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Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1991

Annual Report

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Prepared for
U.S. Nuclear Regulatory Commission

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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 Forms 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 sixth 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 (ANL-E) in Illinois and Oak Ridge National Laboratory (ORNL). The study is designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over a 20-year period.

FIN No. A6876—Field Lysimeter Investigations:
Low-Level Waste Data Base Development Program

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ACRONYMS AND ABBREVIATIONS

ANL–E	Argonne National Laboratory–East
ASTM	American Society for Testing and Materials
DAS	data acquisition system
DOE	U.S. Department of Energy
INEL	Idaho National Engineering Laboratory
MHC	moisture holding capacity
NRC	U.S. Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
TAN–607	Test Area North Building 607
TMI–2	Three Mile Island Unit 2
TP	technical position on waste form
VES	vinyl ester–styrene

Field Lysimeter Investigations: Low–level Waste Data Base Development Program for Fiscal Year 1991

EXECUTIVE SUMMARY

The March 28, 1979 accident at Three Mile Island Unit 2 (TMI–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. Fifty EPICOR–II prefilters with high concentrations of radionuclides were transported to the Idaho National Engineering Laboratory for interim storage before final disposal at a commercial disposal facility in the State of Washington. Research is being conducted on materials from four of those EPICOR–II prefilters under three tasks of the Field Lysimeter Investigations: Low–Level Waste Data Base Development Program.

For resin solidification, Portland Type I–II cement and vinyl ester–styrene (VES) waste forms incorporating ion–exchange resin waste from EPICOR–II prefilters are periodically subjected to the tests specified in the "Technical Position on Waste Form" issued by the U.S. Nuclear Regulatory Commission. Waste form performance data are obtained as a result of the work. EPICOR–II resin waste forms made with Portland Type I–II cement and DOW VES have been compression tested, and the results compared to similar waste forms tested earlier in the program. No tests were performed this fiscal year.

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 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 nuclide release and movement, as well as environmental conditions, will be obtained over a 20–year period.

Each month, data stored on a cassette tape are retrieved from the data acquisition system and are translated into an IBM PC–compatible disk file. At least quarterly, water is drawn from the porous cup soil–water samplers and the lysimeter leachate collection compartment. Those water samples are analyzed for beta– and gamma–producing nuclides.

Results of the sixth year of data acquisition are presented in this report. These results show that radionuclides are continuing to move from the waste forms and through the soil column. Also, some data on waste–form performance are presented. 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.

INTRODUCTION

The March 28, 1979 accident at Three Mile Island Unit 2 (TMI-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, or high-integrity container, was developed during that storage period for use in disposing 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 (DOE), 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 (TAN-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. (EG&G Idaho) 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 being 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, are 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" (TP),[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 compression-test procedures to ensure compliance with stability requirements. During this reporting period, no waste forms were compression-tested. However, the earlier 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

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[a] Mention of specific products and/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.

physical, chemical, and microbiological environment of typical disposal sites in the eastern United States.[1,2,3] It is intended that the lysimeters monitor release of nuclides from the 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 sixth year of lysimeter operation, as well as cumulative data on water balance and nuclide content of water samples. Data for this report were retrieved from the data acquisition system (DAS) 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 tested. No tests were conducted during this reporting period.

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 and data from the first four years of operation are contained in earlier reports.[8,9,10,11,12,13]

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 A is a mixture of synthetic organic ion-exchange resins from PF-7 (phenolic cation, strong acid cation, and strong base anion resins), while Type B 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 A contains 25% Sr-90, while Type B 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 forms 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 waste forms with an average diameter of 4.8 cm and a height of 7.6-cm (137.5 cm^3). A complete description of waste form manufacture is given in Reference 14. Bench testing of similar waste forms, per the requirement of the Branch Technical Position on Waste Form, is described in Reference 15.

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, 316L stainless steel (Figure 1). Internally, the lysimeter is divided into two sections, the upper volume being 1532 L and the lower being 396 L. A 3.8-cm, Schedule 40 stainless steel pipe serves as an access to the lower compartment. Soil, instrumentation, and waste forms are contained in the upper compartment, while the lower compartment serves as a leachate collector.

Table 1. Lysimeter waste form composition.

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

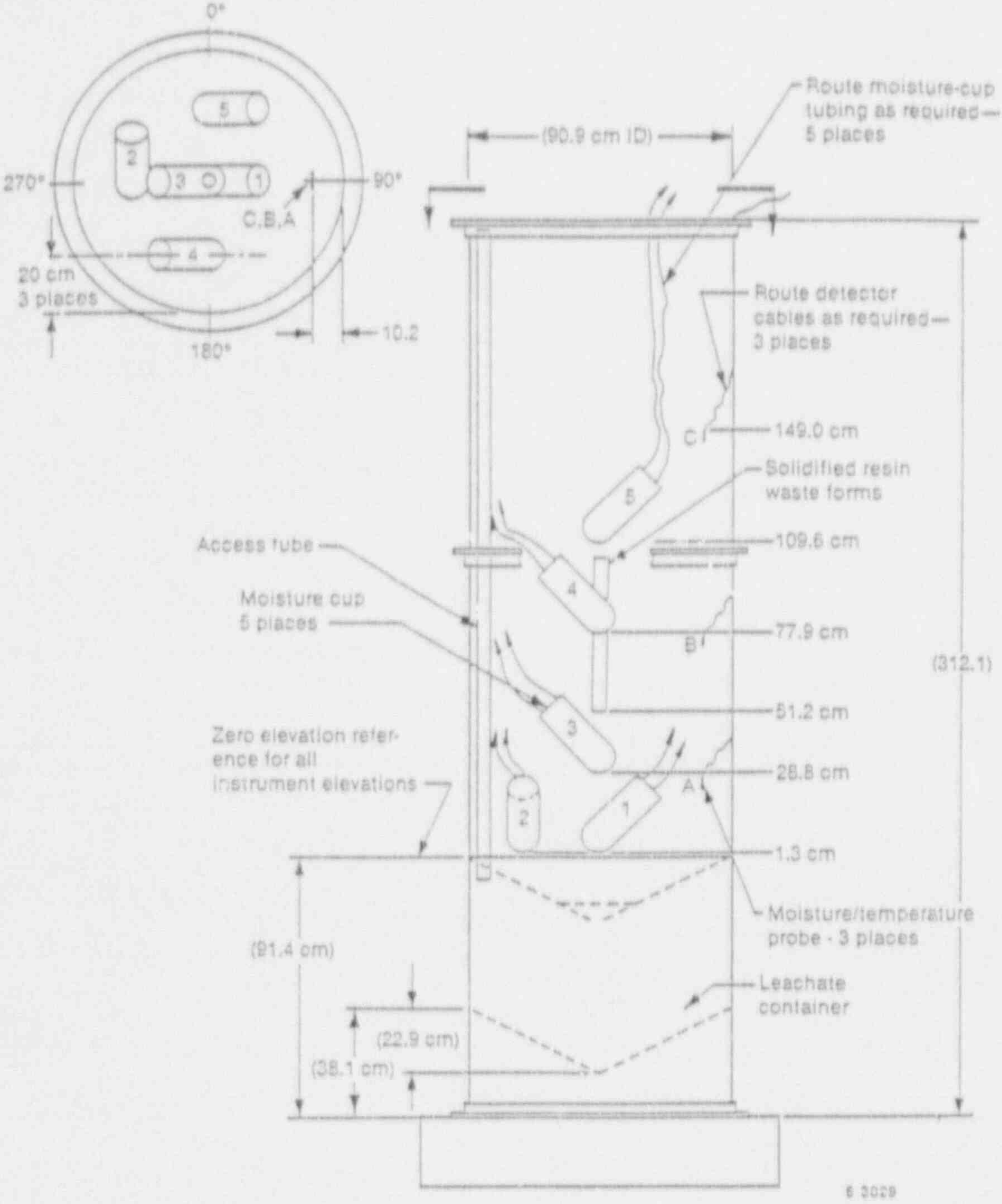

Figure 1. EPICOR–II lysimeter vessel component locations.

Four lysimeters at each field site are filled with soil; the remaining one is a control filled with an inert silica sand.[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 the midwestern soil. It is a Morley silt loam with the surface layer removed. The resulting subsurface soil is a clay loam. Soil at the ORNL was not found to be a suitable substitute for Barnwell soil; therefore, acceptable soil was transported to the ORNL from the Savannah River Plant adjacent to the Barnwell facility in South Carolina.

Each of the lysimeters is consecutively numbered 1 through 5, with 1 through 4 containing soil and number 5 being the sand–filled control. The waste form type found in each lysimeter is given in Table 1.

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.[8]

Data output from the DAS is stored on a cassette tape and, after retrieval, is translated to an IBM PC–compatible disk file. Hard copy from these files is provided either graphically or in a printed format. The graphic display presents data over an extended time period. The graphic presentation was 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 1990 through July 1991. In addition, information on water balance and nuclide content in soil water and leachate is a compilation of data from the initiation of the project (ANL–E, August 1, 1985; ORNL, June 1, 1985) through June 1991. Many of the data are displayed in graphic format so that information can be correlated easily with time.

Weather Data. Precipitation, air temperature, wind speed, 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 9. In October 1990, the anemometer at ANL–E ceased normal operation (Figure 4). The cause was due to mechanical failure. Total official precipitation (measured by reference rain gauges near each site) for the period was 106.2 cm at ANL–E and 149.1 cm at ORNL. ANL–E, for the second consecutive year since 1985, was well above the normal annual rainfall of 85.2 cm,[17] while ORNL continued to be near the normal annual rainfall of 138.8 cm.[18] This is the third time in the past five years that ORNL has equalled or exceeded the normal amount of yearly precipitation. The monthly precipitation pattern at each site can be seen from the histograms in Figures 2 and 6. Figure 10 shows the cumulative pattern of precipitation for both sites since the initiation of field work. By the end of this reporting period, there had been a cumulative total of 531.8 cm at ANL–E while ORNL had received a total of 786.8 cm.

Air temperature data from ANL–E (Figure 3) show that there were periods of freezing temperatures from December 1990 until the first part of March 1991. For the second year, ORNL experienced periods of freezing temperatures from mid–November until mid–February (Figure 7).

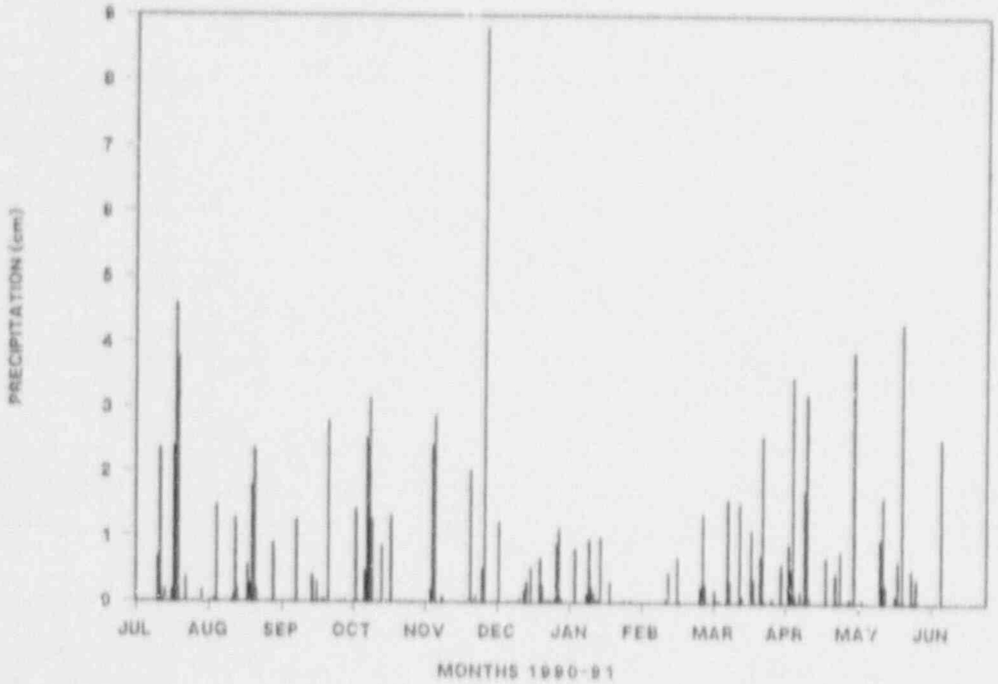

Figure 2. ANL–E weather data–precipitation.

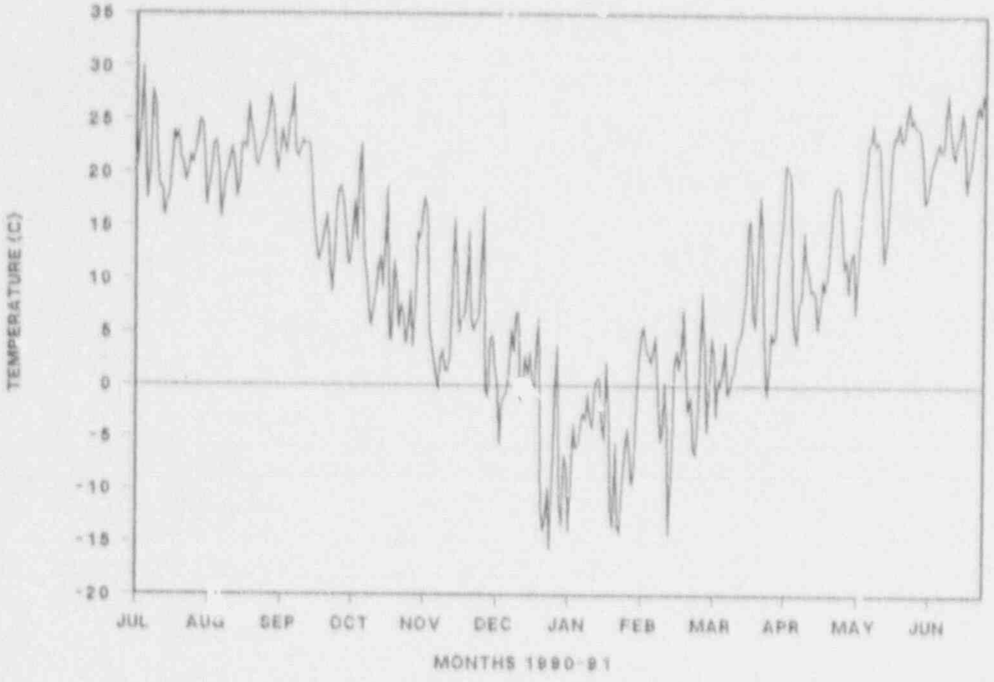

Figure 3. ANL–E weather data–air temperature.

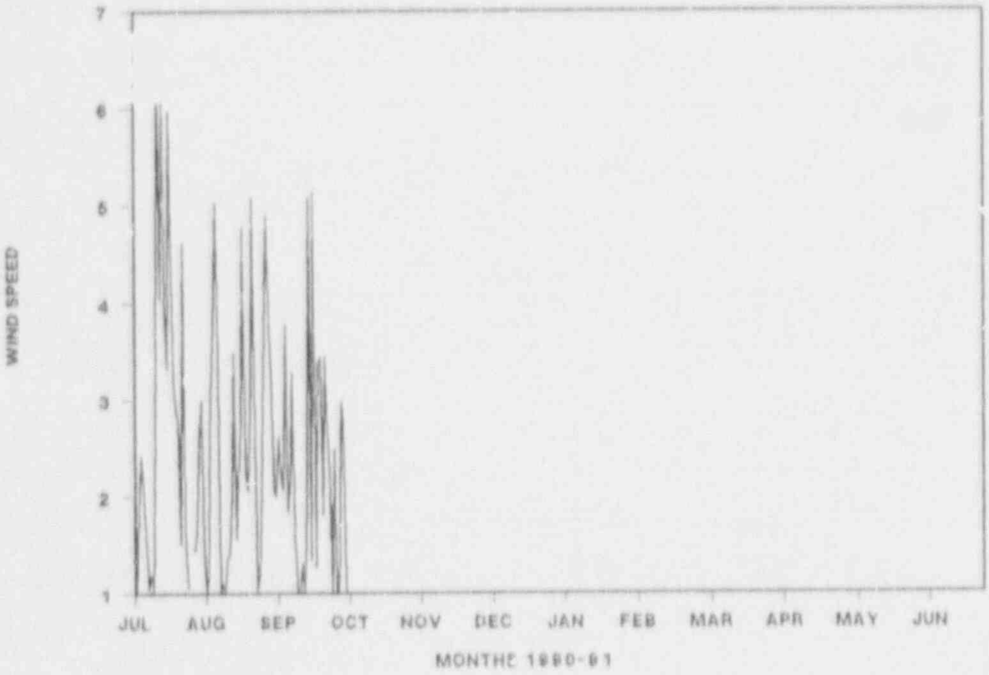

Figure 4. ANL-E weather data–wind speed.

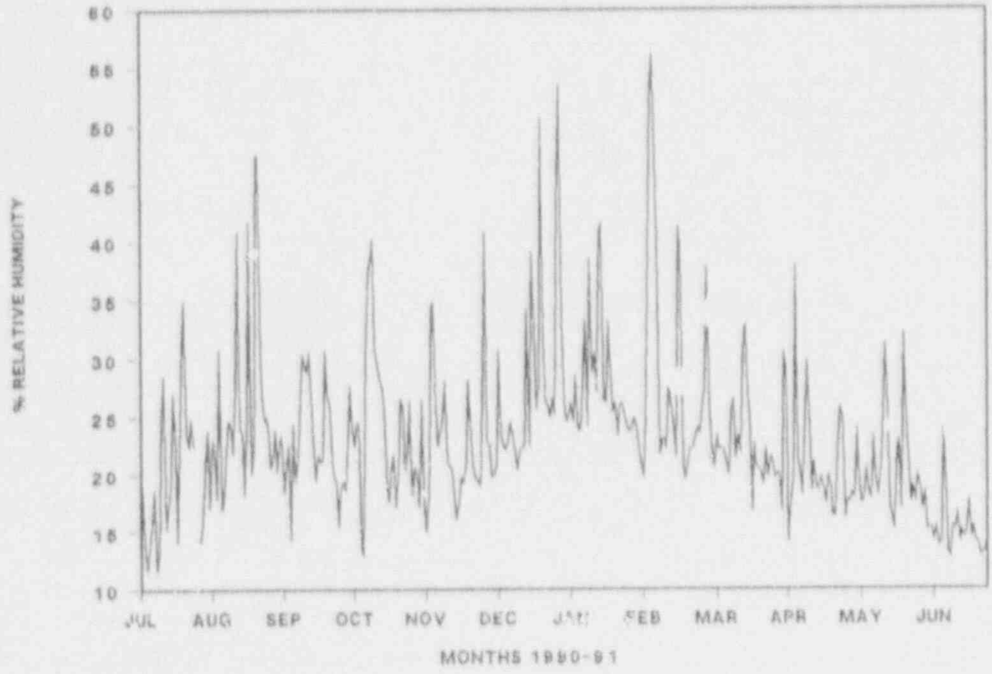

Figure 5. ANL-E weather data–relative humidity.

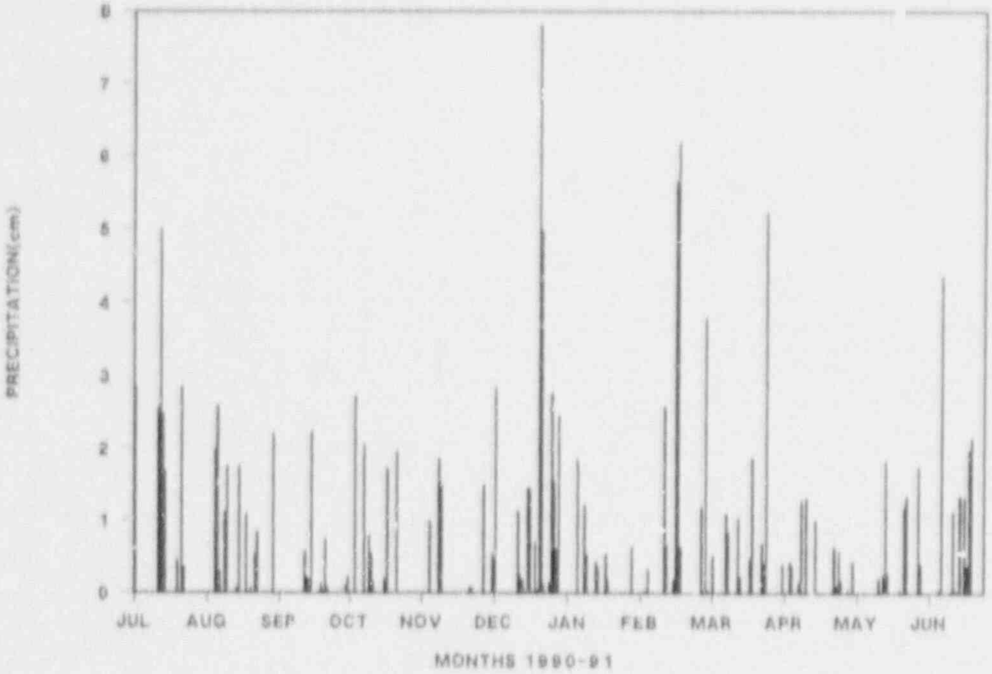

Figure 6. ORNL weather data–precipitation.

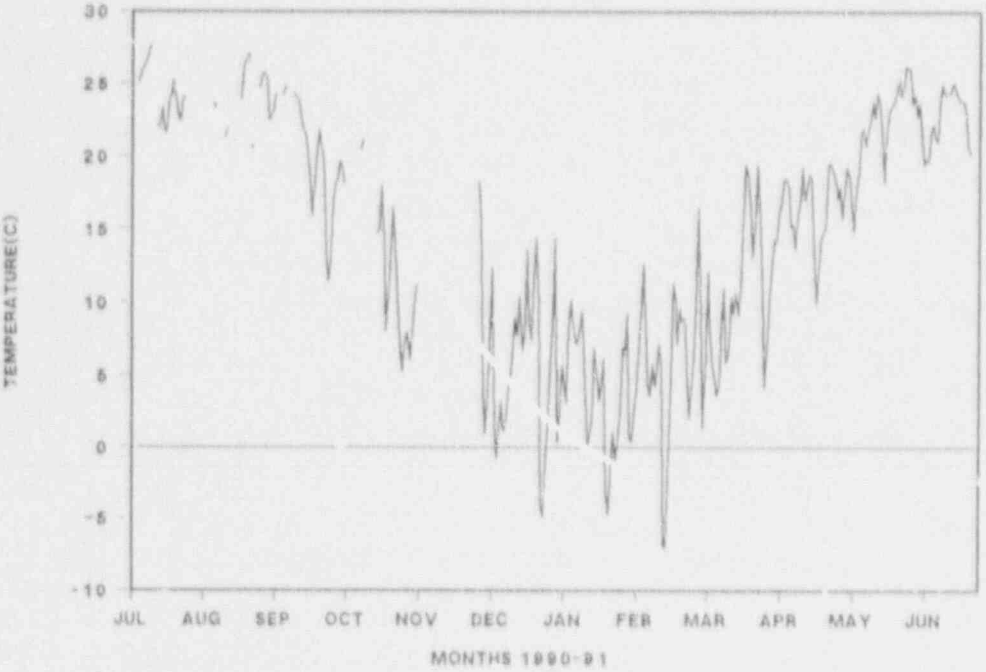

Figure 7. ORNL weather data–air temperature.

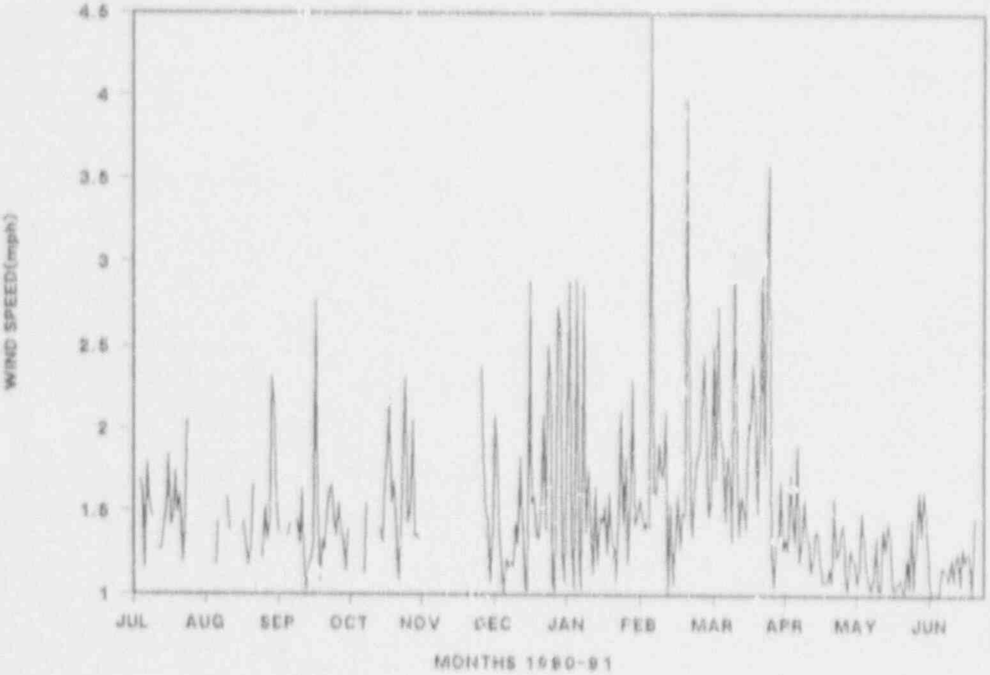

Figure 8. ORNL weather data–wind speed.

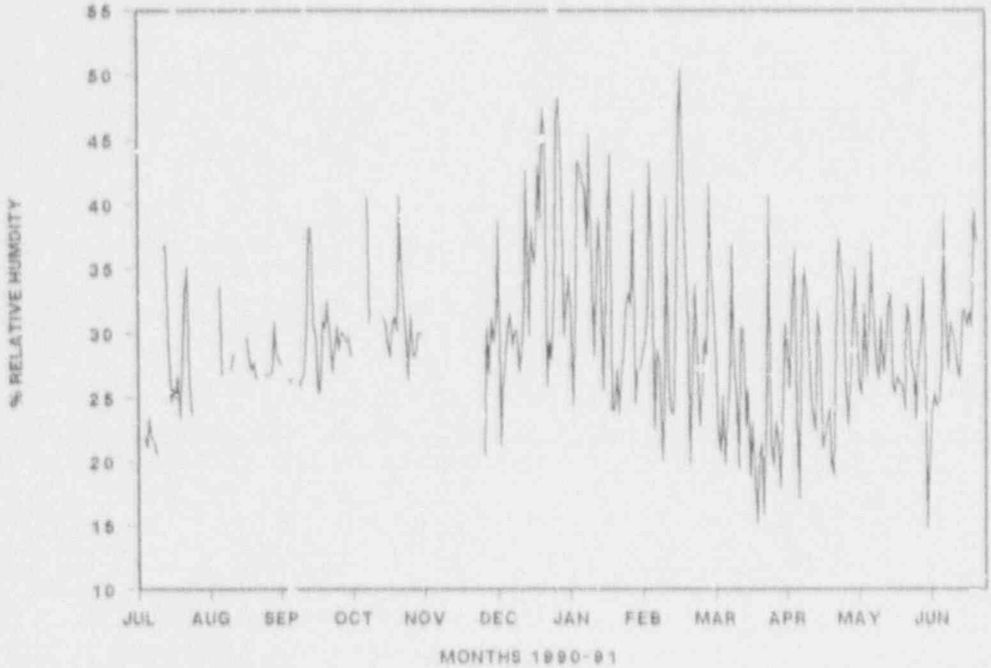

Figure 9. ORNL weather data–relative humidity.

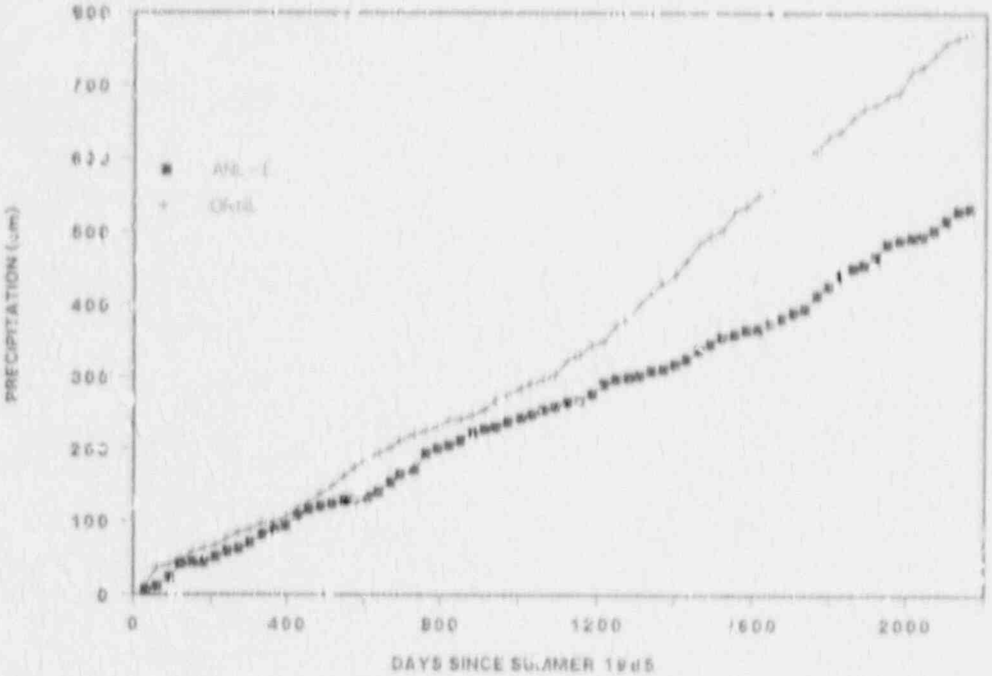

Figure 10. ANL-E and ORNL cumulative precipitation

Lysimeter Soil Temperature Data. 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.

The lysimeter soil temperature data recorded at ANL-E and ORNL during the reporting period are shown in Figures 11 through 19. 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, there have been a number of temperature probe failures at ANL-E. During the last three reporting periods, it was obvious that all the temperature probes in ANL-4 and one in ANL-2 had failed; therefore, data from these probes have not been included in the report. During the 1989–1990 reporting period, it appeared that the probes in ANL-3 were not functioning properly. Further deterioration of these probes and one in ANL-5 has been seen during this reporting period. From past experience, it would appear that the probes have been damaged by corrosion of the metal parts.[10] At the present time, a more damage-resistant replacement for these probes has not been found. Erratic behavior of some ORNL probes was seen during this reporting period. It has not been determined if this was due to the probes or faulty recording by the DAS.

Lysimeter Soil Moisture Data. Data from the moisture probes at both ANL-E and ORNL, shown in Figures 20 through 29, indicate that the lysimeter soil columns at both sites have remained moist during the reporting period. The probe

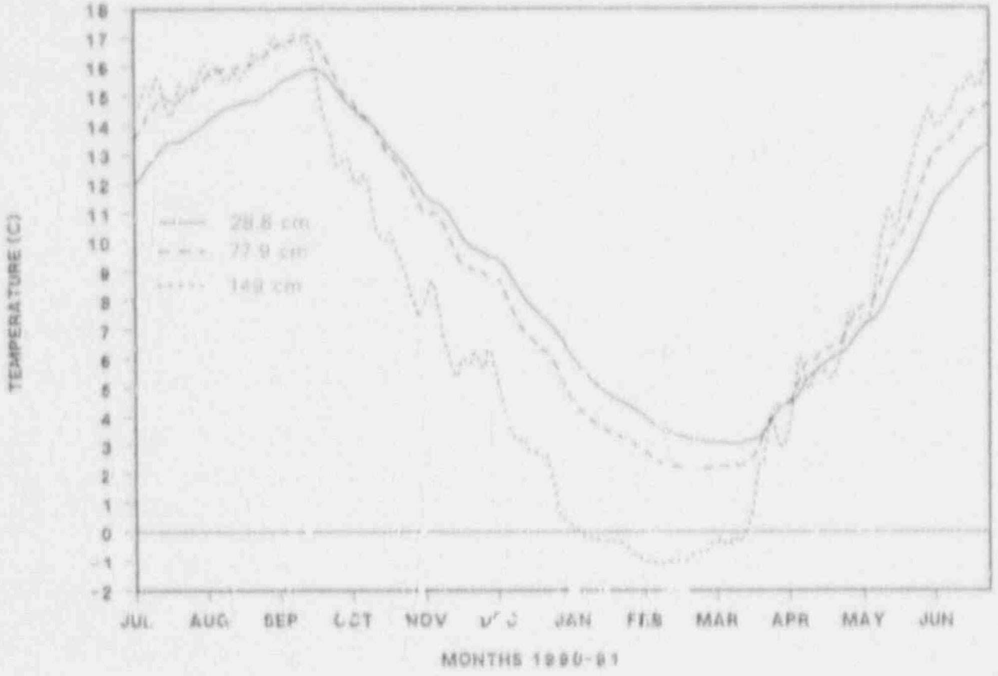

Figure 11. ANL-E lysimeter 1 soil temperature.

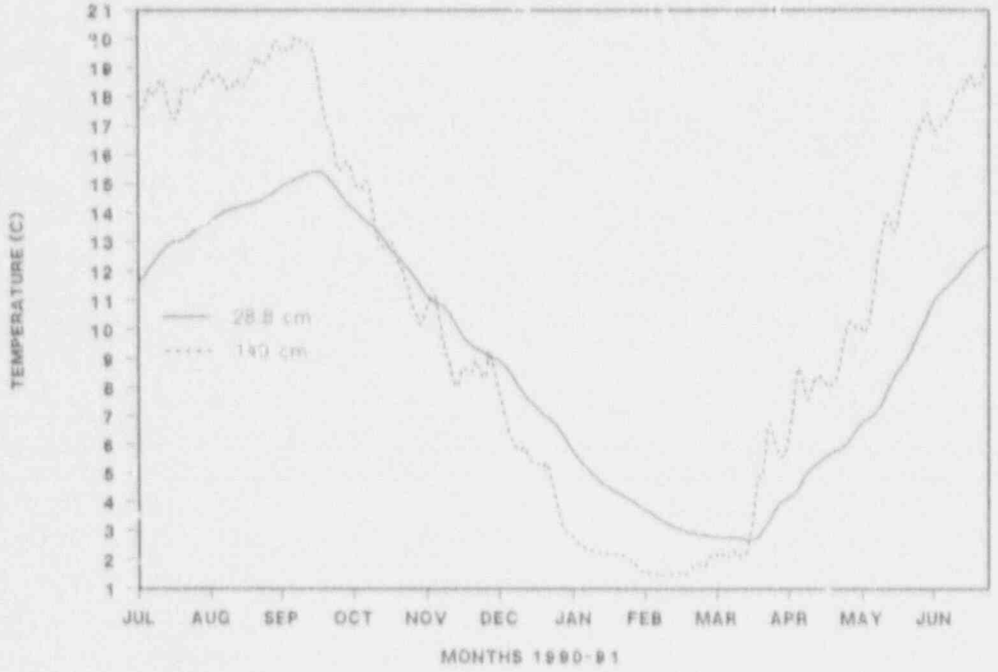

Figure 12. ANL-E lysimeter 2 soil temperature.

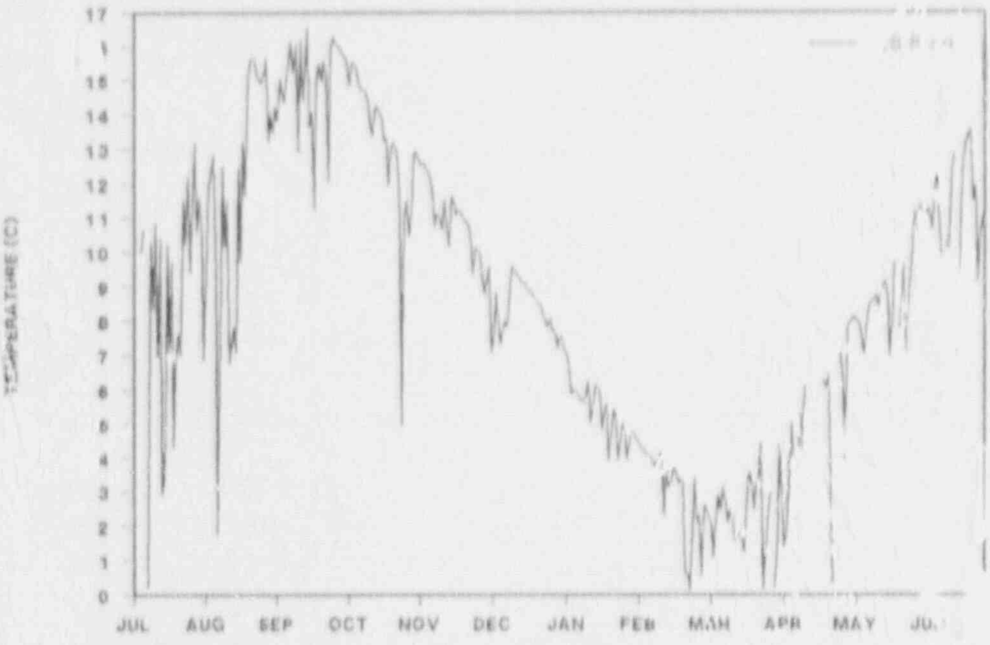

Figure 13. ANL–E lysimeter 3 soil temperature.

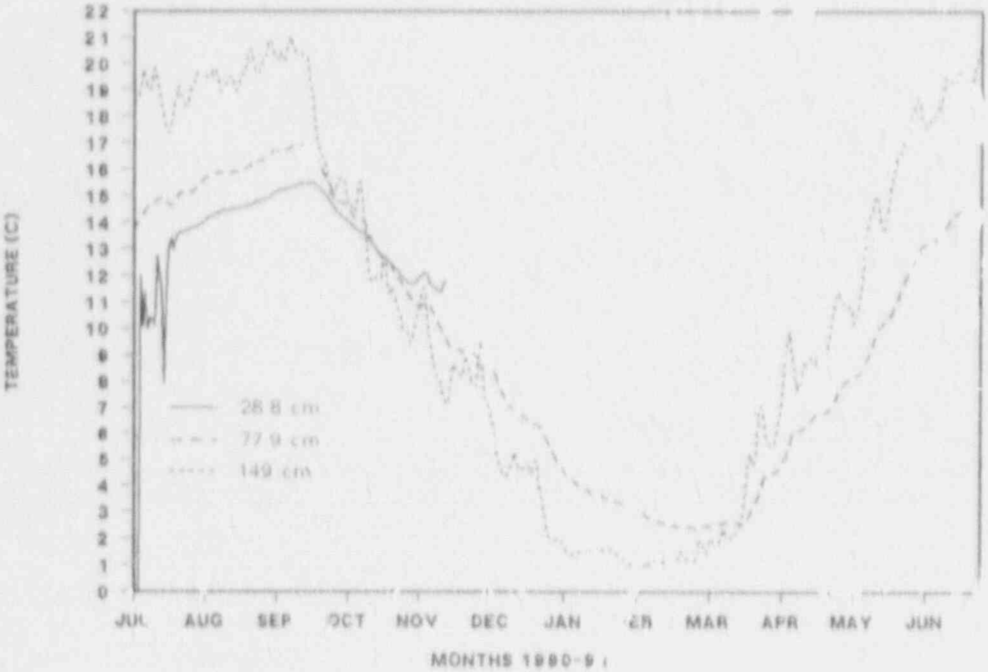

Figure 14. ANL–E lysimeter 5 soil temperature.

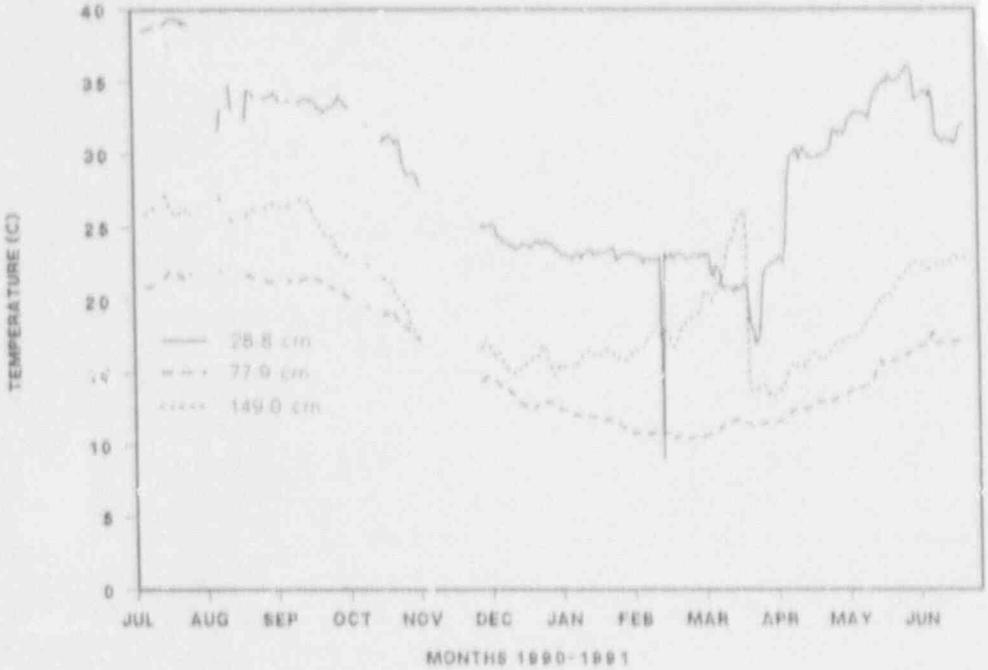

Figure 15. ORNL lysimeter 1 soil temperature.

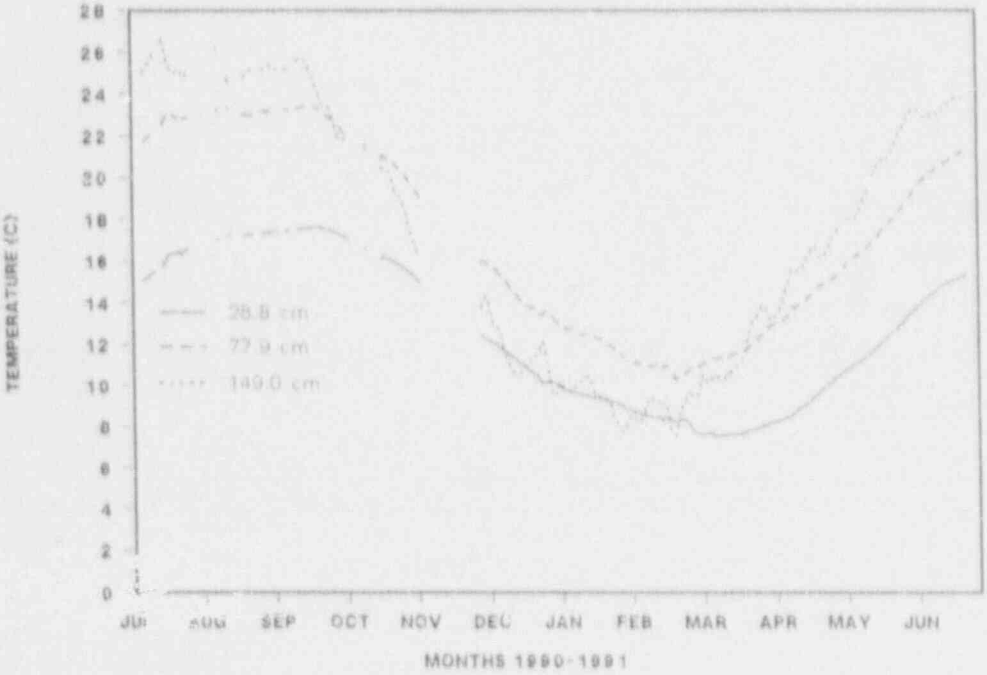

Figure 16. ORNL lysimeter 2 soil temperature.

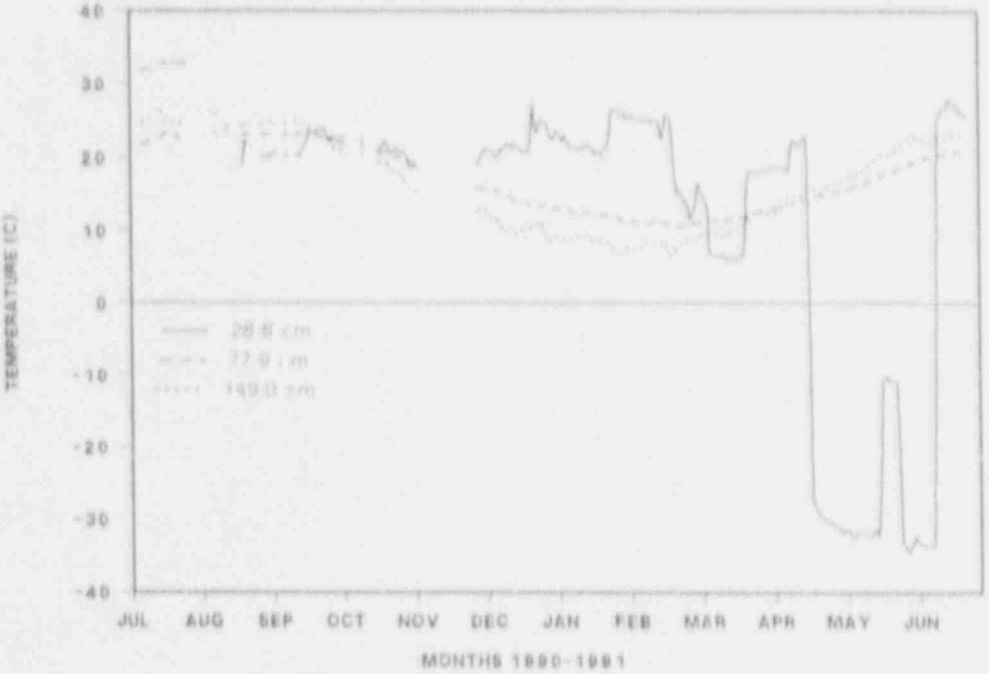

Figure 17. ORNL lysimeter 3 soil temperature.

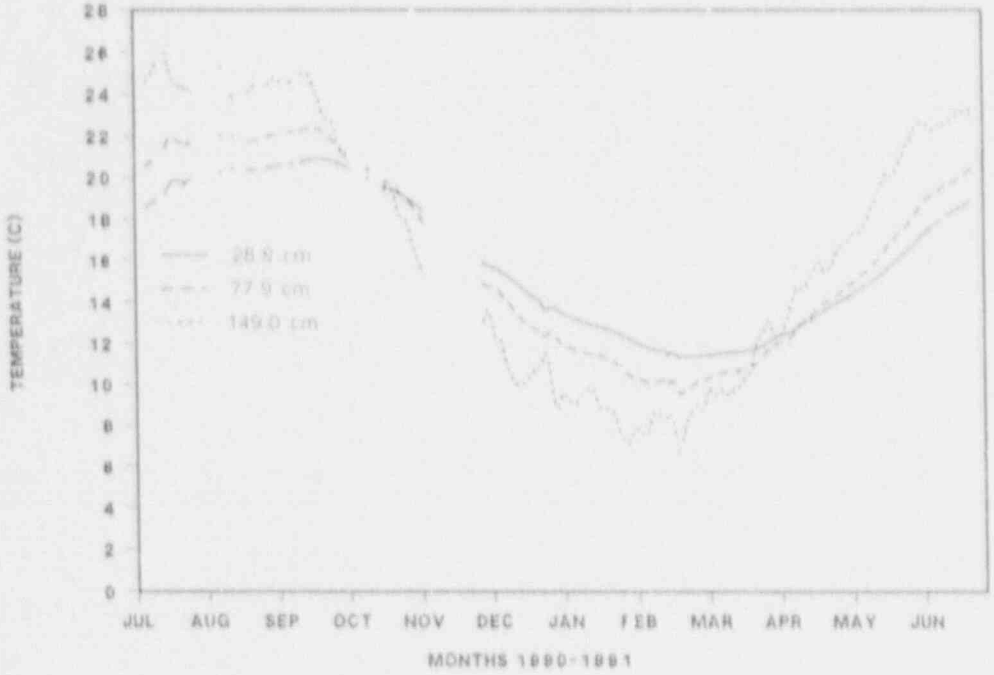

Figure 18. ORNL lisimeter 4 soil temperature.

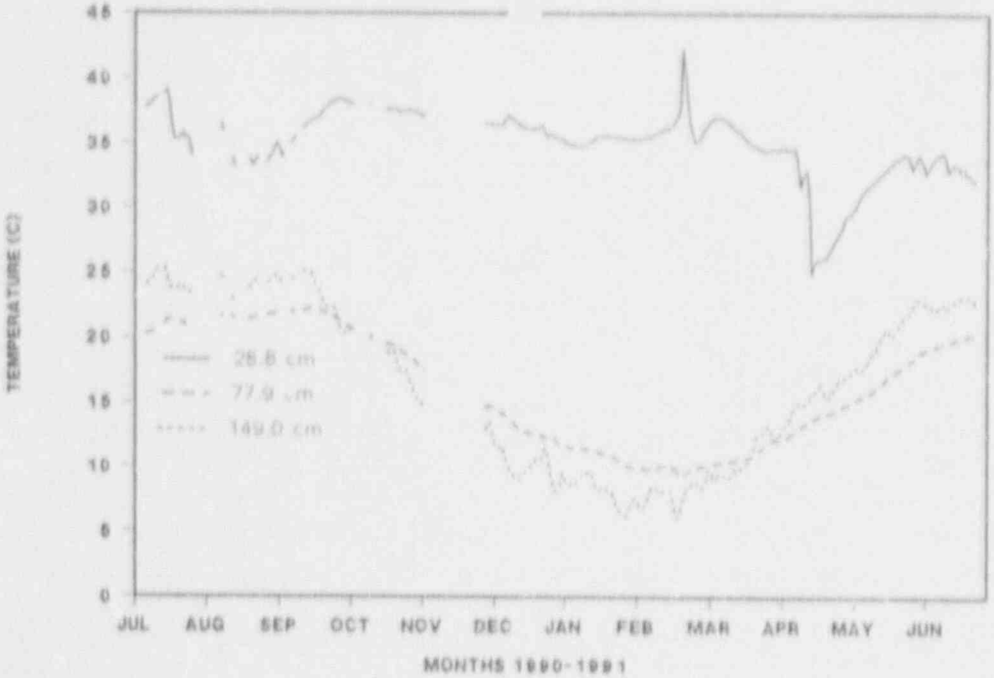

Figure 19. ORNL lysimeter 5 soil temperature.

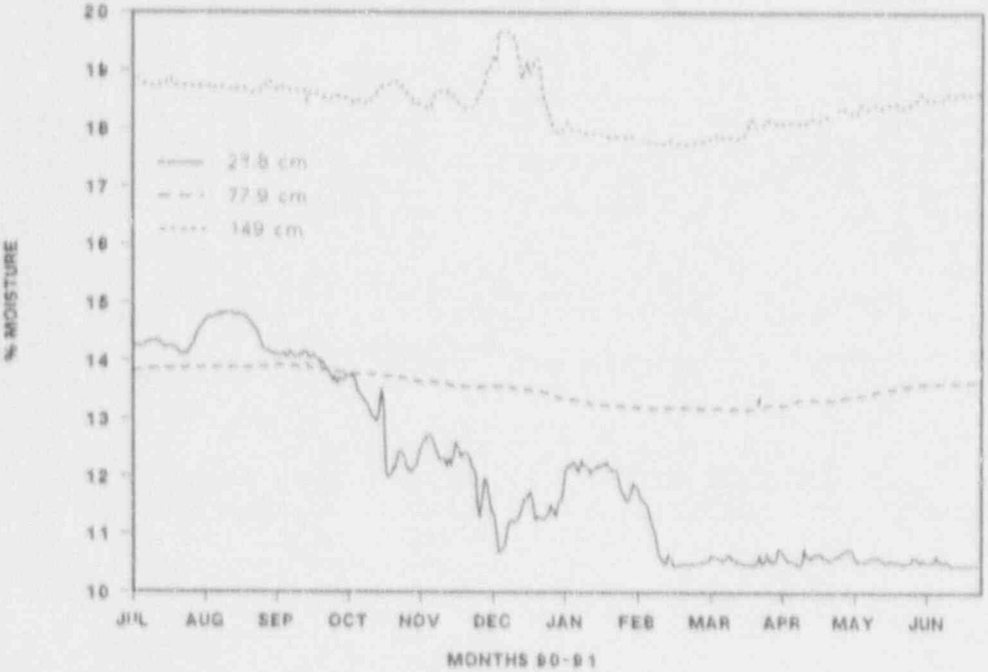

Figure 20. ANL-E lysimeter 1 soil moisture.

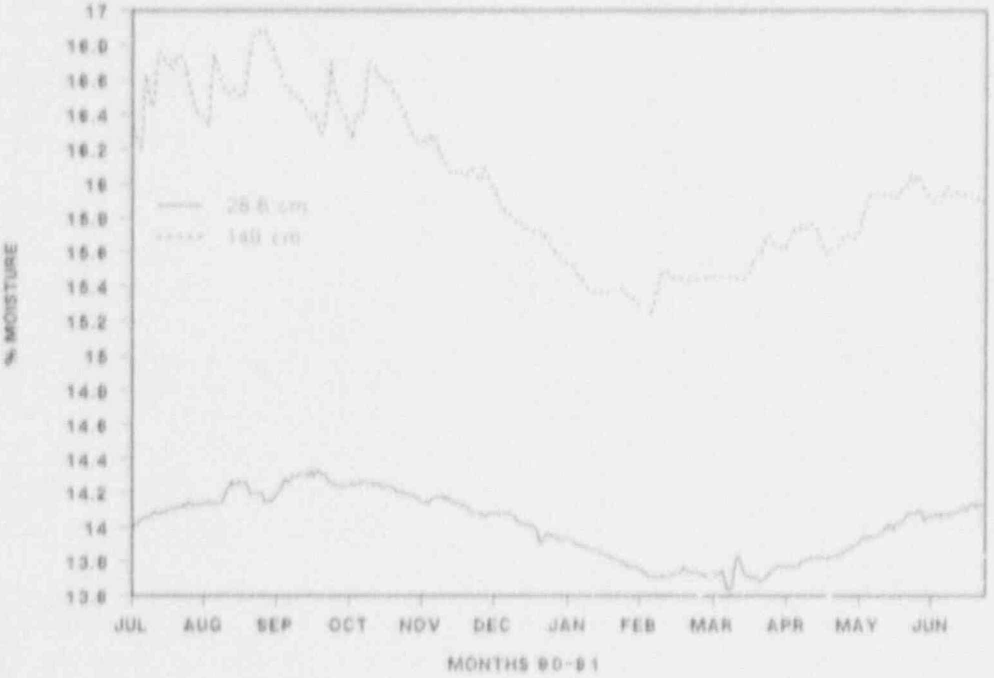

Figure 21. ANL–E lysimeter 2 soil moisture.

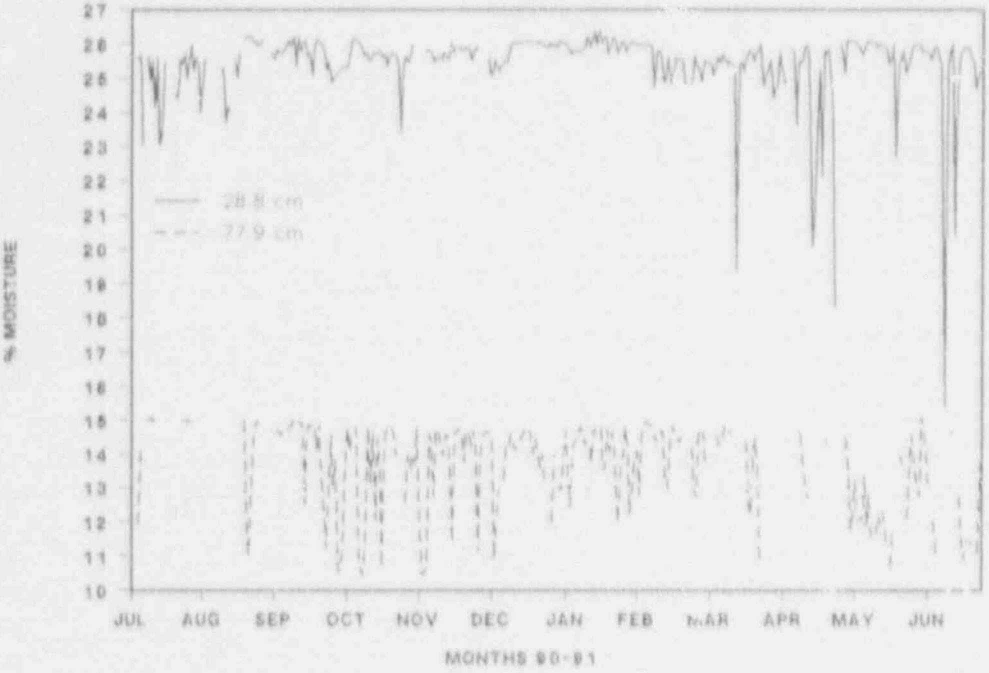

Figure 22. ANL–E lysimeter 3 soil moisture.

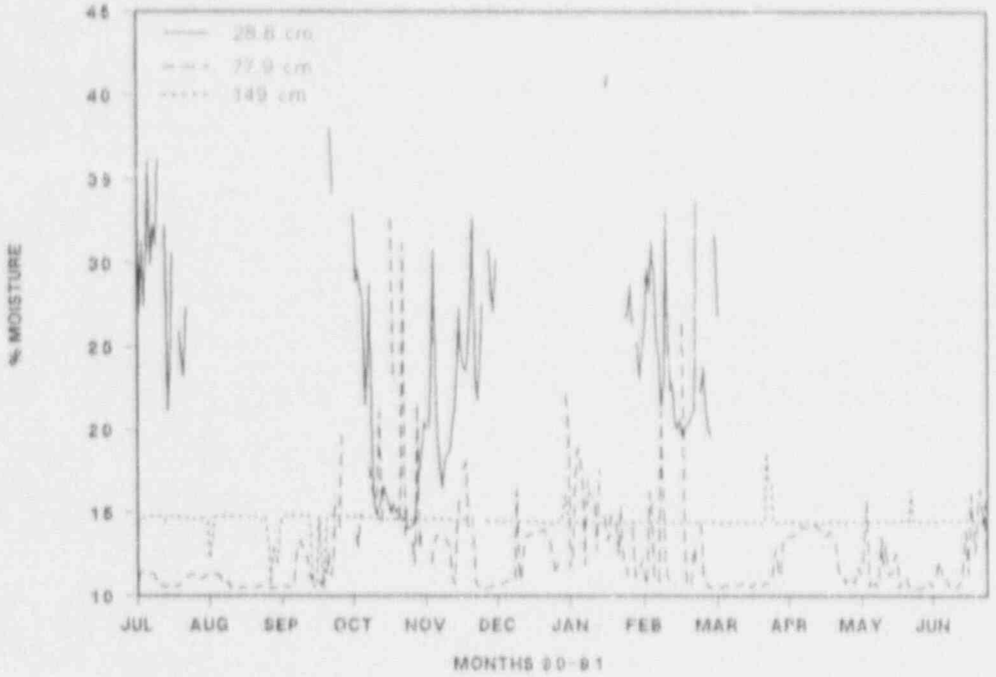

Figure 23. ANL–E lysimeter 4 soil moisture.

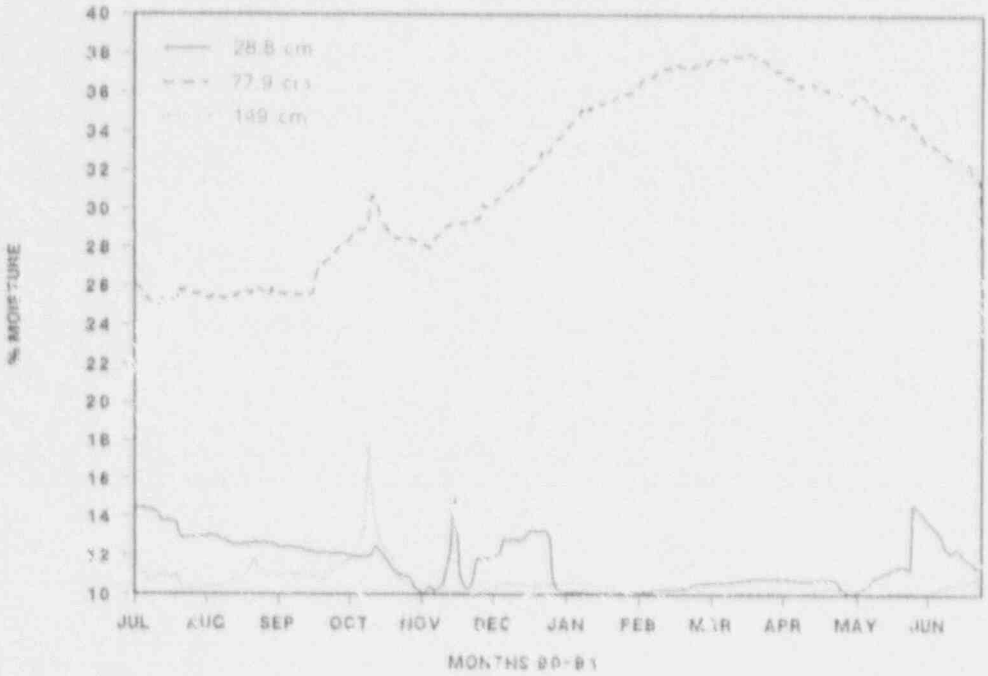

Figure 24. ANL–E lysimeter 5 soil moisture.

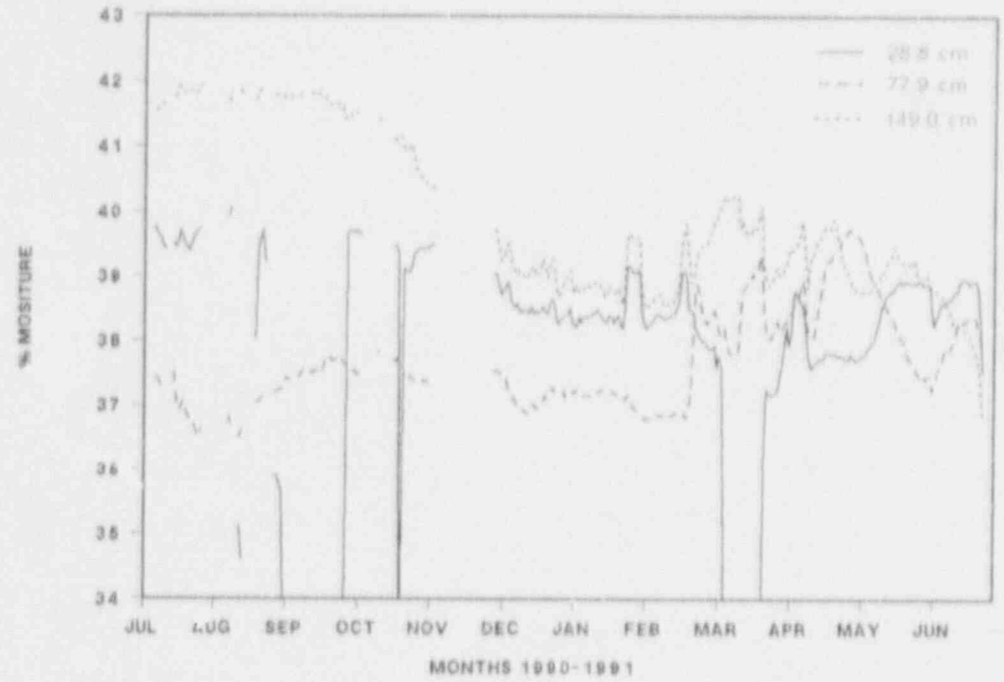

Figure 25. ORNL lysimeter 1 soil moisture.

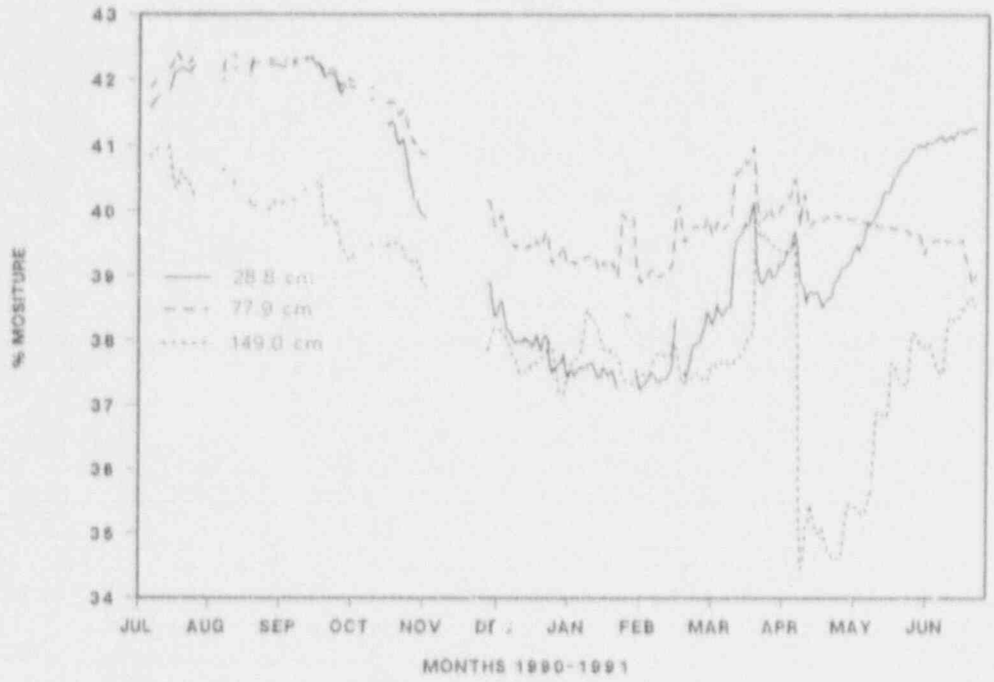

Figure 26. ORNL lysimeter 2 soil moisture.

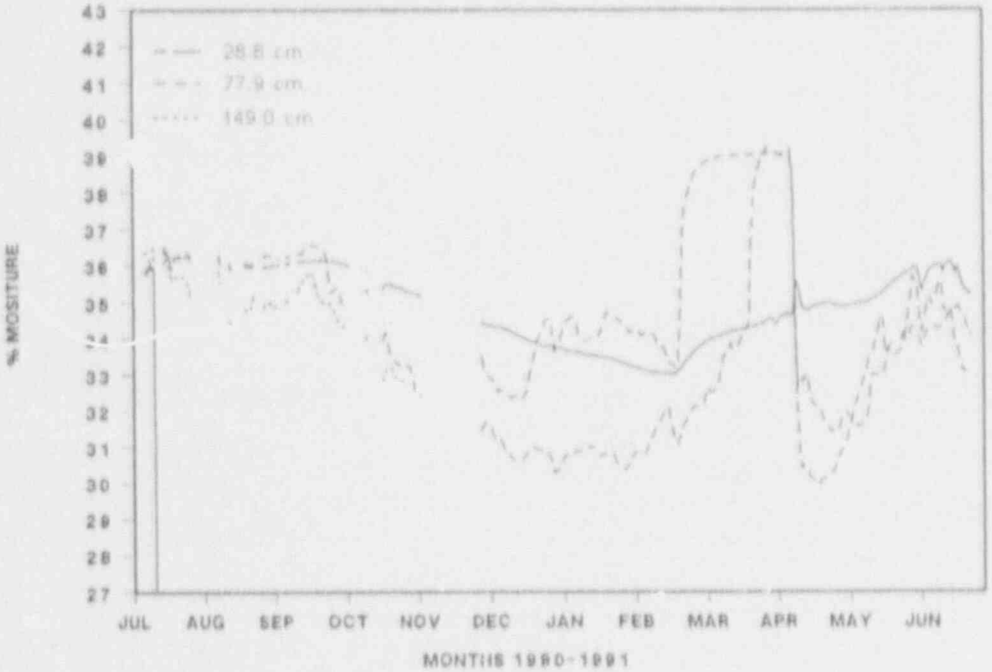

Figure 27. ORNL lysimeter 3 soil moisture.

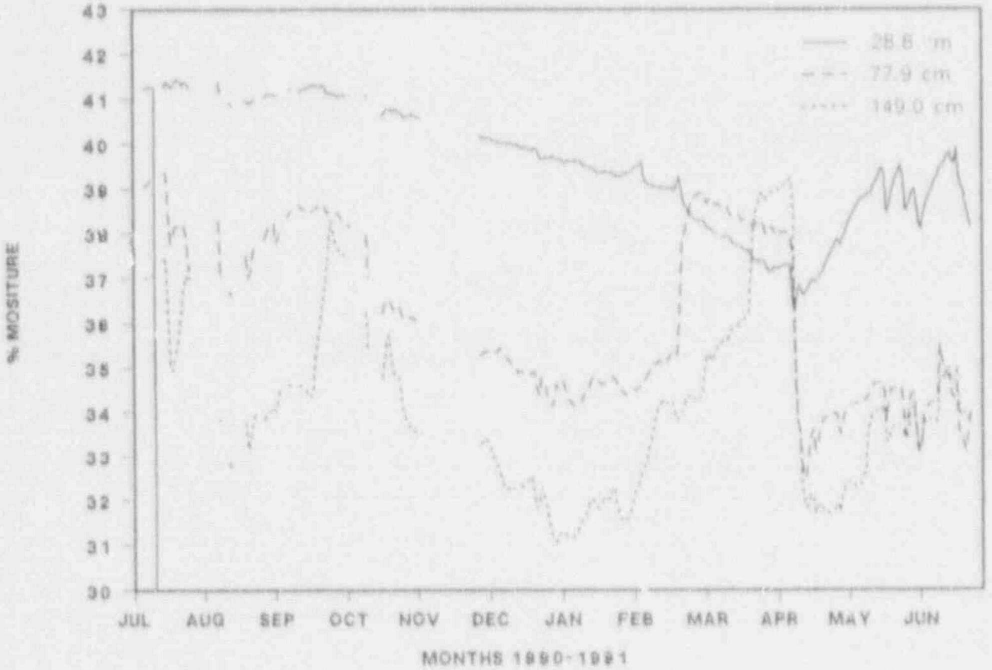

Figure 28. ORNL lysimeter 4 soil moisture.

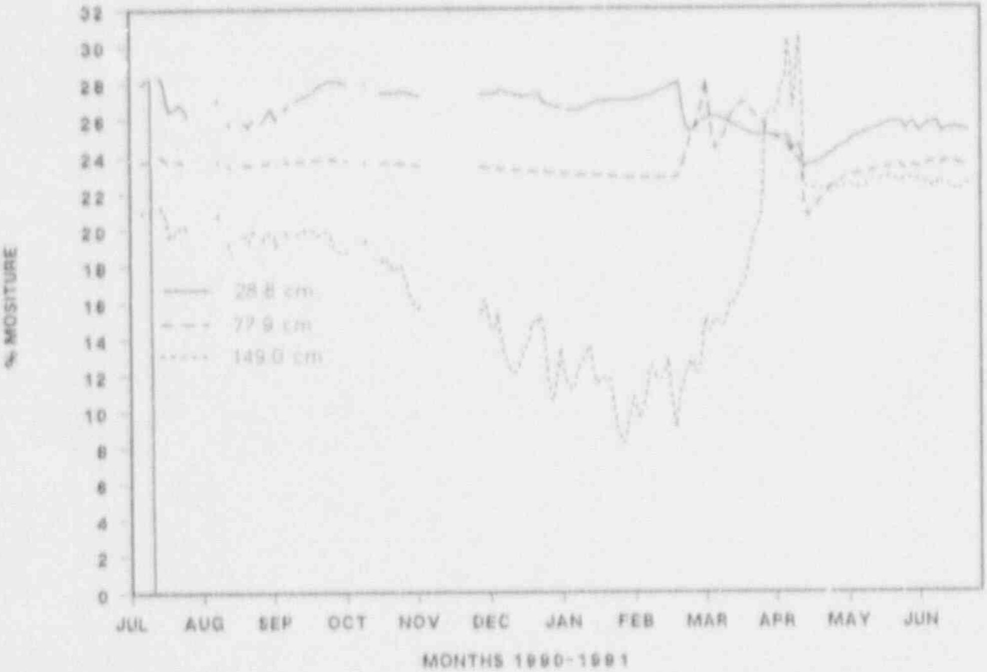

Figure 29. ORNL lysimeter 5 soil moisture.

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 moisture detected for the lysimeters at each site was relatively similar and not excessive. There was a coefficient of variation of 37.4% at ANL–E and 19.6% at ORNL. The probes continue to serve their original purpose of providing some indication of the status 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.[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 probe data for ANL–E lysimeters range between 3.2 and 19.2%. These values are still within a reasonable range given the use of the information. While data from the ORNL probes continue to overestimate the actual percent soil moisture, these data have remained consistent year after year.

In addition to using the probe and gravimetric data to calculate soil moisture during the summer of 1991, a neutron moisture detecting probe was used at ANL–E. Operation of the neutron probe using initial calibration curves produced data that was comparable to gravimetric values (see Table 2). A new calibration curve will be calculated, and it is expected that the neutron probe data will fit more closely with gravimetric measurements.

Soil moisture (as gravimetrically determined) at each sampling depth has remained uniformly consistent between intrasite lysimeters during the past several years (Figures 30 and 31). The uniformity of soil moisture in the ANL–E lysimeters (Figure 30) continues to be of interest given the long–term, nonuniform decrease in water infiltration into ANL–1, 2, 4, and now 3 as well as the drying of the surface soil horizon due to the

Table 2. Moisture profile of ANL–E lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	% Moisture (dry weight) Gravimetric	% Moisture (dry weight) Neutron probe
1	0–41	11.0	
1	41–62	14.9	14.5
1	62–82	17.9	17.9
1	82–107	19.1	
1	107–133	19.0	
1	133–153	20.4	20.8
1	153–182	21.4	
1	182–202	21.5	22.5
2	0–41	11.2	
2	41–62	11.3	17.5
2	62–82	14.4	20.3
2	82–107	18.7	
2	107–133	19.7	
2	133–153	20.0	22.6
2	153–182	21.1	
2	182–202	20.6	23.5
3	0–41	12.3	
3	41–62	15.5	19.1
3	62–82	20.5	23.0
3	82–107	21.8	
3	107–133	20.2	
3	133–153	20.6	23.4
3	153–182	19.6	
3	182–202	22.1	24.2
4	0–41	13.0	
4	41–62	17.6	19.1
4	62–82	20.4	21.4
4	82–107	21.5	
4	107–133	21.5	
4	133–153	22.1	22.8
4	153–182	22.7	
4	182–202	22.8	23.2

a. Samples were collected on July 16, 1991.

Table 3. Moisture profile of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	%Moisture (dry weight) gravimetric
1	0–25	15.0
1	25–50	16.2
1	50–75	17.1
1	75–100	18.1
1	100–125	17.8
1	125–150	18.4
2	0–25	15.1
2	25–50	16.4
2	50–75	16.9
2	75–100	16.7
2	100–125	17.8
2	125–150	18.4
3	0–25	15.4
3	25–50	15.8
3	50–75	16.9
3	75–100	16.9
3	100–125	17.2
3	125–150	18.1
4	0–25	15.0
4	25–50	15.9
4	50–75	16.6
4	75–100	17.1
4	100–125	17.7
4	125–150	18.3

a. Samples were collected on August 5, 1991.

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 1991	Percent difference between actual and probe
ANL 1	14.7 ± 3.3	18.2 ± 3.4	19.2
ANL 2	15.0 ± 1.0	17.1 ± 4.2	12.3
ANL 3	19.6 ± 8.3[b]	19.0 ± 3.4	3.2
ANL 4	17.3 ± 6.4	20.2 ± 3.4	14.4
ORNL 1	38.0 ± 1.7	17.1 ± 1.3	122.2
ORNL 2	39.4 ± 1.1	16.9 ± 1.1	133.1
ORNL 3	34.2 ± 0.9	16.7 ± 1.0	104.8
ORNL 4	36.4 ± 2.7	16.8 ± 1.2	116.7

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a. July 1990 through June 1991.

b. Average from two probes.

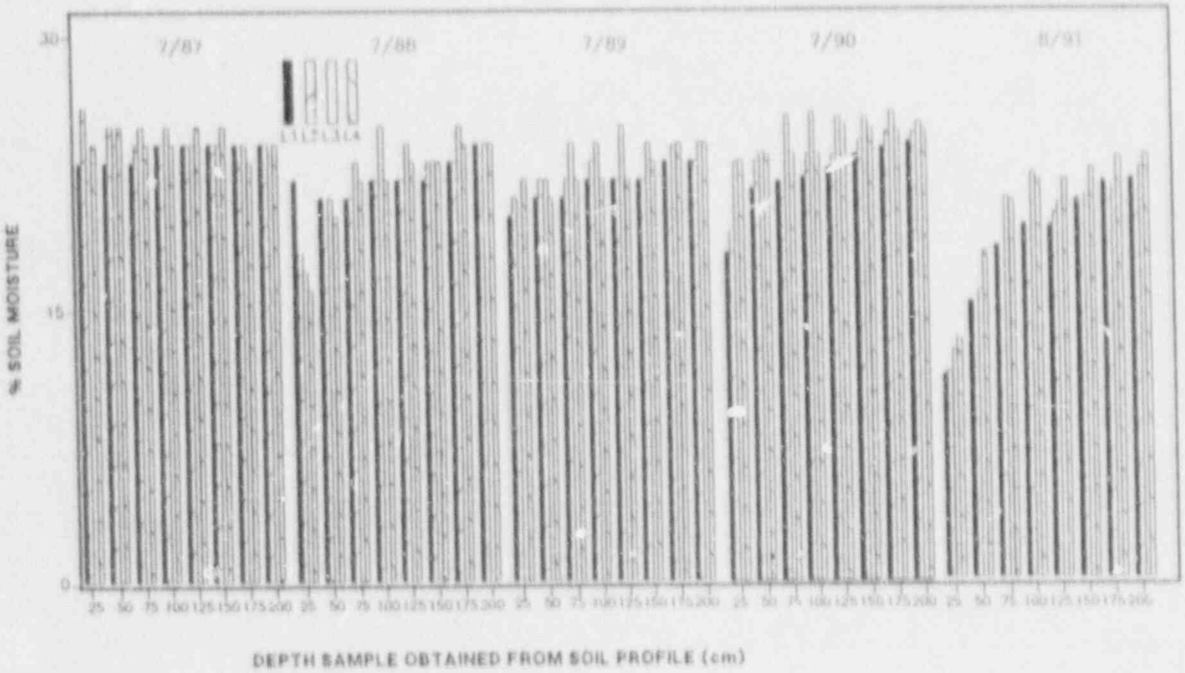

Figure 30. Moisture profile of ANL–E lysimeters 1 through 4 by year based on gravimetric measurement of water content.

Figure 31. Moisture profile of ORNL lysimeters 1 through 4 by year based on gravimetric measurement of water content.

lack of summer precipitation. Action to improve drainage of these lysimeters has been taken; however, it is obvious that initial drainage rates cannot be restored. Observations of surrounding indigenous soils have confirmed that this soil has a low permeability. Therefore, the present conditions are now thought to be indicative of what would be found if a disposal trench were constructed in the same soil. It was decided in FY 1989 that no further efforts would be 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. Total quantities of water removed from the three lysimeters during the year were

- ANL 1, 395 L
- ANL 2 363 L
- ANL 3, 74 L
- ANL 4, 273 L.

It is apparent from data presented in Figures 30 and 31 that after initial wetting, the water storage 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 8, 9, 10, 11, 12, and 13). However, due to a lack of summer precipitation, there was a decrease in quantity of water stored in the ANL–E lysimeters. At the time of the 1991 sampling, the average soil moisture of ANL–E soils had decreased from 56.1% to 45.8% of the soil moisture holding capacity (MHC) while at ORNL this value remained approximately the same: 39.2% for 1990 and 38.0% for 1991. These values have remained fairly constant from year to year.

By using the cumulative rainfall data from each site since the time the lysimeters were placed in operation (Figure 10), it is possible to calculate the approximate volume of water that has been received by the exposed lysimeter surfaces (6489.5 cm^2). The cumulative volume of precipitation received by each ANL–E lysimeter was

3451.1 L; at ORNL, this value was 5008.6 L. The volume of the precipitation that has passed through the lysimeters can be seen in Figures 32 and 33. It has become apparent with time that the throughput of precipitation is dependent on site conditions and lysimeter fill material. At ANL–E, an average of 1468.7 ± 608.2 L, with a range of 27.3 to 66.4% of total precipitation, has passed through the soil lysimeters, while for the control, this value was 3529 L or 102.3% of the calculated available precipitation. For ORNL, the values were 4509.0 ± 34.2 L (90.0%) for the soil-filled lysimeters and 5203 L (103.9%) for the control. These data are comparable to the previous year's data (Reference 13). 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 in Figures 32 and 33). Also, the small deviation in total 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 total volumes of precipitation that have moved through the lysimeters represent an average of [illegible].07 pore volumes for the ANL–E soil lysimeters and 6.35 pore volumes for soil lysimeters at ORNL, while the controls at ANL–E and ORNL were 7.67 and 8.05 pore volumes, respectively.

Radionuclide Analysis. Water samples are normally collected on a quarterly basis from leachate collectors and moisture cups of each of the lysimeters during the 12-month period. During this reporting period, however, sampling occurred in three of the four quarters. At each sampling, only water from the leachate collectors (1 L of collected quantity) and those cups (0.1 L or as noted of the collected quantity) closest to the waste forms (cup 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

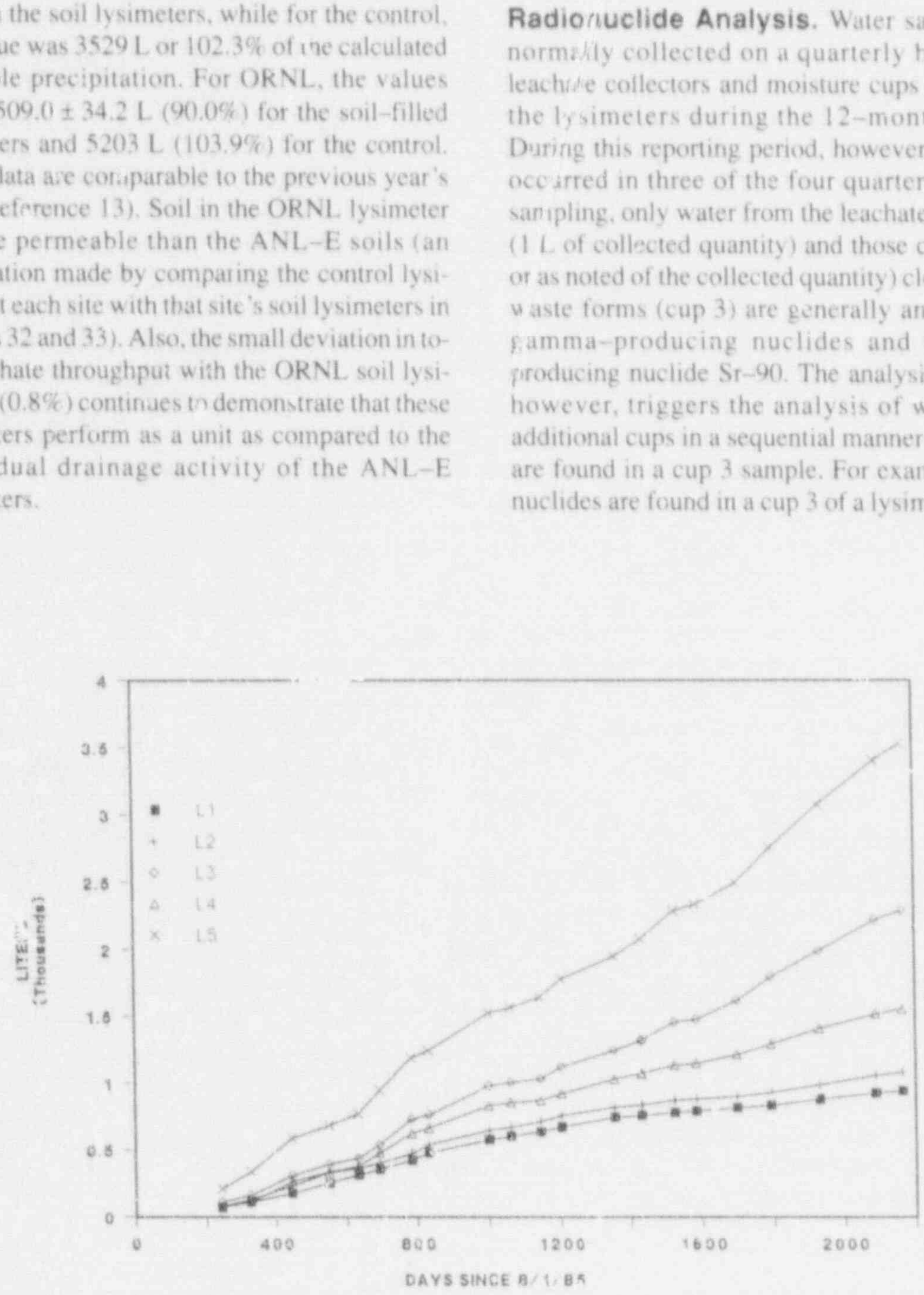

Figure 32. ANL–E cumulative volume of leachate from lysimeters.

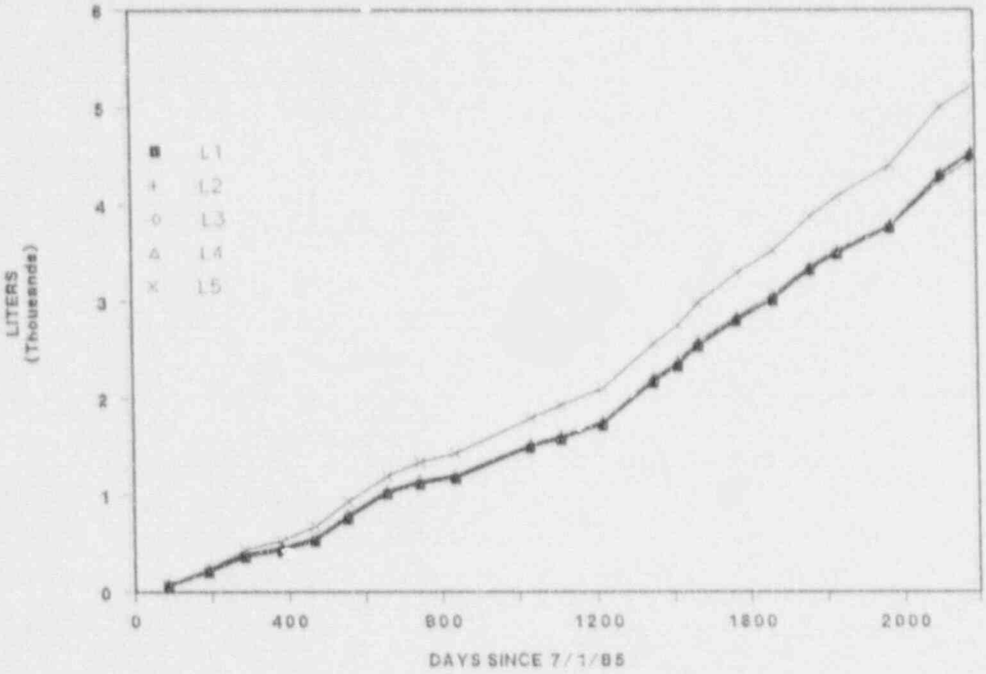

Figure 33. ORNL cumulative volume of leachate from lysimeters.

from cup 1 (directly below cup 3), then cup 4, followed by cup 2 (see Figure 6 for cup placement) should be analyzed. Because of funding levels, however, it has not been possible to follow this protocol. Until recently, only water samples from cups 3 are routinely analyzed at the sites. During this sampling period, water from cups 1 was also analyzed and reported.

Tabulated results of beta and gamma analysis for the samples taken during the period are found in Tables 5 and 6. Three samples were taken at each site during the 12-month period. The cumulative amounts of nuclides as determined in water samples obtained from lysimeter cup 3 and leachate collectors for all sampling periods are displayed graphically in Figures 34 through 42. It should be noted that water samples are once again being obtained from ORNL cup 3-3, which malfunctioned during the previous year. The lack of data from this cup during the 1989–1990 sampling period could give the erroneous impression that Sr-90 was not obtained from this source during that period of time (Figure 35).

As has been reported in the past,[9,10,11,12,13] 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 continue in all cups 3 at both ANL-E (range of 14 to 132% increase) and ORNL (range of 51 to 620% increase) (Tables 5 and 6; Figures 34 and 35). There continues to be standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a cumulative total of 747,622 pCi from 3-3 at ANL-E (an increase of 38% over last year) (Figure 34) and 61,604 pCi from 1-3 at ORNL (65% increase over last year) (Figure 35). It is of interest that the releases into ANL 3-3 and ORNL 1-3 are almost linear, indicating a continuance of an established rate of release. In addition, a significant increase in SR-90 (472% increase) release is now occurring in ORNL 3-3 (Figure 35).

During the past 12 months, leachate water from the [illegible] (sand-filled) lysimeters at each site contained amounts of Sr-90 at least an order

Table 5. Results of beta and gamma analysis of ANL soil moisture and leachate samples, year 6 (1990 – 1991).

Sample identification	Concentration (pCi/L)[a]								
	Co-60			Cs-137			Sr-90		
	Nov	Apr	July	Nov	Apr	July	Nov	Apr	July
Lys 1[b]	<20	<20	<20	<15	<15	<15	<1	<1	<1
Lys 2	<40	<20	<40	<35	<15	<35	<1	<1	<1
Lys 3	<40	<40	<20	<35	<35	<15	39 ± 1	43 ± 1	39 ± 2
Lys 4	<20	<40	<40	<15	<35	<35	<1	<1	59 ± 4
Lys 5	<20	<40	<20	<15	<35	<15	565 ± 4	661 ± 4	576 ± 8
Lys 1–3[c]	<30	<20	<20	<20	<15	272 ± 60	7732 ± 34	7892 ± 41	4055 ± 31
Lys 2–3	<50	<20	<40	322 ± 41	184 ± 26	509 ± 60	3420 ± 27	3767 ± 37	4169 ± 36
Lys 3–3	<50	<40	<40	<50	<35	232 ± 90	8.6E5 ± 3340	1.0E6 ± 3161	8.8E5 ± 2871
Lys 4–3	<50	<20	<20	<50	<15	<15	7144 ± 47	9046 ± 53	1.1E4 ± 58
Lys 5–3	<50	<40	<40	2.3E4 ± 1318	3.9E4 ± 2498	5.6E4 ± 2480	1.1E4 ± 216	1.1E4 ± 177	1.7E4 ± 197
Lys 1–1[c]	<50	<40	<20	<50	<35	<15	<10	<10	NA[d]
Lys 2–1	<30	<20	<20	<20	<15	<15	<10	<10	<10
Lys 3–1	<50	<40	<40	<50	<35	<35	570 ± 12	1350 ± 23	876 ± 7
Lys 4–1	<30	<20	<20	<20	<15	<15	<10	<10	<10
Lys 5–1	<30	<40	<40	<20	<35	<35	808 ± 17	621 ± 15	961 ± 7

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a. Concentration ± 2 sigma

b. 1–L subsample from leachate collector

c. Total moisture cup sample size is ≈ 0.1–L.

d. Sample not available for analysis.

Table 6. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 6 (1990–1991).

Sample identification	Concentration[a] (Pci/L)					
	CO–60			Cs–137		
	Nov 90	Apr 91	Jun 91	Nov 90	Apr 91	Jun 91
Lys 1[b]	0.5 ± 5.9	0.8 ± 5.9	29.7 ± 27.0	–0.8 ± 5.7	1.6 ± 5.7	8.1 ± 35.1
Lys 2	0.5 ± 4.1	0.5 ± 4.1	22.4 ± 23.8	–0.8 ± 3.5	0.8 ± 4.1	8.1 ± 35.1
Lys 3	6.5 ± 4.3	–1.6 ± 6.5	21.6 ± 29.7	2.2 ± 4.9	1.4 ± 5.7	8.1 ± 35.1
Lys 4	3.0 ± 5.9	0.8 ± 6.2	2.7 ± 37.8	–0.8 ± 7.0	1.1 ± 6.5	8.1 ± 37.8
Lys 5	–2.7 ± 5.9	0.3 ± 5.1	16.2 ± 37.8	24.1 ± 4.3	37.8 ± 5.4	156.6 ± 32.4
Lys 1–3[c]	6.2 ± 21.6	10.8 ± 37.8	19.7 ± 22.1	8.6 ± 22.7	5.4 ± 32.4	5.4 ± 29.7
Lys 2–3	21.6 ± 32.4	2.7 ± 37.8	8.1 ± 40.5	27.0 ± 27.0	–8.1 ± 37.8	10.8 ± 35.1
Lys 3–3	13.5 ± 32.4	15.1 ± 21.6	25.3 ± 24.3	8.1 ± 37.8	–5.4 ± 29.7	2.7 ± 32.4
Lys 4–3	–2.7 ± 29.7	1.9 ± 18.5	5.4 ± 40.5	7.6 ± 25.4	4.6 ± 23.0	2.7 ± 32.8
Lys 5–3	8.6 ± 23.8	21.6 ± 32.5	19.7 ± 23.8	892 ± 54	2271 ± 2970	2970 ± 270
Lys 1–1[c]	11.1 ± 14.1	–7.8 ± 26.2	18.9 ± 40.5	2.7 ± 23.2	7.3 ± 22.7	8.1 ± 40.5
Lys 2–1	–11.1 ± 24.6	8.1 ± 35.1	7.6 ± 26.2	4.6 ± 29.7	–16.2 ± 30.4	0.5 ± 25.9
Lys 3–1	35.1 ± 32.4	–5.4 ± 40.5	1.1 ± 4.9	2.7 ± 29.7	7 ± 37.8	0.5 ± 25.9
Lys 4–1	–2.7 ± 40.5	–8.1 ± 32.4	1.1 ± 5.4	18.9 ± 29.7	2.7 ± 27.0	0.8 ± 3.2
Lys 5–1	–2.7 ± 29.7	–2.7 ± 27.0	2.7 ± 29.7	–5.4 ± 25.1	54.1 ± 21.6	32.4 ± 21.6
Lys 1[b]	<8.1	<8.1	2.7 ± 83	15.7 ± 4.9	15.1 ± 4.6	70.2 ± 10.8
Lys 2	<8.1	<8.1	2.7 ± 92	0.8 ± 2.7	0.5 ± 4.9	13.5 ± 5.7
Lys 3	<8.1	<8.1	2.7 ± 83	0.5 ± 3.8	1.9 ± 3.0	4.8 ± 4.3
Lys 4	<8.1	<8.1	24 ± 95	0.0 ± 4.9	2.4 ± 4.9	51.3 ± 8.1
Lys 5	<8.1	<8.1	54 ± 81	541 ± 27	351 ± 27	486 ± 27

Table 6. (continued).

Sample identification	Concentration[a] (Pci/L)					
	Sb–125			Sr–90		
	Nov 90	Apr 91	Jun 91	Nov 90	Apr 91	Jun 91
Lys 1–3[c]	—[d]	—	—	9.2F4 ± 2703	7.0E4 ± 2703	8.1E4 ± 2700
Lys 2–3	—	—	—	1.0E4 ± 270	8920 ± 270	1.0E4 ± 270
Lys 3–3	—	—	—	1.0E5 ± 2703	9.5E4 ± 2703	1.4E5 ± 2700
Lys 4–3	—	—	—	622 ± 54	351 ± 27	1485 ± 54
Lys 5–3	—	—	—	730 ± 54	351 ± 27	1.1E4 ± 270
Lys 1–1[c]	—	—	—	784 ± 27	1135 ± 54	1701 ± 81
Lys 2–1	—	—	—	12.4 ± 4.9	2.7 ± 9.5	164.7 ± 27
Lys 3–1	—	—	—	1.1 ± 7.6	3.8 ± 7.0	8.6 ± 84
Lys 4–1	—	—	—	10.0 ± 8.9	11.4 ± 10.0	5.7 ± 84
Lys 5–1	—	—	—	27.0 ± 24.3	40.5 ± 13.5	64.8 ± 16.2

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a. Concentration ± 2 sigma.

b. 1–L subsample from leachate collection.

c. Total moisture cup sample ≈ 0.1–L sample size.

d. Sample not available.

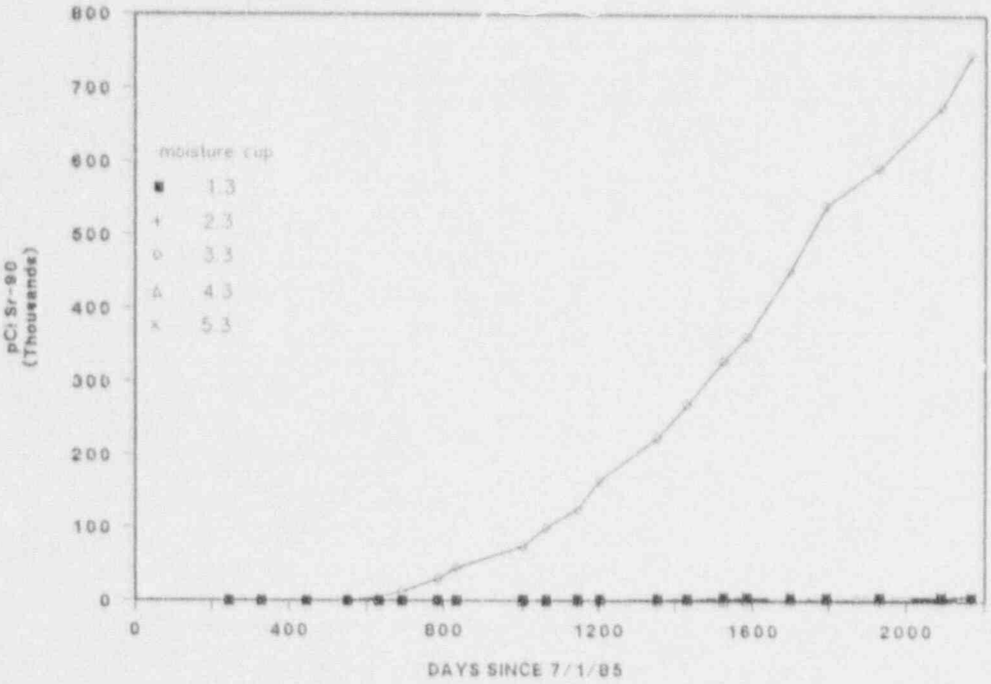

Figure 34. ANL–E cumulative SR–90 collected in moisture cup number 3.

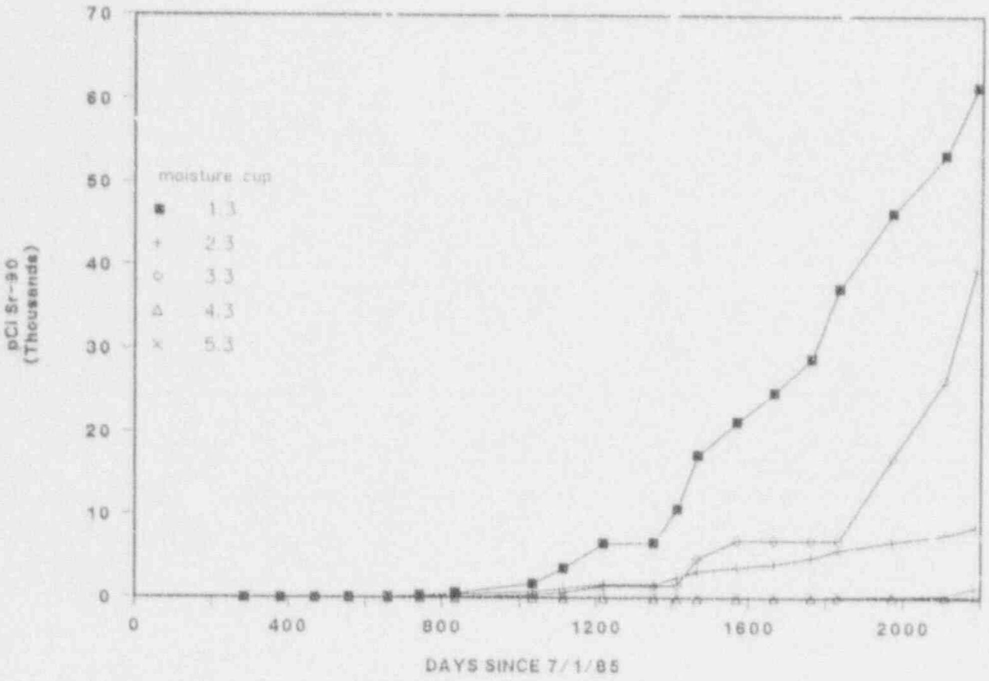

Figure 35. ORNL cumulative SR–90 collected in moisture cup number 3.

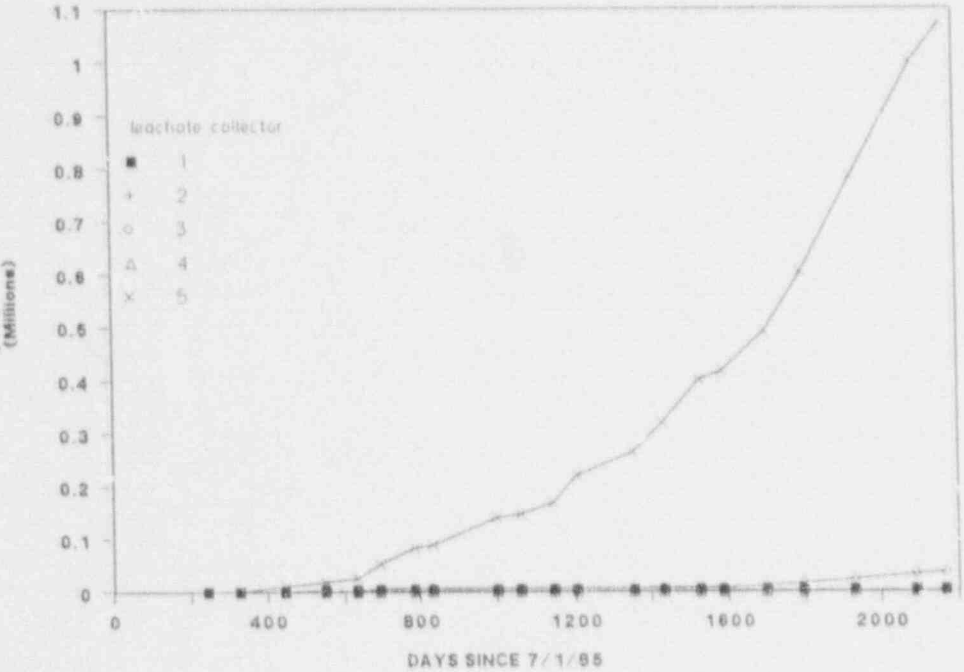

Figure 36. ANL–E cumulative SR–90 collected in lysimeter leachate collectors.

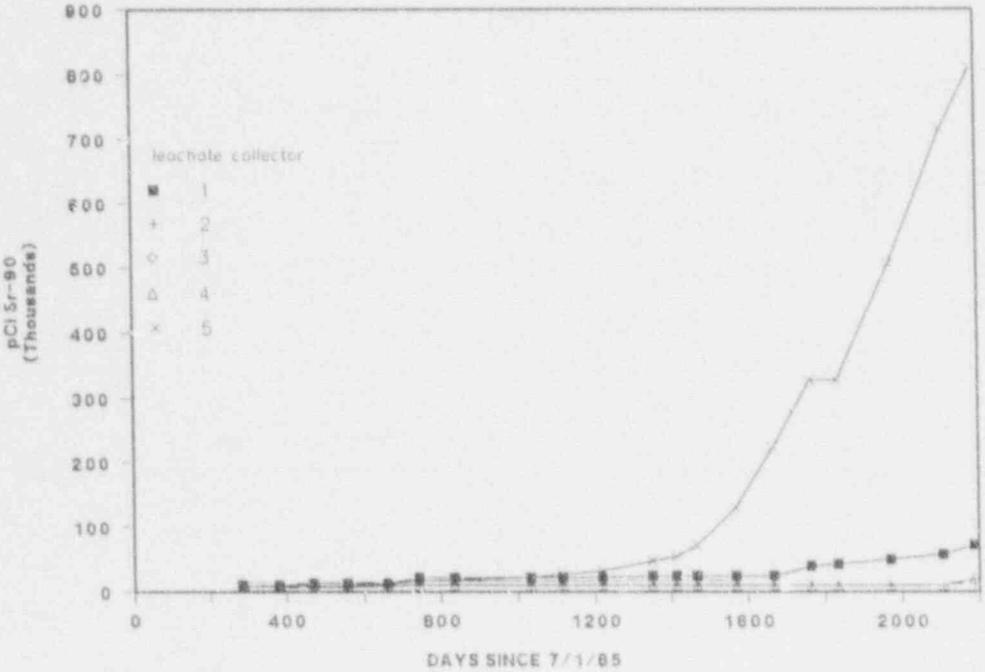

Figure 37. ORNL cumulative Sr–90 collected in lysimeter leachate collectors.

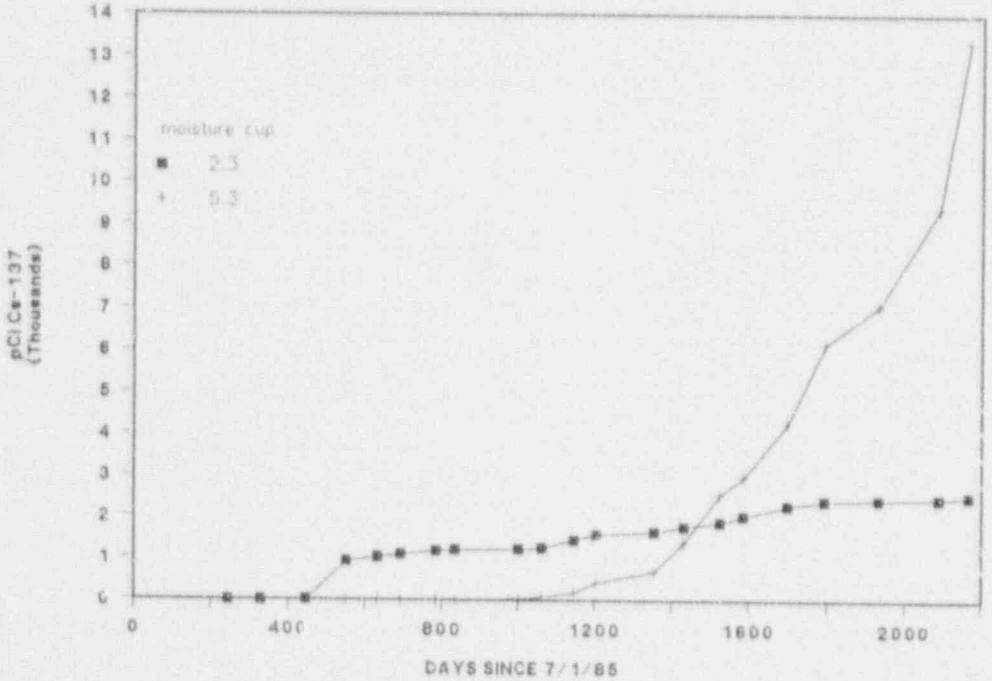

Figure 38. ANL–E cumulative Cs–137 collected in moisture cup number 3.

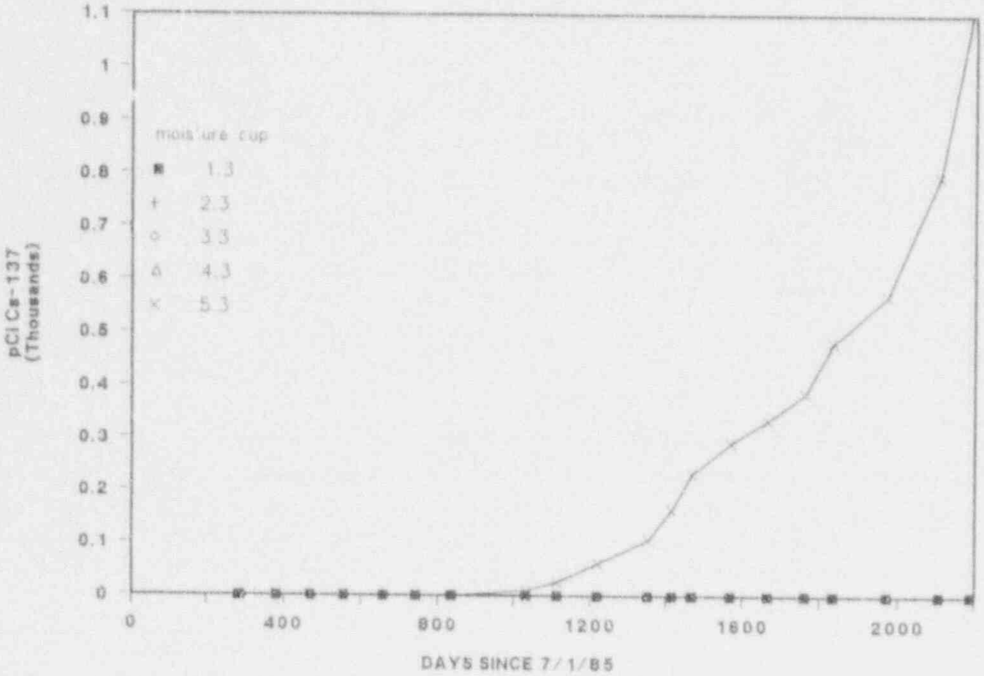

Figure 39. ORNL cumulative Cs–137 collected in moisture cup number 3.

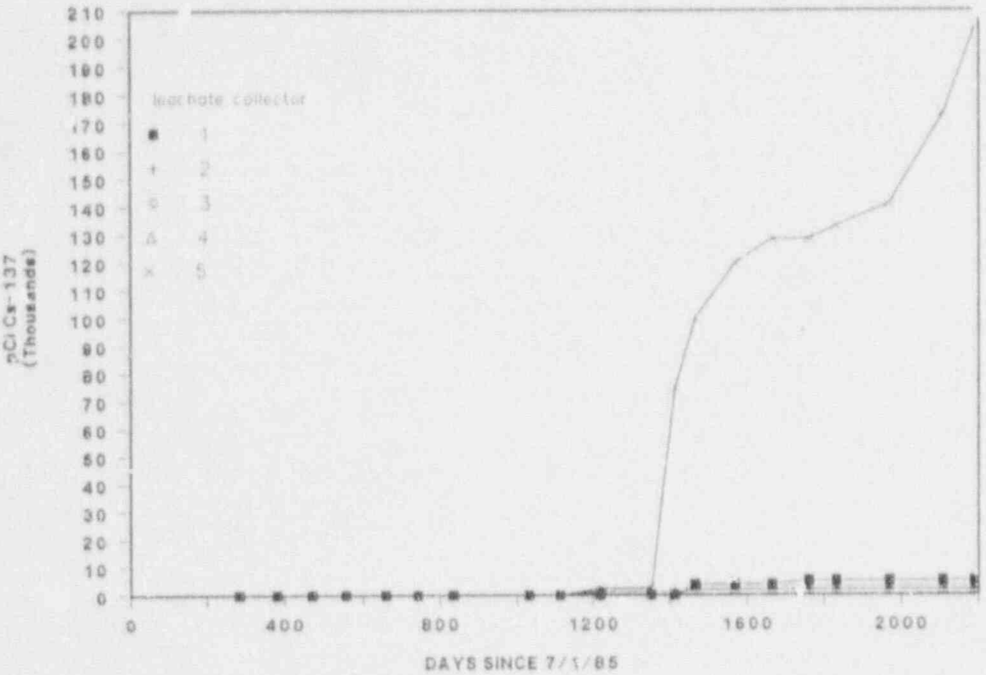

Figure 40. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

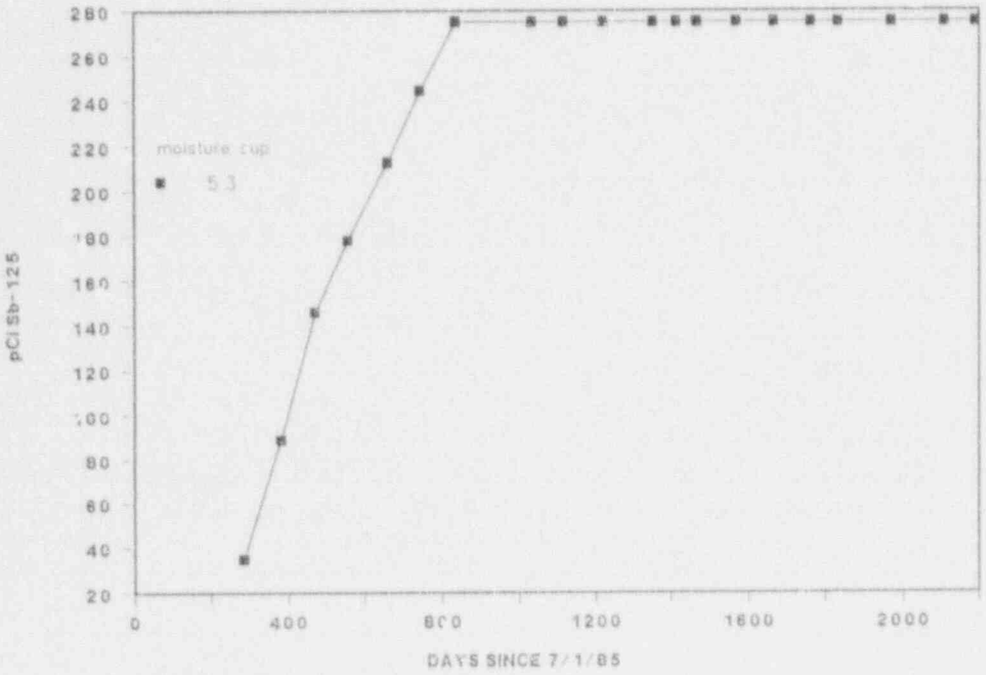

Figure 41. ORNL cumulative Sb-125 collected in moisture cup number 3.

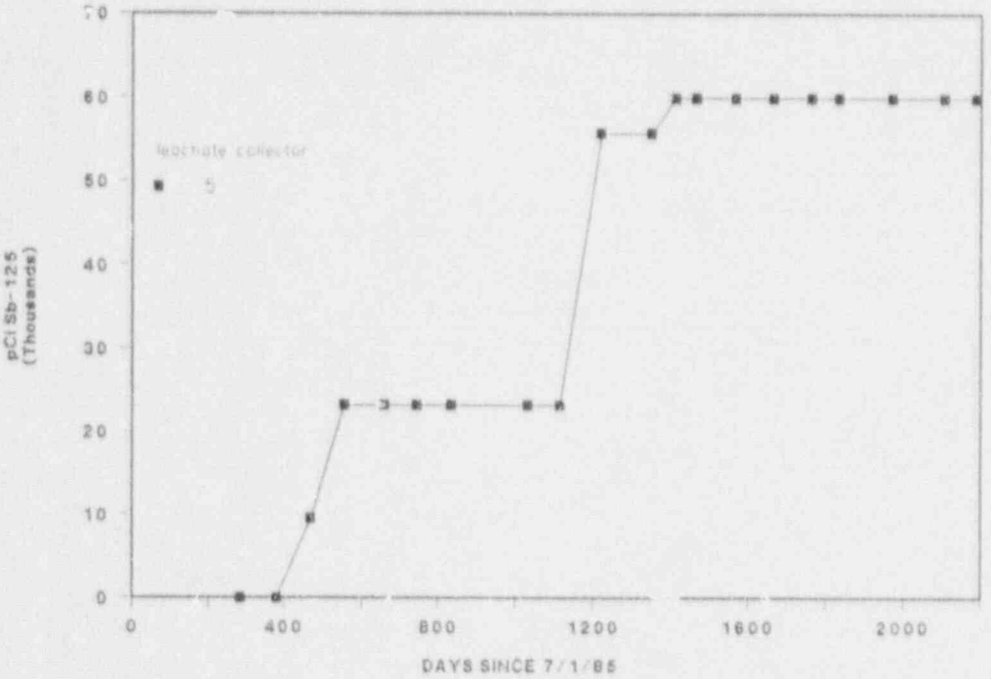

Figure 42. ORNL cumulative Sb–125 collected in lysimeter leachate collector.

of magnitude larger than the soil lysimeters (Figures 36 and 37). This is comparable to the previous year's findings (References 9, 10, 11, 12, and 13). In the soil–filled lysimeters, there were intersite–comparable cumulative amounts of Sr-90 in cups from lysimeters ANL-E 1, 2, and 4 and ORNL 1, 2, and 3 (Table 7). For leachates intersite–comparable quantities of Sr–90 were found in ANL–E 1, 2, and 4 (same as cups) and ORNL 1, 2, and 4 (Table 7). Except for ANL-E 1 and 2 and ORNL 3, there was an increase in the total cumulative quantity of Sr-90 released in the leachate water (Tables 5 & 6). It was noted that the leachate water from ANL-E 4 received a 748% increase in Sr-90 over that of last year. For the ORNL lysimeters 1, 2, and 4, the total amount of the nuclide released in leachate water was comparable to or greater than that in the cups. These data follow a trend seen in some of last year's data and make it appear that a pulse of Sr-90 could be moving through the soil columns of the ORNL lysimeters.

The percent of total Sr–90 being measured in the leachate water and cup 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 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 four times that of ANL (Table 7).

Gamma–producing nuclides continue to occur with regularity at ANL–E and are again present at ORNL. ANL 2–3, below a cement waste form containing large amounts of Cs–137, continues to receive Cs–137 (Table 5), with another significant increase in the quantities of this nuclide appearing again this year (Figure 38). Cesium–137 began appearing in ANL 5–3 in 1987. The quantity of this nuclide has increased in each of the sampling periods since that time,[15] with abrupt increases during the last two years (Figure 38). There continues to be no sustained occurrence of Cs–137

Table 7. Comparison of total Sr–90 and Cs–137 inventory per lysimeter to total amounts in lysimeter water.

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	ORNL	ANL	ORNL	ANL	ORNL	ANL	ORNL
1	Cement	34	339	27	401	—	—	—	2.0
2	Cement	113	264	49	396	0.2	—	—	0.1
3	VES	2729	146	134	64	—	—	—	0.7
4	VES	120	8	54	456	—	—	—	0.1
5	Cement	49	42	5887	25000	4.3	0.1	—	14.2

in any of the ANL leachate water. Detectable amounts of Cs–137 have been consistently found in water from ORNL–5 and sporadically in the other ORNL waters though not during this sampling period (Figure 40 and Table 6). Measurable amounts of Cs–137 began to occur in ORNL 5–3 during the May 1988 sample and have continued in subsequent samplings for a total of 1095 pCi (128% increase over last year). Breakthrough of Cs–137 into the ORNL 5 leachate collector occurred in November 1988, some seven months after its occurrence in the moisture cup ORNL 5–3 (Figure 40). Thus far, a total of 203,677 pCi have passed through the lysimeter. It should be noted that the rate of Cs–137 occurrence in both ORNL 5 leachate water and cup 3 has increased significantly during the past 12 months.

In addition to finding Cs–137 in water from ORNL, both Cs–137 and Sr–90 have been discovered for the first time at the surface of ORNL–5 fill material. Radionuclide activity was detected during a routine gamma survey of the lysimeter surfaces. More activity was found near the center than at the edges. Core samples were obtained from 0 to 2.5 cm and from 2.5 to 5 cm depths at the lysimeter center, for analysis of Cs and Sr–90. Results showed that there was 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 depth 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. There are two possibilities why those nuclides are present at these locations: (a) the contamination was the result of radionuclide–loss during installation of the waste forms; (b) there has been an upward migration from the buried waste forms. The first possibility seems remote, however, since no nuclides were found in a survey after waste form installation and there appears to be no mechanism to initiate the second phenomena of upward movement (i.e., transpiration or evaporation). The contamination appears to decrease with depth. Several factors seem to contradict upward movement. During the reporting period, 100% of the precipitation was recovered in the leachate collector. Experience has also shown that the sand used in the control lysimeters has little or no water retention capability. The sand also has no capillary structure to draw radionuclide containing salts upward.

There have been no occurrences of Sb–125 again this year. This is the second year in a row that Sb–125 has not been found in ORNL 5 leachate water, and the third year for its absence in ORNL cup 5–3.

By using a matrix, several comparisons can be made based on the data. Such a matrix is seen in Table 7, which provides intra– and intersite data. Overall, of the nuclides contained in the waste forms,[8] there has been a greater recovery of Sr–90 in terms of quantity and percent of inventory than other nuclides. Next would be Cs–137 followed by Sb–125 (not listed in Table 7). Compared to Sr–90, the recovery of Cs–137 appears insignificant. There have been occurrences of Cs–137 in cups 3 of the ORNL soil lysimeters during the past two years, but none this year. However, this nuclide has been consistently occurring in ORNL 5–3 (Figure 39) and in the leachate collector of the ORNL–5 lysimeter (Figure 40). Cesium-137 has also occurred in the moisture cups of ANL 2 and 5 but not in the leachate water. More Cs–137 has passed through the ORNL lysimeters than those at ANL.

At ANL–E, a comparison of Sr–90 occurrence in cup 3 and the leachate collectors (Table 7) indicates the beginning of an independence of waste form performance. This behavior might be influenced by the amount of water passing through the ANL-E lysimeters (Figure 32). However, a lack in uniformity is also seen with the ORNL data (Table 7) and these lysimeters have very uniform water movement (Figure 33).

More Sr–90 continues to be found in ANL–3–3 and now ANL–4–3 (VES waste forms) than in the other ANL cups 3 (Table 7). As was noted last year the effect appears to be moderated by the distance traveled in soil from the waste form to the leachate collector. Movement of the nuclide into the leachate of ANL–5 is much greater than that of the other lysimeters, thus providing continued evidence of the moderating effect of soil. Greater quantities of Sr–90 appear to be moving through the ORNL lysimeter in comparison to the ANL–E lysimeter. 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 0.025% of the Sr–90 contained in ORNL–5 has now been recovered in leachate from that lysimeter. Recovery of Sr–90 in the ORNL cups is now about the same for those lysimeters containing the cement waste forms and one of the two containing VES waste forms. These data together with those from ANL 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. A past evaluation has shown that data obtained from the operation of the lysimeters does have a relationship with performance assessment code parameters.[12] The operational lysimeters provide continuous data from the near–field (that area comprised of the waste form and surrounding soil) that directly relates to waste form stability. It was found that information obtained from the data includes the mass balance of released constituents, solubility of radionuclides in a site–specific geochemical system, as well as an indication of the retardation or dispersion of released constituents during transport to the far–field. Also, soil–pore water chemistry (radioactive and inorganic 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 inputs into process level and performance assessment codes since they represent a field data set that contains complete information that characterizes environmental, hydrogeological, geochemical, and waste form effects.

During this reporting period, the collected lysimeter data were used as inputs for the computer code MIXBATH.[16] Use of this model is intended to predict the release of nuclides from a waste form in a failed container surrounded by a porous medium containing a solute. The solute is treated as a well–stirred fluid (i.e., a mixing bath), and solute concentration is calculated using a mass balance that depends on the solute flow rate, the amount of partitioning between the porous medium and solute, the size of the mixing bath, the radioactive decay rate, and the rate of nuclide release from the waste form. Modeling of the waste form is accomplished using a one–dimensional finite difference model. MIXBATH has the capability to simultaneously consider three waste form release mechanisms: diffusion, dissolution, and surface rinse limited by partitioning.

Releases of Cs–137 and Sr–90 from the waste forms were modeled. The most appropriate release process was considered to be diffusion from a cylinder (the shape of the waste forms). The waste form diffusion coefficients for Cs–137 were available from data in reference 15 while those for Sr–90 were obtained based on measurements of similar waste forms of equal size.[19] Calculations for the mass balance of the solute concentration required a Darcy velocity (volumetric flow rate per area), which could not be calculated from the available data. These data were estimated from lysimeter leachate collector analytical data.[13] Soil/water distribution coefficients were estimated from previous published work.[20] Tables 8, 9, and 10 list the values used for the most important parameters. These include the soil/water partition coefficients (K_d) and decay constants, the diffusion coefficients (D) for each waste form and isotope, and the Darcy velocities of the soils. The K_d values used were assumed to fall between the upper and lower boundaries for the model parameters in soils (Table 8). With the Unimin sand, the best curve fit was obtained using an assumed $K_d = 0$. It should be noted that the VES waste form diffusion coefficient for Sr–90 listed in Table 9 is approximately six orders of

Table 8. Partition coefficients (cm^3/g) of three soils used in lysimeters.

Radionuclide	Value used	Model parameters Lower boundary	Model parameters Upper boundary
Morley silt loam			
Cs–137	10^{3}	10^{1}	10^{5}
Sr–90	$10^{0.9}$	10^{0}	10[illegible]
C horizon of fuquay sandy loam			
Cs–137	10^{3}	10^{1}	10^{5}
Sr–90	$10^{0.9}$	10^{0}	10^{3}
Unimin silica oxide sand (inert material)			
Cs–137	0[a]	10^{1}	10^{5}
Sr–90	0[a]	10^{0}	10^{3}

[a] The value assumed for essentially inert material.

	Decay constants (s^{-1})
Cs–137	7.28×10^{-10}
Sr–90	7.57×10^{-10}

Table 9. Diffusion coefficients of waste forms and radionuclides used in lysimeters (cm^2/s).

Waste form	Radionuclide Cs–137[a]	Radionuclide Sr–90[b]
Vinyl ester–styrene	3.30×10^{-14}	1.35×10^{-8}
Portland type I–II cement	5×10^{-11}	4×10^{-10}

[a] See Reference 15.

[b] See Reference 19.

Table 10. Darcy velocities of soils used in lysimeters.

Lysimeter no.	Darcy velocity (cm/s)	
	ANL	ORNL
1	9.42×10^{-7}	3.07×10^{-6}
2	1.10×10^{-6}	3.10×10^{-6}
3	1.65×10^{-6}	3.12×10^{-6}
4	1.34×10^{-6}	3.16×10^{-6}
5	2.59×10^{-6}	3.60×10^{-6}

magnitude larger than that for Cs–137. The cause for this discrepancy is the use of a literature value for the Sr–90 and a bench leach test value for Cs–137. This highlights the necessity of using waste–form–specific parameteric values.

Results of this preliminary lysimeter performance assessment modeling produced data for which the parametric information available was broad enough for accurate predictions. Of course, there were also data in which predicted and measured values were in poor agreement. Such differences appeared to be the result of a lack of waste–form–specific diffusion coefficient data, together with the low cumulative concentration of nuclides in some of the lysimeter leachate waters. Figure 43 shows plots of predicted and measured Sr–90 cumulative activity versus time for ORNL 5. Two predictions are shown using diffusion coefficients of 4×10^{-10} cm^2/s and 5×10^{-11} cm^2/s. With the latter, the MIXBATH prediction and measured values agree within one order of magnitude. Releases of this magnitude appear to be consistent with those measured during other work using these waste forms.[15] Use of the diffusion coefficient of 4×10^{-10} cm^2/s gave results that were five orders of magnitude greater than actual values. These data indicate that the determination and use of waste–form–specific diffusion coefficients for model input is important.

The results as shown in Figure 43 indicated that there is insufficient cumulative radionuclide activity as of this reporting period for code validation. However, there were sufficient data to show similarities between the predicted and measured curves. These plots appear to be typical of the predictions made about Sr–90 release from both cement and VES waste forms. Strontium–90 diffusion coefficients used for prediction are probably much greater than actual values. Data from the lysimeter project have indicated that for VES, Cs–137 and Sr–90 diffusion coefficients are probably of the same order of magnitude.

Data from a comparison of cumulative Cs–137 activity from ORNL–3 appears to give a reasonable prediction (Figure 44). This demonstrates how the measured value of the diffusion coefficient and close approximations of the partition coefficient (Table 8) can significantly increase the accuracy of the prediction. From these data and those from Sr–90, it appears that MIXBATH performed adequately for the purposes of this preliminary performance assessment. It helped identify those areas in which additional data (diffusivity values, soil K_d values, and soil hydraulic properties) will be required in order to use the lysimeter data effectively in performance assessment modeling.

One other fact that the model has shown is that data on this project have not been gathered for a significantly long period of time to provide indications of future trends. It is projected that several more years of data collection will be required for development of a satisfactory data base. This conclusion is strengthened when there is a comparison of nuclide releases between the soil and sand–filled controls. It is apparent from the low activity present in leachate waters collected from the soil lysimeter as compared to waters collected

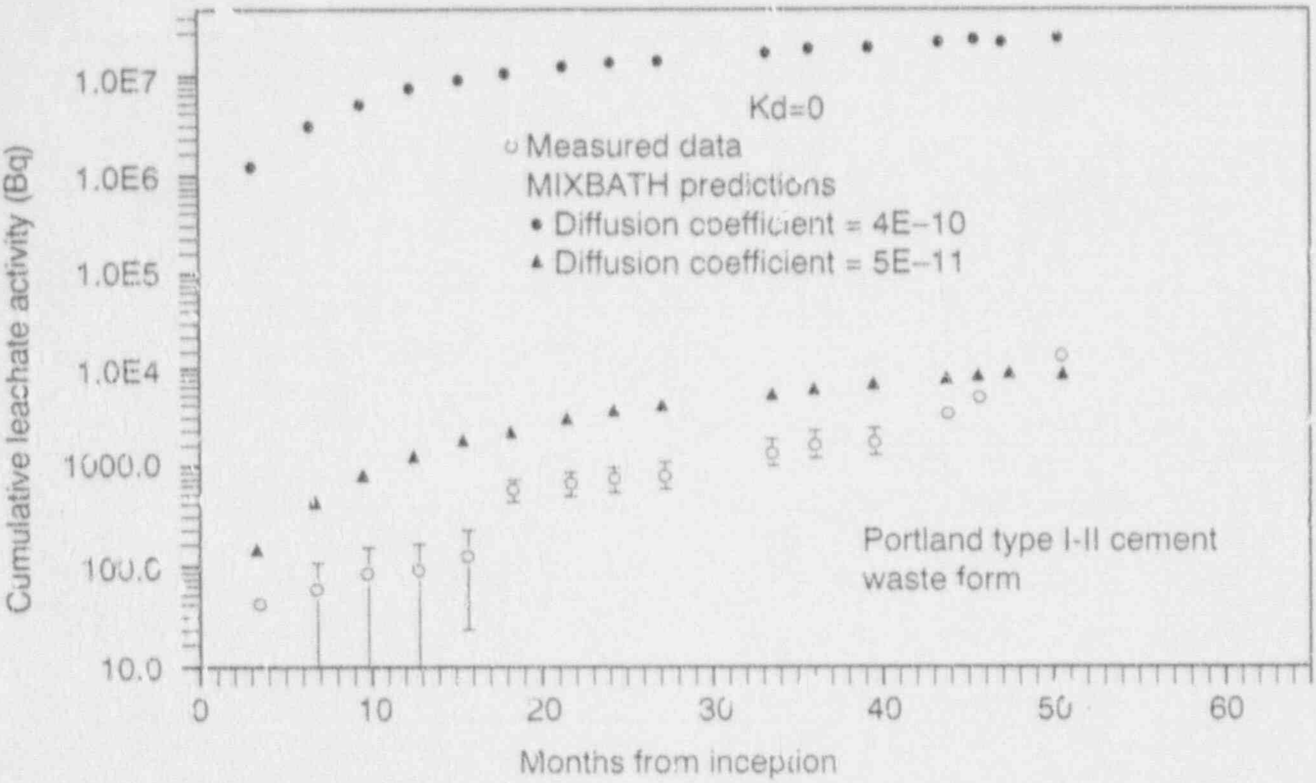

Figure 43. Comparison of Sr–90 cumulative activities for measured data from ORNL lysimeter 5 leachate collector MIXBATH predicted results

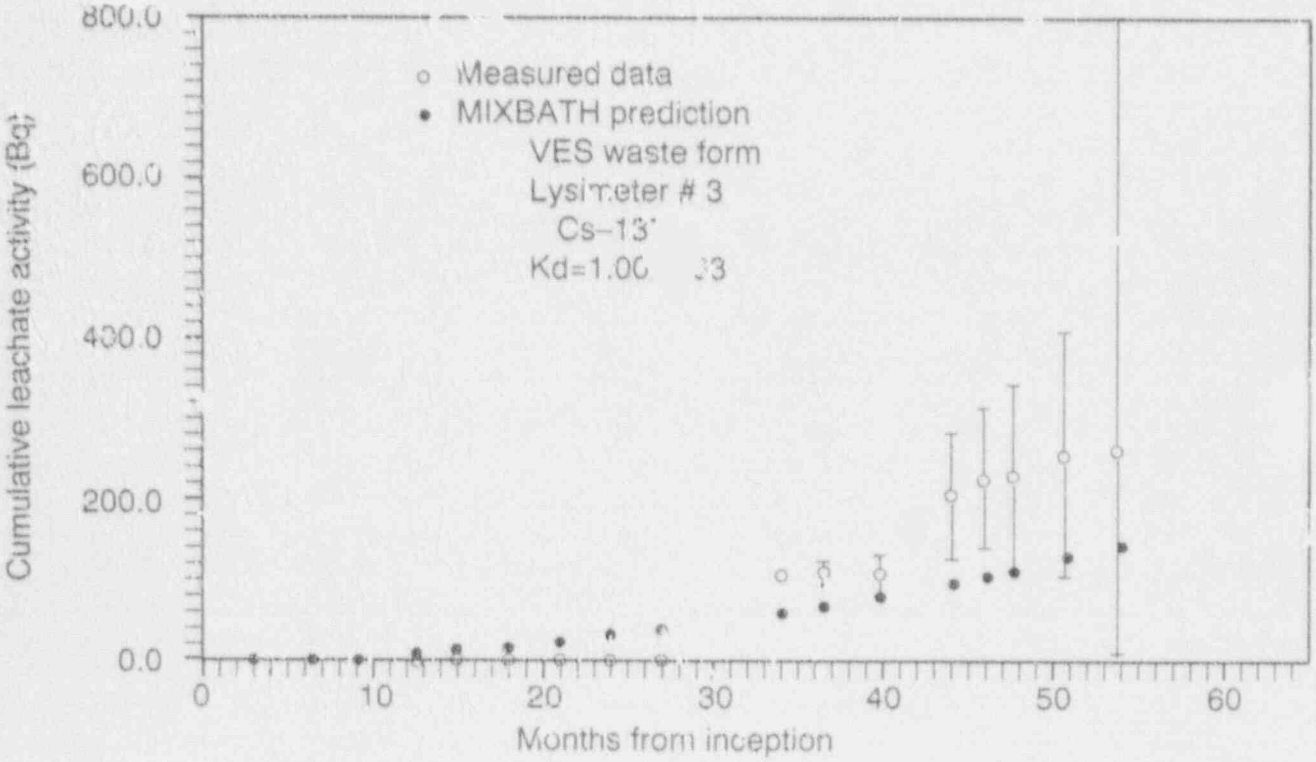

Figure 44. Comparison of Cs–137 cumulative activities for measured data at ORNL lysimeter 3 leachate collector MIXBATH prediction.

from the sand lysimeter that the main body of activity has not yet migrated to the bottom of the soil lysimeter and could require years to do so.

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 1991 was analyzed for the major ionic species listed in Table 11. The justification for the choice of ions is also provided in the table. At ANL–E, cups 1 and 3 were sampled on lysimeter 1; cups 1, 3, and 5 on lysimeters 3 and 5; and cups 2 and 3 on lysimeters 2 and 4 (because cups 1 were inoperative in those lysimeters). Cups 1 and 3 water samples were sampled in 1991 at ORNL. Data from ANL–E in 1991 and ORNL in 1988 showed that ionic concentrations in the soil water were not introduced by the precipitation (see Reference 12 and Table 12). It appears that the waste forms could have been 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 12 and 13 and

Table 11. Ionic species analyzed for in 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, 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 12. ANL–E results of chemical speciation, lysimeter moisture cups 1, 2, 3, and 5, July 1991.

		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)
RAIN 1	—	2.3	.22	.038	<.5	1.0	.37	5.5	<3	3.7
RAIN 2		3.0	.21	.045	<.5	1.2	.38	5.5	<3	3.7
Lys 1–1	Cement	89	13	14	<.5	52	3.7	<.5	<3	45
Lys 1–3		68	12	9.7	<.5	39	3.0	<.5	<3	42
Lys 2–2	Cement	89	13	13	<.5	52	4.9	<.5	<3	41
Lys 2–3		82	19	15	<.5	62	3.2	<.5	<3	57
Lys 3–1	VES	85	6.6	11	<.5	48	2.1	3.0	<3	28
Lys 3–3		79	8.2	16	<.5	47	8.2	<.5	<3	28
Lys 3–5		84	3.4	16	<.5	48	1.8	<.5	<3	28
Lys 4–2	VES	80	6.5	10	<.5	45	4.6	<.5	<3	40
Lys 4–3		81	4.4	10	<.5	45	2.4	<.5	<3	41
Lys 5–1	Cement	5.5	0.6	11	<.5	2.8	0.7	6.2	<3	7.0
Lys 5–3		6.8	10	30	<.5	3.6	1.1	5.5	<3	6.9
Lys 5–5		4.4	0.8	25	<.5	2.1	0.9	12	<3	7.0

Table 13. ORNL results of chemical speciation for lysimeter moisture cup 1 and 3, July 1991.

Sample	Solidification agent	Cation					Anion			
		Ca (mg/L)	Na (mg/L)	[illegible] (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	41	5.2	20	0.09	1.5	0.91	0.57	0.2	26.0
Lys 1–3		53	4.5	26	2.59	1.7	0.55	47	0.2	21.3
Lys 2–1	Cement	40	3.7	20	0.05	1.2	0.79	13.0	0.2	9.6
Lys 2–3		36	4.7	34	2.47	1.1	[illegible]	6.9	0.2	7.6
Lys 3–1	VES	34	1.9	22	0.09	0.9	0.85	46	0.2	6.7
Lys 3–3		120	4.9	31	0.38	2.0	2.43	39	0.2	1.5
Lys 4–1	VES	5.4	4.8	16	0.15	0.8	[illegible]	1.64	0.2	15.8
Lys 4–3		4.9	6.9	16	0.15	1.0	1.33	1.14	0.2	[illegible]
Lys 5–1	Cement	9.2	0.3	10	1.24	3.4	4.03	1.96	0.2	4.2
Lys 5–3		11	2.3	29	2.47	4.2	0.79	7.77	0.2	1.0

Figures 45, 46, 47, and 48). It appears that the cement and VES waste forms performed similarly at both sites. With a few exceptions, the ORNL 1991 cation data (Table 13 and Figure 47) closely resemble those of 1988 and 1989. However, the 1989 and 1991 anion concentrations (Table 13 and Figure 48) were considerably decreased from 1988 and actually showed little of the cup-to-cup variability found in 1988. ANL-E 1991 data are similar, in most cases, to ORNL 1991 data when compared in Figures 45, 46, 47, and 48. While these early data are interesting, no correlation can be made with radionuclide movement as yet.

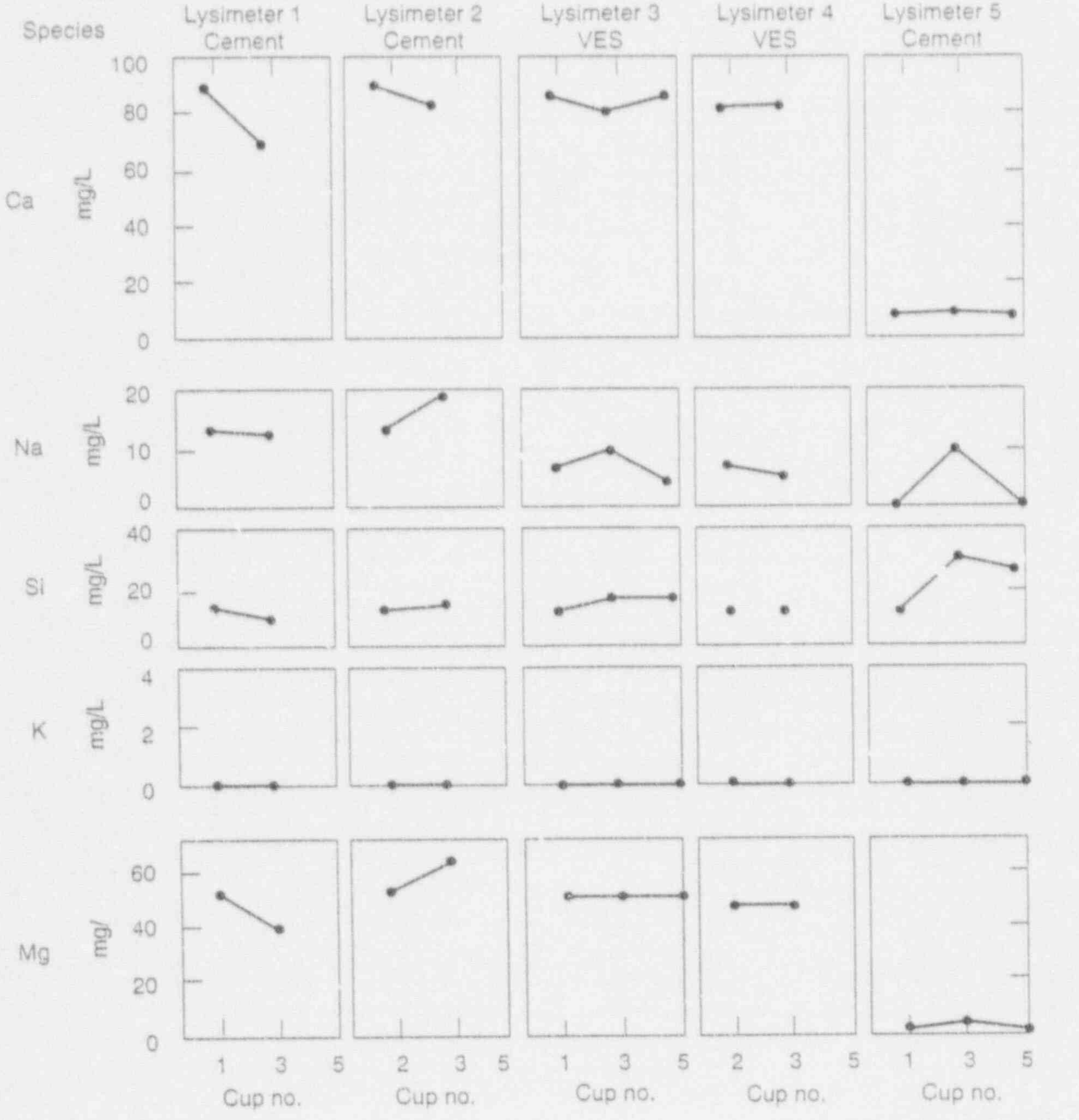

Figure 45. Results of chemical speciation at ANL-E-cations.

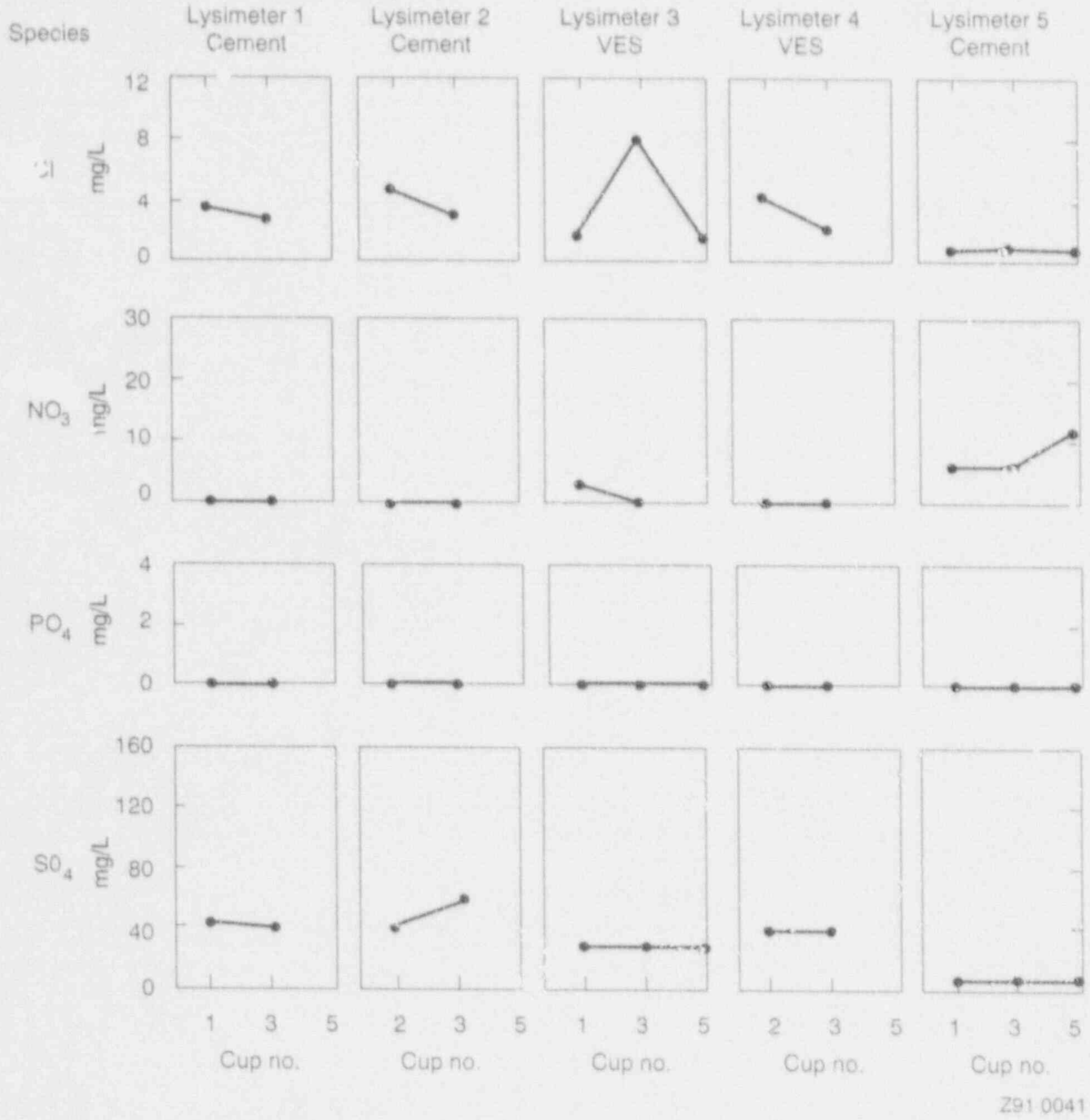

Figure 46. Results of chemical speciation at ANL–E–anions.

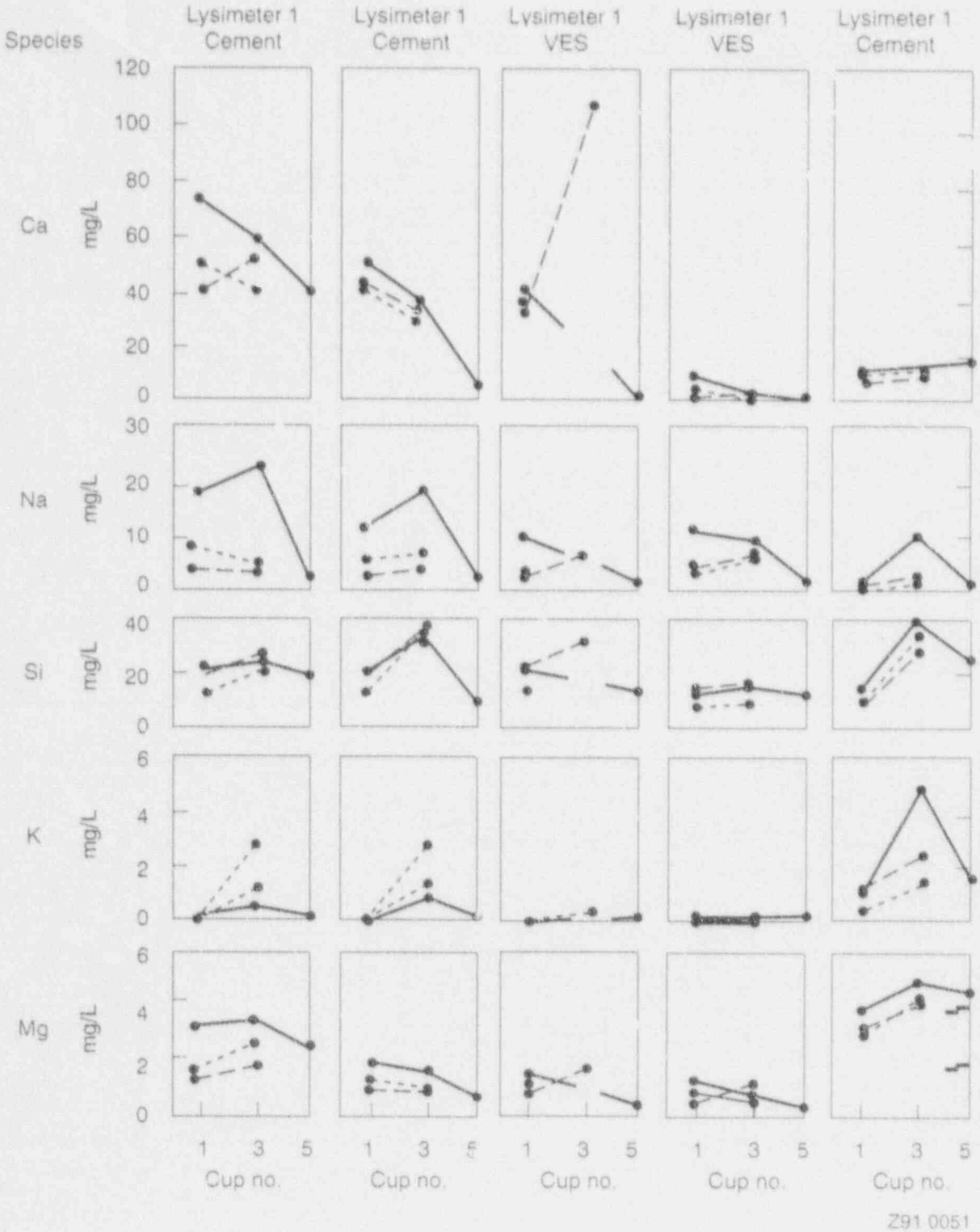

Figure 47. Results of chemical speciation at ORNL–cations.

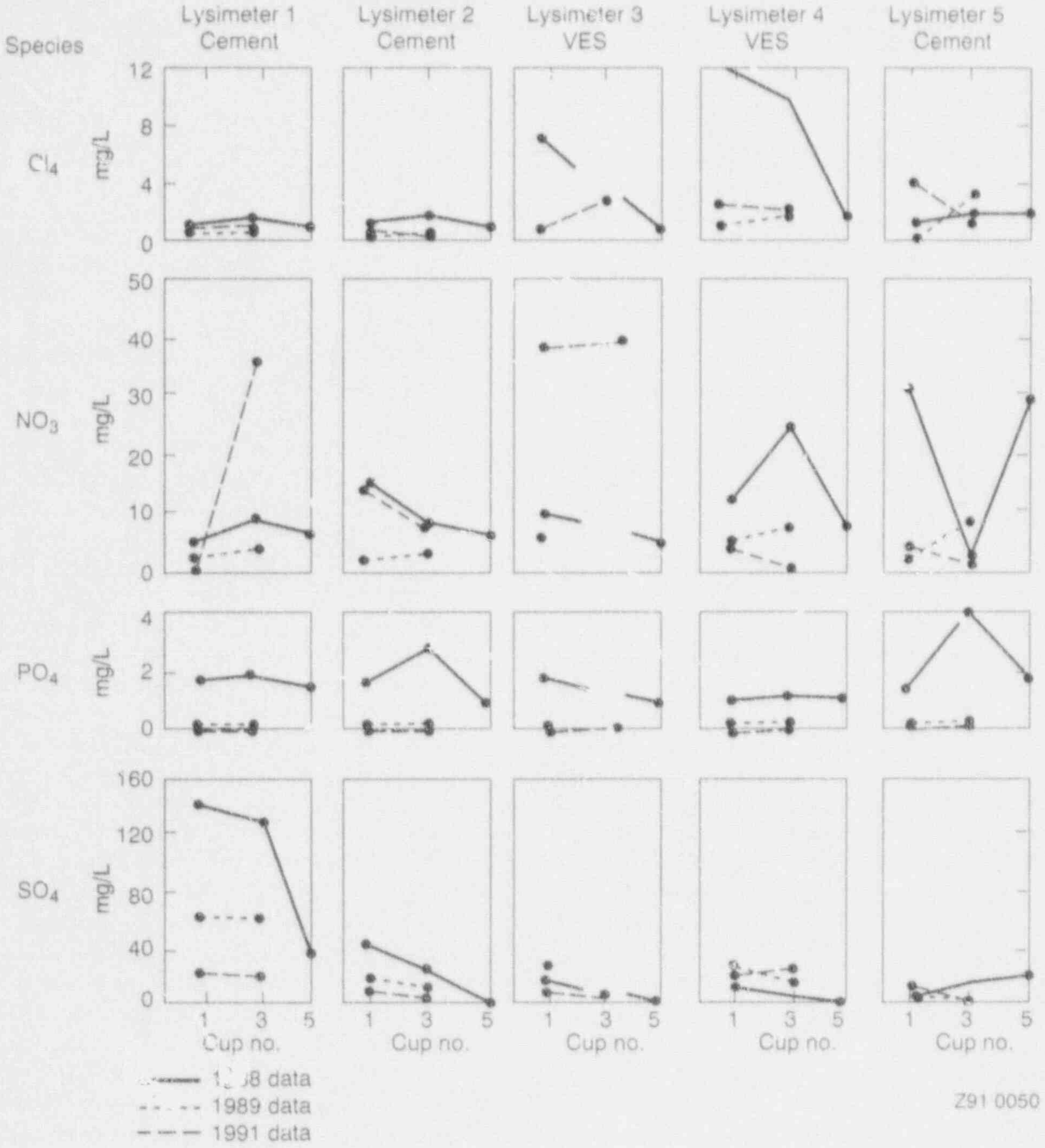

Figure 48. Results of chemical speciation at ORNL–anions.

CONCLUSIONS

Resin Solidification

No new conclusions were generated for compression testing under resin solidification.

Field Testing

Lysimeter operation during the sixth year at ANL-E and ORNL has been successful. Analyses of data collected during the past 72 months are continuing to show a pattern in nuclide availability and movement such that the cumulative data are beginning to provide insight on waste form performance.

There has been 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. Compared to Sr–90, the occurrence of Cs–137 appears insignificant. There have been occurrences of Cs–137 in the moisture cups of the ORNL during the past two years, but none this year. Cesium–137 has been found in the moisture cup of ANL 2 but not in any leachate waters. More of this nuclide has been collected in leachate water at ORNL than at ANL.

On a cumulative basis, more Sr–90 is being removed from the ORNL lysimeters. This could be a result of the different environmental conditions of the two sites. During the past 48 months, Sr–90 continues to be found in higher concentrations in leachate water from the control lysimeters at both sites, with more found at ORNL. These data continue to reinforce the assumption that the limiting step in receiving Sr–90 in the leachate is not release of the nuclide from the waste forms (since Sr–90 is found in cup 3 samples), but rather it is the soil characteristics (including soil and quantity of soil water) that limit movement.

Recovery of Sr–90 from the ORNL cups is now about the same order of magnitude for those lysimeters containing the cement waste forms and one of the two containing VES waste forms. In general, more cumulative nuclides have been recovered from the two lysimeters containing cement waste forms than from those containing VES. ANL cumulative Sr–90 data also show that comparable amounts of the nuclide have been collected in the moisture cups of lysimeters containing cement waste forms and one of those with VES. However, water from one lysimeter containing VES has had an order of magnitude more cumulative Sr–90 than the other VES or cement containing lysimeters. Cesium–137 has only occurred with consistency in the leachate water of ORNL. Cumulative quantities, while minimal, are comparable for all soil lysimeters. As a conclusion, data from the two sites cannot be used to conclusively demonstrate which type of waste form is preferable for nuclide retention. It appears that at this time that releases of Sr–90 and Cs–137 from cement and VES are comparable.

It was possible to initiate limited performance assessment modeling. In lysimeters with experimentally determined diffusion coefficients, where there were high enough leachate concentrations of nuclides for comparison between predicted and experimental results, the computer code MIXBATH was used to predict the observed leachate concentrations of Cs–137 within one order of magnitude. MIXBATH worked well as a first approximation during performance assessment of the EPICOR–II waste forms. MIXBATH was also successfully used in estimating releases of Sr–90 from EPICOR–II waste forms using estimated diffusion coefficient values for Sr–90.

Results of the modeling effort showed that there is not yet enough long term EPICOR–II lysimeter data on which to base the validity of transport models. It clearly demonstrated several more years of data collection will be necessary to accomplish this task. Also, to limit model/prediction uncertainties, several parameters need to be measured in the laboratory. The most important include the soil partition coefficients and waste–form–specific diffusion coefficients. For more detailed modeling, soil flow parameters, such as hydraulic conductivity (as a function of moisture content) and longitudinal and transverse dispersivity need to be known.

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10. SUPPLEMENTARY NOTES

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 Forms 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 sixth 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 (ANL–E) in Illinois and Oak Ridge National Laboratory (ORNL). 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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