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Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1995

Annual Report

Prepared by
J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, I. L. Larsen, T. M. Sullivan

Idaho National Engineering Laboratory
Lockheed Idaho Technologies Company

Prepared for
U.S. Nuclear Regulatory Commission

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Prepared by
J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow,[1] W. E. Sanford,[2] I. L. Larsen, T. M. Sullivan[3]

Idaho National Engineering Laboratory
Lockheed Idaho Technologies Company
Idaho Falls, ID 83415

P. Reed, NRC Project Manager

Prepared for
Division of Regulatory Applications
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001
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[1]Argonne National Laboratory, Argonne IL
[2]Oak Ridge National Laboratory, Oak Ridge, TN
[3]Brookhaven National Laboratory, Upton, NY

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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 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 ion-exchange resins, (c) obtaining performance information on solidified ion-exchange resins in a disposal environment, and (d) determining the condition of liners used to dispose the ion-exchange resins.

Compressive test results of 12-year-old cement and vinyl ester-styrene solidified waste form samples are presented, which show effects of aging and self-irradiation.

Results of the tenth 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 form samples 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.

Job Code A6876—Field Lysimeter Investigations:
Low-Level Waste Data Base Development Program

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EXECUTIVE SUMMARY

Research is being conducted at the Idaho National Engineering Laboratory on materials from four liners containing organic and inorganic ion-exchange resin 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 are periodically subjected to the tests specified in the "Technical Position on Waste Form" issued by the U.S. Nuclear Regulatory Commission. Waste form sample 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. 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 10 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 form samples 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 form samples. 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
CFR	cumulative fractional release
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 1995 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 form samples 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 sample test methods and

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.

acceptable results for compliance with the waste form sample requirements of 10 CFR 61. In this study, EPICOR-II waste form samples 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 cement waste form sample strength has stabilized after increasing with age for 8 years. In the case of VES, the waste form sample strength has begun to decrease.

Solidified waste form samples 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 form samples in typical low-level waste disposal environments, (b) compare field results with short-term laboratory leach studies, (c) compare field results with DOE Special Waste Program field test results, (d) develop a low-level radioactive waste field leach-rate data base for use in performance assessment source term calculations, and (e) use the DUST source term computer code to compare predicted cumulative radionuclide 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 form samples 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 10 years of lysimeter operation, as well as cumulative data on water balance and chemical species and nuclide content of water samples. Data for this report were retrieved from the data acquisition system (DAS), from chemical speciation of water samples, and from beta and gamma analyses of lysimeter water samples.

RESIN SOLIDIFICATION

In this study, EPICOR-II waste form samples solidified with portland type I–II cement and VES were periodically subjected to compression testing per ASTM C39 over a 12-year time span.[7] The samples were tested dry using sulfur leveling caps poured per ASTM C617.[8] Normally, one sample 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. This fiscal year, two samples of each were tested.

On August 9, 1995, eight 12-year-old waste-form samples were compression-tested using a Tinuis Olsen 60,000-lb testing machine calibrated on July 18, 1995. 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 samples tested in FY-95 are shown in Figures 1 through 8. The concrete samples broke in a vertical cracking pattern with flaking. The VES samples bulged in the center of the cylinder as had most previous VES samples. Vertical cracking took place within the bulging center area. The VES samples seemed to be very ductile. Examination of the VES samples containing organic/inorganic resin indicates that failures were more like brittle fractures. Large vertical cracks are evident in these samples. Similar failures were experienced with the irradiated VES waste form samples tested earlier in this program (Reference 10).

Figure 9 shows the strength data from these and previous compressive strength tests[10,11] of sample age (taken at ages after fabrication) of 1 month, 2 years, 7 years, 8 years, 11 years, and 12 years. The cement sample type containing organic resins experienced an increase in strength during the last year. The strength of the cement samples containing organic/inorganic resins stabilized in the twelfth year after exhibiting very high and low strengths in past years. The VES samples exhibited a drop in strength between 8 and 12 years, which was most pronounced in the last test. It appears that strength was affected by accumulated radiation dose after a steady increase of strength with age to 8 years for all four sample types.

Table 1. Compression test result of 12-year-old EPICOR-II waste-form samples.

Specimen	Material of construction	Measured β and γ radiation at contract (R/h)	Area (in^2)	Ultimate load (lb)	Compressive strength (psi)
C1-31	Organic ion-exchange resin in	2	2.68	16,950	6,335
C1-35	portland cement	9	2.68	12,650	4,720
				Average	5,525
C2A-10	Organic/inorganic ion-exchange	12	2.66	14,250	5,360
C2A-29	resin in portland cement	12	2.63	15,500	5,895
				Average	5,620
D1-13	Organic ion-exchange resin in	9	2.60	8,550	3,290
D1-14	VES	7	2.51	9,875	3,840
				Average	3,565
D2-1	Organic/inorganic ion-exchange	30	2.55	9,250	3,630
D2-12	resin in VES	30	2.54	8,925	3,520
				Average	3,575

One portland cement sample, C2A-6, failed far below the expected strength at 8 years, and C2A-14 showed a comparable low strength at 11 years. However, the average strength of C2A-10 and 29 in FY-95 was at the expected higher value. It would be anticipated that the cement samples 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 samples containing organic/inorganic resin may have been caused by radiation effects on the contained organic ion-exchange resin, which serves as aggregate to the cement; however, that phenomenon would have been more likely to adversely effect the all-organic resin. That was not the case. 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 form samples that were uniform in weight and radiation dose, while the VES batches contained some less uniform waste form samples. However, many of the cement waste form samples exhibited surface blemishes in the form of air bubbles. It is suspected that air bubbles were also contained within the interior of those cement samples. Such discontinuities would detrimentally affect the strength of these brittle waste form samples.

The cement compressive strength demonstrates the impact of data scatter resulting from employing only one waste form of each type in earlier test years. In order to economize the few remaining waste forms, only four were normally 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. Due to the high radiation dose, 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, while the results at 12 years (as-cast) are the average of two data points. These results all appear to be very consistent (Table 1).

A review of irradiated waste form compressive test results (Reference 10) shows that cement sample 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 samples hold toward the upper limit while the organic/inorganic C2 samples fall near the lower limit. It is noted that the strength of sample C2A-7 tested in the seventh year at 6,404 psi was well above that standard deviation as were the average strengths of both cement types in the 12-year test results.

The VES compressive strength data of Figure 9 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 sample, D1A-1. The D1 specimens tested at 12 years showed a significant decrease in average strength from the previous year also (17%). The end of increase in strength was thought to be the result of total radiation dose on these waste forms. The 11-year organic/inorganic-resin-containing waste form sample, D2-34, showed a 20% drop in strength from the 8-year test sample, 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. That same effect was observed in the average strength the D2 samples exhibited during the testing at 12 years, where a drop of 12% was seen.

These waste form samples have been stored in shielded drums throughout the project. Waste form contact dose measurements indicate that the

samples have experienced a total beta and gamma dose of 0.6×10^6 to 3.3×10^6 rad. That dose has caused serious degradation of the polyethylene sample containers, which are discolored, embrittled, and cracked. In order to preserve sample identity, a number of those containers were replaced with cardboard tubes during the FY-94 test operation.

The doses applied to the irradiated waste forms of Reference 10 were two orders of magnitude higher than the above-estimated doses after 12 years. The resulting average compressive strength of the four externally irradiated waste form types is shown in Figure 9 and represents the projected 300-year end-of-life strength of these materials. The actual strength may be somewhat higher in that the test irradiations were accomplished in less than 30 days and could have caused accelerated degradation of material due to the higher dose rate. Those irradiations were also somewhat higher in total dose than would be expected at end-of-life.

Figure 1. Sample C1-31 organic ion-exchange resin in portland cement after compression test, front view.

Figure 2. Sample C1-35 organic ion-exchange resin in portland cement after compression test, front view.

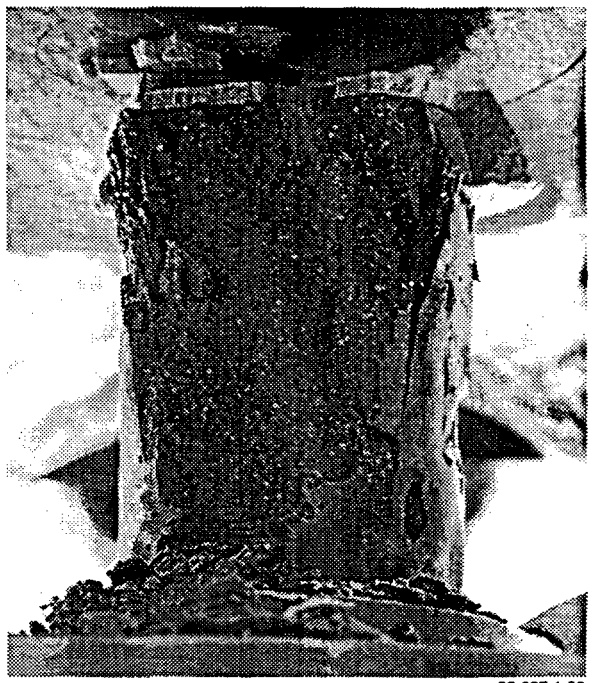

Figure 3. Sample C2A-10 organic/inorganic ion-exchange resin in portland cement after compression test, side view.

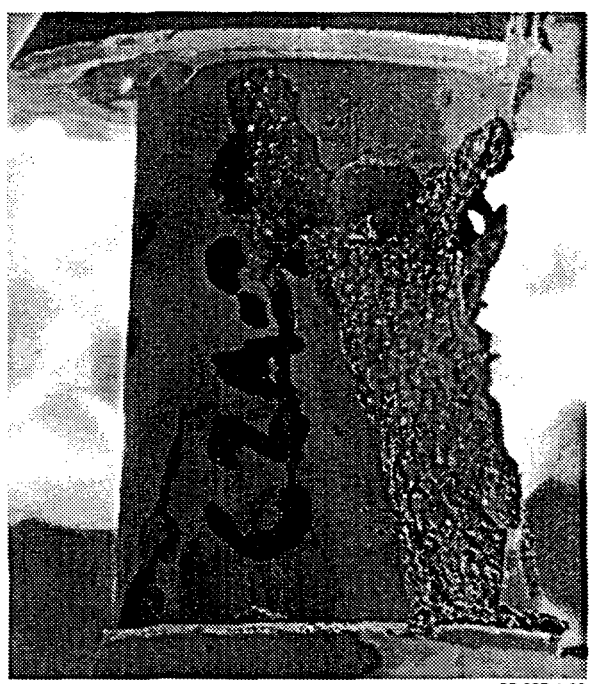

Figure 4. Sample C2A-29 organic/inorganic ion-exchange resin in portland cement after compression test, front view.

Figure 5. Sample D1-13 organic ion-exchange resin in VES after compression test, front view.

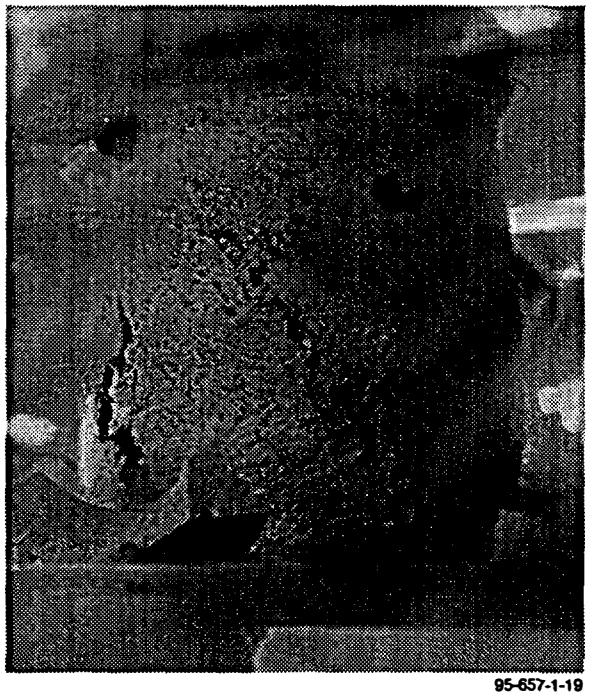

Figure 6. Sample D1-14 organic ion-exchange resin in VES after compression test, side view.

Figure 7. Sample D2-1 organic/inorganic ion-exchange resin in VES after compression test, side view.

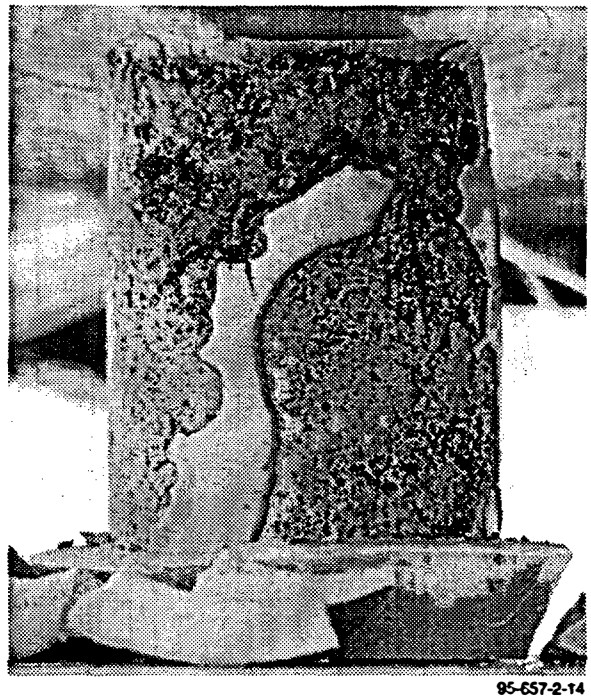

Figure 8. Sample D2-12 organic/inorganic ion-exchange resin in VES after compression test, side view.

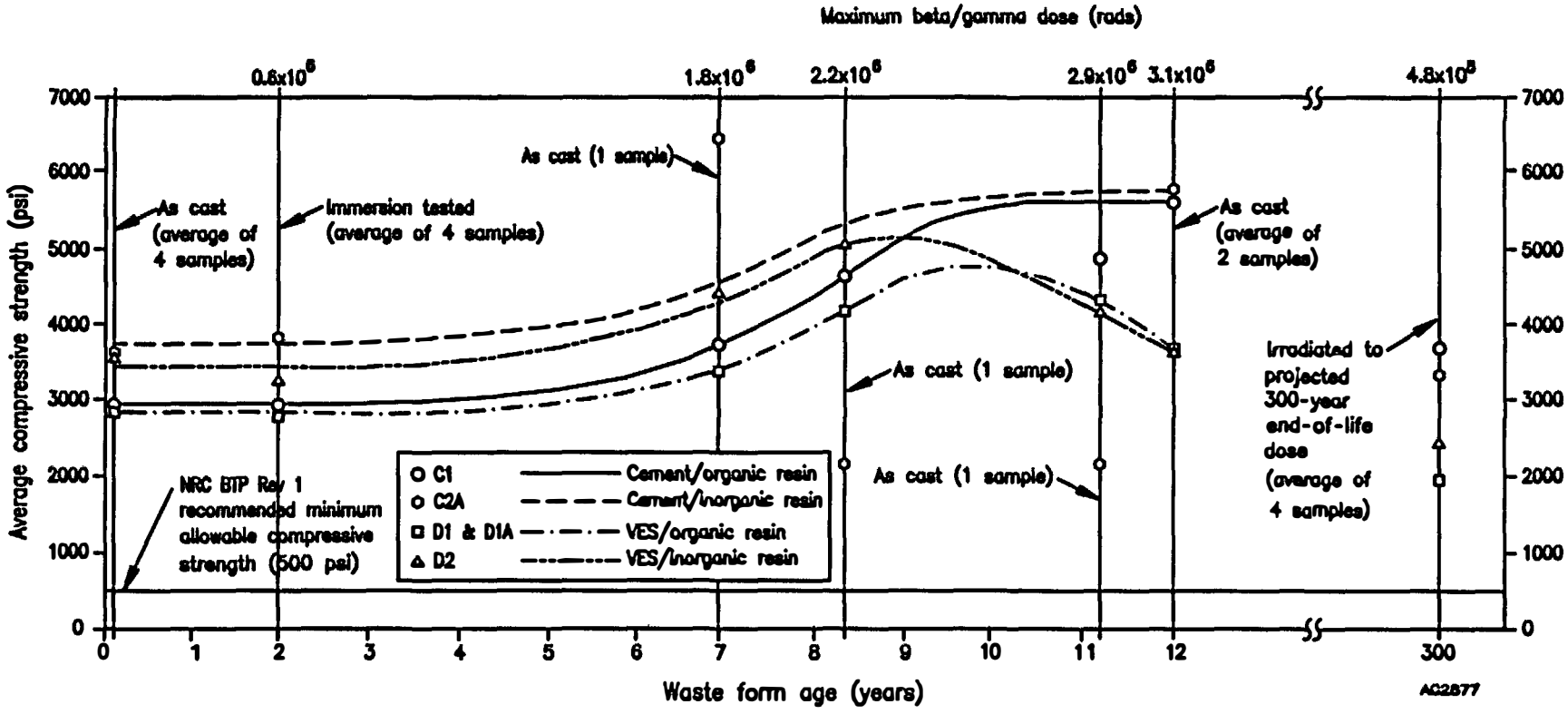

Figure 9. Variation of waste form compressive strength with age caused by self-irradiation and curing.

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 9 years of operation are contained in earlier reports.[11,13–20]

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 samples 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 samples 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 samples stacked end-to-end to form a 1-L waste volume. Table 2 shows the types of samples 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 10). 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 10. 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

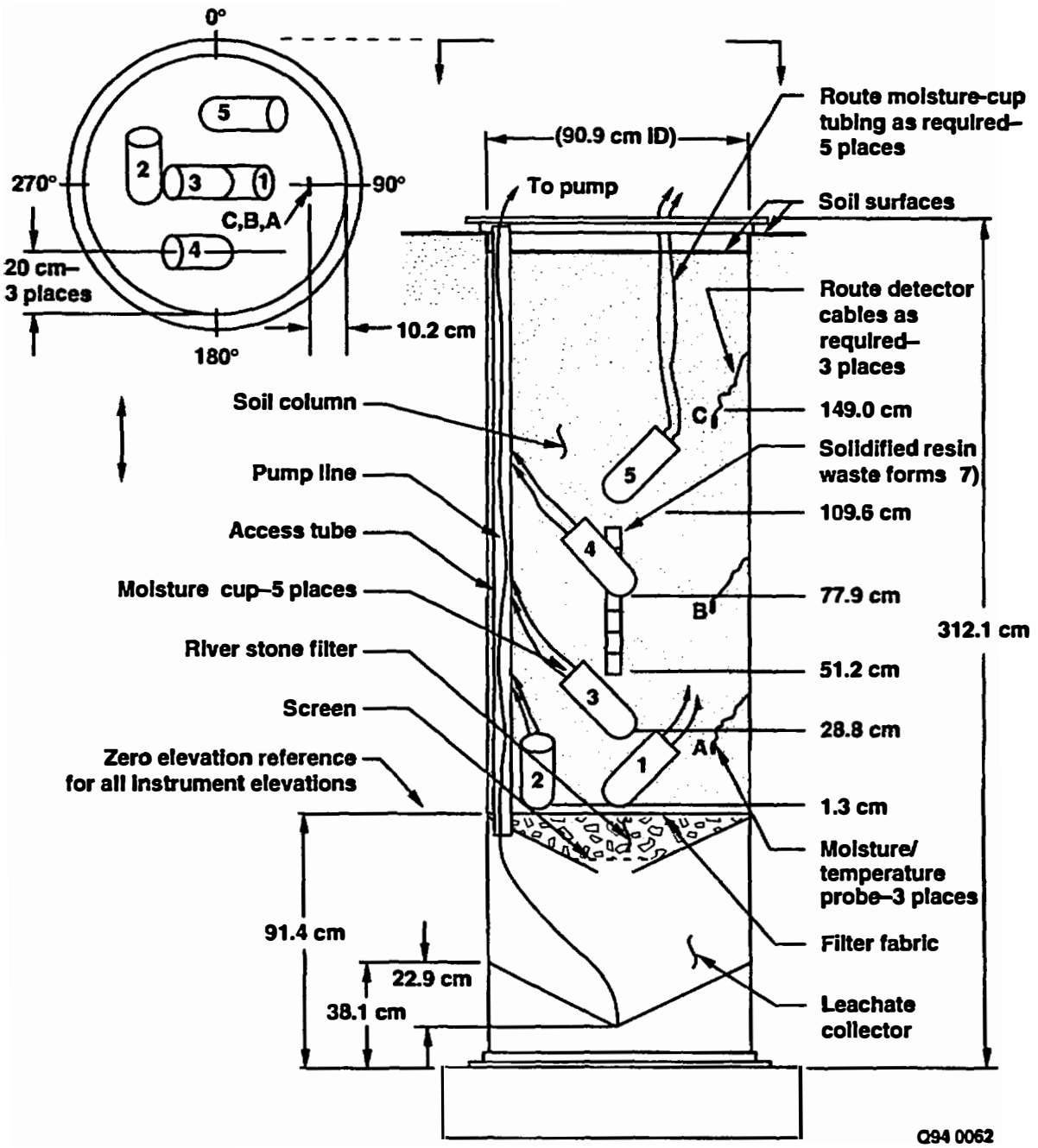

Figure 10. EPICOR-II lysimeter vessel component locations.

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 usually contains DAS data from ANL-E and ORNL. However, due to technical problems with the ORNL DAS during the year, which resulted in recording data during only the months of July 1994 and April and May of 1995, only DAS data from those months are presented. ANL-E data are obtained from July 1994 through June 1995. Information on water balance and on nuclide chemical species content in soil water and leachate from both sites is available. It has been compiled from the initiation of the project (ANL-E—August 1, 1985; ORNL—June 1, 1985) through June 1995. Many of the data are displayed in graphic format so that information can be correlated easily with time.

Weather Data. Precipitation and air temperature, as recorded by the ANL-E DAS during the 12-month reporting period, are presented in Figures 11 and 12. ORNL precipitation data, provided by a nearby measuring station, are found in Figure 13. Figure 14 shows ORNL air temperature data. 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 95.4 cm at ANL-E and 115.3 cm at ORNL. This year, ANL-E was 12% above the normal annual rainfall while ORNL was 17% below normal (ANL-E—85.2 cm; ORNL—138.8 cm).[21,22] This is the second time in the past 9 years that ORNL has not equalled or exceeded the normal amount of yearly precipitation. The monthly precipitation pattern for each site can be seen from the histograms in Figures 11 and 13. Figure 15 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 918 cm at ANL-E, while ORNL had reached a total of 1,338 cm.

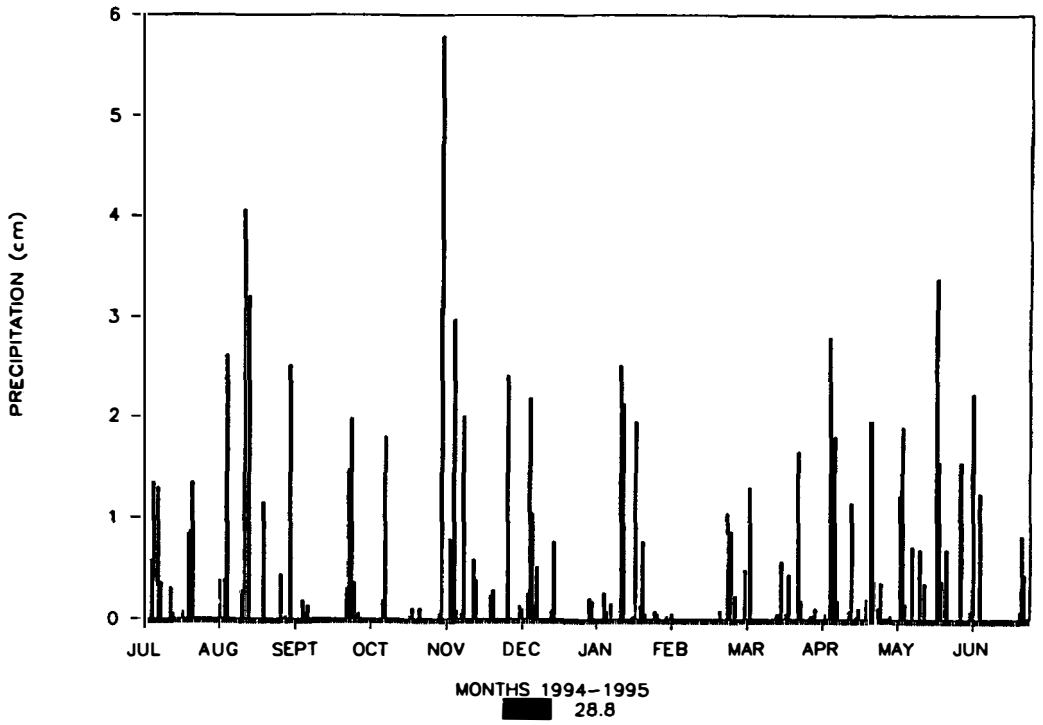

Figure 11. ANL-E weather data—precipitation.

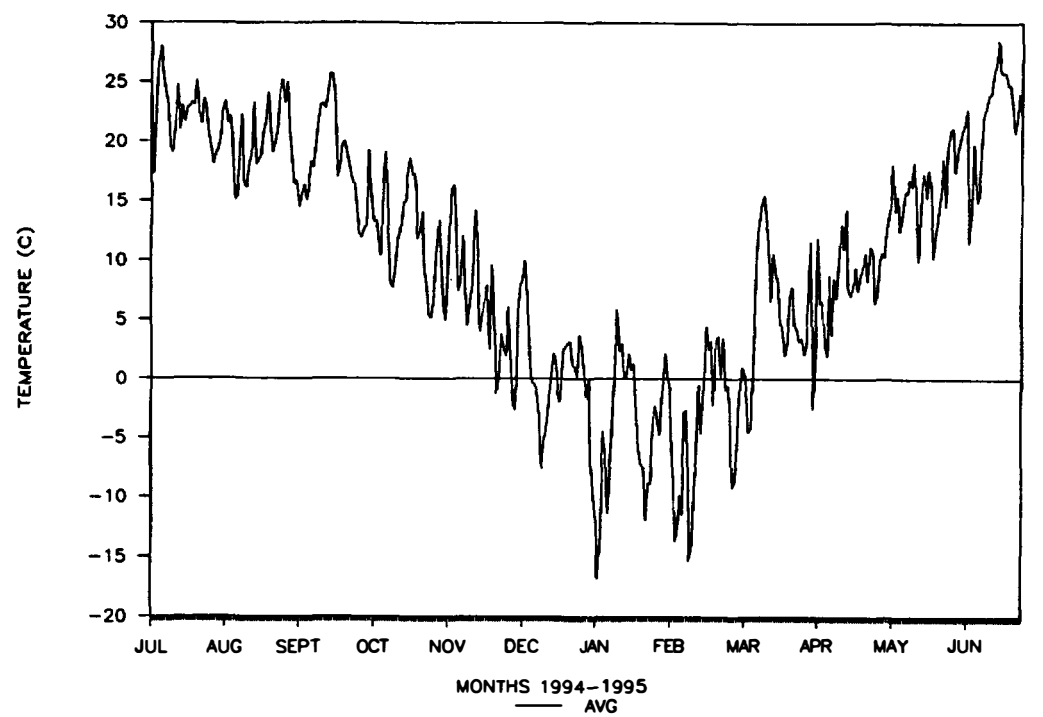

Figure 12. ANL-E weather data—air temperature.

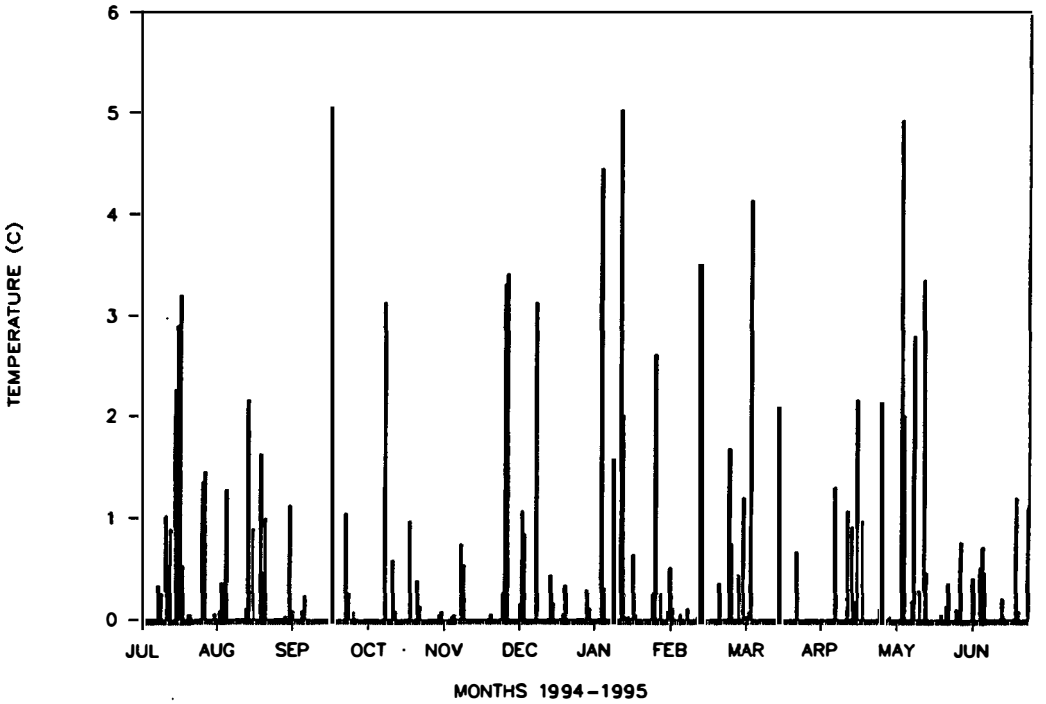

Figure 13. ORNL weather data—precipitation.

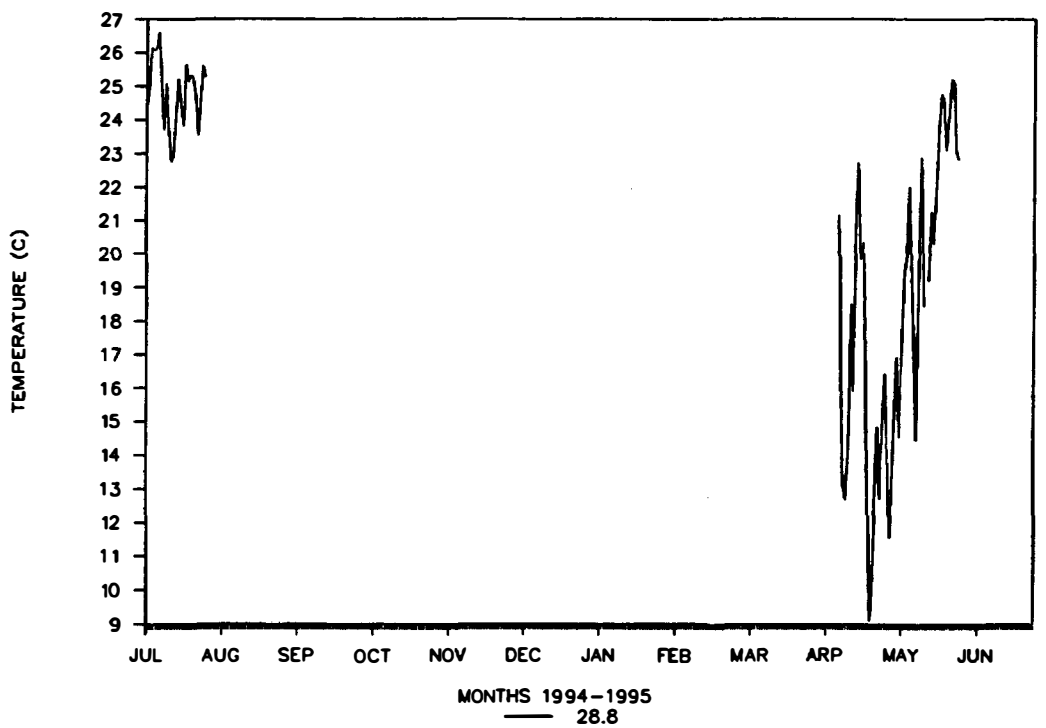

Figure 14. ORNL weather data—air temperature.

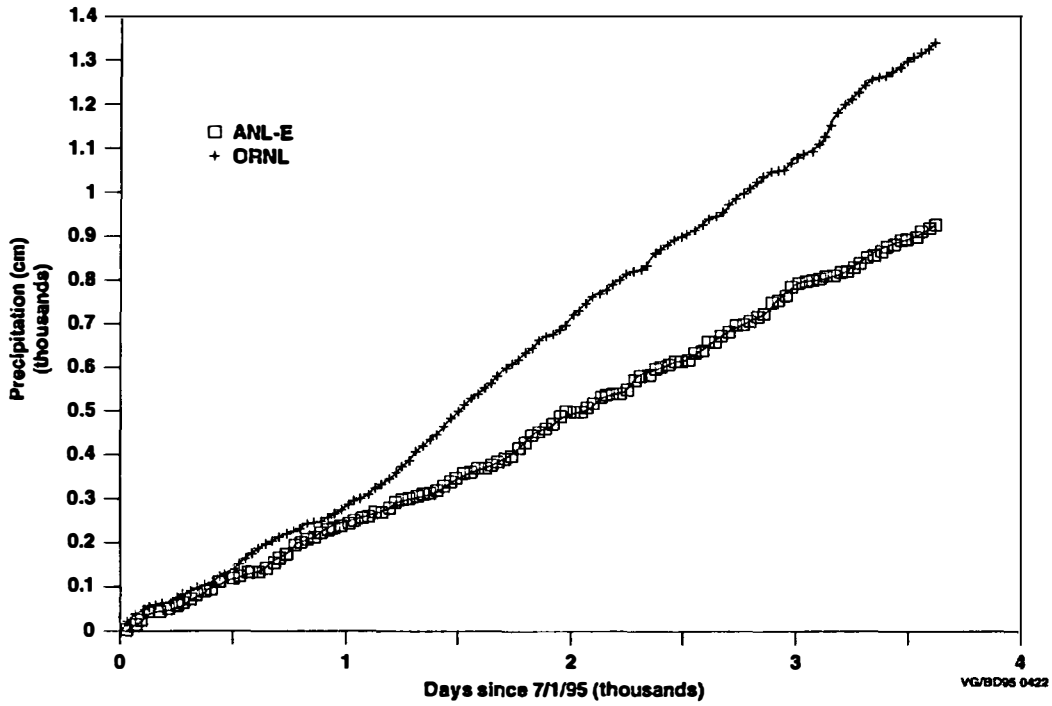

Figure 15. ANL-E and ORNL weather data—cumulative precipitation.

Air temperature data from ANL-E (Figure 12) show that periods of freezing temperatures occurred from early November 1994 until mid March 1995.

Lysimeter Soil Temperature Data. The lysimeter soil temperature data recorded at ANL-E during the reporting period are shown in Figures 16 through 19. 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 ANL-E. Soil temperatures recorded at ORNL are shown in Figures 20 through 24. As mentioned earlier, technical problems with the DAS at ORNL resulted in the loss of over 9 months of data.

Past reports have detailed the failure of some temperature probes at ANL-E. This year, faults were found with one temperature probe in ANL-2, two in ANL-3, all in ANL-4, and two 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). Most of the ORNL temperature probes appear to have been functioning properly. However, several of the probes at ORNL have outputs that are not consistent. The probes at 28.8 cm (lowest in soil column) in ORNL-1, 3, and 5 all appear questionable as does ORNL-1 at 77.9 cm.

Lysimeter Soil Moisture Data. Data from the moisture probes at ANL-E (shown in Figures 25 through 29) and ORNL (shown in Figures 30 through 34) indicate that the lysimeter soil columns at that site have remained moist during the reporting period. As explained previously, the data from ORNL was collected only during the months of July 1994 and April through mid-May 1995. 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

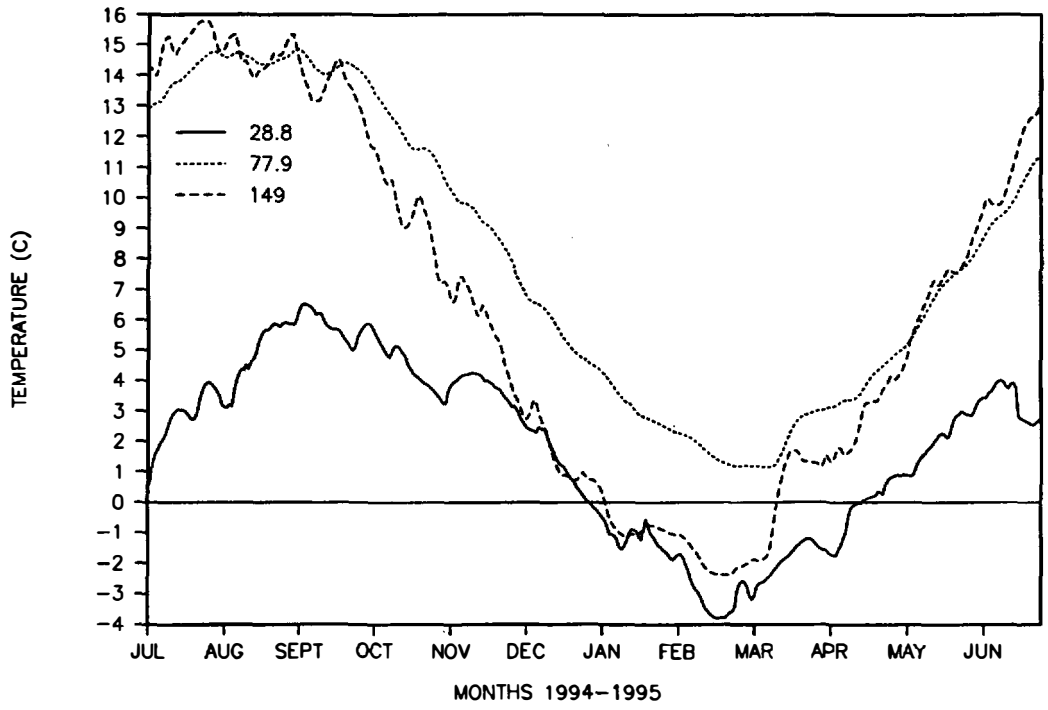

Figure 16. ANL-E lysimeter 1 soil temperature.

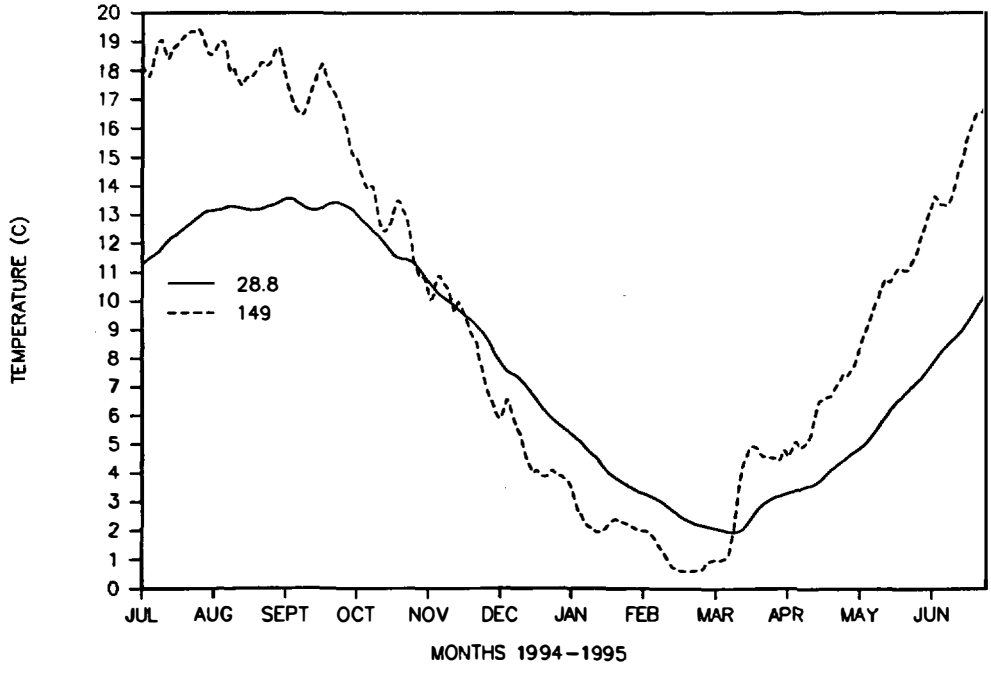

Figure 17. ANL-E lysimeter 2 soil temperature.

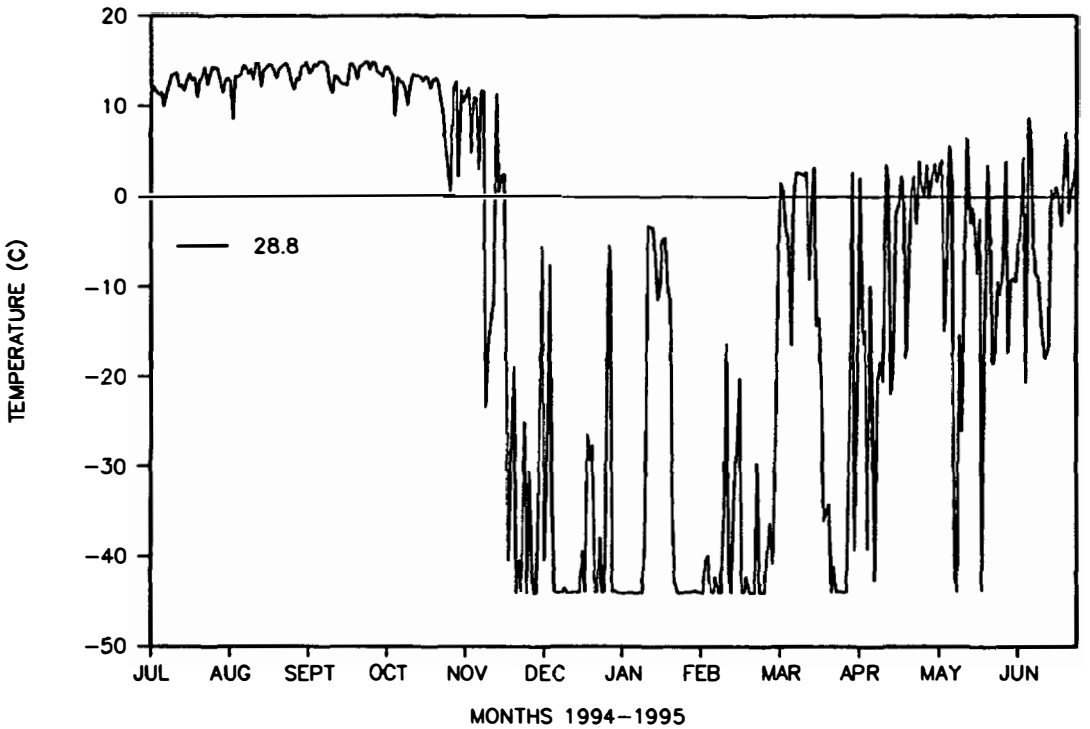

Figure 18. ANL-E lysimeter 3 soil temperature.

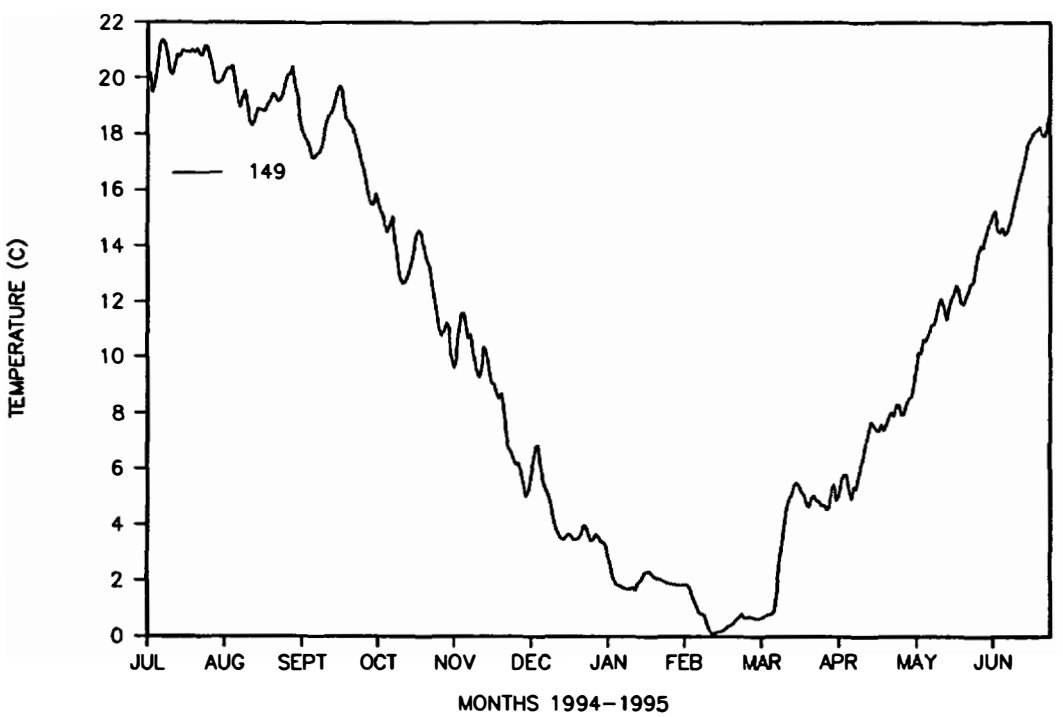

Figure 19. ANL-E lysimeter 5 soil temperature.

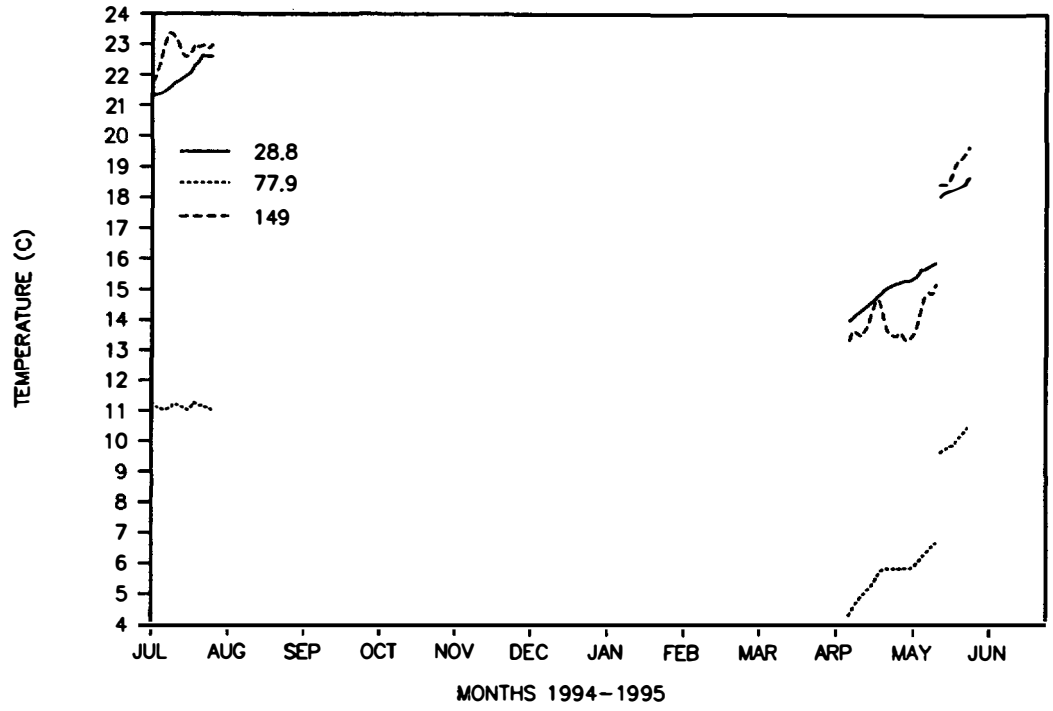

Figure 20. ORNL lysimeter 1 soil temperature.

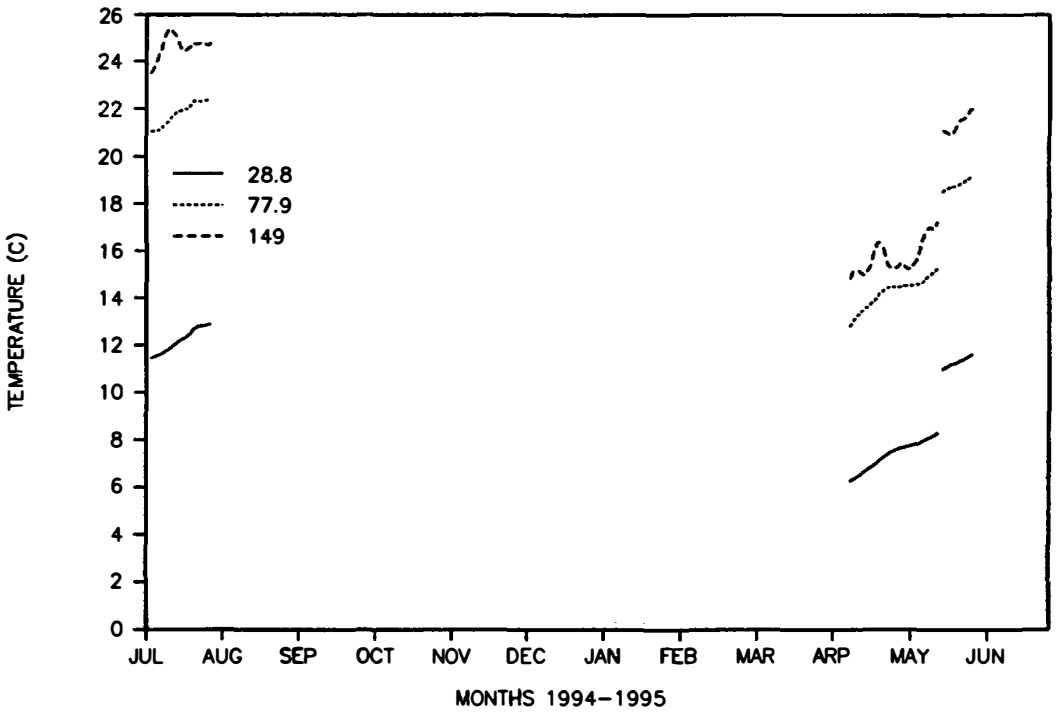

Figure 21. ORNL lysimeter 2 soil temperature.

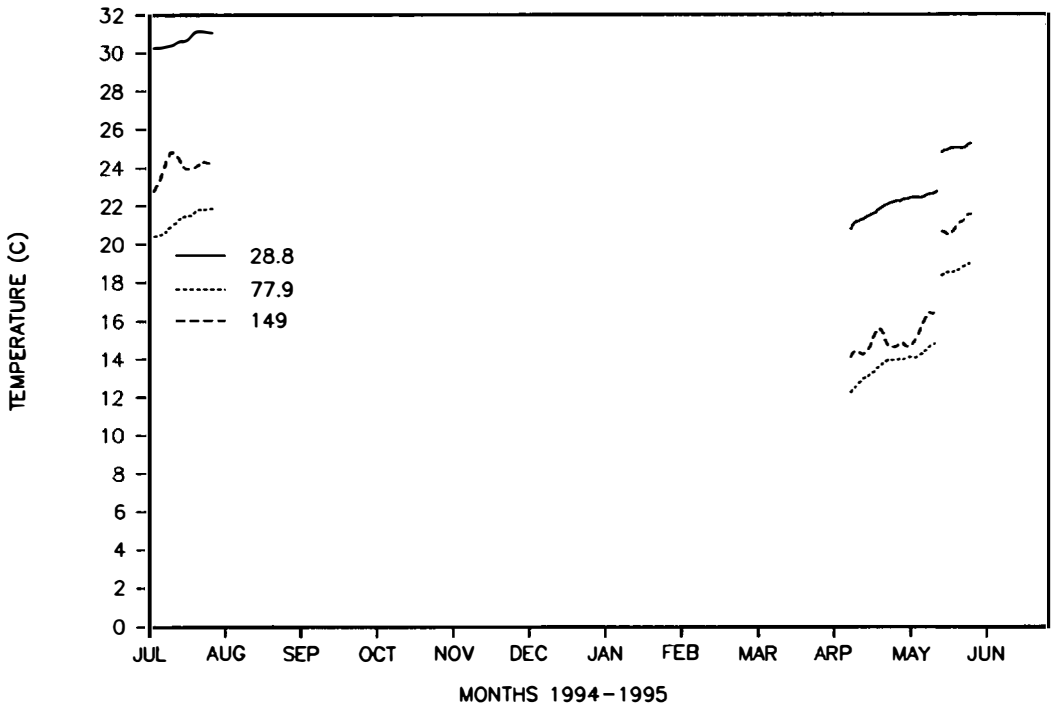

Figure 22. ORNL lysimeter 3 soil temperature.

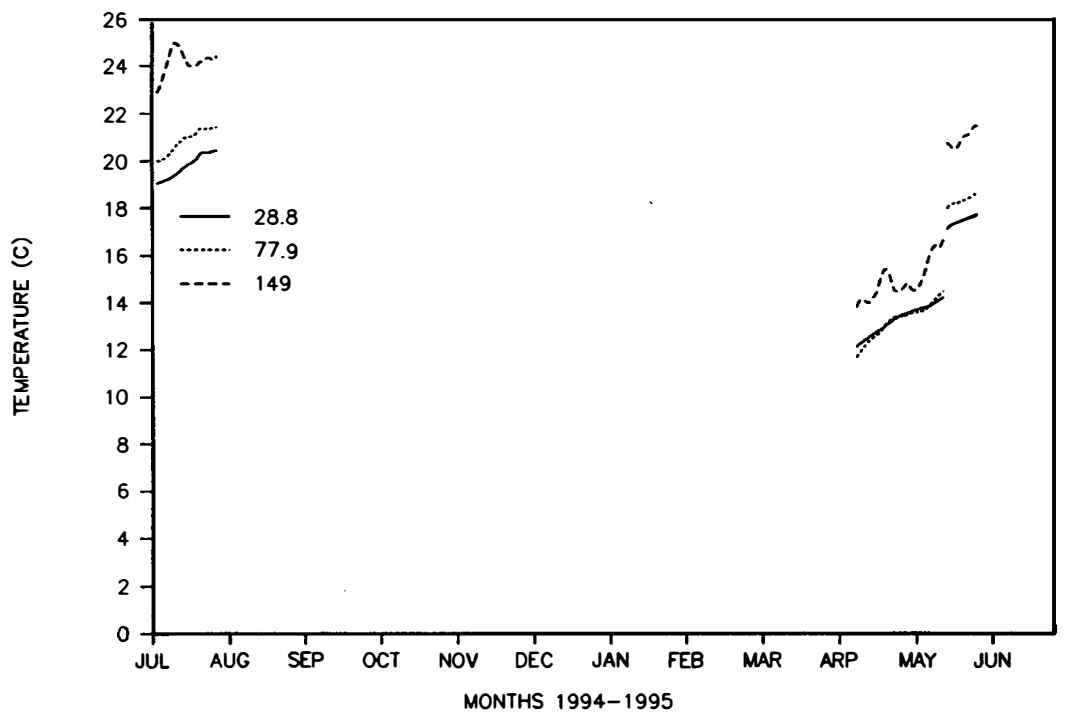

Figure 23. ORNL lysimeter 4 soil temperature.

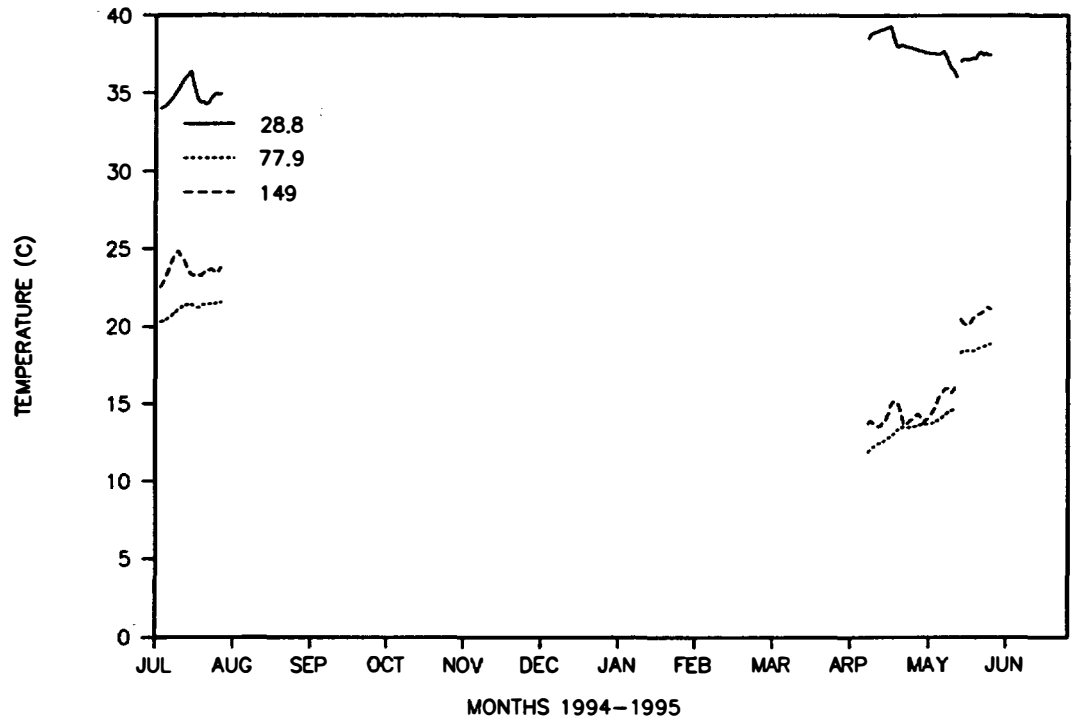

Figure 24. ORNL lysimeter 5 soil temperature.

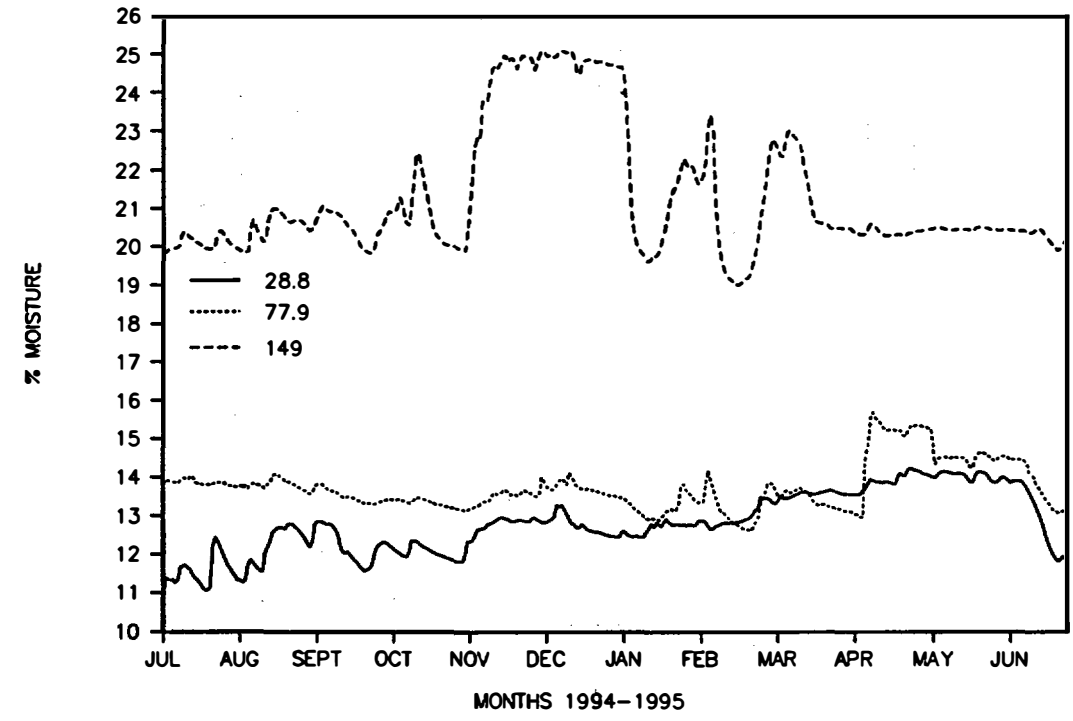

Figure 25. ANL-E lysimeter 1 soil moisture.

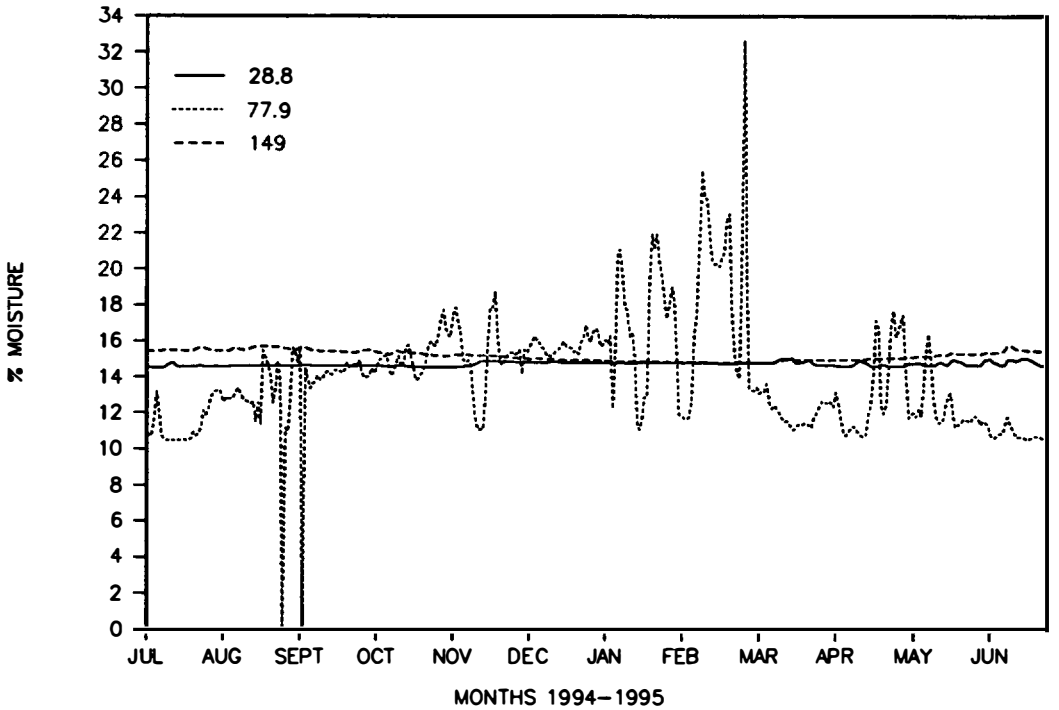

Figure 26. ANL-E lysimeter 2 soil moisture.

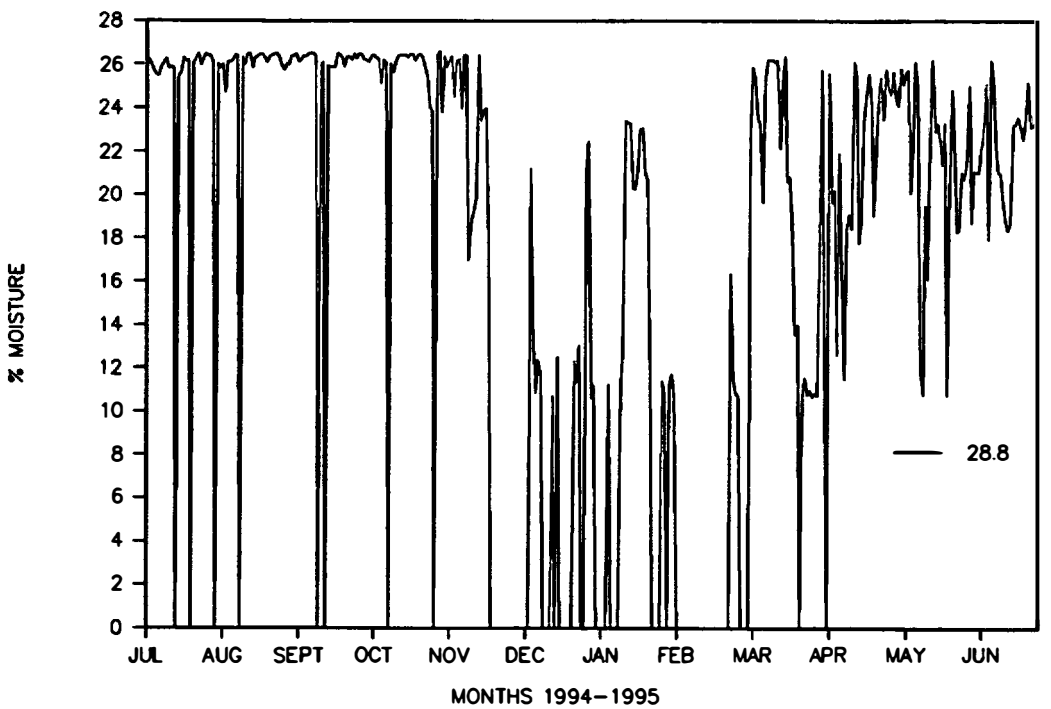

Figure 27. ANL-E lysimeter 3 soil moisture.

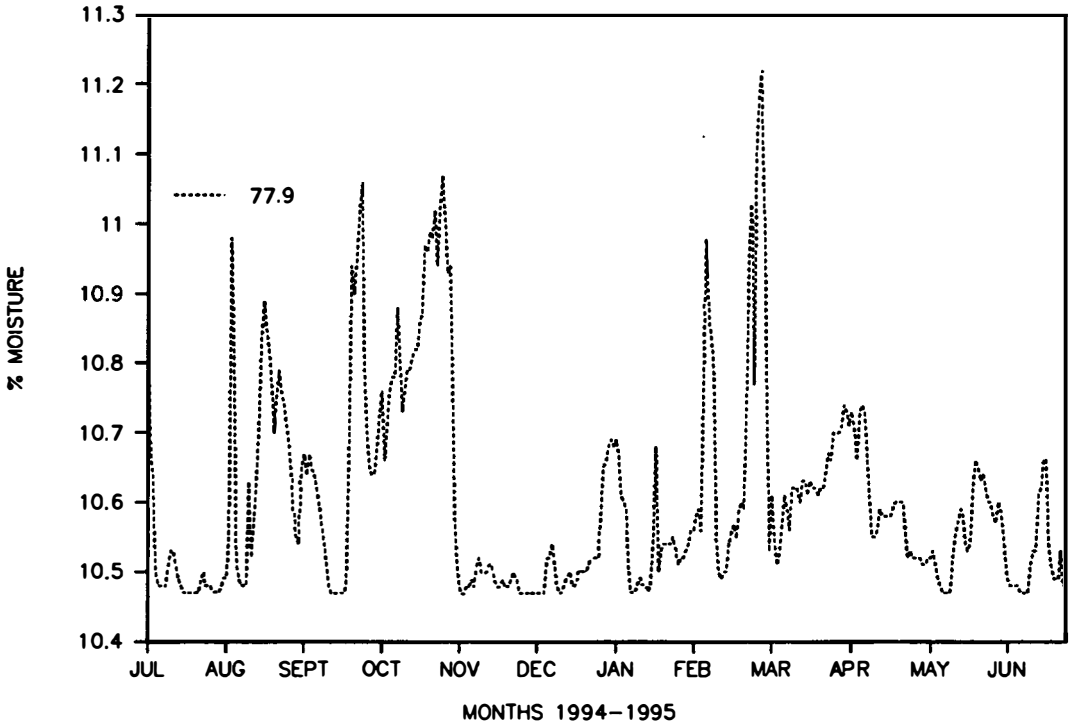

Figure 28. ANL-E lysimeter 4 soil moisture.

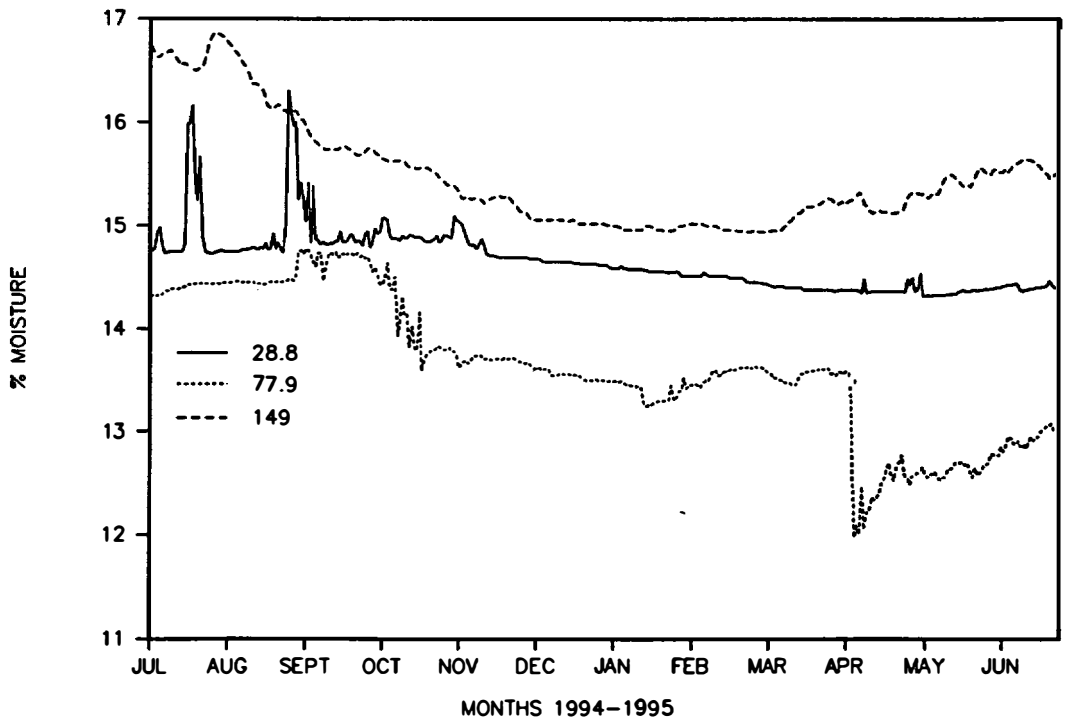

Figure 29. ANL-E lysimeter 5 soil moisture.

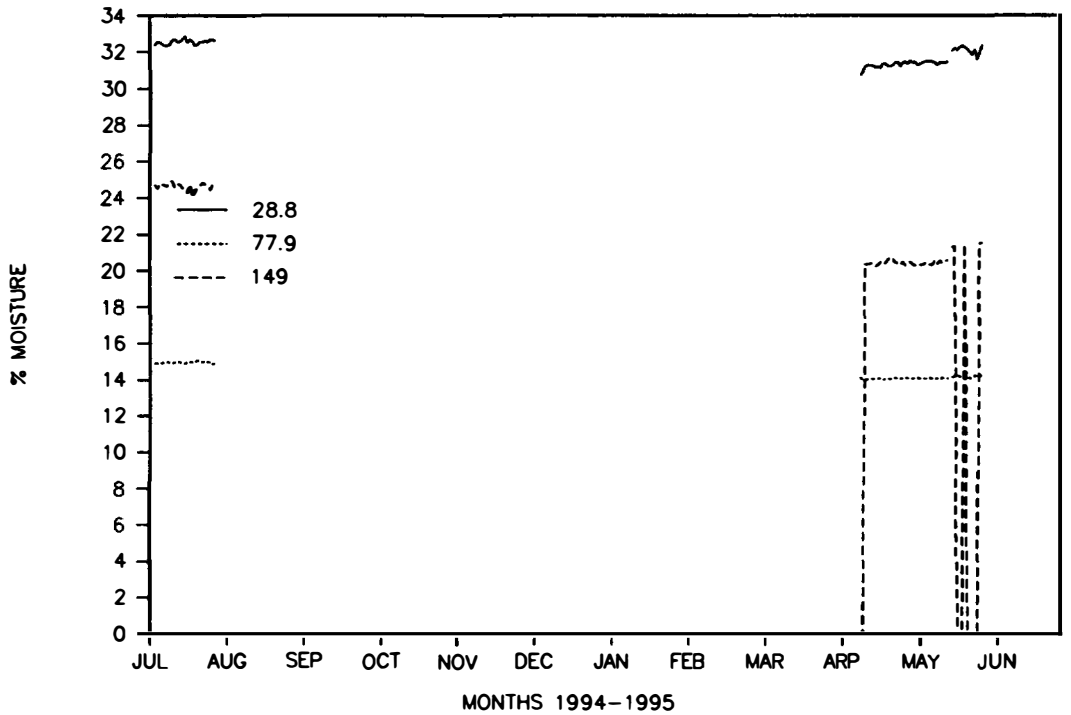

Figure 30. ORNL lysimeter 1 soil moisture.

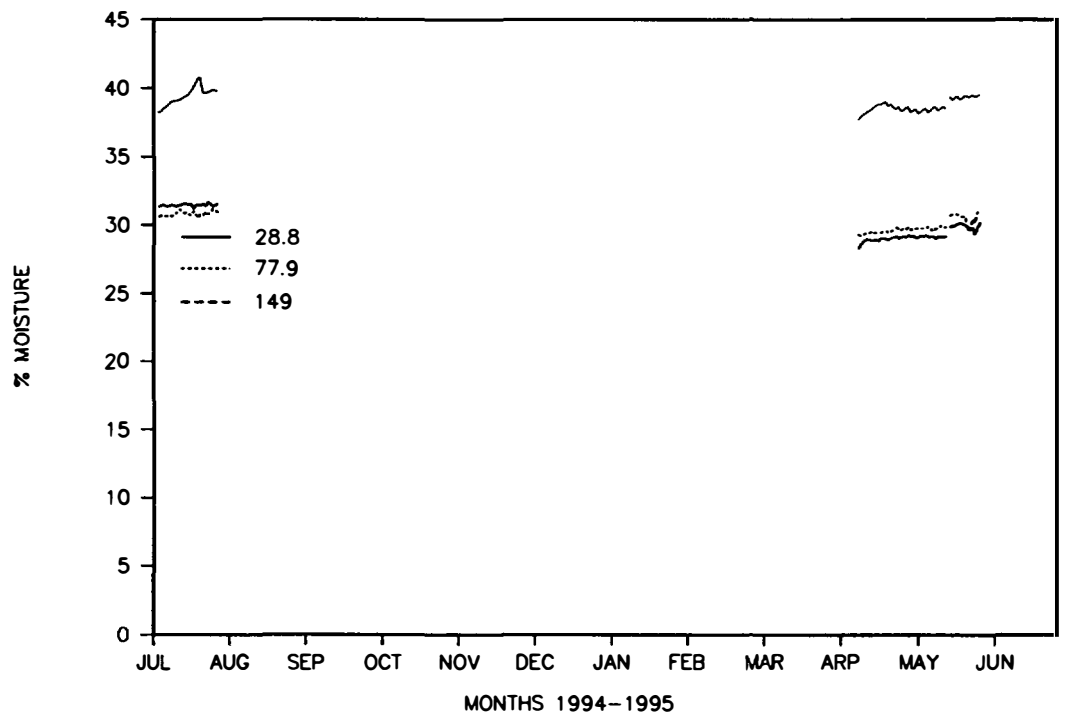

Figure 31. ORNL lysimeter 2 soil moisture.

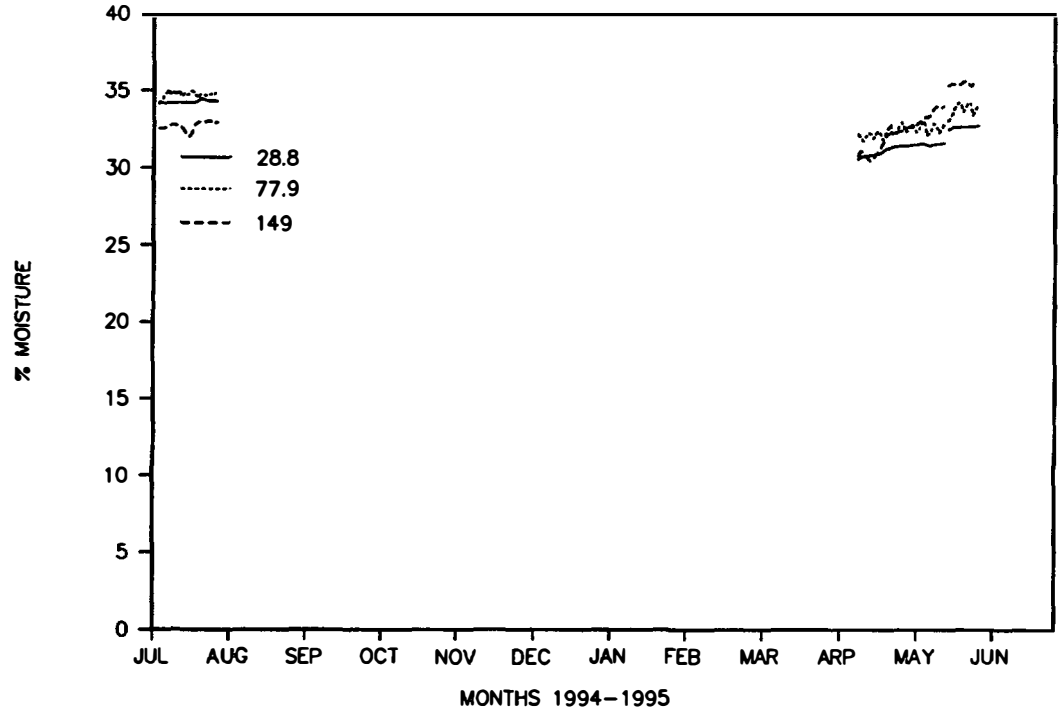

Figure 32. ORNL lysimeter 3 soil moisture.

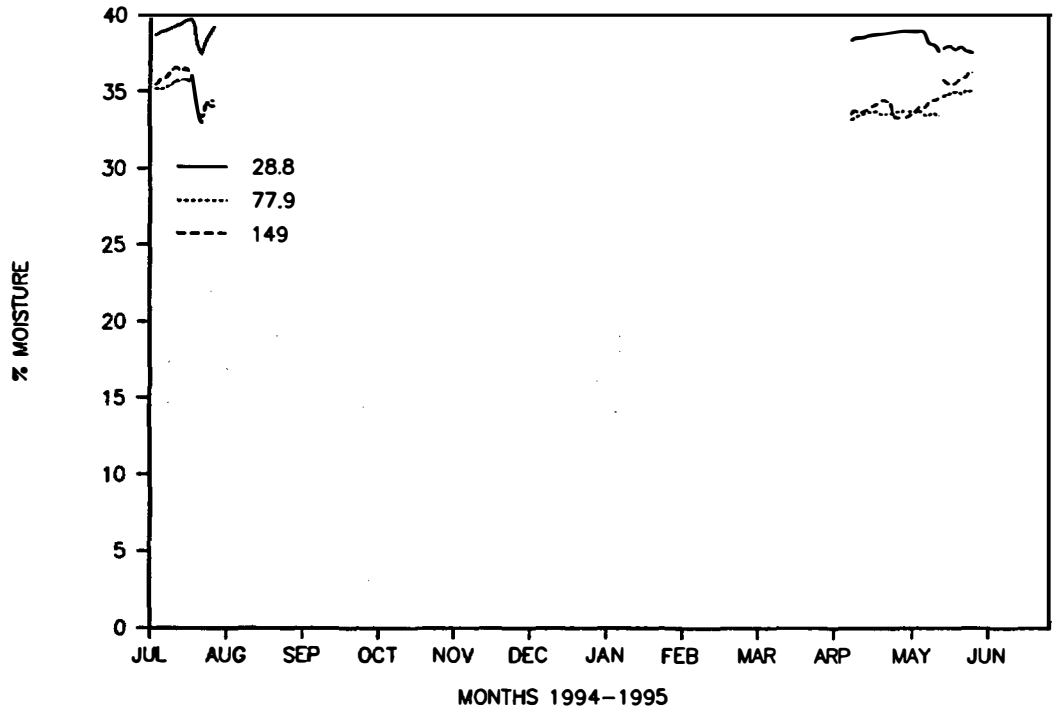

Figure 33. ORNL lysimeter 4 soil moisture.

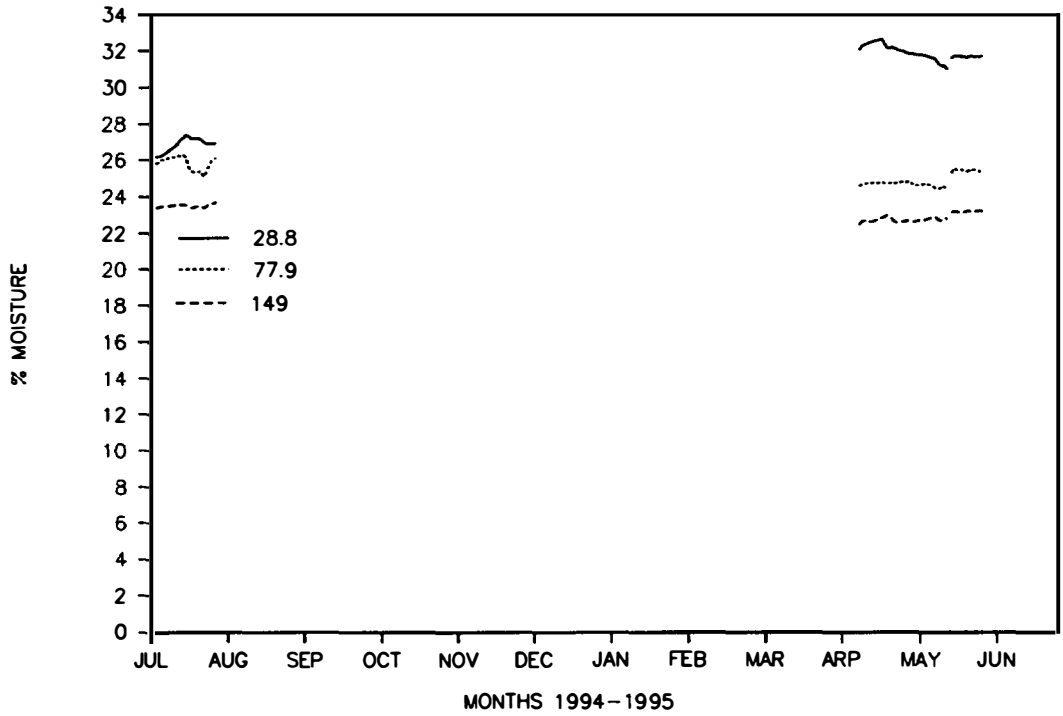

Figure 34. ORNL lysimeter 5 soil moisture.

similar and not excessive. There was a coefficient of variation (CV) of 20.3% at ANL-E and 5.1% at ORNL. The moisture 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). Several probes at ONRL are questionable, specifically those in ORNL-1.

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 moisture probe data (Table 5). Percent differences between the gravimetric data and moisture probe data for ANL-E lysimeters range between 20.4 and 52.0%. These values have increased during this reporting period, but are still within a reasonable range given the use of the information.

In addition to using the moisture probe and gravimetric data to calculate soil moisture starting the summer of 1991, a neutron moisture detecting probe was used at ANL-E. Operation of the neutron probe, using 1991 calibration curves, produced data that were comparable to gravimetric overall average values within 4.1%, but underestimated those values (see Table 3). The variability between gravimetric 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 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 15.2%. In spite of the difference between gravimetric and measured soil moisture at the two sites, the neutron probe does provide a rapid estimate of moisture in the soil column.

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	17.3	—
1	41–62	19.0	20.2
1	62–85	21.2	20.2
1	82–107	20.2	—
1	107–133	21.1	—
1	133–153	22.1	20.2
1	153–182	22.4	—
1	182–202	22.9	22.7
2	0–41	16.7	—
2	41–62	19.5	18.1
2	62–82	23.0	18.6
2	82–107	24.0	—
2	107–133	24.7	—
2	133–153	23.3	19.6
2	153–182	23.1	—
2	182–202	23.3	21.9
3	0–41	18.3	—
3	41–62	21.3	18.3
3	62–82	23.2	22.5
3	82–107	22.8	—
3	107–133	23.0	—
3	133–153	22.7	20.7
3	153–182	22.3	—
3	182–202	23.2	25.0
4	0–41	18.3	—
4	41–62	20.1	20.1
4	62–82	22.7	21.2
4	82–107	22.7	—
4	107–133	23.0	—
4	133–153	23.5	21.8
4	153–182	23.0	—
4	182–202	23.3	23.2

a. Samples were collected on July 31, 1995.

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	13.1	13.2
1	41–62	14.8	16.0
1	62–85	15.0	16.5
1	82–107	16.3	16.7
1	107–133	16.6	15.9
1	133–153	16.3	17.1
1	153–182	18.0	17.3
1	182–202	15.8	17.9
2	0–41	11.0	12.6
2	41–62	12.5	14.7
2	62–82	19.6	15.0
2	82–107	12.7	14.6
2	107–133	15.3	14.1
2	133–153	14.9	15.7
2	153–182	15.6	15.7
2	182–202	10.9	16.3
3	0–41	11.8	13.0
3	41–62	10.2	15.9
3	62–82	9.7	16.3
3	82–107	13.5	17.1
3	107–133	13.2	15.4
3	133–153	13.6	16.7
3	153–182	16.6	17.3
3	182–202	16.2	18.6
4	0–41	9.4	11.3
4	41–62	10.8	16.7
4	62–82	10.2	17.6
4	82–107	13.7	17.4
4	107–133	8.9	16.1
4	133–153	11.9	17.8
4	153–182	16.6	18.2
4	182–202	18.4	19.5

a. Samples were collected on June 21, 1995.

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 moisture probes for preceding 12-month period[a]	Average percent moisture for soil column determined gravimetrically for summer 1995	Percent difference between gravimetric and probe
ANL-1	15.9 ± 4.6	20.8 ± 1.8	23.6
ANL-2	14.6 ± 0.6	22.2 ± 2.5	34.2
ANL-3	17.6[b]	22.1 ± 1.6	20.4
ANL-4	10.6[b]	22.1 ± 1.8	52.0
ORNL-1	31.9[b,c]	15.7 ± 1.4	103.2
ORNL-2	33.1 ± 5.0[c]	14.1 ± 2.9	134.8
ORNL-3	33.0 ± 0.4[c]	13.1 ± 2.5	151.9
ORNL-4	35.9 ± 2.4[c]	12.5 ± 3.5	187.2

a. July 1994 through June 1995.

b. Average from one probe.

c. Data available only during July 1994 and May through June 1995.

Soil moisture (as gravimetrically determined) at each sampling depth has remained uniformly consistent between intrasite lysimeters during the past several years (Figures 35 and 36). The uniformity of soil moisture in the ANL-E lysimeters (Figure 35) 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 (Tables 3, 4, and 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 except ANL-3. Water accumulation at ANL-E during the last 12 months occurred in lysimeters 1, 2, 3, and 4. A total of 247 L of water was removed from ANL-1, 150 L from ANL-2, 19 from ANL-3, and 181 L from ANL-4 over the year.

As shown in Figures 35 and 36, 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 20). At the time of the 1995 sampling, the average soil moisture of ANL-E soils had decreased from 54.3 to 53.6% of the soil moisture holding capacity, while at ORNL, this value decreased from 35.2% for 1994 to 30.9% for 1995 (this value is nearly 10% less than that for 1993—39.4% versus 30.9%). This year, it is noted that the ORNL percent moisture at depth was not as uniform as has been seen in past years (Figure 36).

By using the cumulative rainfall data from each site since the lysimeters were placed in operation (Figure 15), 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

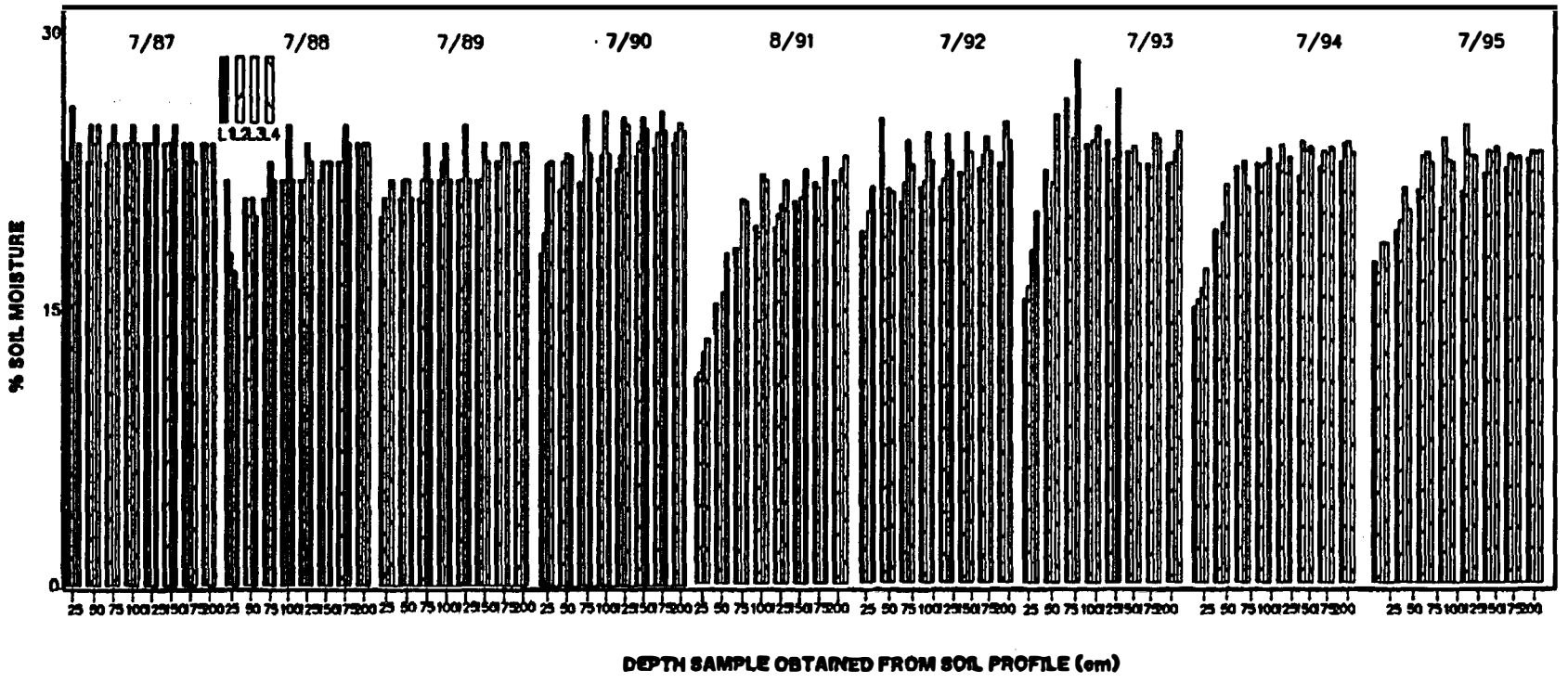

Figure 35. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.

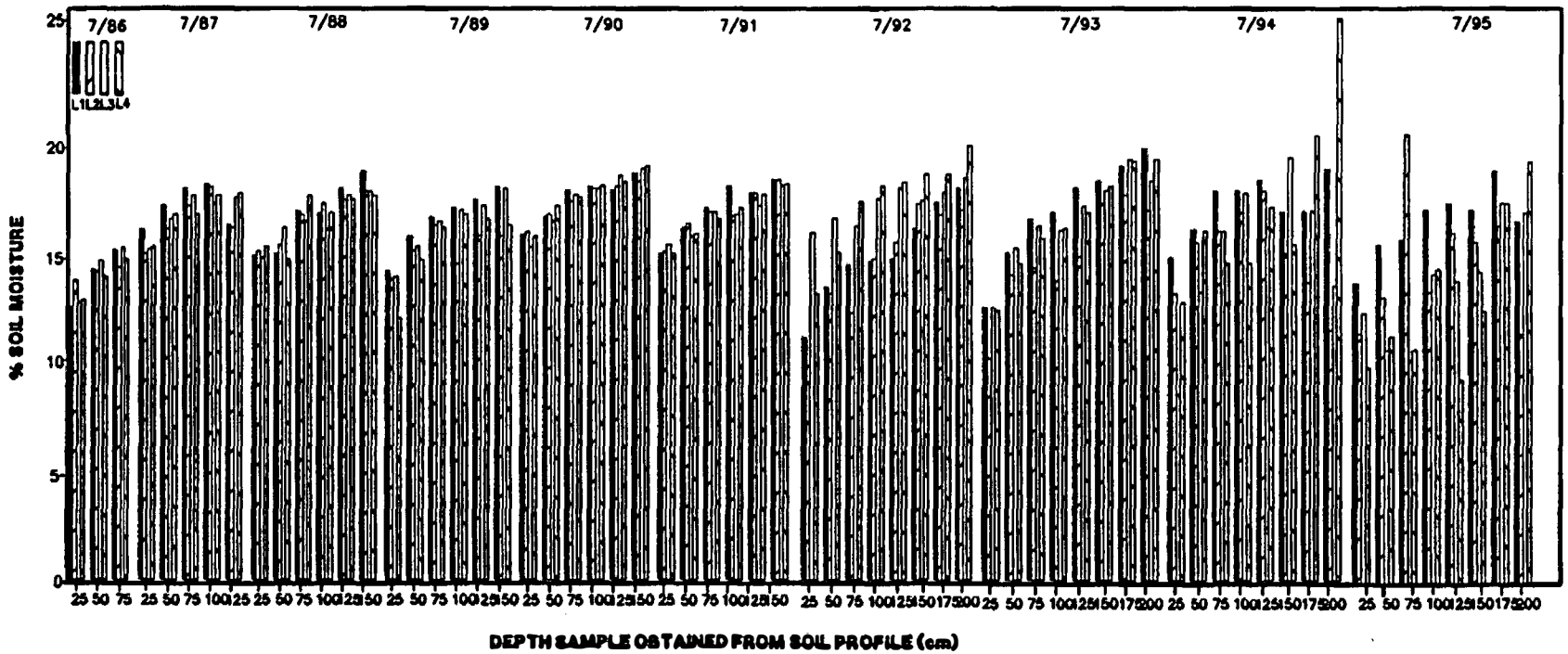

Figure 36. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.

by each ANL-E lysimeter has now reached 5,957.4 L; at ORNL, this value is 8,682.9 L. Figures 37 and 38 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 2,461 ± 1,058 L, with a range of 24.7 to 65.9% of total precipitation received, has passed through the soil lysimeters, while for the control, this value was 6,081 L or 102.1% of the calculated available precipitation. For ORNL, the values were 7,785 ± 164 L (89.7%) for the soil-filled lysimeters and 8,864 L (102.1%) for the control. These trends are comparable to the previous year's trends (Reference 20). 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 37 and 38). Also, the small deviation in total yearly leachate throughput with the ORNL soil lysimeters (2.1%) 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 43.0%.

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 465 ± 37 L (CV 7.8%) of water that was removed as a combination of leachate and standing water. During the previous year, this volume was 288 ± 14 L (CV 4.9%).

The total volumes of precipitation that have moved through the lysimeters represent an average of 3.4 pore volumes for the ANL-E soil lysimeters and 12.4 pore volumes for soil lysimeters at ORNL, while the controls at ANL-E and ORNL were 13.3 and 13.7 pore volumes, respectively. These data show that the ORNL soil lysimeters have had an average of 3.6 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 10 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 5 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. The collection was made during the fourth quarter 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 39 through 47.

As has been reported in the past (References 11 and 13 through 20), 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 all cups 3 at both ANL-E (range of 17 to 42%) and ORNL (range of 13 to 62%) (Tables 6 and 7; Figures 39 and 40). There continues to be standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a

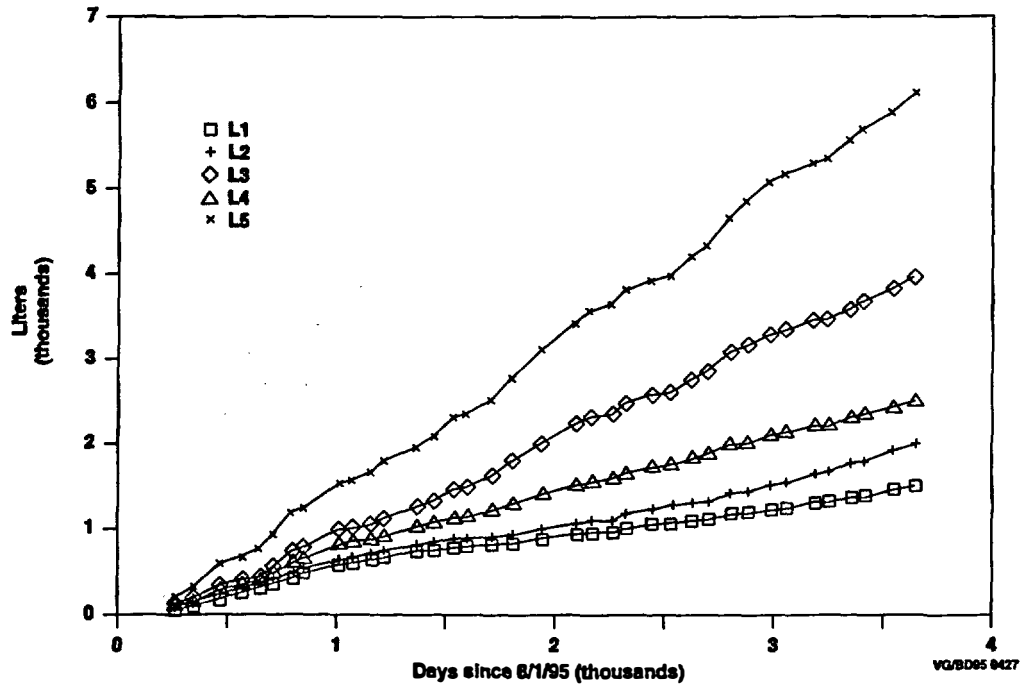

Figure 37. ANL-E cumulative volume of leachate from lysimeters.

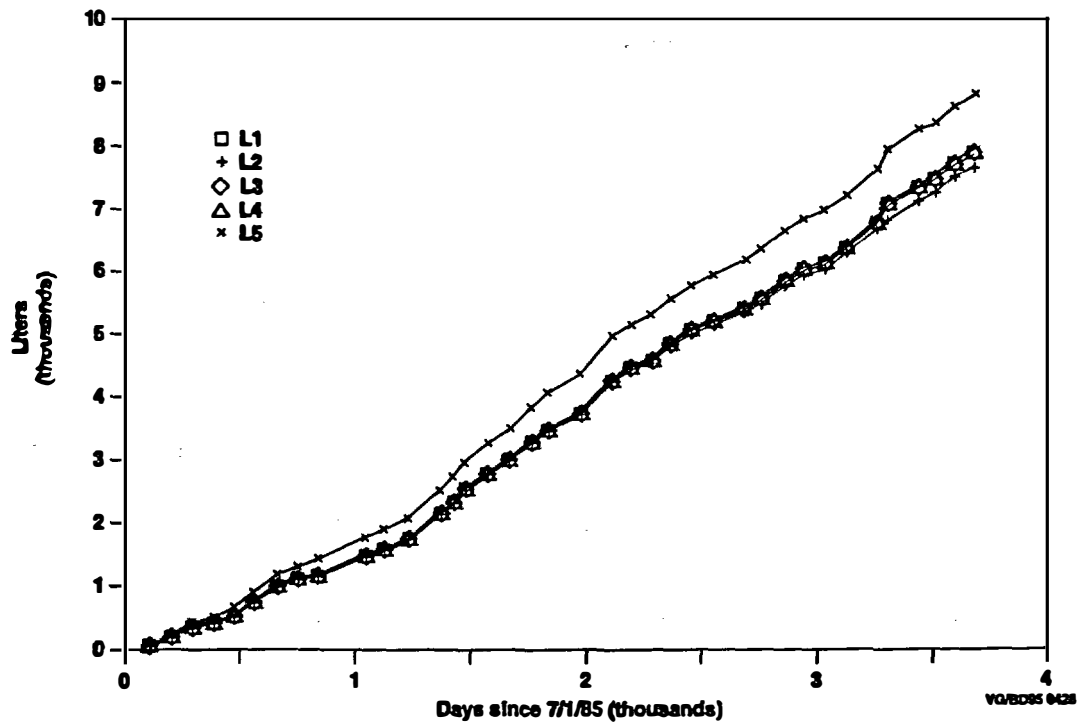

Figure 38. ORNL cumulative volume of leachate from lysimeters.

Table 6. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 10 (1994–1995).

	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
Sample identification	Oct 94	Dec 94	Apr 95	Jul 95	Oct 94	Dec 94	Apr 95	Jul 95	Oct 94	Dec 94	Apr 95	Jul 95
Lys 1[b]	<200	<200	<200	<100	<50	<50	<50	<50	<0.6	<0.6	<0.6	<0.4
Lys 2	<200	<200	<200	<100	<50	<50	<50	<50	<0.6	<0.6	<0.6	<0.4
Lys 3	<200	<200	<200	<100	<50	<50	<50	<50	457 ± 5	449 ± 6	293 ± 5	330 ± 7
Lys 4	<200	<200	<200	<100	<50	<50	<50	<50	15 ± 1	14 ± 1	9 ± 1	8.0 ± 0.5
Lys 5	<200	<200	<200	<100	<50	<50	<50	<50	1,617 ± 22	1,537 ± 21	1,462 ± 21	1,400 ± 25
Lys 1-3[c]	<200	<200	<200	<100	<50	<100	<50	<100	2.6E+4 ± 400	3.5E+4 ± 469	4.1E+4 ± 494	3.9E+4 ± 400
Lys 2-3	<200	<200	<200	<100	600 ± 20	320 ± 30	230 ± 10	410 ± 40	7,800 ± 60	1.0E+4 ± 70	4,350 ± 50	6,100 ± 55
Lys 3-3	<200	<200	<200	<100	70 ± 14	<100	60 ± 8	<100	115E+4 ± 1.6E+4	122E+4 ± 1.9E+4	150E+4 ± 2.2E+4	124E+3 ± 1.7E+4
Lys 4-3	<200	<200	<200	<100	<50	<100	<50	<50	4.5E+4 ± 522	4.0E+4 ± 490	3.7E+4 ± 484	5.1E+4 ± 493
Lys 5-3	<200	<200	<200	<100	<15.6E+4 ± 910	33.6E+4 ± 670	23.6E+4 ± 700	46.6E+4 ± 460E+2	4.1E+4 ± 503	3.6E+4 ± 490	4.3E+4 ± 517	5.4E+4 ± 520
Lys 1-1[c]	<200	<200	<200	<100	<50	<50	<50	<50	25 ± 2	28 ± 2	32 ± 2	32 ± 2
Lys 2-1	<200	<200	<200	<100	—	—	<50	<50	—	11 ± 4	<6	7 ± 3
Lys 3-1	<200	<200	<200	<100	<50	<50	<50	<50	7,600 ± 90	8,500 ± 100	1.2E+4 ± 113	9,730 ± 80
Lys 4-1	<200	<200	<200	<100	<50	<50	<50	<50	507 ± 13	—	1,080 ± 35	1,070 ± 16
Lys 5-1	<200		<200	<100	<50	<50	<50	<50	2,875 ± 55	1,650 ± 40	1,800 ± 40	1,430 ± 35
Lys 1-2	—	—	<50	—	—	—	<100	—	—	—	2.5 ± 1.1	—
Lys 2-2	—	—	<50	—	—	—	<100	—	—	—	<1.5	—
Lys 3-2	—	—	<50	—	—	—	<100	—	—	—	6.9 ± 1.3	—
Lys 4-2	—	—	<50	—	—	—	<100	—	—	—	1,076 ± 39	—
Lys 5-2	—	—	<50	—	—	—	<100	—	—	—	3,550 ± 60	—

a. Concentration ± 2 sigma.

b. One-L subsample from leachate collector.

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

Table 7. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 10 (1994–1995).

Sample identification	Concentration (pCi/L)[a]							
	Co-60				Cs-137			
	Sep 94	Jan 95	Apr 95	Jun 95	Sep 94	Jan 95	Apr 95	Jun 95
Lys 1[b]	2.7 ± 43.2	–4.3 ± 22.4	0.3 ± 8.2	13.5 ± 40.5	13.5 ± 40.5	1.1 ± 22.9	0.5 ± 7.8	72.9 ± 5.4
Lys 2	10.8 ± 27.0	–2.7 ± 27.0	2.4 ± 4.6	–2.7 ± 35.1	–2.7 ± 35.1	–3.8 ± 25.1	0.8 ± 5.1	18.8 ± 4.6
Lys 3	17.8 ± 23.5	13.0 ± 23.4	0.3 ± 4.3	16.2 ± 35.1	16.2 ± 35.1	8.9 ± 19.2	–0.3 ± 4.3	7.6 ± 3.5
Lys 4	–8.1 ± 45.9	16.2 ± 15.1	–2.4 ± 8.1	–2.7 ± 32.4	–2.7 ± 32.4	1.6 ± 20.3	3.2 ± 5.9	7.0 ± 4.0
Lys 5	–10.8 ± 48.6	–16.2 ± 62.1	3.5 ± 5.4	2,973 ± 270	2,973 ± 270	1,595 ± 81.1	594 ± 27.0	540 ± 27.0
Lys 1-3[c]	–29.7 ± 86.5	–62.2 ± 164	18.9 ± 113.5	13.5 ± 70.3	–10.8 ± 89.2	27.0 ± 138	21.6 ± 108	2.7 ± 83.8
Lys 2-3	21.6 ± 48.6	24.3 ± 97.3	54.1 ± 102.7	21.6 ± 78.4	18.9 ± 43.2	32.4 ± 100	48.6 ± 116	297 ± 81.1
Lys 3-3	–27.0 ± 94.6	—	75.7 ± 121.6	–32.4 ± 121	29.7 ± 64.9	—	254 ± 151	–51.3 ± 122
Lys 4-3	13.5 ± 27.0	–10.8 ± 154	64.9 ± 64.9	–5.4 ± 86.5	45.9 ± 48.6	–16.2 ± 143	–35.1 ± 129	–13.5 ± 116
Lys 5-3	–21.6 ± 102.7	—	–10.8 ± 121.6	21.6 ± 78.4	2.1E+4 ± 540	—	2.0E+4 ± 540	5.1E+4 ± 2,703
Lys 1-1[c]	5.4 ± 70.3	81.1 ± 351	13.5 ± 113.5	–2.7 ± 78.4	784 ± 108	108 ± 324	51.3 ± 108	29.7 ± 94.6
Lys 2-1	2.7 ± 67.6	–27.0 ± 56.8	8.11 ± 113.5	–24.3 ± 108	21.6 ± 48.6	232 ± 43.2	13.5 ± 100	73.0 ± 62.2
Lys 3-1	43.2 ± 59.46	–29.7 ± 59.5	45.9 ± 102.7	8.1 ± 59.5	40.5 ± 70.3	32.4 ± 40.5	140 ± 75.7	240 ± 78.4
Lys 4-1	21.6 ± 48.6	8.1 ± 75.7	24.3 ± 113.5	18.9 ± 102	8.1 ± 48.6	64.9 ± 100	67.6 ± 102.7	–5.4 ± 102
Lys 5-1	–13.5 ± 91.89	–13.5 ± 164.9	–18.9 ± 113.5	–13.5 ± 121	132 ± 62.2	129 ± 83.7	2.7 ± 127	216 ± 97.3
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				Sr-90			
	Sep 94	Jan 95	Apr 95	Jun 95	Sep 94	Jan 95	Apr 95	Jun 95
Lys 1[b]	—[e]	—[e]	—[e]	—[e]	1,567 ± 54	1,783 ± 162	1,567 ± 81.0	1,621 ± 54.0
Lys 2	—[e]	—[e]	—[e]	—[e]	62.2 ± 5.4	159 ± 56.7	110 ± 10.8	124 ± 8.1
Lys 3	—[e]	—[e]	—[e]	—[e]	29.7 ± 2.7	91.9 ± 56.8	48.6 ± 8.1	67.6 ± 5.4
Lys 4	—[e]	—[e]	—[e]	—[e]	1.1 ± 1.2	0.0 ± 35.1	0.3 ± 2.9	1.1 ± 1.8
Lys 5	—[e]	—[e]	—[e]	—[e]	2,486 ± 108	2,675 ± 216	1,621 ± 81.0	2,378 ± 81.1
Lys 1-3[c]	—[e]	—[e]	—[e]	—[e]	7.3E+4 ± 2,703	9,189 ± 540	5.7E+4 ± 2,702	6.2E+4 ± 2,703
Lys 2-3	—[e]	—[e]	—[e]	—[e]	9,729 ± 270	8,918 ± 270	8,918 ± 270.3	8,108 ± 270
Lys 3-3	—[e]	—[e]	—[e]	—[e]	32.4E+4 ± 2.7E+4	—	23.5E+4 ± 2,702	26.4E+4 ± 5,405
Lys 4-3	—[e]	—[e]	—[e]	—[e]	1.6E+4 ± 270	1.3E+4 ± 540	1.4E+4 ± 270	1.4E+4 ± 270
Lys 5-3	—[e]	—[e]	—[e]	—[e]	945 ± 216	—	2.0E+4 ± 270	2.6E+4 ± 810
Lys 1-1[c]	—[e]	—[e]	—[e]	—[e]	1.9E+4 ± 270	5.1E+4 ± 2,701	2.3E+4 ± 540	1.9E+4 ± 540
Lys 2-1	—[e]	—[e]	—[e]	—[e]	1,243 ± 54	2,702 ± 270	973 ± 54.0	1,405 ± 54.0
Lys 3-1	—[e]	—[e]	—[e]	—[e]	703 ± 27.0	757 ± 108	1,459 ± 54.0	2,162 ± 54.0
Lys 4-1	—[e]	—[e]	—[e]	—[e]	17.0 ± 8.1	149 ± 83.7	124 ± 18.9	78.4 ± 16.2
Lys 5-1	—[e]	—[e]	—[e]	—[e]	1,216 ± 54.1	2,324 ± 297	621 ± 27.0	946 ± 54.0
Lys 1-2[d]	—	—	—	—	—	—	—	102 ± 18.9
Lys 2-2[d]	—	—	—	—	—	—	—	51.3 ± 13.5
Lys 3-2[d]	—	—	—	—	—	—	—	45.9 ± 13.5
Lys 4-2[d]	—	—	—	—	—	—	—	-1.4 ± 9.7
Lys 5-2[d]	—	—	—	—	—	—	—	946 ± 54.1

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.

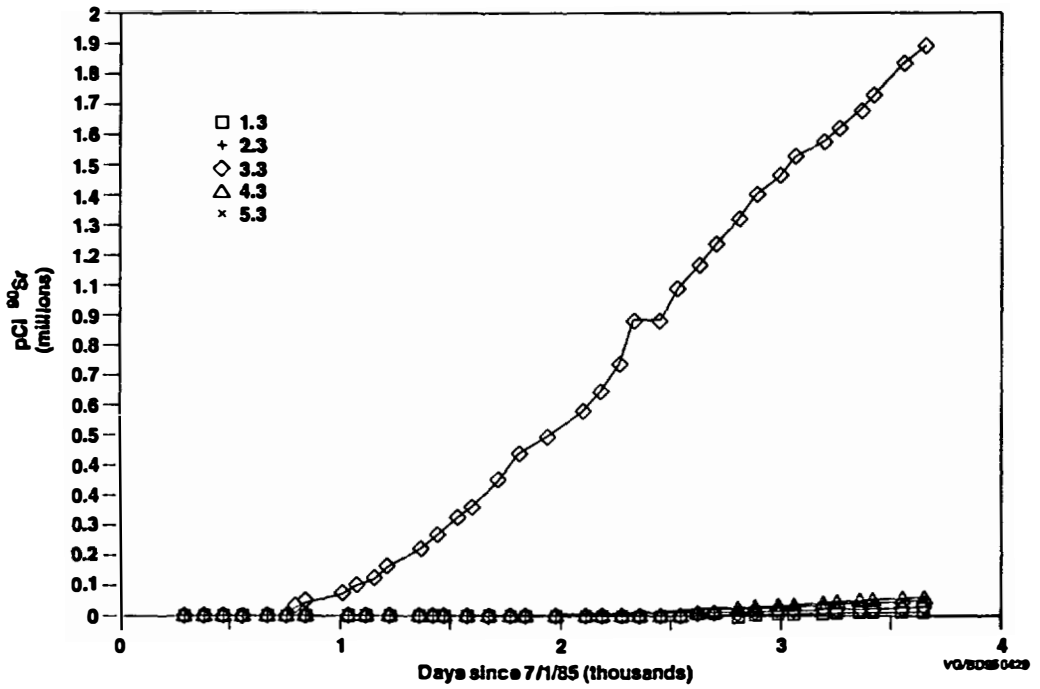

Figure 39. ANL-E cumulative Sr-90 collected in moisture cups number 3.

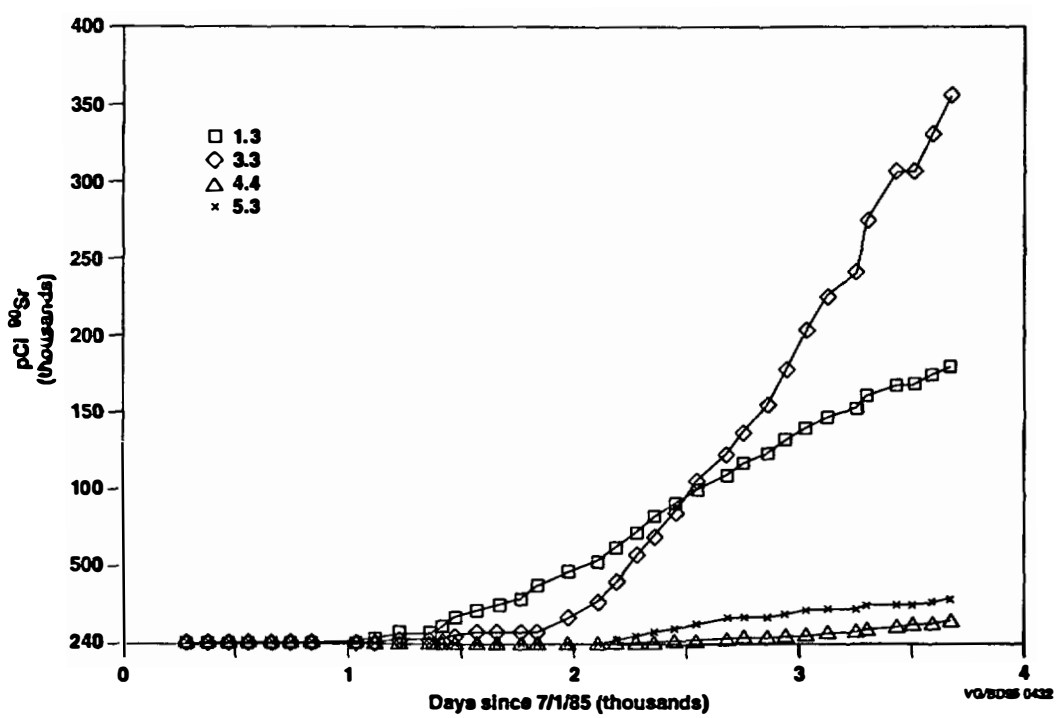

Figure 40. ORNL cumulative Sr-90 collected in moisture cups number 3.

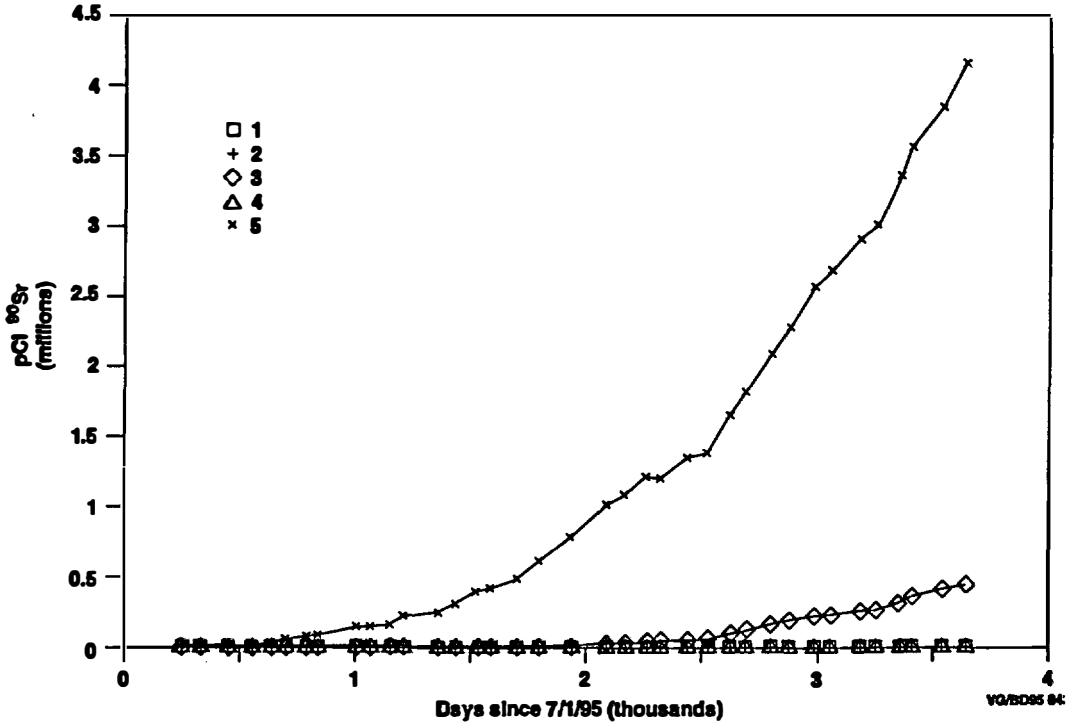

Figure 41. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

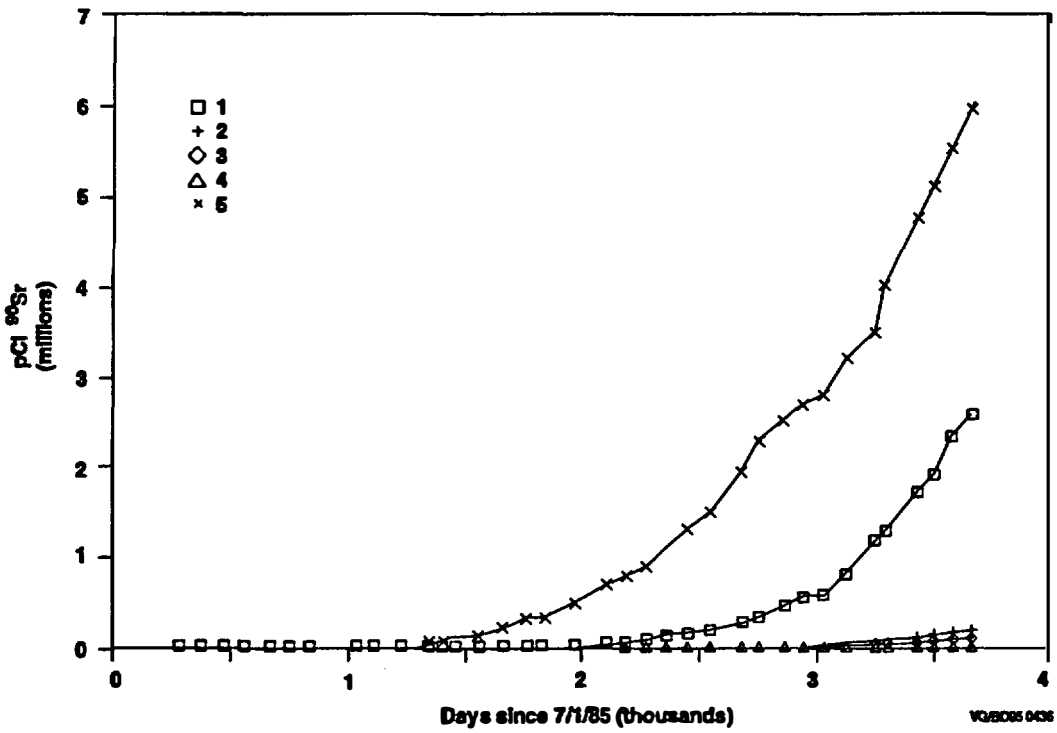

Figure 42. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

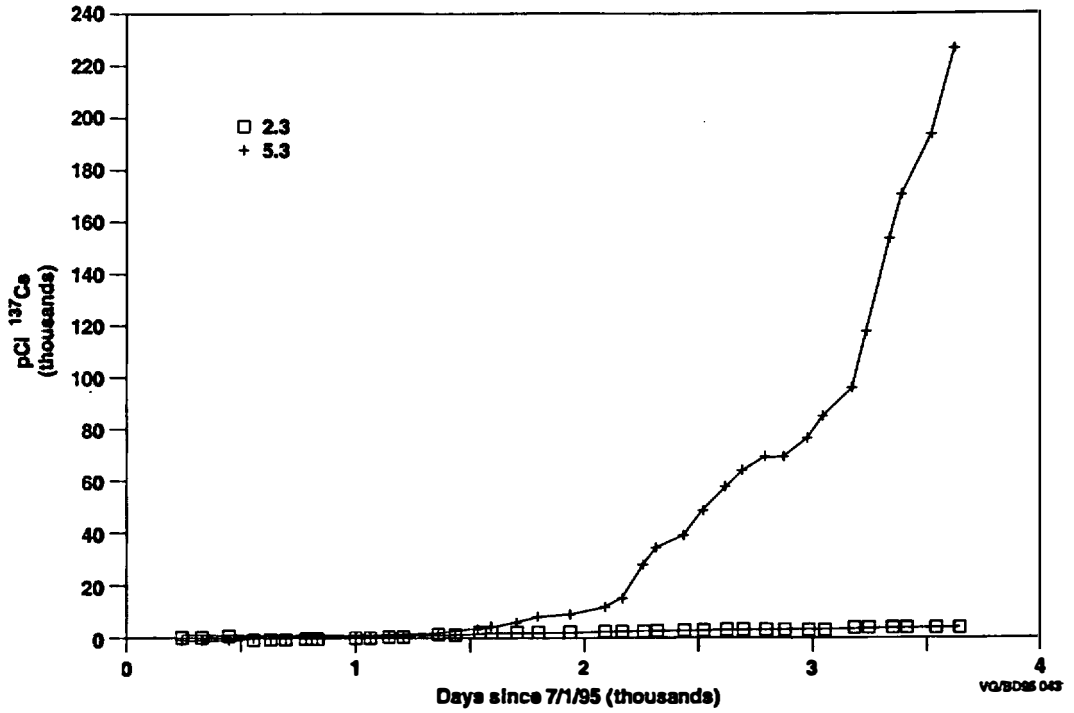

Figure 43. ANL-E cumulative Cs-137 collected in moisture cups number 3.

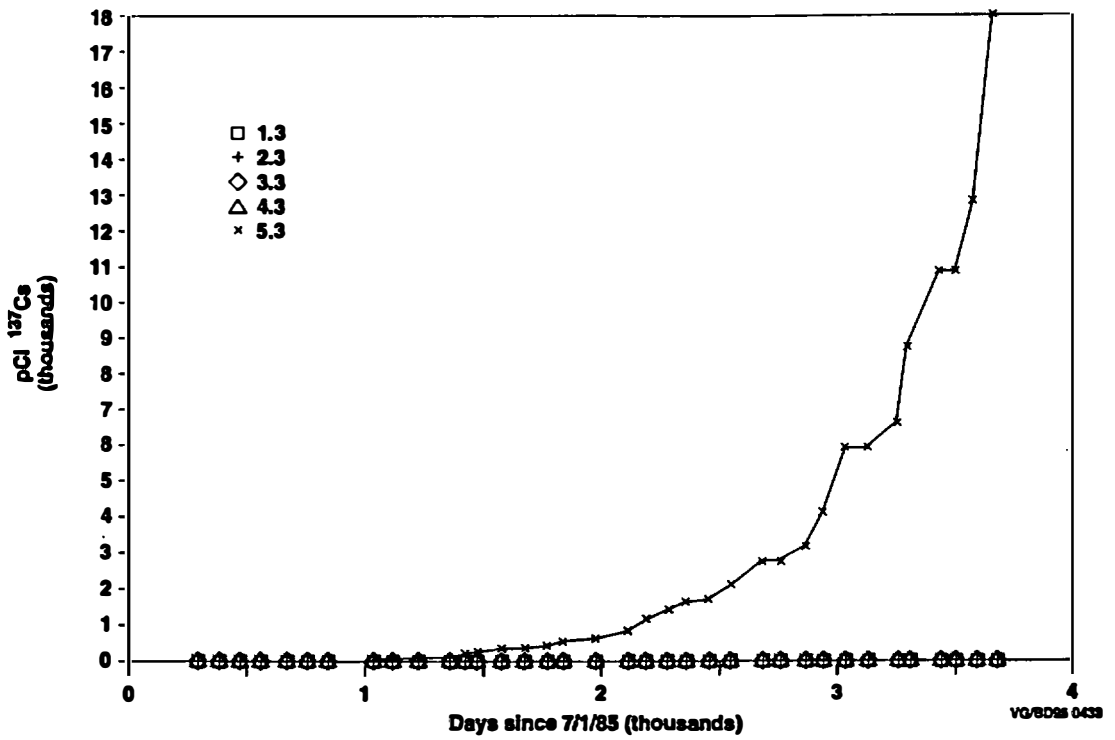

Figure 44. ORNL cumulative Cs-137 collected in moisture cups number 3.

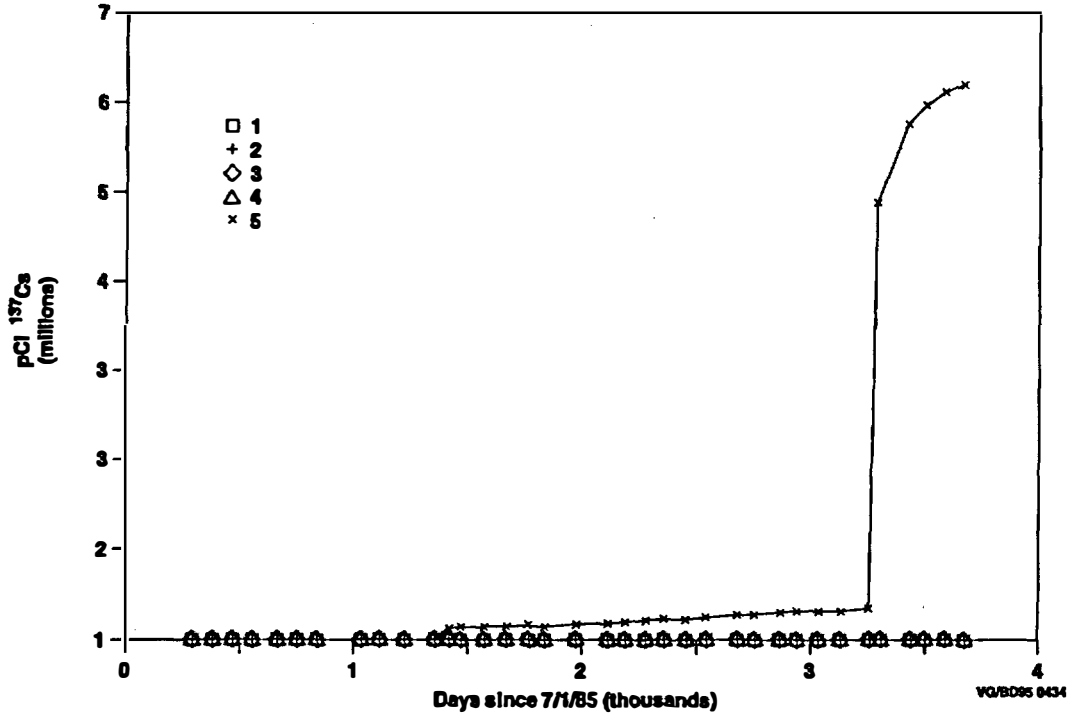

Figure 45. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

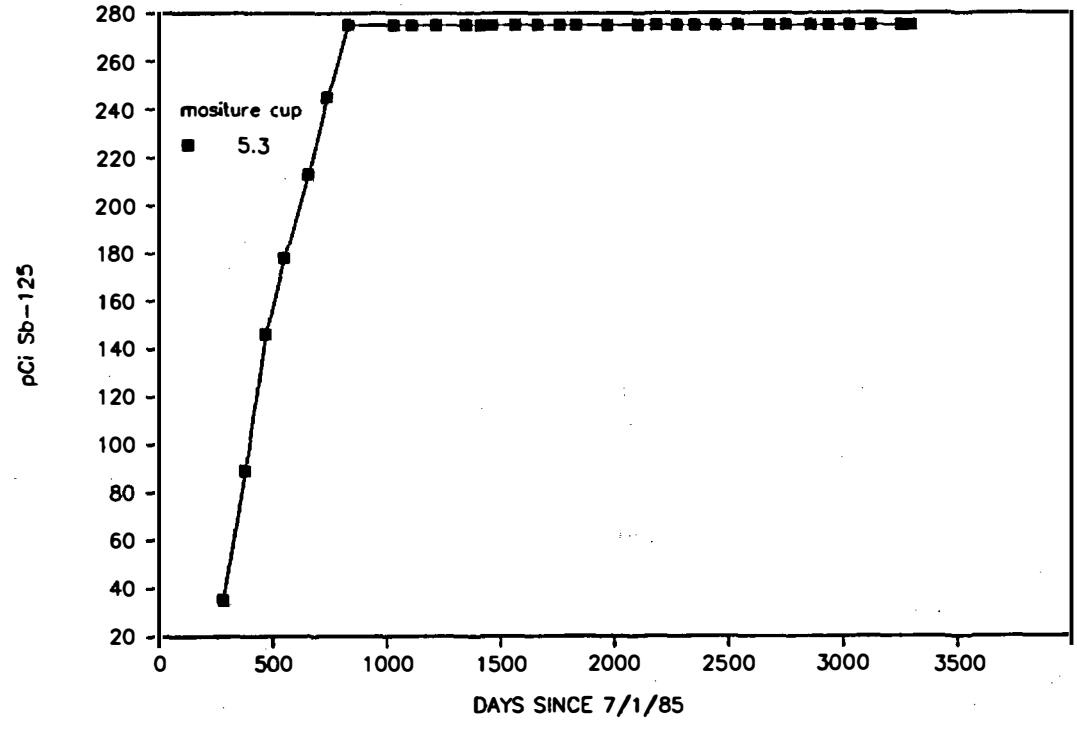

Figure 46. ORNL cumulative Sb-125 collected in moisture cups number 3.

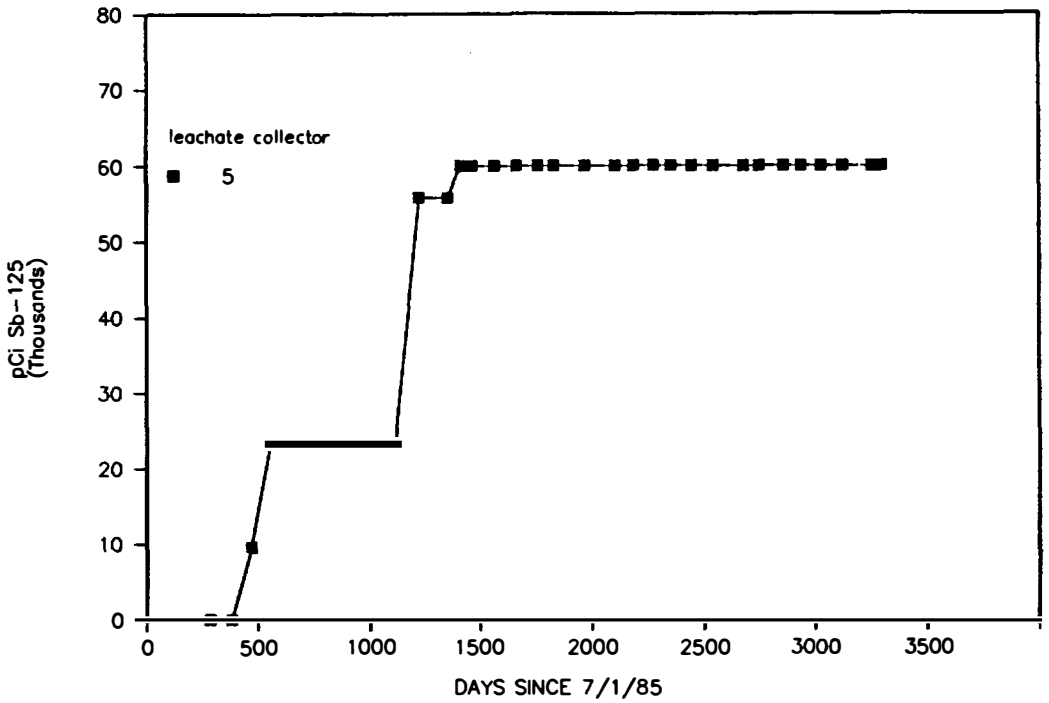

Figure 47. ORNL cumulative Sb-125 collected in lysimeter leachate collectors.

cumulative total of 1,902,175 pCi from 3-3 at ANL-E (an increase of 17% over last year) (Figure 39) and 357,308 pCi from 3-3 at ORNL (30% increase over last year), which continues to increase beyond ORNL 1-3 (Figure 40). 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 (20%) continues in ORNL 5-3 as well as a 62% increase into ORNL 4-3 (a decrease from the 100% of the last 2 years) (Figure 40). The above data show that significant quantities of Sr-90 continue to be transported from the waste form samples.

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 continue to be at least one order of magnitude larger than the largest cumulative release from a soil lysimeter at ANL-E (Figure 41). This is comparable to the previous years' findings (References 11 and 13 through 20). There have been substantial releases of Sr-90 from the soil-filled lysimeters at ORNL. Strontium-90 content in leachate from lysimeters 1 through 3 has increased over 100% (Figure 42). A larger release of Sr-90 in ORNL 1 (100%) increased the total to 2,596,758 pCi and maintains it in the same order of magnitude as ORNL 5 (5,980,597 pCi). There continued to be an increase in the total cumulative quantity of Sr-90 released in the leachate water in all lysimeters at both sites except ANL-E 1 and 2 (Tables 6 and 7).

For ORNL lysimeters 1, 2, and 4, the percent of total inventory of the nuclide released in leachate water continued to be greater than that in the cups (Table 8). These data follow a trend seen over the past 66 months and make it appear that a pulse of Sr-90 could be moving through the soil columns of those three 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 over an order of magnitude difference for both locations).

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).

Table 8. Percent of total Sr-90 and Cs-137 inventory per lysimeter released to moisture cups and leachate water through July 1995.

Lysimeter number	Solidification agent	Percent total inventory Sr-90				Percent total inventory Cs-137			
		Moisture cups		Leachate water		Moisture cups		Leachate water	
		ANL-E	ORNL	ANL-E	ORNL	ANL-E	ORNL	ANL-E	ORNL
1	Cement	1.4E-4	9.7E-4	0.4E-4	143.0E-4	—[a]	—	—	1.7E-6
2	Cement	4.4E-4	7.8E-4	0.7E-4	46.0E-4	0.2E-6	—	—	0.1E-6
3	VES	69.4E-4	13.0E-4	16.1E-4	4.5E-4	—	—	—	1.4E-6
4	VES	14.7E-4	3.3E-4	2.2E-4	4.8E-4	—	—	—	0.4E-6
5	Cement	2.7E-4	8.8E-4	225.0E-4	1,812.0E-4	71.9E-6	1.3E-6	—	434.0E-6

a. Percent released is essentially equal to zero.

Perhaps this represents a difference in how the environment at the two sites affects the movement of Sr-90 being released from the waste form samples. 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 7.5 to 8.0 times that of ANL-E (Table 8). From a waste form standpoint, the cement waste form samples 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 form samples than occurs at ORNL (Table 8).

Gamma-producing nuclides continue to occur with regularity at both sites. ANL 2-3, below a cement waste form sample containing large amounts of Cs-137, continues to receive significant quantities of Cs-137 (Table 6; Figure 43). Since Cs-137 began appearing in ANL 5-3, the quantity of this nuclide has continued to dramatically increase in each of the sampling periods. This year, there was an increase of 49% as compared to a 51% increase last year (Figure 43). ANL 2-3 received sporadic releases of Cs-137 this year. However, 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 44) and have continued in subsequent samplings for a total of 17,861 pCi, which is another 100% increase over the previous year. Detectable amounts of Cs-137 have been consistently found in leachate water from ORNL-5 and sporadically in the other ORNL waters (i.e., lysimeter 4 this year) (Figure 45 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 44 and 45). This year, there was an increase of 28% (as compared to a 1,567% increase last year) for a total cumulative release of 6,215,226 pCi (Table 7). It appears that a pulse of Cs-137 has moved through the system.

For the sixth year in a row, Sb-125 has not been found in ORNL-5 leachate water, as shown in Figures 46 and 47. Also, this is the seventh 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 form samples (Reference 12), a greater recovery of Sr-90 has continued to occur 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 3-3 and possibly ORNL 2-3 (Table 7). On the other hand, this nuclide continues to consistently occur in ORNL 5-3 (Figure 44) and in the leachate collector of the ORNL-5 lysimeter (Figure 45). As noted above, Cs-137 has also occurred in the moisture cups of ANL-E lysimeters 2 and 5. However, 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 37 and 38). 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 form samples 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). It could be assumed that nuclide leaching from these waste form samples 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 39 and 40 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 form samples (range of 176 to 2,874% 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 (293 to 4,356%). So it appears that there is a general trend for more Sr-90 to be removed from the higher loaded waste form samples with a subsequent movement through the soil column.

The assumption of a uniform percent release of Sr-90 from the waste form samples, 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 form samples (124 to 472%), while two of the five have the higher Sr-90 released to the leachate collector water (311 and 732%).

A greater percentage of Sr-90 continues to be found in ANL 3-3 and ANL 4-3 (which both contain VES waste form samples) 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 (4 and 7 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 616 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 0.181% 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 (13 to 402 times higher).

This is the third 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 totaled 355 pCi. While most of the soil-filled lysimeter cups have seen only traces, this year cup 4-2 had a total of 108 pCi. 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 form samples.

Recovery of Sr-90 in cups 3 at ANL-E is higher for those lysimeters containing VES waste form samples than for those with cement waste form samples. Recovery of Sr-90 in the cups at ORNL is comparable for those lysimeters containing the cement waste form samples and those containing VES waste form samples. In the leachate collectors, more Sr-90 is seen in the ANL-E lysimeters containing VES waste form samples, while more is seen in those ORNL lysimeters holding cement waste form samples. These data continue to indicate that cement and VES have generally 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 (CFRs) of radionuclides to leachate collector waters from 10 years of field testing EPICOR-II waste form samples in lysimeters to releases from bench-leach-testing similar waste form samples in demineralized and sea waters as reported in References 10 and 15. Releases observed in the lysimeters are at least two orders of magnitude less for Sr-90 in soil and at least four orders of magnitude less for Cs-137 in soil. It is interesting to note that release of Sr-90 in the sand-filled lysimeters is only one or two orders of magnitude less than bench-test results with demineralized water.

CFRs have been plotted with time in Figures 48 through 54 for both cups number 3 and leachate collector waters. These plots are similar to cumulative release curves of Figures 39 through 47, where cup 3 with VES waste form samples at both sites having the highest measured Sr-90 CFR (Figures 48 and 49). Cup 4-3 at ANL-E, below another VES waste form sample, is the only other ANL-E standout while all of the ORNL cups 3

Table 9. Cumulative fractional releases from lysimeter field testing through June 1995 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	Cement[a]	Sr-90	7.8E-2	—	—	—
Bench,[a] INEL	7	Cement[a]	Cs-137	9.0E-2	—	—	—
Bench,[a] INEL	7	VES[a]	Sr-90	4.5E-2	—	—	—
Bench,[a] INEL	7	VES[a]	Cs-137	4.0E-2	—	—	—
Bench,[a] INEL	7	Cement	Cs-137	4.8E-2	9.0E-2	—	—
Bench,[a] INEL	24	Cement	Cs-137	2.3E-2	2.6E-2	—	—
Bench,[a] INEL	7	VES	Cs-137	2.1E-3	6.4E-2	—	—
Bench,[a] INEL	24	VES	Cs-137	3.0E-4	1.3E-2	—	—
Field, ANL-E	7	Cement	Sr-90	—	—	3.7E-7	2.3E-4
Field, ANL-E	24	Cement	Sr-90	—	—	7.0E-7	—
Field, ANL-E	7	VES	Sr-90	—	—	1.6E-5	—
Field, ANL-E	24	VES	Sr-90	—	—	2.2E-6	—
Field, ORNL	7	Cement	Sr-90	—	—	1.4E-4	—
Field, ORNL	24	Cement	Sr-90	—	—	4.6E-5	1.8E-3
Field, ORNL	7	VES	Sr-90	—	—	4.5E-6	—
Field, ORNL	24	VES	Sr90	—	—	4.8E-6	—
Field, ORNL	7	Cement	Cs-137	—	—	1.7E-8	—
Field, ORNL	24	Cement	Cs-137	—	—	1.4E-9	4.3E-6
Field, ORNL	7	VES	Cs-137	—	—	1.4E-8	—
Field, ORNL	24	VES	Cs-137	—	—	3.8E-9	—

a. Waste form samples were irradiated before test.

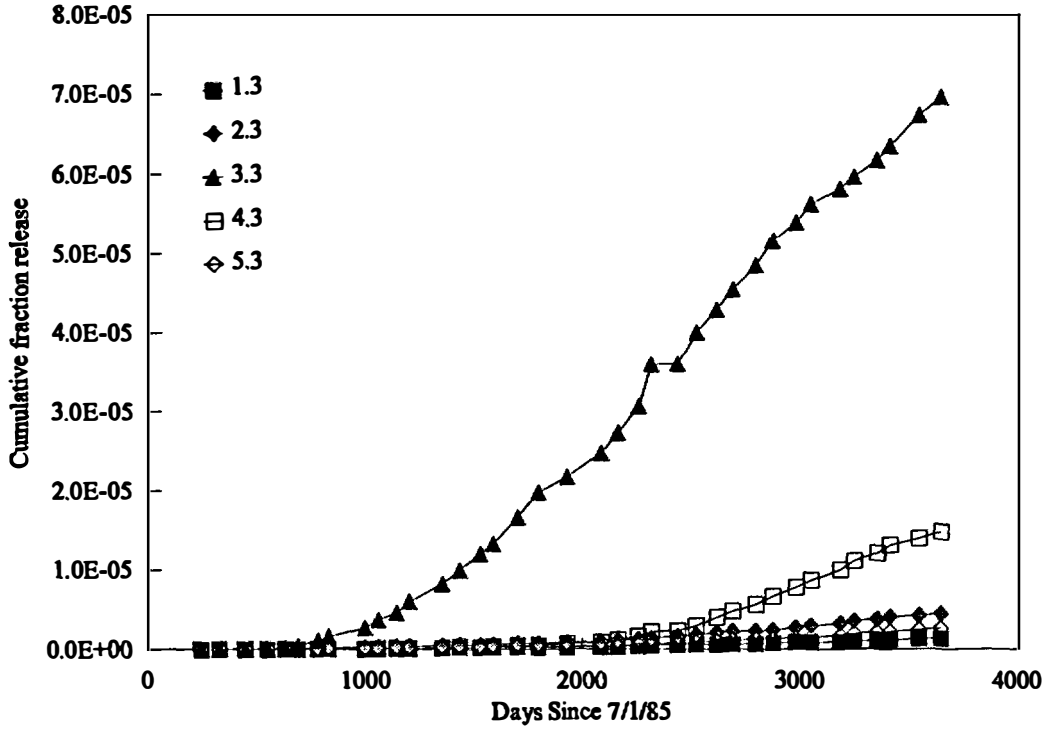

Figure 48. ANL-E cumulative fraction release of Sr-90 collected in moisture cups number 3.

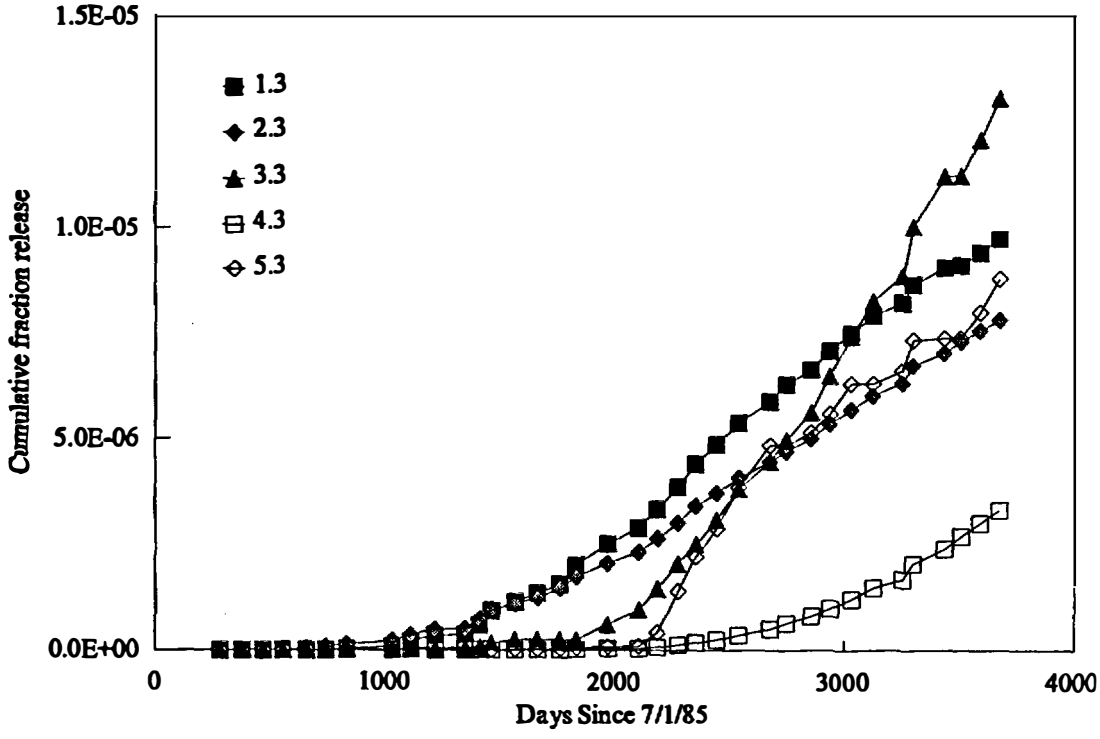

Figure 49. ORNL cumulative fraction release of Sr-90 collected in moisture cups number 3.

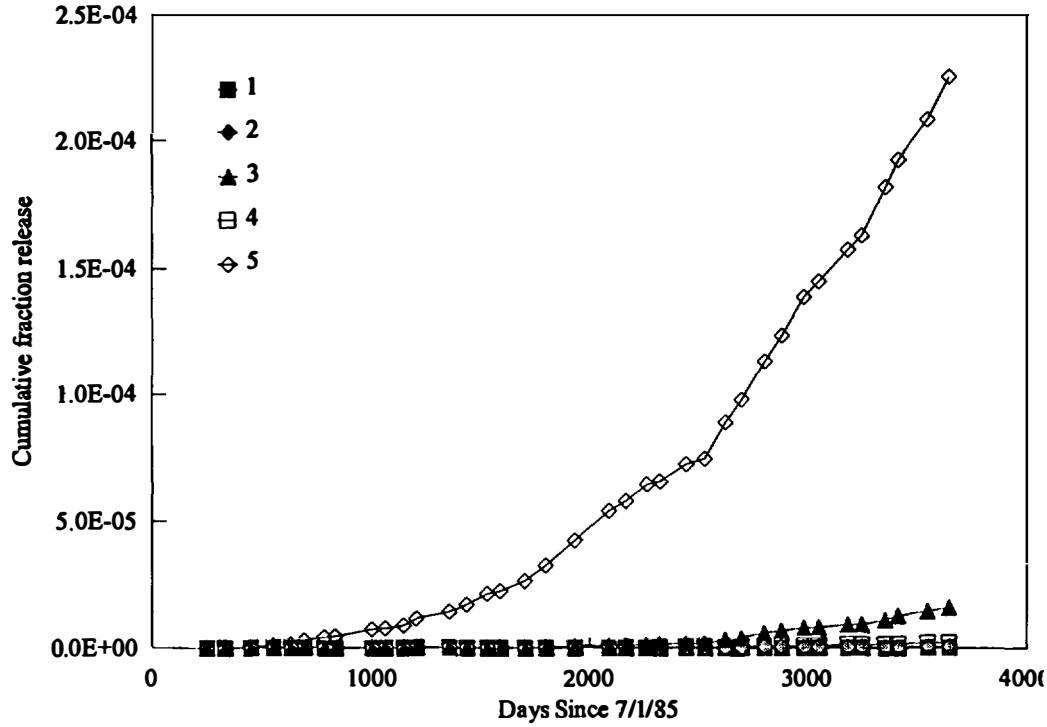

Figure 50. ANL-E cumulative fraction release of Sr-90 collected in lysimeter leachate collectors.

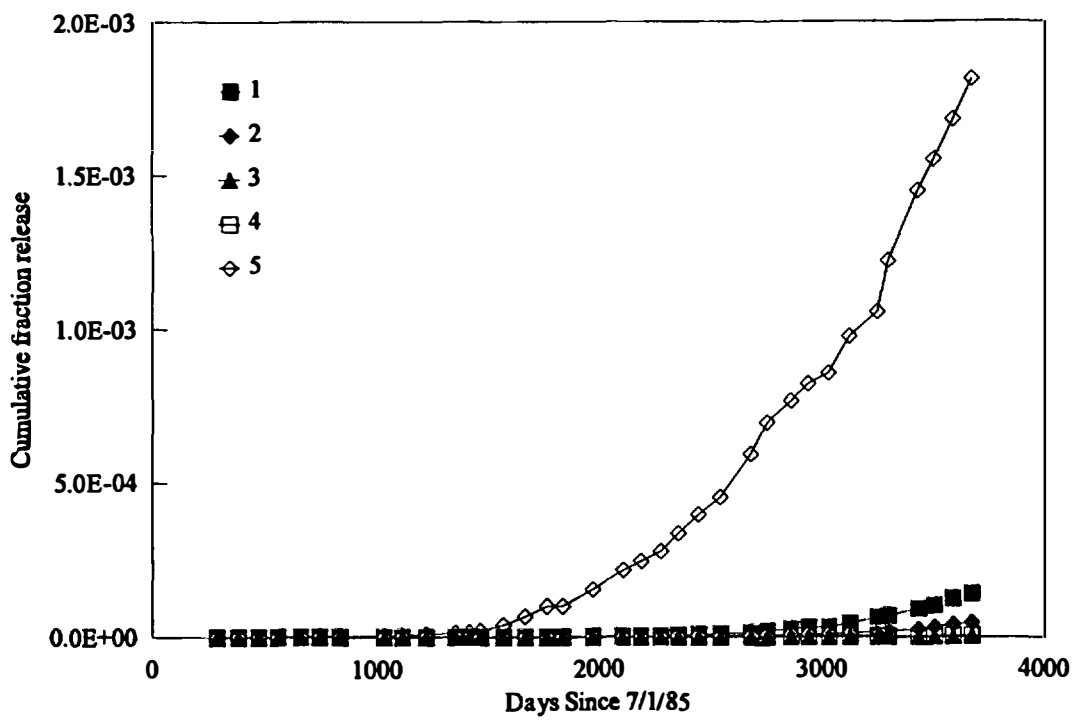

Figure 51. ORNL cumulative fraction release of Sr-90 collected in lysimeter leachate collectors.

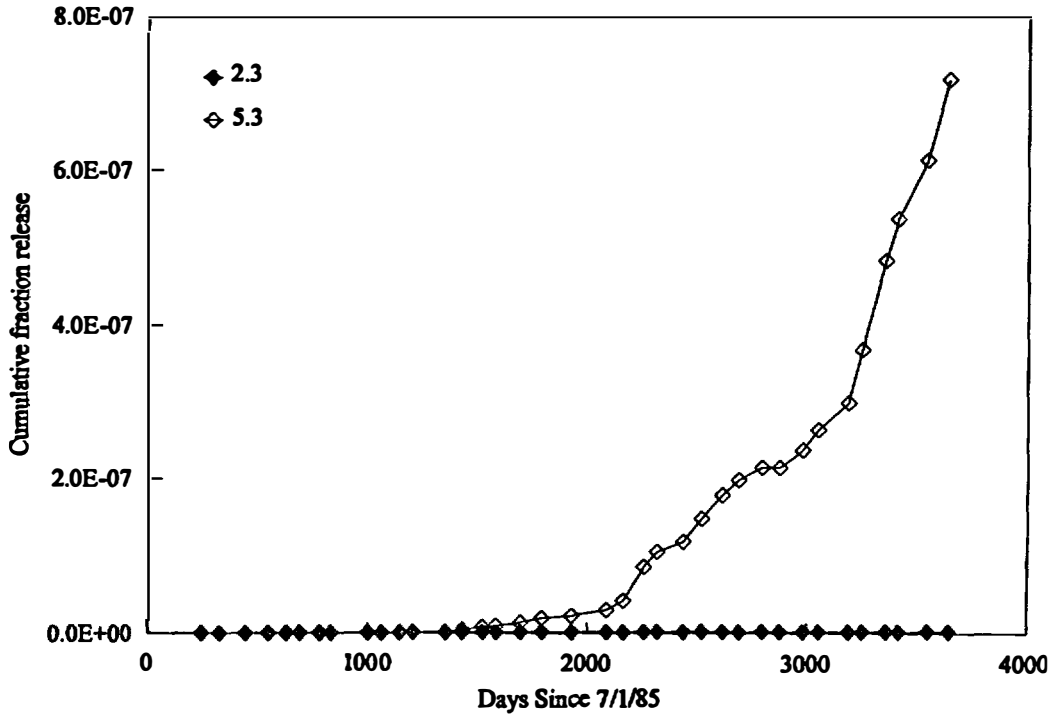

Figure 52. ANL-E cumulative fraction release of Cs-137 collected in moisture cups number 3.

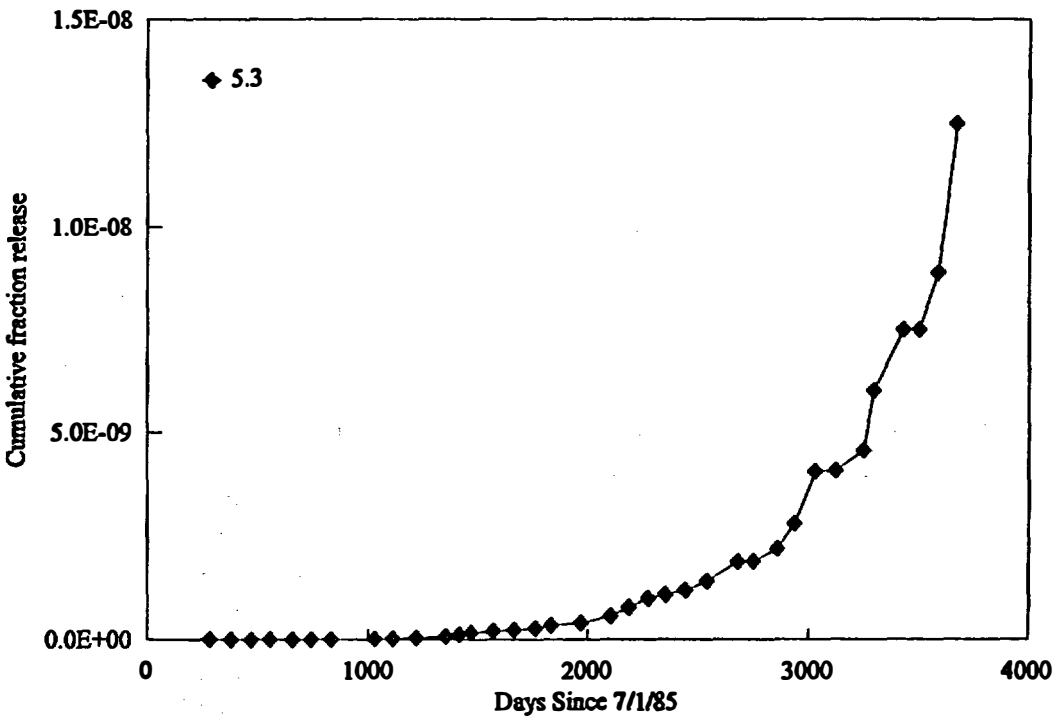

Figure 53. ORNL cumulative fraction release of Cs-137 collected in moisture cup number 3.

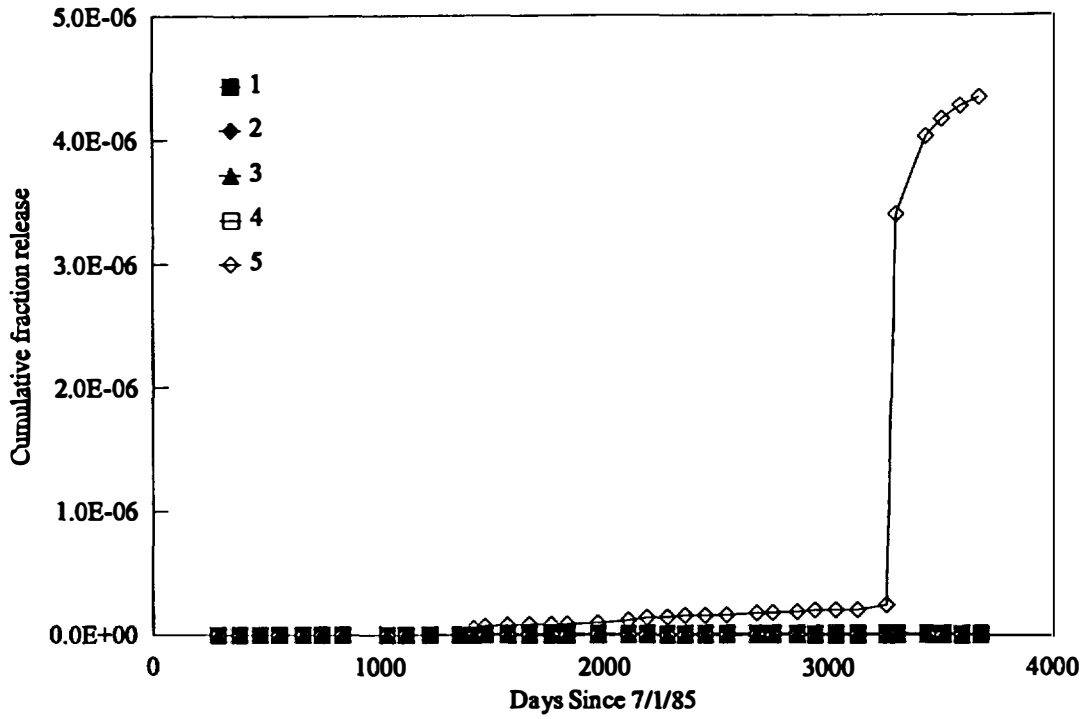

Figure 54. ORNL cumulative fraction release of Cs-137 collected in lysimeter leachate collectors.

show significantly high CFRs. There is little difference between cumulative releases and CFRs to collectors (see Figures 41 and 42 compared to 50 and 51), with sand-filled lysimeter collectors receiving much higher CFRs than soil-filled units (as seen in Table 9). The Cs-137 plots of CFR exhibit similar features to those of cumulative release (Figures 52 through 54 compared to 43 through 45), with the exception that only ORNL cup 5-3 is presented from that site. At the present rate of increase (Figures 48 through 54), these CFRs will be of similar magnitude in a few 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. Surface 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 the wall, and the other was collected from the center of the lysimeter directly above the buried waste form (located approximately 100 cm below the sand surface). These sand cores were sectioned into 5-cm segments. Radiocesium and strontium activity were measured for each segment.

The analyses show that Cs-137 and Cs-134 were present throughout the length of the center core (Table 10). Cesium-137 was also found in all segments of the side core, but Cs-134 was only found in the upper half of that core. Cesium-134 is an

Table 10. Cesium (Cs) and strontium (Sr) analyses for sand core segments from the center (Core C) and side (Core S) and root fragments from the center of ORNL lysimeter 5 collected on January 31, 1994.

Segment number		Depth (cm)	Sand									Plant Root	
			Segment dry weight (g)		Cs-137 (pCi/g)		Cs-134 (pCi/g)		Sr-90 (pCi/g)	Ratio[b] Cs-137/Cs-134		Ratio[e] Cs-137/Sr-90	Sample weight (g)	Cs-137 (pCi/g)
Core C	Core S		Core C	Core S	Core C	Core S	Core C	Core S	Core C	Core C	Core S	Core C		
—[a]	1	76.5-71.5	—	126.11	—	0.26	—	ND[c]	—	—	d		—	—
1	2	71.5-66.5	100.68	139.80	598.1	0.20	1.5	ND	1.5	399		450	0.0134	18,900
1	—	71.5-66.5	17.08	—	704.5	—	1.6	—	—	448			—	—
1	—	71.5-66.5	17.16	—	660.8	—	1.6	—	—	425			—	—
2	3	66.5-61.5	118.92	120.32	1,303.4	0.22	1.8	ND	3.5	724		650	0.0172	20,660
2	—	66.5-61.5	17.48	—	2,241	—	2.3	—	—	969			—	—
2	—	66.5-61.5	20.37	—	1,550	—	1.8	—	—	882			—	—
3	4	61.5-56.5	121.53	131.40	356.7	0.19	0.7	ND	2.0	484		200	0.0301	20,480
3	—	61.5-56.5	19	—	400.7	—	0.8	—	—	498			—	—
3	—	61.5-56.5	14.85	—	376	—	0.7	—	—	517			—	—
4	5	56.5-51.5	115.25	109.76	490.2	0.24	1.1	ND	2.1	447		300	0.0234	22,540
5	6	51.5-46.5	117.07	115.29	403.3	0.17	0.8	ND	2.7	492		200	0.0216	27,520
6	7	46.5-41.5	125.28	141.24	1,594	0.19	3.2	ND	7.6	491		300	0.0224	27,360
7	8	41.5-36.5	129.06	113.21	37,283.1	0.40	30.4	ND	14.1	466		1,000	0.0220	81,970
8	9	36.5-31.5	121.14	124.99	551.2	1.14	1.4	ND	1.5	404		400	0.0302	13,620
9	10	31.5-26.5	116.32	117.30	866.6	38.9	2.3	0.08	3.5	376	467	300	0.0196	10,150
10	11	26.5-21.5	122.86	135.38	5,484.2	6.1	10.9	ND	7.6	475		750	0.0463	21,580
11	12	21.5-16.5	117.94	108.01	2,032.4	2.6	4.4	ND	16.0	458		200	0.0256	5,990
12	13	16.5-11.5	125.78	104.74	1,513	3.5	3.6	ND	0.5	423		3,400	0.1049	3,850
13	14	11.5-6.5	94.99	117.22	711.7	9.0	1.8	0.02	0.2	390	557	4,400	0.0615	5,940
14	15	6.5-0	150.30	142.25	715.2	53.6	1.6	0.12	0.6	451	462	1,200	0.3105	8,570

a. No measurement was taken at this location.
b. Theoretical ratio of Cs-137/Cs-134 = 399.
c. ND = none detected.
d. Blank indicates not enough information available to calculate ratio.
e. Theoretical ratio of Cs-137/Sr-90 = 440.

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 55 indicate that the ratio in all segments of the center core fall close to this value, except for segment 2 (66.5 to 61.5 cm in depth). This confirms that the cesium seen throughout the length of the sand core is a result of upward vertical transport from the waste form sample.

There are three peaks seen in the cesium content (Figure 55): one at 24 cm, a large peak at 39 cm, and a smaller peak at 64 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 56. 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. 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 10 and Figure 56). 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.

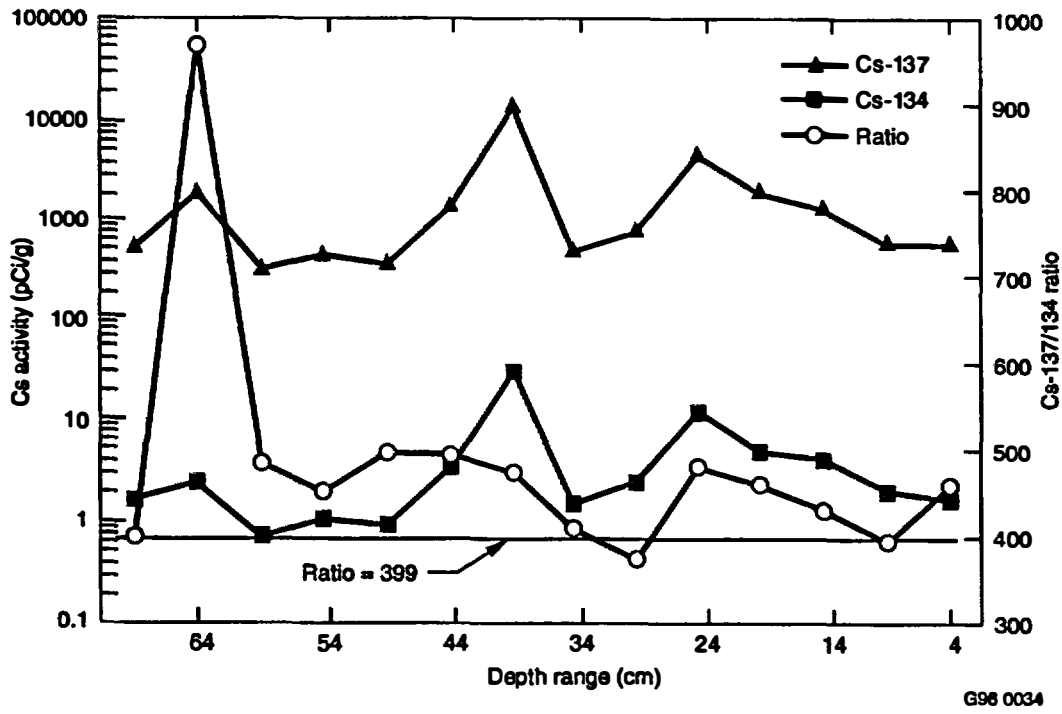

Figure 55. Cesium analyses of the center sand core from ORNL lysimeter 5.

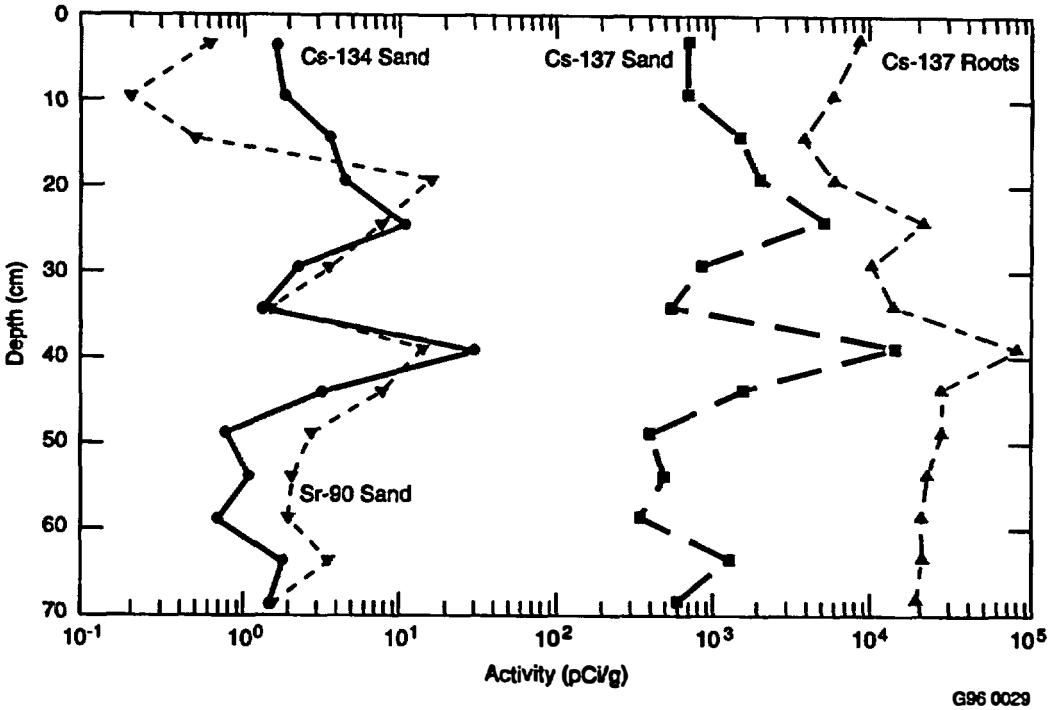

Figure 56. Cesium-137 and Sr-90 associated with plant roots and sand from the center core taken from ORNL lysimeter 5.

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 Figure 57 presents this ratio versus depth. 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 evaporation enhanced by transpiration through 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.

A comparison of the Cs-137 concentrations in the center and side cores (Figure 58) shows that Cs-137 concentrations are much lower at the edge of the lysimeter as might be expected. Those areas are dependant on dispersion as well as evaporation to move the nuclides to them. The obvious peaks of concentration in the center core are less apparent and appear to be delayed about 5 cm.

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.

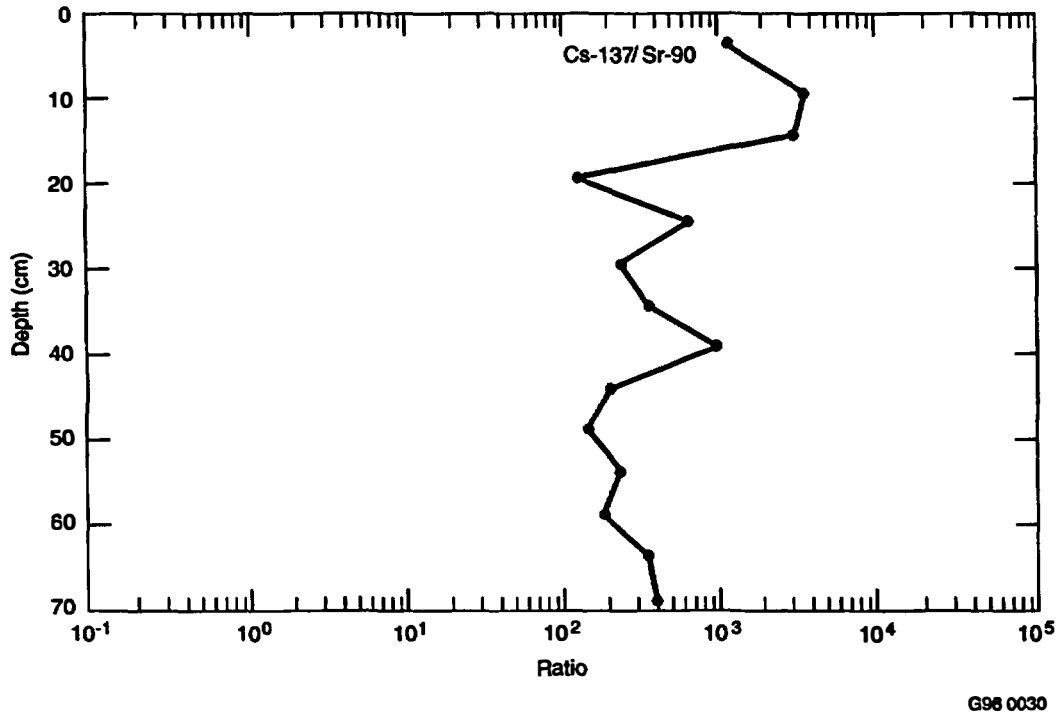

Figure 57. Ratio of Cs-137 to Sr-90 from analysis of center sand core from ORNL lysimeter 5.

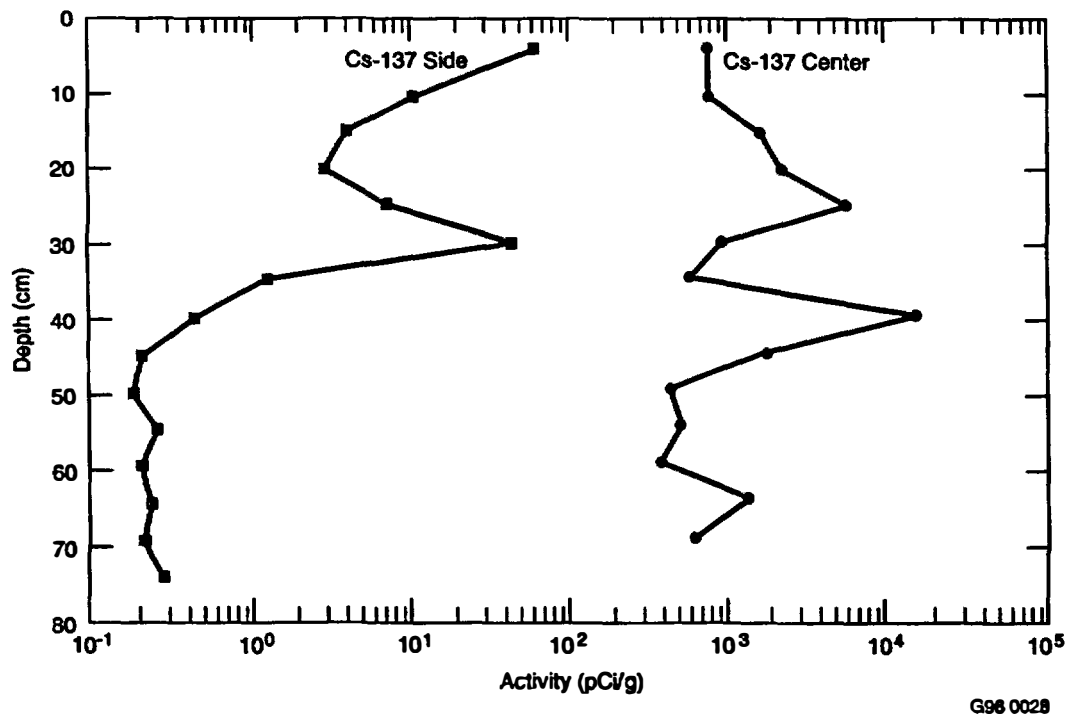

Figure 58. Comparison of Cs-137 analysis from center versus side sand cores from ORNL lysimeter 5.

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 45). 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 four occurrences in which the lysimeters have accumulated water that moved high into the soil column, above the waste form samples. This happened as a result of unusually heavy precipitation events between samplings. It is possible that the high water level caused additional releases of radionuclides, which then moved up with evaporation and became fixed to organic matter at the surface, which prevented reentry. Further analysis of soil cores and flooding data is planned.

Scientists at ORNL performed gamma radiation surveys on samples that were collected from soil lysimeters 1 through 4 for gravimetric moisture analyses in 1993 and 1994. Measurable but insignificant amounts (below 1 pCi/g) of gamma-producing radionuclides were detected. It was 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 10 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 cumulative fractional release rates of radionuclides from waste form samples, the mass balance of released constituents, the solubility of radionuclides in a site-specific geochemical system, as well as the retardation and 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, and hydraulic conductivity 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 11. The data could be used in such codes as PATHRAE,[23] PRESTO,[24] and others to predict the stability of waste form samples 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 (a control) 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

Table 11. 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

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 form samples 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 form samples 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 form samples, and from the lysimeter leachate collector, located 61 cm below the bottom of the waste form samples. 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 form samples. DUST is a one-dimensional code that can model release by a finite difference method or by a mixing cell cascade approach, and has the ability to simultaneously model three different types of release mechanisms: diffusion, dissolution, and surface rinse. The mixing cell model is limited in that it does not take diffusional release into consideration. Therefore, for these simulations, the finite difference model was selected because it is more flexible and capable of handling a variety of different parameters. A further description of the models in the code is given in References 25 and 26.

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 27 and 28 and by fitting the model predictions to the data. The cumulative leachate activity collected from the lysimeters over the first 10 years of the experiment, which was used to make comparisons to the DUST code predictions, represented 0.18% and 0.023% of the total inventory of Sr-90 in lysimeters 5, at ORNL and ANL-E, respectively. At ORNL, the collected amount represented less than 0.0043% of the Cs-137 inventory in lysimeter 5, while nothing has been collected in ANL-E lysimeter 5 (Table 12).

The cumulative activity collected from the lysimeters is 0.18% of total inventory for Sr-90 and less than 0.005% for Cs-137 (Table 8). 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 form samples. 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 0.0033 Ci of Sr-90 and 1.432 Ci of Cs-137 (Reference 12). The PF-24 waste form at ANL-E had a total initial inventory of 0.0185 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 (partition) 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 59 and 60 are obtained using a

Table 12. Total and collected Ci amounts of Sr-90 and Cs-137 in lysimeters 5 through July 1995.

	Total inventory (Ci)	Amount collected (Ci)	Percent collected
ORNL-5 Cs-137	1.432	6.22E-6	0.0043
ORNL-5 Sr-90	0.0033	5.98E-6	0.1812
ANL-5 Sr-90	0.0185	4.16E-6	0.0225

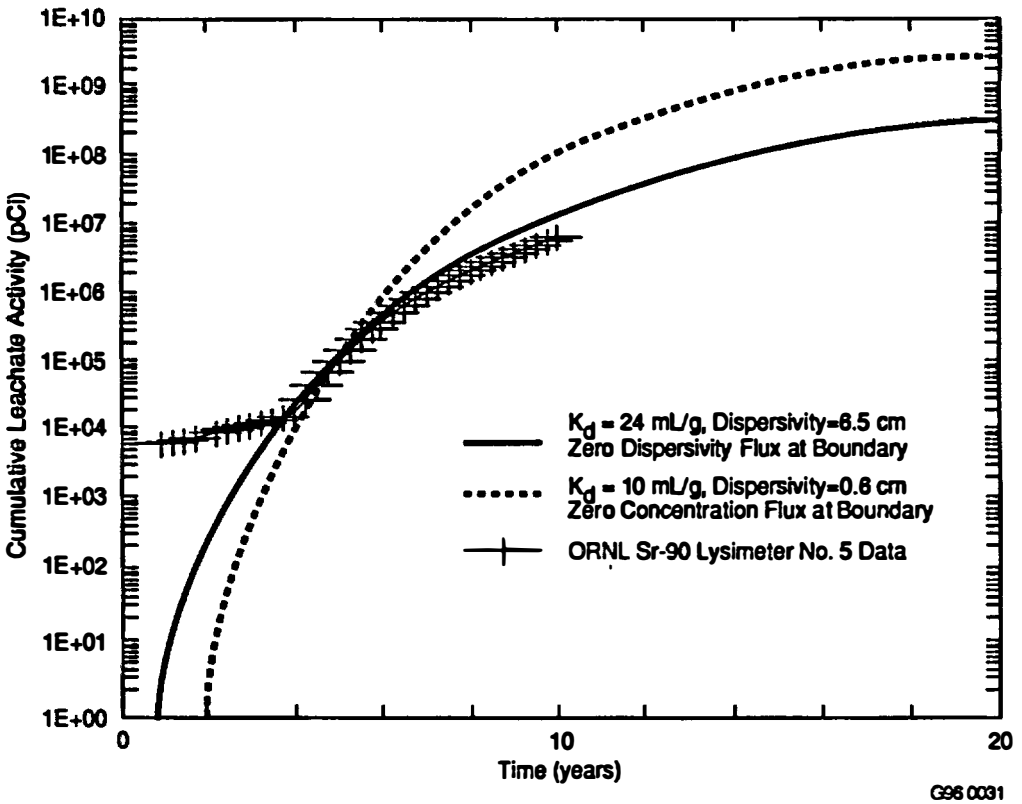

Figure 59. Ten years of data for Sr-90 at ORNL lysimeter 5, compared with two sets of estimated K_d and dispersivity values.

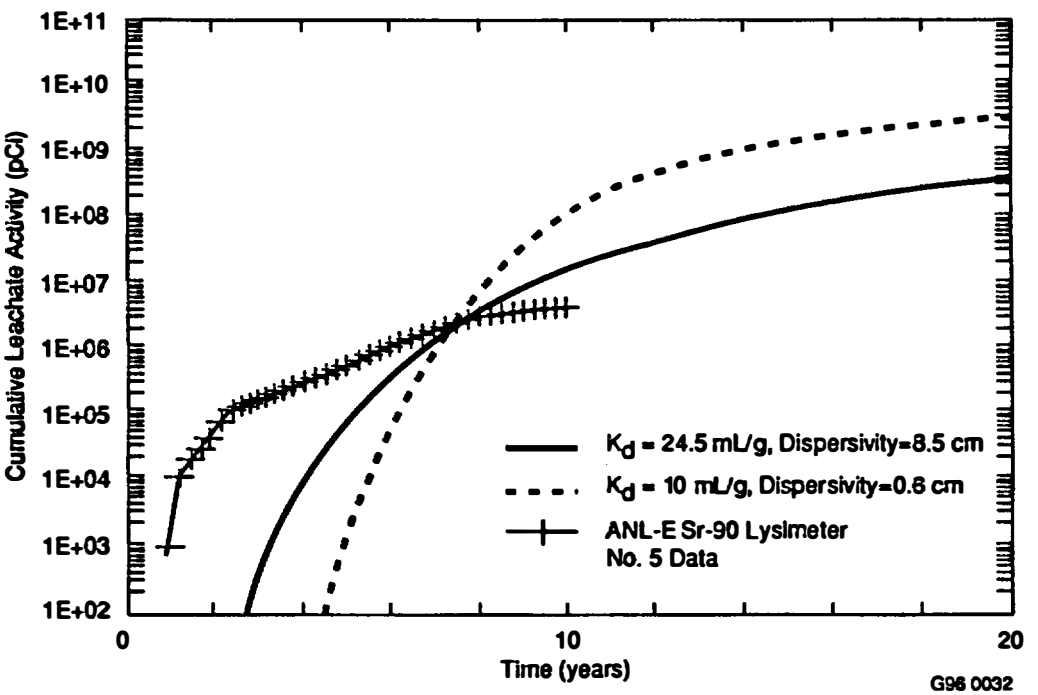

Figure 60. Ten years of data for Sr-90 at ANL-E lysimeter 5, compared with two sets of estimated K_d and dispersivity values for 20 years.

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 59, the actual data for Sr-90 from ORNL lysimeter 5 for 10 years are compared with the DUST code predicted releases in case 1 using zero dispersive flux BC, K_d = 24 mL/g, and dispersivity = 8.5 cm. Also shown are predicted releases of case 2 using zero concentration flux BC, K_d = 10 mL/g, 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 10 years. The lack of measured dispersivity coefficient and K_d further necessitates obtaining fractional release data over a longer term.

Figure 50 shows the actual data for Sr-90 at ANL-E lysimeter 5, which covers a period of 10 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 mL/g and a dispersivity of 8.5 cm. Case 2 has a K_d of 10 mL/g 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 10 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 1995 was analyzed for the major ionic species listed in Table 13. The justification for the choice of ions is also provided in the table. At ANL-E, cups 1, 3, and 5 were sampled on lysimeters 1, 2, 3, and 5, and cups 2, 3, and 5 on lysimeter 4. Cups 1, 3, and 5 were sampled on lysimeters 1, 3, 4, and 5, and cups 1 and 3 on lysimeter 2 in 1995 at ORNL. Data from precipitation samples at ANL-E in 1991 and ORNL in 1989 showed that ionic concentrations in the soil water were not introduced by the precipitation (Reference 11 and 15). It appears that the waste form samples 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 14 and 15 and Figures 61, 62, 63, and 64).

Table 13. 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.

It appears that the cement and VES waste form samples performed similarly at both sites. The ORNL 1995 soil lysimeter data (Table 15 and Figures 63 and 64) closely resemble those of 1988, 1989, 1991, 1992, 1993, and 1994 cation and anion concentrations, and actually show little cup-to-cup variability. ANL-E 1995 data are similar, except for lower calcium, to previous years' data and to ORNL 1995 data when compared in Figures 61, 62, 63, and 64. The inert sand-filled lysimeter results are almost identical except for higher chlorine and NO_3 concentration at ORNL. While these data are interesting, no correlation has yet been made with radionuclide movement.

Table 14. ANL-E chemical speciation results from lysimeter moisture cups 1, 2, 3, and 5, May 1995.

Sample	Solidification agent	Cation					Anion			
		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	80	9.0	12	0.67	45	3.0	0.32	<0.5	31
Lys 1-3		76	10.6	12	0.55	41	2.8	<0.1	<0.5	31
Lys 1-5		73	4.0	13	0.55	31	2.7	<0.1	<0.5	33
Lys 2-1	Cement	63	5.0	10	0.22	32	1.8	<0.1	<0.5	19
Lys 2-3		59	5.4	10	0.30	30	3.0	<0.1	<0.5	13
Lys 2-5		43	2.0	8.0	0.41	17	2.4	0.44	<0.5	14
Lys 3-1	VES	77	3.5	10	0.66	37	2.1	1.2	<0.5	24
Lys 3-3		77	5.4	14	0.44	40	3.0	<0.1	<0.5	22
Lys 3-5		83	2.2	14	0.65	37	3.7	0.78	<0.5	24
Lys 4-2	VES	76	3.6	8.0	0.58	32	2.4	<0.1	<0.5	24
Lys 4-3		84	6.5	9.0	0.79	39	1.7	<0.1	<0.5	32
Lys 4-5		69	2.2	12	0.46	33	1.8	<0.1	<0.5	23
Lys 5-1	Cement	8.0	1.6	8.0	0.91	3.2	3.7	2.0	<0.5	6.0
Lys 5-3		9.0	9.4	28	4.4	4.0	3.8	2.7	<0.5	7.0
Lys 5-5		8.1	1.4	231	1.5	3.1	4.8	5.2	<0.5	9.0

Table 15. ORNL chemical speciation results from lysimeter moisture cups 1, 3, and 5, June and July 1995.

Sample	Solidification agent	Cation					Anion			
		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	32	1.8	18	<2	1.2	0.76	3	<0.5	12
Lys 1-3		31	2	26	2.5	1.1	0.8	4.8	<0.5	11
Lys 1-5		33	0.18	19	<2	0.99	1.2	5.3	<0.5	9.9
Lys 2-1	Cement	36	1.4	18	<2	1.2	<0.1	7.7	<0.5	7.4
Lys 2-3		31	1.7	31	<2	1.0	<0.1	5.8	<0.5	5.4
Lys 2-5		NA	NA	NA	NA	NA	NA	NA	NA	NA
Lys 3-1	VES	28	1.6	20	<2	0.7	0.99	4.2	<0.5	4.6
Lys 3-3		28	2.1	28	<2	0.95	0.9	4.5	<0.5	4
Lys 3-5		1.5	0.33	19	<2	0.47	1.0	3.6	<0.5	2.3
Lys 4-1	VES	7.4	2.2	15	<2	1.1	1.6	4.1	<0.5	15
Lys 4-3		6.9	3.1	8.2	<2	1	1.2	7.7	<0.5	16
Lys 4-5		9.3	0.3	10	<2	1.5	1.5	3	<0.5	16
Lys 5-1	Cement	8.6	0.3	8.3	<2	3.4	6.6	8.7	<0.5	5.8
Lys 5-3		10	1	17	2.6	4.1	NA	NA	NA	NA
Lys 5-5		NA	NA	NA	NA	NA	2.2	10	<0.5	5.3

NA = No sample was taken from this cup in this reporting period.

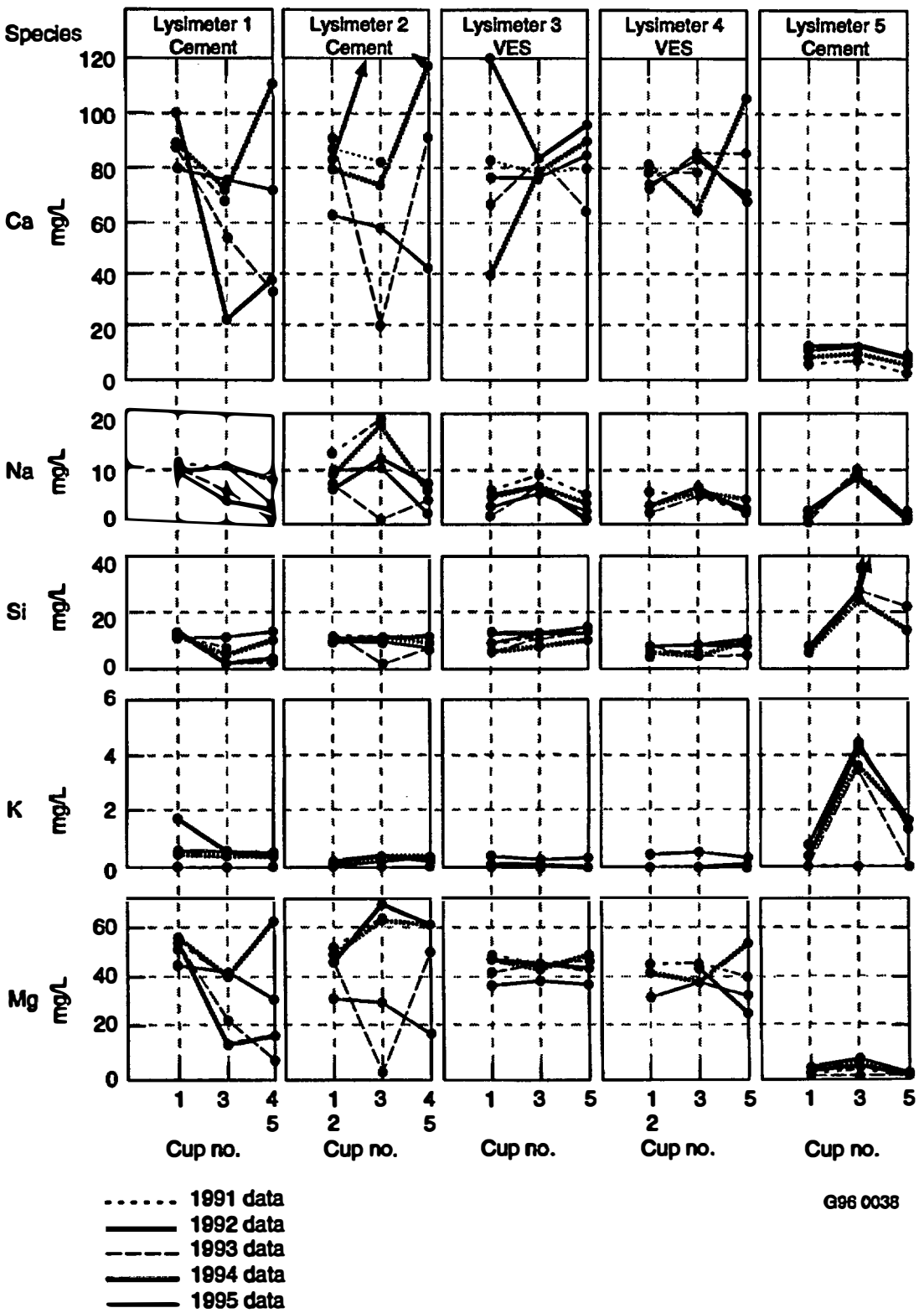

Figure 61. Results of chemical speciation at ANL-E—cations.

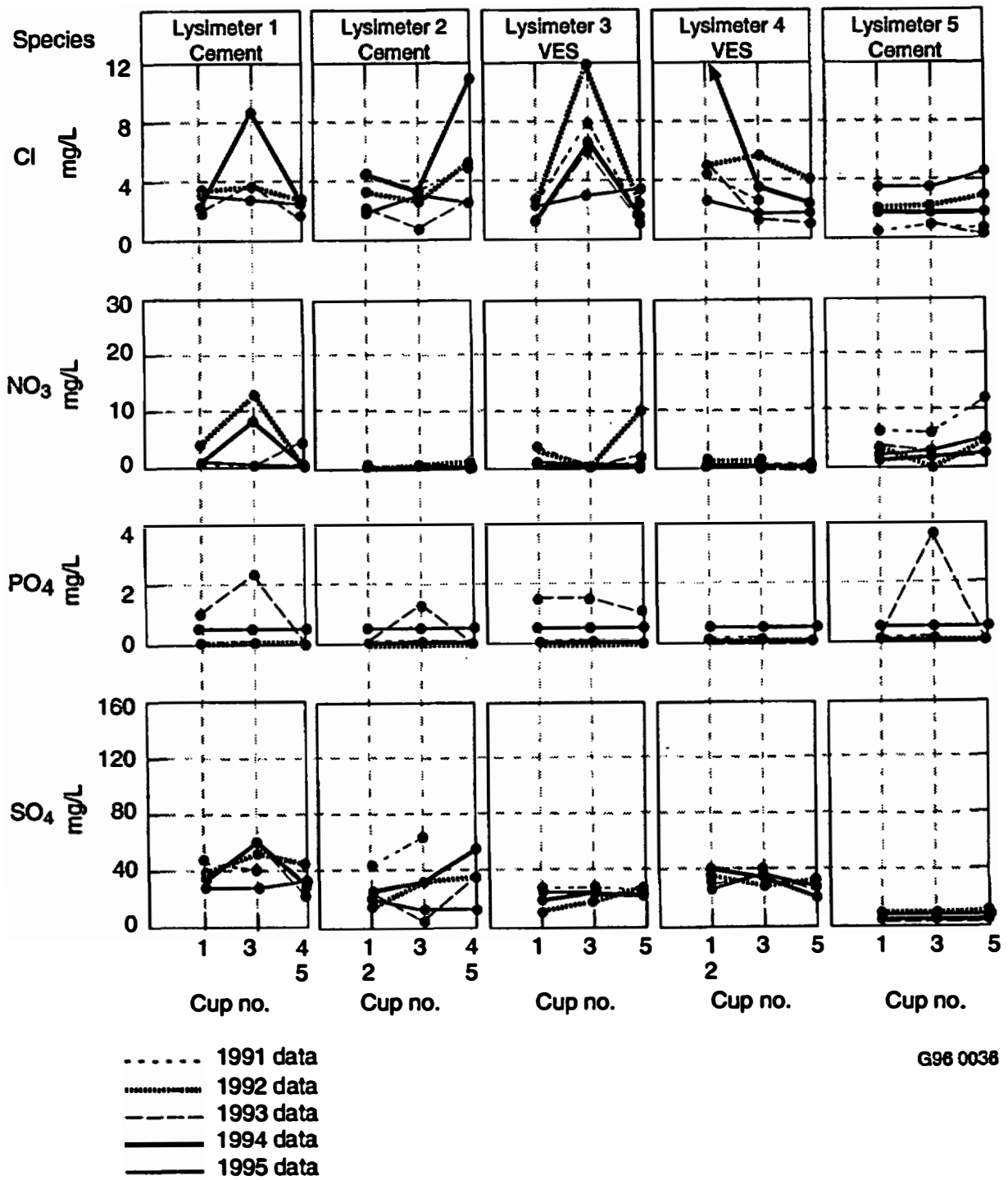

Figure 62. Results of chemical speciation at ANL-E—anions.

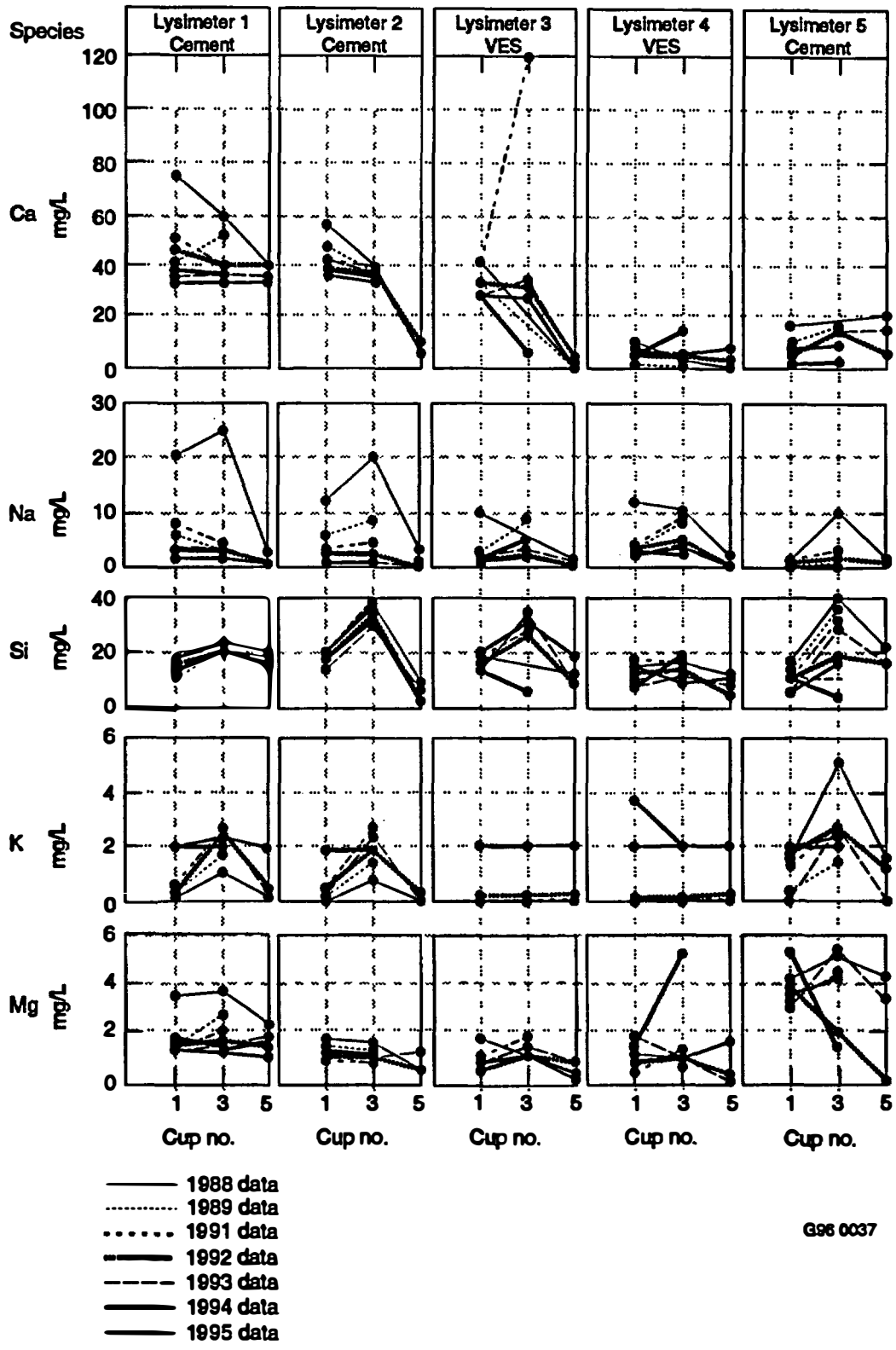

Figure 63. Results of chemical speciation at ORNL—cations.

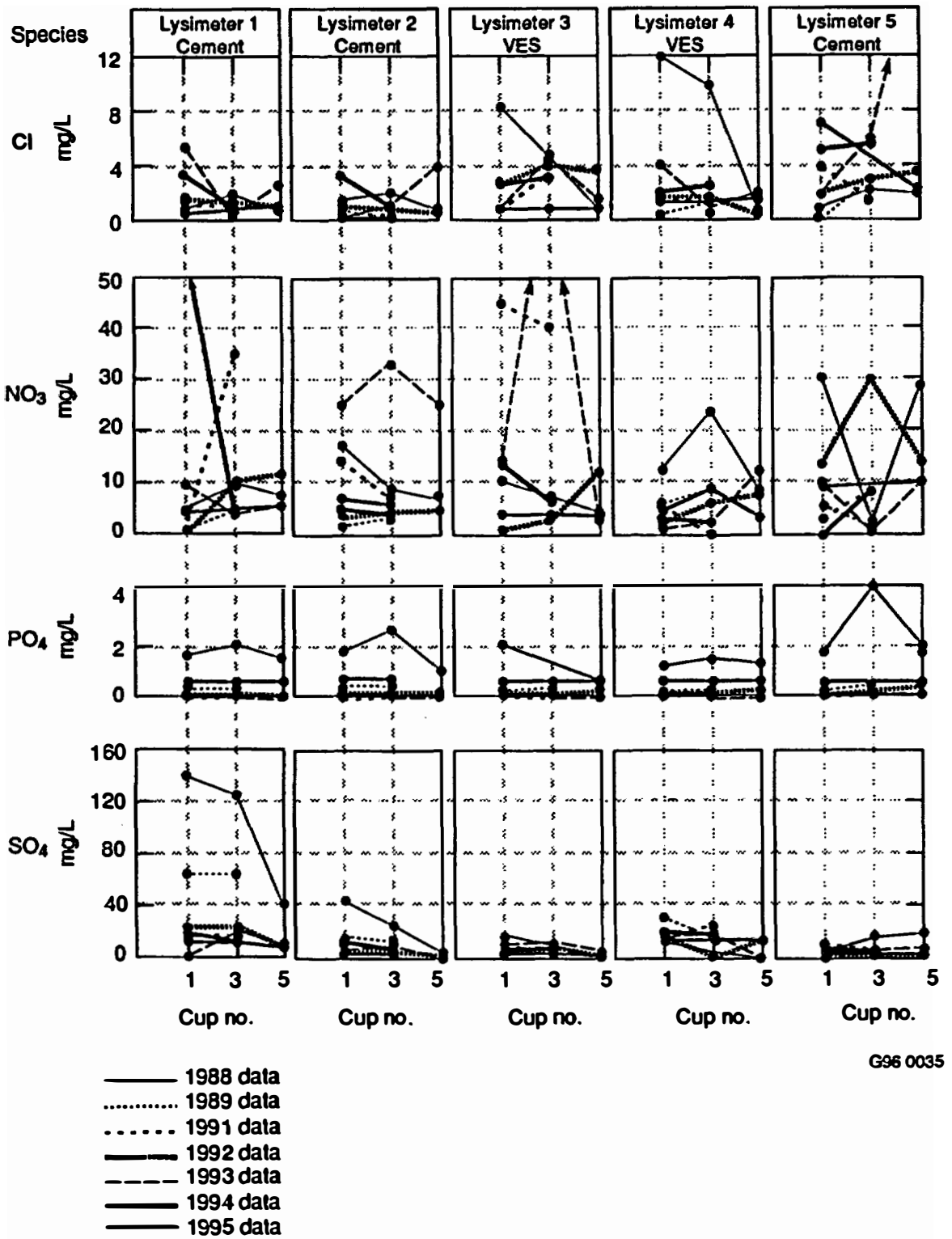

Figure 64. Results of chemical speciation at ORNL—anions.

CONCLUSIONS

Resin Solidification

Both cement waste form types have exhibited an increase in strength with age and self irradiation dose. After a buildup over the first 8 years, that increase seems to have stabilized at about 5,500 psi for both types. In the interval from 7 through 11 years, the cemented organic/inorganic resin waste form sample data were scattered very high and low. The cemented all-organic resin waste samples exhibited more consistent results with one low point at 11 years. Comparing these data to the irradiated compressive strength data obtained during INEL testing in 1985 indicates that the strength of the cemented wastes will slowly decrease with age to about 3,000 psi in the future.

Both VES waste form types also exhibited an increase in strength with age and self-irradiation dose through 8 years. The maximum strength of the VES samples was between 4,500 and 5,000 psi. However, at 11 years, strength had dramatically decreased with the organic/inorganic resin waste form sample (20%). At 12 years, both types of waste form samples suffered a further strength decrease (16% for all-organic and 11% for organic/inorganic). Comparing these data to those of the irradiation test compressive strength data of 1985 confirms that the VES sample strength will continue to decrease with age to about 2,000 psi in the next few years.

The total beta/gamma exposure dose experienced by the 12-year-old waste form samples was between 0.6 and 3.1E+6 Rads. That is two orders of magnitude less than the doses received by the irradiated samples of earlier INEL tests, and these aged samples had not reached the low strength exhibited by those earlier samples.

Field Testing

The lysimeter experiment during the 10 years of operation has been successful. Analyses of data collected during those 10 years 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 72 months) and Co-60. Compared to Sr-90, the occurrence of Cs-137, Sb-125, and Co-60 appear insignificant, except for the leachate from the collection of ORNL-5.

On a cumulative basis, a larger amount of Sr-90 is being removed in leachate water to the collectors 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. During the past 96 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 received in the leachate collector eight times more of the available source or inventory of Sr-90 than the control lysimeter at ANL-E. Such data continue to reinforce the assumption that the limiting step in receiving Sr-90 in leachate water is not release of the nuclide from the waste form samples, but rather, movement 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).[29,30,31] SRL has found that Sr-90 will move from buried waste form samples, readily migrate through the soil column, and appear in collected leachate water.[29] 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 form samples at PNL has shown that Sr-90 does not move in those soils.[30,31]

Percent recovery of Sr-90 from the ORNL cups is the same order of magnitude for those lysimeters containing the cement waste form samples 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 form samples 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 form samples are an order of magnitude larger than in those containing cement waste form samples.

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 that releases of Sr-90 from cement and VES are comparable but dependent on environmental influences. Release 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.[29] This interesting difference should be studied further. The data reported by SRL[29] and PNL[30] agree that Cs-137 is more readily released from cement than from VES, while PNL[31] has observed Cs-137 release in trace amounts only from masonry cement waste forms, and none from portland cement and VES waste forms.

A comparison of cumulative fractional releases from field testing of EPICOR-II waste form samples in lysimeters to the releases from bench-leach-testing of similar waste form samples shows that lysimeter releases are at least two orders of magnitude less for Sr-90 in soil and at least four 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. Plots of CFRs are very similar to plots of cumulative releases.

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 such as evaporation enhanced by transpiration 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 4 years, refinements made it possible to successfully model some of the lysimeter Sr-90 releases using the DUST computer code. 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 10-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 dispersivity and partition coefficients. The sensitivity of predicted releases to these model parameters make it essential that site-specific soil data be collected for those parameters. Releases of Sr-90 to cups 2 is helping to define dispersivity as it becomes available. Laboratory testing is also planned to determine partition coefficients of lysimeter soils and sands and to better model the release patterns from the lysimeters.

REFERENCES

1. R. C. Schmitt and H. W. Reno, *Program Plan of the EPICOR and Waste Research and Disposition Program of the Technical Support Branch*, EGG-TMI-6521, revised December 1983.

2. J. W. McConnell, Jr., *EPICOR-II Resin/Liner Research Plan*, EGG-TMI-6198, March 1983.

3. J. W. McConnell, Jr. and R. D. Sanders, Sr., *EPICOR-II Resin Degradation Results from First Samples of PF-8 and PF-20*, NUREG/CR-4150, EGG-2176, Idaho National Engineering Laboratory, EG&G Idaho, Inc., May 1985.

4. R. D. Sanders, Sr. and J. W. McConnell, Jr., *EPICOR-II Resin Degradation Results from Second Samples of PF-8 and PF-20*, NUREG/CR-4608, EGG-2452, Idaho National Engineering Laboratory, EG&G Idaho, Inc., October 1986.

5. J. W. McConnell, Jr., D. A. Johnson, and R. D. Sanders, Sr., *Radiation Degradation in EPICOR-II Ion Exchange Resins, Final Report*, NUREG/CR-5594, EGG-2603, Idaho National Engineering Laboratory, EG&G Idaho, Inc., September 1990.

6. U.S. Nuclear Regulatory Commission, "Technical Position on Waste," Revision 1, Low-Level Waste Management Branch, Washington, D.C., January 1991.

7. American Society for Testing and Materials, ASTM C39, "Standard Test Method for Compressive Strength of Cylindrical Concrete Specimens," 1972.

8. American Society for Testing and Materials, ASTM C617, "Standard Method of Capping Cylindrical Concrete Specimens," 1976.

9. R. M. Neilson, Jr. and J. W. McConnell, Jr., *Solidification of EPICOR-II Resin Waste Forms*, GEND-INF-055, August 1984.

10. R. M. Neilson, Jr. and J. W. McConnell, Jr., *EPICOR-II Resin Waste Form Testing*, NUREG/CR-4637, EGG-2457, Idaho National Engineering Laboratory, EG&G Idaho, Inc., October 1986.

11. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, D. S. Wickliff, and R. R. Brey, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1991*, NUREG/CR-5229, EGG-2577, Vol. 4, Idaho National Engineering Laboratory, EG&G Idaho, Inc., January 1992.

12. R. D. Rogers, J. W. McConnell, Jr., E. C. Davis, and M. W. Findlay, *Field Testing of Waste Forms Containing EPICOR-II Ion Exchange Resins Using Lysimeters*, NUREG/CR-4498, EGG-2438, Idaho National Engineering Laboratory, EG&G Idaho, Inc., June 1986.

13. R. D. Rogers, J. W. McConnell, Jr., M. W. Findlay, and E. C. Davis, *Lysimeter Data from EPICOR-II Waste Forms—Fiscal Year 1986*, EGG-TMI-7417, October 1986.

14. R. D. Rogers, J. W. McConnell, Jr., M. W. Findlay, and E. C. Davis, *Lysimeter Data from EPICOR-II Waste Forms—Fiscal Year 1987*, EGG-TMI-8004, March 1988.

15. R. D. Rogers, J. W. McConnell, Jr., E. C. Davis, and J. D. Jastrow, *Annual Report of the TMI-2 EPICOR-II Resin/Liner Investigation: Low-Level Waste Data Base Development Program for Fiscal Year 1988*, NUREG/CR-5229, EGG-2553, Vol. 1, Idaho National Engineering Laboratory, EG&G Idaho, Inc., December 1988.

16. J. W. McConnell, Jr., R. D. Rogers, E. C. Davis, and J. D. Jastrow, *Annual Report of the TMI-2 EPICOR-II Resin/Liner Investigation: Low-Level Waste Data Base Development Program for Fiscal Year 1989*, NUREG/CR-5229, EGG-2577, Vol. 2, Idaho National Engineering Laboratory, EG&G Idaho, Inc., February 1990.

17. J. W. McConnell, Jr., R. D. Rogers, D. A. Johnson, J. D. Jastrow, and D. S. Wickliff, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1990*, NUREG/CR-5229, EGG-2577, Vol. 3, Idaho National Engineering Laboratory, EG&G Idaho, Inc., December 1990.

18. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, and D. S. Wickliff, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1992*, NUREG/CR-5229, EGG-2577, Vol. 5, Idaho National Engineering Laboratory, EG&G Idaho, Inc., February 1993.

19. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, and T. M. Sullivan, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1993*, NUREG/CR-5229, EGG-2577, Vol. 6, Idaho National Engineering Laboratory, EG&G Idaho, Inc., May 1994.

20. J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow, W. E. Sanford, and T. M. Sullivan, *Annual Report of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program for Fiscal Year 1994*, NUREG/CR-5229, INEL94/0278, Vol. 7, Idaho National Engineering Laboratory, May 1995.

21. N. W. Gokhert, T. L. Duffy, and J. Sedlet, *Environmental Monitoring at Argonne National Laboratory, Annual Report for 1982*, ANL-E-83-26, March 1983.

22. DOE, *Environmental Monitoring Report, United States Department of Energy, Oak Ridge Facilities, Calendar Year 1983*, Y/UB-19, June 15, 1984.

23. "PATHRAE-EPA: A Low-Level Radioactive Waste Environmental Transport and Risk Assessment Code," EPA 520/1-87-028, December 1987, Developed by V. Rogers and C. Hang.

24. "PRESTO-EPA-POP: A Low-Level Radioactive Waste Environmental Transport and Risk Assessment Code," EPA 520/1-87-024-1, December 1987, developed by D. E. Fields, C. A. Little, F. Parraga, V. Rogers, and C. Hang.

25. T. M. Sullivan, *Selection of Models to Calculate the LLW Source Term*, NUREG/CR-5773, BNL-NUREG-52295, Brookhaven National Laboratory, October 1991.

26. T. M. Sullivan, *Disposal Unit Source Term (DUST) Data Input Guide*, NUREG/CR-6041, BNL-NUREG-52295, 1993.

27. D. Isherwood, *Geoscience Data Base Handbook for Modeling a Nuclear Waste Repository*, NUREG/CR-0912, Vol. 1, 1981.

28. R. A. Freeze and J. A. Cherry, *Groundwater*, Chapter 9, pp. 430–432, Prentice-Hall, Inc., Englewood Cliffs, NJ, 1979.

29. S. B. Oblath and M. W. Grant, *Special Wasteform Lysimeters Initial Three-year Monitoring Report*, SRL-DP-1712, November 1985.

30. M. B. Walter, R. J. Serne, T. L. Jones, and S. B. McLaurine, *Chemical Characterization, Leach, and Desorption Studies of Solidified Low-Level Wastes*, PNL-6047, December 1986.

31. T. L. Jones and R. J. Serne, *Special Waste-form Lysimeters—April 1984–1992 Data Summary and Preliminary Interpretation,* PNL-8955, October 1994.

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J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow[1], W. E. Sanford[2], I. L. Larson, T. M. Sullivan[3]

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Idaho National Engineering Laboratory
Lockheed Idaho Technologies Company
P.O. Box 1625
Idaho Falls, Idaho 83415

[1] Argonne National Laboratory, Argonne, IL
[2] Oak Ridge National Laboratory, Oak Ridge, TN
[3] Brookhaven National Laboratory, Upton, NY

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P.R. Reed, NRC Project Manager

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 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 ion-exchange resins, (c) obtaining performance information on solidified ion-exchange resins in a disposal environment, and (d) determining the condition of liners used to dispose the ion-exchange resins.

Compressive test results of 12-year-old cement and vynyl ester-styrene solidified waste forms are presented, which show effects of aging and self-irradiation.

Results of the tenth 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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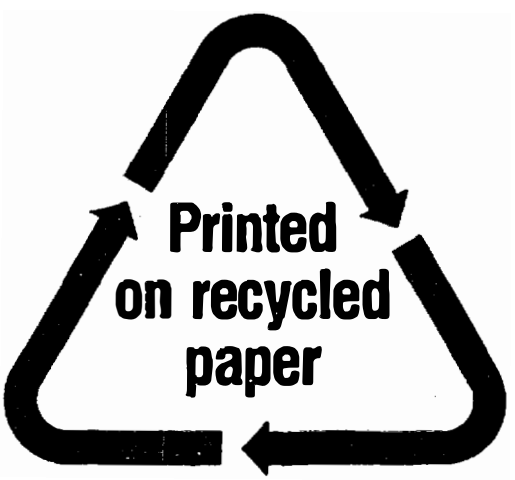

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