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Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1994

Annual Report

Prepared by
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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 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.

Compressive test results of 11-year-old cement and vinyl ester-styrene solidified waste forms are presented, which show effects of aging and self-irradiation.

Results of the ninth 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.

For resin solidification, Portland type I–II cement and vinyl ester-styrene (VES) waste-form specimens 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, and the results are compared to data obtained from similar waste-form specimens tested earlier in the 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, which has run for 9 years, 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

Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1994 Annual Report

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 the INEL on materials from those EPICOR-II prefilters.[1,2] That work is now funded and 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, tests are being performed to characterize solidified ion-exchange media, and experiments are being conducted to field-test solidified wastes using lysimeters.

The results of resin degradation from studies of the first and second sampling, as described in References 3 and 4,were compared with those of the third sampling described in Reference 5. 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,"[6] provides guidance to waste generators on waste form test methods and acceptable results for

a. References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof.

compliance with the waste form requirements of 10 CFR 61. In this study, EPICOR-II waste forms were subjected to the recommended tests of Reference 6 and, on an annual basis, are subjected to the specified compression test to ensure compliance with stability requirements. The data indicate that the waste form strength has stabilized after increasing with age for 8 years.

Solidified waste forms containing EPICOR-II ion-exchange resin waste are currently being field-tested using lysimeters. The objectives of the field testing task are to (a) examine the performance of the waste forms in typical low-level waste disposal environments, (b) compare field results with bench leach studies and with DOE Special Waste Program field test results, (c) develop a low-level radioactive waste field leach rate data base for use in performance assessment source term calculations, and (d) use the DUST computer code to compare predicted cumulative release to actual field data.

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 and 2). 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 the test period.

This report contains data from 9 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.

As an extension of the lysimeter field-testing task, a Phase 2 experimental design was developed. That second-generation experiment will expose commercial nuclear power station low-level waste to the environments of several typical disposal sites. The preliminary mechanical and instrumentation design, along with installation procedures, were completed during this reporting period and submitted to the NRC.

RESIN SOLIDIFICATION

In this task, EPICOR-II waste forms solidified with Portland type I–II cement and VES are periodically subjected to compression testing per ASTM C39.[7] The specimens were tested dry using sulfur leveling caps poured per ASTM C617.[8] One specimen of each type of ion-exchange resin waste form (all organic and organic resins with zeolite) in each solidification agent (cement and VES) are tested.

On September 29, 1994, four 11-year-old waste-form samples were compression-tested using a Tinuis Olsen 60,000-lb testing machine calibrated on September 28, 1994. The samples had mass and contact radiation dose readings similar to those tested in 1984.[9,10] The test results are presented in Table 1.

The tested specimens are shown in Figures 1, 2, 3, and 4. The concrete specimens broke in a vertical cracking pattern with very little flaking. One VES specimen, D1A-29, bulged in the center of the cylinder as had all previous VES specimens. Vertical cracking took place within the bulging center area. That VES specimen seemed to be very ductile. Examination of the second VES specimen, D2-34, indicates that it failed in brittle fracture (Figure 4). A large vertical crack is evident in this specimen. Similar brittle failures were experienced with the irradiated VES waste forms tested earlier in this program (Reference 10).

Figure 5 shows the strength data from these and previous compressive strength tests[10,11] of specimen age (taken at ages after fabrication) of 1 month, 2 years, 7 years, 8 years, and 11 years. Three of the four sample types experienced no change in strength during the last 3 years. The VES specimen containing organic/inorganic resins (D2-34), which suffered brittle fracture, exhibited a drop in strength between 8 and 11 years. It appears that strength was affected by accumulated radiation dose after a steady increase with age to 8 years.

One Portland cement specimen, C2A-6, failed far below the expected strength at 8 years, and C2A-14 has shown a comparable low strength at 11 years. It would be anticipated that the cement specimens would retain strength to a higher radiation dose than VES because low-level radiation does not adversely affect cement strength. The low strength exhibited by the cement specimens containing organic/inorganic resin may be caused by radiation effects on the contained ion-exchange resin, which serves as aggregate to the cement. It is noted that this same cement waste-form type exhibited an abnormally high strength at 7 years. An examination of the data from Reference 10 revealed that the cement batches contained waste forms that were very uniform in weight and radiation dose, while the VES batches

Table 1. Compression test result of 11-year-old EPICOR-II waste-form samples.

Specimen	Material of construction	Estimated radiation at contract (R/h)	Area (in^2)	Ultimate load (lb)	Compressive strength (psi)
C1-24	Organic ion-exchange resin in Portland cement	4.5	2.68	12,750	4,765
C2A-14	Organic/inorganic ion-exchange resin in Portland cement	6.5	2.69	5,675	2,110
D1A-29	Organic ion-exchange resin in VES	6.5	2.60	11,100	4,270
D2-34	Organic/inorganic ion-exchange resin in VES	7.5	2.59	10,450	4,035

contained many nonuniform waste forms. However, many of the cement waste forms exhibited surface blemishes in the form of air bubbles. It is suspected that air bubbles were also contained within the interior of those cement waste forms. Such discontinuities would detrimentally affect strength.

The cement compressive strength demonstrates the impact of data scatter resulting from employing only one waste form of each type in each year of testing. In order to economize the few remaining waste forms, only four are tested during each period, one of each type. Thus, one waste form with no imperfections containing higher than average amounts of either zeolite aggregate or cement could result in an unusually high compressive strength for that period. Also, a waste form containing an air bubble inclusion could produce a low strength for the period. The failed waste forms have not been examined closely enough to locate specific initiators of failure. The results shown in Figure 5 from 1 month (as-cast) and 2 years (immersion tested) are the average of four data points at each value and appear to be very consistent.

A review of irradiated waste form compressive test results (Reference 10) shows that cement specimen strength after irradiation varied considerably. The strength of waste forms containing all organic resins (C1) varied from 1,740 to 5,230 psi, averaging 3,640 psi with a standard deviation of 1,440 psi. Strength of waste forms with organic/inorganic resins (C2A) varied from 1,860 to 5,200 psi, averaging 3,310 psi with a standard deviation of 1,710 psi. Reference 10 noted that standard deviations for the Portland cement waste form compressive strength data were large and that material effects induced by gamma radiation may have been responsible for that variation. All of the compressive strength results from 8 and 11 years fall within the standard deviations of the original irradiated waste form tests. The organic C1 specimens hold toward the upper limit while the organic/inorganic C2 specimens fall near the lower limit. It is noted that the strength of specimen C2A-7 tested in the seventh year at 6,404 psi was well above the standard deviation of any other specimen strength.

The VES compressive strength data of Figure 5 show a steady increase with age (and subsequent radiation dose) through 8 years for both organic and organic/inorganic resin waste forms. At 11 years, the organic-resin-containing waste form, D1A-29, exhibited strength almost identical to the 8-year-old specimen, D1A-1. The end of increase in strength could have been caused by a difference in makeup of D1A-29, which was at the end of that batch pour. Or it was the result of total radiation dose on this waste form. The 11-year organic/inorganic-resin-containing waste form, D2-34, showed a 20% drop in strength from the 8-year test specimen, D2-35, probably as a result of radiation-induced cross-linking of the VES polymer. That effect would result in a more brittle failure as experienced by D2-34.

Further testing planned over the next several years should help to clarify these results.

These waste-form specimens have been stored in shielded drums throughout the project. Dose measurements indicate that the specimens have experienced a total gamma dose of 0.5×10^6 to 1.2×10^6 rad. That dose has caused serious degradation of the polyethylene specimen containers, which are discolored, embrittled, and cracked. In order to preserve specimen identity, a number of those containers were replaced with cardboard tubes during the last test operation.

Figure 1. Sample C1-24 organic ion-exchange resin in Portland cement after compression test.

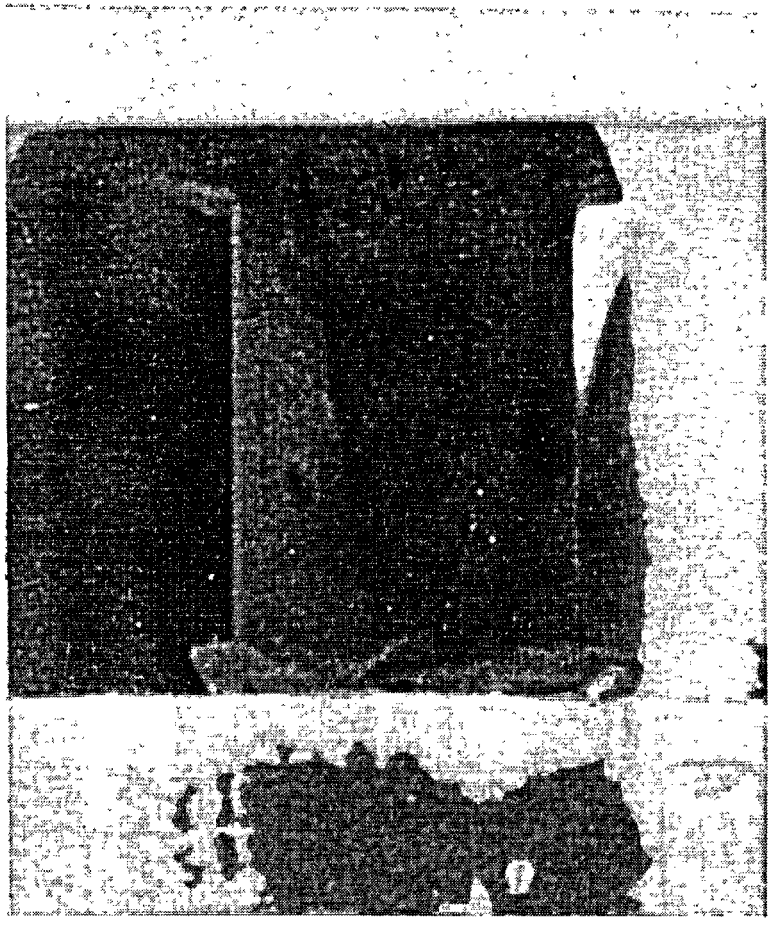

Figure 2. Sample C2A-14 organic/inorganic ion-exchange resin in Portland cement after compression test.

Figure 3. Sample D1A-29 organic ion-exchange resin in VES after compression test.

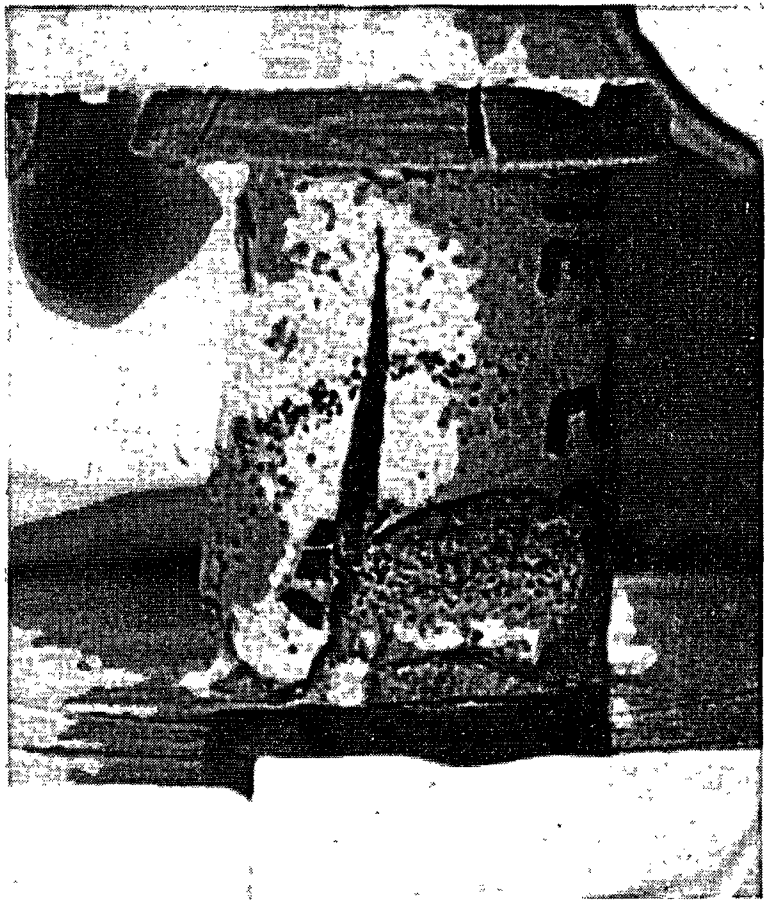

Figure 4. Sample D2-34 organic/inorganic ion-exchange resin in VES after compression test.

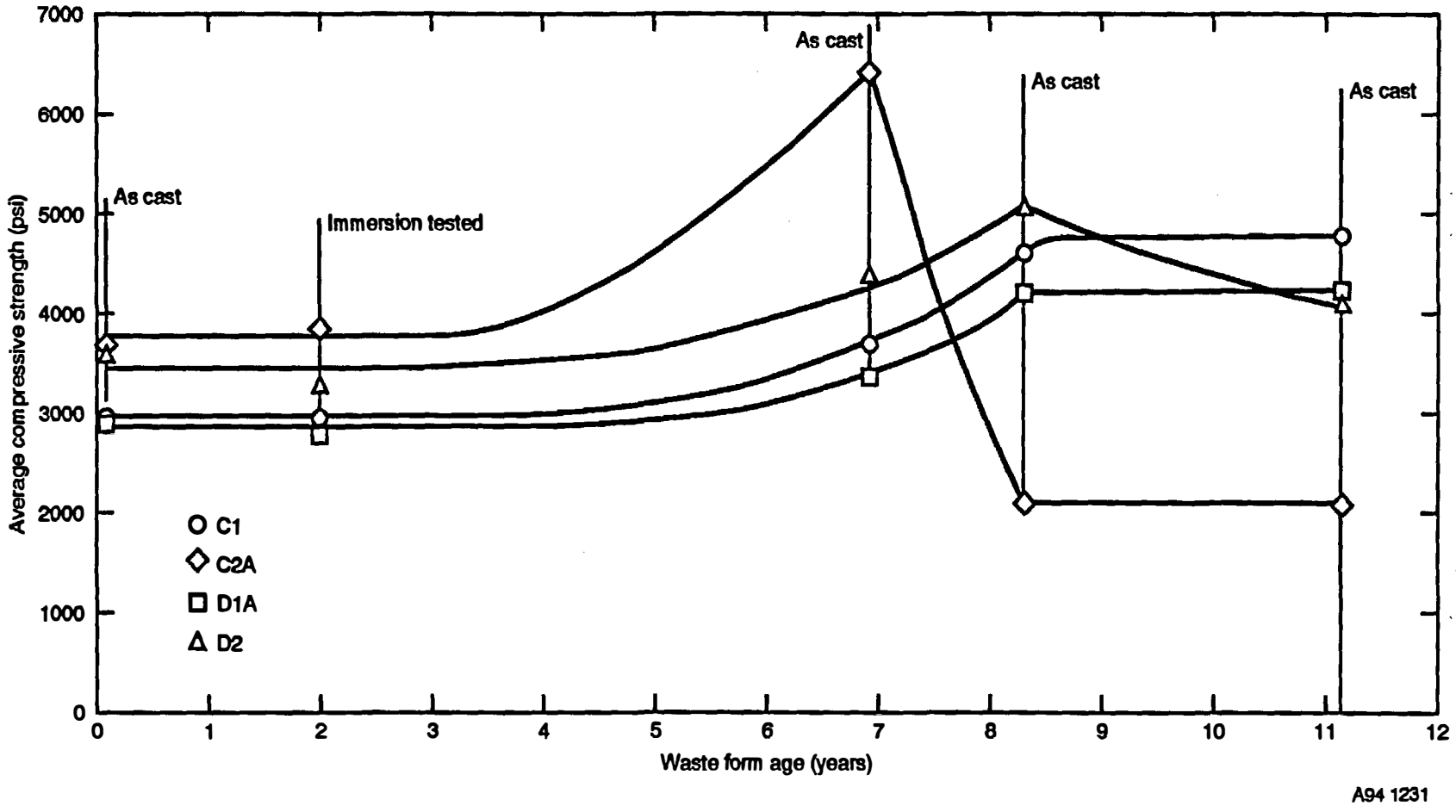

Figure 5. Variation of waste form compressive strength with age caused by self-irradiation and curing.

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 12 while data from the first 8 years of operation are contained in earlier reports.[11,13–19]

Description of Waste Forms. Waste forms used in the field test are composed of solidified EPICOR-II prefilter resin wastes. Two waste types were used in the solidification project. One is a mixture of synthetic organic ion-exchange resins from prefilter PF-7 (phenolic cation, strong acid cation, and strong base anion resins), and the other is a mixture of synthetic organic ion-exchange resins from prefilter PF-24 (strong acid cation and strong base anion resins) with an inorganic zeolite. PF-7 waste contains 5% Sr-90, while PF-24 waste contains about 1% Sr-90. Of the other radionuclides in those wastes, Cs-137 and Cs-134 are the major constituents, with traces of Co-60 and Sb-125 included.

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 2 shows the types of specimens placed in the lysimeters. A complete description of waste form manufacture is given in Reference 9. Bench testing of similar waste forms, per the requirement of the Branch Technical Position on Waste Form, is described in Reference 10.

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 6). 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 provides 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 6. The cups are numbered 1 through 5 as noted. The lower compartment serves as a leachate collector, which is emptied and sampled through the 3.8-cm pipe.

Table 2. Lysimeter waste form composition.

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

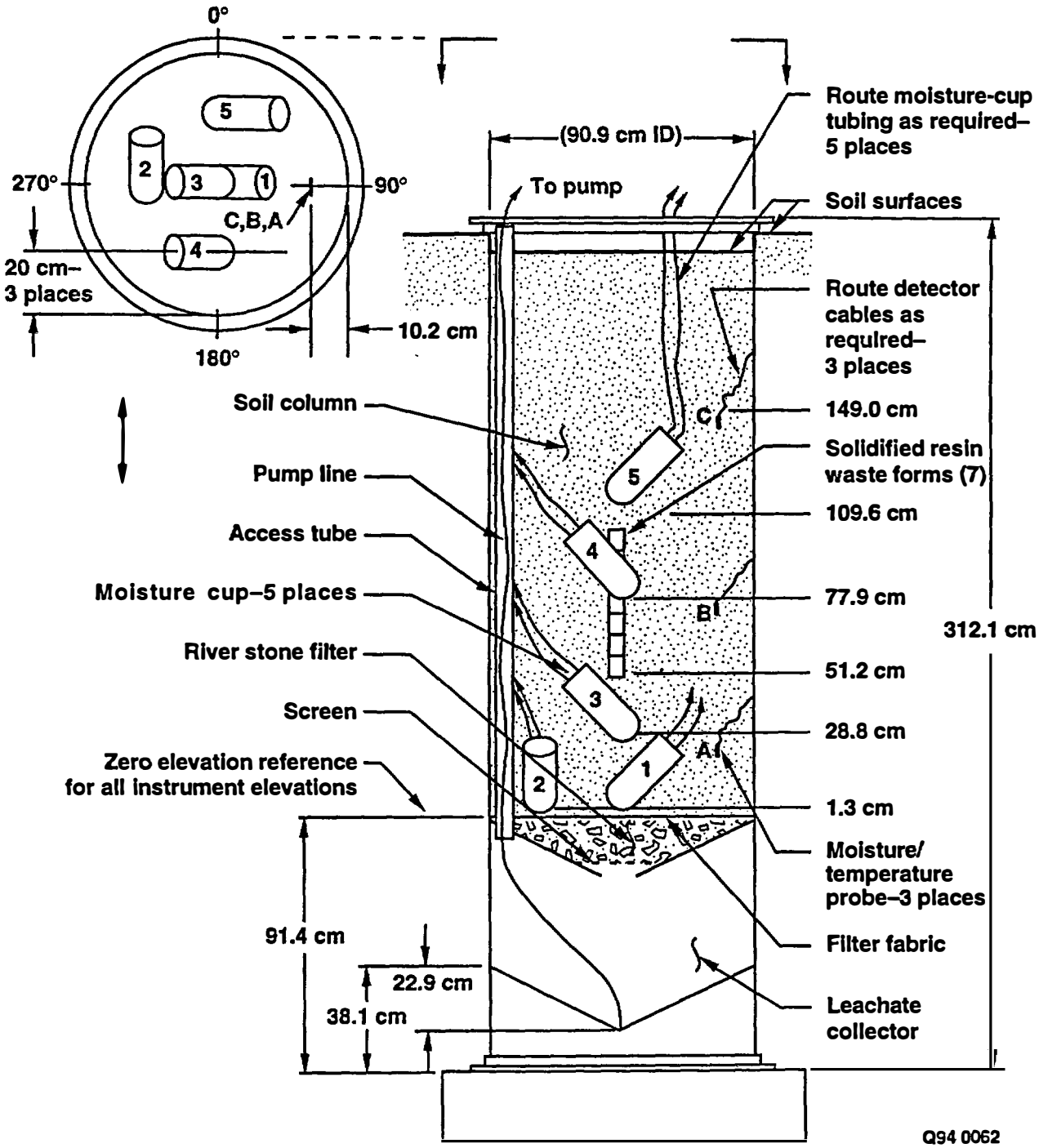

Figure 6. EPICOR-II lysimeter vessel component locations.

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 12). 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 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 12).

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 alphanumeric 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 1993 through June 1994. 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 1994. Many of the data are displayed in graphic format so that information can be correlated easily with time. There were periods of time when the DAS was not in operation at either site. These were caused by 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 7 through 11. Because of sensor 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 80.5 cm at ANL-E and 177.9 cm at ORNL. This year, ANL-E was near the normal annual rainfall while ORNL was 28% above normal (ANL-E—85.2 cm; ORNL—138.8 cm).[20,21] This is the seventh time in the past 8 years that ORNL has equalled or exceeded the normal amount of yearly precipitation. The monthly precipitation pattern for each

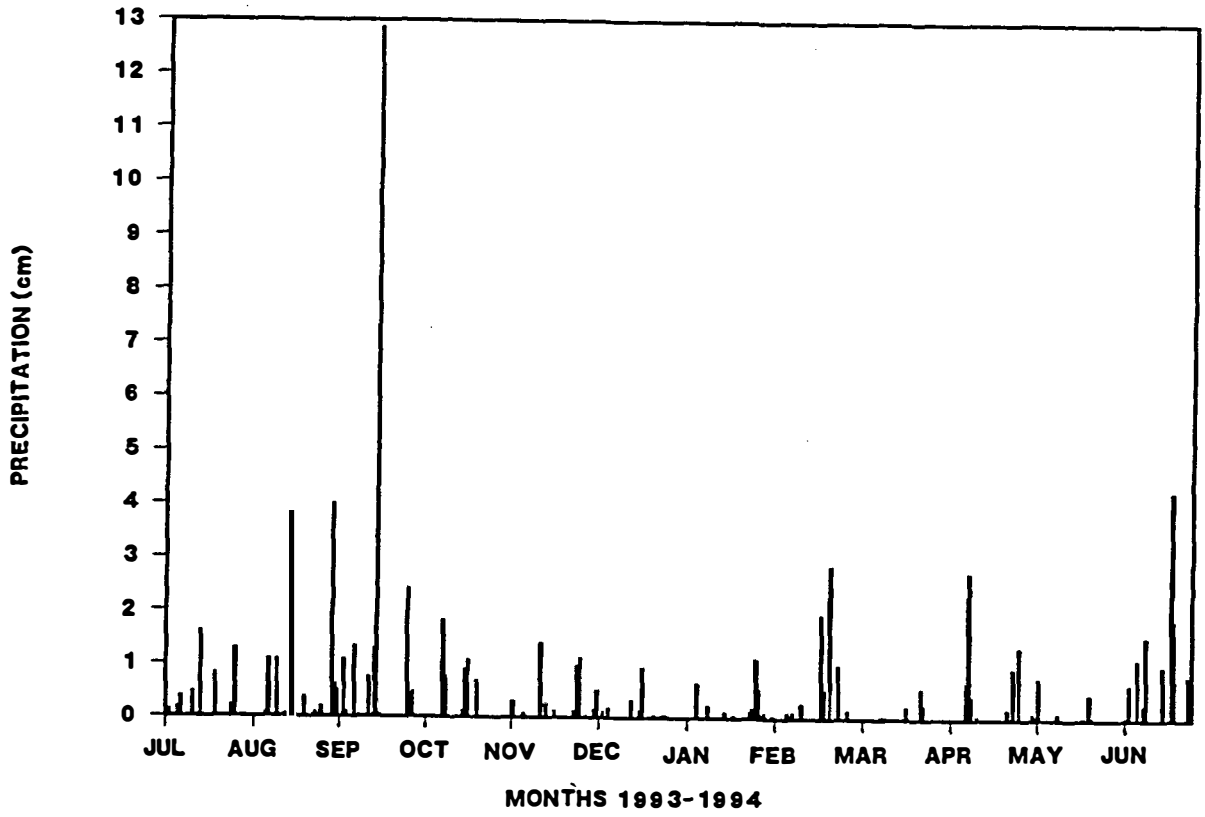

Figure 7. ANL-E weather data—precipitation.

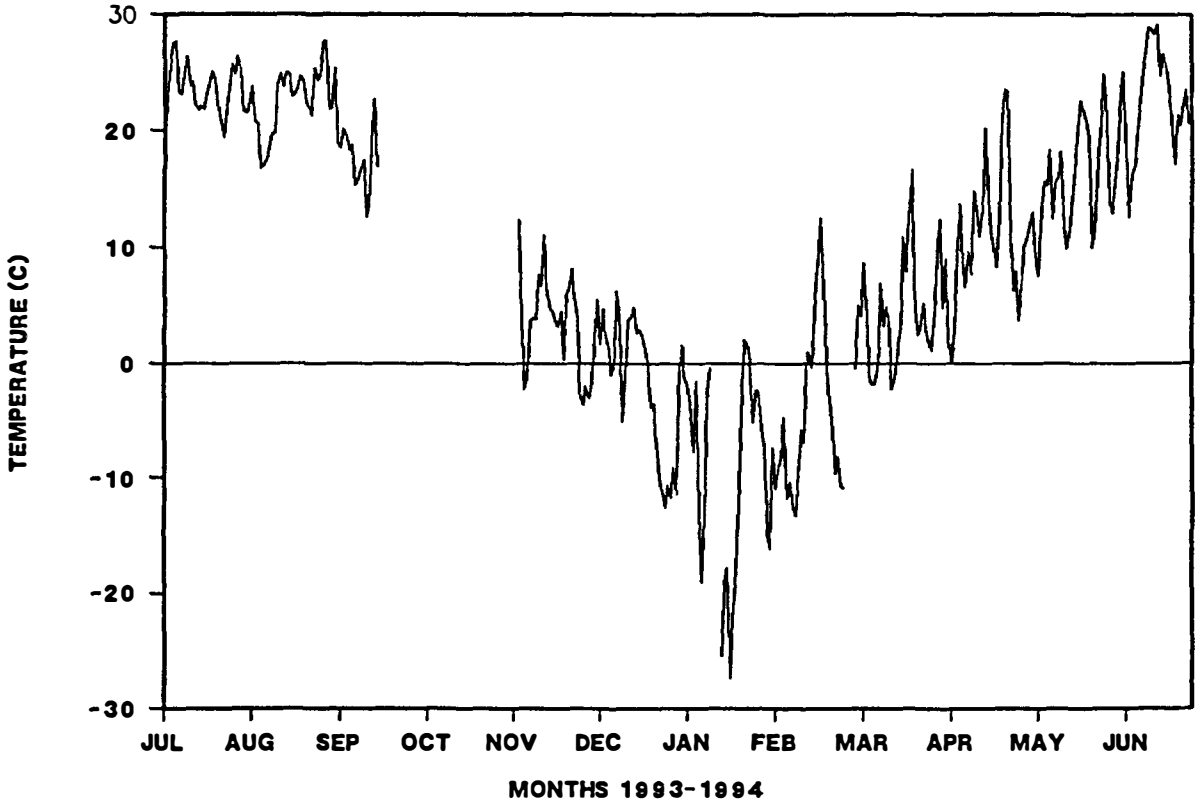

Figure 8. ANL-E weather data—air temperature.

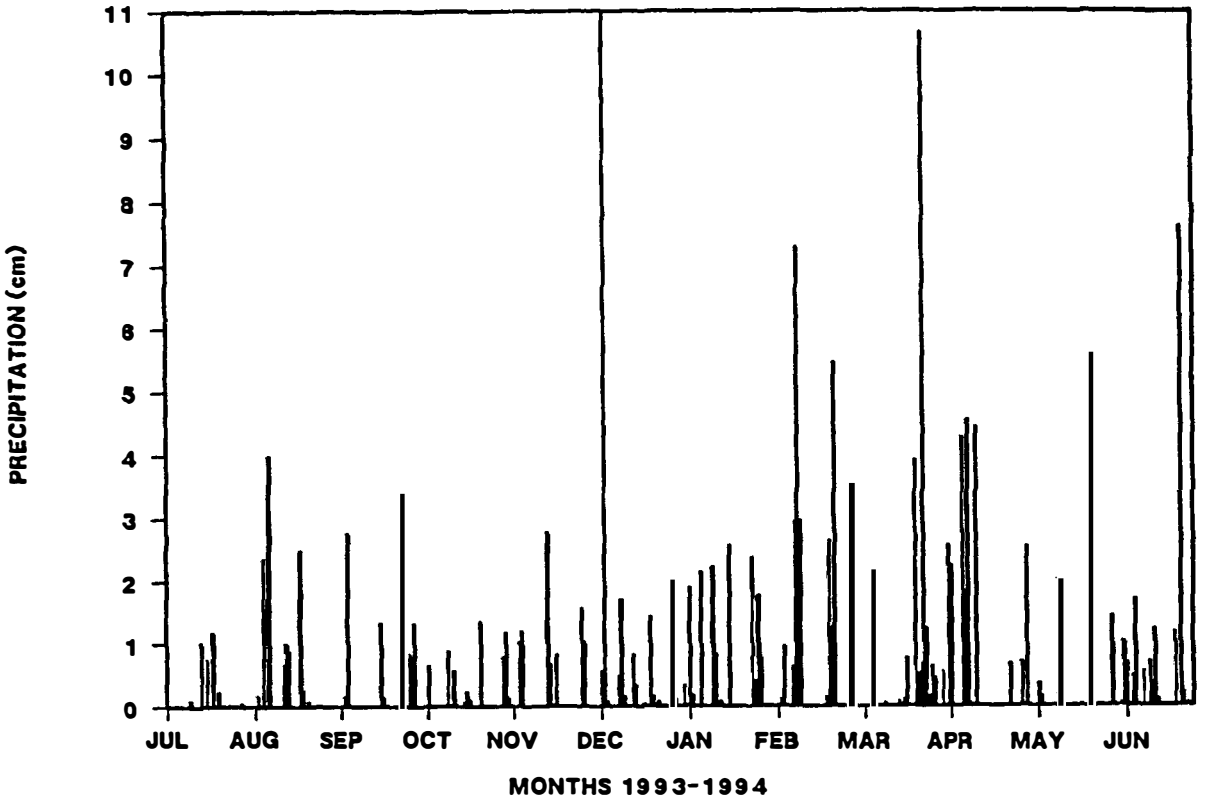

Figure 9. ORNL weather data—precipitation.

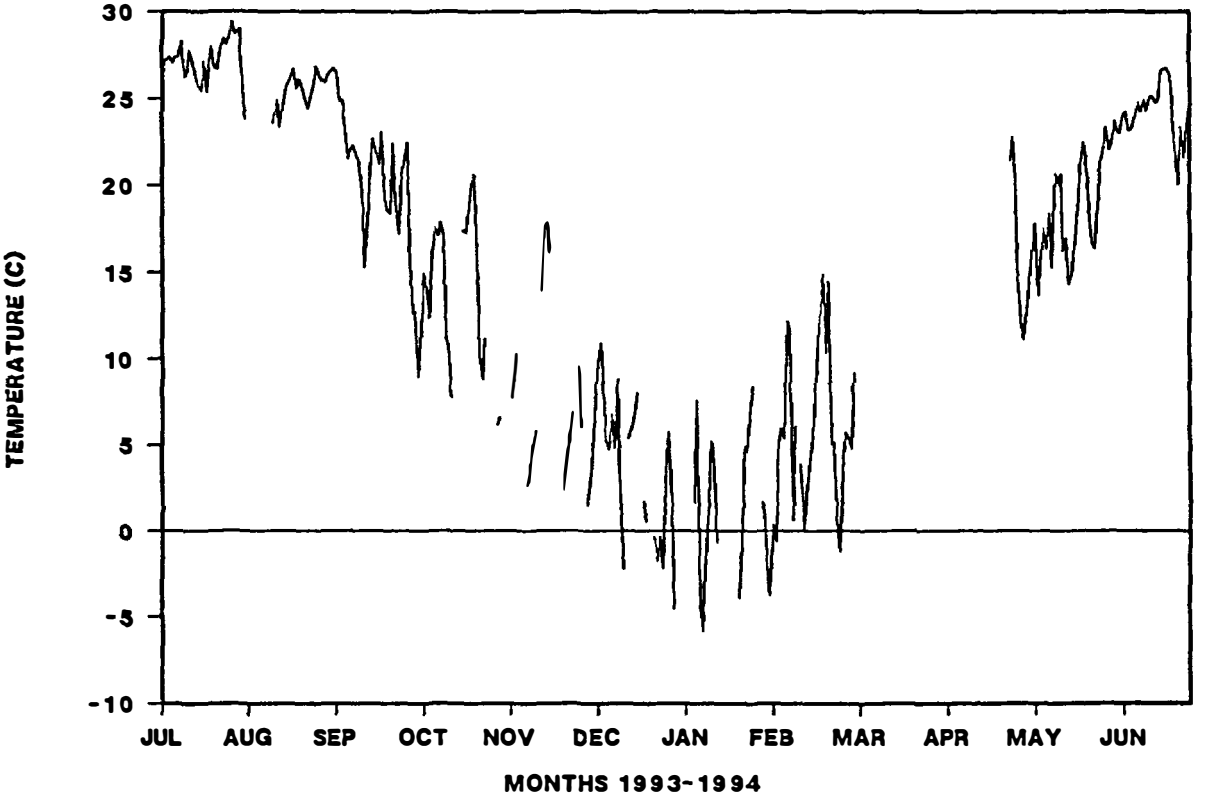

Figure 10. ORNL weather data—air temperature.

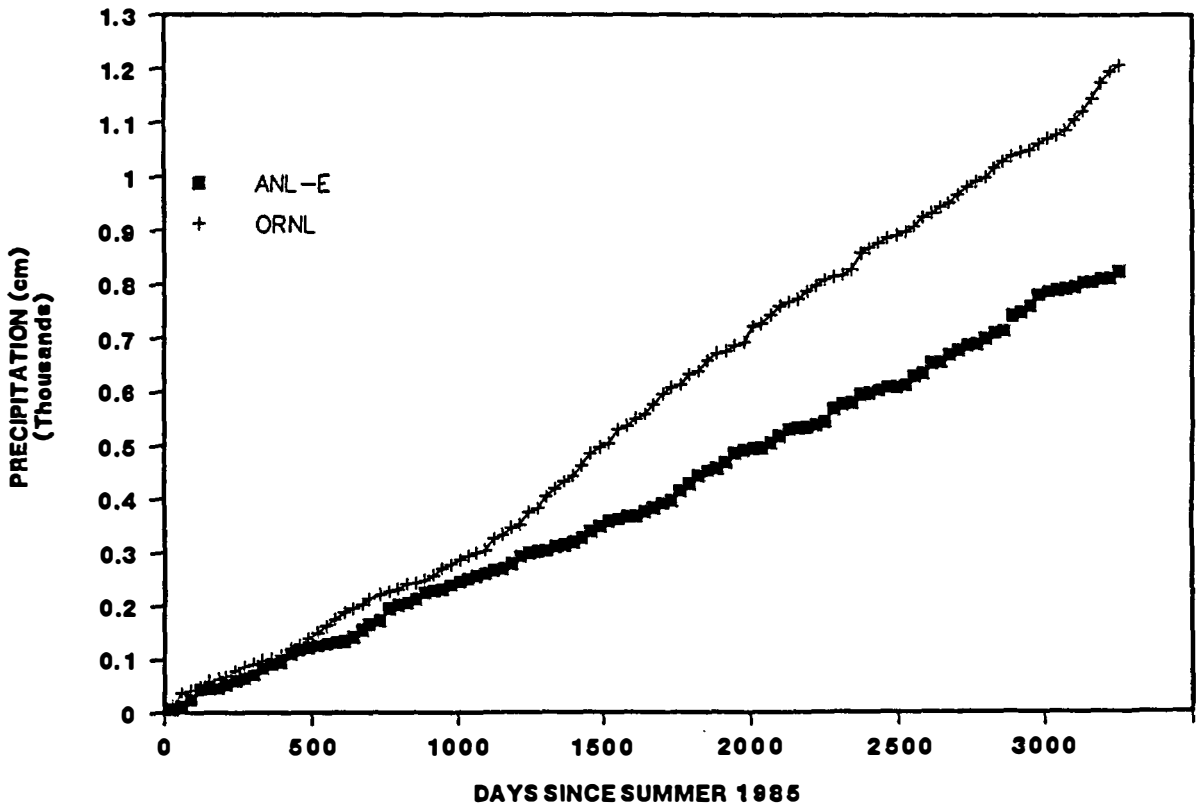

Figure 11. ANL-E and ORNL weather data—cumulative precipitation.

site can be seen from the histograms in Figures 7 and 9. Figure 11 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 823.0 cm at ANL-E, while ORNL had reached a total of 1,223.1 cm.

Air temperature data from ANL-E (Figure 8) show that periods of freezing temperatures occurred from early November 1993 until mid March 1994. ORNL experienced freezing temperatures from early December through late February (Figure 10).

Lysimeter Soil Temperature Data. The lysimeter soil temperature data recorded at ANL-E and ORNL during the reporting period are shown in Figures 12 through 20. As in past years, 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. There continues to be a direct correspondence between air temperature and soil temperature at both locations.

Past reports have detailed the failure of some temperature probes at ANL-E. Faults were found with one temperature probe in ANL-2, two in ANL-3, all in ANL-4, and one in ANL-5. During this reporting period, an additional probe failed in ANL-5. Data from these failed probes were not included in the report. The probes have probably been damaged by corrosion of the metal parts (Reference 14). A more damage-resistant replacement for these probes has been found, but the new components have not been procured or installed. ORNL probes are all continuing to provide output. As mentioned in the last report, 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,

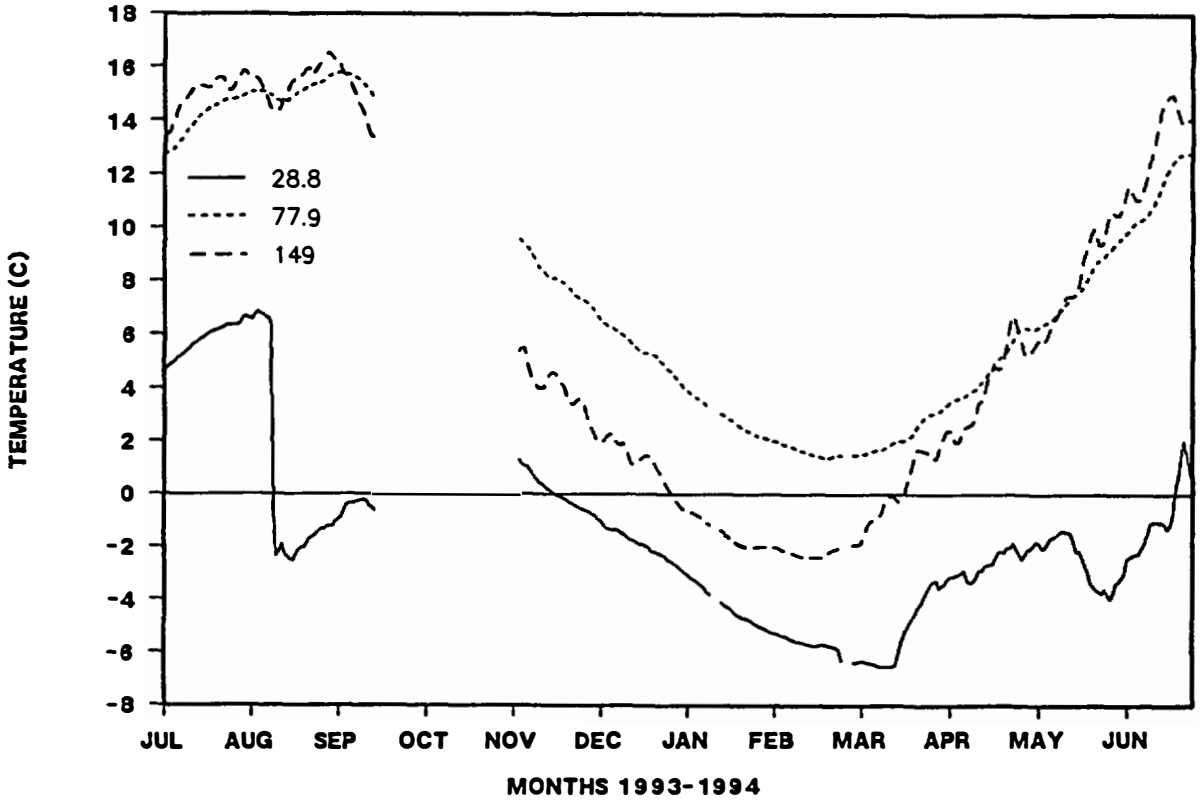

Figure 12. ANL-E lysimeter 1 soil temperature.

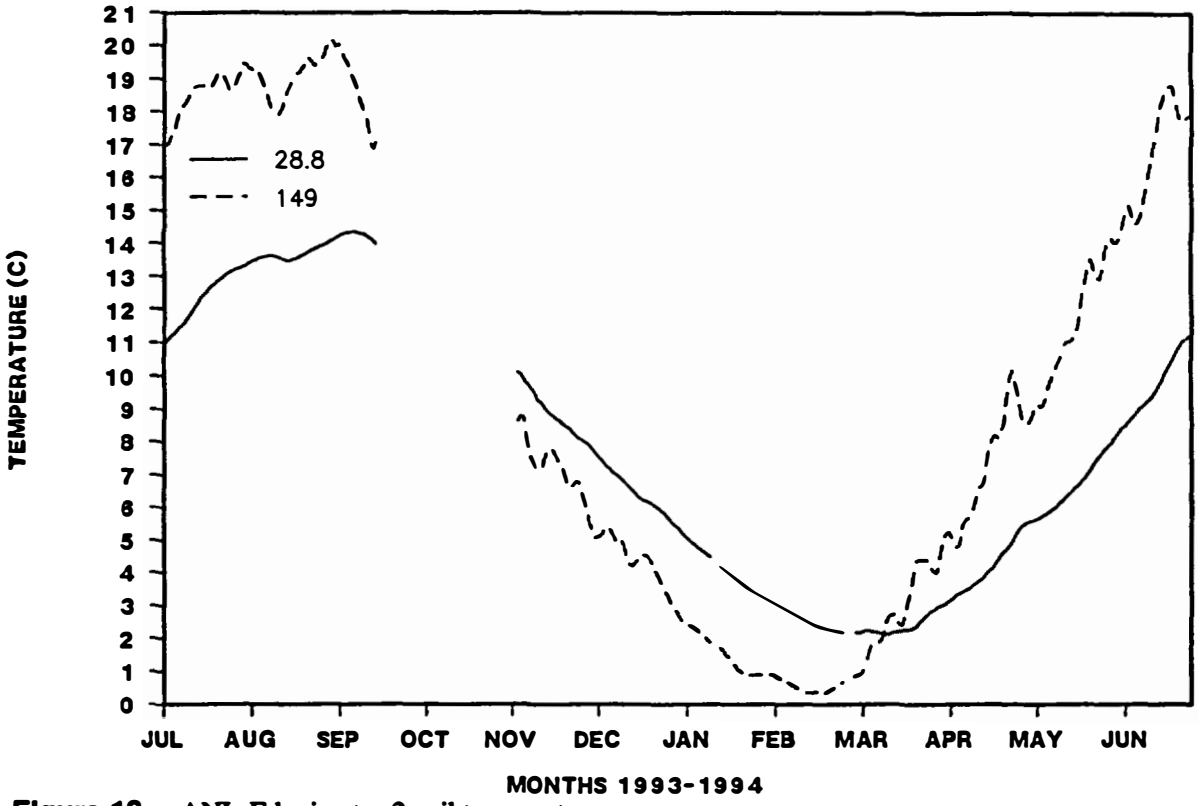

Figure 13. ANL-E lysimeter 2 soil temperature.

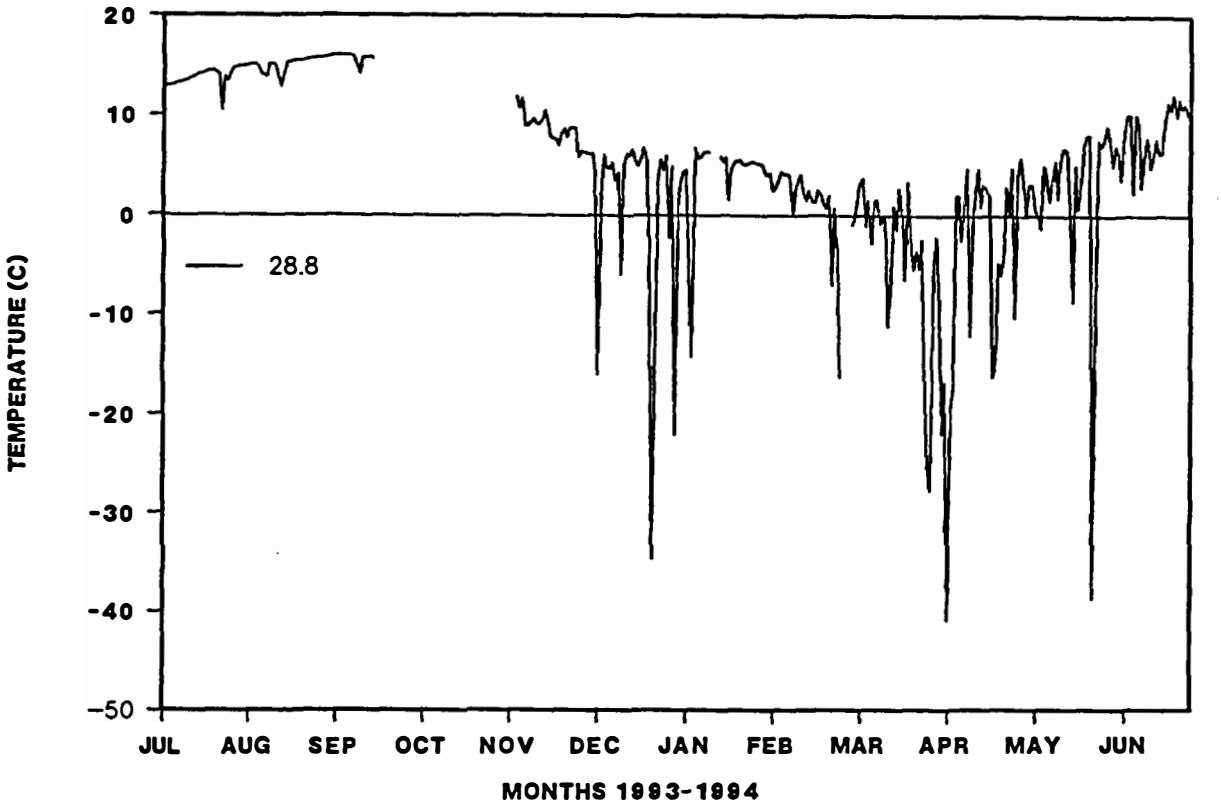

Figure 14. ANL-E lysimeter 3 soil temperature.

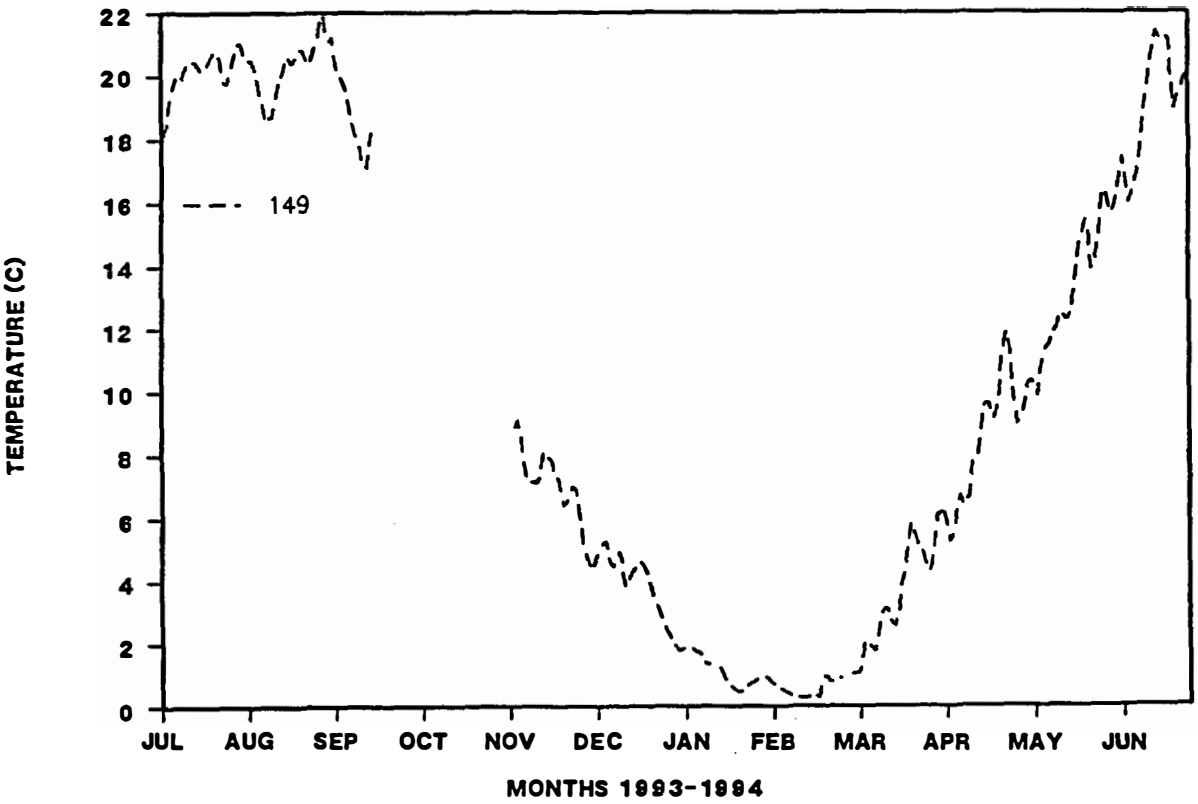

Figure 15. ANL-E lysimeter 5 soil temperature.

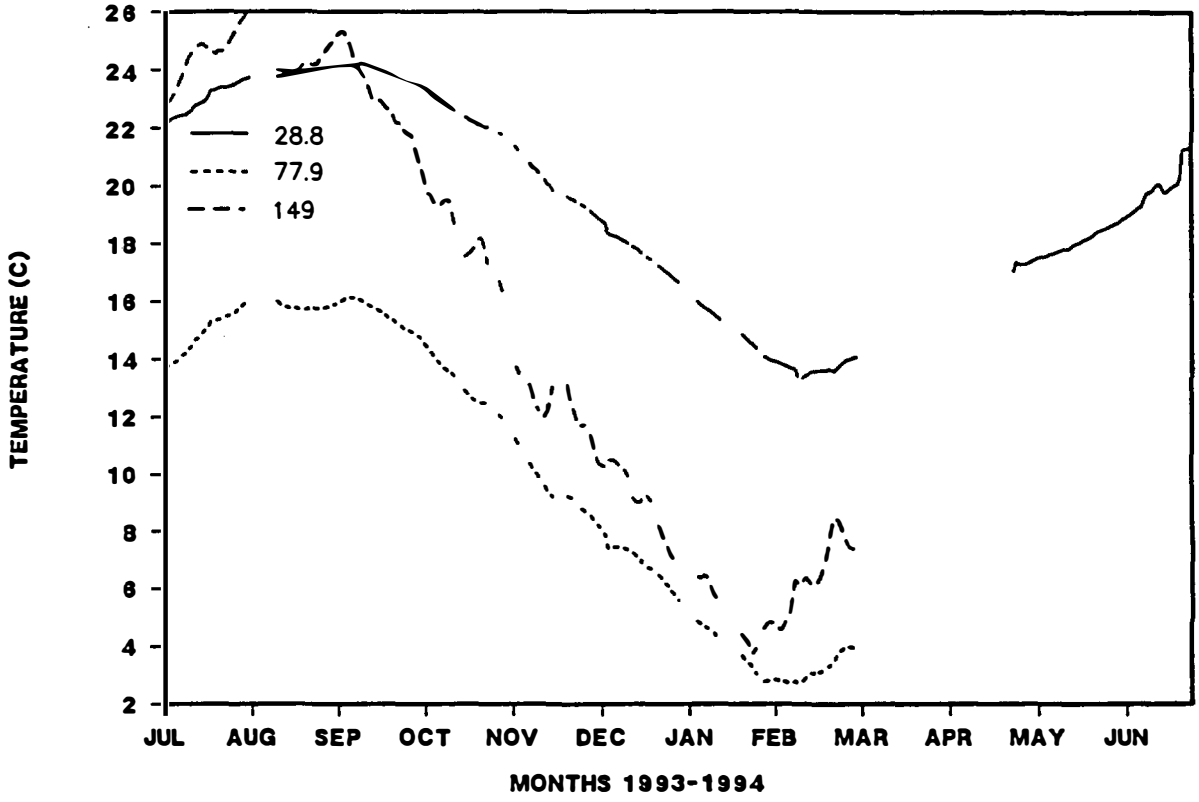

Figure 16. ORNL lysimeter 1 soil temperature.

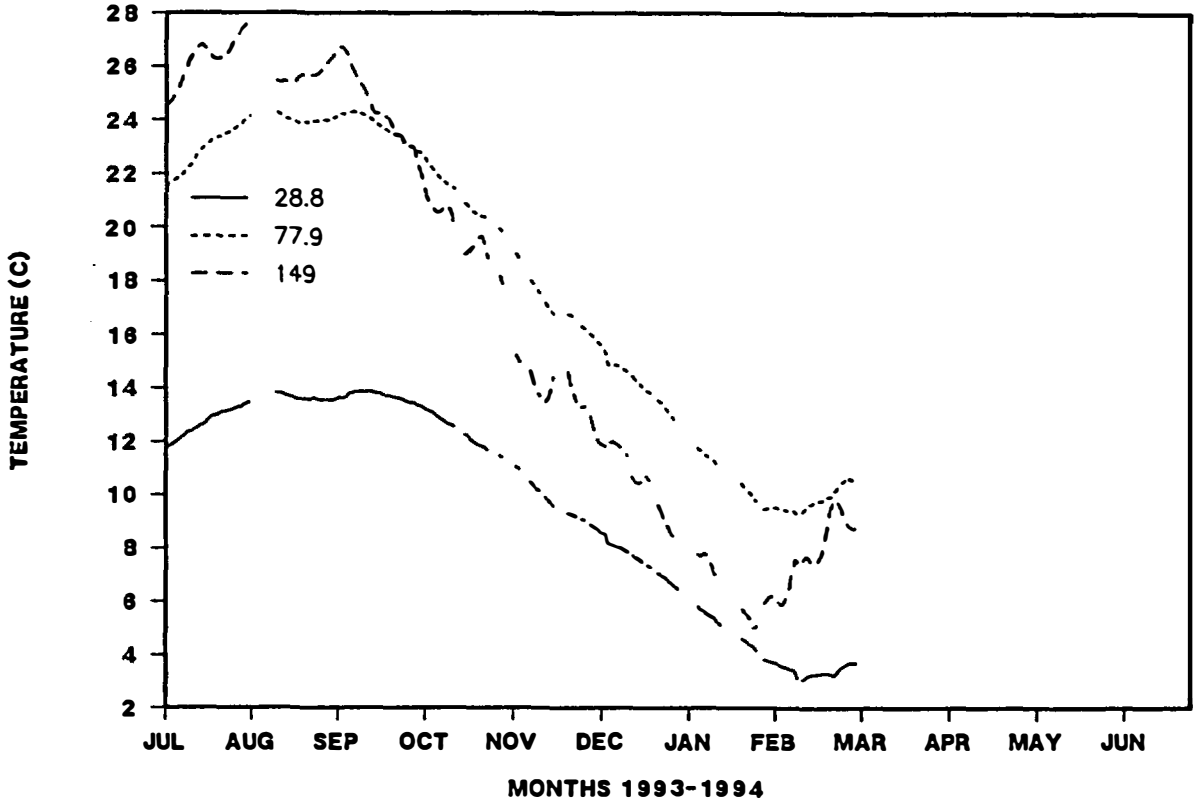

Figure 17. ORNL lysimeter 2 soil temperature.

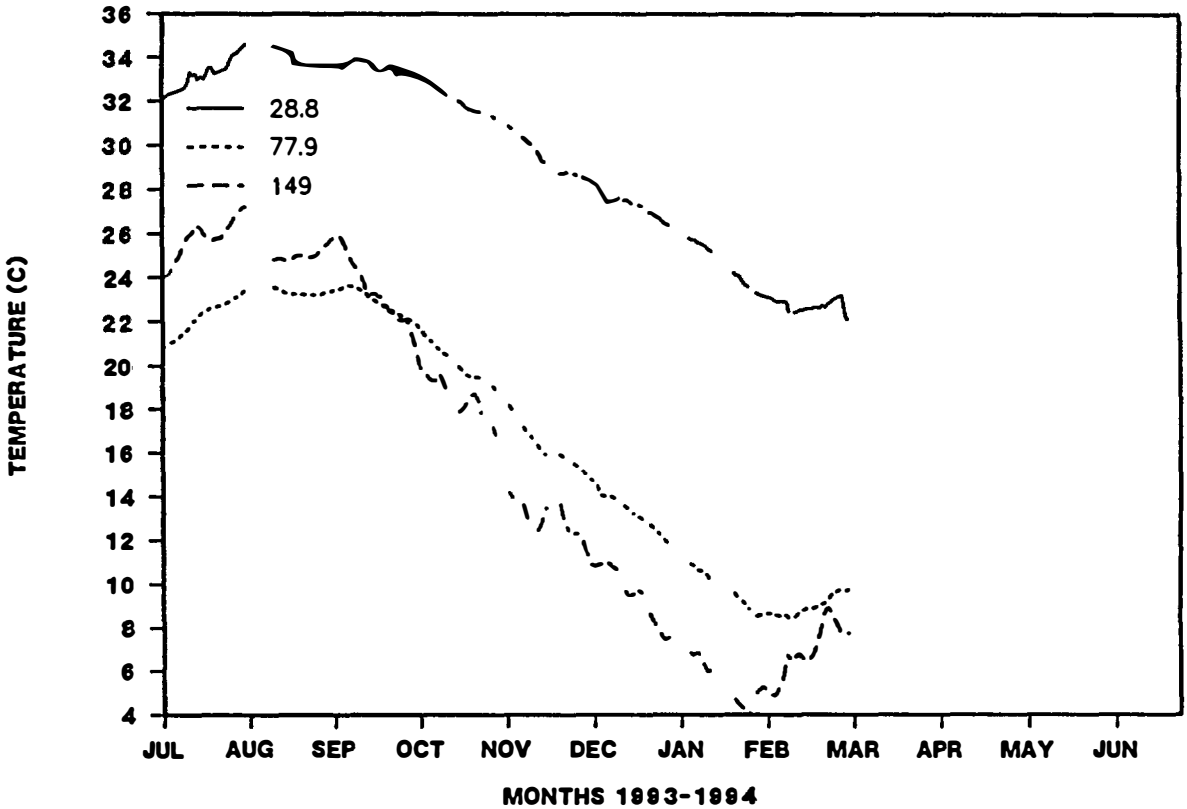

Figure 18. ORNL lysimeter 3 soil temperature.

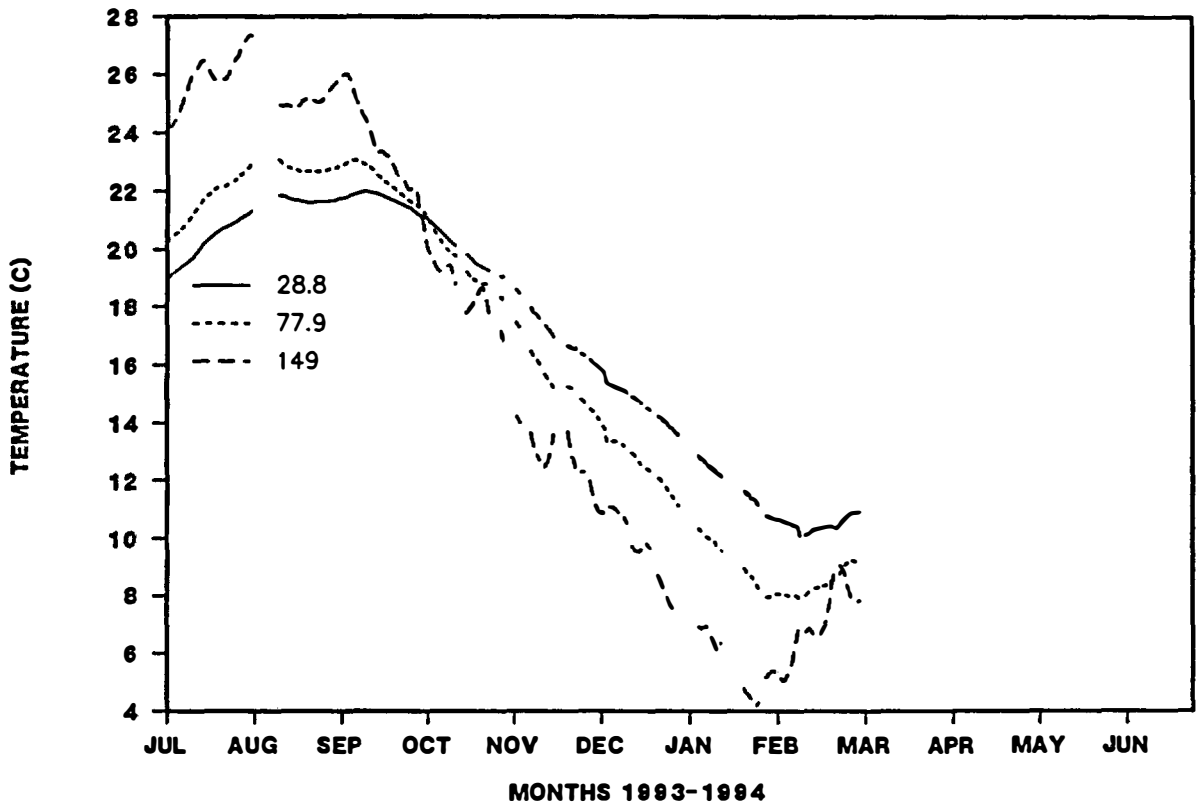

Figure 19. ORNL lysimeter 4 soil temperature.

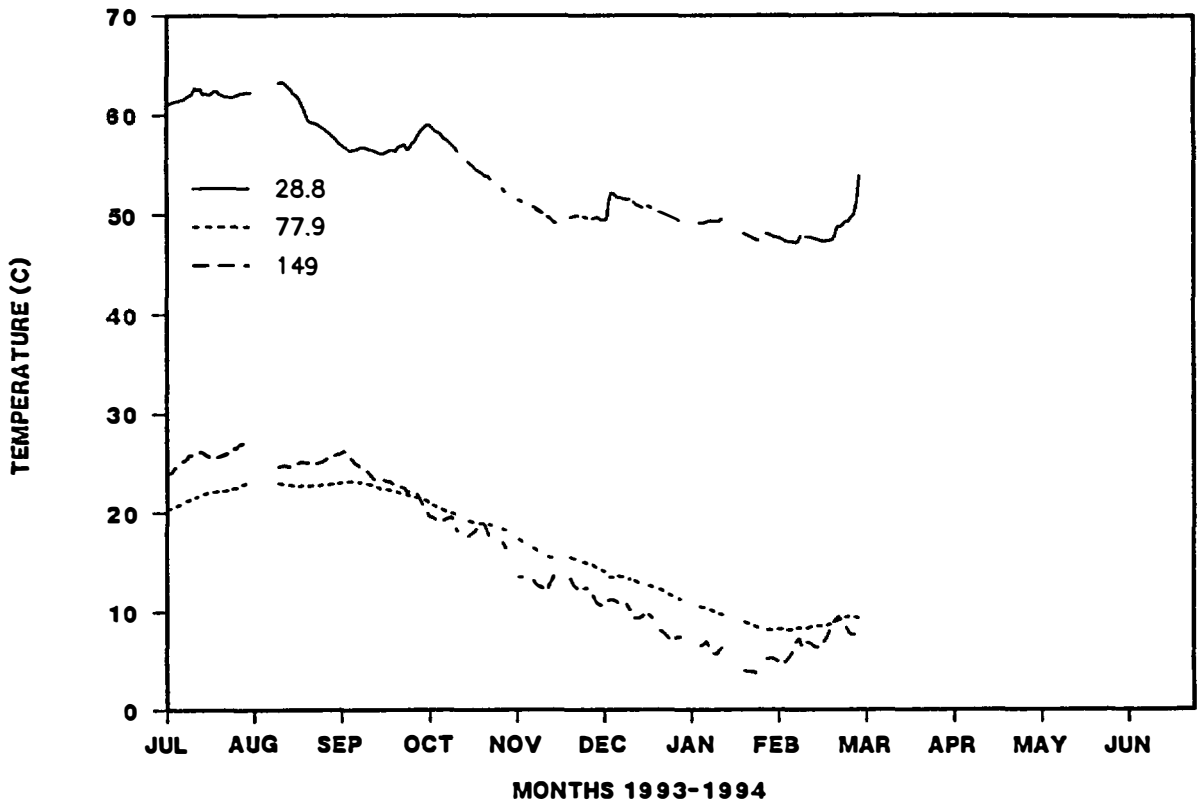

Figure 20. ORNL lysimeter 5 soil temperature.

shown in Figures 21 through 30, 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 34.4% at ANL-E and 14.2% 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 14).

Soil moisture in the soil column of the lysimeters at each site is quantified gravimetrically once each year (see Tables 3 and 4). 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 5). Percent differences between the gravimetric data and moisture probe data for ANL-E lysimeters range between 20.3 and 33.3%. These values have increased during this reporting period, but are still 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 10.4%, but underestimated those values (see Table 3). 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

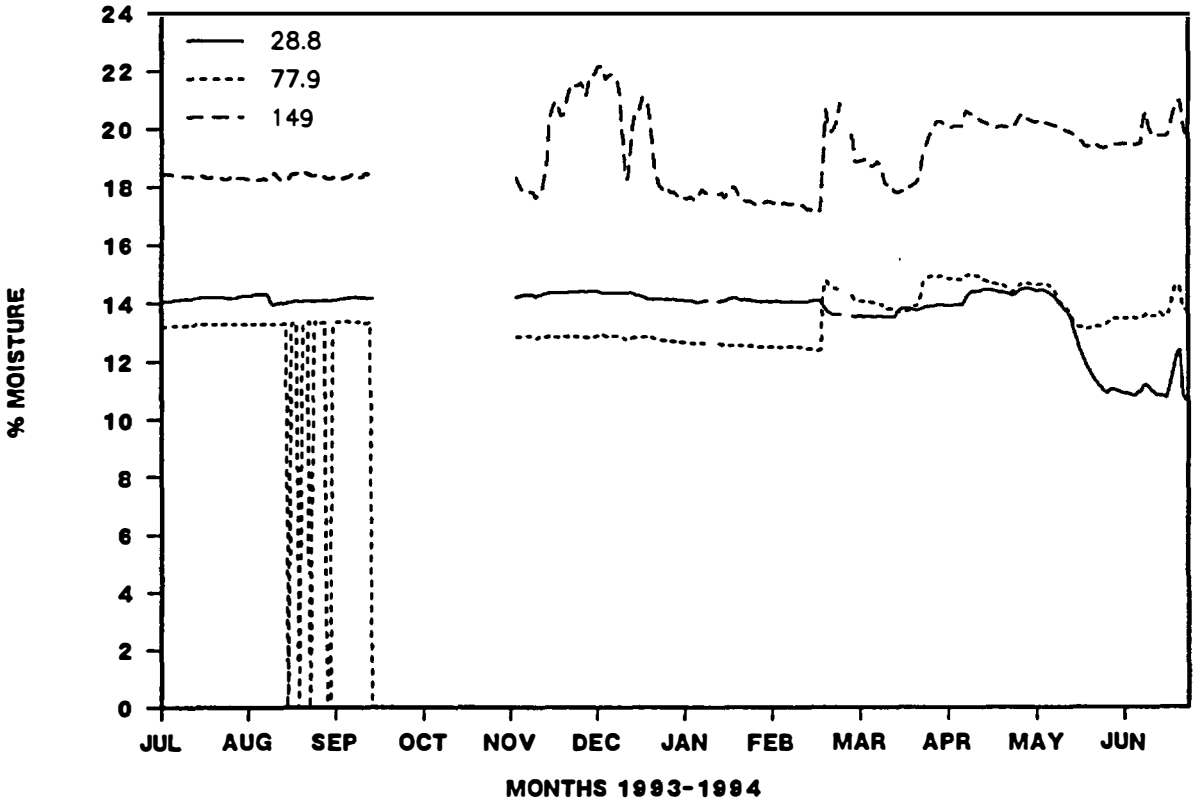

Figure 21. ANL-E lysimeter 1 soil moisture.

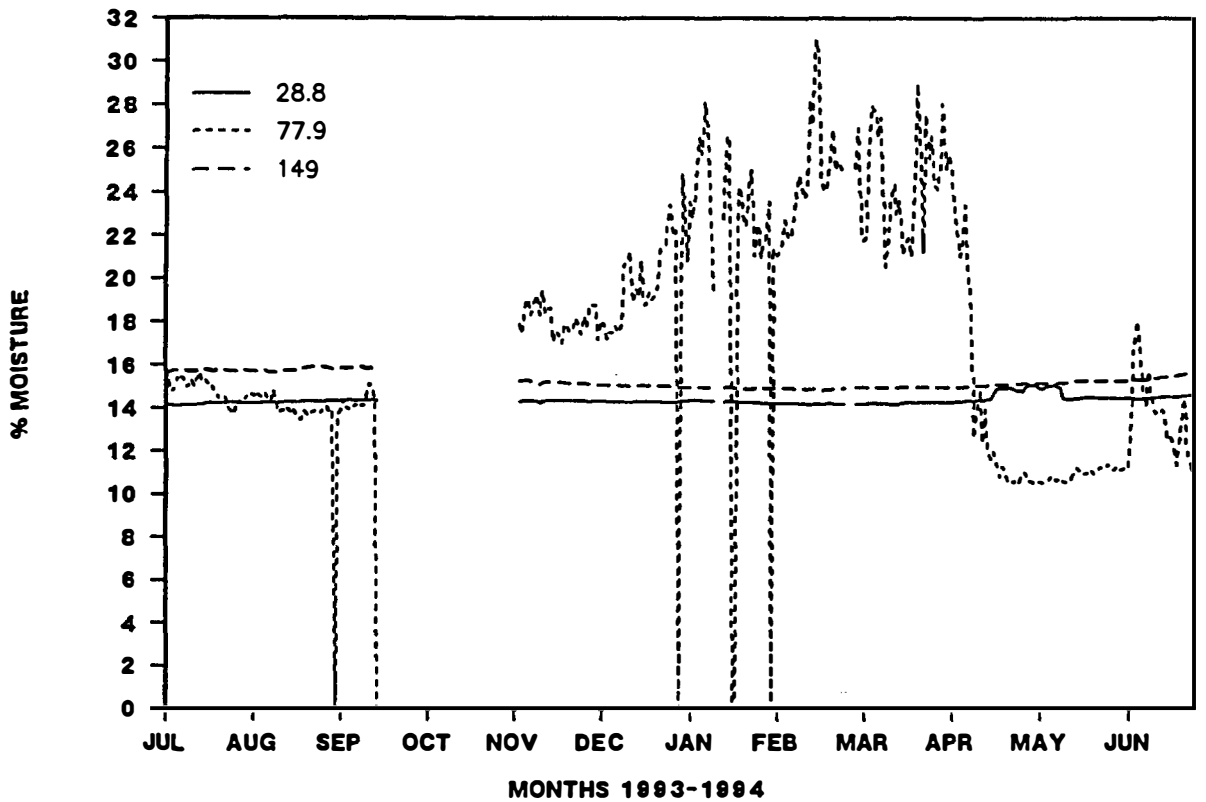

Figure 22. ANL-E lysimeter 2 soil moisture.

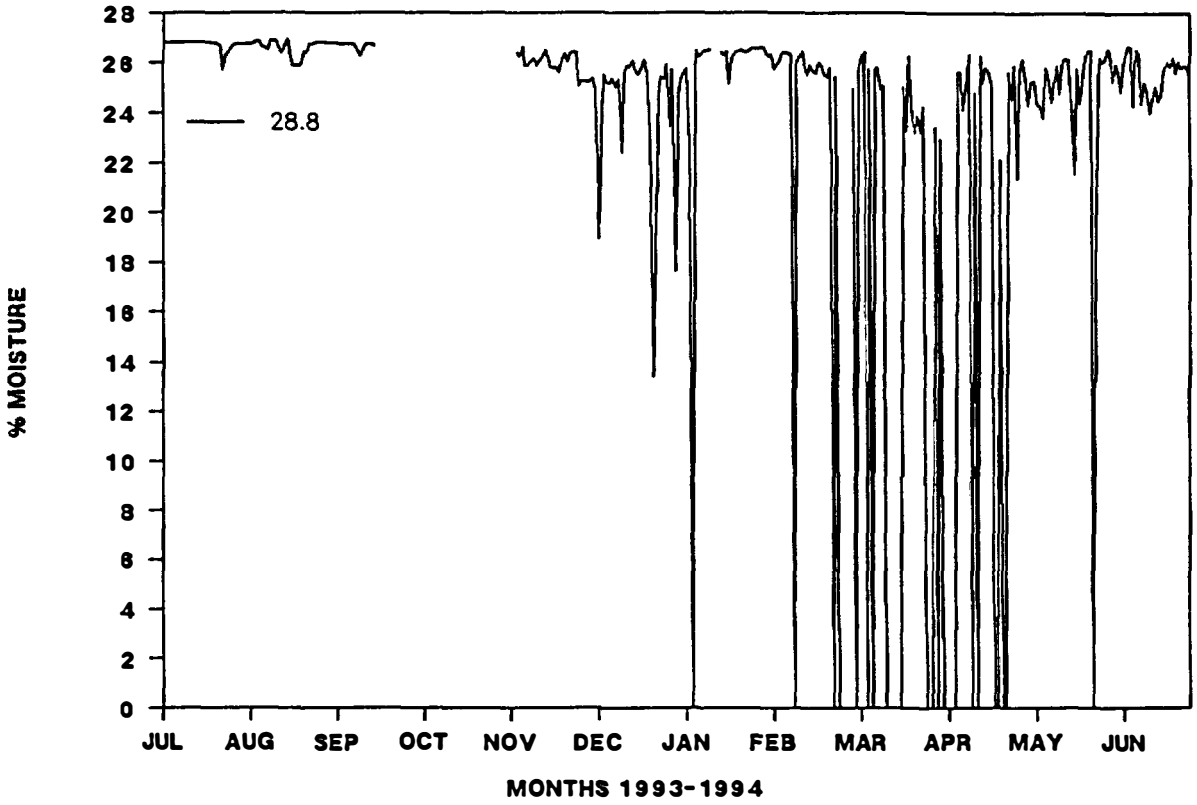

Figure 23. ANL-E lysimeter 3 soil moisture.

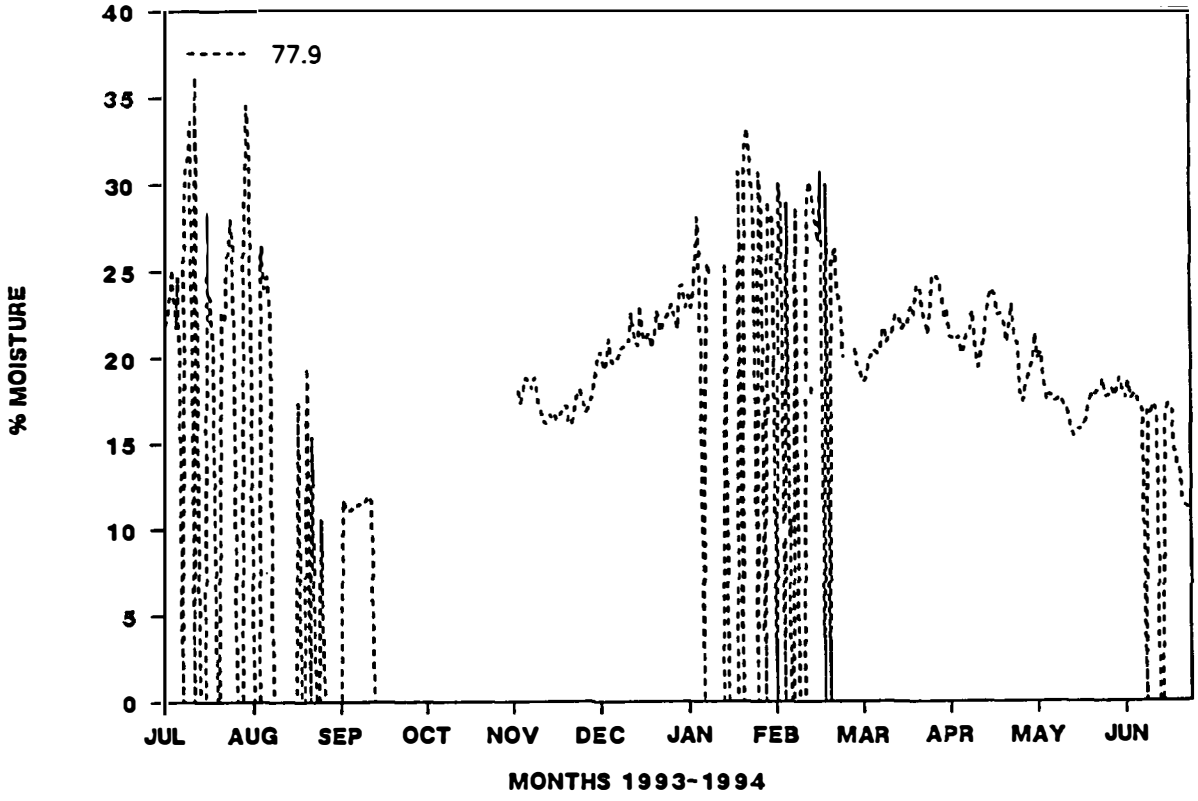

Figure 24. ANL-E lysimeter 4 soil moisture.

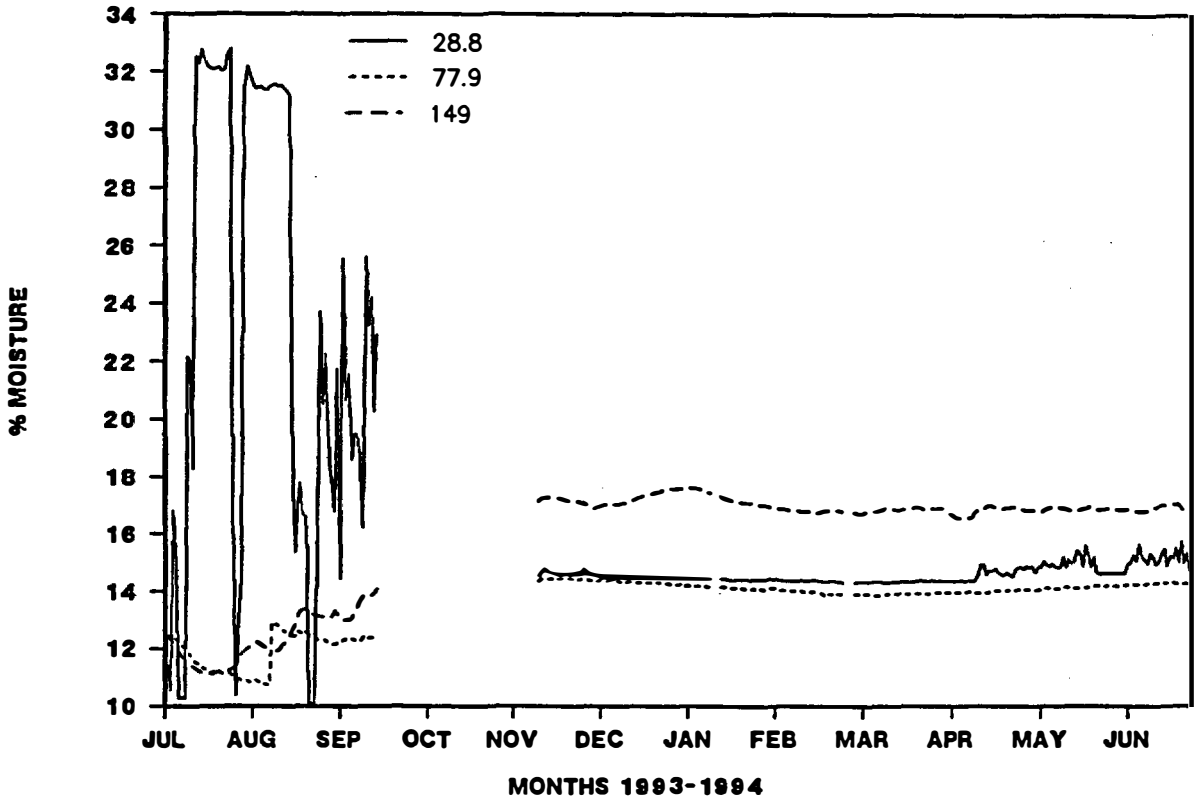

Figure 25. ANL-E lysimeter 5 soil moisture.

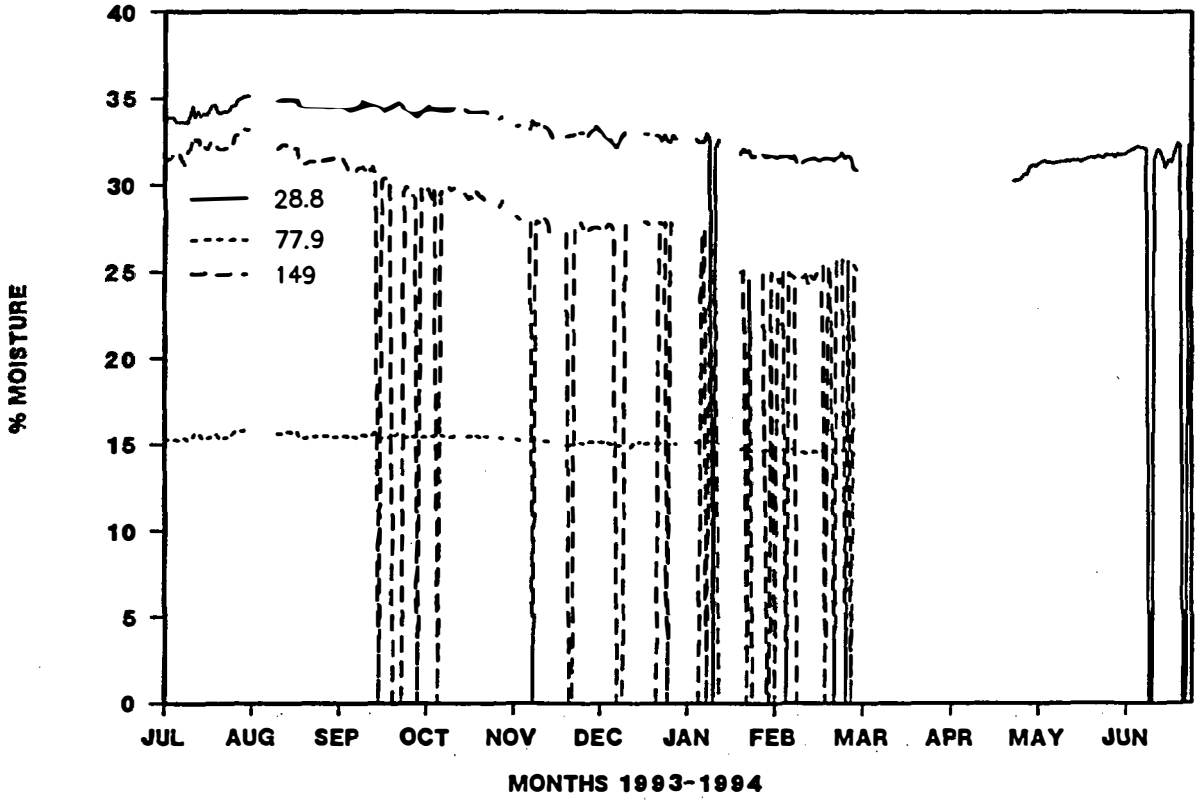

Figure 26. ORNL lysimeter 1 soil moisture.

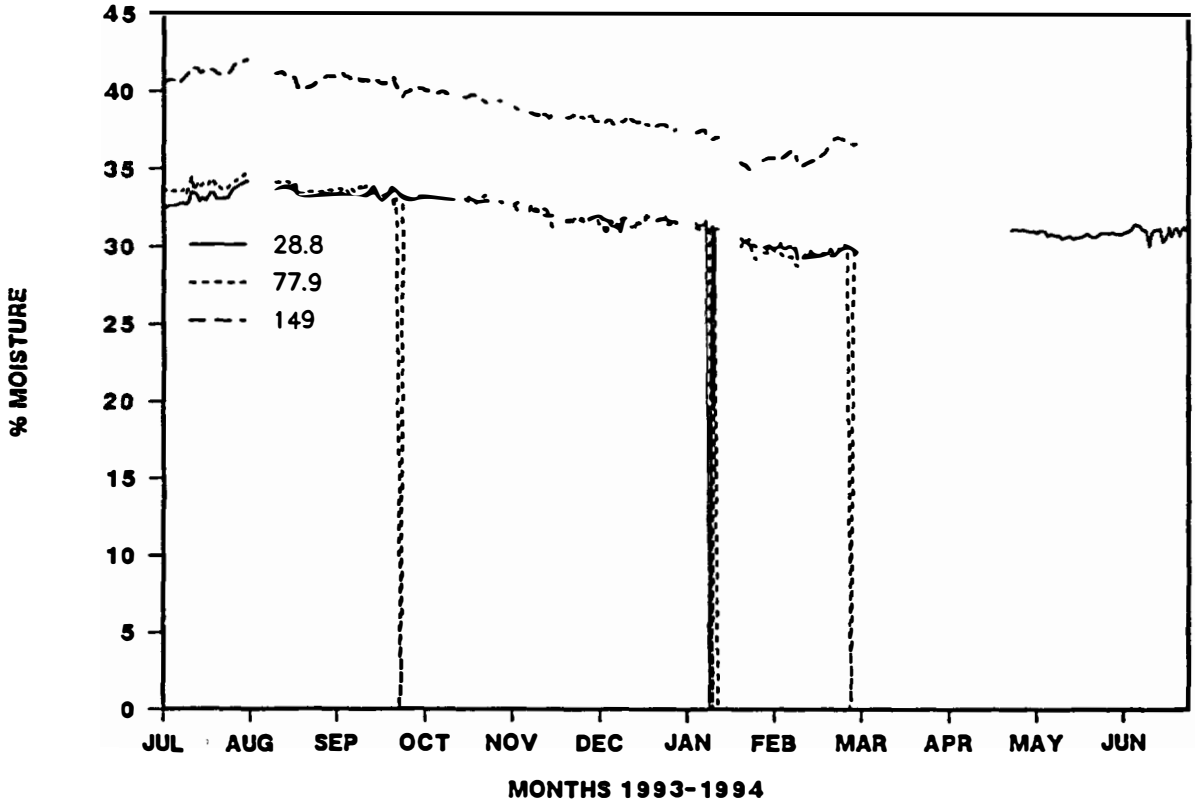

Figure 27. ORNL lysimeter 2 soil moisture.

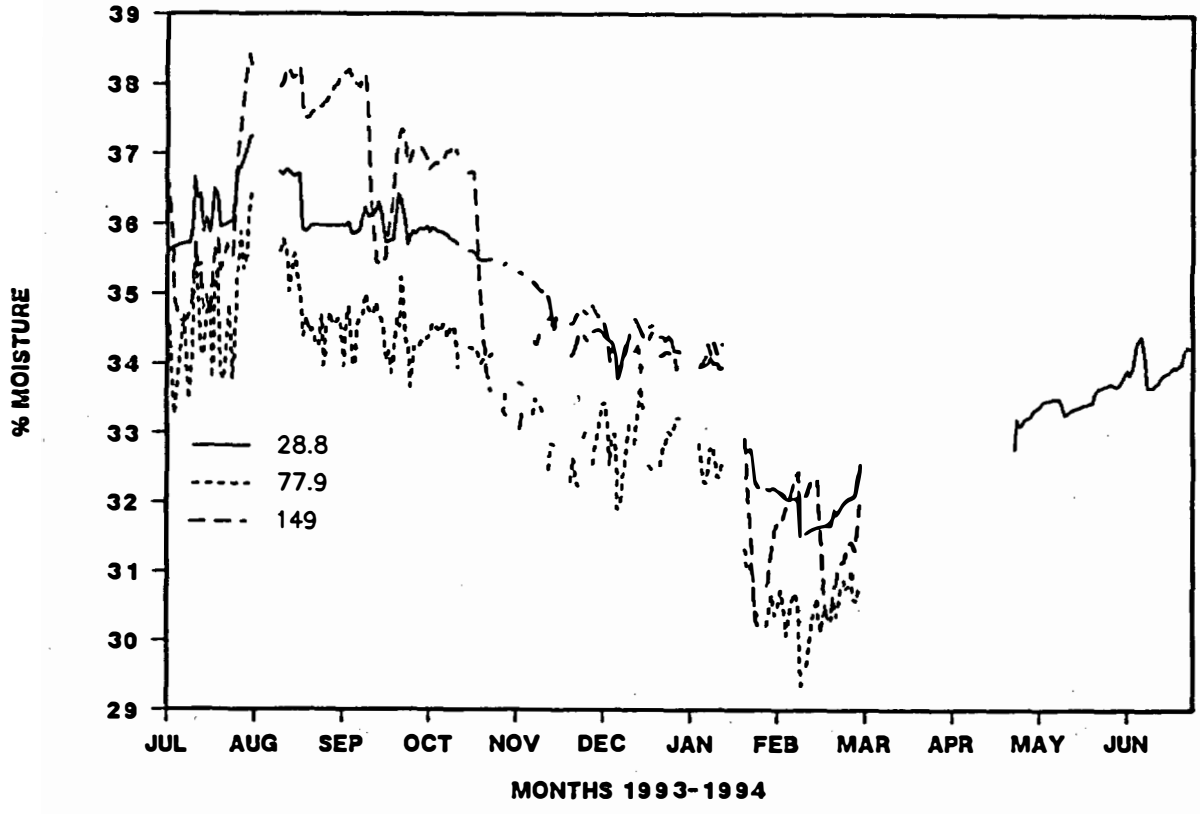

Figure 28. ORNL lysimeter 3 soil moisture.

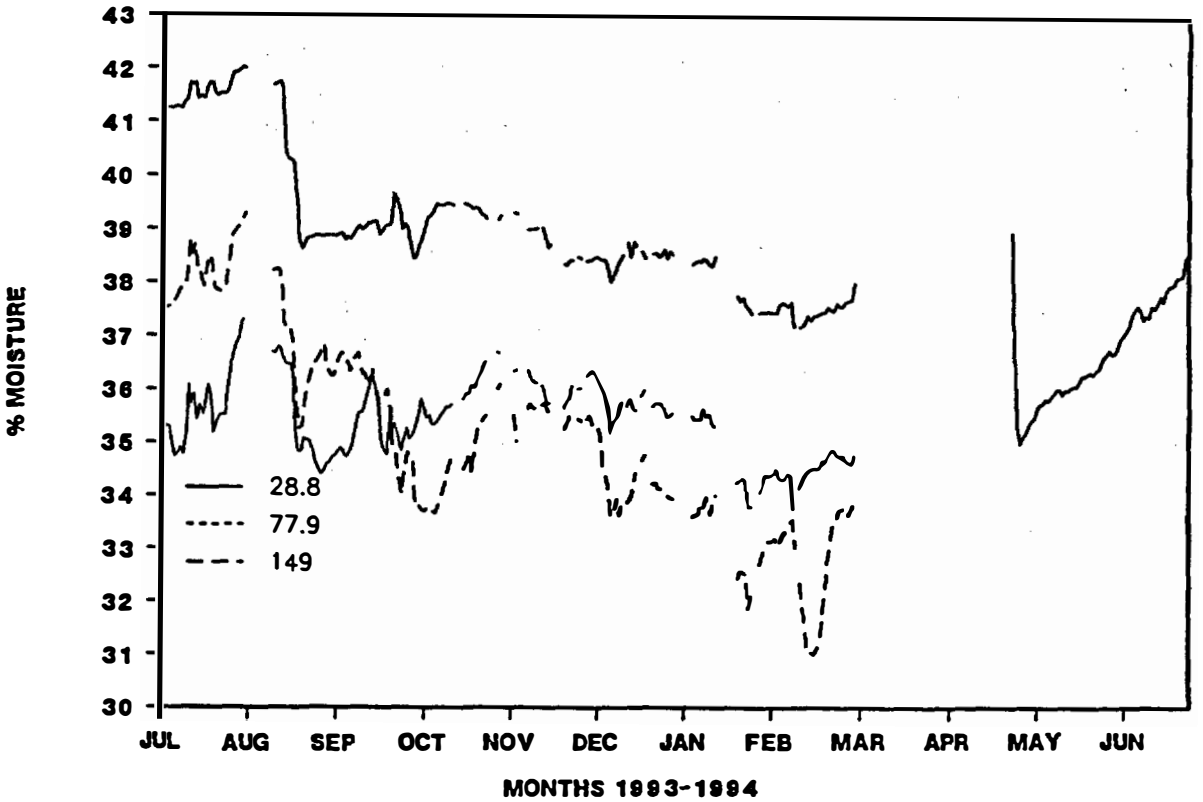

Figure 29. ORNL lysimeter 4 soil moisture.

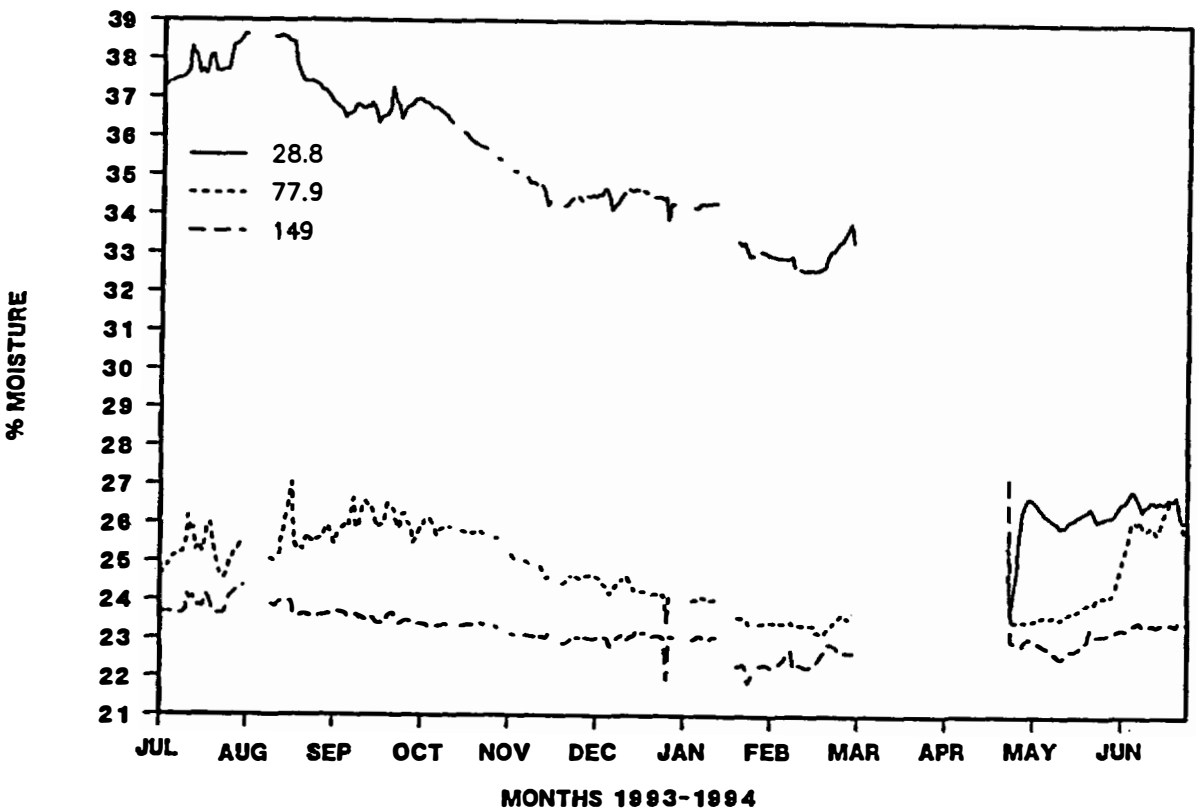

Figure 30. ORNL lysimeter 5 soil moisture.

Table 3. 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	15.3	—
1	41–62	19.6	19.2
1	62–85	23.1	20.1
1	82–107	23.3	—
1	107–133	23.3	—
1	133–153	22.6	20.7
1	153–182	23.1	—
1	182–202	23.4	20.6
2	0–41	15.7	—
2	41–62	18.6	16.4
2	62–82	20.4	18.4
2	82–107	23.1	—
2	107–133	24.3	—
2	133–153	24.5	18.8
2	153–182	24.0	—
2	182–202	24.4	19.6
3	0–41	16.3	—
3	41–62	20.0	17.3
3	62–82	23.4	21.2
3	82–107	23.3	—
3	107–133	22.7	—
3	133–153	24.0	19.9
3	153–182	23.8	—
3	182–202	24.5	20.7
4	0–41	17.4	—
4	41–62	22.1	19.4
4	62–82	22.0	21.4
4	82–107	24.1	—
4	107–133	23.6	—
4	133–153	24.2	21.5
4	153–182	24.2	—
4	182–202	23.9	21.6

a. Samples were collected on July 21, 1994.

Table 4. 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	14.4	12.7
1	41–62	15.7	16.5
1	62–85	17.4	17.0
1	82–107	17.4	17.2
1	107–133	17.9	16.4
1	133–153	16.5	17.5
1	153–182	16.5	17.6
1	182–202	18.4	18.2
2	0–41	12.8	13.1
2	41–62	15.1	15.2
2	62–82	15.6	15.5
2	82–107	16.4	15.6
2	107–133	17.4	14.4
2	133–153	14.6	16.3
2	153–182	13.3	16.0
2	182–202	11.2	14.6
3	0–41	8.3	14.1
3	41–62	12.9	16.6
3	62–82	15.6	17.0
3	82–107	17.3	17.4
3	107–133	15.8	15.6
3	133–153	18.9	17.1
3	153–182	16.5	17.7
3	182–202	13.2	18.9
4	0–41	12.4	12.6
4	41–62	15.6	17.6
4	62–82	14.2	18.6
4	82–107	14.2	18.4
4	107–133	16.7	16.6
4	133–153	15.0	18.3
4	153–182	19.9	18.8
4	182–202	25.1	20.2

a. Samples were collected on June 13, 1994.

Table 5. 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 1994	Percent difference between actual and probe
ANL-1	14.9 ± 3.1	21.7 ± 2.9	31.3
ANL-2	14.6 ± 0.9	21.9 ± 3.3	33.3
ANL-3	26.7[b]	22.2 ± 2.8	20.3
ANL-4	16.5 ± 6.4	22.7 ± 2.3	27.3
ORNL-1	27.3 ± 10.3	16.8 ± 1.3	62.5
ORNL-2	36.1 ± 4.4	14.6 ± 2.0	147.3
ORNL-3	35.5 ± 0.8	14.8 ± 3.3	139.9
ORNL-4	38.5 ± 3.0	16.6 ± 4.1	131.9

a. July 1993 through June 1994.

b. Average from one probe.

ORNL in 1992. This year's data are given in Table 4. Comparison of the neutron probe results to gravimetric results, in overall average values, shows that the probe overestimated by 5.1%. In spite of the difference between actual and measured soil moisture at the two sites, the neutron probe does provide a rapid 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 31 and 32). The uniformity of soil moisture in the ANL-E lysimeters (Figure 31) continues to be of interest given the long-term, nonuniform decrease in water infiltration into the ANL-E soil lysimeters. The lysimeters appear to have nearly the same stored water based on gravimetric data (Table 5). While action to improve drainage of the ANL-E lysimeters was taken early in the experiment, initial drainage rates cannot be restored. 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 precipitation, which resulted in drier surface soils, less water ponded on the surface of the lysimeters. During the previous reporting period (Reference 19), 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, and 4. From ANL-1, 146 L of water were removed, 95 L from ANL-2, and 82 L from ANL-4.

As shown in Figures 31 and 32, the amount of moisture within the deeper horizons of the lysimeter soil columns at ANL-E appears to have remained fairly constant (see Table 3 and References 11 and 13 through 19). At the time of the 1994 sampling, the average soil moisture of ANL-E soils had decreased from 56.3% to 54.3% of the soil moisture holding capacity, while at ORNL, this value decreased from 39.4% for 1993 to 35.2% for 1994. This year, it is noted that the ORNL percent moisture at depth was not as uniform as has been seen in past years (Figure 32).

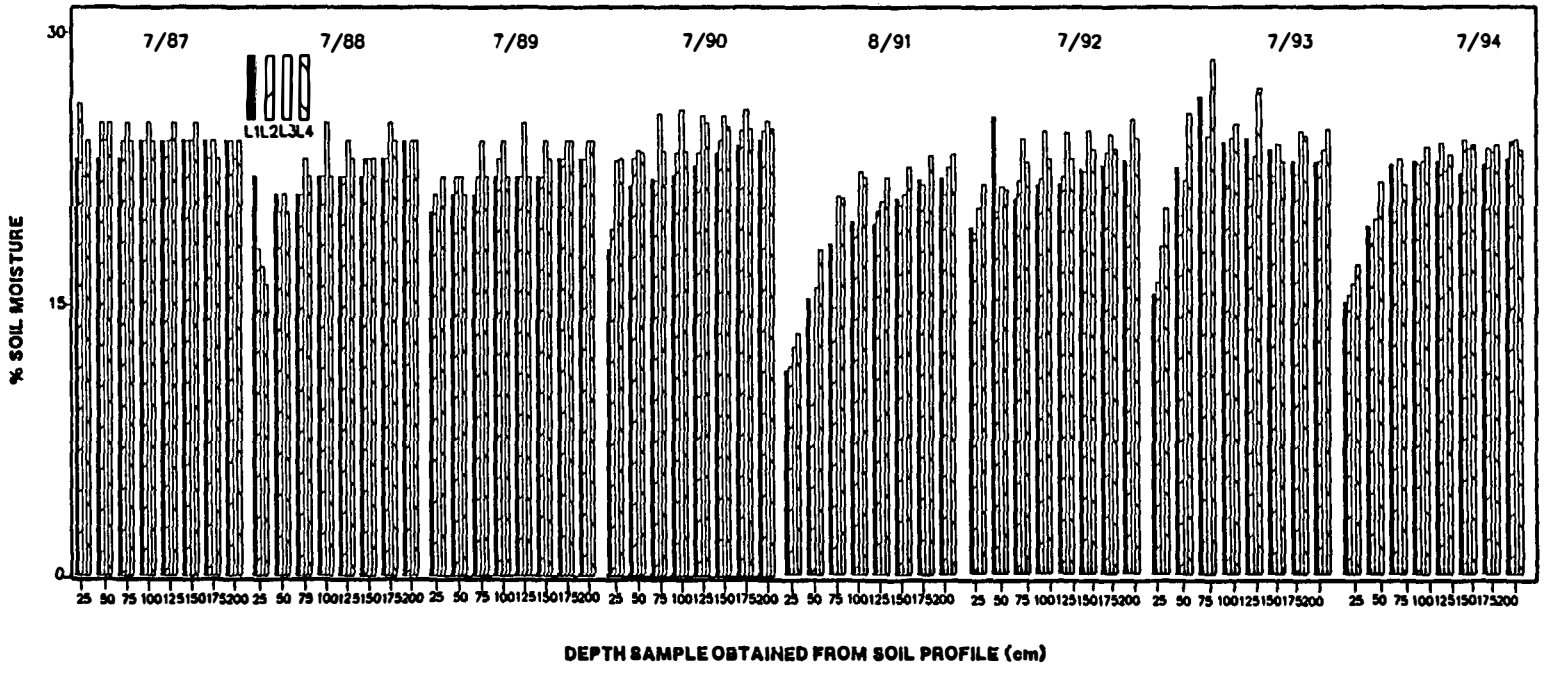

Figure 31. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.

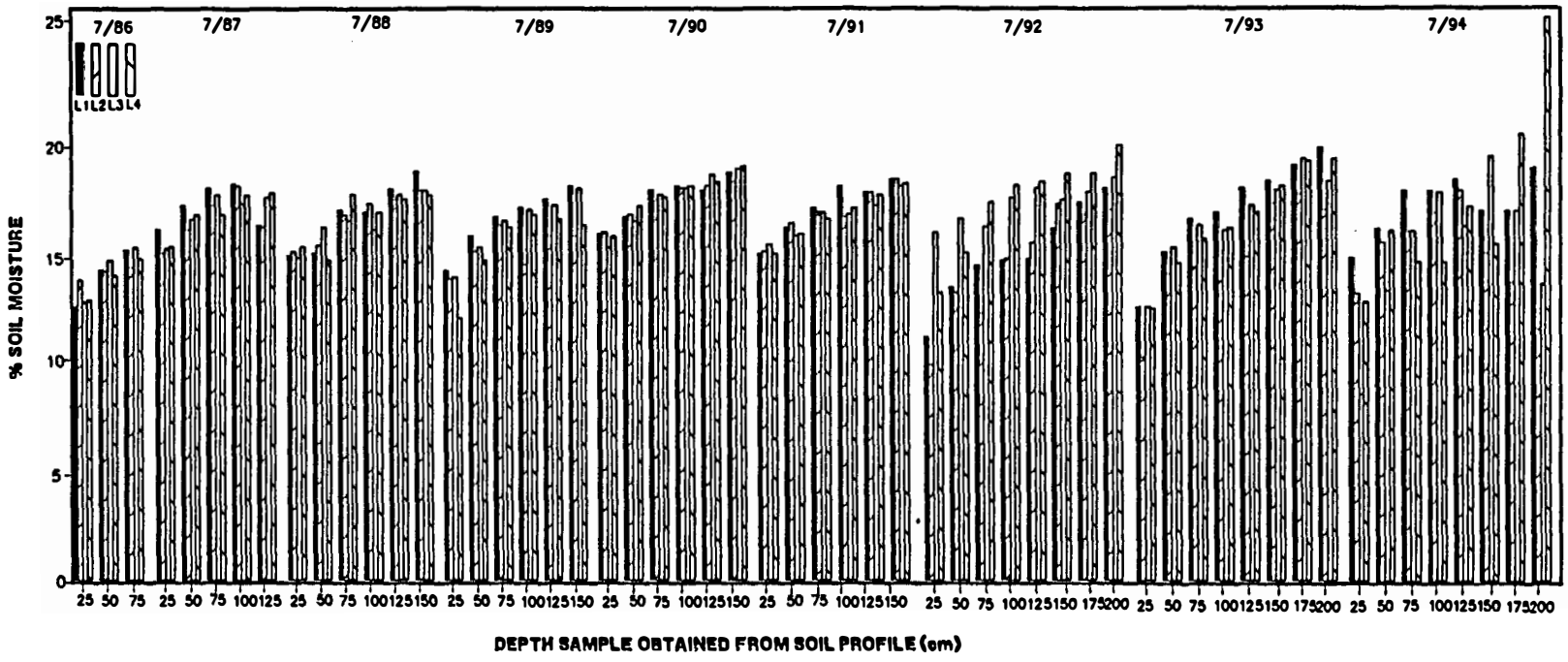

Figure 32. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.

By using the cumulative rainfall data from each site since the lysimeters were placed in operation (Figure 11), 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 has now reached 5,338.3 L; at ORNL, this value is 7,894.5 L. Figures 33 and 34 show the volumes of precipitation that have passed through the lysimeters. The throughput of precipitation is dependent on site conditions and lysimeter fill material. At ANL-E, an average of 2,146 ± 939 L, with a range of 24.3 to 64.4% of total precipitation received, has passed through the soil lysimeters, while for the control, this value was 5,318 L or 99.6% of the calculated available precipitation. For ORNL, the values were 7,066 ± 127 L (89.5%) for the soil-filled lysimeters and 7,997 L (101.3%) for the control. These trends are comparable to the previous year's trends (Reference 19). 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 33 and 34). Also, the small deviation in total yearly leachate throughput with the ORNL soil lysimeters (1.8%) continues to demonstrate that these lysimeters perform as a unit as compared to the individual drainage activity of the ANL-E lysimeters with a deviation of 43.8%.

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 288 ± 14 L (CV 4.9%) of water that was removed as a combination of leachate and standing water. During the previous year, this volume was 613 ± 14 L (CV 4.9%).

The total volumes of precipitation that have moved through the lysimeters represent an average of 3.0 pore volumes for the ANL-E soil lysimeters and 11.3 pore volumes for soil lysimeters at ORNL, while the controls at ANL-E and ORNL were 11.6 and 12.4 pore volumes, respectively. These data show that the ORNL soil lysimeters have had an average of 3.8 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 6 for cup placement). Because of funding levels, however, it has not been possible to follow this protocol. During the first 5 years of operation, water samples from only cups 3 were routinely analyzed at the sites. However, for the past 4 years, water from cups 1 has also been analyzed and reported. Water from cups 2 was collected again during the third quarter for analysis at ANL-E; however, the collection was not made at ORNL (Tables 6 and 7, respectively).

Tabulated results of beta and gamma analysis for the liquid samples taken during the period are found in Tables 6 and 7. 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 35 through 41.

As has been reported in the past (References 11 and 13 through 19), not all nuclides are appearing consistently in either the water obtained from the cups or the leachate collectors. The nuclide that continues to appear with the most regularity at both sites is Sr-90. Consistent, significant, increasing occurrences of this nuclide continue in

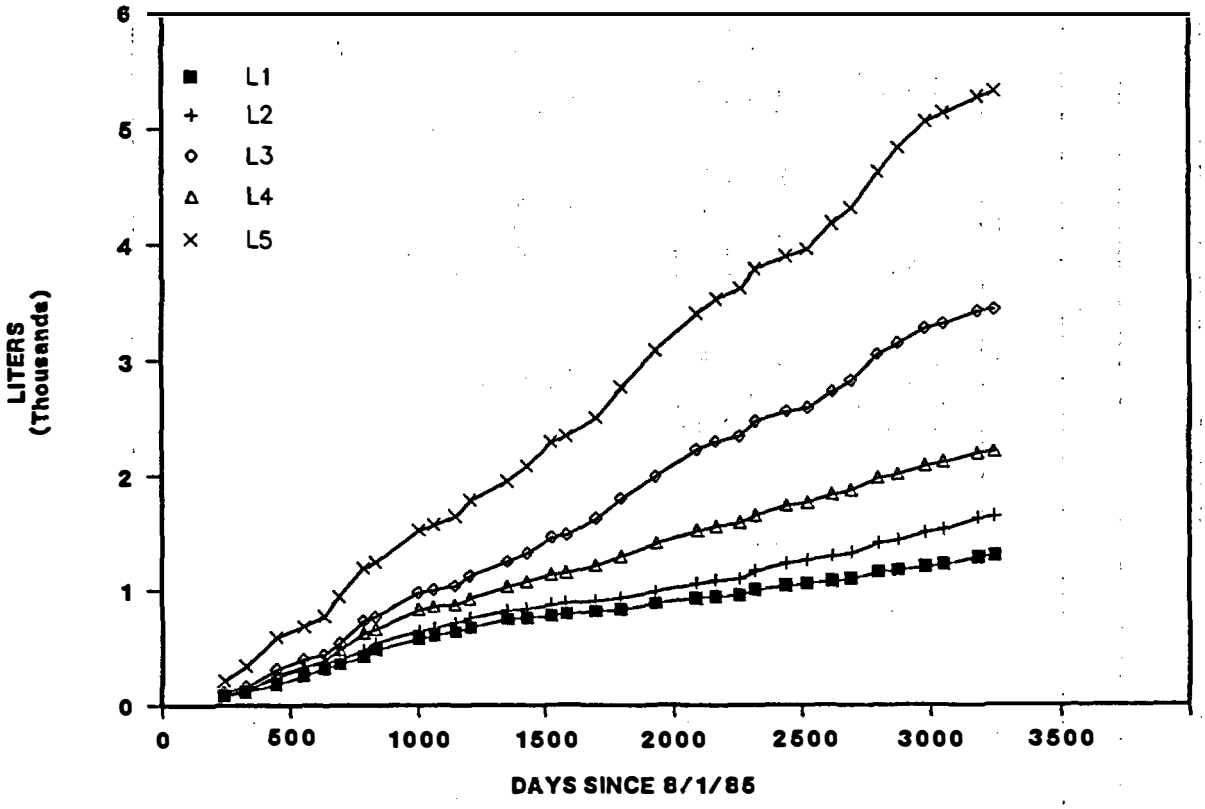

Figure 33. ANL-E cumulative volume of leachate from lysimeters.

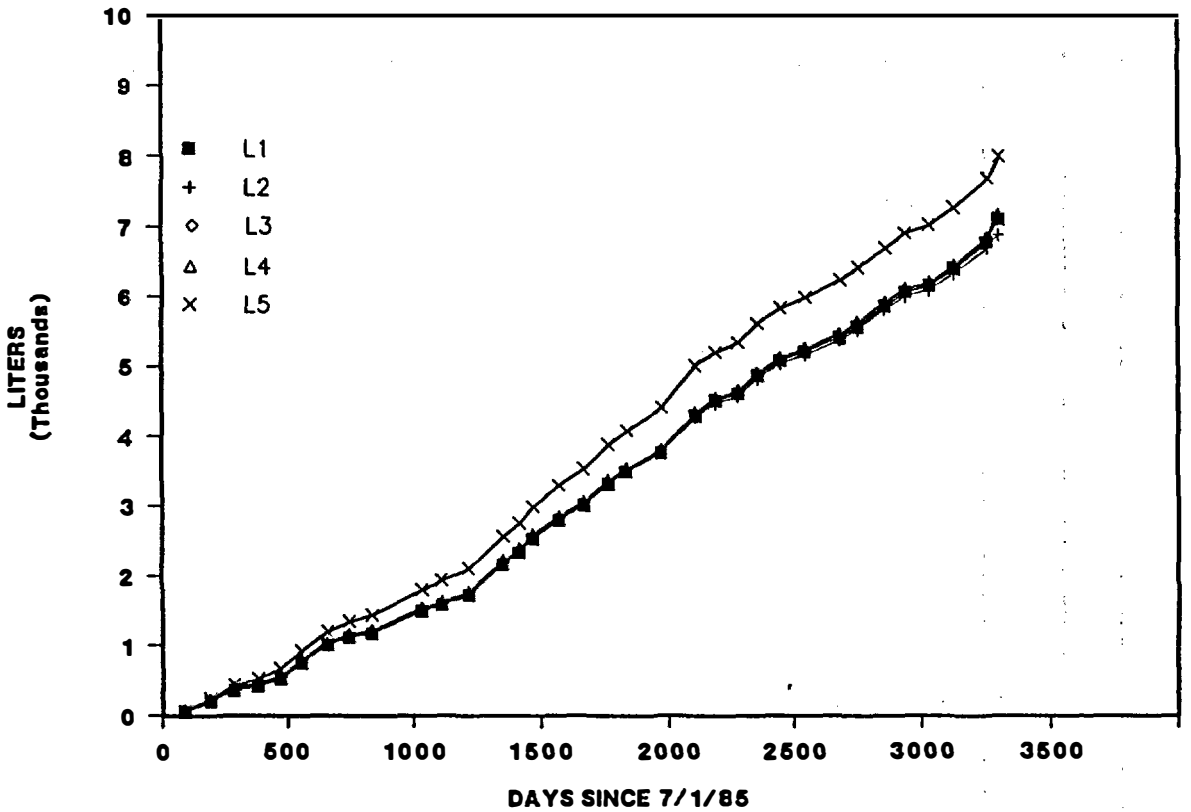

Figure 34. ORNL cumulative volume of leachate from lysimeters.

Table 6. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 9 (1993–1994).

	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
Sample identification	Sep 93	Dec 93	Apr 94	Jun 94	Sep 93	Dec 93	Apr 94	Jun 94	Sep 93	Dec 93	April 94	June 94
Lys 1[b]	<50	<50	<50	<200	<100	<100	<100	<400	0.3 ± 0.3	0.68 ± 0.28	0.4 ± 0.2	2.3 ± 0.4
Lys 2	<50	<50	<50	<200	<100	<100	<100	<400	0.7 ± 0.3	1.8 ± 0.3	0.8 ± 0.2	1.8 ± 0.4
Lys 3	<50	<50	<50	<200	<100	<100	<100	<400	250 ± 6	128 ± 4	268 ± 5	115 ± 5
Lys 4	<50	<50	<50	<200	<100	<100	<100	<400	6.2 ± 0.4	13.9 ± 0.6	14 ± 1.0	5.8 ± 0.6
Lys 5	<50	<50	<50	<200	<100	<100	<100	<400	1,240 ± 25	1,470 ± 30	1,685 ± 30	2,150 ± 40
Lys 1-3[c]	119 ± 27	334 ± 80	<50	<200	388 ± 24	277 ± 23	<100	<400	3.8E+4 ± 500	2.9E+4 ± 400	1.5E+4 ± 300	2.4E+4 ± 379
Lys 2-3	137 ± 35	94 ± 21	<50	<200	340 ± 27	210 ± 15	190 ± 19	<400	9,915 ± 70	9,000 ± 70	8,300 ± 70	1.0E+4 ± 60
Lys 3-3	<50	<50	<50	<200	<100	175 ± 25	113 ± 19	<400	122E+4 ± 1.7E+4	127E+4 ± 1.8E+4	104E+4 ± 2E+4	102E+4 ± 1.9E+4
Lys 4-3	<50	<50	<50	<200	316 ± 65	155 ± 40	<100	<400	4.9E+4 ± 500	4.0E+4 ± 450	5.8E+4 ± 550	5.1E+4 ± 640
Lys 5-3	<50	2,830 ± 85	<50	<200	17.5E+4 ± 530	11.7E+4 ± 470	18.3E+4 ± 5.5E+2	27E+4 ± 1.0E+3	7.1E+4 ± 470	3.9E+4 ± 450	4.4E+4 ± 500	5.4E+4 ± 385
Lys 1-1[c]	<50	<50	<50	<200	288 ± 36	200 ± 14	<100	<400	13.8 ± 1.6	19 ± 3	28 ± 3	24 ± 3
Lys 2-1	<50	<50	<50	<200	—	—	<100	<400	1.2 ± 1.4	2.1 ± 1.2	—	—[d]
Lys 3-1	143 ± 24	96 ± 19	<50	<200	287 ± 22	<100	<100	<400	5,700 ± 80	5,100 ± 80	7,100 ± 90	3,900 ± 100
Lys 4-1	<50	<50	<50	<200	320 ± 24	155 ± 40	<100	<400	22.3 ± 1.4	343 ± 22	350 ± 20	425 ± 12
Lys 5-1	<50	960 ± 95	<50	<200	232 ± 36	345 ± 35	<100	<400	1,424 ± 28	1,900 ± 30	2,200 ± 40	2,500 ± 50
Lys 1-2	—	—	<50	—	—	—	<100	—	—	—	28 ± 3.0	—
Lys 2-2	—	—	<50	—	—	—	<100	—	—	—	1.7 ± 1.9	2.9 ± 2.8
Lys 3-2	—	—	<50	—	—	—	<100	—	—	—	19.6 ± 4.1	—
Lys 4-2	—	—	<50	—	—	—	<100	—	—	—	1.6 ± 1.3	—
Lys 5-2	—	—	<50	—	—	—	<100	—	—	—	3,100 ± 30	—

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 7. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 9 (1993–1994).

Sample identification	Concentration (pCi/L)[a]							
	Co-60				Cs-137			
	Sep 93	Dec 93	Apr 94	Jun 94	Sep 93	Dec 93	Apr 94	Jun 94
Lys 1[b]	49 ± 11	–2.2 ± 11	–14 ± 65	14 ± 43	19 ± 40	–7.0 ± 14	35 ± 32	2.7 ± 51
Lys 2	2.7 ± 54	–0.3 ± 7.0	–5.4 ± 57	11 ± 51	2.7 ± 57	–0.5 ± 7.0	2.7 ± 54	11 ± 46
Lys 3	–32 ± 62	3.5 ± 11.9	–24 ± 65	–11 ± 57	–19 ± 59	1.6 ± 14	24 ± 35	11 ± 54
Lys 4	32 ± 35	–13 ± 22	14 ± 43	–5.4 ± 57	35 ± 40	–1.3 ± 15	11 ± 46	–21 ± 59
Lys 5	–2.7 ± 45	–3 ± 12	11 ± 43	14 ± 49	28 ± 57	13 ± 7.0	127 ± 43	1.46E+4 ± 270
Lys 1-3[c]	–5.4 ± –113	–29 ± 113	8.1 ± 46	19 ± 81	97 ± 68	16 ± 95	–32 ± 76	103 ± 49
Lys 2-3	56 ± 70	–40 ± 119	–2.7 ± 68	–8.1 ± 108	–32 ± 103	14 ± 78	57 ± 38	5.4 ± 103
Lys 3-3	22 ± 105	2.7 ± 97	5.4 ± 54	2.1 ± 122	30 ± 103	24 ± 76	–5.4 ± 62	38 ± 78
Lys 4-3	35 ± 70	14 ± 70	38 ± 38	–16 ± 108	49 ± 81	–11 ± 84	32 ± 73	38 ± 78
Lys 5-3	14 ± 81	–11 ± 92	–11 ± 70	30 ± 70	1.8E+4 ± 270	214 ± 89	7,027 ± 270	2.1E+4 ± 540
Lys 1-1[c]	–65 ± 127	5.4 ± 45	–14 ± 46	49 ± 19	27 ± 97	8.1 ± 86	70 ± 35	249 ± 62
Lys 2-1	–8.1 ± 100	2.7 ± 92	–19 ± 84	35 ± 62	30 ± 89	8.1 ± 97	54 ± 54	27 ± 59
Lys 3-1	–13 ± 113	22 ± 92	–11 ± 40	51 ± 57	32 ± 89	24 ± 73	–2.7 ± 49	86 ± 46
Lys 4-1	22 ± 70	–19 ± 97	19 ± 48	–11 ± 121	104 ± 84	16 ± 78	43 ± 65	135 ± 68
Lys 5-1	22 ± 57	11 ± 76	40 ± 32	27 ± 81	76 ± 73	235 ± 100	159 ± 62	324 ± 81
Lys 1-2[d]	—	—	—	—	—	—	—	—
Lys 2-2[d]	—	—	—	—	—	—	—	—
Lys 3-2[d]	—	—	—	—	—	—	—	—
Lys 4-2[d]	—	—	—	—	—	—	—	—
Lys 5-2[d]	—	—	—	—	—	—	—	—

Table 7. (continued).

Sample identification	Concentration (pCi/L)[a] Sb-125 Sep 93	Sb-125 Dec 93	Sb-125 Apr 94	Sb-125 Jun 94	Sr-90 Sep 93	Sr-90 Dec 93	Sr-90 Apr 94	Sr-90 Jun 94
Lys 1[b]	—[e]	—[e]	—[e]	—[e]	351 ± 27	865 ± 54	972 ± 81	351 ± 27
Lys 2	—[e]	—[e]	—[e]	—[e]	22 ± 8.6	43 ± 5.4	32 ± 13	62 ± 8.2
Lys 3	—[e]	—[e]	—[e]	—[e]	10 ± 7.3	15 ± 4.6	7.8 ± 7.8	27 ± 5.4
Lys 4	—[e]	—[e]	—[e]	—[e]	3.2 ± 5.4	0.3 ± 2.7	4.3 ± 6.8	1.9 ± 2.3
Lys 5	—[e]	—[e]	—[e]	—[e]	1,000 ± 54	1,567 ± 108	649 ± 54	1,783 ± 297
Lys 1-3[c]	—[e]	—[e]	—[e]	—[e]	7.8 ± 2,703	7.6E+4 ± 5,405	5.4E+4 ± 2,703	8.1E+4 ± 2,702
Lys 2-3	—[e]	—[e]	—[e]	—[e]	1.1 ± 540	1.1E+4 ± 540	1.0E+4 ± 270	1.4E+4 ± 540
Lys 3-3	—[e]	—[e]	—[e]	—[e]	25.7 ± 8,108	22.9E+4 ± 1.3E+4	16.2E+4 ± 5,405	32.4E+4 ± 2.7E+4
Lys 4-3	—[e]	—[e]	—[e]	—[e]	1.0E+4 ± 810	1.2E+4 ± 2,432	8,378 ± 540	1.7E+4 ± 810
Lys 5-3	—[e]	—[e]	—[e]	—[e]	2.4 ± 1,081	3.2 ± 13	9,459 ± 1,891	2.5E+4 ± 1,081
Lys 1-1[c]	—[e]	—[e]	—[e]	—[e]	1.0E+4 ± 540	1.4E+4 ± 540	1.4E+4 ± 1,081	2.1E+4 ± 1,081
Lys 2-1	—[e]	—[e]	—[e]	—[e]	257 ± 35	486 ± 54	540 ± 54	170 ± 13
Lys 3-1	—[e]	—[e]	—[e]	—[e]	224 ± 35	324 ± 54	324 ± 54	146 ± 11
Lys 4-1	—[e]	—[e]	—[e]	—[e]	12 ± 13	35 ± 19	32 ± 19	4.3 ± 2.9
Lys 5-1	—[e]	—[e]	—[e]	—[e]	151 ± 32	186 ± 35	259 ± 43	405 ± 27
Lys 1-2[d]	—	—	—	—	—	—	—	—
Lys 2-2[d]	—	—	—	—	—	—	—	—
Lys 3-2[d]	—	—	—	—	—	—	—	—
Lys 4-2[d]	—	—	—	—	—	—	—	—
Lys 5-2[d]	—	—	—	—	—	—	—	—

a. Concentration ±2 sigma.

b. One-L subsample from leachate collector.

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

d. No samples were taken from cups number 2 in this reporting period.

e. None detected.

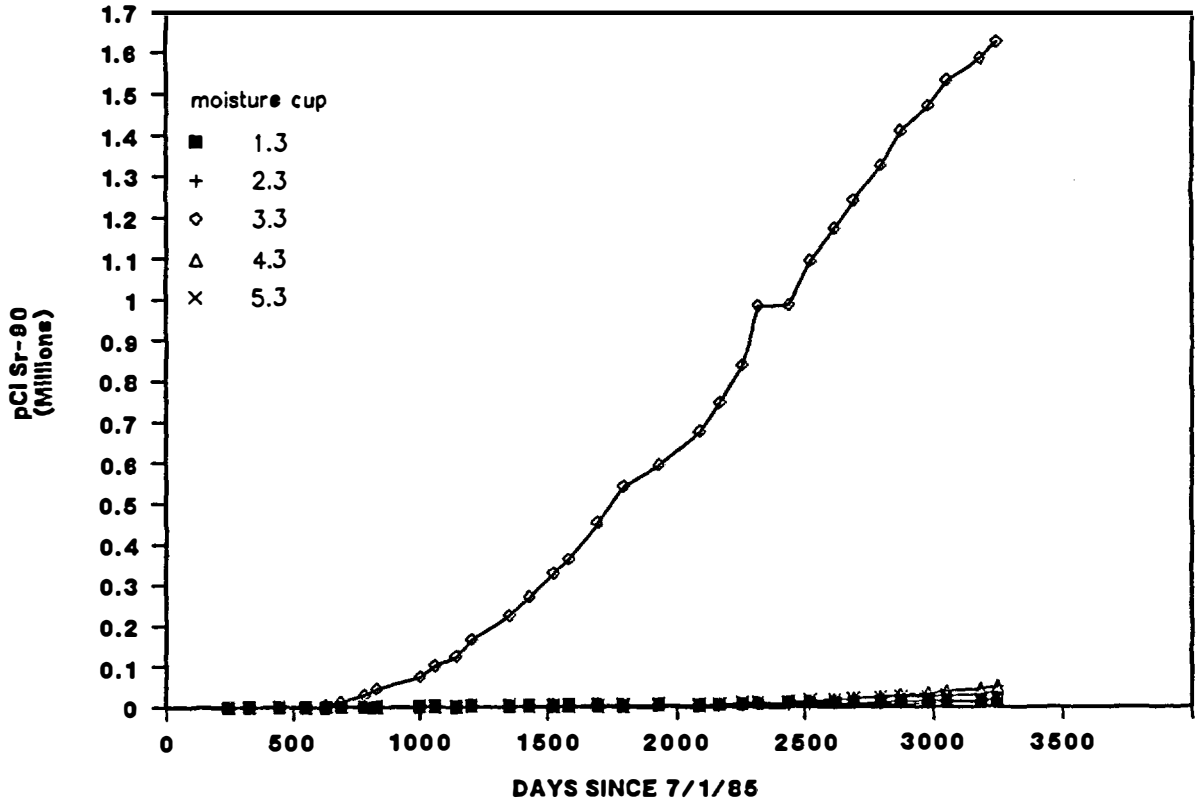

Figure 35. ANL-E cumulative Sr-90 collected in moisture cups number 3.

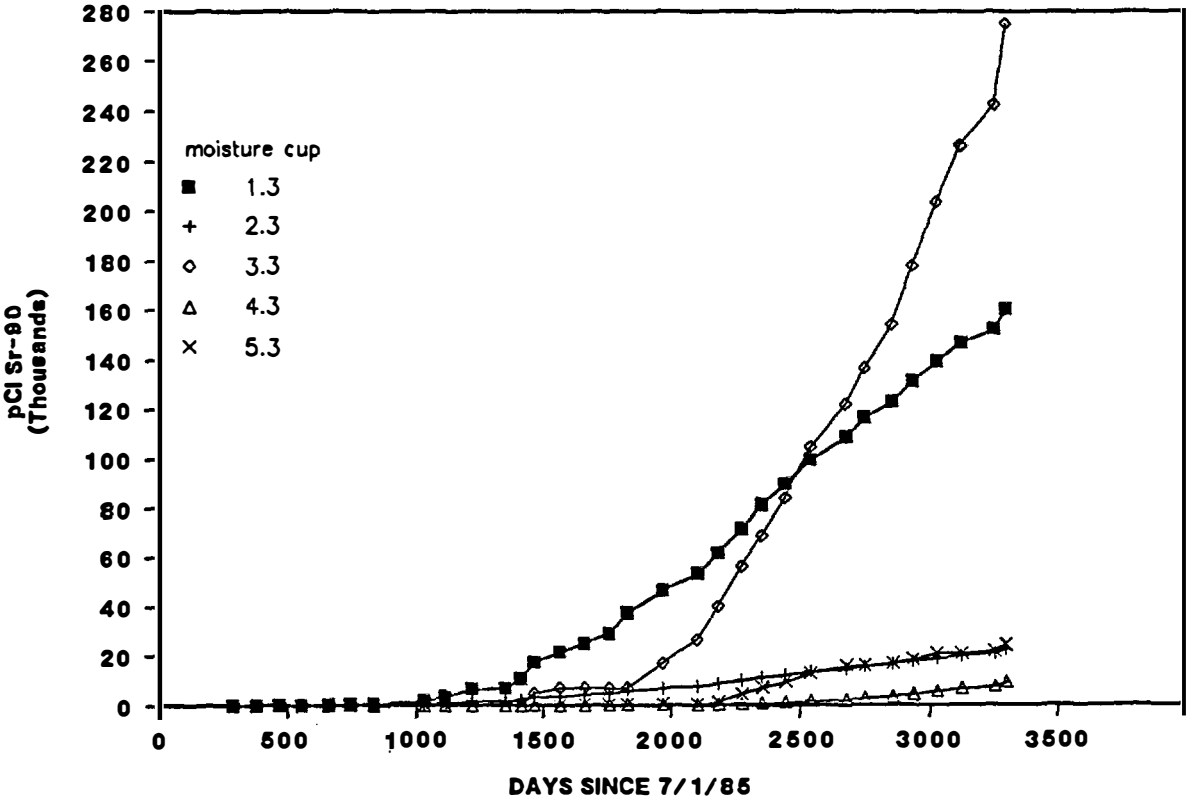

Figure 36. ORNL cumulative Sr-90 collected in moisture cups number 3.

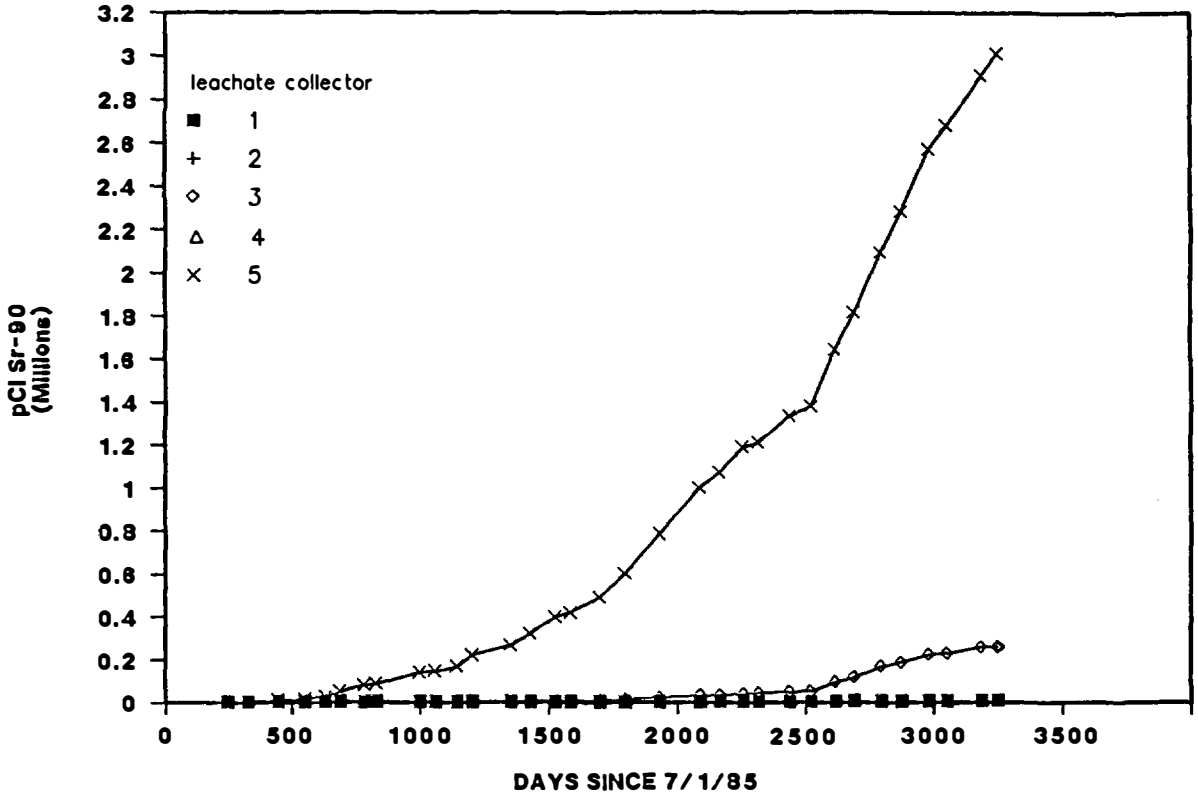

Figure 37. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

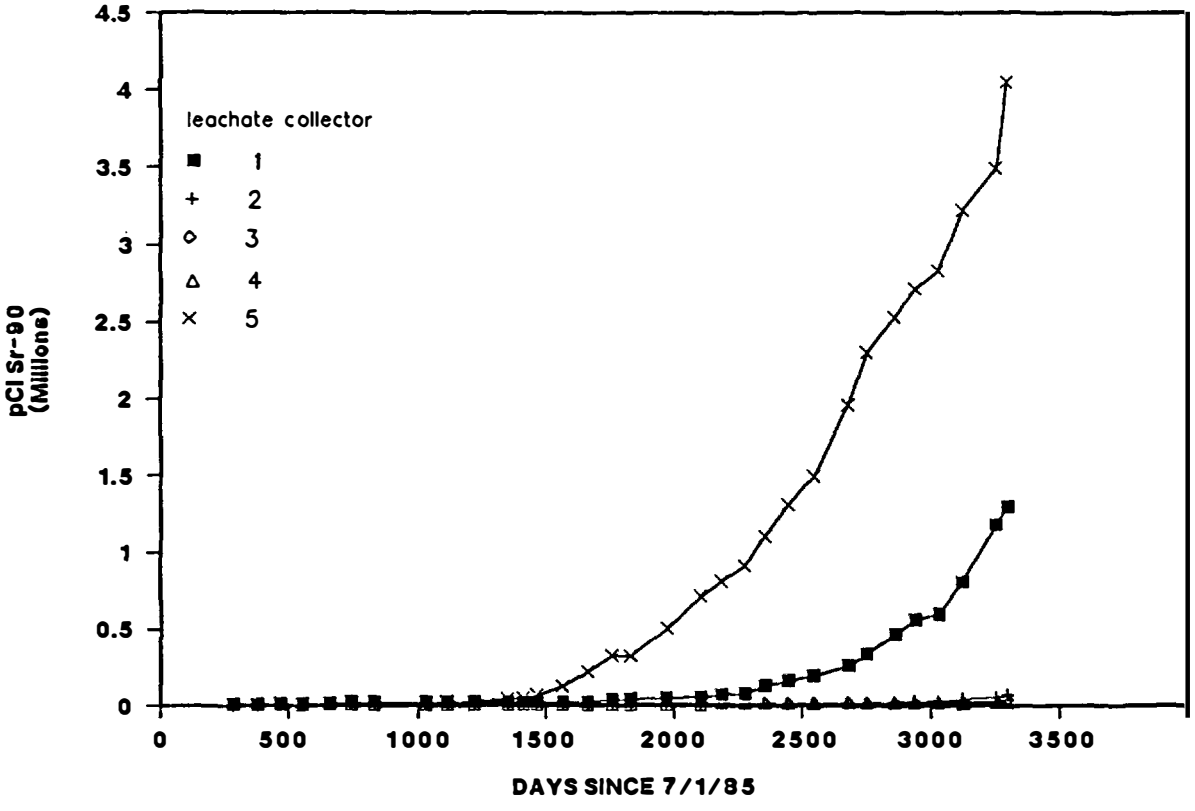

Figure 38. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

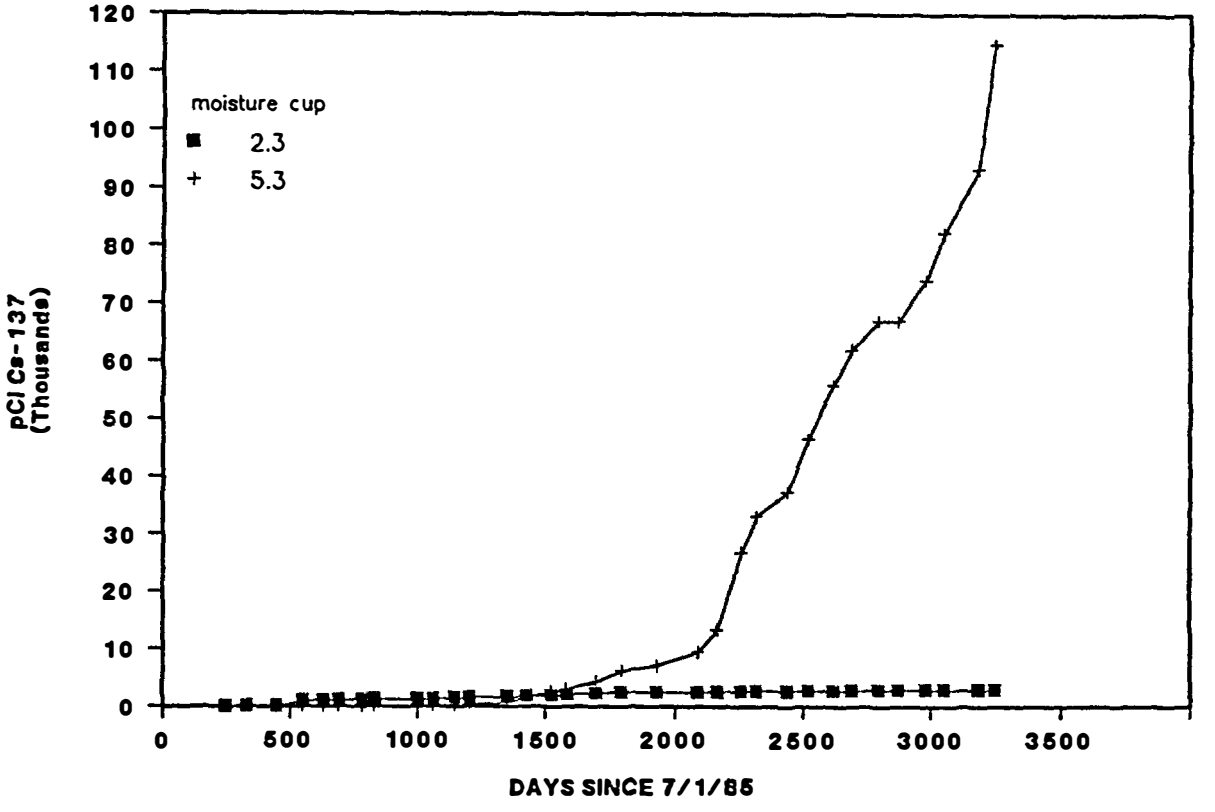

Figure 39. ANL-E cumulative Cs-137 collected in moisture cups number 3.

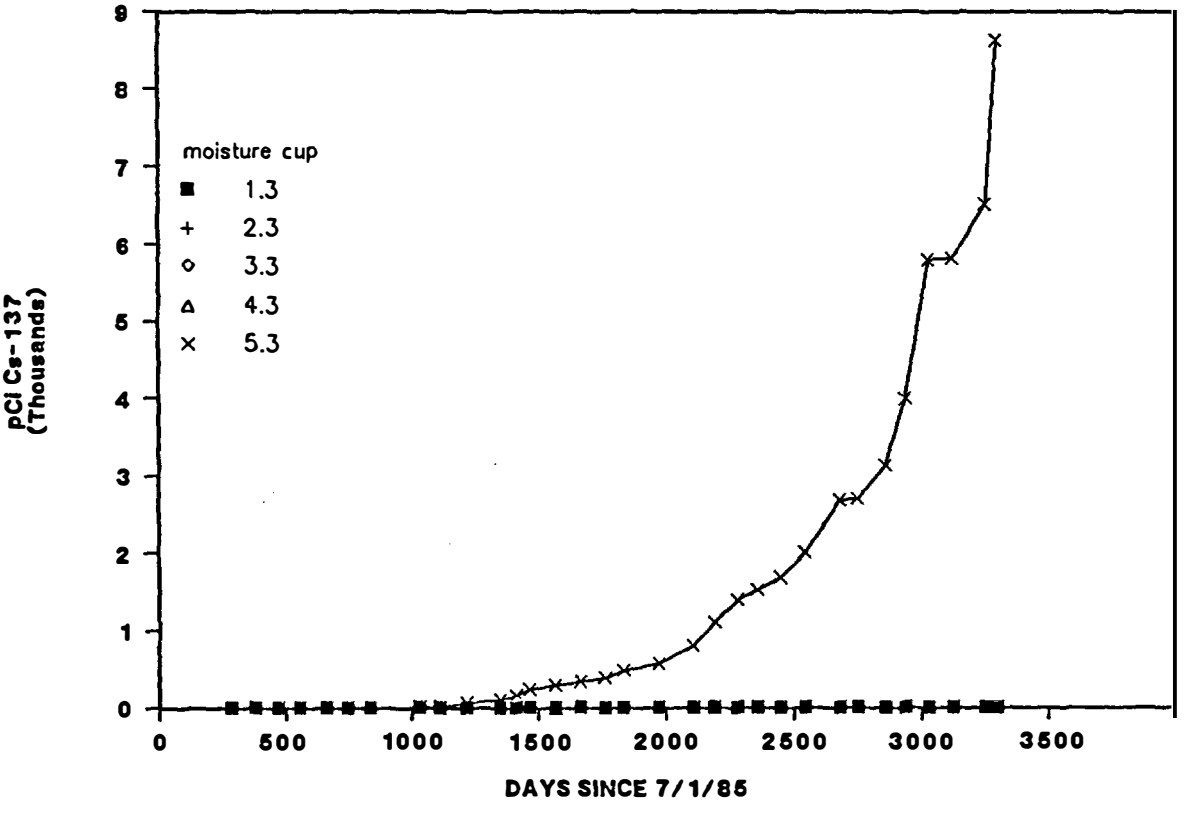

Figure 40. ORNL cumulative Cs-137 collected in moisture cups number 3.

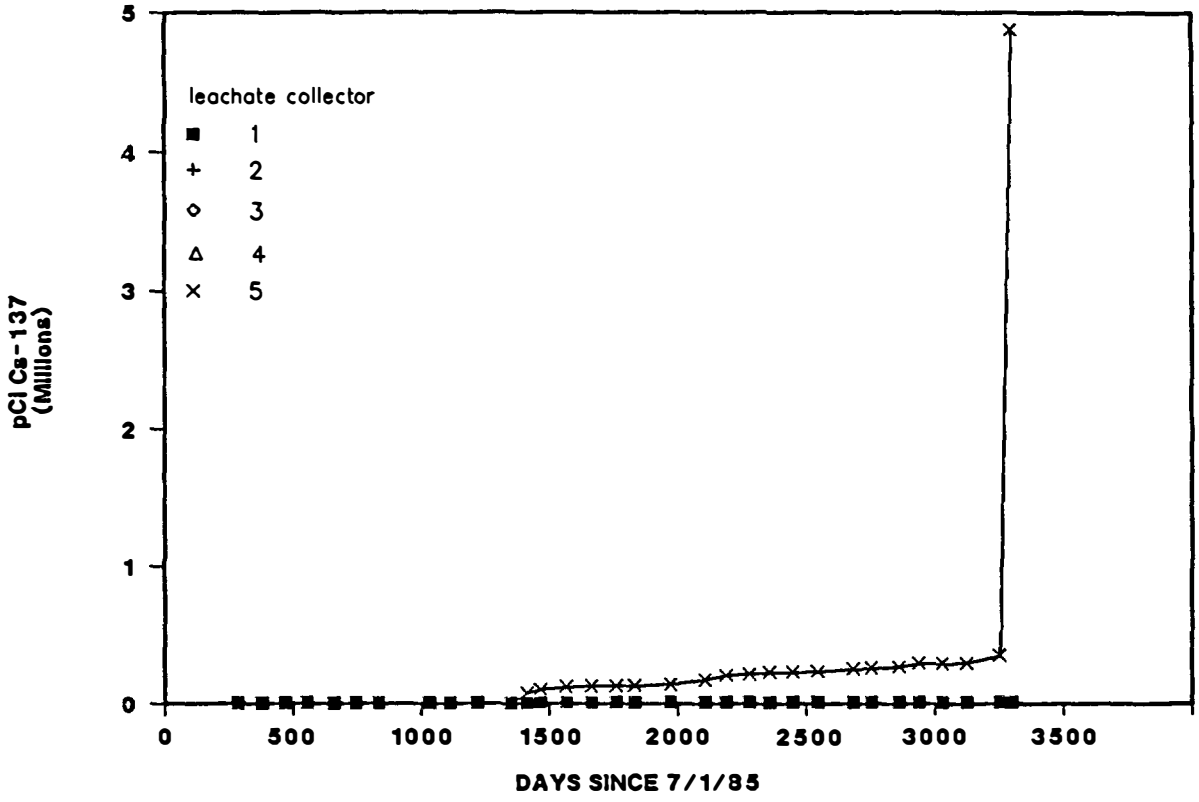

Figure 41. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

all cups 3 at both ANL-E (range of 28 to 66%) and ORNL (range of 22 to 107%) (Tables 6 and 7; Figures 35 and 36). There continues to be standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a cumulative total of 1,628,875 pCi from 3-3 at ANL-E (an increase of 15% over last year) (Figure 35) and 274,877 pCi from 3-3 at ORNL (55% increase over last year), which continues to increase beyond ORNL 1-3 (Figure 36). 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 (32%) continues in ORNL 5-3 as well as a 107% increase into ORNL 4-3 (second year in a row that over 100% increase occurred) (Figure 36). 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; at ANL-E, these values are at least one order of magnitude larger than the largest cumulative release from a soil lysimeter (Figures 37 and 38). This is comparable to the previous year's findings (References 11 and 13 through 19), except a larger release in ORNL 1 (130%) increased the total Sr-90 to 1,295,843 pCi and places it in the same order of magnitude as ORNL 5 (4,045,025 pCi). 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 8). There was an increase in the total cumulative quantity of Sr-90 released in the leachate water in all lysimeters at both sites except ORNL 4 (Tables 6 and 7). For ORNL lysimeters 1 and 2, the percent of total inventory of the nuclide released in leachate water continues to be greater than that in the cups (Table 8). These data follow a trend seen over the past 54 months and make it appear that a pulse of Sr-90 could be moving through the soil columns of those two ORNL lysimeters. For the control lysimeters at both sites, there was substantially more Sr-90 in the leachate than in cups 3 (there continues to be two orders of magnitude difference for both locations).

Table 8. Percent of total Sr-90 and Cs-137 inventory per lysimeter released to moisture cups and leachate water through July 1994.

		Percent total inventory Sr-90				Percent total inventory Cs-137			
		Moisture cups		Leachate water		Moisture cups		Leachate water	
Lysimeter number	Solidification agent	ANL-E	ORNL	ANL-E	ORNL	ANL-E	ORNL	ANL-E	ORNL
1	Cement	1.0E-4	8.8E-4	3.7E-5	7.1E-3	—[a]	—	—	2.0E-6
2	Cement	3.5E-4	6.7E-4	7.0E-5	2.0E-3	9.0E-7	—	—	1.0E-7
3	VES	5.9E-3	1.0E-3	9.5E-4	1.4E-4	—	—	—	1.5E-6
4	VES	1.1E-3	1.5E-4	1.5E-4	4.7E-4	—	—	—	3.0E-7
5	Cement	2.0E-4	7.4E-4	1.7E-2	1.2E-1	3.8E-5	4.0E-7	—	1.8E-3

a. Percent released is essentially equal to zero.

The percent of total Sr-90 being measured in the leachate water and cups 3 continues to be inconsistent between the two sites (Table 8). 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 from 6.5 to 7.5 times that of ANL-E (Table 8). From a waste form standpoint, the cement waste forms at ORNL are releasing more Sr-90 into both Cups 3 and the leachate than those at ANL-E. On the other hand (except for the leachate water from ANL-4), there is more release of Sr-90 from the ANL-E lysimeters with the VES waste forms than occurs at ORNL (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 6; Figure 39). Since Cs-137 began appearing in ANL 5-3, the quantity of this nuclide has dramatically increased in each of the sampling periods. This year, there was a significant increase of 51% (Figure 39). The remaining ANL-E cups 3 received sporadic releases of Cs-137 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 40) and have continued in subsequent samplings for a total of 8,618 pCi, which is an 1,160% 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 4 years (Figure 41 and Table 7). Breakthrough of Cs-137 into the ORNL-5 leachate collector occurred in November 1988, some 7 months after its occurrence in moisture cup ORNL 5-3 (Figures 40 and 41). This year, there was a dramatic increase of 1,567% for a total cumulative release of 4,871,810 pCi (Table 7).

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

By using a matrix (as in Table 8), several comparisons can be made based on the intra- and intersite data. Overall, of the nuclides contained in the waste forms (Reference 12), 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 8). Compared to Sr-90, the recovery of Cs-137 continues to appear 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 continued reoccurrence in ORNL 1-3 (Table 7). On the other hand, this nuclide has been consistently occurring in ORNL 5-3 (Figure 40) and in the leachate collector of the ORNL-5 lysimeter (Figure 41). Cesium-137 has also occurred in the moisture cups of ANL-E lysimeters 2 and 5. This year, Cs-137 was also seen in cups from the other three ANL-E lysimeters (Table 6) 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 of the soil-filled lysimeters (Table 8) 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 in these lysimeters. This behavior might be influenced by the amount of water passing through the lysimeters (Figures 33 and 34). In the case of the ORNL lysimeters, which have had as much as five times more water pass through, there has been considerably more Sr-90 in the leachate collectors than in the cups (except ORNL-3) (Table 8). The influence of the soil column can be seen with a comparison between the soil-filled lysimeters and the sand-filled controls. At both locations, large quantities of Sr-90 have passed through the controls.

As seen from Table 2, 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 12). 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). This is the second year in which Sr-90 has been detected in ANL-E cups 2, which are located at the bottom outside edge of the soil columns. The amounts observed in the sand-filled lysimeter 5-2 cup total 435 pCi, while the soil-filled lysimeter cups have seen only traces. Outward movement of Sr-90 to these cups provides an indication of dispersivity in these soil and sand columns, which may be applied to model the release from those waste forms. 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 each site when compared to other cups at that same site (Table 8). Figures 35 and 36 show that cumulative total quantities of Sr-90 in water retrieved from cups 3 are higher from the soil lysimeters with the higher loaded waste forms (range of 37 to 97% higher). The same was also true for the four soil lysimeters at each site when the quantity of Sr-90 in leachate collector water is compared (43 to 97%). 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 8). 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 (31 to 554%), while two of the five have the higher Sr-90 released to the leachate collector water (255 and 518%).

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 8). 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 amount of Sr-90 than the other ANL-E collectors but a significant amount less than the cups 3 of those lysimeters (7 and 6 times, respectively). The amount of available nuclide that continues to move into the leachate collector of sand-filled ANL-5 is much greater than that of the other ANL-E lysimeters (14 to 446 times greater), 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 0.123% of the Sr-90 contained in ORNL-5 has now been recovered in leachate from that lysimeter. The amount of available Sr-90 that has moved into the ORNL-5 leachate collector remains significantly higher than the other ORNL collectors (17 to 904 times higher).

Recovery of Sr-90 in the cups at both sites is comparable for those lysimeters containing the cement waste forms and those containing VES waste forms. These data 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 8). Soil type and precipitation (environmental factors) appear to be the controlling factors.

Cumulative Fractional Releases Compared. Table 9 is a comparison of cumulative fractional releases to leachate collectors from field testing EPICOR-II waste forms in lysimeters to releases from bench-leach-testing similar waste forms in demineralized and sea waters as reported in References 10 and 15. Releases observed in the lysimeters are at least four orders of magnitude less for Sr-90 in soil and at least five orders of magnitude less for Cs-137 in soil. It is interesting to note that release of Sr-90 in the sand-filled lysimeter is

only one or two orders of magnitude less than bench-test results with demineralized water. At the present rate of increase (Figures 37 and 38), these cumulative fractional releases will be of similar magnitude in a couple of years.

Upward Migration of Radionuclides at ORNL. 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 cesium and Sr-90. Analysis detected Cs-137, Cs-134, and Sr-90. 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.

On January 31, 1994, two cores of sand 80 cm long from lysimeter number 5 were collected to be analyzed for cesium and strontium. One core was collected from the side of the lysimeter near

Table 9. Cumulative fractional releases from lysimeter field testing compared to those from bench leach testing (10,15).

				Cumulative fractional release			
						Leachate collectors	
Test type	Prefilter number	Solidification agent	Radionuclide	Demineralized water	Seawater	Soil	Sand
Bench,[a] INEL	7	VES[a]	Sr-90	4.5E-2	—	—	—
Bench,[a] INEL	7	Cement[a]	Sr-90	7.8E-2	—	—	—
Bench,[a] INEL	7	VES[a]	Cs-137	4.0E-2	—	—	—
Bench,[a] INEL	7	Cement[a]	Cs-137	9.0E-2	—	—	—
Bench,[a] INEL	7	VES	Cs-137	2.1E-3	6.4E-2	—	—
Bench,[a] INEL	7	Cement	Cs-137	4.8E-2	9.0E-2	—	—
Bench,[a] INEL	24	VES	Cs-137	3.0E-4	1.3E-2	—	—
Bench,[a] INEL	24	Cement	Cs-137	2.3E-2	2.6E-2	—	—
Field, ANL-E	7	VES	Sr-90	—	—	9.5E-6	—
Field, ANL-E	7	Cement	Sr-90	—	—	3.7E-7	1.7E-4
Field, ANL-E	24	VES	Sr-90	—	—	1.5E-6	—
Field, ANL-E	24	Cement	Sr-90	—	—	7.0E-7	—
Field, ORNL	7	VES	Sr-90	—	—	1.4E-6	—
Field, ORNL	7	Cement	Sr-90	—	—	7.1E-5	—
Field, ORNL	24	VES	Sr90	—	—	4.7E-6	—
Field, ORNL	24	Cement	Sr-90	—	—	2.0E-5	1.2E-3
Field, ORNL	7	VES	Cs-137	—	—	1.5E-8	—
Field, ORNL	7	Cement	Cs-137	—	—	2.0E-8	—
Field, ORNL	24	VES	Cs-137	—	—	3.0E-9	—
Field, ORNL	24	Cement	Cs-137	—	—	1.0E-9	1.8E-5

a. Waste forms were irradiated before test.

the wall and has not yet been analyzed. The other core was collected from the center of the lysimeter directly above the buried waste form (located approximately 100 cm below the sand surface). The sand core was sectioned into 5-cm segments. Radiocesium and strontium activity were measured for each segment.

The analyses show that Cs-137 and Cs-134 are present throughout the length of the core (Table 10). Cesium-134 is an activation product that is formed in the core of nuclear reactors and does not occur naturally; therefore, the ratio of Cs-137 to Cs-134 in the sand segments can be used to determine if there was an outside source of cesium added to the lysimeter. By decay-correcting the original ratio of the waste form to the date of sand collection (and assuming that both radioisotopes behave chemically identical), the ratio should be equal to 399. The data presented in Table 10 and Figure 42 indicate that the ratio in all segments fall close to this value, except to segment 2 (70 to 75 cm in depth; the reason for this has not yet been determined). This strongly suggests that the cesium seen throughout the length of the sand core is a result of upward vertical transport from the waste form.

Table 10. Cesium and strontium analyses for sand core segments from ORNL lysimeter 5 collected on January 31, 1994.

Segment number	Depth (cm)	Sample weight (g)	Cs-137 (pCi/g)	Cs-134 (pCi/g)	Ratio[a] (137/134)	Sr-90 (pCi/g)	Ratio[b] (Cs-137/Sr-90)
1	80–75	100.68	598.1	1.5	399	1.5	450
1	80–75	17.08	704.5	1.573	448	—	—
1	80–75	17.16	660.8	1.556	425	—	—
2	75–70	118.92	1,303.4	1.8	724	3.5	650
2	75–70	17.48	2,241	2.313	969	—	—
2	75–70	20.37	1,550	1.758	882	—	—
3	70–65	121.53	356.7	0.737	484	2.0	200
3	70–65	19	400.7	0.805	498	—	—
3	70–65	14.85	376	0.727	517	—	—
4	65–60	115.25	490.2	1.097	447	2.1	300
5	60–55	117.07	403.3	0.819	492	2.7	200
6	55–50	125.28	1,594	3.246	491	7.6	300
7	50–45	129.06	37,283.1	89.51	417	14.1	1,000
7	50–45	17.79	14,160	30.4	466	—	—
8	45–40	121.14	551.2	1.363	404	1.5	400
9	40–35	116.32	866.6	2.306	378	3.5	300
10	35–30	122.86	5,484.2	22.58	243	7.6	750
10	35–30	17.5	5,172	10.88	475	—	—
11	30–25	117.94	2,032.4	4.438	458	16.0	200
12	25–20	125.78	1,513	3.574	423	0.5	3,400
13	20–15	94.99	711.7	1.823	390	0.2	4,400
14	15–8.5	150.3	715.2	1.585	451	0.6	1,200

a. Theoretical ratio of Cs-137/Cs-134 = 399.

b. Theoretical ratio of Cs-137/Sr-90 = 440.

There are three peaks seen in the cesium content (Figure 42): one at 30 to 35 cm, a large peak at 45 to 50 cm, and a smaller peak at 70 to 75 cm. These peaks may be indicative of some sort of periodic movement of the cesium, but further laboratory study is necessary before this can be determined.

During the sectioning of the core, it was noticed that there was a fine plant root present throughout the depth of the core. The root material was extracted from each segment and counted. The results are presented in Table 10 and Figure 43. Cesium-137 activity is associated with the roots, and the peaks in the root data occur at the same depths as do the peaks in the sand activity (Figure 44). It can be seen that there are higher concentrations of Cs-137 associated with the roots than with the sand. Sand from the deepest three segments was analyzed three separate times. The first time was the whole segment, and the other two times were subsamples of the sand. Segment 2 (Table 10) has a fairly wide range of activities between the whole segment and the two subsamples, suggesting that the activity in the sand is not evenly distributed. This could be a result of the root being involved in the transport process.

Strontium-90 analysis results show that there is significant strontium throughout the entire depth of the core (Table 11 and Figure 45). Peaks occur in the distribution at the same depths as for cesium in both the sand and roots. This suggests that the same mechanism may be involved for transporting strontium upward as for cesium. Strontium and cesium behave very differently chemically, but if the process of migration is more physical than chemical, then the ratio of Cs-137 to Sr-90 should be similar at all depths. Table 10 includes a tabulation of this ratio versus depth, and it can be seen that the ratios are similar for most of the segments, indicating that the upward transport is possibly related to a physical phenomenon such as evapotranspiration enhanced by the presence of the root. The fact that the sand has a very low cation-exchange capacity is probably the reason that the physical aspect of migration is so evident.

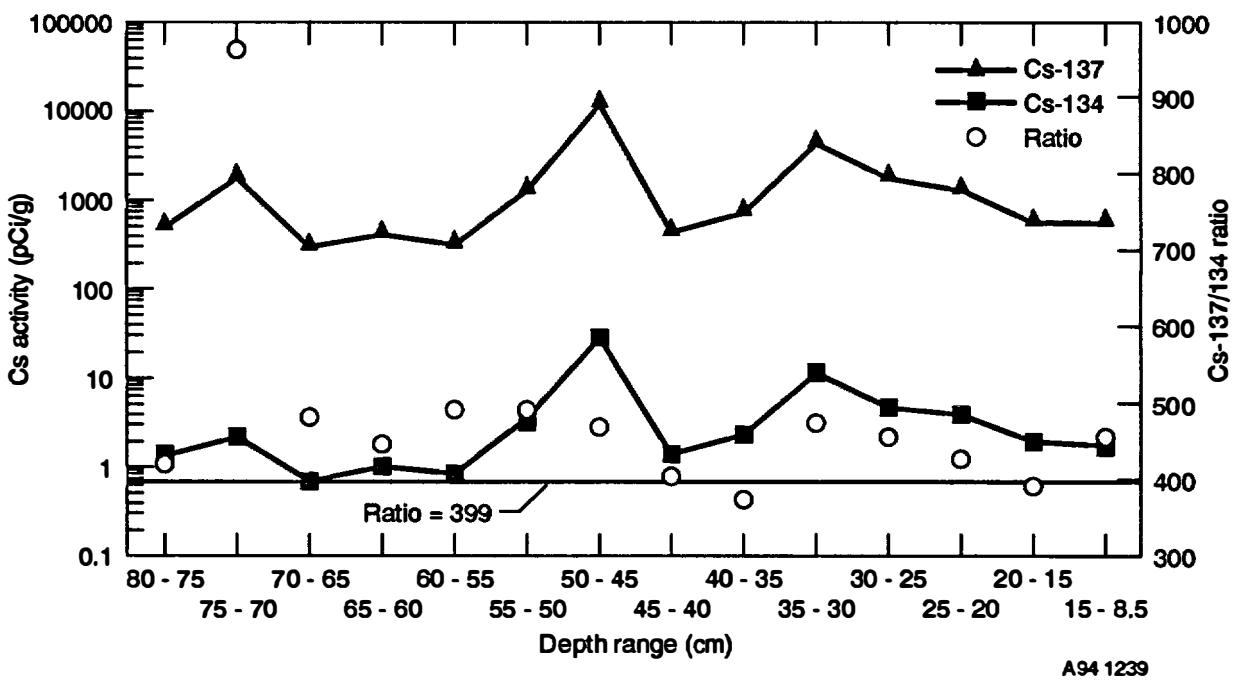

Figure 42. Cesium analyses of sand core from ORNL lysimeter 5.

Table 11. Cesium-137 analyses on root fragments from segments of sand from ORNL lysimeter 5.

Segment number	Depth (cm)	Weight (g)	Cs-137 (pCi/g)
1	80–75	0.0134	18,990
2	75–70	0.0172	20,660
3	70–65	0.0301	20,480
4	65–60	0.0234	22,540
5	60–55	0.0216	27,520
6	55–50	0.0224	27,360
7	50–45	0.022	81,970
8	45–40	0.0302	13,620
9	40–35	0.0196	10,150
10	35–30	0.0463	21,580
11	30–25	0.0256	5,990
12	25–20	0.1049	3,850
13	20–15	0.0615	5,940
14	15–8.5	0.3105	8,570

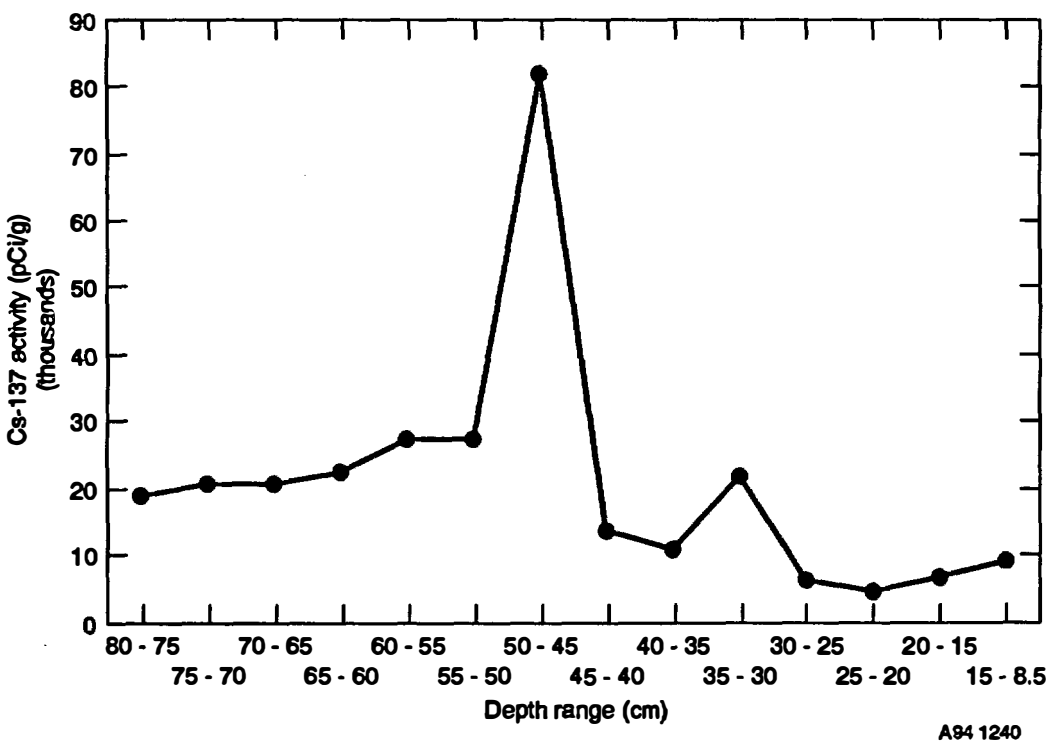

Figure 43. Cesium-137 associated with plant roots from ORNL lysimeter 5.

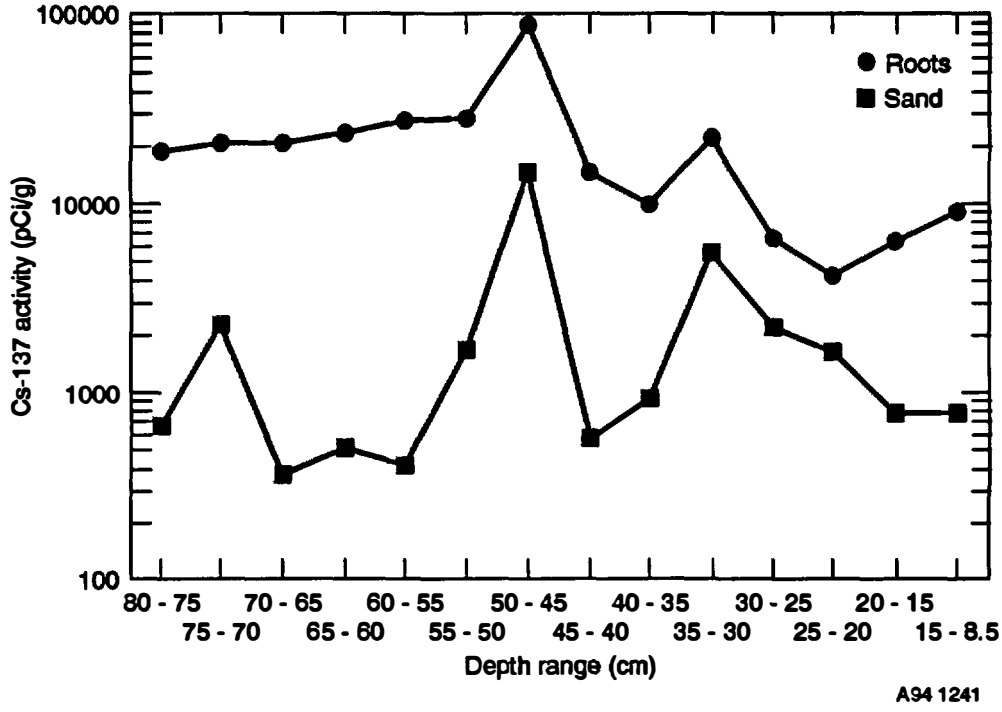

Figure 44. Comparison of Cs-137 associated with plant roots and sand from ORNL lysimeter 5.

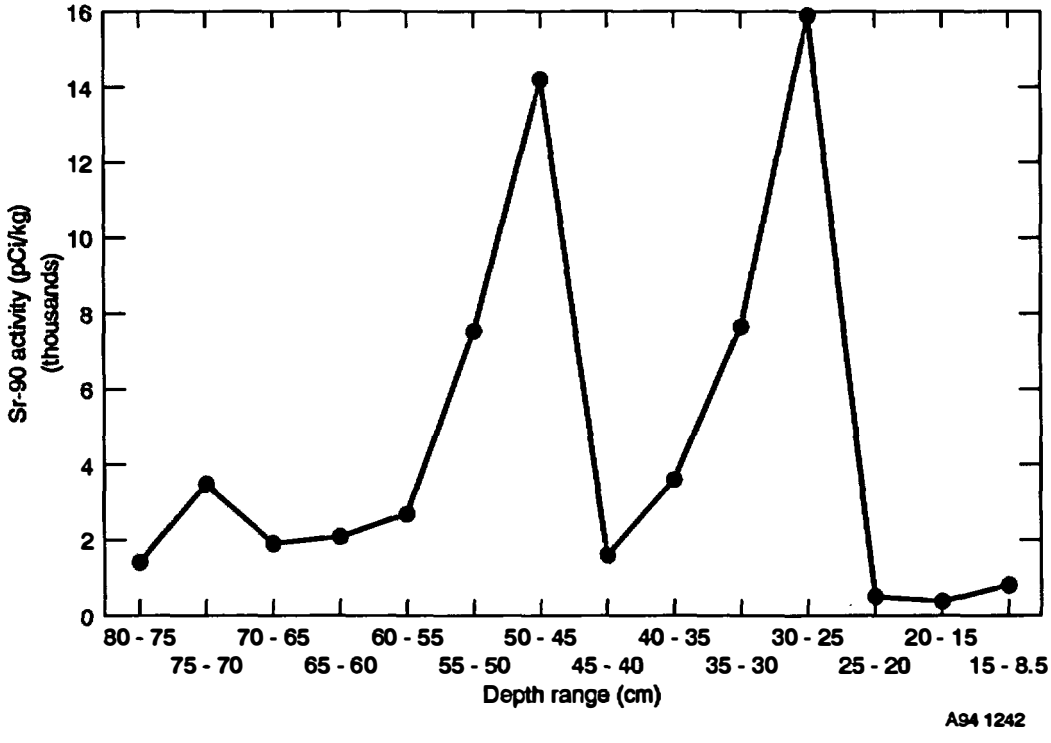

Figure 45. Strontium-90 analyses of sand core from ORNL lysimeter 5.

One further check on the possibility of an outside source of radionuclides can be made. If the original Cs-137/Sr-90 ratio of the waste form is decay-corrected to the sampling date, the value should be 440. A mass balance on the total cesium and strontium in the core was made by determining the average concentration of each isotope, ignoring the upper three segments because of the large errors associated with the strontium analysis (Table 10). The average Cs-137 concentration is 2,590 pCi/g, and the average strontium-90 concentration is 5.6 pCi/g, giving a ratio of 460. The similarity of the measured ratio to the theoretical ratio is further evidence that there has been no strontium or cesium added from an outside source. This ratio is not similar to the downward movement of Cs-137 and Sr-90 as described in the Radionuclide Data section of this report. There it is shown that Sr-90 makes up a majority of the radionuclides detected in the leachate for as yet unexplained reasons.

It is important to find out how cesium migrated more than 1 m upward from the waste form to the surface of the sand. 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 confirms that assumption (Table 7 and Figure 41). 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. 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.

A third theory involves lysimeter flooding. There have been several occurrences in which the lysimeters have accumulated water that moved high into the soil column, above the waste forms. This happened as a result of unusually heavy precipitation events. It is possible that the high water level caused an upward movement of radionuclides, which then became fixed to organic matter, which prevented reentry. Further analysis of soil cores is planned.

Scientists at ORNL performed gamma radiation surveys on soil samples that were collected from lysimeters 1 through 4 for gravemetric moisture analyses in 1993 and 1994. Measurable but insignificant amounts (below 1 pCi/g) of gamma-producing radionuclides were detected. ORNL scientists determined that soil-filled lysimeters are not experiencing upward migration of radionuclides.

Use of Lysimeter Data for Performance Assessment Radionuclide Release Calculations. It is becoming apparent, through operational experience and cumulative data provided by the NRC lysimeter array during the past 9 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 12. The data could be used in such codes as PATHRAE,[22] PRESTO,[23] 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-94 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 inert 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. PF-7 waste was a mixture of synthetic organic ion-exchange resins (phenolic cation, strong acid cation, and strong base anion). PF-24 waste 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 6) along with a leachate container located at the bottom of the lysimeter (Reference 12). 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 developed by Brookhaven National Laboratory (BNL) was used by BNL for the third year 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

Table 12. 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_v	=	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_v
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

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 References 24 and 25.

The sand-filled lysimeters 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 soil-filled lysimeters were substantially lower and the data were not sufficient to model. At ANL-E, lysimeter 5 contained PF-7 resin waste solidified in cement; at ORNL, lysimeter 5 contained PF-24 resin waste, 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 determined in bench leach testing and are presented in Reference 10. 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 11). The soil bulk density values were 1.55 g/cm^3 at ANL-E and 1.60 g/cm^3 at ORNL (Reference 12). Moisture content values were calculated using the effective soil porosity and the fraction of saturation values found in Reference 19. In lysimeter 5 at both sites, the moisture content was calculated as 21%. The dispersivity and retardation coefficient have not been measured for Sr-90 or Cs-137; therefore, they were estimated based on data in References 26 and 27 and by fitting the model predictions to the data. The cumulative leachate activity collected from the lysimeters over the first 9 years of the experiment, which was used to make comparisons to the DUST code predictions, represented 0.12% and 0.017% of the total inventory of Sr-90 in lysimeters 5, at ORNL and ANL-E, respectively. At ORNL, the collected amount represented less than 0.0018% of the Cs-137 inventory in lysimeter 5, while nothing has been collected in ANL-E lysimeter 5 (Table 13).

The cumulative activity collected from the lysimeters is 0.12% of total inventory for Sr-90 and less than 0.002% for Cs-137 (Table 12). Therefore, either the waste form release rates are much lower than anticipated, or transport processes are controlling release through the soil column. At the Cs-137 level, it is possible that random fluctuations are being seen, and release patterns may not develop for several more years.

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 PF-7 waste form had a total initial inventory of 3.29E-3 Ci of Sr-90 and 1.432 Ci of Cs-137

Table 13. Total and collected Ci amounts of Sr-90 and Cs-137 in lysimeter 5 through July 1994.

	Total inventory (Ci)	Amount collected (Ci)	Percent collected
ORNL-5 Cs-137	1.432	25.8E-6	1.8E-3
ORNL-5 Sr-90	0.0034	4.1E-6	1.2E-1
ANL-5 Sr-90	0.0184	3.1E-6	1.7E-2

(Reference 12). The PF-24 waste form at ANL-E had a total initial inventory of 1.84E-2 Ci of Sr-90 (Reference 12). Cesium-137 was not modeled at ANL-E for lack of sufficient releases.

Three parameters are known to strongly influence release through the soil column. They are retardation (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 have been modeled in which either K_d or dispersivity were varied to best match the actual release data from the lysimeters.

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 46 and 47 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.

As shown in Figure 46, the actual data for Sr-90 from ORNL lysimeter 5 for 9 years are compared with the DUST code predicted releases in case 1 using zero dispersive flux BC, $K_d = 24$, and dispersivity = 8.5 cm. Also shown are predicted releases of case 2 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 initial stabilization of the test data. Case 2 releases less activity over 4 years than case 1; however, 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. This reflects the higher K_d value of case 1, which reduces the travel time through the soil, thereby causing higher releases. Also case 1 is a better fit to the data through 9 years. The lack of measured dispersivity coefficient and K_d further necessitates obtaining fractional release data over a longer term.

Figure 47 shows the actual data for Sr-90 at ANL-E lysimeter 5, which covers a period of 9 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 K_d of 24.5 and a dispersivity of 8.5 cm. Case 2 has a K_d of 10 and a dispersivity of 0.6 cm. Case 2 releases less activity over about 7 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 9 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. The effort to analyze water samples obtained from moisture cups for some major cation and anion species was initiated at ORNL in 1988 and at ANL-E in 1991. It is anticipated that such data will prove useful as an 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 sampling period in 1994 was analyzed for the major ionic species listed in Table 14. The justification for the choice of ions is

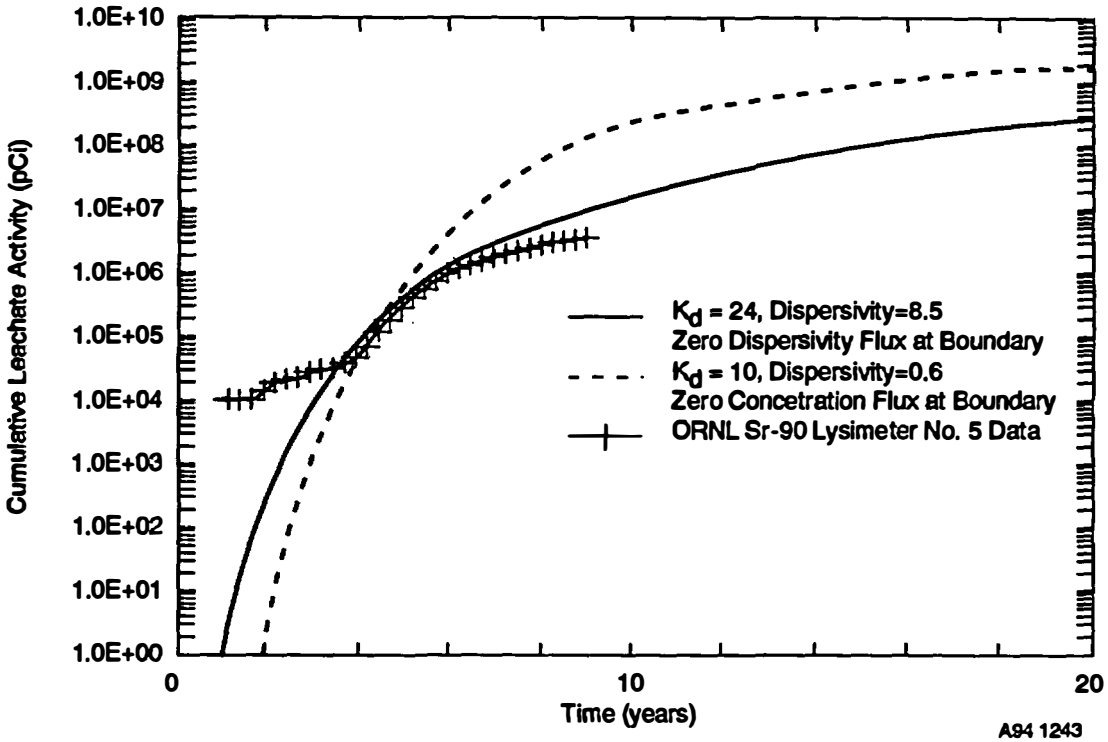

Figure 46. Nine years of data for Sr-90 at ORNL lysimeter 5, compared with two sets of estimated K_d and dispersivity values.

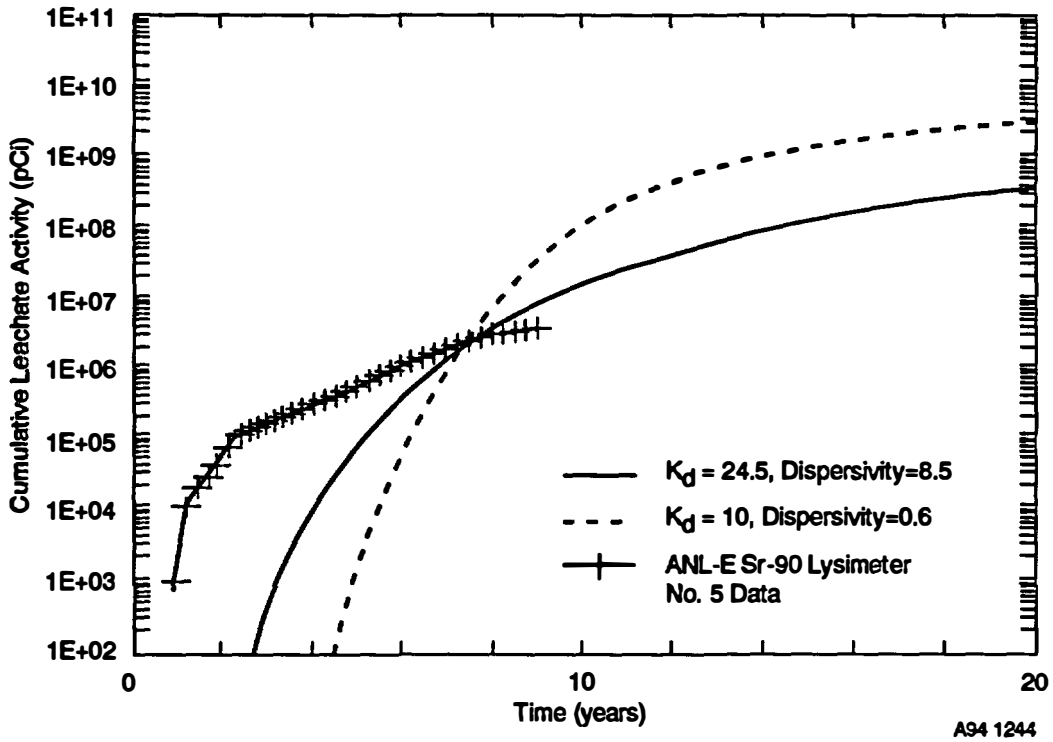

Figure 47. Nine years of data for Sr-90 at ANL-E lysimeter 5, compared with two sets of estimated K_d and dispersivity values for 20 years.

Table 14. 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.

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 and 3 water samples were sampled in 1994 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 and 15). 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 15 and 16 and Figures 48, 49, 50, and 51).

It appears that the cement and VES waste-form specimens performed similarly at both sites. With the exception of elevated potassium, the ORNL 1994 soil lysimeter data (Table 16 and Figures 50 and 51) closely resemble those of 1988, 1989, 1991, 1992, and 1993 cation and anion concentrations, and actually show little cup-to-cup variability. ANL-E 1994 data are similar, in most cases, to previous year data and to ORNL 1994 data when compared in Figures 48, 49, 50, and 51. The inert sand-filled lysimeter results are almost identical except for higher NO_3 concentration at ORNL. While these data are interesting, no correlation has yet been made with radionuclide movement.

Table 15. ANL-E chemical speciation results from lysimeter moisture cups 1, 2, 3, 4, and 5, April 1994.

		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	100	9.5	13	1.8	54	2.4	0.6	0.7	37
Lys 1-3		24	3.0	3.2	0.6	13	8.5	7.3	<0.5	57
Lys 1-5		37	1.6	6.4	0.4	16	1.9	<0.1	<0.5	28
Lys 2-2	Cement	85	6.7	11	0.3	46	4.3	<0.1	<0.5	23
Lys 2-3		145	13.0	14	0.6	58	3.6	<0.1	<0.5	36
Lys 2-4		115	7.0	14	0.3	61	11	<0.1	<0.5	56
Lys 3-1	VES	119	4.8	15	0.9	45	1.6	1.1	<0.5	21
Lys 3-3		86	6.1	14	0.3	47	6.6	<0.1	<0.5	22
Lys 3-5		93	2.6	15	0.2	45	1.5	0.4	<0.5	24
Lys 4-1	VES	NA	NA	NA	NA	NA	13	<0.1	<0.5	36
Lys 4-3		88	5.4	10	0.2	43	3.5	<0.1	<0.5	34
Lys 4-5		67	2.4	11	0.5	27	2.4	<0.1	<0.5	17
Lys 5-1	Cement	9.4	2.5	8.4	0.6	3.9	1.6	1.5	<0.5	4.6
Lys 5-3		9.8	8.6	25	4.6	4.4	1.7	1.4	<0.5	5.6
Lys 5-5		8.3	1.1	108	1.4	3.3	1.6	2.3	<0.5	6.0

NA = no sample.

Table 16. ORNL chemical speciation results from lysimeter moisture cups 1, 3, and 5, June 1994.

		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	37	3	18	<2	1.7	2.9	58	<0.5	18
Lys 1-3		33	2.2	23	2.1	1.4	1.3	4.7	<0.5	14
Lys 1-5[a]		—	—	—	—	—	—	—	—	—
Lys 2-1	Cement	38	1.8	18	<2	1.4	2.9	4.5	<0.5	10
Lys 2-3		30	2.1	28	<2	1.0	1.4	3.7	<0.5	8.2
Lys 2-5[a]		—	—	—	—	—	—	—	—	—
Lys 3-1	VES	30	1.9	19	<2	0.9	2.5	12	<0.5	6.1
Lys 3-3		6.1	4.6	11	<2	1.1	2.8	5.9	<0.5	6.7
Lys 3-5[a]		—	—	—	—	—	—	—	—	—
Lys 4-1	VES	6.1	3.6	7.3	3.7	0.9	2.2	3.5	<0.5	22
Lys 4-3		12	1.3	19	<2	5.4	2.6	3.5	<.0.5	19
Lys 4-5[a]		—	—	—	—	—	—	—	—	—
Lys 5-1	Cement	1.2	0.2	12	<2	5.1	4.9	<0.1	<0.5	4.7
Lys 5-3		2.4	0.3	3.8	<2	1.1	5.5	5.9	<0.5	6.2
Lys 5-5[a]		—	—	—	—	—	—	—	—	—

a. No samples were taken from cups number 5 in this reporting period.

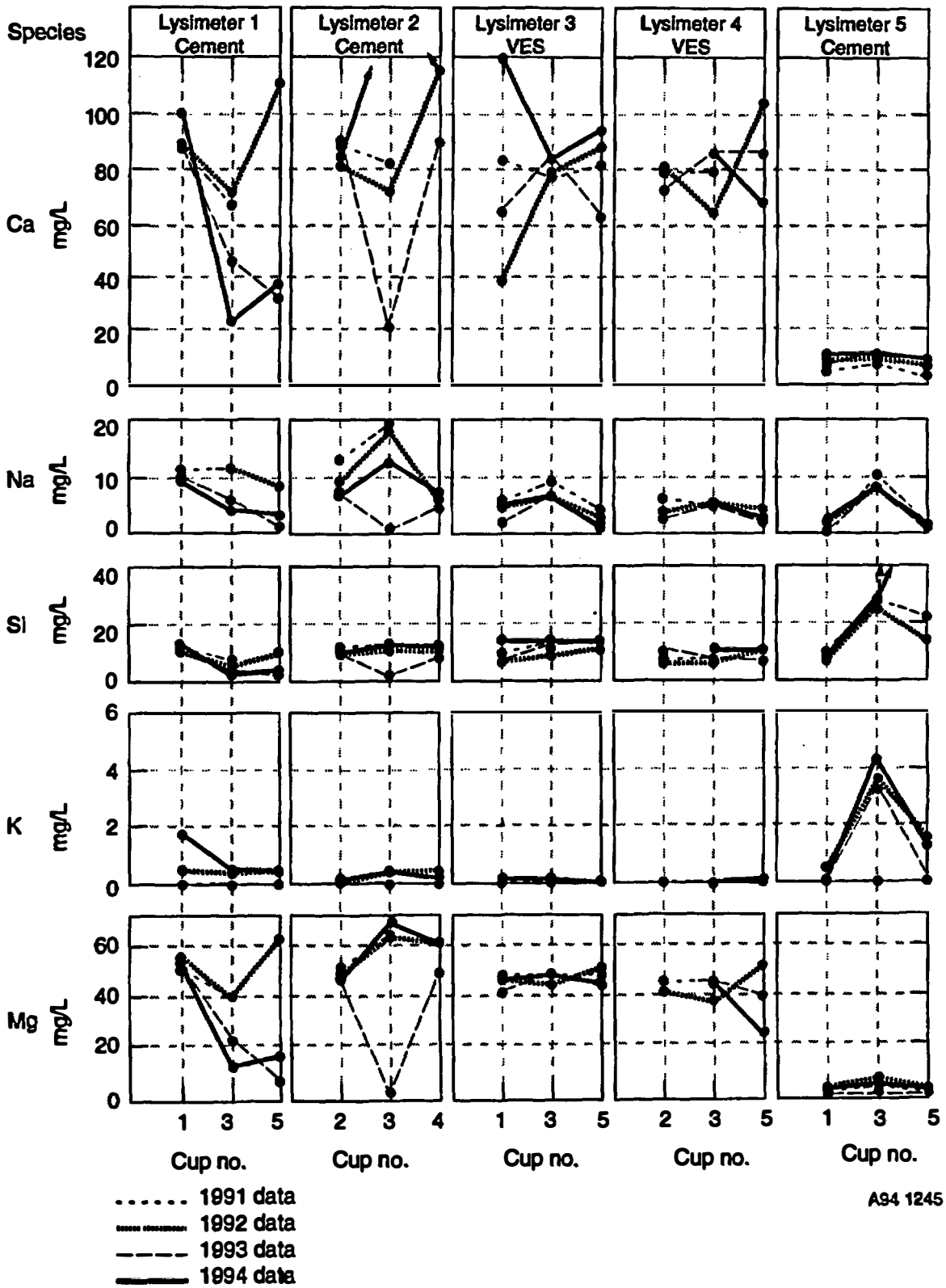

Figure 48. Results of chemical speciation at ANL-E cations.

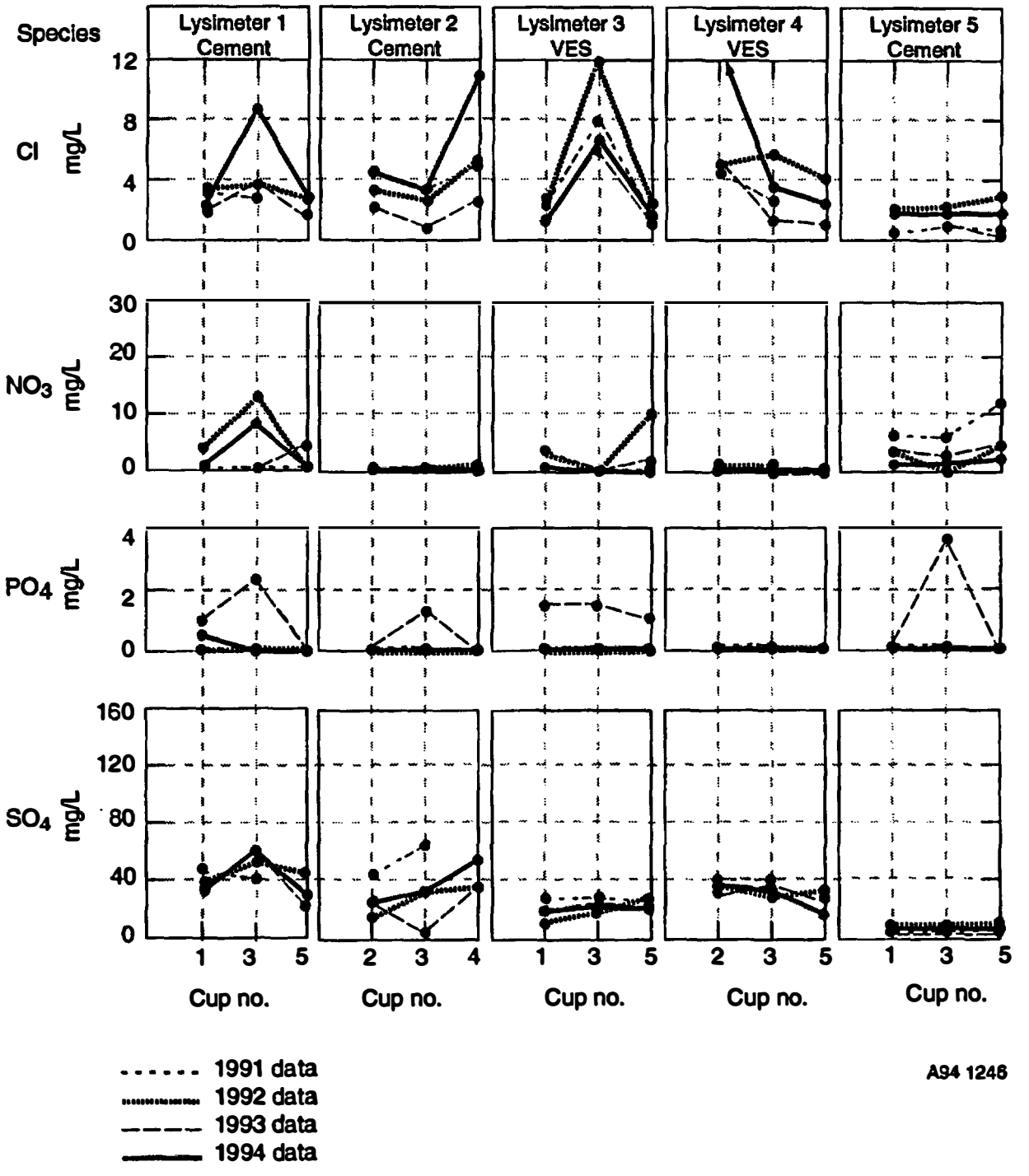

Figure 49. Results of chemical speciation at ANL-E anions.

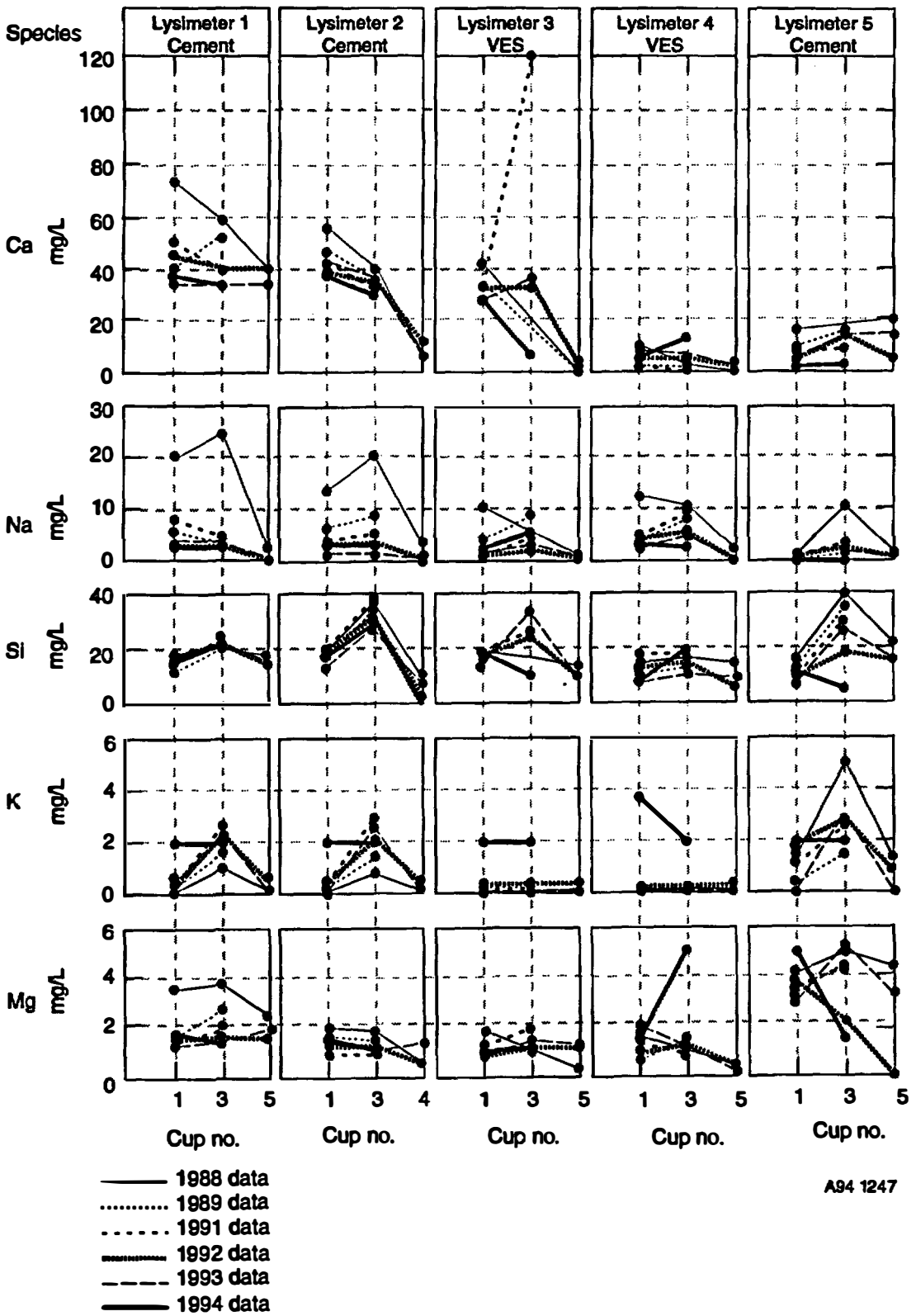

Figure 50. Results of chemical speciation at ORNL cations.

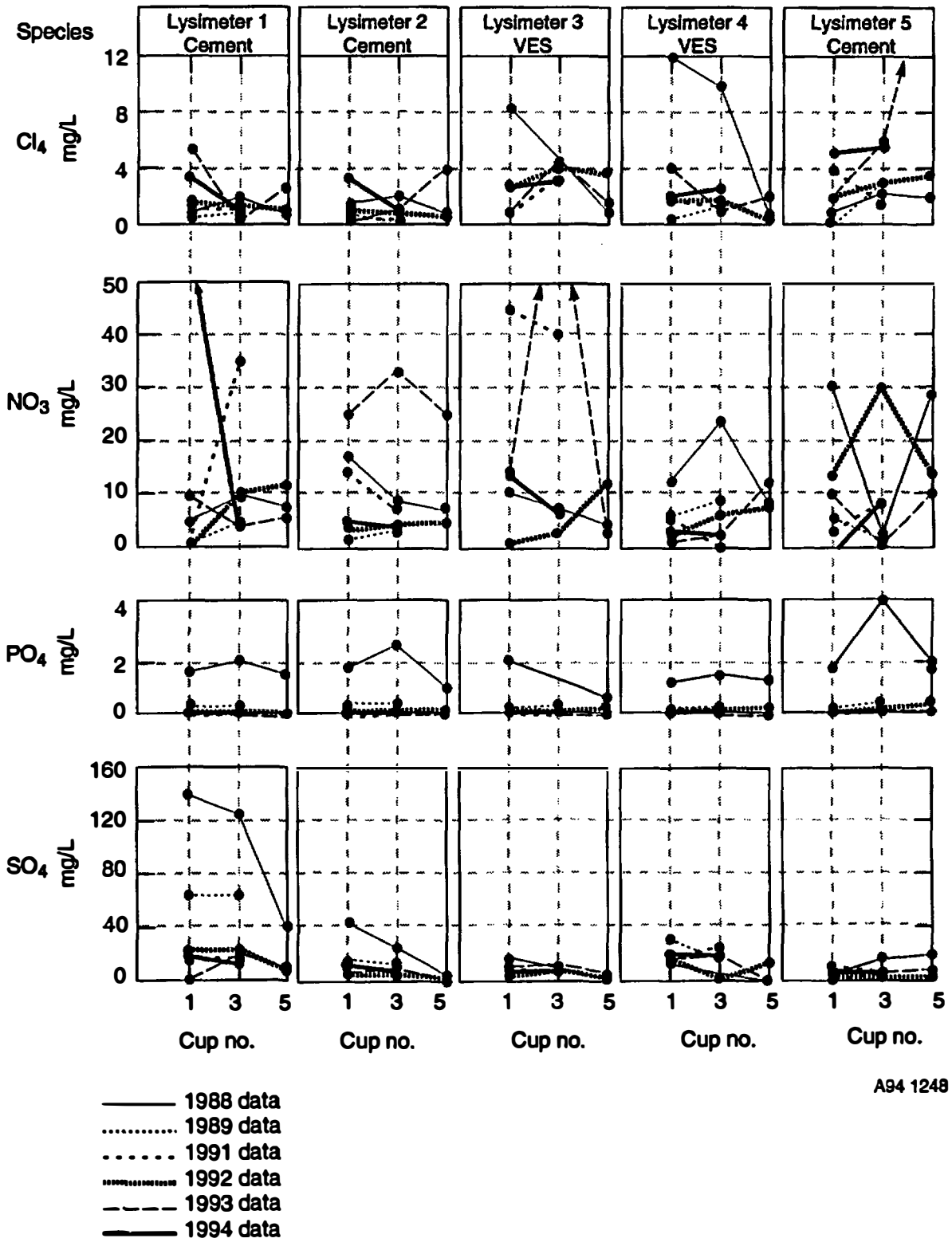

Figure 51. Results of chemical speciation at ORNL anions.

CONCLUSIONS

Resin Solidification

The 11-year-old waste forms fabricated with EPICOR-II ion-exchange resins exhibited the same or a slight decrease in ultimate compressive strength with age when compared to the strength of waste forms that are 8 years old. It appears that strength has been affected by accumulated radiation dose after a steady increase with age to 7 years. One Portland cement specimen failed far below the expected strength after 8 years and has shown a comparable low strength at 11 years. It would be anticipated that the cement specimens would retain strength to a higher radiation dose than VES because low-level radiation does not adversely affect cement strength. Therefore, the low strength exhibited by the one cement type may be caused by radiation effects on the contained ion-exchange resin, which serves as aggregate to the cement. It is noted that this same cement waste form type exhibited an abnormally high strength after 7 years. The compression tests planned over the next several years should confirm the observed trends.

Field Testing

The lysimeter experiment during the 9 years of operation has been successful. Analyses of data collected during the past 108 months continue to show a pattern in nuclide availability and movement such that the cumulative results are beginning to provide a better insight to 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 60 months) and Co-60. Compared to Sr-90, the occurrence of Cs-137, Sb-125, and Co-60 appear insignificant.

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 soil-filled 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 soil-filled lysimeters containing VES waste forms are an order of magnitude larger than in those containing cement waste forms.

Cesium-137 has been found in leachate collector water from the sand-filled control lysimeter at ORNL for the third year but was not detected in the leachate collector water of any lysimeters at ANL-E this year.

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 from cement and VES are comparable but dependent on environmental influences. Releases of Cs-137 has been in such small quantities that a diffinitive statement on comparison of release cannot be made. These data still differ from those obtained at SRL. Those data show that cement minimizes the release of Sr-90.[28] This interesting difference should be studied further. The data reported by SRL[28] and PNL[29] agree that Cs-137 is more readily released from cement than from VES, while PNL[30] has observed Cs-137 release in trace amounts only from masonry cement waste forms, and none from Portland cement and VES waste forms.

On a cumulative basis, a larger amount of Sr-90 is being removed in leachate water from the ORNL soil lysimeters versus those at ANL-E. This is thought to be a result of the difference in soils as well as in environmental conditions between the two sites. 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, but rather, movement that is limited by environmental characteristics (including soil and quantity of soil water). This conclusion is supported by data from lysimeter work at Savannah River Laboratory (SRL) and Pacific Northwest Laboratory

(PNL).[28,29,30] SRL has found that Sr-90 will move from buried waste forms, readily migrate through the soil column, and appear in collected leachate water.[28] It is not surprising, then, that Sr-90 moves through soil in the ORNL lysimeters, since that soil originated at SRL.[12] On the other hand, lysimeter work with waste forms at PNL has shown that Sr-90 does not move in those soils.[29,30]

During the past 84 months, Sr-90 continues to be found in similar concentrations in leachate water from the sand-filled control lysimeters at both sites, with a slightly more rapid accumulation at ORNL, which now has had seven times more of the available source of Sr-90 released than the control lysimeter at ANL-E.

A comparison of cumulative fractional releases from field testing of EPICOR-II waste forms in lysimeters to releases from bench-leach-testing similar waste forms shows that lysimeter releases are at least four orders of magnitude less for Sr-90 in soil and at least five orders of magnitude less for Cs-137 in soil. Releases of Sr-90 in sand-filled lysimeters are only one or two orders of magnitude less than bench test results.

Cesium-137, Cs-134, and Sr-90 are present throughout the upper 80 cm of the inert sand in ORNL lysimeter 5 directly above the waste form. The ratio of Cs-137/Cs-134 indicates that the radionuclides are from the buried waste form and not from an outside source and were transported vertically upward by some physical mechanism enhanced by the presence of a plant root.

The results from a preliminary evaluation that was carried out in FY-91 indicated that in lysimeters with experimentally determined diffusion coefficients, a computer code could be tested for performance assessment modeling. This held true where there were high enough leachate concentrations of nuclides for comparison between predicted and experimental results. In the last 3 years, refinements made it possible to successfully model some of the lysimeter Sr-90 releases using the DUST computer code. Once again, as was the case last year, it was strongly recommended by BNL 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.

DUST-predicted cumulative release of Sr-90 from ORNL lysimeter 5, which was plotted over the 9-year data collection period, show a reasonable fit to the field data. The accuracy of the DUST modeling study was limited, however, by the lack of soil and waste form radionuclide release data. The sensitivity of predicted releases to the model parameters (soil dispersivity and retardation coefficients and waste form diffusion coefficient) make it essential that site-specific soil data be collected for those parameters. Releases of Sr-90 to cups 2 will help define dispersivity as it becomes available. In addition, longer-term tests are needed to provide larger cumulative releases to better model the release patterns from the lysimeters.

The Phase 2 lysimeter field test preliminary experimental equipment design, which will use commercial low-level waste, was completed.

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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.

Compressive test results of 11-year-old cement and vynyl ester-styrene solidified waste forms are presented, which show effects of aging and self-irradiation.

Results of the ninth 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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