical waste forms measuring 4.8 cm in diameter and 7.6 cm in height. They are stacked one on top of the other in the lysimeters forming a height of 53.2 cm and a volume of 1 L. The waste forms were solidified in either vinyl ester-styrene or Portland Type I–II cement. The waste streams included two resin types. Type I was a

Table 8. Relationship between performance assessment code parameters and lysimeter data.

Code parameters		Data collected from lysimeters
Q = Inventory		Known inventory is introduced by experimental design
P = Annual percolation		Amount of rainfall on lysimeter; amount of evapotranspiration
S = Fraction of saturation		Soil moisture content
V_s = Water velocity		Mass or volume of effluent water per unit time
R = Retardation factor		Mass or volume of effluent solute per unit time relative to V_s
d_s = Soil bulk density		From experimental design of lysimeter
P_s = Effective soil porosity		Can be estimated for saturated conditions from mass of effluent water, volume of soil, soil bulk density
I_r = Inventory released		Radionuclide concentrations in soil pore water and in effluent
V_w = Trench volume		From experimental design of lysimeter
Cw = Radionuclide concentration		Radionuclide concentration in effluent
Mi = molality		Effluent concentrations
MIN = Minerals dissolved or precipitated		From mineralogical characterization of soil at end of experiment

mixture of synthetic organic ion-exchange resins (phenolic cation, strong acid cation, and strong base anion). Type II resin was a mixture of synthetic ion-exchange resins (strong acid cation and strong base anion resins) with inorganic zeolite. Each lysimeter is equipped with five moisture collecting cups and three soil moisture/temperature probes, which are located at various elevations in the lysimeter (Figure 1) along with a leachate container located at the bottom of the lysimeter (Reference 8). Below the fill material, a layer of filter fabric was placed between the soil or sand and the gravel bed. A gravel bed is located below the filter fabric. The height of the gravel bed was set to 10 cm in these modeling studies. The data used in this study were collected from moisture cup 3, located approximately 23 cm from the bottom of the waste forms, and from the lysimeter leachate collector, located 61 cm below the bottom of the waste forms. The radionuclides found to date in the leachate waters have been primarily Cs-137 and Sr-90.

The Disposal Unit Source Term (DUST) code was used to model the release of Cs-137 and Sr-90 from the lysimeter waste forms. DUST is a one-dimensional code that can model release by a finite difference method or by a mixing cell cascade approach, and has the ability to simultaneously model three different types of release mechanisms: diffusion, dissolution, and surface rinse. The mixing cell model is limited in that it does not take diffusional release into consideration. Therefore, for these simulations, the finite difference model was selected because it is more flexible and capable of handling a variety of different parameters. A further description of the models in the code is given in Reference 22.

Lysimeters 5 at ORNL and ANL-E were chosen for study of the release of Cs-137 and Sr-90 from Portland Type I–II cement because releases from other lysimeters were substantially lower and the data were not sufficient to model. At ANL-E, lysimeter 5 contained resin waste type I solidified in cement; at ORNL, lysimeter 5 contained resin waste Type II, which was also solidified in Portland Type I–II cement (see Table 1). Diffusional release is believed to be the controlling mechanism for a cement-solidified waste. The waste form diffusion coefficients for Portland Type I–II cement were presented in Reference 17. Measured values were 9.6E-10 cm^2/s for Sr-90 and 5E-11 cm^2/s for Cs-137. The Darcy velocities ranged from 2.59E-6 cm/s at ANL-E to 3.6E-6 cm/s at ORNL (Reference 14). The soil bulk density values were 1.55 g/cm^3 at ANL-E and 1.60 g/cm^3 at ORNL (Reference 8). Moisture content values were calculated using the effective soil porosity and the fraction of saturation values found in Reference 11. In lysimeter 5 at both sites, the moisture content was calculated as 21%. The

distribution coefficients have not been measured for Sr-90 or Cs-137; therefore, they were estimated by fitting the model predictions to the data. The cumulative leachate activity collected from the lysimeters over the first seven years of the experiment, which was used to make comparisons to the DUST code predictions, represented 0.045% and 0.008% of the total inventory of Sr-90 in lysimeters 5 at ORNL and ANL-E, respectively. At ORNL, the collected amount represented less than 8.6E-5% of the Cs-137 inventory in lysimeter 5 while nothing has been collected in ANL-E lysimeter 5 (Table 9).

Concentrations and predicted releases were matched to moisture cup 3 and the lysimeter leachate collector. The concentrations and releases were taken at 23 and 51 cm below the waste forms. In this report, the cumulative leachate activity collected 51 cm beneath the waste form is used as the performance measure. Initial amounts of Cs-137 and Sr-90 varied at ORNL and ANL-E because the control lysimeters contained different resin types. In ORNL lysimeter 5, the type I waste form had a total initial inventory of 3.29 E-3 Ci of Sr-90 and 1.432 Ci of Cs-137 (Reference 8). The type II waste form at ANL-E had a total initial inventory of 1.84E-2 Ci of Sr-90 (Reference 8). Cesium-137 was not modeled at ANL-E for lack of sufficient releases.

The cumulative activity collected from the lysimeters is less than 5E-2% in comparison to the total inventory for Sr-90 and less than 9E-5% for Cs-137 (Table 9). Therefore, either the waste form release rates are much lower than anticipated, or transport processes are controlling release through the soil column. At that level, it is possible that random fluctuations (noise) are being seen, and release patterns may not develop for several more years.

Three parameters are known to strongly influence release through the soil column. They are distribution coefficient (K_d) and dispersivity, which together control transport from the waste form through the soil column, and waste form diffusion, which controls waste form release rates. Several cases were modeled where either K_d, dispersivity, or waste form diffusion coefficients were varied to best match the actual release data from the lysimeters.

An exponentially decaying waste form release rate of 1.75E-6 exp (-1t) Ci/yr was chosen, where (1) is the decay constant for Sr-90 and (t) is the time; also chosen were a dispersivity of 10.5 cm and K_d values of between 4.5 and 4.8 (Figure 37). In doing so, a very good fit to the data was obtained, although the parameters used are highly unlikely. The waste form is releasing approximately 0.01% of inventory per year, i.e., 0.07% over seven years. The experimentally measured release from lysimeter 5 at ANL-E was 0.007%.

The domain of the model was extended to 52 cm below the waste form. This ensures that boundary conditions (BCs) will not significantly affect the predicted concentrations. Therefore, the results in Figures 38 and 39 are obtained using a bottom BC of zero dispersive flux. A concentration trace continued to be taken at the location of the filter fabric, which is 51 cm below the waste form.

Table 9. Total and collected Ci amounts of Sr-90 and Cs-137 in lysimeter 5 through July 1992.

	Total amount (Ci)	Amount collected (Ci)	Percent collected
ORNL Cs-137	1.432	0.23E-6	8.6E-5
ORNL Sr-90	3.39E-3	1.6E-6	4.5E-2
ANL-E Sr-90	1.84E-2	1.4E-6	7.6E-3

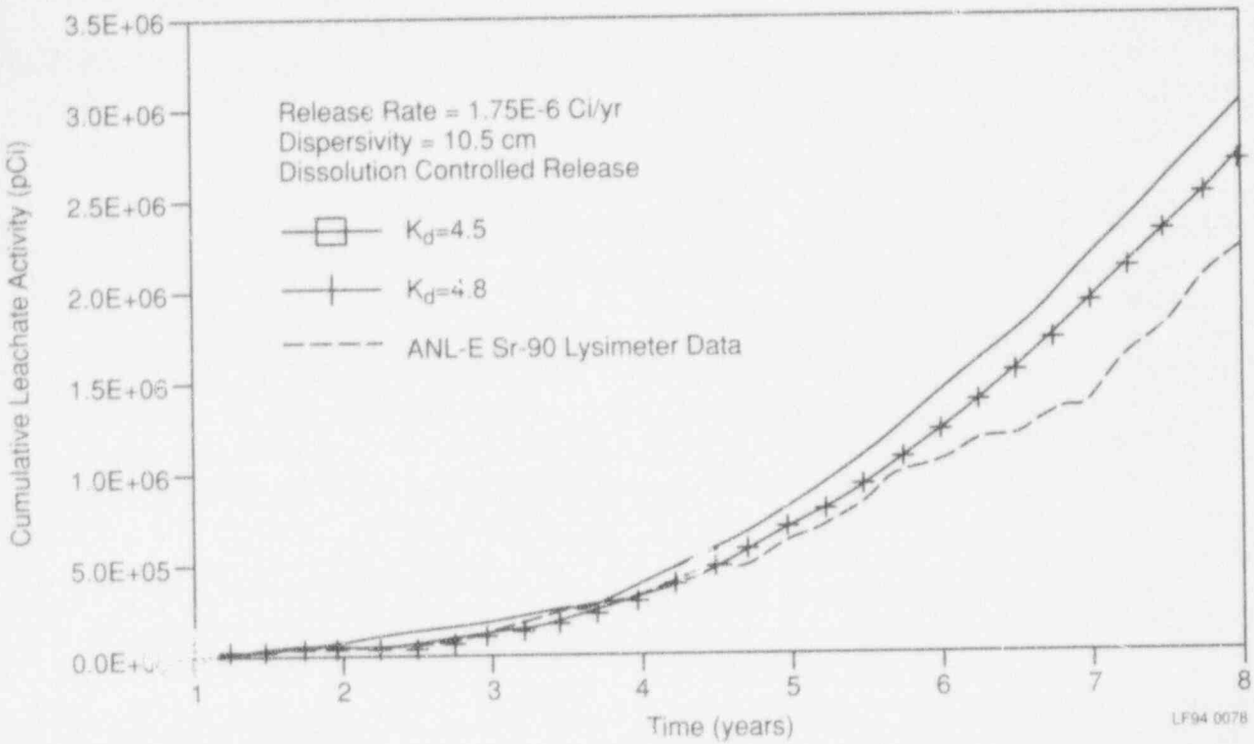

Figure 37. Data for Sr-90 at ANL-E lysimeter 5, compared with the effects of K_d values on predicted releases with an exponentially decaying waste form release rate.

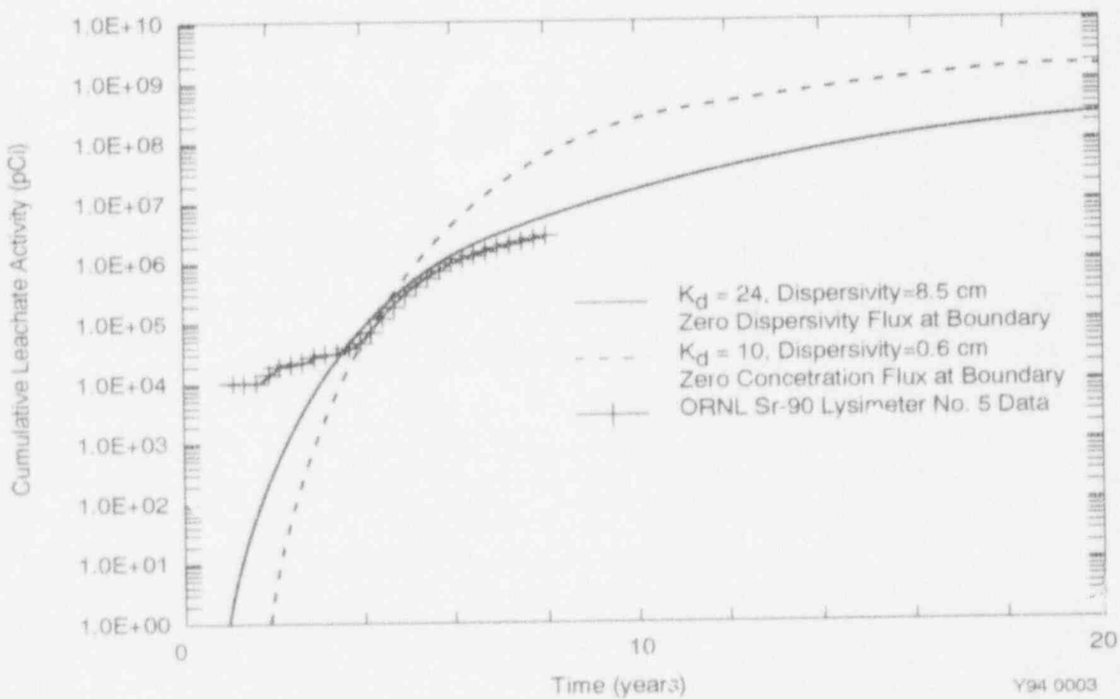

Figure 38. Data for Sr-90 at ORNL lysimeter 5, compared with two sets of estimated K_d and dispersivity values.

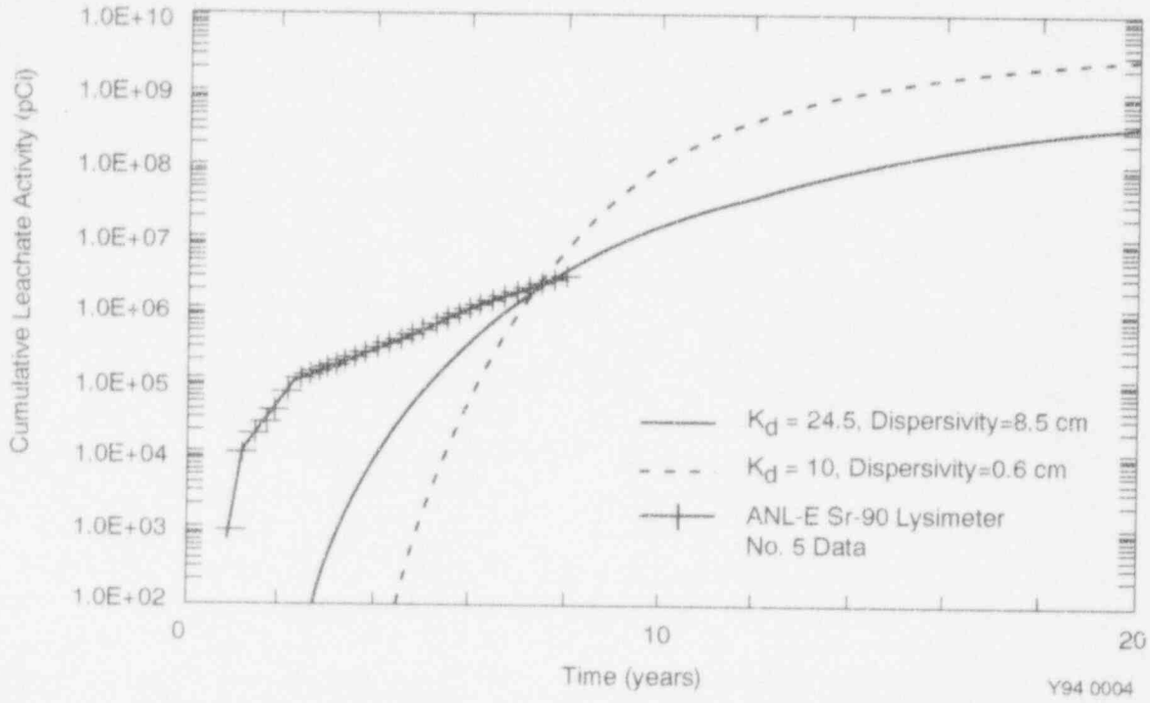

Figure 39. Eight years of data for Sr-90 at ANL-E lysimeter 5, compared with two sets of estimated K_d and dispersivity values for 20 years.

As shown in Figure 38, the actual data for Sr-90 from ORNL lysimeter 5 for eight years are compared with the DUST code predicted releases using zero dispersive flux BC, K_d = 24, and dispersivity = 8.5 cm. Also shown are predicted releases using zero concentration flux BC, K_d = 10, and dispersivity = 0.6 cm. The measured waste form diffusion coefficient of 9.6E-10 cm^2/s was used. The predicted releases of zero dispersive flux BC show a very good fit to the actual data after three years. The DUST curve that is generated with the zero dispersive flux BC is rising at a much more shallow slope than the zero concentration BC curve, indicating lower predicted releases over 20 years.

Figure 39 shows the actual data for Sr-90 at ANL-E lysimeter 5, which covers a period of eight years. In addition, the DUST predictions of 20 years of cumulative leachate activity is plotted in two cases, using dispersive flux BCs. The measured waste form diffusion coefficient of 9.6E-10 cm^2/s was used. Case 1 has a dispersivity of 8.5 cm and a K_d of 24.5. Case 2 has a dispersivity of 0.6 cm and a K_d of 10. Case 2 releases less activity over eight years than Case 1; however, at 20 years, the amount of activity released by case 2 is an order of magnitude higher than the amount in case 1. Over 20 years, case 2 will have released 33% of the total Sr-90 inventory, whereas case 1 will have released 3.3% of the total Sr-90 inventory. Case 1, also, is a better fit to the actual data at eight years, indicating a predicted higher dispersivity and K_d than previously thought.

Major Cation and Anion Analysis. A clear understanding of the factors that influence movement of radionuclides through the lysimeter soils is not available in the literature. A preliminary effort was initiated at ORNL in 1988 and at ANL-E in 1991 to analyze water samples obtained from moisture cups for some major cation and anion species. It is anticipated that such data could prove useful as a first indication of deterioration of waste form solidifying material. It could also indicate the presence of major ions, which could enhance radionuclide transport by either forming soluble complex formations with radionuclides [e.g., Sr-90

$(HCO_3)_2$—an electrically neutral dissolved species) or by causing movement as a result of competition with radionuclides for the limited number of soil exchange sites (e.g., K^+ versus Cs^+). These data, together with future analysis of the mineralogical composition of the lysimeter soil, could be used to develop equilibrium geochemical modeling, which could in turn be used to calculate the concentration of various radionuclide complexes in the soil solution.

A portion of the water obtained at ORNL and ANL-E during one summer sampling period in 1993 was analyzed for the major ionic species listed in Table 10. The justification for the choice of ions is also provided in the table. At ANL-E, cups 1, 3, and 5 were sampled on lysimeters 1, 3, 4, and 5; and cups 2, 3, and 4 on lysimeter 2. Cups 1, 3, and 5 water samples were sampled in 1993 at ORNL. Data from precipitation samples at ANL-E in 1991 and ORNL in 1988 showed that ionic concentrations in the soil water were not introduced by the precipitation (Reference 11). It appears that the waste forms could be an influencing factor either as the source of ions or possibly by causing replacement of ions from the surrounding soil such as the exchange of soil calcium for released cesium (see Tables 11 and 12 and Figures 40, 41, 42, and 43). It appears that the cement and VES waste-form specimens performed similarly at both sites. With a few exceptions, the ORNL 1993 soil lysimeter data (Table 12 and Figures 42 and 43) closely resemble those of 1988, 1989, 1991, and 1992 cation and anion concentrations and actually show little cup-to-cup variability. ANL-E 1993 data are similar, in most cases, to ORNL 1993 data when compared in Figures 40, 41, 42, and 43. The inert sand-filled lysimeter results are almost identical except for NO_3 at ORNL, which had a higher concentration. While these early data are interesting, no correlation has been made with radionuclide movement.

Table 10. Ionic species analyzed from lysimeter moisture cup water samples.

Ionic species	Justification
Na^+	Indicator of weathering reactions if Na-feldspars are present.
Mg^{2+}	Forms complexes with bicarbonate and carbonate.
Ca^{2+}	In the absence of calcium minerals, this may be an indicator of cement breakdown. Forms complexes with bicarbonate and carbonate. An indicator of Sr behavior.
K^+	Indicator of weathering reactions if K-feldspars or illite are present. Competes with Cs for exchange sites.
H_4SiO_4	Indicator of weathering reactions. Concentrations of dissolved silica above saturation with quartz may indicate weathering of the zeolite.
Alkalinity	Bicarbonate and carbonate form complexes with Ca, Mg, and Sr. Typically the major anion in soil solutions.
SO_4^{2-}	Second most abundant anion in soil waters. Forms complexes with most cations.
PO_4^{3-}	Complex forming anion. Sorbs on iron oxide surfaces. Indicator of Sb behavior.
NO_3^-	Needed for charge balance calculation.
Cl^-	Needed for charge balance calculation.

Table 11. ANL-E chemical speciation results from lysimeter moisture cups 1, 2, 3, 4, and 5, June 1993.

		Cation					Anion			
Sample	Solidification agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	88	11	12	<1	53	2.0	0.32	0.94	38
Lys 1-3		45	6.6	5.9	<1	23	3.6	1.2	2.4	48
Lys 1-5		32	0.89	2.8	<1	7.8	2.0	4.4	<0.5	20
Lys 2-2	Cement	89	7.9	11	<1	48	2.3	<0.1	<0.5	27
Lys 2-3		20	0.30	2.3	<1	4.1	0.98	1.7	1.5	6.1
Lys 2-4		90	5.0	10	<1	49	2.1	0.21	<0.5	36
Lys 3-1	VES	67	3.3	7.9	<1	41	1.5	4.4	1.6	20
Lys 3-3		83	6.0	14	<1	48	6.4	0.48	1.7	27
Lys 3-5		62	2.3	16	<1	46	1.5	1.8	1.2	26
Lys 4-1	VES	75	4.2	11	<1	47	4.9	0.28	<0.5	34
Lys 4-3		86	5.4	9.4	<1	45	1.6	0.23	<0.5	35
Lys 4-5		86	2.8	9.5	<1	40	1.5	<0.1	<0.5	30
Lys 5-1	Cement	6.6	<0.3	7.4	<1	3.0	0.57	3.7	<0.5	4.4
Lys 5-3		8.3	8.5	28	3.6	4.2	1.3	4.3	3.5	5.6
Lys 5-5		6.9	<0.3	168	<1	2.9	0.98	4.8	<0.5	5.2

Table 12. ORNL chemical speciation results from lysimeter moisture cups 1, 3, and 5, July 1993.

		Cation					Anion			
Sample	Solidification agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	34	3.1	18	<1	1.3	5.3	9.2	<3	2.4
Lys 1-3		33	2.8	23	2.3	1.4	0.6	3.8	<3	18
Lys 1-5		35	0.34	19	<1	1.8	2.3	5.4	<3	11
Lys 2-1	Cement	41	2.2	16	<1	1.4	<1.0	25	<3	7.7
Lys 2-3		36	2.4	28	2.3	1.1	1.3	33	<3	6.8
Lys 2-5		9.9	0.78	7	<1	1.2	3.9	25	<3	1.6
Lys 3-1	VES	30	1.6	17	<1	0.75	1.2	13	<3	4.9
Lys 3-3		37	4.3	33	<1	1.2	4.6	79	<3	4.6
Lys 3-5		3	0.25	9.9	<1	1.2	1.7	2.9	<3	2
Lys 4-1	VES	8.7	3.7	8.2	<1	1.9	4.2	1.4	<3	17
Lys 4-3		6.1	4.5	10	<1	0.98	1	2.4	<3	18
Lys 4-5		1.8	0.24	9	<1	0.39	2	12	<3	3.4
Lys 5-1	Cement	6.3	0.23	7.2	<1	3	1.7	9.2	<3	7.6
Lys 5-3		13	1.7	27	2.7	5.0	5.8	<1	<3	5.0
Lys 5-5		13	0.28	18	<1	3.3	26	11	<3	6.1

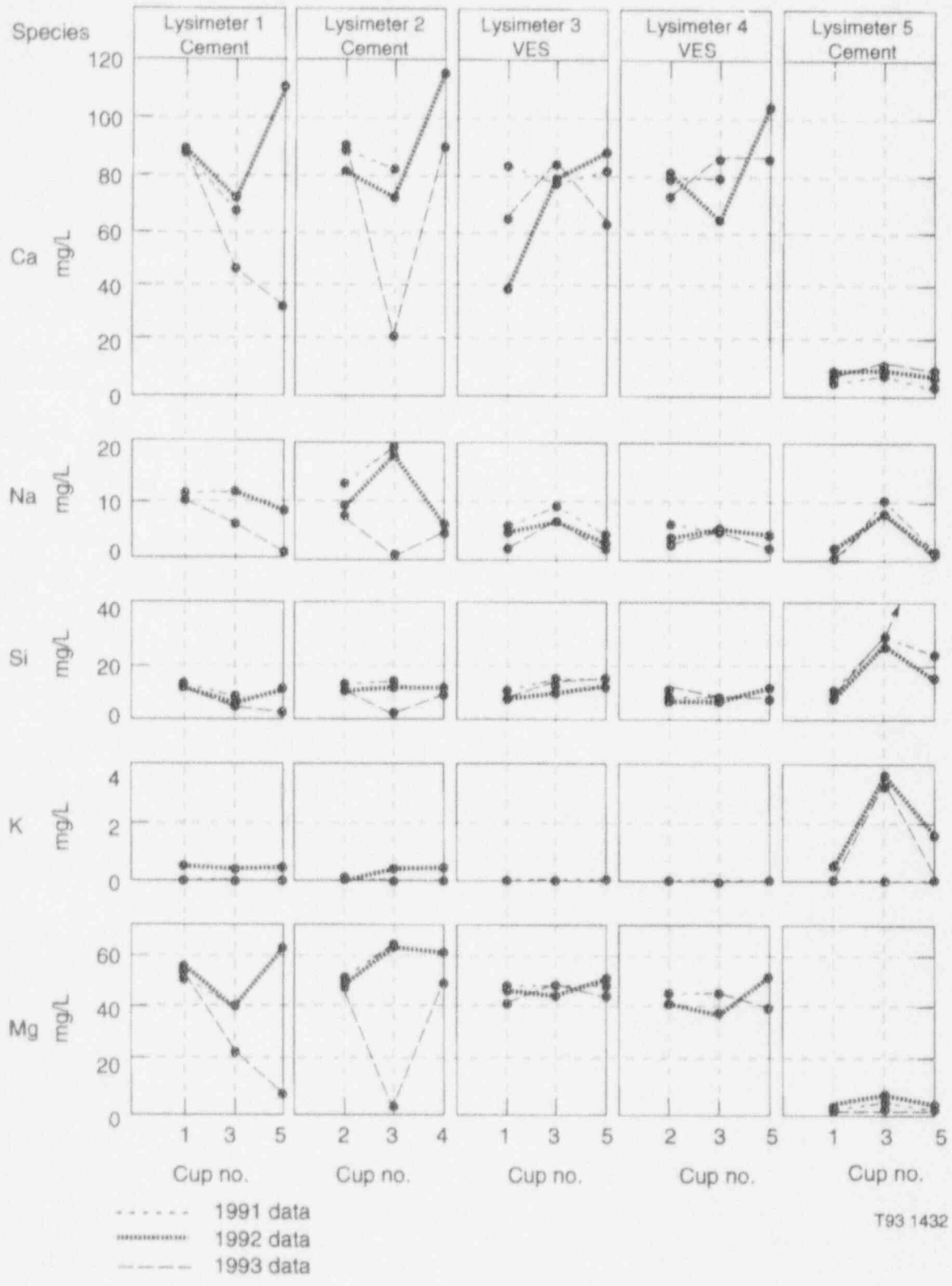

Figure 40. Results of chemical speciation at ANL-E cations.

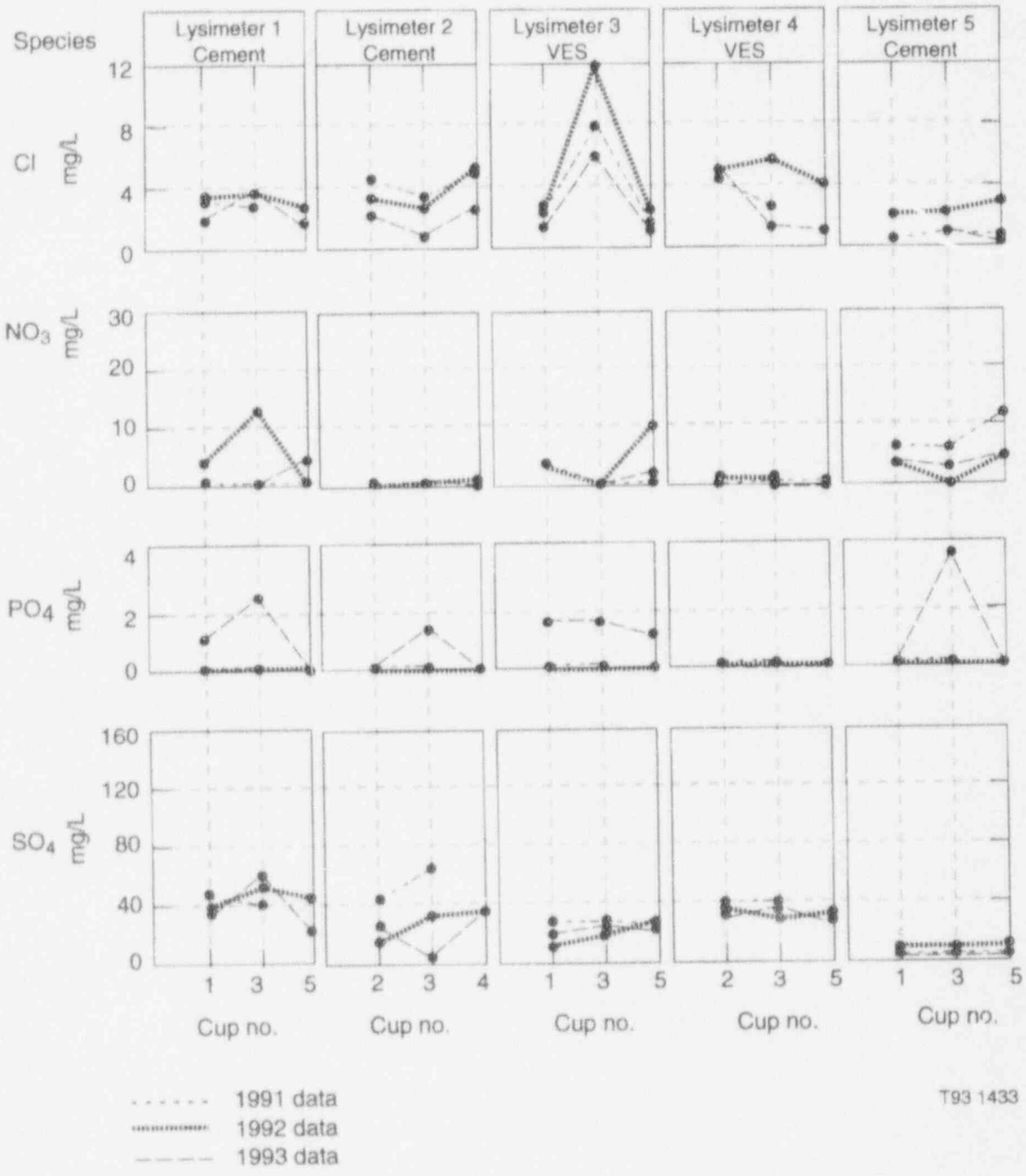

Figure 41. Results of chemical speciation at ANL-E anions.

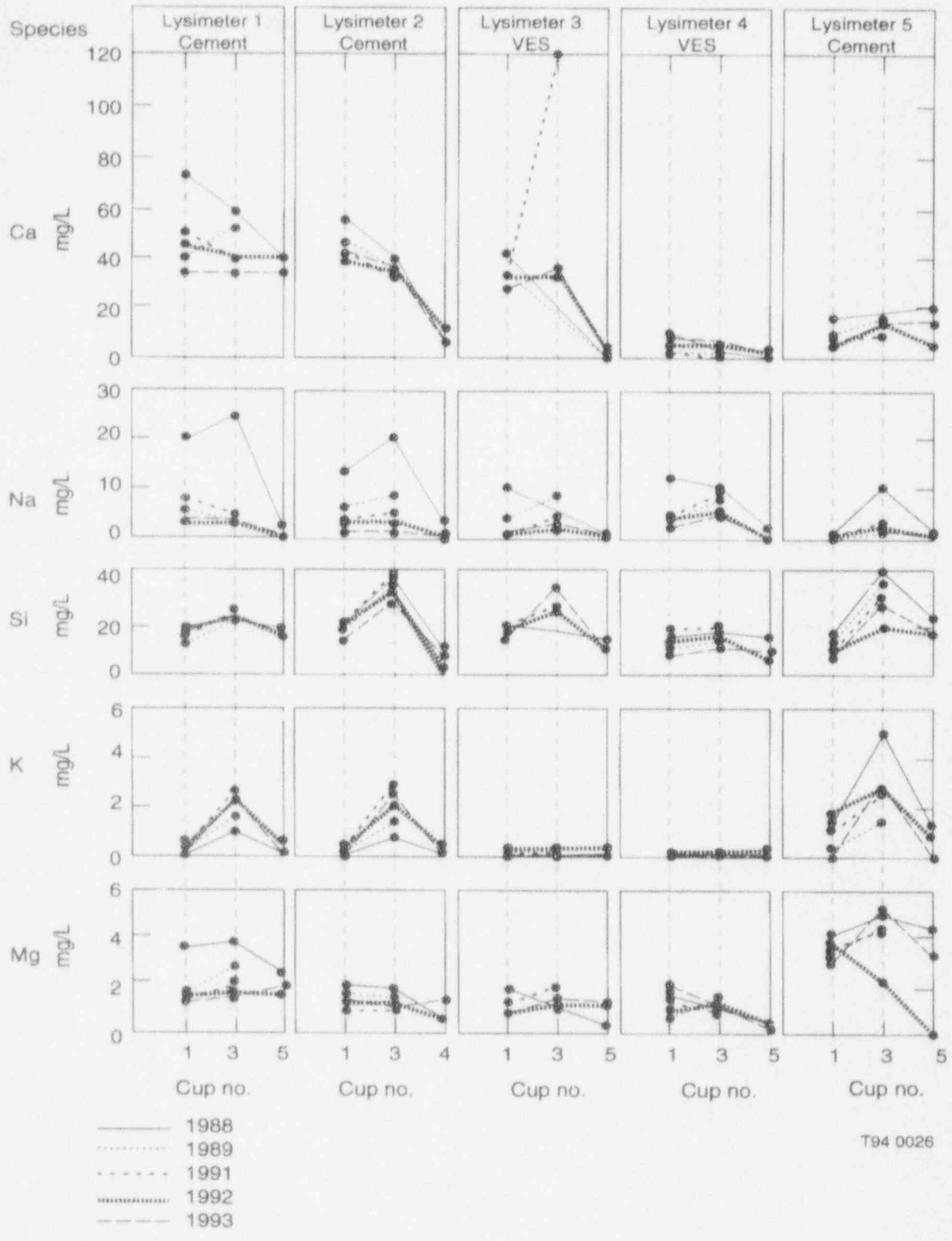

Figure 42. Results of chemical speciation at ORNL cations.

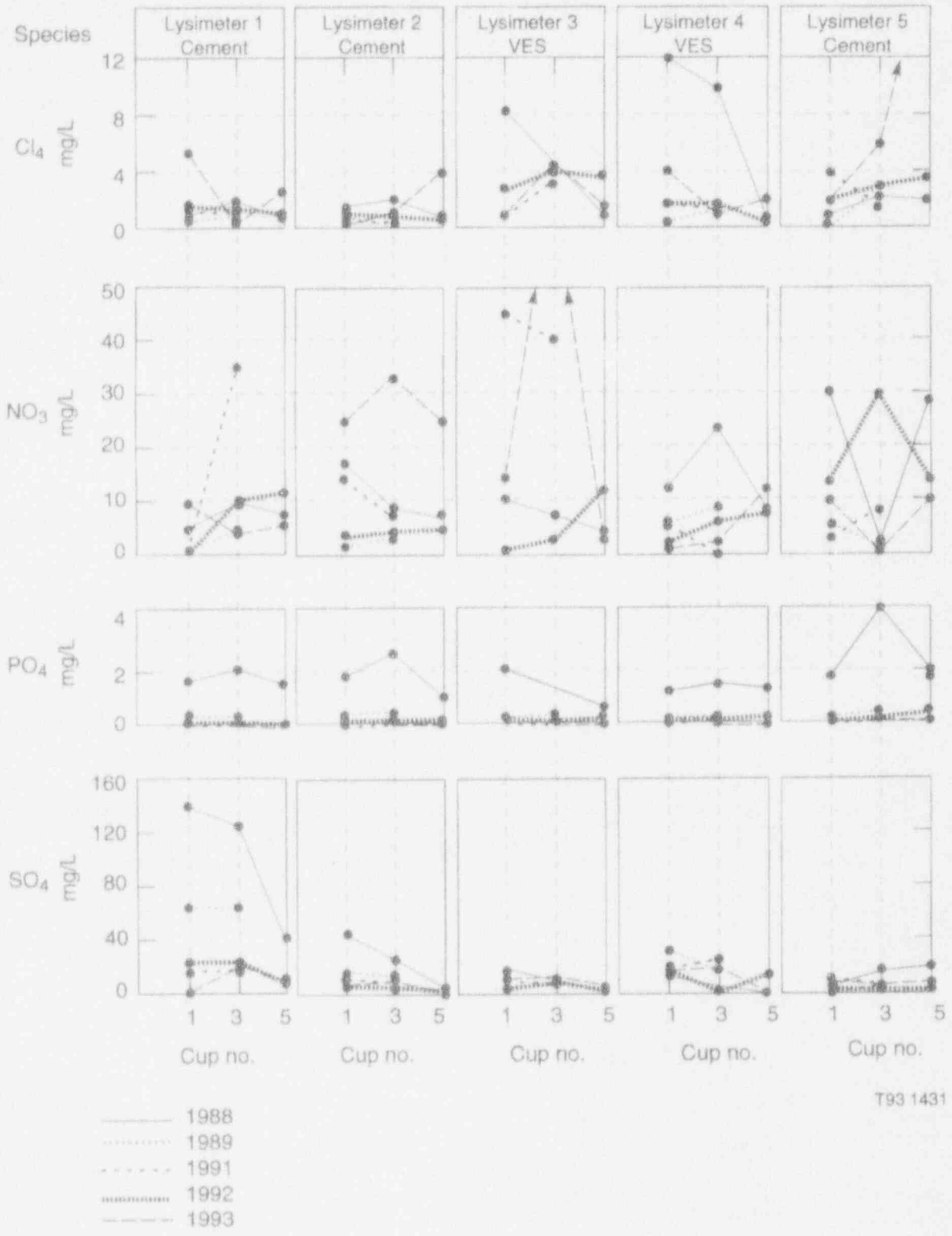

Figure 43. Results of chemical speciation at ORNL anions.

CONCLUSIONS

Field Testing

The lysimeter experiment during the eight years of operation has been successful. Analyses of data collected during the past 96 months continue to show a pattern in nuclide availability and movement such that the cumulative results are beginning to provide an insight on waste form performance.

There continues to be a greater recovery of Sr-90 in terms of quantity and percent of inventory than other nuclides. Next in abundance is Cs-137, followed by Sb-125 (this nuclide has not been detected for the past 48 months) and Co-60. Compared to Sr-90, the occurrence of Cs-137 appears insignificant.

On a cumulative basis, a larger amount of Sr-90 is being removed in leachate water from the ORNL soil lysimeters. This is thought to be a result of the difference in soils as well as in environmental conditions between the two sites. During the past 72 months, Sr-90 continues to be found in equal concentrations in leachate water from the sand-filled control lysimeters at both sites, with a slightly more rapid accumulation at ORNL, which now has hao six and one half times more of the available source of Sr-90 released than the control lysimeter at ANL-E. Such data continue to reinforce the assumption that the limiting step in receiving Sr-90 in leachate water is not release of the nuclide from the waste forms (since Sr-90 is found in larger quantities in leachate water at ORNL rather than in cups), but rather, the movement is limited by environmental characteristics (including soil and quantity of soil water). This conclusion is supported by data from lysimeter work at SRL and Pacific Northwest Laboratory (PNL).[23,24] SRL has found that Sr-90 will move from buried waste forms, migrate through the soil column, and appear in collected leachate water.[23] It is not surprising, then, that Sr-90 moves through soil in the ORNL lysimeters, since that soil originated at SRL.[8] On the other hand, lysimeter work with waste forms at PNL has shown that Sr-90 does not move in those soils.[24]

Percent recovery of Sr-90 from the ORNL cups is the same order of magnitude for those lysimeters containing the cement waste forms and one of the two containing VES waste forms. In general, at ORNL, a larger percentage of Sr-90 has been recovered from the two lysimeters containing cement waste forms than from those containing VES. ANL-E cumulative Sr-90 data show that amounts of Sr-90 collected in the moisture cups of the two lysimeters containing VES waste forms are larger than in those containing cement waste forms.

Cesium-137 again has been found in leachate water from the sand-filled control lysimeters at both sites for the second year.

As a conclusion, data from the two sites have not yet demonstrated which type of solidification product is preferable for nuclide retention. It appears at this time that releases of Sr-90 and Cs-137 from cement and VES are comparable but dependent on environmental influences. These data still differ from those obtained at SRL. Those data show that cement minimizes the release of SR-90.[23] This interesting difference should be studi: 3 further. Both data reported herein and data reported by SRL and PNL agree that Cs-137 ;s more readily released from cement than from VES.

On two occasions, lysimeter data have been reviewed to determine the possibility of using these data to initiate limited performance assessment modeling. The results from a preliminary evaluation that was carried out in FY-91 indicated that in lysimeters with experimentally determined diffusion coefficients, where there were high enough leachate concentrations of nuclides for comparison between predicted and experimental results, a computer code could be tested. Last year, further refinements made it possible to model some of the lysimeter Sr-90 release data using the DUST computer code. Once again, as has been the case of others using these data, it was

strongly recommended that the lysimeter experiments be continued. Rapidly increasing radionuclide release showed that data from future years could be used to obtain a reliable, quantitative understanding of nuclide movement through the use of numerical codes.

The numerical study continues to be hampered by the lack of soil data. It is important to know the site-specific soil distribution coefficient (K_d) and dispersivity values to better predict the release characteristics in the lysimeters.

Boundary conditions have little effect on predicted cumulative activity release; however, they play an important role in predicted concentrations. Concentration profiles are developing slowly, and further releases should, therefore, continue to be monitored. Further numerical studies are planned.

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11. ABSTRACT (200 words or less)

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program, funded by the U.S. Nuclear Regulatory Commission, is (a) studying the degradation effects in EPICOR-II organic ion-exchange resins caused by radiation, (b) examining the adequacy of test procedures recommended in the Branch Technical Position on Waste Form to meet the requirements of 10 CFR 61 using solidified EPICOR-II resins, (c) obtaining performance information on solidified EPICOR-II ion-exchange resins in a disposal environment, and (d) determining the condition of EPICOR-II liners.

Results of the eighth year of data acquisition from the field testing are presented and discussed. During the continuing field testing, both Portland type I-II cement and Dow vinyl ester-styrene waste forms are being tested in lysimeter arrays located at Argonne National Laboratory-East in Illinois and at Oak Ridge National Laboratory. The study is designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over a 20-year period.

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