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Field Lysimeter Investigations–Test Results

Low-Level Waste Data Base Development Program: Test Results for Fiscal Years 1994 and 1995

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DISCLAIMER

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ABSTRACT

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

Results of the final 2 (10 total) years of data acquisition from operation of 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 experimental equipment is described and results of waste form characterization using tests recommended by the NRC's "Technical Position on Waste Form" are presented. The study is designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over a 20-year period. At the end of the tenth year, the experiment was closed down. Examination of soil and waste forms is planned to be conducted next and will be reported later.

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

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EXECUTIVE SUMMARY

The 28 March 1979 accident at Three Mile Island Unit 2 released approximately 560,000 gal of contaminated water to the auxiliary and fuel handling buildings. The water was decontaminated using a three-stage demineralization system called EPICOR-II, which contained organic and inorganic ion-exchange media. The first stage of the system was designated the prefilter, and the second and third stages were called demineralizers. Fifty EPICOR-II prefilters with high concentrations of radionuclides were transported to the Idaho National Engineering Laboratory for interim storage before final disposal at a commercial disposal facility in the State of Washington. Research is being conducted on materials from four of those EPICOR-II prefilters under three tasks of the TMI-2 EPICOR-II Resin/Liner Investigation: Low-Level Waste Data Base Development Program.

In the first task, Resin Degradation, the changes caused by contained radioactivity were observed in the ion-exchange resin from two EPICOR-II prefilters. Three resin samplings were made over a period of 6 years from PF-8 and PF-20. Results of this study were presented in three NUREG/CR reports.

For the second task, Resin Solidification, portland type I–II cement and vinyl ester-styrene (VES) waste forms incorporating ion-exchange resin waste from EPICOR-II prefilters were subjected to the tests specified in the "Technical Position on Waste Form" issued by the Nuclear Regulatory Commission. Waste form performance data were obtained and reported in two NUREG/CR reports as a result of the work.

The third task, Field Testing, which is reported here, is an ongoing examination of the effect of disposal environments on solidified ion-exchange resin wastes from EPICOR-II prefilters. The purpose of this task, using lysimeter arrays at Oak Ridge National Laboratory and Argonne National Laboratory-East in Illinois, is to expose samples of ion-exchange resin (which were solidified during the Resin Solidification task) to the actual physical, chemical, and microbiological conditions of a disposal environment. The study is designed so that continuous data on nuclide release and movement, as well as environment conditions, can be obtained over a 20-year period.

Experimental equipment includes lysimeter vessels, instruments, leachate samplers, weather stations, and a data acquisition system at each test site. Each month, data stored on a cassette tape are retrieved from the data acquisition system. At least quarterly, water is drawn from the porous cup soil-water samplers and the lysimeter leachate collection compartment. Those water samples are analyzed for beta- and gamma-producing nuclides and chemical species.

Results of the final 2 of a total of 10 years of data acquisition, which are presented in this report, show that radionuclides are moving from the waste forms 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. The experiment was closed down at the end of the tenth year of operation. Examination of soil and waste forms is planned to be conducted next and will be reported later.

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.

Field Lysimeter Investigations: Low-Level Waste Data Base Development Program Lysimeter Test Results for Fiscal Years 1994 and 1995

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 U.S. Nuclear Regulatory Commission (NRC) recommended limits for low-level wastes. Those prefilters were transported to the Idaho National Engineering Laboratory for interim storage before final disposal. A special overpack (high-integrity container) was developed during that storage period to dispose 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, while four were retained for research purposes. Those prefilters used in the research were stored in temporary storage casks and were later disposed at the Radioactive Waste Management Complex at the Idaho National Engineering Laboratory.

Under the EPICOR and Waste Research and Disposition Program, continuing research has been conducted by the INEL on materials from those four 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. Three studies were initiated on organic ion-exchange resins from selected prefilters: (a) the resins were examined to measure radiation degradation, (b) tests were performed to characterize solidified ion-exchange resin waste forms, and (c) experiments are being conducted to field-test solidified wastes using lysimeters.

The Resin Degradation studies examined the radiation degradation caused by contained radionuclides to the organic ion-exchange resin from EPICOR-II prefilters PF-8 and PF-20. Three resin samplings were made over a period of 6 years. Those examinations were completed, and the results were published in three NUREG/CR reports.

In the tests performed in the Resin Solidification task, the EPICOR-II wastes were solidified from two of those prefilters, PF-7 and PF-24, through the use of portland type I–II cement and vinyl ester-styrene (VES), a proprietary solidification agent developed and supplied by the Dow Chemical Company. The formulations used for the immobilization of EPICOR-II wastes were developed to produce waste forms meeting the regulatory requirements of 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Wastes."[3] The NRC Low-Level Waste Management Branch, in its "Technical Position on Waste Form"[4] (BTP), which has been replaced by the BTP Revision 1,[5] provides guidance to waste generators on waste form test methods and acceptable results for compliance with the waste form requirements of 10 CFR 61. In that study, EPICOR-II waste forms were subjected to the recommended NRC test procedures to ensure compliance with the BTP stability requirements

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, recommendations, or favoring by the United States Government or any agency thereof.

and to characterize the waste forms. The solidification studies were completed and reported.

In the Field Testing task discussed in this report, waste forms fabricated under the Resin Solidification task are presently being field-tested at two locations using lysimeters. Experiments were installed at Argonne National Laboratory-East and Oak Ridge National Laboratory to study the effects of disposal environments on those waste forms.

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program is exposing waste forms to the physical, chemical, and microbiological environment of typical disposal sites; monitoring release and movement of radionuclides from those waste forms; and comparing the results with short-term laboratory leach test results. This program has been operating lysimeters for 10 years to obtain information on the performance of radioactive waste forms in a disposal environment and investigate waste form stability per requirements of 10 CFR 61. The experiment measures the releases of radionuclides from the waste forms and subsequent transport through soil columns to sampling locations within the lysimeters. This study was developed to field test waste forms composed of solidified ion-exchange resins from EPICOR-II. The resins used in the study are significant because they have high loadings of radionuclides and are the commercial types used by the nuclear industry.

The NRC has enacted regulations that link low-level radioactive waste acceptance criteria to the long-term satisfactory performance of the disposal facility. Under 10 CFR 61, commercially generated low-level radioactive waste is classified as Class A, B, or C. Class B and Class C wastes must be stabilized into waste forms or placed in containers designed to remain stable for a minimum of 300 years. To verify the 300-year stability, the NRC recommends the use of the short-term standardized tests mentioned earlier with the intention that such tests would provide information relevant to near-surface disposal performance objectives.

A central requirement for disposing low-level radioactive waste is the need for a detailed understanding of the waste form behavior because the radionuclide source from those wastes is the driving force behind the disposal site performance. A major requirement in any site licensing is the performance assessment, which is used to evaluate the proposed disposal site. Assumptions regarding the radionuclide release from buried waste forms have a direct bearing on the outcome of the performance assessment. This has resulted in a very real need to obtain accurate data on the long-term field performance of these wastes.

The objectives of the Field Testing program are to (a) examine the performance of the waste forms in typical low-level waste disposal environments, (b) compare field results with short-term laboratory leach studies, (c) compare field results with Department of Energy 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) apply the DUST source term computer code to compare predicted cumulative radionuclide releases to the lysimeter waste form releases.

The results of the first 4 years of operation were presented in annual reports (References 6 through 9) and were discussed in a topical report (Reference 10). The results of the second 4 years of operation were also presented in annual reports (References 11 through 14) and were discussed in another topical report (Reference 15). This report discusses the results obtained during the last 2 of a total of 10 years of operation of the experiment, which were presented in References 16 and 17. The experiment was closed down at the end of the tenth year of operation. Examination of soil and waste forms is planned to be conducted next and will be reported later.

MATERIALS AND METHODS USED FOR FIELD TESTING

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

This report contains data from the final 2 years of lysimeter operation,[16,17] including cumulative data on water balance and nuclide content of water samples. Data for this report were retrieved from a data acquisition system (DAS) at each site and from beta, gamma, cation, and anion analyses of lysimeter leachate samples. A detailed description of the experimental system is given in Reference 18.

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 (phenolic cation, strong acid cation, and strong base anion resins) from PF-7, and the other is a mixture of synthetic organic ion-exchange resins (strong acid cation and strong base anion resins) with an inorganic zeolite from PF-24. Waste form solidification was described in Reference 19.

Portland type I–II cement and VES were used to solidify both types of resin wastes. In all, 267 waste forms were prepared by combining the resin waste with either cement or VES and allowing the mixture to harden in polyethylene molds 4.8 cm in diameter and 10.2 cm high. Four batches of waste forms were prepared using cement, two batches for each waste type (PF-7 and PF-24). Also, four batches of waste forms were prepared using VES, two batches for each waste type. Table 1 gives the formulations used.[19] The completed waste forms had an average dimension of 4.8 cm in diameter and 7.6 cm high (137.5 cm^3) (Figure 1).

Table 1. Batch formulations for waste forms containing EPICOR-II wastes.

	Formulation weight percentage[a]						
Batch	Waste type	As-received waste	Added water	Decanted waste total[b]	Portland type I–II cement	Additional water	Vinyl ester-styrene
C1	PF-7	15.6	8.5	24.1	62.7	13.2	—
C1A	PF-7	15.6	8.5	24.1	62.7	13.2	—
C2A	PF-24	16.8	7.2	24.0	62.5	13.5	—
C2B	PF-24	16.5	7.0	23.5	61.4	15.1	—
D1	PF-7	40.9	20.3	61.3	—	—	38.7
D1A	PF-7	38.9	22.6	61.5	—	—	38.5
D2	PF-24	43.1	18.3	61.4	—	—	38.6
D2A	PF-24	34.9	14.9	49.8	—	—	50.2

a. Does not include catalyst and promoter, which constitutes a total of approximately 1 wt%.

b. Decanted waste total is the as-received waste plus added water.

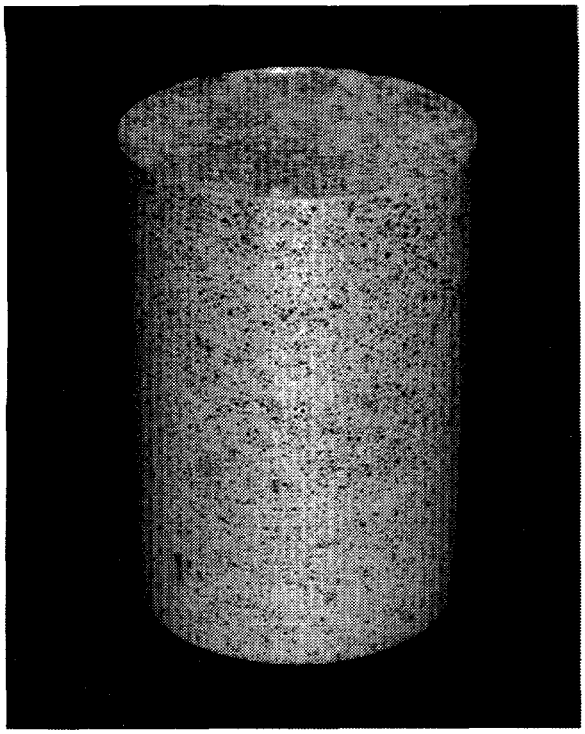

Figure 1. An example of an EPICOR-II prefilter waste form.

Aliquots (0.1 to 0.3 g each) of dried EPICOR-II resin wastes were analyzed by gamma spectroscopy and Sr-90 analysis to determine the radionuclide contents. PF-7 contains 5% Sr-90, while PF-24 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. The average resin radionuclide activities are given in Table 2 and Reference 20.

Radioactive EPICOR-II waste forms were characterized by testing in accordance with recommendations in the BTP to determine the presence of any free-standing liquid, as-prepared compressive strength, and homogeneity.[20] During the tests, no free-standing liquid was observed on any of the waste forms. The compressive strengths of all the as-prepared waste forms tested exceeded the 350-kPa minimum strength required by the BTP (Table 3). The high compressive strengths and the appearance of the waste forms after failure indicated that the waste forms were homogeneous.

Environmental tests were also conducted on the waste forms in accordance with BTP recommendations to determine thermal stability, leachability, immersion stability, radiation stability, leachability after irradiation, and biodegradability.[20] The results of those tests are summarized in the following paragraphs.

No thermal instability was noted in testing. Average compression test data are given in Table 3 for the thermally cycled waste forms. The BTP required that waste forms should have compressive strengths greater than 350 kPa after thermal cycling. All thermally cycled waste forms had compressive strengths two orders of magnitude above the required minimum.

The cement and VES waste forms containing wastes from both PF-7 and PF-24 were found to be resistant to leaching. All waste forms tested had leachability indexes greater than 6.0, as required by the BTP (Table 4).

Immersion stability was determined by testing the compressive strength of waste forms that had been immersed for 90 days in both seawater and

Table 2. Activity content of EPICOR-II resin wastes.

Waste type	Nuclide	Activity content[a] ± 1σ (Ci/g dry resin)
PF-7	Cs-134	7.73E-5 ± 2.83E-7
	Cs-137	1.17E-3 ± 9.90E-5
	Sr-90	6.92E-5 ± 7.21E-6
PF-24	Cs-134	3.30E-4 ± 5.80E-5
	Cs-137	4.99E-3 ± 3.04E-4
	Sr-90	1.18E-5 ± 6.36E-7

a. Cs-134 and -137 as of September 20, 1983; Sr-90 as of October 25, 1983.

Table 3. Compressive strengths of EPICOR-II waste forms.

		Compressive strength ± 1σ (psi)				
Binder	Waste type	As-prepared	Thermal cycled	Immersion tested	Radiation stability	Biodegradability
PC	PF-7	2,930 ± 480	4,740 ± 90	2,960 ± 780	3,640 ± 1,440	2,260 ± 740
PC	PF-24	3,620 ± 720	5,670 ± 650	3,850 ± 1,200	3,310 ± 1,710	—
VES	PF-7	2,900 ± 150	2,770 ± 330	2,770 ± 300	1,930 ± 560	—
VES	PF-24	3,580 ± 190	4,060 ± 70	3,270 ± 320	2,420 ± 810	—

PC = Portland type I–II cement.

VES = Vinyl ester-styrene.

Table 4. Effect of gamma irradiation on the leachability index.

				Leachability index			CFR	
Binder	Waste type	Leachant	Gamma dose (rad)	Cs-134	Cs-137	Sr-90	Cs-137	Sr-90
PC	PF-7	DI	0	10.3	10.3	—	4.7E-2	—
PC	PF-7	DI	5.3E+8	9.4	9.3	9.0	9.1E-2	7.8E-2
PC	PF-24	DI	0	10.6	10.4	—	2.3E-2	—
PC	PF-24	DI	5.4E+8	10.0	9.9	—	2.2E-2	—
PC	PF-7	SW	0	9.6	9.5	—	9.0E-2	—
PC	PF-7	SW	5.3E+8	10.0	9.9	—	4.6E-2	—
PC	PF-24	SW	0	10.4	10.3	—	2.6E-2	—
PC	PF-24	SW	5.4E+8	10.9	10.8	—	1.2E-2	—
VES	PF-7	DI	0	12.4	12.2	—	2.0E-3	—
VES	PF-7	DI	5.7E+8	9.8	9.7	9.7	4.1E-2	4.5E-2
VES	PF-24	DI	0	14.0	13.8	—	3.4E-4	—
VES	PF-24	DI	4.9E+8	12.3	12.2	—	3.0E-3	—
VES	PF-7	SW	0	9.4	9.3	—	6.4E-2	—
VES	PF-7	SW	5.7E+8	8.8	8.7	—	1.2E-1	—
VES	PF-24	SW	0	10.9	10.7	—	1.3E-2	—
VES	PF-24	SW	4.9E+8	10.0	9.8	—	3.9E-2	—

PC = Portland type I–II cement.

VES = Vinyl ester-styrene.

DI = Demineralized water.

SW = Synthetic seawater.

CFR = Cumulative fractional release.

deionized water. All specimens exhibited strengths well above the required 350 kPa, as shown in Table 3.

In the radiation degradation test, the total gamma irradiation dose received by the waste forms was larger than the total dose of beta and gamma radiation that the waste forms would have received through self-irradiation by the end of 300 years. All irradiated specimens had compressive strengths far in excess of the 350 kPa required by the BTP (Table 3).

Even though leachability after irradiation testing is not required by the BTP, tests were conducted. Table 4 lists the average leachability indexes for irradiated waste forms. All leachability indexes are above the value of 6.0 recommended by the BTP.

The data of cumulative fractional release with time for irradiated cement waste form C1-5 and irradiated VES waste form D1-1 (resins from PF-7) are plotted in Figure 2 for Sr-90 and Cs-137.[8] The fractional releases were nearly identical for the two radionuclides from a specific waste form. It is noted that the cement waste form exhibited the higher fractional release of both Sr-90 and Cs-137, about 8% of the total inventories, while the VES fractional releases were about 4.5% of the inventories. The leach indices for the waste forms are also given. The cement leach indices were comparable for Sr-90 and Cs-137 (9.0 and 9.3) and lower than those of the VES (9.7). Also, the Sr-90 leached more rapidly from both types of waste forms than did Cs-137. This was particularly evident in the case of the VES waste form where nearly all the leachable Sr-90 had been removed in 5 days.

Figure 3 presents fractional release of Cs-137 over time in demineralized water from unirradiated portland type I-II cement and VES waste forms containing PF-7 and PF-24 resins.[8] These data illustrate the lower leachability (higher leachability index) of VES compared with cement for the EPICOR-II resin waste forms. The waste forms containing PF-24 resins exhibited better leach characteristics for Cs-137, probably because those resins contained inorganic zeolite, which does not degrade with the radiation doses observed in the EPICOR-II prefilters.

A comparison of the information of Figures 2 and 3 shows that the fractional release of the waste forms was higher with a higher irradiation dose. This effect was more pronounced with VES waste forms.

VES and cement waste forms were placed in nutrient-rich media to test the growth of the applied species of fungi and bacteria.[21] The VES waste forms supported fungal growth, but not bacterial. The cement waste forms were not affected by and did not support their growth. Also, the cement waste forms did not chemically or radiologically prevent the growth of fungi. Only cement waste forms from PF-7 were subjected to compression tests after exposure to microbial attack. The results are given in Table 3.

A complete description of waste form manufacture is given in Reference 19; bench testing of those EPICOR-II waste forms, according to the recommendations of the BTP, is further described in References 8, 20, and 21.

Description of Test Sites

Field testing is being conducted at Argonne National Laboratory-East (ANL-E) and Oak Ridge National Laboratory (ORNL). Both laboratories have set aside field sites that cover areas of approximately 116 m^2. These field sites have been dedicated to testing solidified EPICOR-II waste forms since the installation of experiments in 1985. Testing is planned to last a total of 20 years, until the year 2005. ANL-E ensured the physical security of the field site by enclosing it with a fence 2.4 m high; the field site at ORNL is enclosed within a larger, controlled-access area. Field locations at each laboratory are shown in Figures 4 and 5. Both sites offer unobstructed exposure to prevailing environmental conditions while providing security from inadvertent personnel exposure to irradiation or contamination.

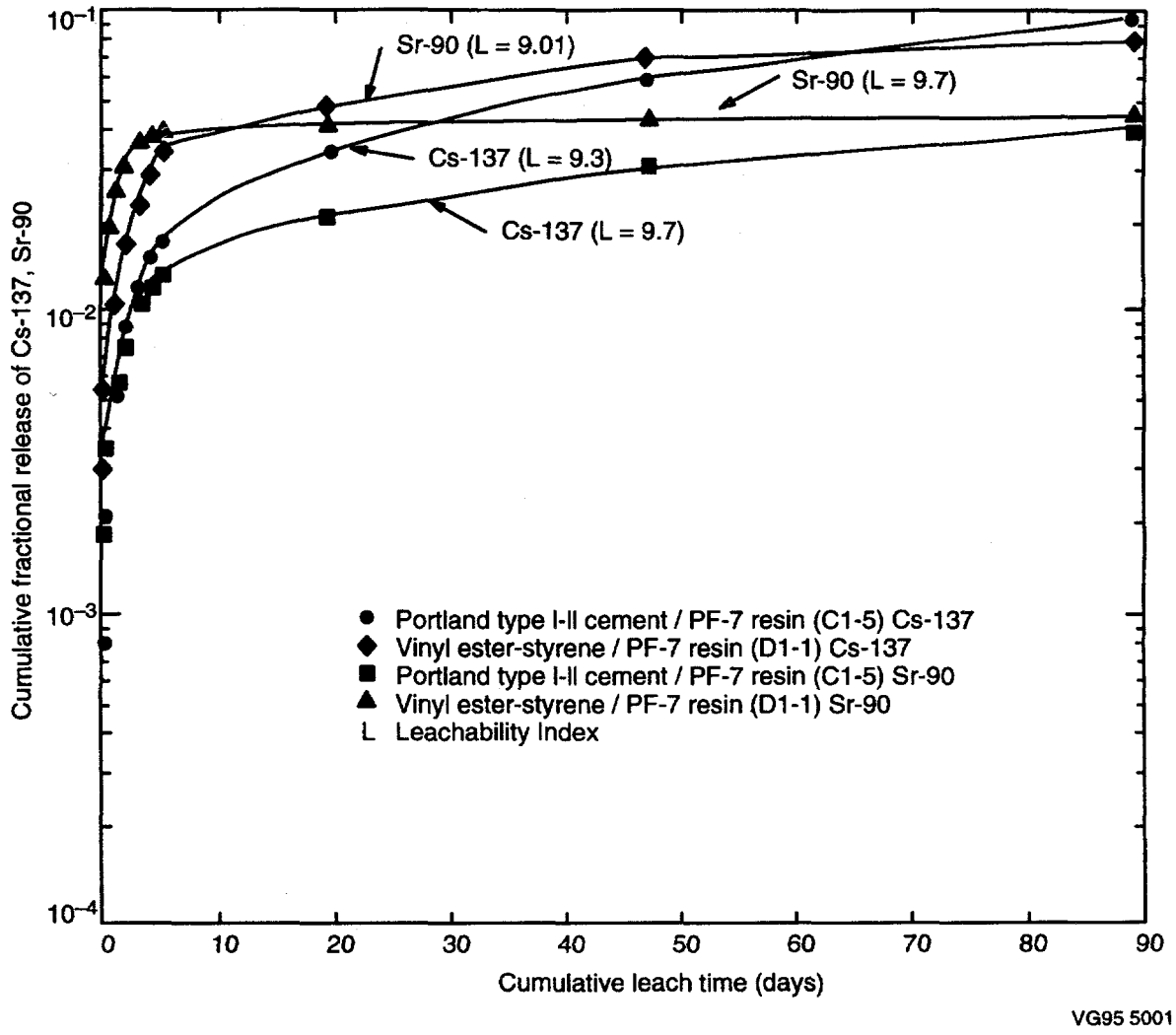

Figure 2. Irradiated EPICOR-II waste form radionuclide cumulative fractional release of Cs-137 and Sr-90 with demineralized water leachant.

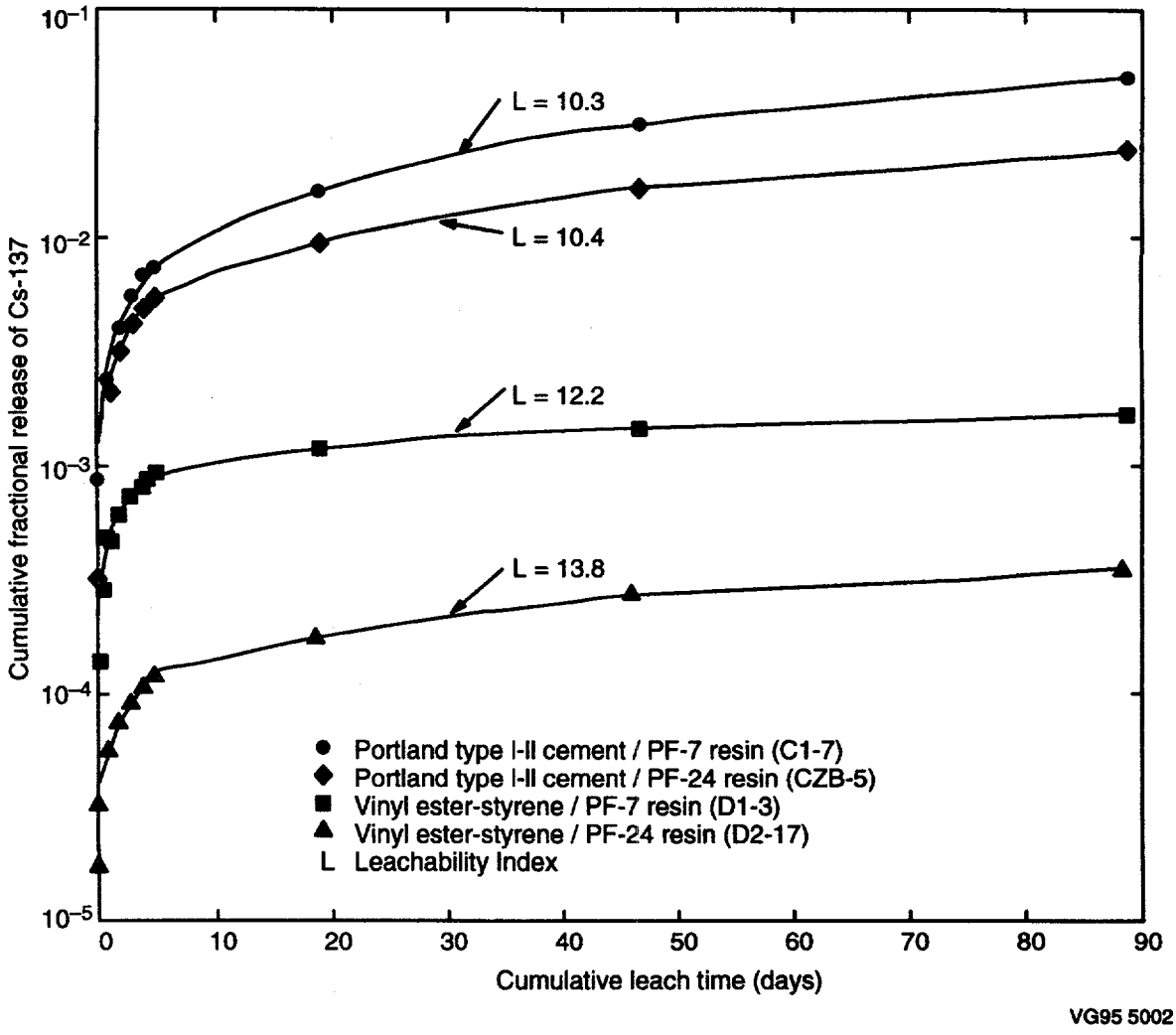

Figure 3. Cumulative fractional release of Cs-137 from unirradiated EPICOR-II waste forms with demineralized water leachant.

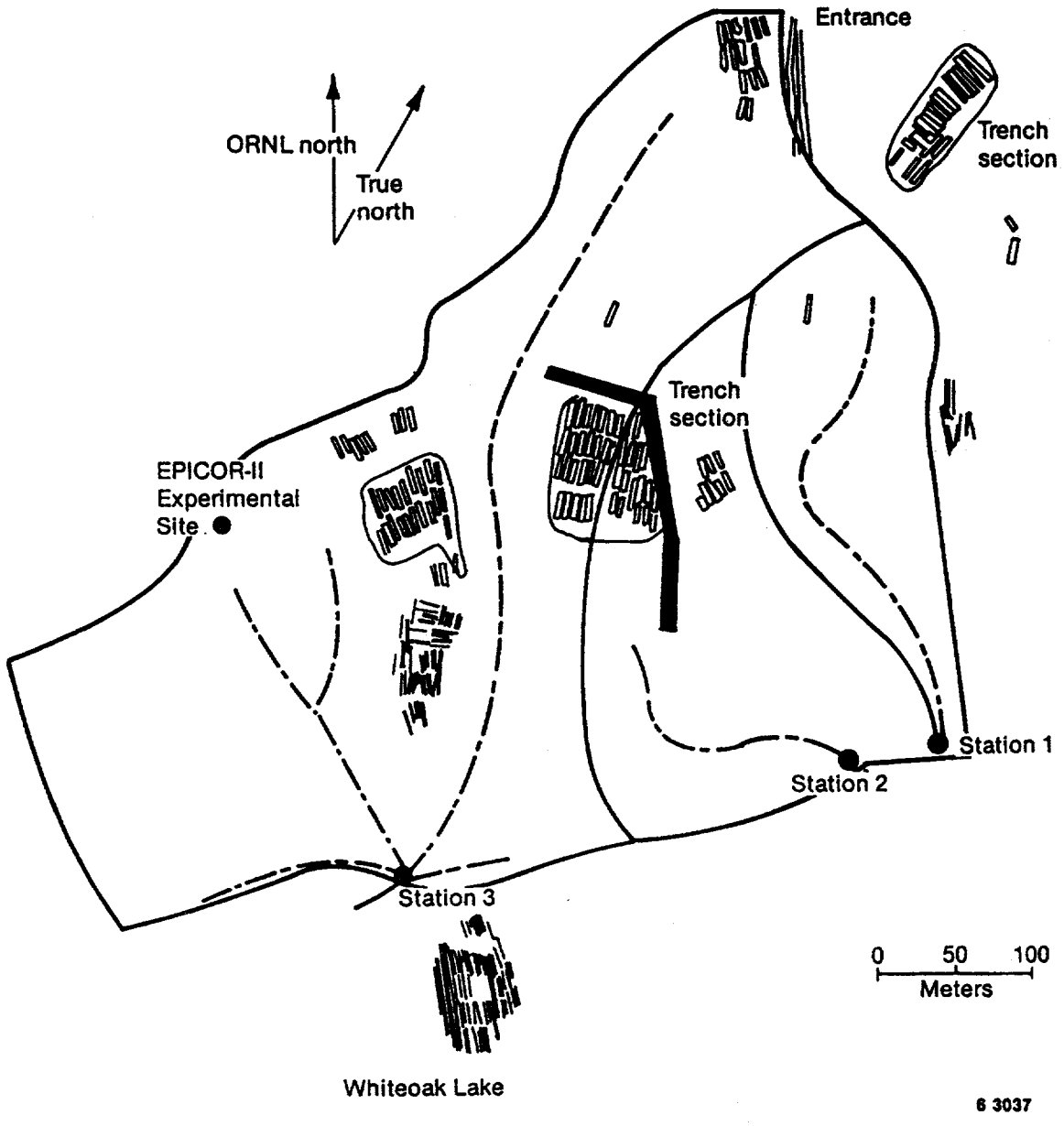

Figure 4. Location of the EPICOR-II lysimeter experiment at ORNL.

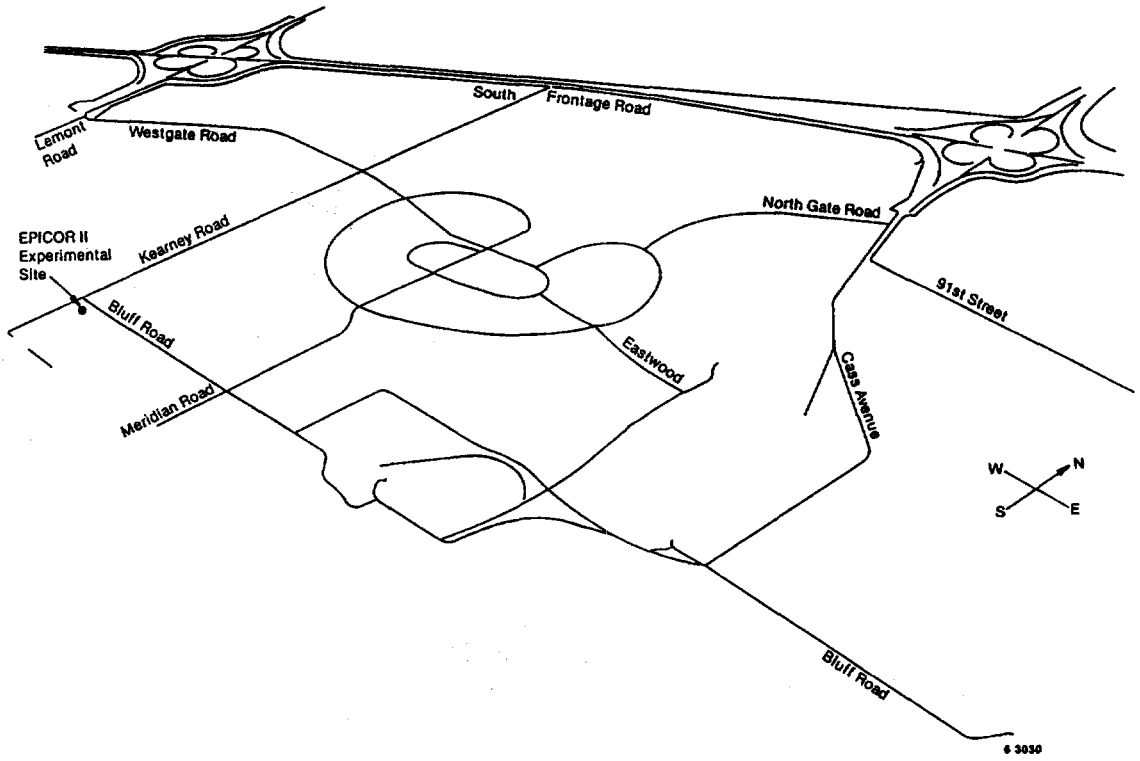

Figure 5. Location of the EPICOR-II lysimeter experiment at ANL-E.

ANL-E is located 43 km southwest of Chicago, Illinois, and 39 km due west of Lake Michigan. It has terrain that is gently rolling and partially wooded, which was formerly prairie and farm land. The area around the testing site has been allowed to return to natural vegetation, while the soil surface of each lysimeter has been weeded frequently to prevent the growth of any vegetative cover. The climate is that of the upper Mississippi Valley, as moderated by Lake Michigan. On average, temperatures of 0°C or colder prevail during the months of December through February, with temperatures near or slightly above 20°C during June through August. The average frost line in soil is 89 cm during the cold months. Precipitation (an average of 85.2 cm) appears to be uniformly distributed during the year, with May through September being the wettest months.[22]

ORNL is located 26 km east of Knoxville, Tennessee, in a broad valley that lies between the Cumberland Mountains to the northwest and the Great Smoky Mountains to the southeast. The coldest month is normally January (4°C), but differences between the mean temperatures of the three winter months of December, January, and February are comparatively small. July is usually the hottest month (24°C), but temperatures vary little during June, July, and August. The average frost line in soil is usually no deeper than 23 cm. Winter and early spring are the seasons of heaviest precipitation, with the monthly maximum normally occurring during January to March, although heavy rain may occur in July. The mean annual precipitation is 134 cm.[23]

Both ANL-E and ORNL sites were supplied with field meteorological stations. These stations consist of a tipping-bucket rain gauge (heated so as to measure the water content of snow), wind speed sensor, wind direction sensor, and air temperature/relative humidity probe. All equipment except the rain gauge is mounted on a 3-m, electrically grounded tripod located adjacent to each lysimeter array. Data from each instrument are processed and stored in real time by the DAS.

Description of Lysimeters

The lysimeters are designed as self-contained units that can be easily disposed after the field test experiment is completed. Each lysimeter is a right-circular cylinder (0.91 m ID by 3.12 m in height) constructed of 12-gauge, 316 L stainless steel (Figure 6). Internally, the lysimeter is divided into two sections, the upper being 1,532 L in volume and the lower being 396 L (Figure 7). A 3.8-cm, Schedule 40, stainless steel pipe provides access to the lower compartment, which serves as a leachate collector.

Instrumentation includes porous cup soil-water samplers by Timco and soil moisture/temperature probes by Soil Test, Inc. The probes are connected to an on-site Campbell Scientific CR-7 DAS, which also collects data from a Campbell Scientific field meteorological station located at each site.

The lysimeters at each site are consecutively numbered 1 through 5; lysimeters 1 through 4 contain soil, and number 5 is used as a control and is filled with an inert silica oxide sand.[18] Each lysimeter contains seven waste forms stacked end to end vertically. Table 5 shows which type of waste form was placed in each lysimeter.

The local indigenous soil at ANL-E met the NRC criterion for Midwestern soil, so it was used for the filler in lysimeters 1 through 4 at ANL-E. It is a Morley silt loam with the surface layer removed. The resulting subsurface soil is a clay loam. Chemical and physical properties of this soil are given in Table 6.

The soil for the ORNL lysimeters was intended to approximate soil found at Barnwell, South Carolina. Because the soil at ORNL was not a suitable substitute for Barnwell soil, soil was transported to ORNL from the Savannah River Plant adjacent to the Barnwell facility in South Carolina. That soil is from the C horizon of a Fuquay sandy loam; chemical and physical properties of that soil are listed in Table 6. The soil is similar texturally to the subsurface soil found at Barnwell.[b] The only apparent difference between the two soils could be pH.

The material to be used as filler in the control lysimeter at each site needed to meet the NRC criterion of low cation-exchange capacity, which is a major contributor to the retention of many radionuclides in soil. Three materials [high-density polyethylene beads, aluminum oxide (Al_2O_3), and inert silica oxide (SiO_2) sand] were evaluated as inert filler. Only silica oxide sand was found to be suitable. This sand was obtained from the Unimin Corporation, Troy, Illinois, under the trade name "Granusil 100."

Several mesh sizes of silica oxide sand were evaluated. They were classified by the manufacturer as very fine/fine, fine/medium, medium/coarse, and coarse. Table 7 provides information on the particle size distribution of these samples, while moisture holding capacity and cation-exchange capacity are listed in Table 8. The physical characteristics of each sample were considered (density, ability to provide rigid support for probes, moisture retention, etc.), along with cost and availability. The fine/medium sand was selected as best suited for use in the control lysimeters.

One final item used as an integral part of the fill material was a layer of a support/filter fabric. That material (DuPont "Typar" style 3401) was placed at the interface of the soil or sand and the gravel bed (see Figure 7). The fabric was placed at the bottom of the soil profile in order to (a) improve separation of the soil and the drainage aggregate, (b) prevent clogging of the drainage aggregate with soil fines, and (c) promote adequate drainage of the lysimeter soil/sand. Before installation, the fabric was tested to determine if it would sorb selected radionuclides. The test involved submersing a 59-cm^2 fabric section for 264 hours in a water solution containing Ce-144, I-131, Ru-103, Sr-85, Cs-137, and Co-60. After soaking, the fabric was rinsed with two washes of distilled water, and the

b. Personal communication between E. C. Davis and V. Rogers, Soil Scientist Office, P.O. Box A, Aiken, South Carolina 29801, April 4, 1984.

Figure 6. Unfilled lysimeter vessel being lowered into position at ORNL.

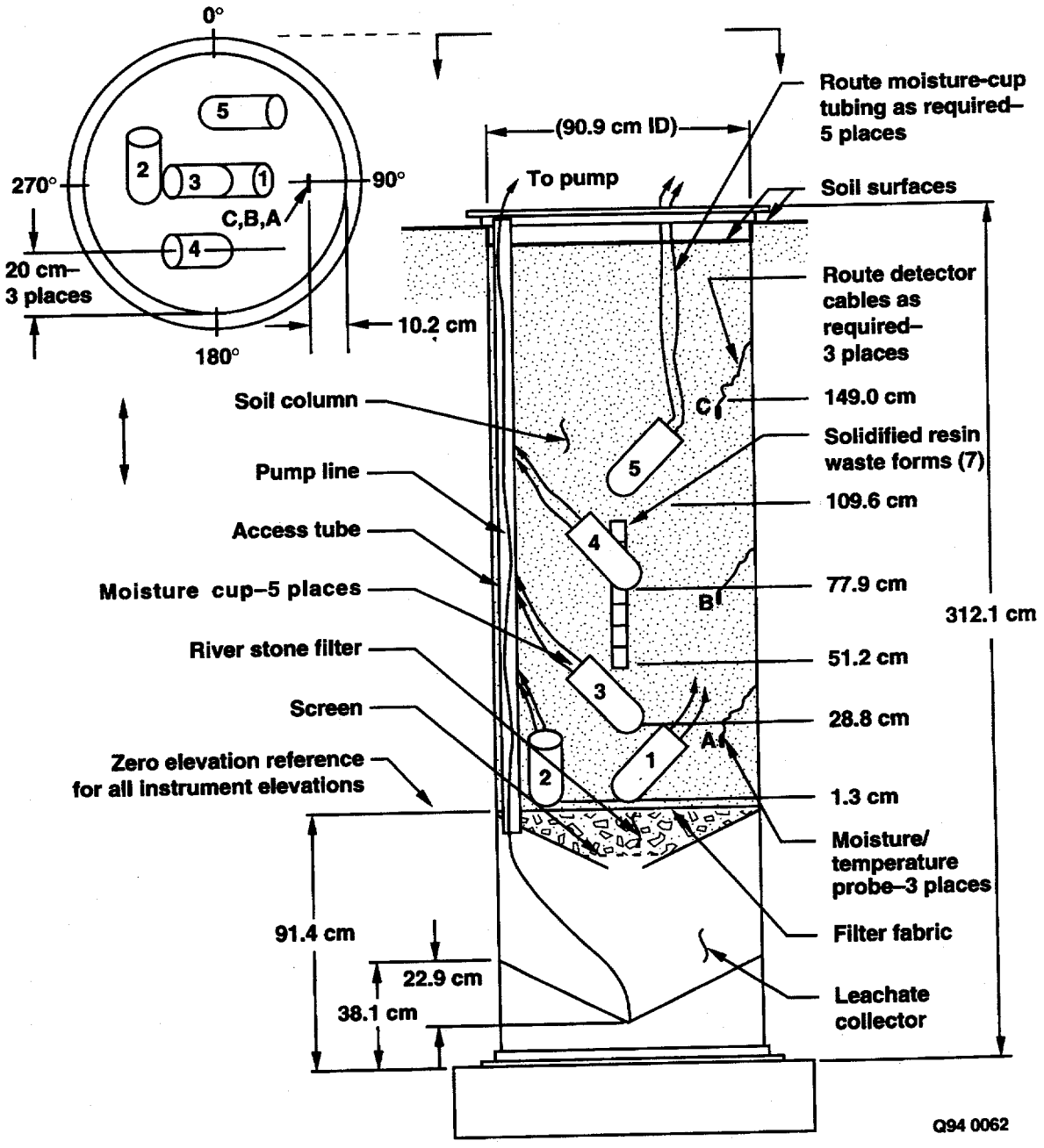

Figure 7. Lysimeter vessel component locations.

Table 5. Lysimeter waste form descriptions and radionuclide inventories.

Lysimeter number	Fill material	Waste form description	Waste form inventory[a] per lysimeter (E+9 pCi)				
			Cs-137	Cs-134	Sr-90	Other[b]	Total
1	Soil	PF-7 resin/cement	312	21	19	3	354
2	Soil	PF-24 resin/cement	1,432	95	3	14	1,544
3	Soil	PF-7 resin/VES	464	31	27	3	525
4	Soil	PF-24 resin/VES	1,928	128	5	18	2,078
5 ANL-E	Silica oxide	PF-7 resin/cement	312	21	19	3	354
5 ORNL	Silica oxide	PF-24 resin/cement	1,432	95	3	14	1,544

a. Cs-137 and Cs-134 activity as of 9/20/83; Sr-90 activity as of 10/25/83.

b. Other includes Co-60 and Sb-125 in trace amounts.

Table 6. Physical and chemical characteristics of soils used at ANL-E and ORNL with comparison of Savannah River Laboratory and Barnwell soils.

Characteristic	Soil		
		ORNL	
	ANL-E	Savannah River Laboratory	Barnwell[a]
Soil bulk density (g/cm^3)	1.74	—[b]	—[b]
Texture (%)			
Sand	29	58	52
Silt	29	2	11
Clay	42	39	38
Clay mineralogy (%)			
Vermiculite	—[b]	10	12
Kaolinite	—[b]	80	77
Percent carbon	4.20	0.07	—[b]
Cation exchange capacity (meq/100 g)	8.4	4.9	8.0
pH (1:1 paste method)	8.3	6.2	4.8[c] to 6.0[d]
Percent moisture-holding capacity	40.6	44.5	—[b]

a. P. L. Piciulo, C. E. Shea, R. Barletta, *Analyses of Soils from the Low-Level Radioactive Waste Disposal Sites at Barnwell, SC, and Richland, WA,* NUREG/CR-4083, Brookhaven National Laboratory, March 1985.

b. Not available.

c. E. B. Fowler, E. H. Essington, W. L. Polzer, *Interactions for Radioactive Wastes with Soils. A Review,* NUREG/CR-1155, Los Alamos Scientific Laboratory, 1979.

d. Personal communication with John N. Fischer, U.S. Geological Survey, Reston, Virginia, 1983.

Table 7. Particle size distribution of Unimin silica oxide sand evaluated for use as inert filler for control lysimeters.

Particle size (mm)	Weight distribution (%)			
	Sample 1 (very fine/fine)	Sample 2 (fine/medium)	Sample 3 (medium/coarse)	Sample 4 (coarse)
0.07–0.09	11.0	—	—	—
0.09–0.10	81.2	—	—	—
0.10–0.12	7.6	—	—	—
0.12–0.15	0.2	2.9	0.1	—
0.15–0.21	—	18.5	0.8	—
0.21–0.30	—	36.6	6.7	0.1
0.30–0.42	—	38.6	46.0	7.4
0.42–0.59	—	3.4	46.4	80.8
0.59–0.84	—	—	0.1	11.7

Table 8. Properties of Unimin silica oxide sand.

Particle size	Cation-exchange capacity (meq/100 g)	Moisture holding capacity (%)
Very fine/fine	0.07	25.6
Fine/medium	0.06	23.0
Medium/coarse	0.05	21.2
Coarse	0.03	20.7

quantity of sorbed nuclides was determined by gamma spectroscopy. Inconsequential amounts of the radionuclides were sorbed to the fabric, as expected (Table 9).

The gravel bed in each lysimeter provides support for the Typar fabric and is intended to promote drainage of water from the soil column. Gravel is prevented from entering the leachate compartment by a screen covering the drainage port (Figure 7). ANL-E used a granitic pea gravel of a 0.64-cm size, while ORNL used crushed silica quartz river rock of the same size. All gravel was prewashed to remove fines.

Data Collection and Analysis

Data from the moisture/temperature probes within the lysimeters, as well as that from the weather station, are collected by, processed in, and stored in a Campbell Scientific Model CR-7 DAS. This programmable unit has multiple processors, 28 differential input channels (the probes and weather station requiring 21 of those channels), excitation for ac or dc resistive measurements, analog outputs, and internal data storage (20,000 data values), as well as output to a cassette tape recorder that provides storage for an additional 180,000 values. The unit weighs 13.6 kg and its dimensions are 43.5 × 30.7 × 5.1 cm. It is housed at each lysimeter site within a heated, environmentally sealed, metal enclosure with dimensions of 60.5 × 60.5 × 35.8 cm.

The DAS has a scan rate of 250 channels/sec, ensuring instantaneous acquisition of data from all data sources during each activation cycle. The DAS collects data during the day and stores the data in memory. At the beginning of each day

Table 9. Extent of nuclide sorption to DuPont 3401 drainage cloth.

Nuclide	Percent sorbed
Ce-144	0.12
I-131	0.07
Ru-103	1.02
Sr-85	0.00
Cs-137	0.86
Co-60	0.00

(0000 h), the system processes the data from the previous day to provide a daily maximum, minimum, and average for each source except for the rain gauge, which provides a total rain value. This processing produces 200 8-character numbers (see Table 10 for example), which are transferred daily to the cassette tape that provides auxiliary storage for up to 112 days of data. The first two characters of each number serve as identifiers.

The cassette tape is retrieved from the DAS each month and translated to an IBM PC-compatible disk file using a Campbell Scientific C20 cassette interface. Once transferred to disk, the data are arranged in tables (see Table 11 for example). These files are printed in either text or graphic format. The graphic format presents data over an extended time period and is used in 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 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.

Table 10. Example of 1-day data block in CR-7 DAS format.

01 + 0104.	02 + 0214.	03 + 0000.	04 + 0.240	05 + 24.76	06 + 084.5	07 + 1.366	08 + 201.1
09 + 22.04	10 + 23.28	11 + 25.73	12 + 24.43	13 + 23.38	14 + 25.69	15 + 65.35	16 + 23.42
17 + 25.60	18 + 20.95	19 + 23.24	20 + 25.71	21 + 19.40	22 + 22.27	23 + 24.72	24 + 36.66
25 + 34.68	26 + 10.04	27 + 39.12	28 + 29.60	29 + 07.92	30 + 07.92	31 + 38.17	32 + 07.59
33 + 07.59	34 + 07.61	35 + 17.58	36 + 10.80	37 + 15.26	38 + 09.21	39 + 0.933	40 + 0.961
41 + 1.015	42 + 0.986	43 + 0.962	44 + 1.014	45 + 1.616	46 + 0.964	47 + 1.012	48 + 0.910
49 + 0.960	50 + 1.014	51 + 0.875	52 + 0.992	53 + 0.992	54 + 0.798	55 + 0.705	56 + 0.042
57 + 0.924	58 + 0.498	59 + 0.000	60 + 0.004	61 + 0.874	62 + 0.006	63 + 0.006	64 + 0.008
65 + 0.163	66 + 0.051	67 + 0.119	68 + 0.031	69 + 22.24	70 + 62.84	71 + 1.000	72 + 0.193
73 + 22.03	74 + 23.28	75 + 25.66	76 + 24.26	77 + 23.37	78 + 25.56	79 + 63.45	80 + 23.42
81 + 25.47	82 + 20.97	83 + 23.23	84 + 25.59	85 + 19.20	86 + 22.28	87 + 24.62	88 + 36.24
89 + 34.27	90 + 09.89	91 + 38.87	92 + 28.85	93 + 07.81	94 + 07.64	95 + 37.98	96 + 07.60
97 + 07.60	98 + 07.60	99 + 16.32	00 + 10.69	01 + 15.04	02 + 08.97	03 + 0.934	04 + 0.961
05 + 1.014	06 + 0.983	07 + 0.963	08 + 1.012	09 + 1.601	10 + 0.964	11 + 1.010	12 + 0.910
13 + 0.960	14 + 1.012	15 + 0.871	16 + 0.939	17 + 0.991	18 + 0.776	19 + 0.685	20 + 0.040
21 + 0.909	22 + 0.470	23 + 0.000	24 + 0.004	25 + 0.863	26 + 0.005	27 + 0.005	28 + 0.007
29 + 0.138	30 + 0.050	31 + 0.115	32 + 0.028	33 + 31.35	34 + 090.4	35 + 09.00	36 + 360.6
37 + 22.08	38 + 23.34	39 + 23.82	40 + 24.86	41 + 23.42	42 + 25.78	43 + 68.22	44 + 23.47
45 + 25.70	46 + 20.99	47 + 23.30	48 + 25.81	49 + 19.66	50 + 22.33	51 + 24.81	52 + 37.01
53 + 34.97	54 + 10.16	55 + 39.39	56 + 30.00	57 + 07.98	58 + 07.69	59 + 38.35	60 + 07.62
61 + 07.61	62 + 07.64	63 + 18.86	64 + 10.89	65 + 15.43	66 + 09.43	67 + 0.935	68 + 0.963
69 + 1.017	70 + 0.996	71 + 0.964	72 + 1.016	73 + 1.640	74 + 0.966	75 + 1.014	76 + 0.911
77 + 0.962	78 + 1.017	79 + 0.881	80 + 0.940	81 + 0.995	82 + 0.814	83 + 0.717	84 + 0.043
85 + 0.937	86 + 0.511	87 + 0.001	88 + 0.005	89 + 0.882	90 + 0.007	91 + 0.007	92 + 0.009
93 + 0.189	94 + 0.052	95 + 0.122	96 + 0.034	97 + 1.366	98 + 0.185	99 + 318.7	00 + 075.3

Table 11. Example of transcribed CR-7 DAS data.

Year: 1985 Day: 237 Time: 0 hrs

Weather data for preceding 24-hour period

Rainfall		Temp (°C)	Relative humidity	Wind speed (mph)	Direction (degrees)
0.00 in.	Avg	19.96	87.50	3.12	244.30
	Max	27.10	95.50	24	360.00
	Min	15.36	59.36	1.00	0.19

Soil conditions

		Lysimeter 1		Lysimeter 2		Lysimeter 3		Lysimeter 4		Lysimeter 5	
Elevation		T(°C)	%M	T(°C)	%M	T(°C)	%M	T(°C)	%M	T(°C)	%M
28.8 cm	Avg	18.3	6.5	18.4	8.9	18.3	12.8	17.5	10.0	17.6	-2.8
	Max	18.3	7.6	18.4	9.3	18.4	12.9	17.6	10.4	17.7	-2.7
	Min	18.3	5.9	18.4	8.6	18.3	12.6	17.5	9.8	17.6	-2.8
77.9 cm	Avg	19.3	6.5	19.5	10.3	19.3	13.2	19.2	11.1	19.0	-1.1
	Max	19.3	7.0	19.6	10.8	19.3	13.2	19.2	11.2	19.0	-1.1
	Min	19.2	5.9	19.6	9.8	19.2	12.9	19.1	10.8	19.0	-1.2
149.0 cm	Avg	20.6	6.3	20.8	12.1	20.5	7.9	20.6	6.6	20.3	-1.6
	Max	20.6	7.0	20.9	12.3	20.5	8.6	20.7	7.5	20.4	-1.3
	Min	20.6	5.9	20.8	11.8	20.5	7.0	20.6	5.9	20.2	-1.8

RESULTS AND DISCUSSION OF FIELD TESTING

This section presents DAS data from the beginning of the experiment (ANL-E—August 1, 1985; ORNL—June 1, 1985) through June 1995. In addition, information on water balance, nuclide, and cation/anion content in soil water and leachate is presented. Many of the data are displayed in graphic format so that information can easily be correlated with time. This information has been presented on an annual basis in References 6, 7, 8, 9, 11, 12, 13, 14, 16, and 17 .

The DAS at ANL-E functioned fairly well during the last 2 years. There was one period of time when the DAS was not in operation from mid-September through November 1994 due to equipment failure that required repair. However, due to technical problems with the ORNL DAS in 1995, only a fraction of the data from that year is presented. Those periods were July 1994 and April through mid-May of 1995. Data loss was caused by failure of the CR-7 data logger, which was replaced with an available CR-10 made by the same manufacturer.

Weather Data

Precipitation, air temperature, wind speed, and relative humidity, as recorded by the ANL-E and ORNL systems during the 24-month reporting period, are presented in Appendix A. Average annual precipitation for the period was 88.0 cm at ANL-E and 146.6 cm at ORNL. ANL-E was at 103% of the normal annual rainfall[22] of 85.2 cm, while ORNL was 106% of the normal annual rainfall[23] of 138.8 cm. The monthly precipitation pattern for each site can be seen from the histograms in Figures A-1 and A-2 and Figures A-5 and A-6 in Appendix A. Figure 8 shows the cumulative precipitation for both sites since the initiation of field work.

In 1995, ANL-E, for the second time since 1985, was well above the normal annual rainfall (12%) after being 5% below normal in 1994, while

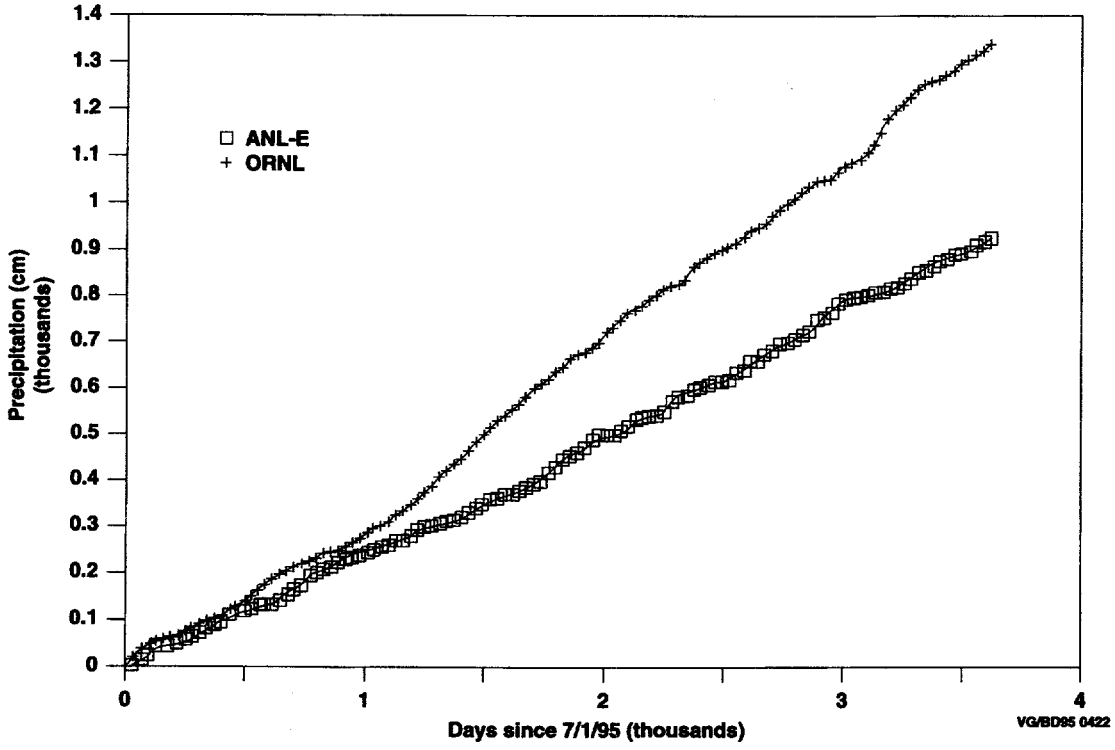

Figure 8. ANL-E and ORNL cumulative precipitation.

ORNL was 28% above the normal annual rainfall in 1994. Last year (1995) was the second time in 9 years that ORNL did not equal or exceed the normal amount of yearly precipitation but was 17% below normal. By the end of this reporting period, there was a cumulative precipitation total of 918 cm at ANL-E, while ORNL received a total of 1,338 cm.

In October 1990, the anemometer at ANL-E ceased normal operation. During 1992, the anemometer at ORNL appears to have failed at times due to mechanical wear of bearings. Because of these failures, windspeed data are not included in this report. Also, relative humidity readings at both sites became questionable in 1993 and are not included in this report.

In June 1986, the ORNL rain gauge was replaced with a Climatronics tipping-bucket gauge, which is designed for episodic high-intensity rainfall. Data from this gauge appear to be accurate; however, the rainfall data recorded by the DAS contain occasional, erroneously high data points. The Weather Measure tipping-bucket rain gauge supplied with the DAS at ANL-E has occasionally failed to produce accurate rainfall readings as well; it appears to be either underreporting precipitation events or sporadically not recording events at all. These malfunctions have not resulted in a loss of rainfall data because both ANL-E and ORNL have mechanical recording rain gauges close to their lysimeter sites. Data from those nearby rain gauges are presented in Figures A-1, A-2, A-5, and A-6 and were used to calculate the total quantities of precipitation received by each site.

Air temperature data from ANL-E show that periods of freezing temperatures occurred each of the past 2 years from early November until mid-March (Figures A-3 and A-4). ORNL experienced periods of freezing temperatures from early December until late February 1994 and not at all in 1995 (Figures A-7 and A-8).

Lysimeter Soil Temperature Data

Soil temperature and moisture sensors (probes) 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 (Figure 7). The function of these probes is to provide data on the physical environment experienced by the buried waste forms, specifically, whether or not they 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 of collected data. Therefore, as long as there are functioning probes in any of the soil lysimeters at each site, data sufficient to satisfy reporting criteria will be available. In addition, temperature data collected during the years of extended service life of the probes will serve as a useful climatological reference for assessing waste form performance in future years.

The lysimeter soil temperature data recorded at ANL-E and ORNL during the reporting period are shown in Figures B-1 through B-18 of Appendix B. The only probe to record a valid freezing temperature was at the 149-cm elevation in ANL-1 (Figures B-1 and B-2). The 28.8-cm probe data for that lysimeter are erroneous. A direct correspondence can be seen between air temperature and soil temperatures at both sites.

Past reports have detailed the failure of some temperature probes at ANL-E. Faults were found with one temperature probe in ANL-2, two in ANL-3, all in ANL-4, and one in ANL-5. During this reporting period, an additional probe failed in ANL-5. Data from these failed probes were not included in this report. Partial deterioration of the remaining ANL-3 probe was seen during this period. The probes have probably been damaged by corrosion of the metal parts (Reference 7). At the present time, a more damage-resistant replacement for these probes has been found, but new components have not been procured or installed. Occasional erratic behavior of some

ORNL probes seen in the past has been reduced to a single spike on several outputs. The bottom temperature probes in ORNL-3 and -5 have consistently indicated elevated soil temperature (Figures B-13 and B-17). Since the abnormal readings began soon after lysimeter installation, it is possible that probes or wiring were damaged at that time. The probe in ORNL-5 was later repaired but continues to read high. The bottom probe in ORNL-1 has also given elevated temperature readings recently (Figure B-9). All of the other temperature probes at ORNL were functioning prior to the failure of the DAS, including the probes at the 77.9-cm elevation, which are close to the waste forms.

Lysimeter Soil Moisture Data

Data from the moisture probes at both ANL-E and ORNL, shown in Figures C-1 through C-20 in Appendix C, indicate that the lysimeter soil columns at both sites have remained moist during the reporting period.

The moisture probe output from the soil column of each lysimeter over time (as determined by averaging the outputs of the three probes in each lysimeter) showed that the variation in detected moisture among the lysimeters at each site was relatively similar and not excessive (Table 12). There was a coefficient of variation maximum (CV) of 28.2% at ANL-E and 10.1% 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 combined moisture/temperature probes at ANL-E are no longer functioning. This condition was discussed in the previous section.

Soil moisture in the soil column of the lysimeters at each site is quantified gravimetrically once each year (see Tables D-1 through D-4 of Appendix D). 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 12). Percent differences between the gravimetric data and moisture probe data for ANL-E lysimeters range between a low of 20.3% in 1994 to a high of 52.0% in 1995. These values have increased significantly during this reporting period, but are well within a reasonable range given the use of the information. As in the past, data from the ORNL probes continue to overestimate the actual percent soil moisture from a low of 62.5% in 1994 to a high of 187.2% in 1995.

In addition to using the moisture probe and gravimetric data to calculate soil moisture starting in 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 7.3%, but underestimated those values (see Tables D-1 and D-2 of Appendix D). 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. Those data are given in Tables D-3 and D-4 of Appendix D. Comparison of the ORNL neutron probe results to gravimetric results, in overall average values, shows that the probe overestimated by 10.2%. In spite of the difference between gravimetric and measured soil moisture at ANL-E, the accuracy appears very good at ORNL. Therefore, it can be said that the use of the neutron probe provides a rapid, accurate estimate of moisture in the soil column.

Soil moisture (as gravimetrically determined) at each sampling depth has remained uniformly consistent between intrasite lysimeters during the past several years (Figures 9 and 10). The uniformity of soil moisture in the ANL-E lysimeters (Figure 9) 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 12, D-1, and D-2). While action to improve drainage of the ANL-E lysimeters was taken early in the

Table 12. Comparison of the average percent moisture values in lysimeter soil column as determined from probe and gravimetric data.

Lysimeter number	Period	Average percent moisture for soil column moisture probes[a]	Average percent moisture for soil column determined gravimetrically	Percent difference between gravimetric and probe
ANL-1	1993–94	14.9 ± 3.1	21.7 ± 2.9	31.3
ANL-2		14.6 ± 0.9	21.9 ± 3.3	33.3
ANL-3		26.7[c]	22.2 ± 2.8	20.3
ANL-4		16.5 ± 6.4	22.7 ± 2.3	27.3
ORNL-1		27.3 ± 10.3	16.8 ± 1.3	62.5
ORNL-2		36.1 ± 4.4	14.6 ± 2.0	147.3
ORNL-3		35.5 ± 0.8	14.8 ± 3.3	139.9
ORNL-4		38.5 ± 3.0	16.6 ± 4.1	131.9
ANL-1	1994–95	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.

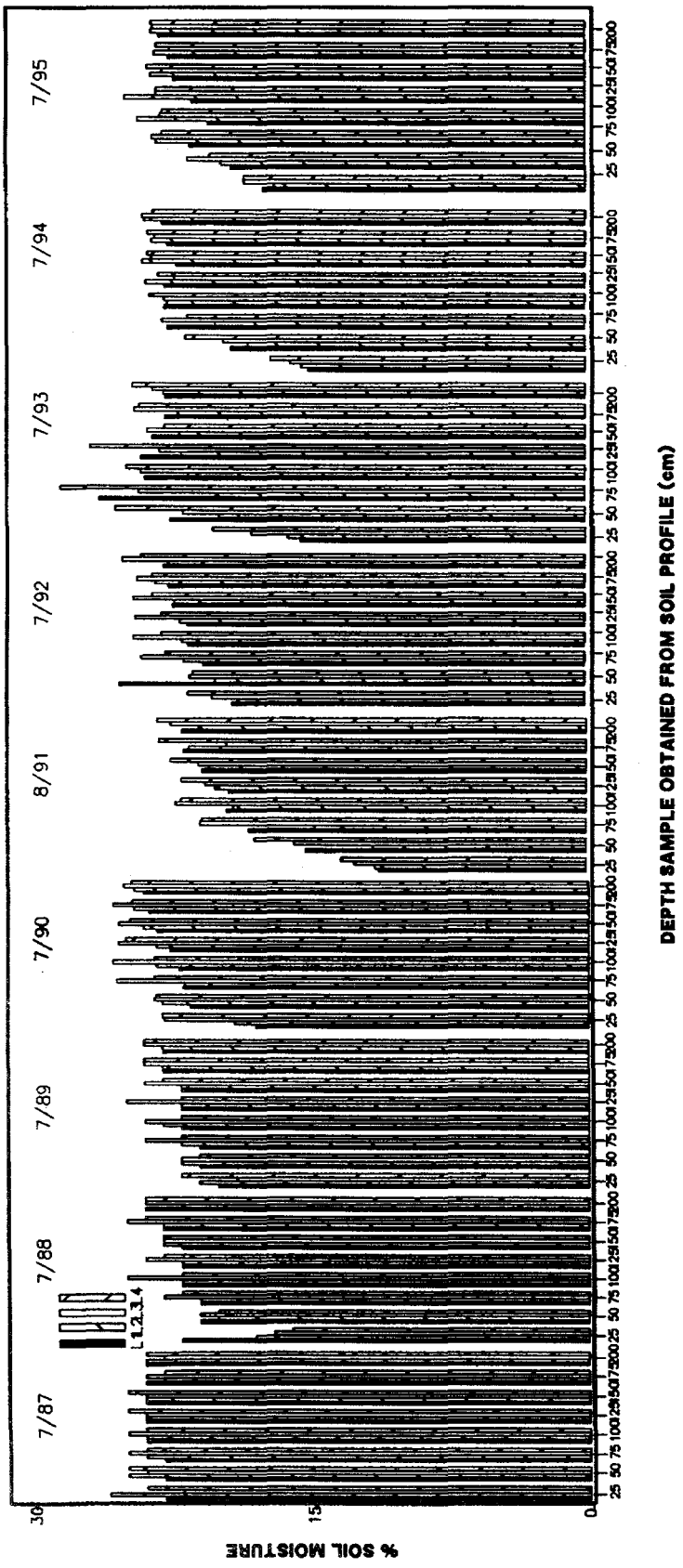

Figure 9. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.

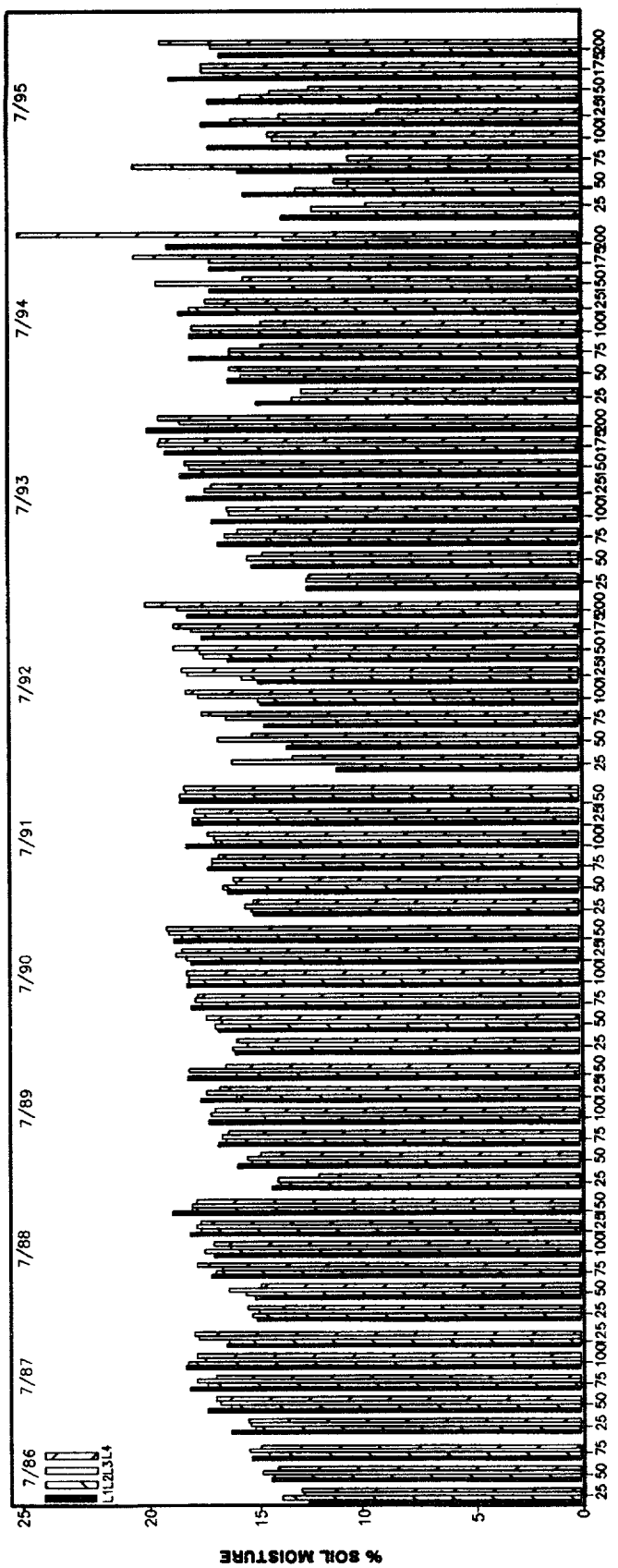

Figure 10. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.

experiment, initial drainage rates cannot be restored. Observations of surrounding indigenous soils have confirmed that this soil has a low permeability after being disturbed. Therefore, the present conditions within the lysimeters are indicative of what would be found if a disposal trench were constructed in the same soil. Since FY 1989, no efforts have been made to improve drainage of these lysimeters. Instead, water is no longer allowed to pond on the soil surface. Water in excess of 2–3 cm in depth is now removed from the lysimeter surfaces. Records of the amounts of water removed are maintained for use in the water balance calculations. Water accumulation at ANL-E during the reporting period occurred in all soil lysimeters and is reported in Table 13.

As shown in Figures 9 and 10, the amount of moisture within the deeper horizons of the lysimeter soil columns at each site appears to have remained fairly constant (see Tables D-1 through D-4). 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 in 1994, while at ORNL, this value decreased from 35.2% for 1994 to 30.9% for 1995. These values have remained fairly constant from year to year. For this reporting period, the ORNL percent moisture at depth was not as uniform as has been seen in the past (Figure 10).

Measurement of Leachate

By using the cumulative rainfall data from each site since the time the lysimeters were placed in operation (Figure 8), it is possible to calculate the approximate volume of water that has been received by the exposed surface (6,489.5 cm^2) of each lysimeter. The cumulative volume of precipitation received by each ANL-E lysimeter has now reached 5,957.4 L; at ORNL, this value is 8,682.9 L. Precipitation per year is listed in Table 14 as well as average volume of leachate through the lysimeters. The volume of precipitation that has passed through the lysimeters can be seen graphically in Figures 11 and 12. The throughput of precipitation is dependent on site conditions and lysimeter fill material. At ANL-E, an average of 2,461 ± 105.8 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 data are comparable year to year and reflect a high percentage of precipitation throughput at ORNL. The ORNL lysimeter soils are more permeable than the ANL-E soils (an observation made by comparing cumulative leachate through the control lysimeter at each site with cumulative leachate through soil lysimeters at that site, which are shown in Figures 11 and 12). 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, which have a deviation of 43.0%.

Table 13. ANL-E water removed from surface of lysimeters after precipitation accumulation.

	Water removed from lysimeter surfaces (L)	
Lysimeter number	1994	1995
ANL-1	146	247
ANL-2	95	150
ANL-3	—	19
ANL-4	82	181

Table 14. Precipitation received and leachate passing through lysimeters at ANL-E and ORNL.

		ANL-E		ORNL	
	Test period	Cumulative volume (L)	Total (%)	Cumulative volume (L)	Total (%)
Precipitation received	1993–94	5,338.3	—	7,894.5	—
	1994–95	5,957.4	—	8,682.9	—
Average leachate passed through soil-filled lysimeters	1993–94	2,146 ± 939	40.2	7,066 ± 127	89.5
	1994–95	2,461 ± 1,058	41.3	7,785 ± 164	89.7
Leachate passed through sand-filled lysimeters	1993–94	5,318	99.6	7,997	101.3
	1994–95	6,081	102.1	8,864	102.1

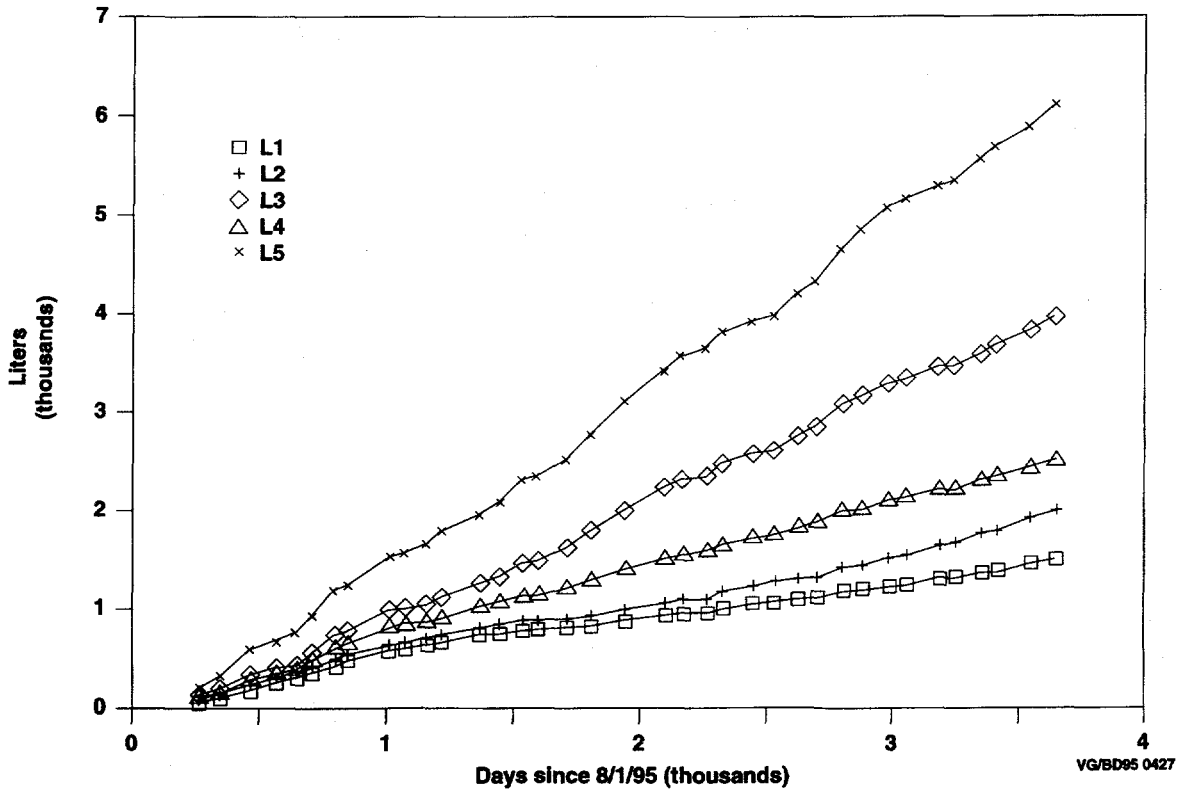

Figure 11. ANL-E cumulative volume of leachate from lysimeters.

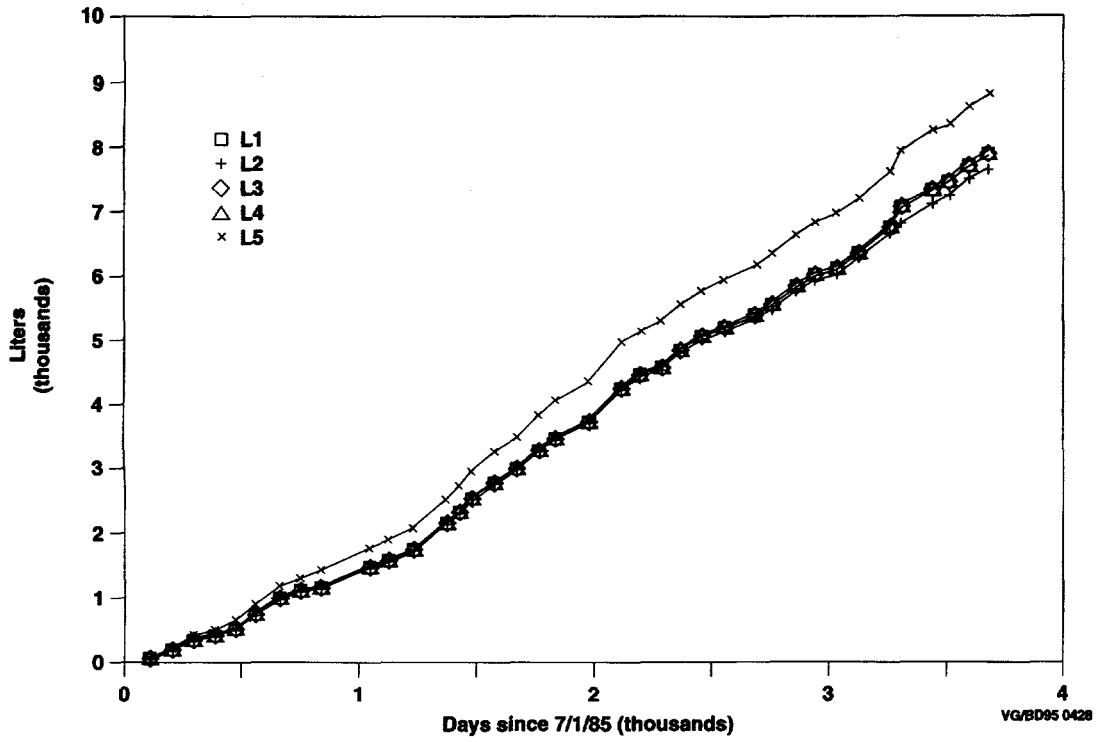

Figure 12. ORNL cumulative volume of leachate from lysimeters.

The data for ANL-E indicate that there is an increasing disparity in water balances for the ANL-E soil lysimeters. However, a comparison of the total amount of water associated with each of these lysimeters (water removed from the surface plus the quantity of leachate) shows that each of the lysimeters is exposed to equal volumes of water. During the past year, each lysimeter had a total of 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 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. The lysimeters at each site received comparable volumes of water; however, those quantities did not move through the lysimeters at each site in equal amounts due to the differences in soil texture and to weather conditions (Figures 11 and 12).

Soil used at ANL-E[22] is heavier (contains more fine material such as silts and swelling clay) than the soil used at ORNL.[23] Therefore, infiltration and percolation of water through the ANL-E soil would be expected to be significantly reduced in comparison to ORNL soil. The effect of weather is not apparent when comparing the sand-filled control lysimeters at the two sites. At both ANL-E and ORNL, 100% of the volume of precipitation passed through those lysimeters. At ANL-E, precipitation came during the months of November through March when the average air temperature was below 0°C. This precipitation then was in the form of freezing rain or snow that would not penetrate the frozen soil surface and could have been blown off (in the case of snow) or lost due to sublimation. Other factors such as generally gustier winds and lower humidity at ANL-E indicate that evaporation of water from the ANL-E lysimeters could have been higher than at ORNL. Also as noted earlier, ANL-E lysimeters 1, 2, 3, and 4 have experienced water ponding during periods of

heavy rainfall. To prevent loss of precipitation, that water was drained from the surface of those lysimeters.

Therefore, if nuclides were mobilized by the water surrounding the waste forms, the greatest opportunity for detection would be found in water from the ORNL site. This is based on two assumptions: (a) the nuclide is water soluble and (b) the soil column does not interfere with nuclide movement.

Radionuclide Analysis

Water samples are normally collected on a quarterly basis from leachate collectors and moisture cups in each of the lysimeters during each 12-month period. At each sampling, water from the leachate collectors (1 L of collected quantity) and those cups (0.1 L of the collected quantity) closest to the waste forms (cup 3) is 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 from cup 1 (directly below cup 3), then cup 4, followed by cup 2, (see Figure 7 for cup placement) should be analyzed. 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, starting in 1991, water from cups 1 has been analyzed and reported. In 1993, water from cups 2 has also been analyzed and reported.

Tabulated results of beta and gamma analysis for the samples taken during the reporting period are found in Tables E-1 through E-4 in Appendix E. Four samples were taken at each site during each 12-month period. The cumulative amounts of nuclides as determined from water samples obtained from lysimeter number 3 cups and leachate collectors for all samplings during this period are given in Tables 15 and 16. Radionuclide cumulative amounts versus time are displayed graphically in Figures 13 through 21.

As has been reported in the past,[6-17] not all nuclides are appearing consistently in the water obtained from either the cups or leachate collectors. The nuclide that appears with the most regularity at both sites is Sr-90 (Tables 15 and 16 and Appendix E). This nuclide consistently occurs in significant amounts in all the number 3 cups at ANL-E and ORNL, and in the number 5 leachate collectors at both sites (Figures 13 through 16). There continues to be standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a cumulative total of 1,902,175 pCi from 3-3 at ANL-E (a 35% increase during this period) (Table 15 and Figure 13) and 357,308 pCi from 3-3 at ORNL (a 101% increase during the period) (Table 16 and Figure 14), which continues to increase beyond ORNL 1-3. 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 continues in ORNL 5-3 as well as in ORNL 4-3 (Figure 14). The above data show that significant quantities of Sr-90 continue to be transported from the waste forms.

As noted in the Resin Solidification section of Reference 8, during laboratory testing of similar waste forms, Sr-90 appears to move from these waste forms more rapidly than Cs-137. While the cumulative totals of Sr-90 appear large when compared to other lysimeter experiments, the highest total cup release, ANL 3-3, represents only about 0.005% of the waste form inventory in that lysimeter (Table 17).

At ANL-E, Sr-90 retrieved from number 3 cups of the soil lysimeters during the tenth year ranges from 380% to 660% of that found in the respective leachate collectors (Table 15), while at ORNL, these values are between 7% and 450% (Table 16). These are increases over previous years and are the result of both an increased quantity of Sr-90 moving into the area near the moisture cups and a decrease in the movement of the nuclide through the entire soil profile into the leachate collectors.

Table 15. ANL-E total cumulative radionuclide Sr-90 and Cs-137 extracted from lysimeters.

Test period	Operating days	Sr-90 in moisture cups (pCi)					Sr-90 in leachate collectors (pCi)					Cs-137 in moisture cups (pCi)	
		1-3	2-3	3-3	4-3	5-3	1	2	3	4	5	2	5
1993–94	2,982	16,232	8,915	1,472,575	34,699	26,247	6,692	2,132	222,763	5,287	2,568,229	2,889	74,071
	3,052	16,522	9,815	1,536,075	38,699	28,977	6,711	2,186	228,292	5,763	2,678,773	2,910	82,261
	3,184	17,122	10,645	1,588,075	44,499	31,617	6,737	2,288	257,316	6,834	2,909,955	2,929	93,241
	3,247	18,082	11,705	1,628,875	49,599	35,897	6,759	2,312	259,276	6,900	3,011,865	2,929	114,841
1994–95	3,352	19,402	12,485	1,686,375	54,069	39,185	6,759	2,312	313,522	8,176	3,363,562	2,989	151,321
	3,410	21,172	13,505	1,735,175	58,821	40,980	6,759	2,312	356,805	8,842	3,562,296	3,021	168,121
	3,542	24,484	13,940	1,840,175	62,531	45,310	6,759	2,312	402,191	9,639	3,845,924	3,044	191,721
	3,641	25,654	14,550	1,902,175	66,101	49,097	6,759	2,312	440,240	10,060	4,158,404	3,085	224,201

Table 16. ORNL total cumulative radionuclide Sr-90 and Cs-137 extracted from lysimeters.

Test period	Operating days	Sr-90 in moisture cups (pCi) 1-3	2-3	3-3	4-3	5-3	Sr-90 in leachate collectors (pCi) 1	2	3	4	5
1993–94	3,031	138,876	18,722	203,377	5,427	20,790	598,753	31,734	24,823	21,075	2,831,321
	3,126	146,476	19,822	226,277	6,627	20,790	812,408	42,140	28,483	21,075	3,226,205
	3,254	151,876	20,822	242,477	7,465	21,736	1,181,768	54,044	28,483	21,075	3,492,295
	3,300	159,976	22,222	274,877	9,165	24,236	1,295,843	65,928	37,258	21,075	4,045,025
1994–95	3,434	167,273	23,195	307,309	10,733	24,330	1,736,170	84,714	45,778	21,348	4,790,825
	3,505	168,192	24,087	307,309	12,090	24,330	1,923,385	103,317	55,438	21,348	5,125,325
	3,588	173,868	24,979	330,822	13,495	26,384	2,332,372	132,027	67,884	21,427	5,545,423
	3,672	180,084	25,790	357,308	14,846	28,979	2,596,758	152,487	79,104	21,592	5,980,597
Test period	Operating days	Cs-137 in moisture cups (pCi) 1-3	2-3	3-3	4-3	5-3	Cs-137 in leachate collectors (pCi) 1	2	3	4	5
1993–94	3,031	0	0	0	0	5,794	5,300	2,040	6,415	6,178	292,324
	3,126	0	0	0	0	5,815	5,300	2,040	6,415	6,178	295,600
	3,254	0	0	0	0	6,518	5,300	2,040	6,415	6,178	347,670
	3,300	0	0	0	0	8,618	5,300	2,040	6,415	6,178	4,871,810
1994–95	3,434	0	0	0	0	10,753	5,300	2,040	6,415	6,178	5,763,710
	3,505	0	0	0	0	10,753	5,300	2,040	6,415	6,178	5,962,960
	3,588	0	0	0	0	12,726	5,300	2,040	6,415	6,178	6,116,406
	3,672	0	0	0	0	17,861	5,300	2,040	6,415	7,333	6,215,226

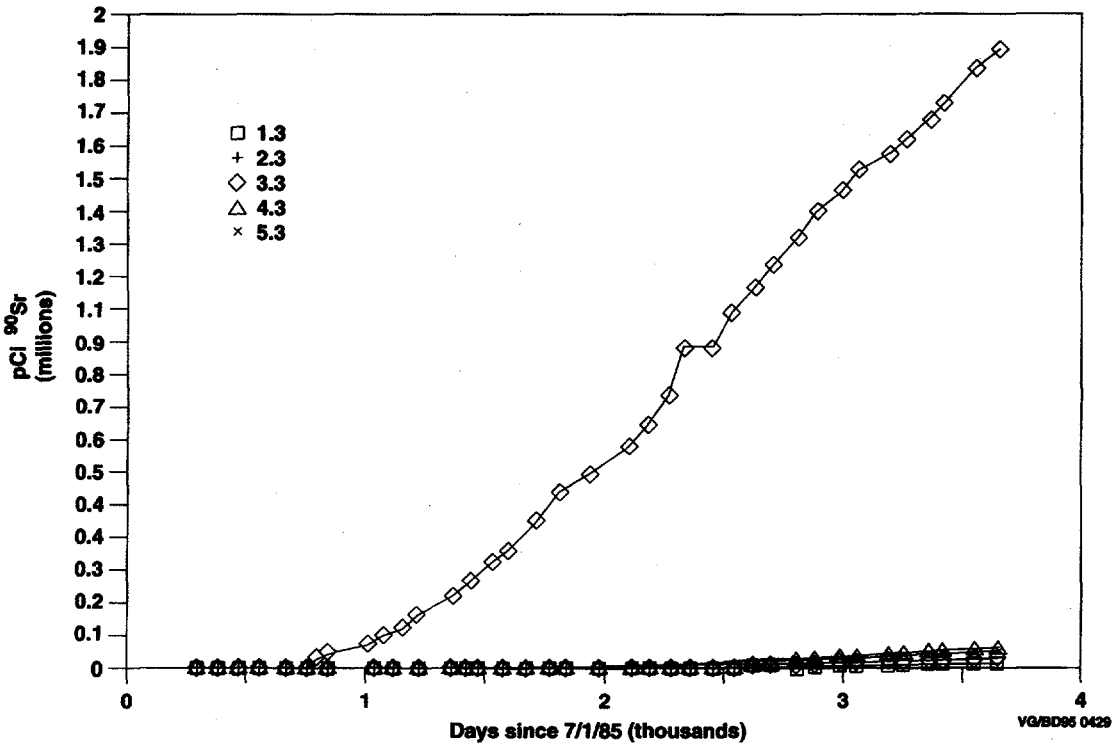

Figure 13. ANL-E cumulative Sr-90 collected in moisture cups number 3.

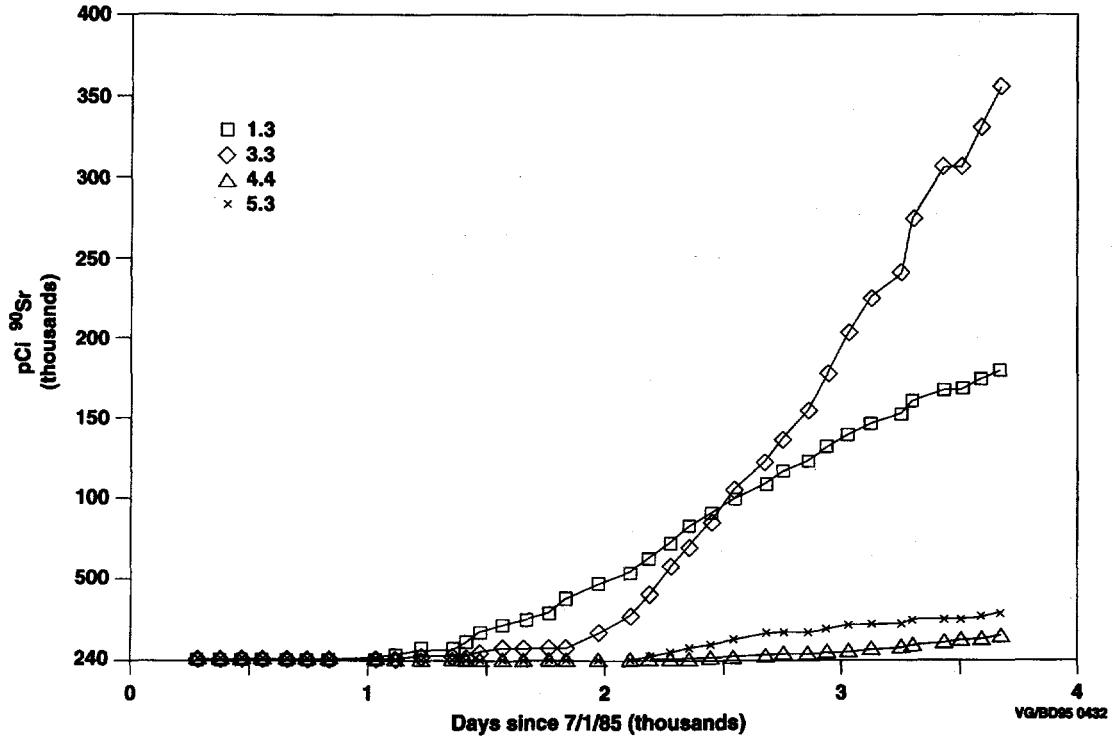

Figure 14. ORNL cumulative Sr-90 collected in moisture cups number 3.

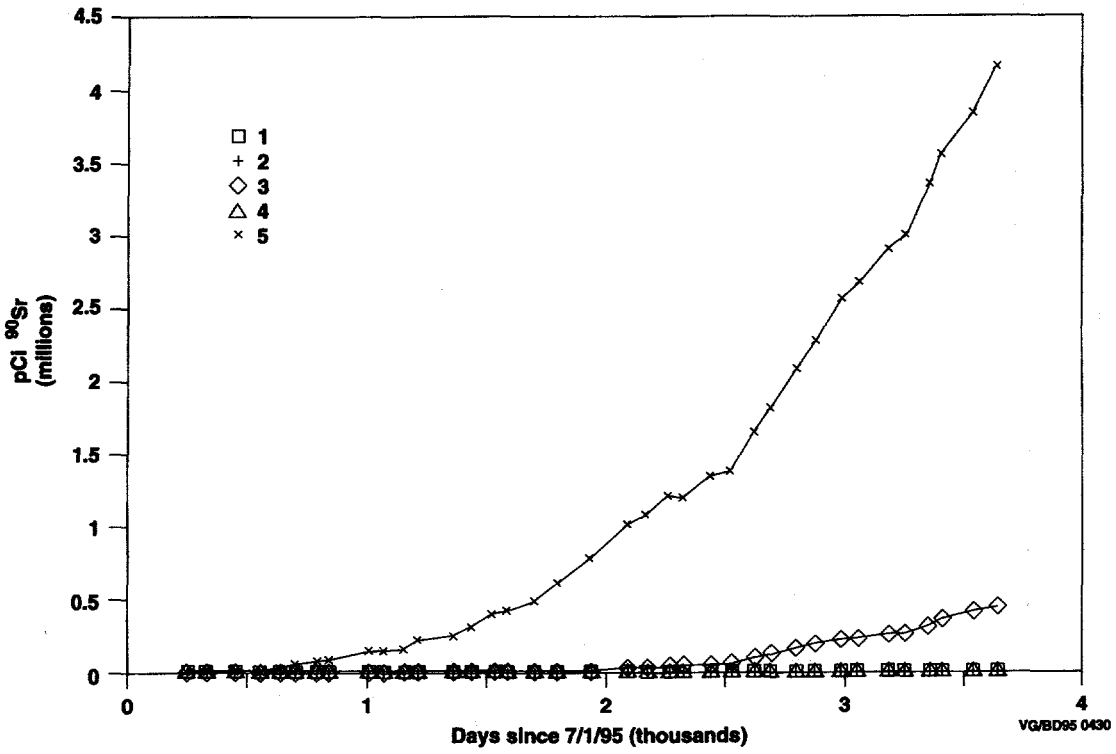

Figure 15. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

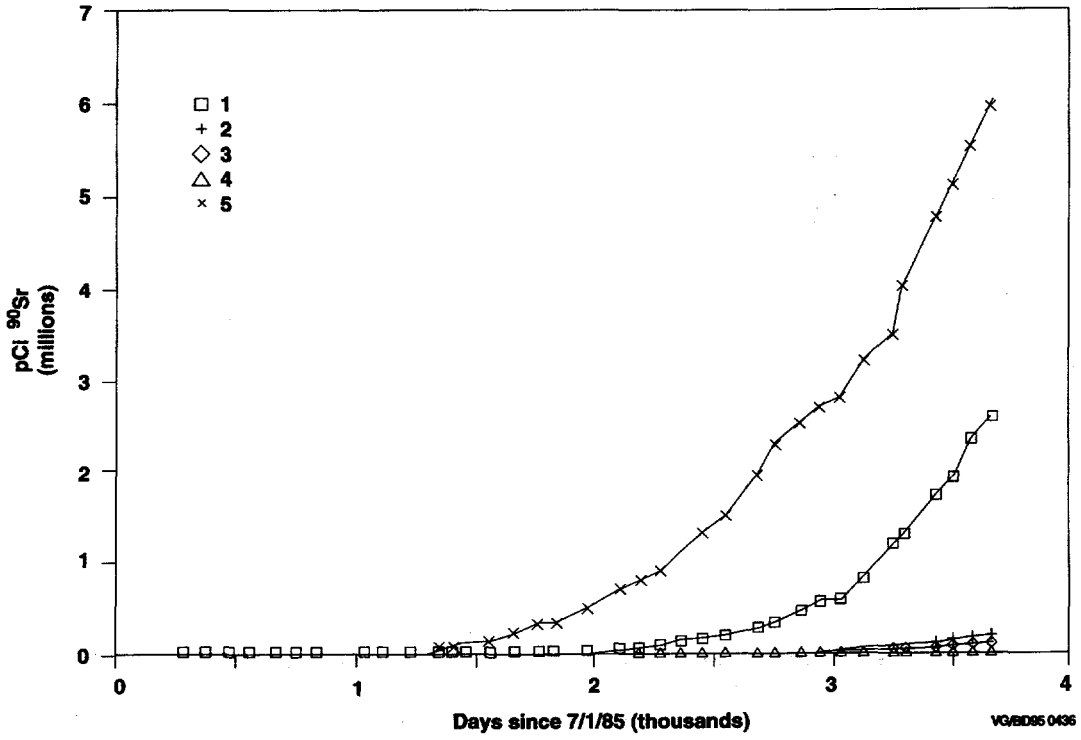

Figure 16. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

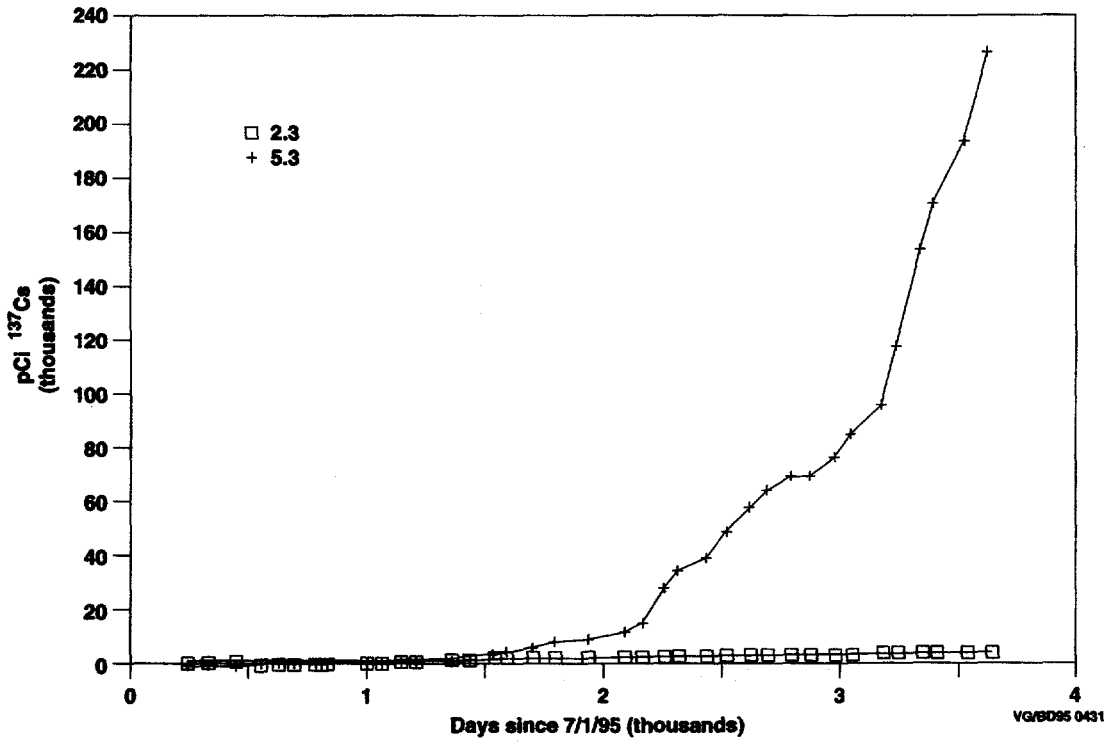

Figure 17. ANL-E cumulative Cs-137 collected in moisture cups number 3.

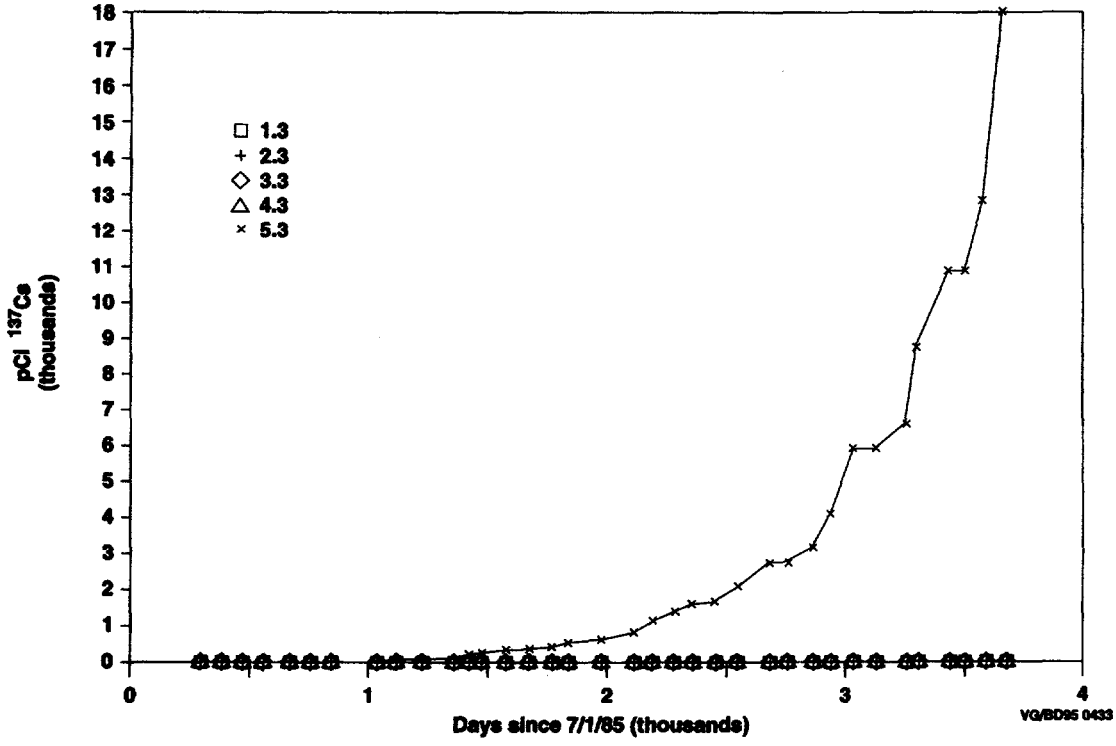

Figure 18. ORNL cumulative Cs-137 collected in moisture cups number 3.

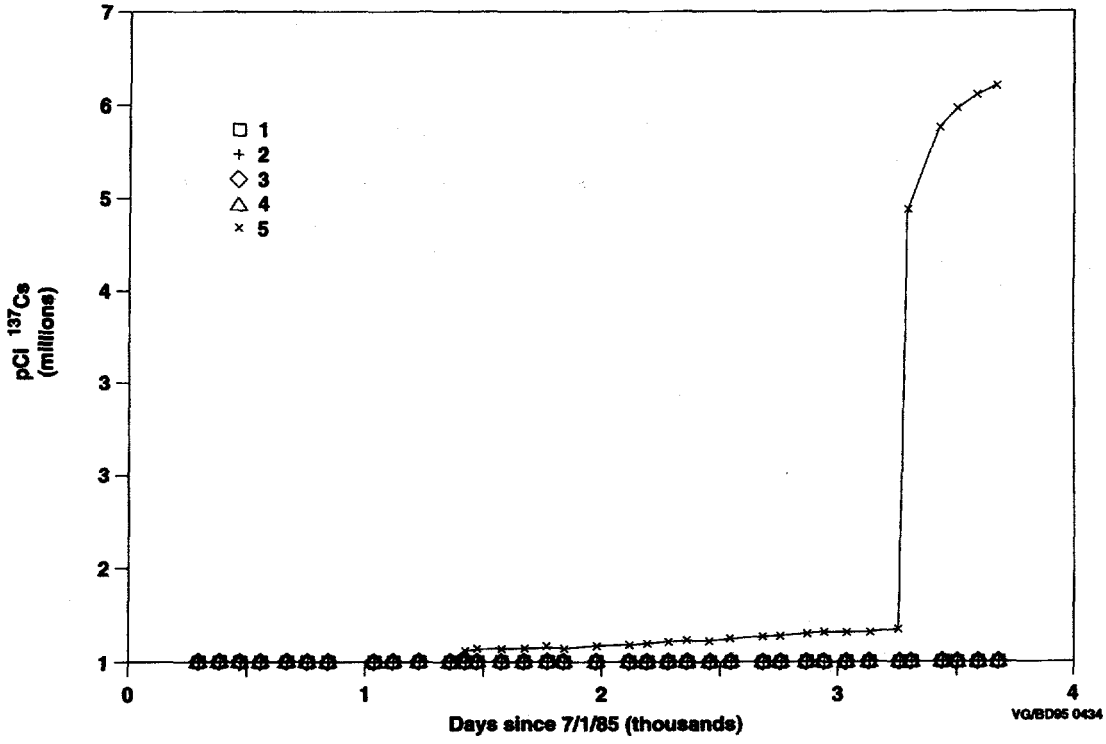

Figure 19. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

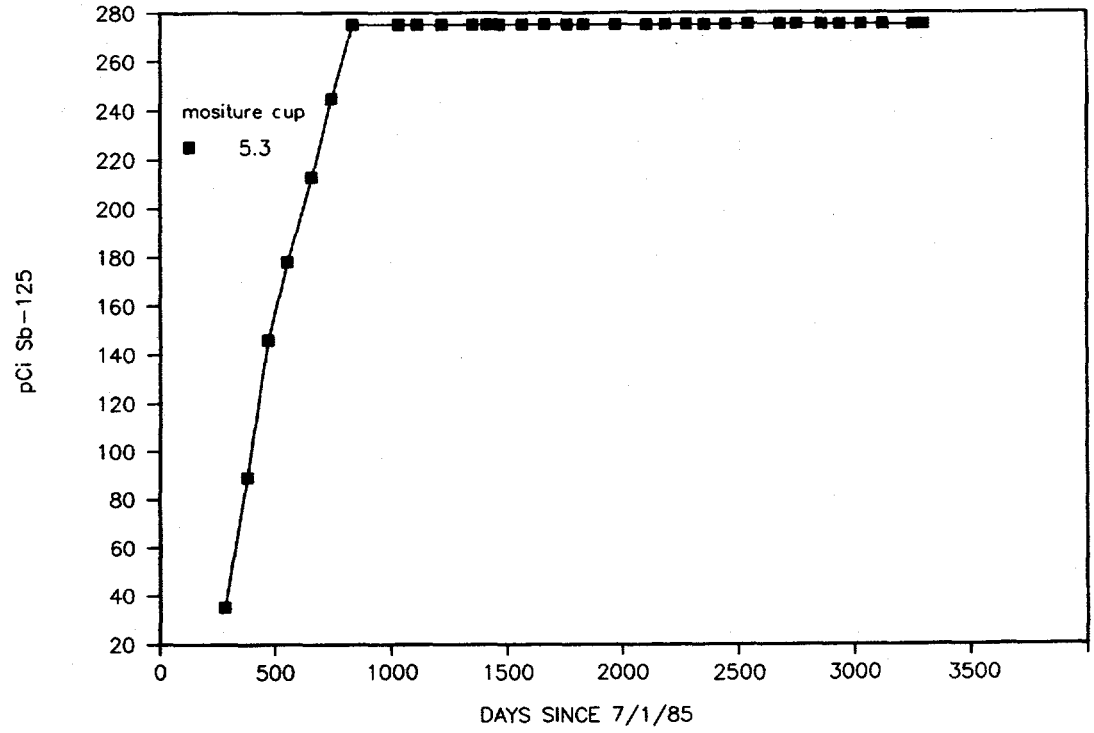

Figure 20. ORNL cumulative Sb-125 collected in moisture cup number 3.

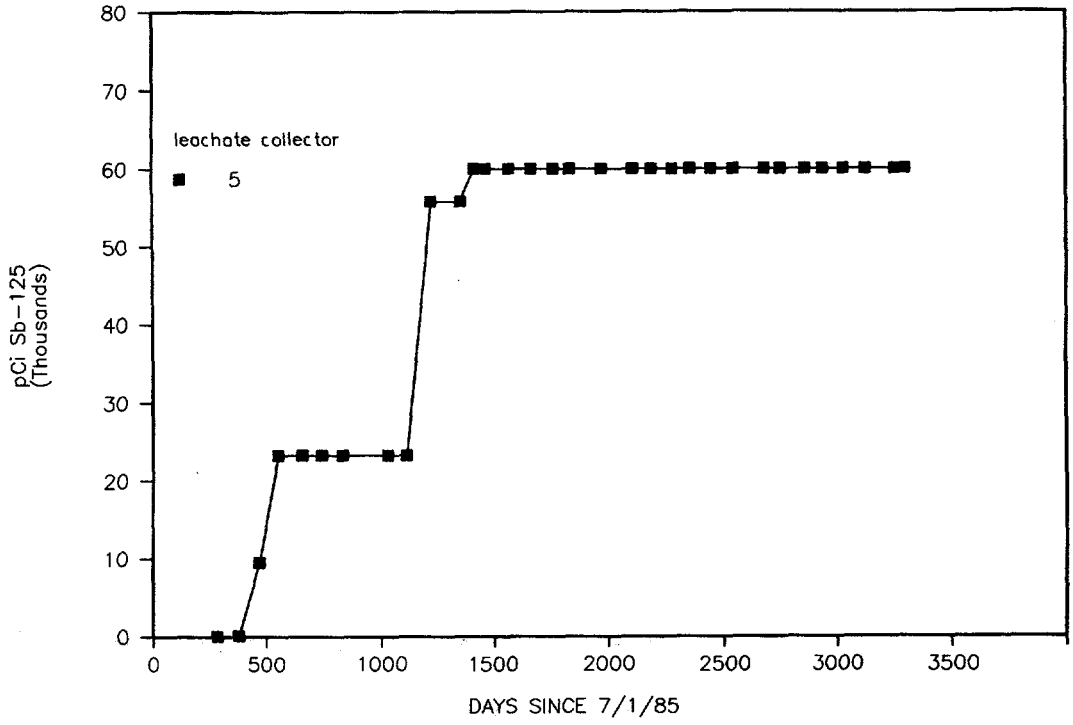

Figure 21. ORNL cumulative Sb-125 collected in lysimeter leachate collectors.

During the past 2 years, 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 any ANL-E soil lysimeter (Figure 15). This is comparable to the previous years' findings (References 6, 7, 8, 9, 11, 12, 13, 14, 16, and 17). There have been substantial releases of Sr-90 from the soil-filled lysimeters at ORNL. Sr-90 content in leachate from lysimeters 1 through 3 has increased between 300 and 500% during the period (Table 16). High Sr-90 release into ORNL-1 placed it within the same order of magnitude as ORNL-5 (2,596,758 to 5,980,597 pCi). For leachates from soil lysimeters, intersite-comparable percentages of total inventory of Sr-90 were found in ANL-3 and 4 and ORNL-3 and 4 (Table 17). There continued to be a significant increase in the total cumulative quantity of Sr-90 released in the leachate water in all lysimeters at both sites this period except ANL-1 and 2 (Tables 15 and 16).

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 17). 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 two ORNL lysimeters. For the control lysimeters at both sites, there was substantially more Sr-90 in the leachate than in cups 3 (there continues to be over two orders of magnitude difference for both locations).

The percent of total Sr-90 being measured in the leachate water and cups 3 continues to be somewhat inconsistent between the two sites (Table 17). Perhaps this represents a difference in how the environment at the two sites affects the movement of Sr-90 being released from the waste forms. This difference is also seen when the percent of total Sr-90 found in the leachate water from the two control lysimeters is examined. The percent passing through the ORNL control was 8.0 times that of ANL-E (Table 17).

Table 17. Percent of total Sr-90 and Cs-137 inventory per lysimeter extracted from moisture cups and leachate water through July 1995.

Lysimeter number	Solidification agent	Liner number	Total inventory[18,19,20] per lysimeter (pCi)[a]		Percent total inventory Sr-90		Percent total inventory Cs-137	
			Sr-90	Cs-137	Moisture cups	Leachate water	Moisture cups	Leachate water
ANL-1	Cement	PF-7	18.5E+9	3.1E+11	1.4E-4	0.4E-4	—[b]	—
ANL-2	Cement	PF-24	3.3E+9	14.3E+11	4.4E-4	0.7E-4	0.2E-6	—
ANL-3	VES	PF-7	27.4E+9	4.6E+11	69.4E-4	16.1E-4	—	—
ANL-4	VES	PF-24	4.5E+9	19.3E+11	14.7E-4	2.2E-4	—	—
ANL-5	Cement	PF-7	18.5E+9	3.1E+11	2.7E-4	225.0E-4	71.9E-6	—
ORNL-1	Cement	PF-7	18.5E+9	3.1E+11	9.7E-4	143.0E-4	—	1.7E-6
ORNL-2	Cement	PF-24	3.3E+9	14.3E+11	7.8E-4	46.0E-4	—	0.1E-6
ORNL-3	VES	PF-7	27.4E+9	4.6E+11	13.0E-4	4.5E-4	—	1.4E–6
ORNL-4	VES	PF-24	4.5E+9	19.3E+11	3.3E-4	4.8E-4	—	0.4E-6
ORNL-5	Cement	PF-24	3.3E+9	14.3E+11	8.8E-4	1,812.0E-4	1.3E-6	434.0E-6

a. Activities of radionuclides have not been decay corrected from date of measurement (9/20/83 for Cs-137 and 10/25/83 for Sr-90).

b. Percent release is essentially equal to zero.

Gamma-producing nuclides continue to occur with regularity at both sites. ANL 2-3, below a cement waste form containing large amounts of Cs-137, continues to receive sporadic quantities of Cs-137 (Tables E-1 and E-2; Figure 17). Since Cs-137 began appearing in ANL 5-3, the quantity of this nuclide has dramatically increased in each of the sampling periods with significant increases during the last 4 years (300% in 2 years) (Figure 17). 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 18) and have continued in subsequent samplings for a total of 17,861 pCi (100% increase in the last year). Detectable amounts of Cs-137 have been consistently found in leachate water from ORNL-5 (Figure 19) and sporadically in the other ORNL waters, though none have been found during the past 3 years except in lysimeter 4 in the last sampling (Table 16). 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 18 and 19). In 1994, there was a dramatic increase of over 1,500% in Cs-137 to that collector. Thus far, a total of 6,215,226 pCi has passed through to that collector. It appears that a pulse of Cs-137 is moving through that coil column.

For 6 years in a row, Sb-125 has not been found in ORNL-5 leachate water. Also, this is the seventh year of its absence in ORNL cup 5-3 (Figures 20 and 21; Tables E-3 and E-4).

By using a matrix (as in Table 17), several comparisons can be made based on the intra- and inter-site data. Overall, of the nuclides contained in the waste forms, 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 17). Compared to Sr-90, the recovery of Cs-137 appears insignificant. There have been significant occurrences of Cs-137 in cups 3 of the ORNL soil lysimeters during past years, and there was evidence of its reoccurrence in ORNL 1-3, 2-3, and 3-3 (Tables E-3 and E-4). On the other hand, this nuclide has been consistently occurring in ORNL 5-3 (Figure 18) and in the leachate collector of the ORNL-5 lysimeter (Figure 19). Cesium-137 has also occurred in the moisture cups of ANL-E lysimeters 2 and 5 but not in the leachate water. More Cs-137 has passed through the ORNL lysimeters than those at ANL-E.

At ANL-E, a comparison of Sr-90 occurrence in cups 3 and the leachate collectors of the soil-filled lysimeters (Table 17) 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 (Table 14; Figures 11 and 12). 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 17). 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 Tables 2, 5, and 17, the lysimeters at both sites have been loaded with waste forms based on solidification agent and total nuclide content. Numbers 1, 2, and 5 were solidified with cement; numbers 3 and 4 with VES. ANL-1, -3, and -5, and ORNL-1 and -3 contain 5% of activity as Sr-90; the others contain 1% of activity as Sr-90 (Reference 18). This provides a total of five matched sets for the sites (ANL-1 and -2, ANL-3 and -4, ORNL-1 and -2, ORNL-3 and -4, and ANL-5 and ORNL-5). It could be assumed that nuclide leaching from these waste forms would be proportional to content (i.e., those with the higher loading would have proportionally larger Sr-90 releases, but the total percent of release should be close to the same).

The first part of this assumption appears to be correct in the case of Sr-90 movement into cups 3 for both sites when compared to other cups at that site (Table 17). Figures 13 and 14 show that cumulative quantities of Sr-90 in water retrieved

from cups 3 are higher from the lysimeters with the higher loaded waste forms (range of 176 to 2,874% more). The same was also true for the four soil lysimeters 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 forms with a subsequent movement through the soil column.

The assumption of a uniform percent release of Sr-90 from the waste forms, however, is not supported by the data (Table 17). For the moisture cup soil water collection, only three of the five sets have a higher total percent released to the cup water from those lysimeters containing the higher loaded waste forms (124 to 472%), and only two of the five have the higher Sr-90 released to the leachate 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 forms) than in the other ANL-E cups 3 (Table 17). As has been noted, the length of the soil column appears to moderate the quantity of the nuclide that travels from the waste form to the leachate collector. The leachate collectors in those same ANL-3 and -4 lysimeters also receive a higher percentage of Sr-90 than the other ANL-E soil lysimeter collectors, but a significant amount less than the cups 3 (4 and 7 times, respectively). The percent 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 percent 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).

Recovery of Sr-90 in cups 3 at ANL-E is higher for those lysimeters containing VES waste forms than for those with cement waste forms. Recovery of Sr-90 in the ORNL cups is comparable for those lysimeters containing the cement waste forms and one of the two containing VES waste forms. In the leachate collectors, more Sr-90 is seen in the ANL-E lysimeters containing VES waste forms, while more is seen in these ORNL lysimeters holding cement waste forms. These data together with those from ANL-E continue to indicate that cement and VES have comparable releases.

On an intersite comparison, it can be seen that larger quantities of Sr-90 and Cs-137 are moving in the ORNL lysimeters (Table 17). Soil type and precipitation (environmental factors) appear to be the controlling factors.

Cumulative Fractional Releases Compared

As described earlier in this report, waste forms from the sample batches were tested to the requirements of the NRC BTP.[4] The test thought to be most representative of field conditions is the bench leach test performed in accordance with the American Nuclear Society "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes," ANS 16.1 (1986). That accelerated test was used as a primary tool to characterize the waste forms that are being tested in the field lysimeters. Table 18 is a comparison of the cumulative fractional releases (CFRs) of radionuclides to leachate collectors from 10 years of field testing EPICOR-II waste forms in lysimeters to releases from bench-leach-testing similar waste forms in demineralized and seawaters as reported in References 8 and 20. 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 lysimeter is only one or two orders of magnitude less than bench-test results with demineralized water. CFRs have been plotted with time in Figures 22 through 28 for both cups number 3 and leachate collector waters.

Table 18. Cumulative fractional releases from lysimeter field testing compared to those from bench leach testing (8,20).

Test type	Prefilter number	Solidification agent	Radio-nuclide	Cumulative fractional release			
				Demineralized water	Seawater	Leachate collectors: Soil	Leachate collectors: Sand
Bench,[a] INEL	7	Cement	Sr-90	7.8E-2	—	—	—
Bench,[a] INEL	7	VES	Sr-90	4.5E-2	—	—	—
Bench,[a] INEL	7	Cement	Cs-137	9.4E-2	—	—	—
Bench,[a] INEL	7	VES	Cs-137	4.6E-2	—	—	—
Bench, INEL	7	Cement	Cs-137	4.8E-2	9.0E-2	—	—
Bench, INEL	24	Cement	Cs-137	2.3E-2	2.6E-2	—	—
Bench, INEL	7	VES	Cs-137	2.1E-3	6.4E-2	—	—
Bench, INEL	24	VES	Cs-137	3.4E-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	Sr-90	—	—	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 forms were irradiated before test.

These plots are similar to cumulative release curves of Figures 13 through 19, with cup 3 with VES waste forms at both sites having the highest measured Sr-90 CFR (Figures 22 and 23). Cup 4-3 at ANL-E, below another VES waste form, is the only other ANL-E standout, while all of the ORNL cups 3 show significantly high CFRs. There is little difference between cumulative release and CFRs to collectors (Figures 24 and 25 compared to Figures 15 and 16) with sand-filled lysimeter collectors receiving much higher CFRs than soil-filled units (as seen in Table 18). The Cs-137 plots of CFR exhibit similar features to those of cumulative release (Figures 26 through 28 compared to 17 through 19) with the exception that only ORNL cup 5-3 is presented from that site. At the present rate of increase (Figures 22 through 28), 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

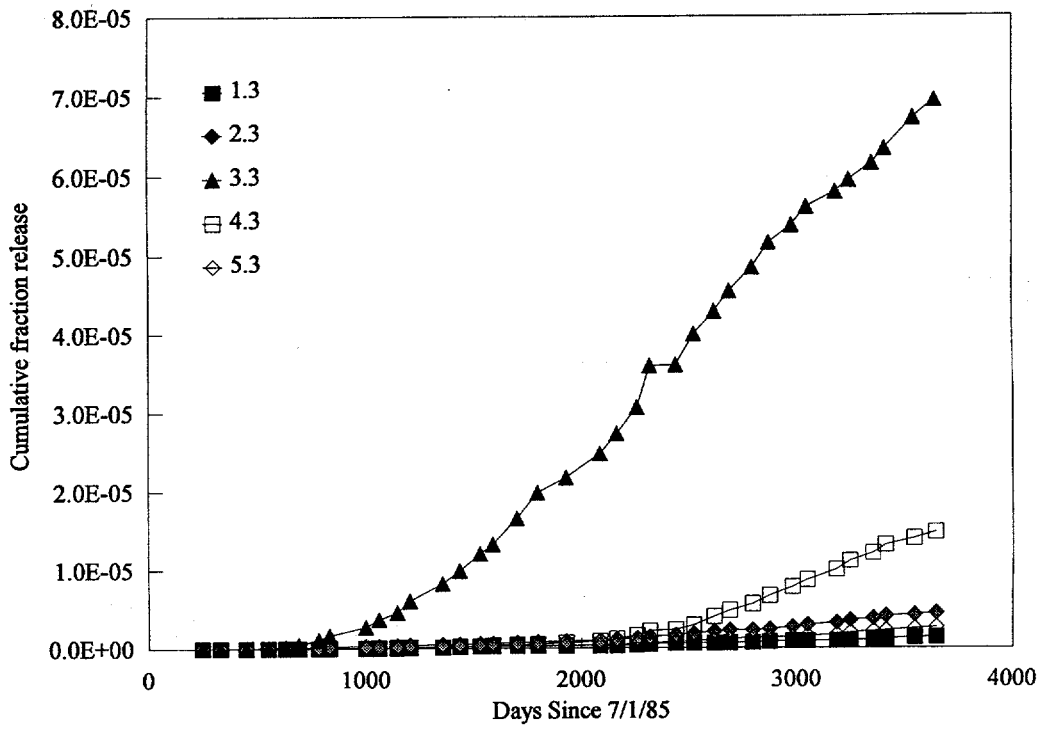

Figure 22. ANL-E cumulative fractional release of Sr-90 collected in moisture cup 3.

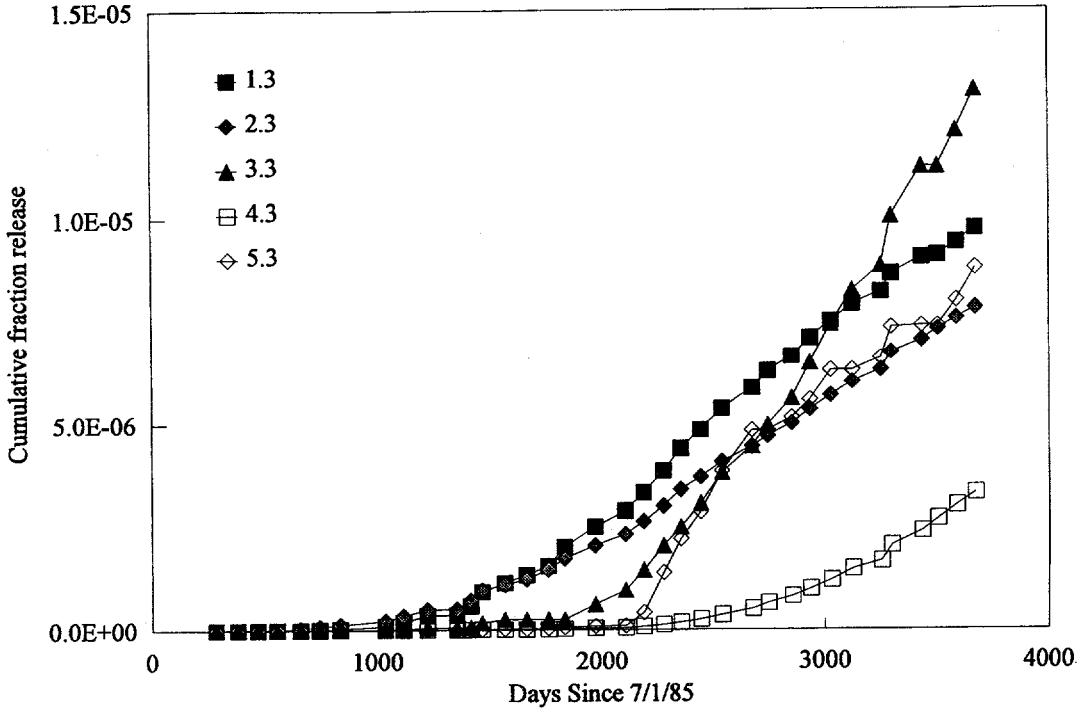

Figure 23. ORNL cumulative fractional release of Sr-90 collected in moisture cup 3.

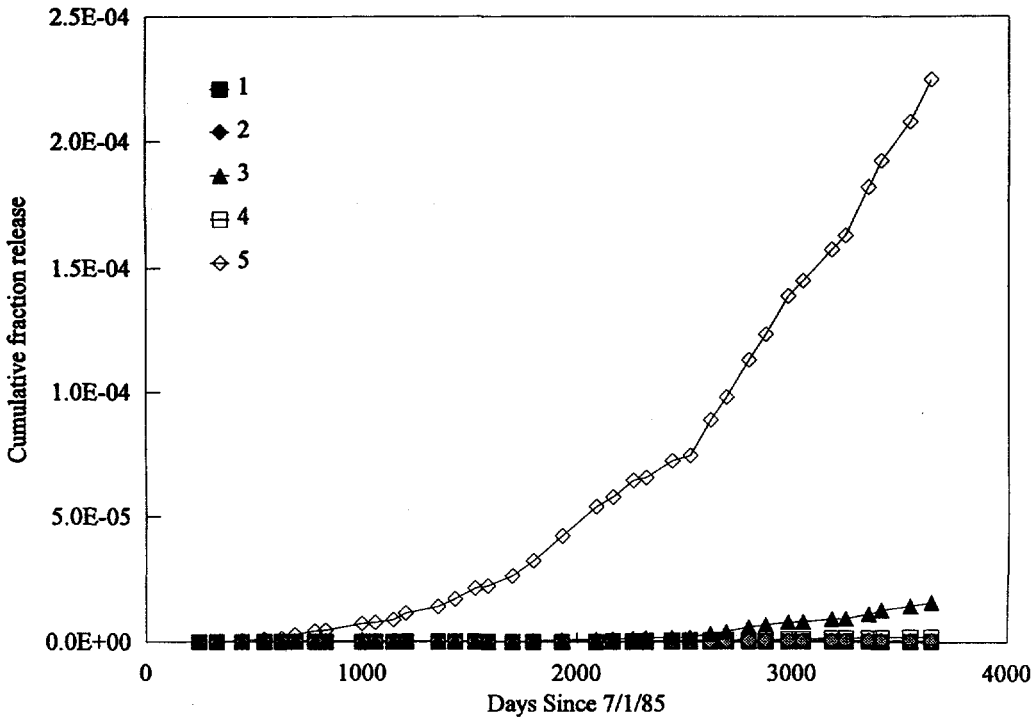

Figure 24. ANL-E cumulative fractional release of Sr-90 collected in lysimeter leachate collectors.

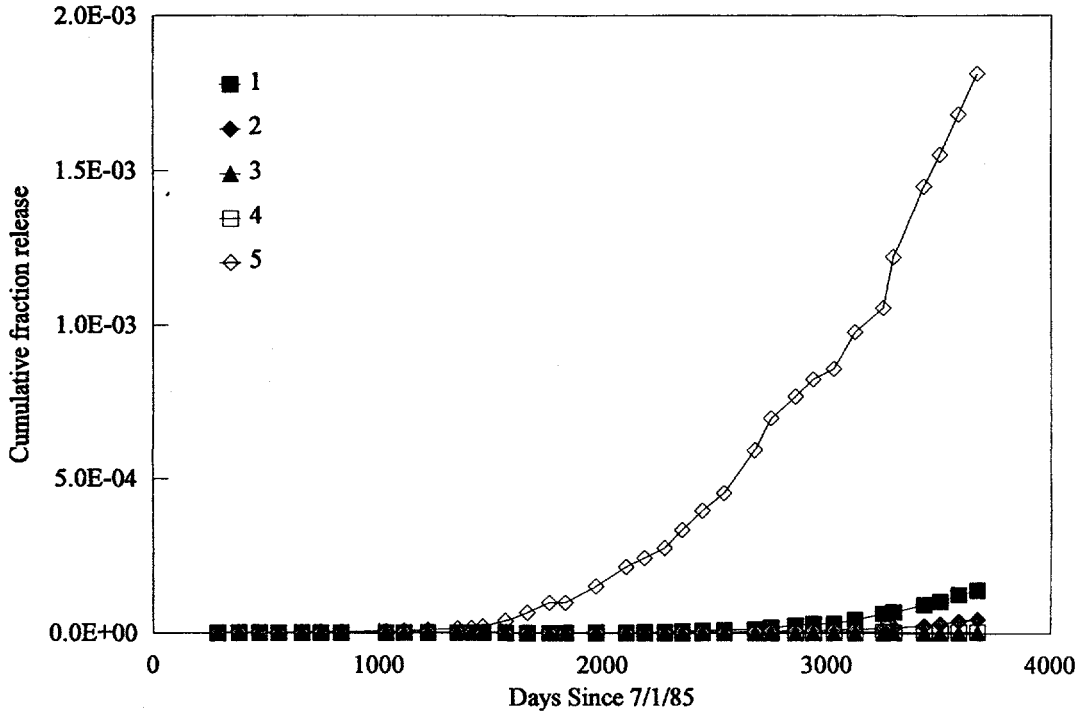

Figure 25. ORNL cumulative fractional release of Sr-90 collected in lysimeter leachate collectors.

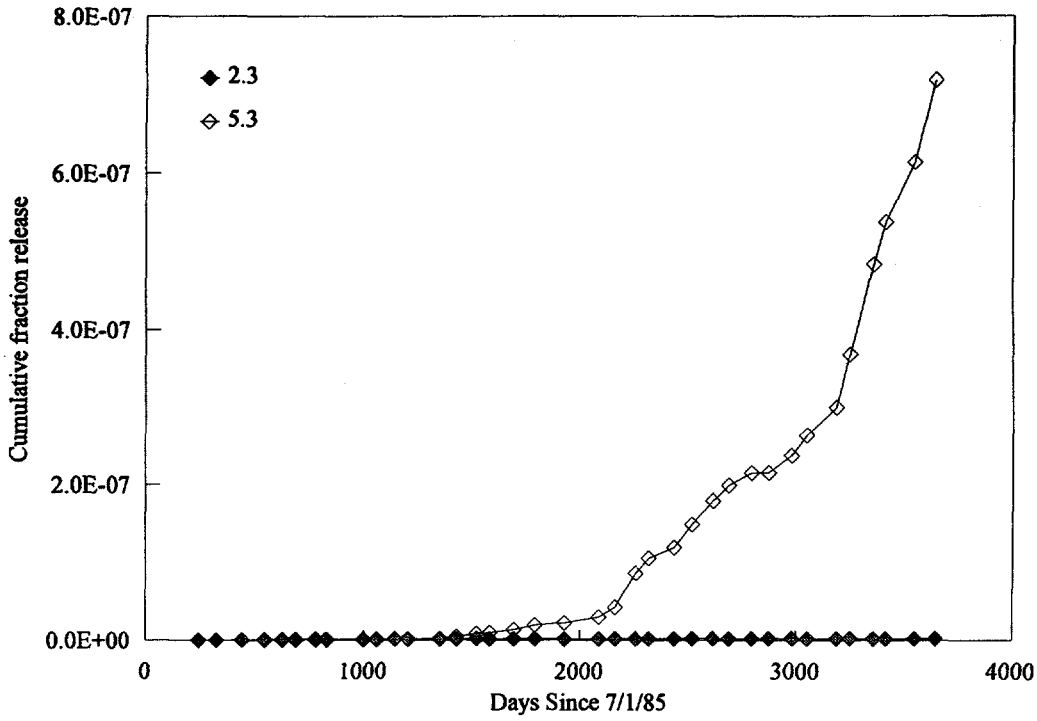

Figure 26. ANL-E cumulative fractional release of Cs-137 collected in moisture cup 3.

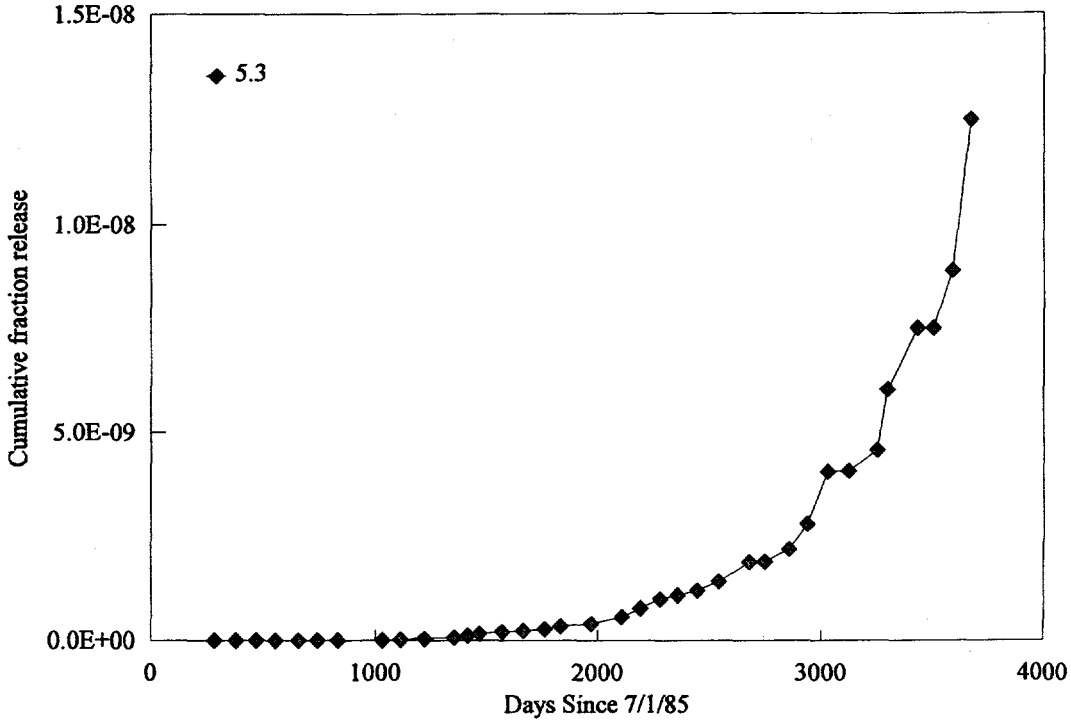

Figure 27. ORNL cumulative fractional release of Cs-137 collected in moisture cup 3.

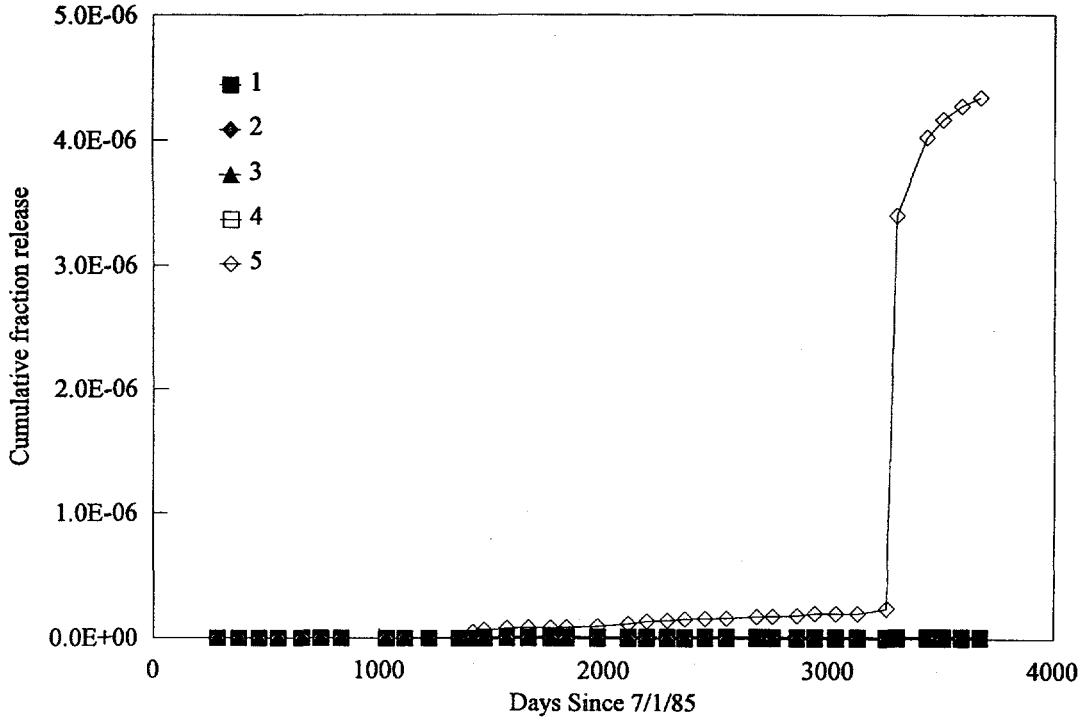

Figure 28. ORNL cumulative fractional release of Cs-137 collected in lysimeter leachate collectors.

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 forms (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. Cesium-137 was also found in all segments of the side core, but Cs-134 was found in only the upper half of that core (Table 19). Cesium-134 is an activation product that is formed in the core of nuclear reactors and does not occur naturally; therefore, the ratio of Cs-137 to Cs-134 in the sand segments can be used to determine if there was an outside source of cesium added to the lysimeter. By decay-correcting the original ratio of the waste form to the date of sand collection (and assuming that both radioisotopes behave chemically identical), the ratio should be equal to 399. The data presented in Table 19 and Figure 29 indicate that the ratio in all segments of the center core fall close to this value, except for segments 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 forms.

Table 19. 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.

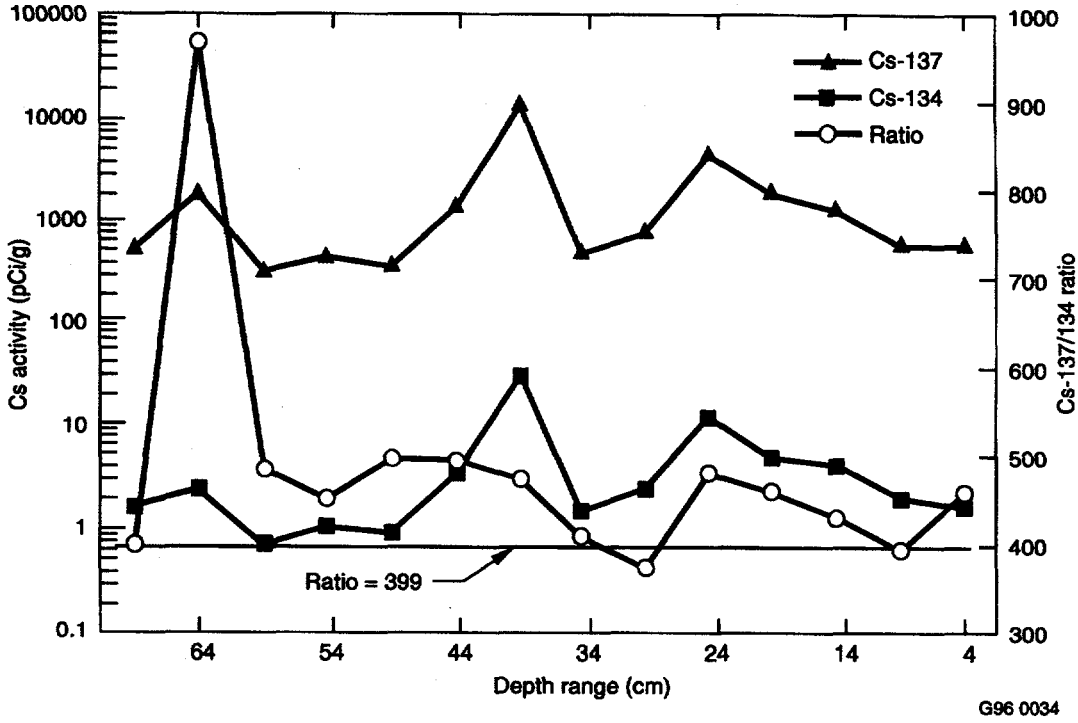

Figure 29. Cesium analyses of the center sand core from ORNL lysimeter 5.

There are three peaks seen in the cesium content (Figure 29): 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 19 and Figure 30. 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 19) 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 19 and Figure 30). Peaks occur in the distribution at the same depths as for cesium in both the sand and roots. This suggests that the same mechanism may be involved for transporting strontium upward as for cesium. Strontium and cesium behave very differently chemically, but if the process of migration is more physical than chemical, then the ratio of Cs-137 to Sr-90 should be similar at all depths. Table 19 includes a tabulation of this ratio versus depth, and Figure 31 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.

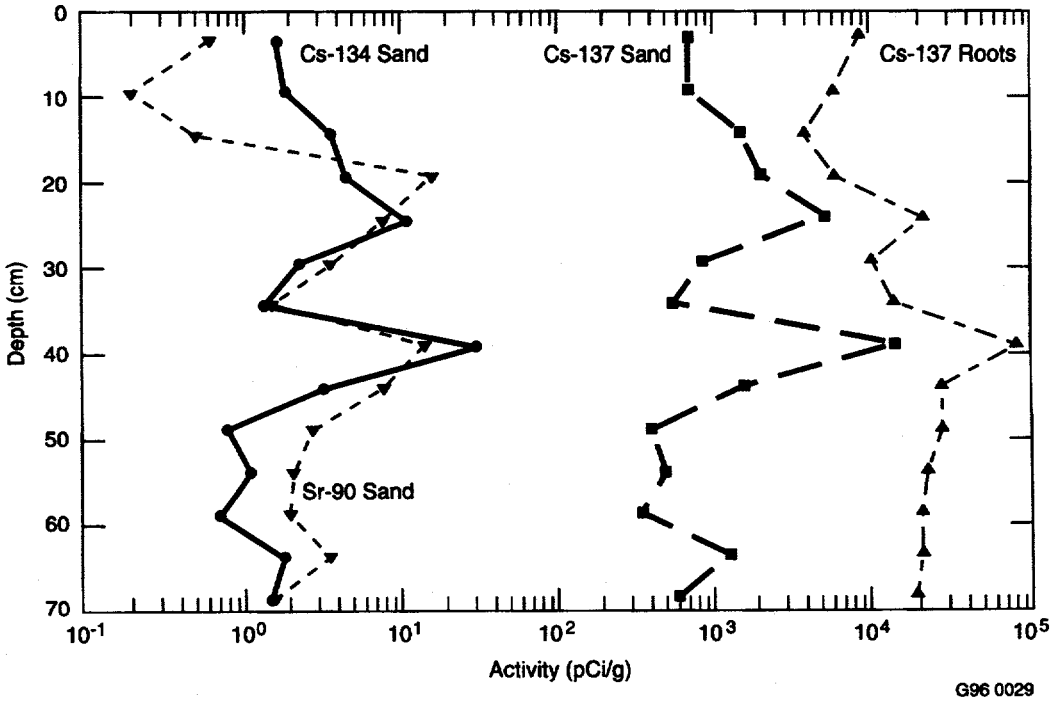

Figure 30. Cesium-137 and Sr-90 associated with plant roots and sand from the center core taken from ORNL lysimeter 5.

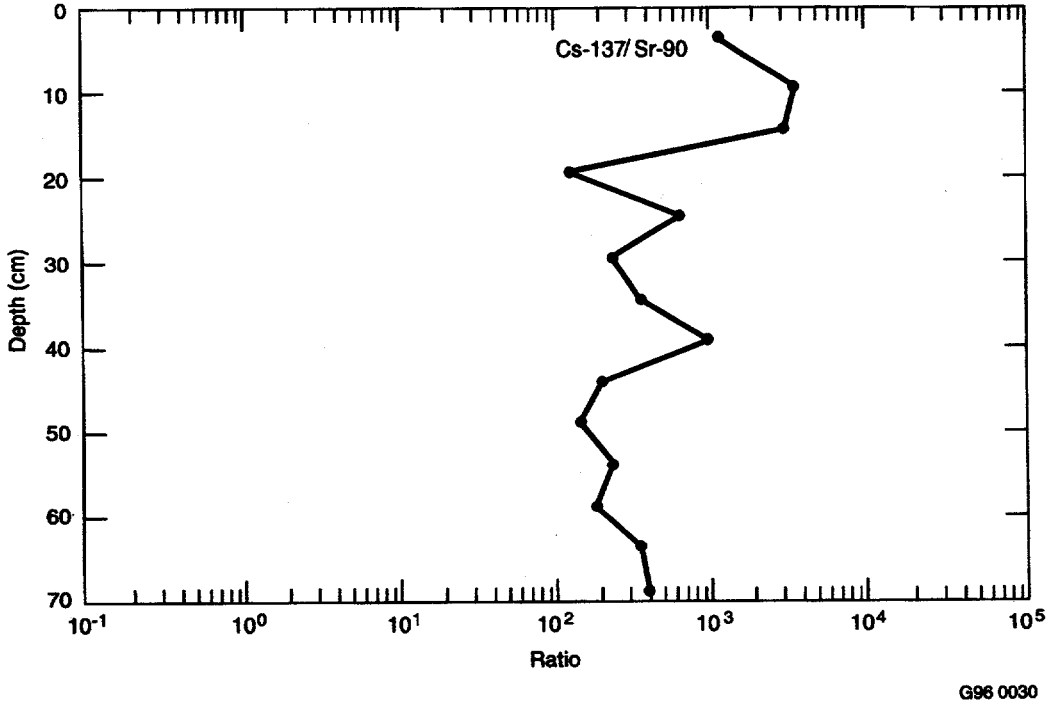

Figure 31. Ratio of Cs-137 to Sr-90 from analysis of center sand core from ORNL lysimeter 5.

A comparison of the Cs-137 concentrations in the center and side cores (Figure 32) 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 in the side core 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 and ignoring the upper three segments because of the large errors associated with the strontium analysis (Table 19). 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 Analysis section of this report. There it is shown that Sr-90 makes up a majority of the radionuclides detected in the leachate.

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 confirm that assumption (Table 15 and Figure 18). 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

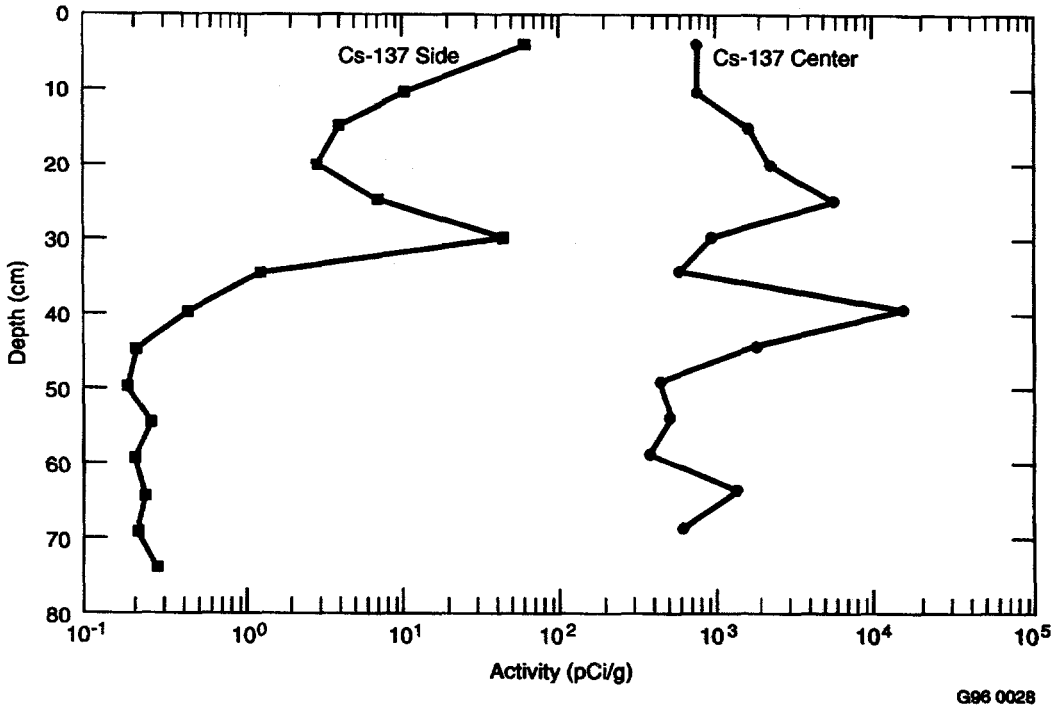

Figure 32. Comparison of Cs-137 analysis from center versus side sand cores from ORNL lysimeter 5.

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 and Source Term 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 forms, mass balance of released constituents, 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 the lysimeter outputs. Such data are invaluable as inputs into process-level and performance assessment codes since they represent a field data set that contains complete information that characterizes environmental, hydrogeological, geochemical, and waste form effects.

The relationship between input parameters for codes and data derived from lysimeter operation is compared in Table 20. The data could be used in such codes at PATHRAE,[24] PRESTO,[25] and others to predict the stability of waste forms for a 300-year period of time.

Source term code studies were performed using the data produced through FY-93 by the ANL-E and ORNL field experiments. A brief summary of the pertinent characteristics of the lysimeters is in order. At each site, four of the lysimeters are filled with soil while the fifth lysimeter (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 Plant adjacent to the Barnwell facility in South Carolina. ANL-E lysimeters are filled with a local soil that represents a typical Midwestern type. It is a morley silt loam with the surface layer removed. Each lysimeter is filled with seven cylindrical waste forms measuring 4.8 cm in diameter and 7.5 cm in height. They are stacked one on top of the other in the lysimeters forming a height of 53.2 cm and a volume of 1 L. The waste forms were solidified in either vinyl ester-styrene or portland type I-II cement. The waste streams included two resin types. PF-7 waste was

Table 20. 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 water 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
C_w	=	Radionuclide concentration	Radionuclide concentration in effluent
M_i	=	Molality	Effluent concentrations
MIN	=	Minerals dissolved or precipitated	From mineralogical characterization of soil at end of experiment

a mixture of synthetic organic ion-exchange resins (phenolic cation, strong acid cation, and strong base anion). PF-24 waste resin was a mixture of synthetic ion-exchange resins (strong acid cation and strong base anion resins) with inorganic zeolite. Each lysimeter is equipped with five moisture collecting cups and three soil moisture/temperature probes, which are located at various elevations in the lysimeter (Figure 7) along with a leachate container located at the bottom of the lysimeter (Reference 18). Below the fill material, a layer of filter fabric was placed between the soil or sand and the gravel bed. A gravel bed is located below the filter fabric. The height of the gravel bed was set to 10 cm in these modeling studies. The data used in this study were collected from moisture cup 3, located approximately 23 cm from the bottom of the waste forms, and from the lysimeter leachate collector, located 61 cm below the bottom of the waste forms. The radionuclides found to date in the leachate waters have been primarily Cs-137 and Sr-90.

The Disposal Unit Source Term (DUST) code developed by Brookhaven National Laboratory (BNL) was used by BNL for the third year to model the release of Cs-137 and Sr-90 from the lysimeter waste forms. DUST is a one-dimensional code that can model release by a finite difference method or by a mixing cell cascade approach, and has the ability to simultaneously 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 26 and 27.

The sand-filled lysimeters 5 at ORNL and ANL-E were chosen for study of the release of Cs-137 and Sr-90 from portland type I-II cement because releases from other lysimeters were substantially lower and the data were not sufficient to model. At ANL-E, lysimeter 5 contained resin waste type I solidified in cement; at ORNL, lysimeter 5 contained resin waste type II, which was also solidified in portland type I-II cement (see Table 5). Diffusional release is believed to be the controlling mechanism for a cement-solidified waste. The waste form diffusion coefficients for portland type I-II cement were presented in Reference 20. 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 12). The soil bulk density values were 1.55 g/cm^3 at ANL-E and 1.60 g/cm^3 at ORNL (Reference 18).

Moisture content values were calculated using the effective soil porosity and the fraction of saturation values found in Reference 14. In lysimeter 5 at both sites, the moisture content was calculated as 21%. The dispersivity and retardation coefficients have not been measured for Sr-90 or Cs-137; therefore, they were estimated based on data in References 28 and 29 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 0.181% 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 0.004 of the Cs-137 inventory in lysimeter 5, while nothing has been collected in ANL-E lysimeter 5 (Table 21).

The cumulative activity collected from the lysimeters is less than 0.2% in comparison to the total inventory for Sr-90 and less than 0.005% for Cs-137 (Table 21). Therefore, either the waste form release rates are much lower than anticipated or transport processes are controlling release through the soil column. At Cs-137 level, it is possible that random fluctuations (noise) are being seen, and release patterns may not develop for several more years.

Concentrations and predicted releases were matched to moisture cup 3 and the lysimeter leachate collector. The concentrations and releases were taken at 23 and 51 cm below the waste forms. In this report, the cumulative leachate activity collected 51 cm beneath the waste form is used as the performance measure. Initial amounts of Cs-137 and Sr-90 varied at ORNL and ANL-E because the control lysimeters contained different resin types. In ORNL lysimeter 5, the type I waste form had a total initial inventory of 0.0033 Ci of Sr-90 and 1.432 Ci of Cs-137 (Table 21 and Reference 18). The PF-24 waste form at ANL-E had a total initial inventory of 0.0185 Ci of Sr-90 (Table 21 and Reference 18). 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 were modeled where 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 33 and 34 are obtained using a bottom BC of zero dispersive flux. A concentration trace continued to be taken at the location of the filter fabric, which is 51 cm below the waste forms.

As shown in Figure 33, the actual data for Sr-90 from ORNL lysimeter 5 for 10 years are compared with the DUST code predicted releases using zero dispersive flux BC, K_d = 24 mL/g, and dispersivity = 8.5 cm. Also shown are predicted releases 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

Table 21. Total and collected Ci amounts of Sr-90 and Cs-137 in lysimeters 5 through July 1995.

	Total amount (Ci)	Amount collected (Ci)	Percent collected
ORNL-5 Cs-137	1.432	6.22E-6	434.0E-6
ORNL-5 Sr-90	0.0033	5.98E-6	1812.0E-4
ANL-5 Sr-90	0.0185	4.16E-6	225.0E-4

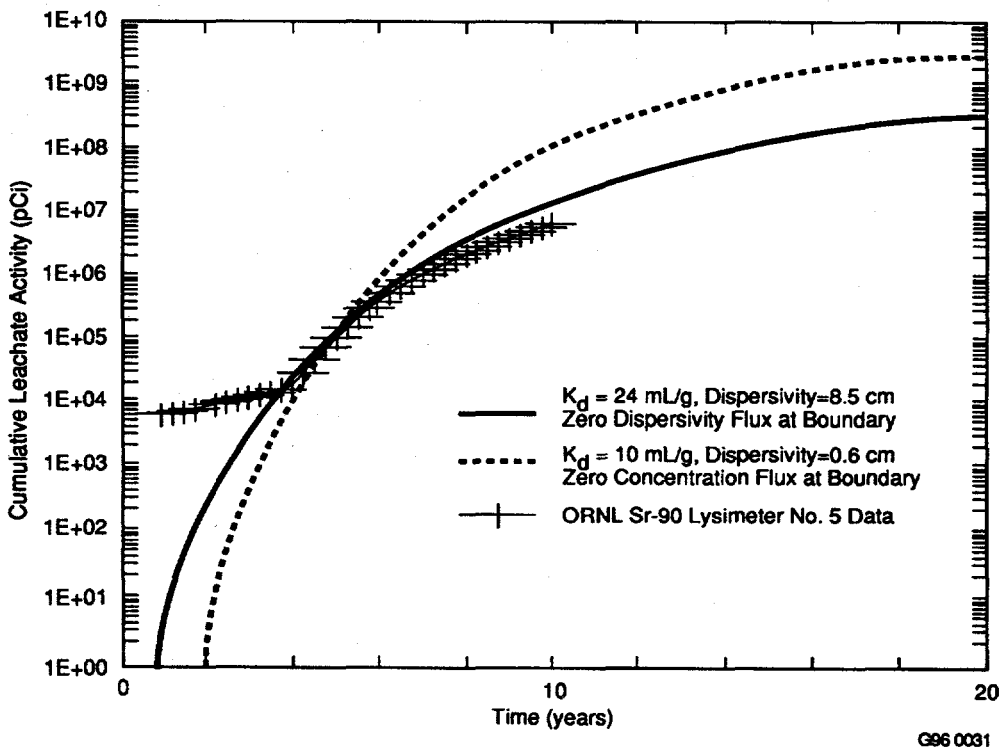

Figure 33. Ten years of data for Sr-90 at ORNL lysimeter 5, compared with two sets of estimated K_d and dispersivity values.

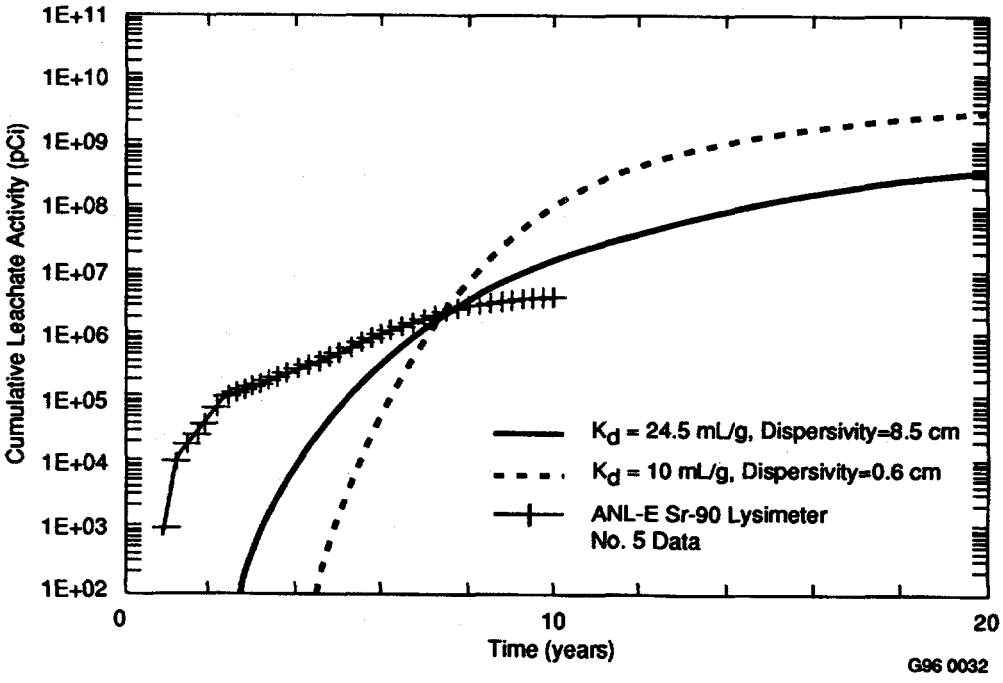

Figure 34. Ten years of data for Sr-90 at ANL-E lysimeter 5, compared with two sets of estimated K_d and dispersivity values for 20 years.

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 34 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 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 the 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 a 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 during the summer sampling periods in 1994 and 1995 was analyzed for the major ionic species listed in Table 22. The justification for the choice of ions is also provided in the table. At ANL-E, cups 1, 3, and 5 were sampled on lysimeters 1, 3, 4, and 5; and cups 2, 3, and 4 on lysimeter 2 in 1994. In 1995, 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 water samples were sampled on all lysimeters both years at ORNL. Data from precipitation samples at ORNL in 1989 and ANL-E in 1991 showed that ionic concentrations in the soil water were not introduced by the precipitation (References 9 and 12). It appears that the waste forms could be an influencing factor either as the source of ions or possibly by causing replacement of ions from the surrounding soil (Tables F-1 through F-4 of Appendix F and Figures 35, 36, 37, and 38). It appears that the cement and VES waste forms performed similarly at both sites. The ORNL 1994 and 1995 soil lysimeter cation and anion data (Tables F-3 and F-4; Figures 37 and 38) closely resemble those of 1989, 1991, 1992, 1993, and 1994, and actually showed little of the cup-to-cup variability found in 1988. ANL-E 1994 and 1995 data are similar (Tables F-1 and F-2; Figures 35 and 36), except for lower calcium, to previous years' data and to ORNL 1994 and 1995 data when compared in Figures 35, 36, 37, and 38. The inert, sand-filled lysimeter results are almost identical except for higher chlorine and NO_3 concentrations at ORNL. While these early data are interesting, no correlation can be made with nuclide movement as yet.

Table 22. 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.

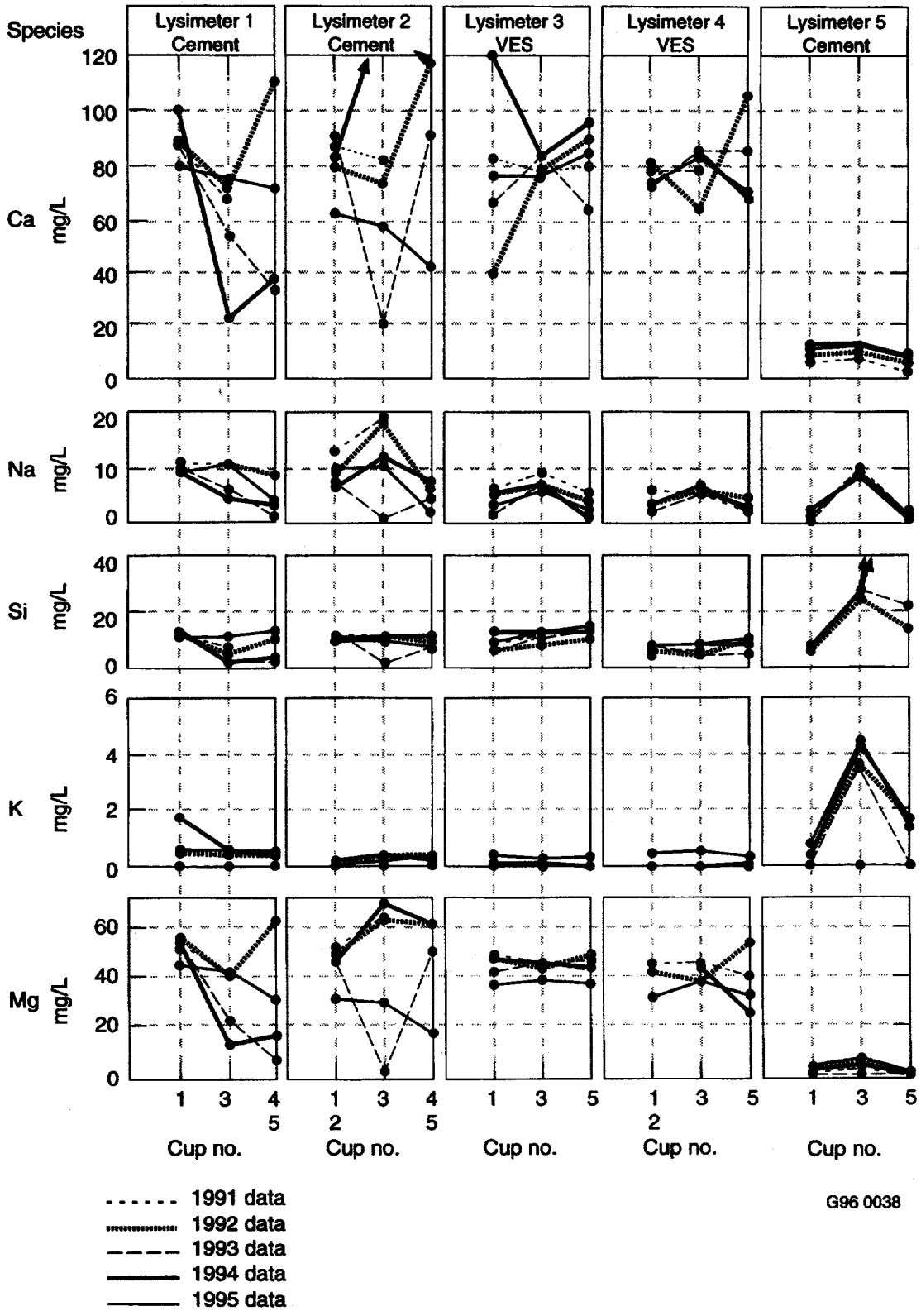

Figure 35. Results of chemical speciation at ANL-E—cations.

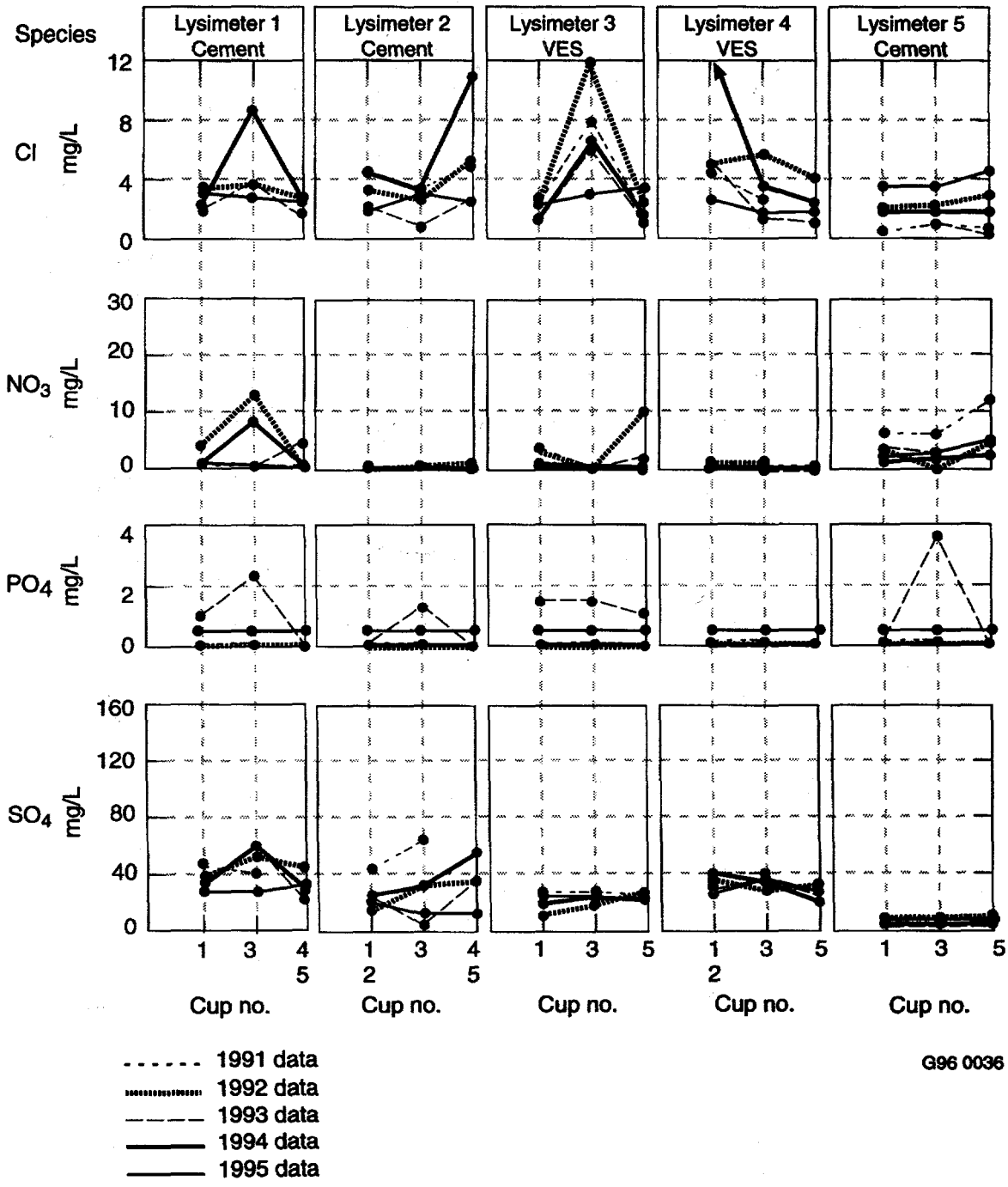

Figure 36. Results of chemical speciation at ANL-E—anions.

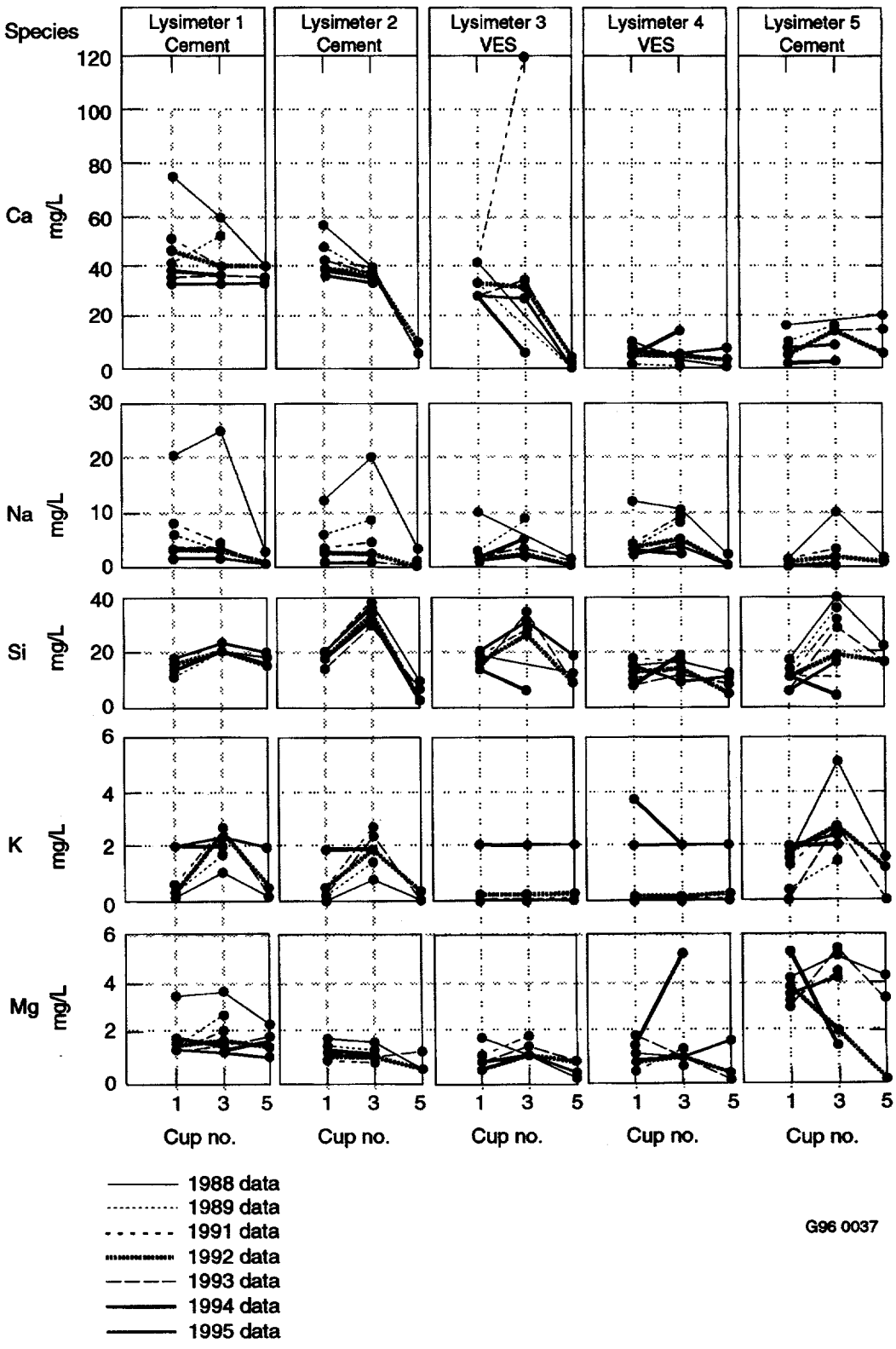

Figure 37. Results of chemical speciation at ORNL—cations.

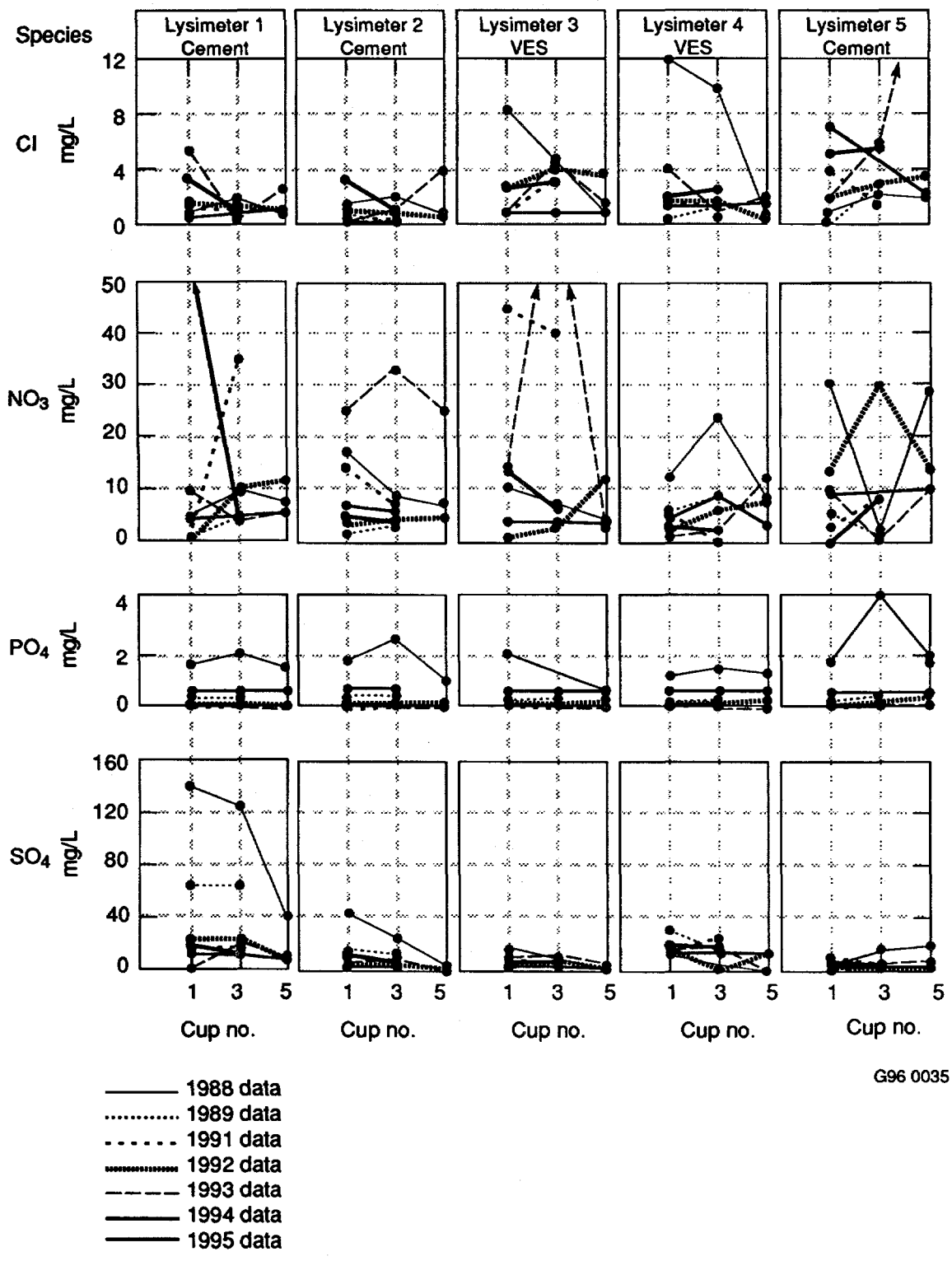

Figure 38. Results of chemical speciation at ORNL—anions.

CONCLUSIONS

The lysimeter experiment has been successful during the 10 years of operation. Analyses of data collected during the past 120 months continue to show a pattern in nuclide availability and movement such that the cumulative results are beginning to provide an insight on waste form performance.

There continues to be a greater recovery of Sr-90 in terms of quantity and percent of inventory than other nuclides. Next in abundance is Cs-137, followed by Sb-125 (this nuclide has not been detected for the past 72 months) and Co-60. Compared to Sr-90, the occurrence of Cs-137, Sb-125, and Co-60 appears insignificant except for the leachate from the collector of ORNL-5.

On a cumulative basis, a larger amount of Sr-90 is being removed in leachate water from the ORNL soil lysimeters versus those at ANL-E. This is thought to be a result of the difference in soils as well as in environmental conditions between the two sites. During the past 96 months, Sr-90 continues to be found in equal concentrations in leachate water from the sand-filled control lysimeters at both sites, with a slightly more rapid accumulation at ORNL, which now has had eight times more of the available source of Sr-90 released than the control lysimeter at ANL-E. Such data continue to reinforce the assumption that the limiting step in receiving Sr-90 in leachate water is not release of the nuclide from the waste forms (since Sr-90 is found in larger quantities in leachate water at ORNL rather than in cups), but rather, the movement is limited by environmental characteristics (including soil and quantity of soil water). This conclusion is supported by data from lysimeter work at Savannah River Laboratory (SRL) and Pacific Northwest Laboratory (PNL).[30,31,32] SRL has found that Sr-90 will move from buried waste forms, migrate through the soil column, and appear in collected leachate water.[30] It is not surprising, then, that Sr-90 moves through soil in the ORNL lysimeters, since that soil originated at SRL.[18] On the other hand, lysimeter work with waste forms at PNL has shown that Sr-90 does not move in those soils.[31]

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

In the past 3 years, Cs-137 has been found with consistency in leachate collector water from the sand-filled lysimeters only at ORNL. In 1992 and 1993, Cs-137 was found in leachate water from the sand-filled control lysimeters at both sites, but was not detected in the leachate collector water of any lysimeter at ANL-E in the last 2 years.

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. These data still differ from those obtained at SRL. Those data show that cement minimizes the release of Sr-90.[30] This interesting difference should be studied further. Both data reported herein and data reported by SRL[30] and PNL[31] agree that Cs-137 is more readily released from cement than from VES, while PNL[32] has observed Cs-137 release in trace amounts only from masonry cement waste forms, and none from portland and VES waste forms.

A comparison of cumulative fractional releases from field-testing of EPICOR-II waste forms in lysimeters to the releases from bench-leach-testing of similar waste forms show that lysimeter releases are at least two 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

CFR 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, shows 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.

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Appendix A

Weather Data

Appendix A
Weather Data

List of Figures

Site	Parameter	Year 1993–94	Year 1994–95
ANL-E	Precipitation	A-1	A-2
	Air temperature	A-3	A-4
	Wind speed	—	—
	Relative humidity	—	—
ORNL	Precipitation	A-5	A-6
	Air temperature	A-7	A-8
	Wind speed	—	—
	Relative humidity	—	—

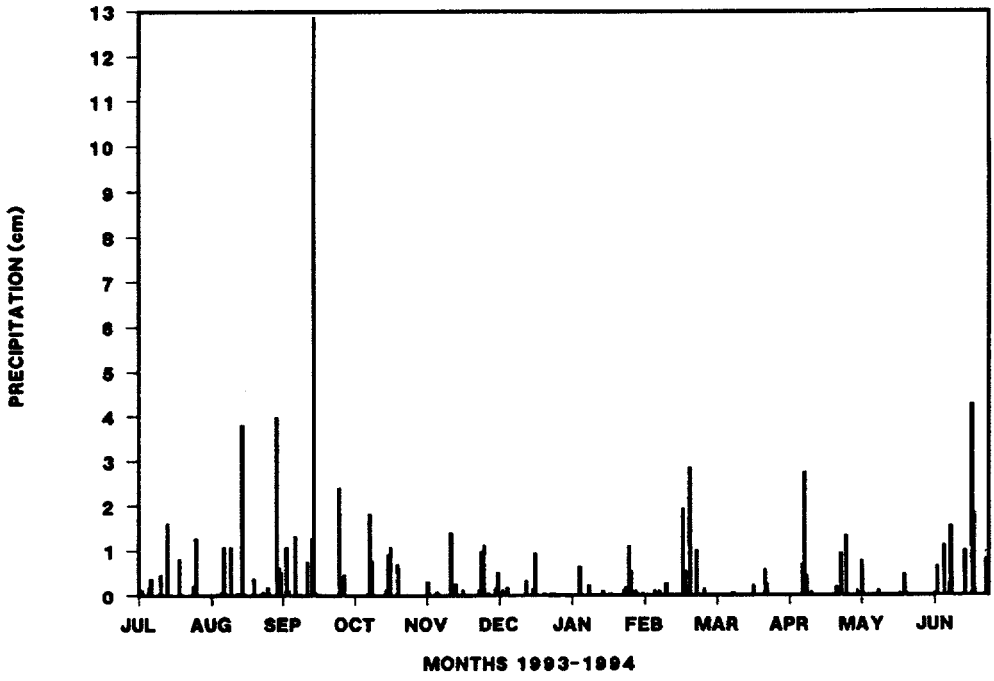

Figure A-1. ANL-E weather data for 1993-94—precipitation.

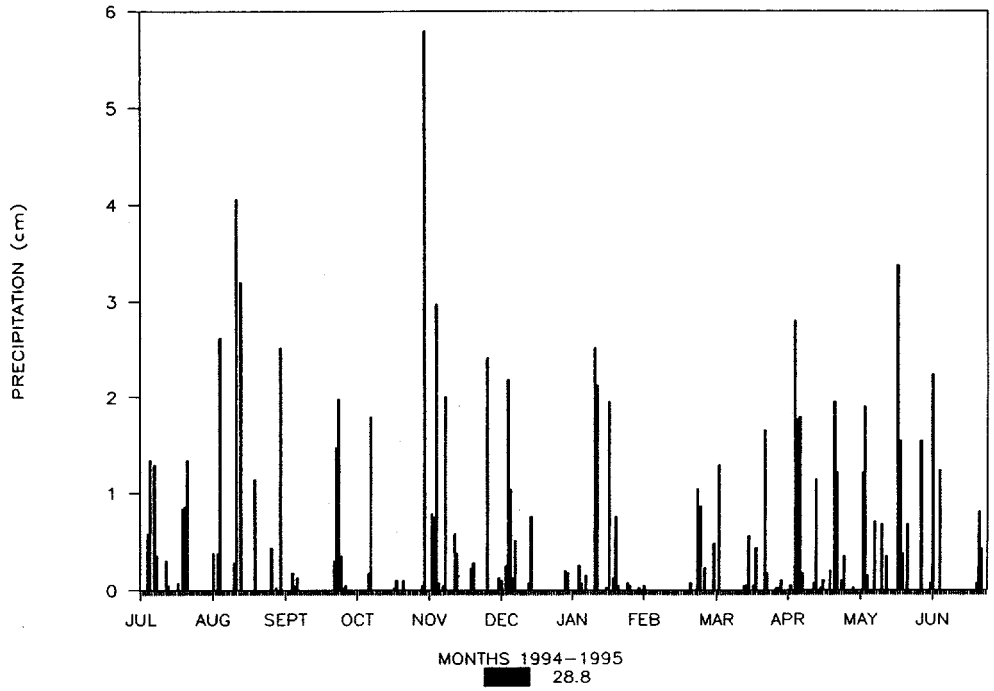

Figure A-2. ANL-E weather data for 1994-95—precipitation.

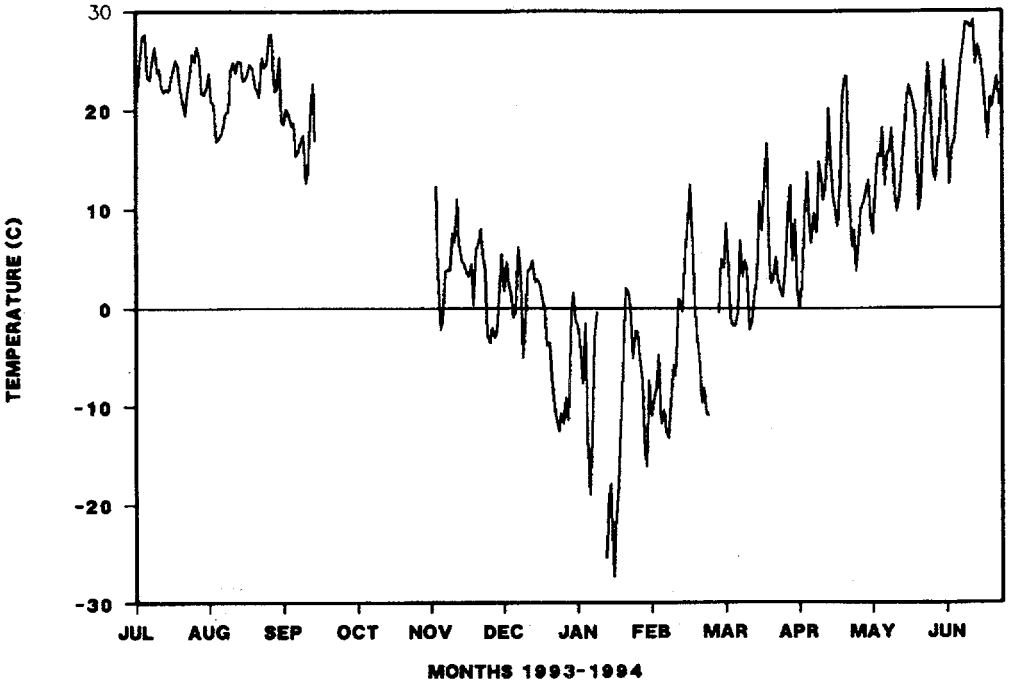

Figure A-3. ANL-E weather data for 1993-94—air temperature.

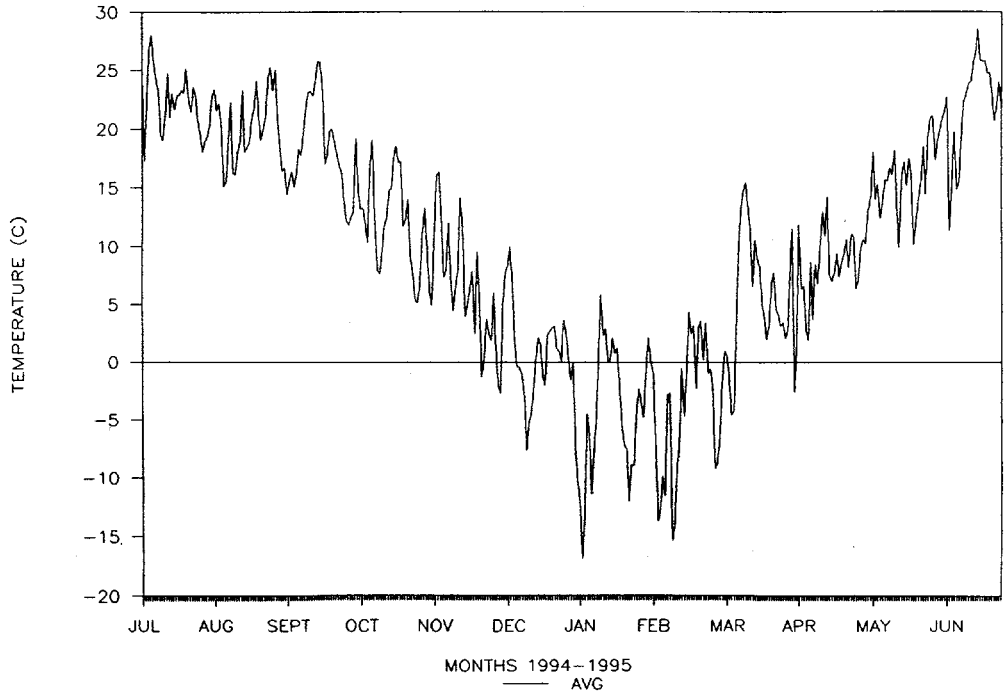

Figure A-4. ANL-E weather data for 1994-95—air temperature.

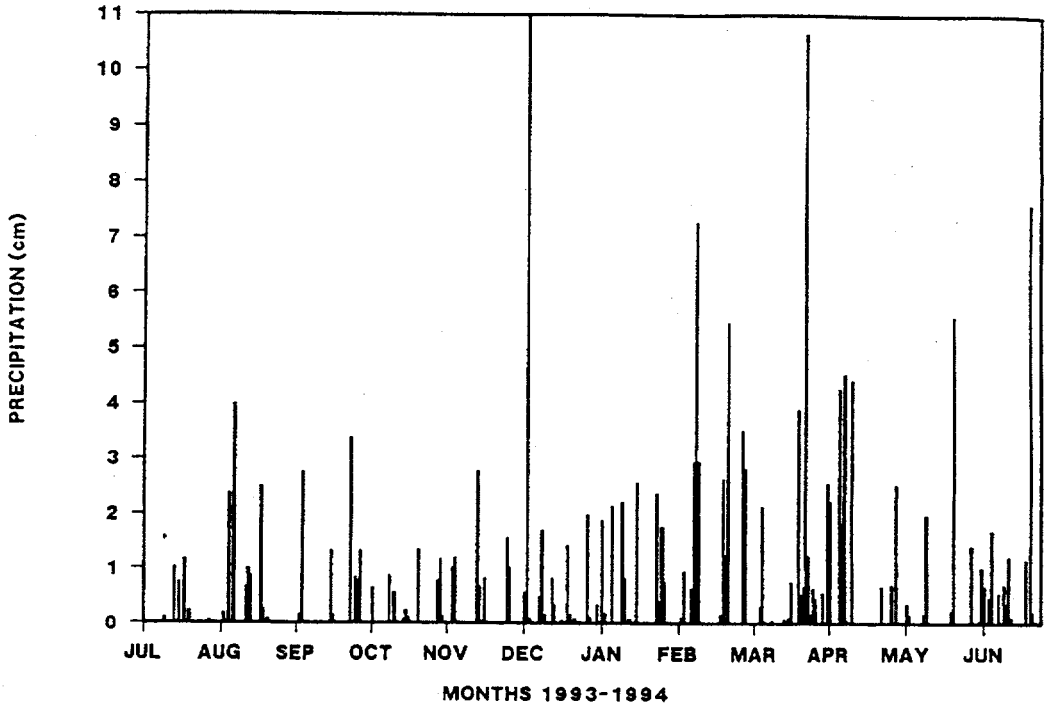

Figure A-5. ORNL weather data for 1993-94—precipitation.

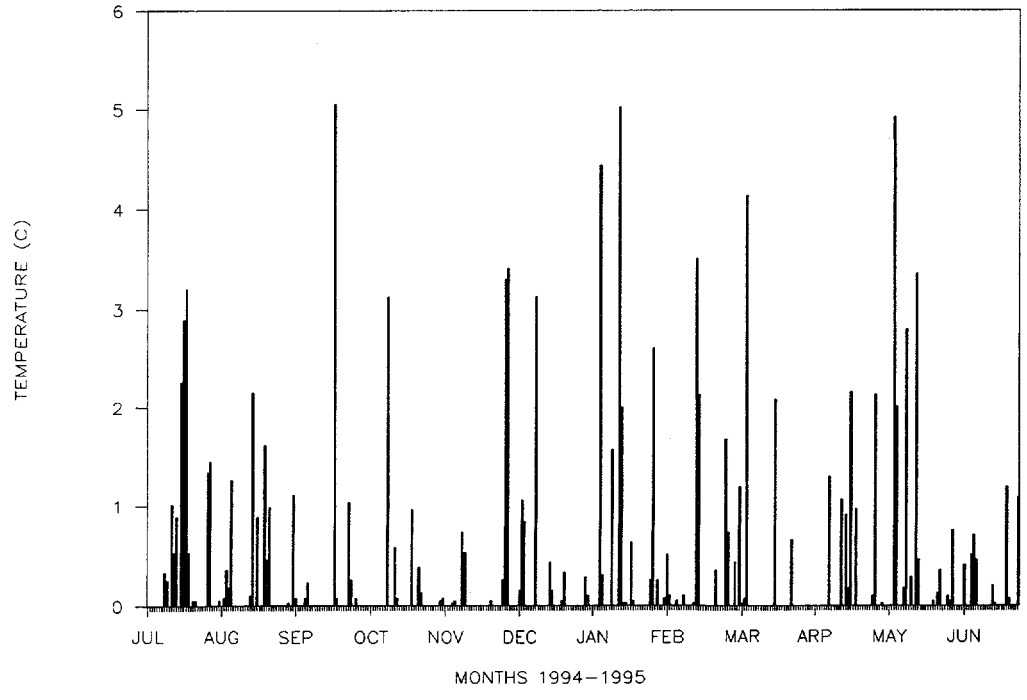

Figure A-6. ORNL weather data for 1994-95—precipitation.

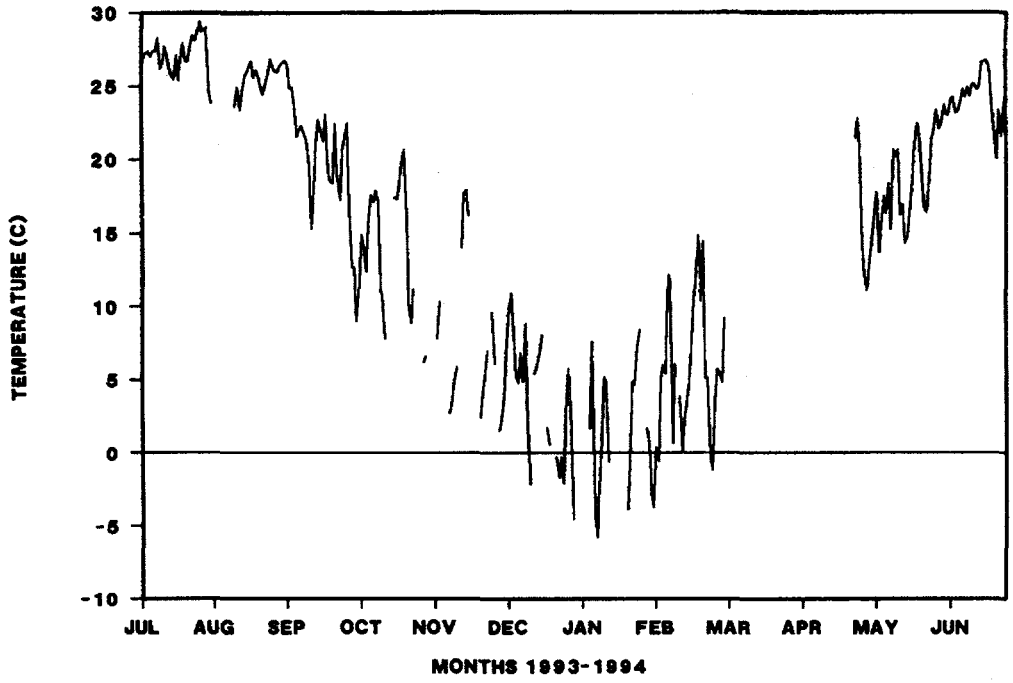

Figure A-7. ORNL weather data for 1993-94—air temperature.

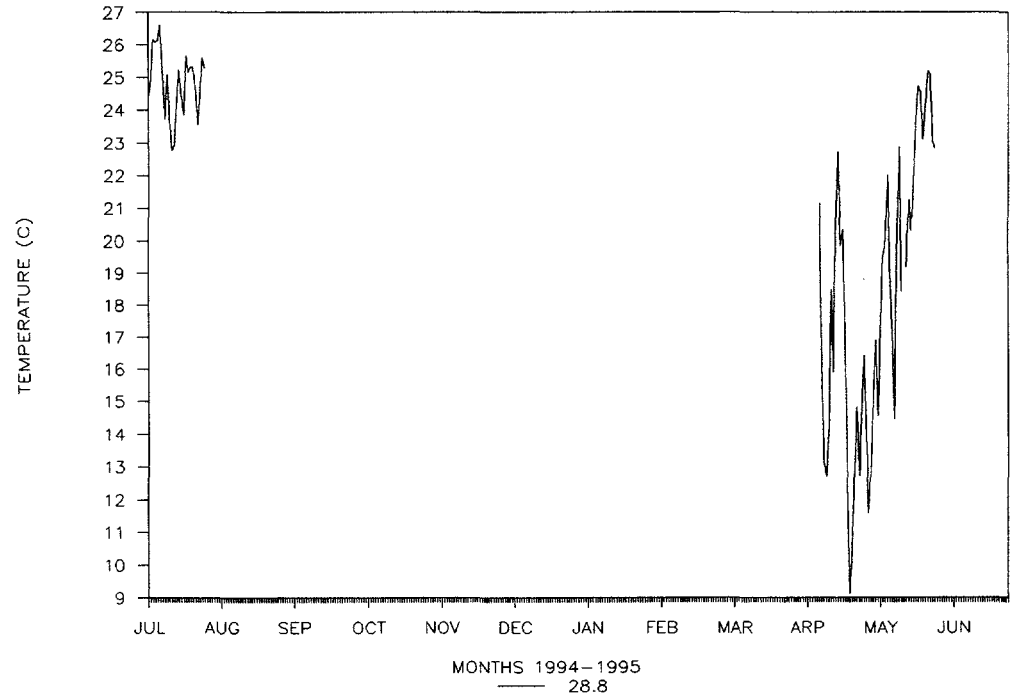

Figure A-8. ORNL weather data for 1994-95—air temperature.

Appendix B

Soil Temperature Data—Resistance Probes

Appendix B
Soil Temperature Data—Resistance Probes

List of Figures

Site	Lysimeter number	Year	
		1993–94	1994–95
ANL-E	1	B-1	B-2
	2	B-3	B-4
	3	B-5	B-6
	4	—	—
	5	B-7	B-8
ORNL	1	B-9	B-10
	2	B-11	B-12
	3	B-13	B-14
	4	B-15	B-16
	5	B-17	B-18

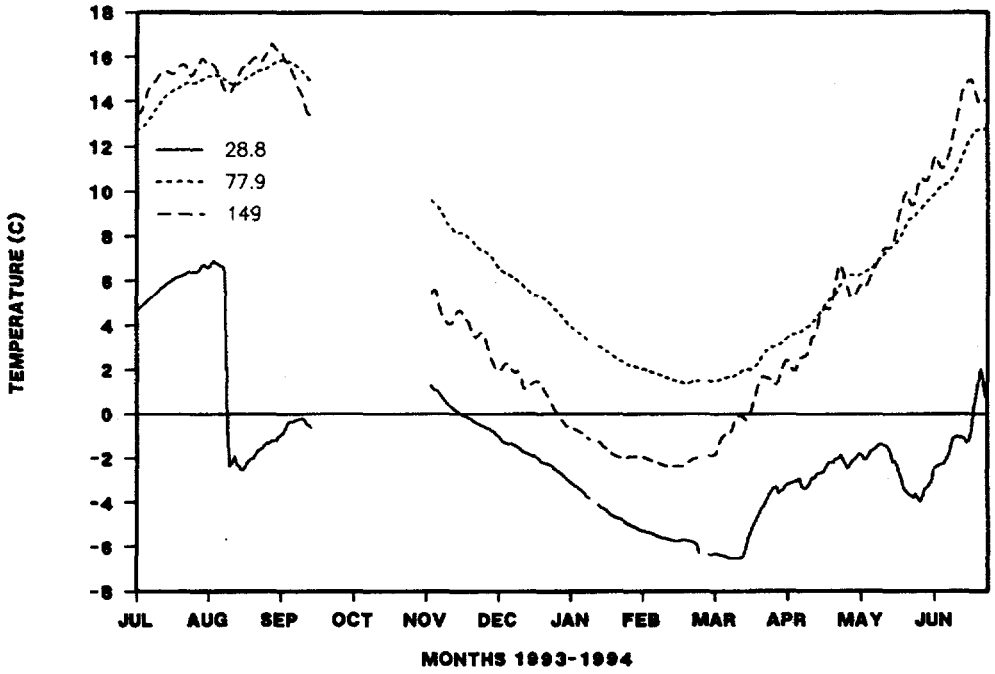

Figure B-1. ANL-E lysimeter 1 soil temperatures for 1993-94.

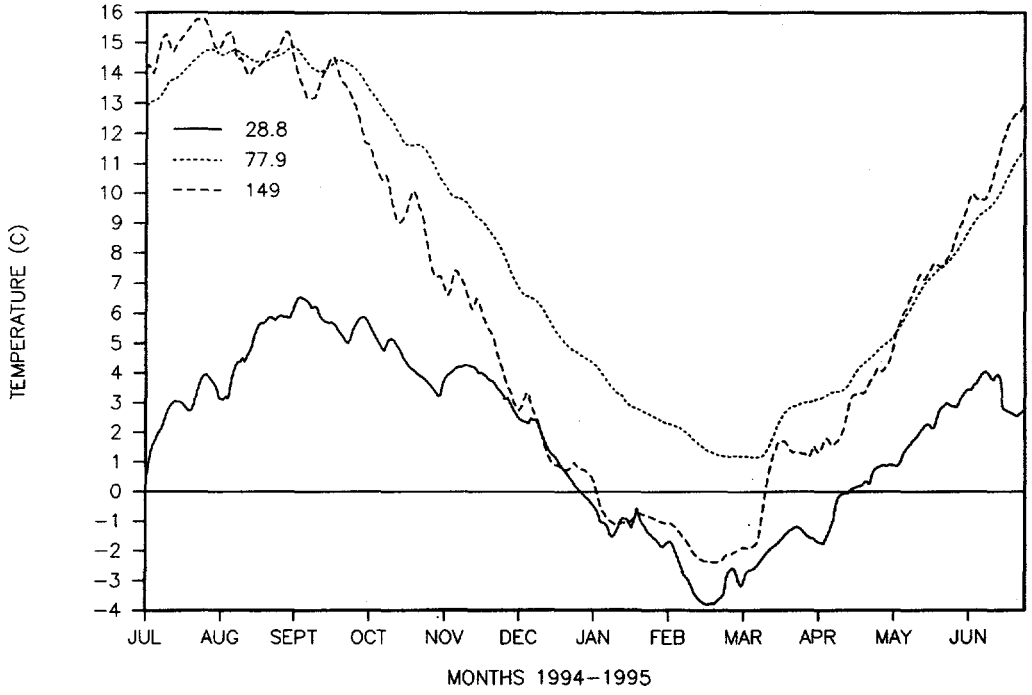

Figure B-2. ANL-E lysimeter 1 soil temperatures for 1994-95.

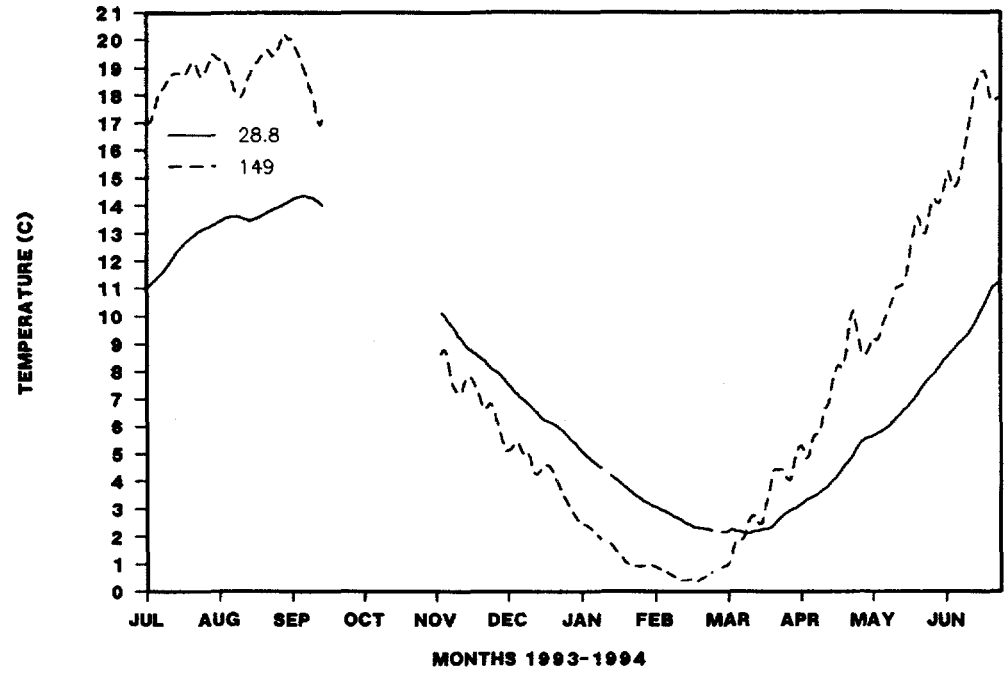

Figure B-3. ANL-E lysimeter 2 soil temperatures for 1993-94.

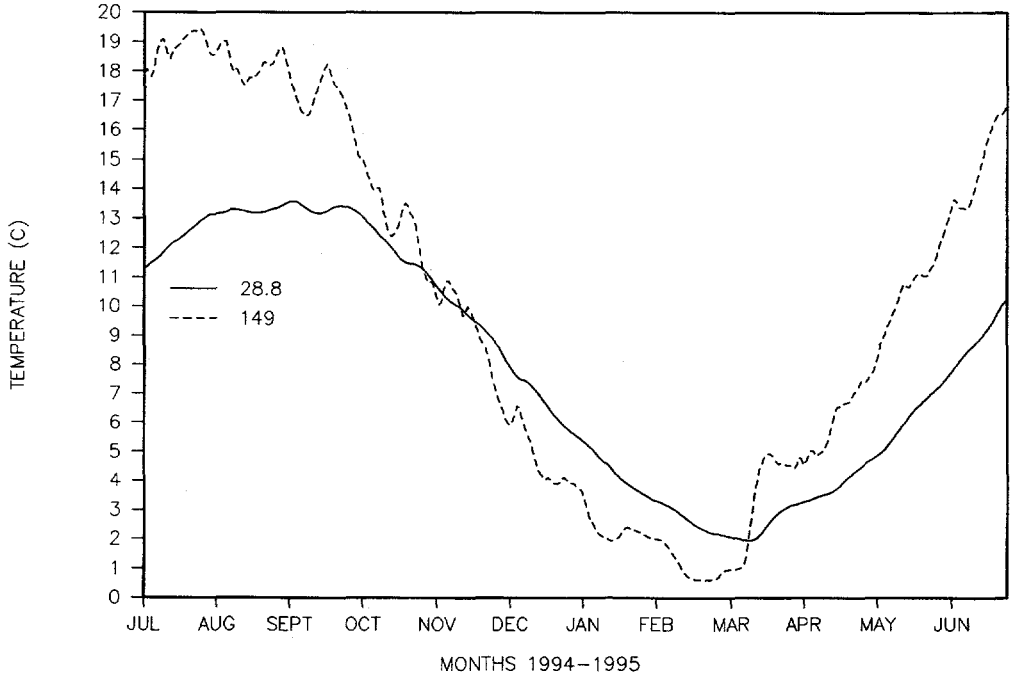

Figure B-4. ANL-E lysimeter 2 soil temperatures for 1994-95.

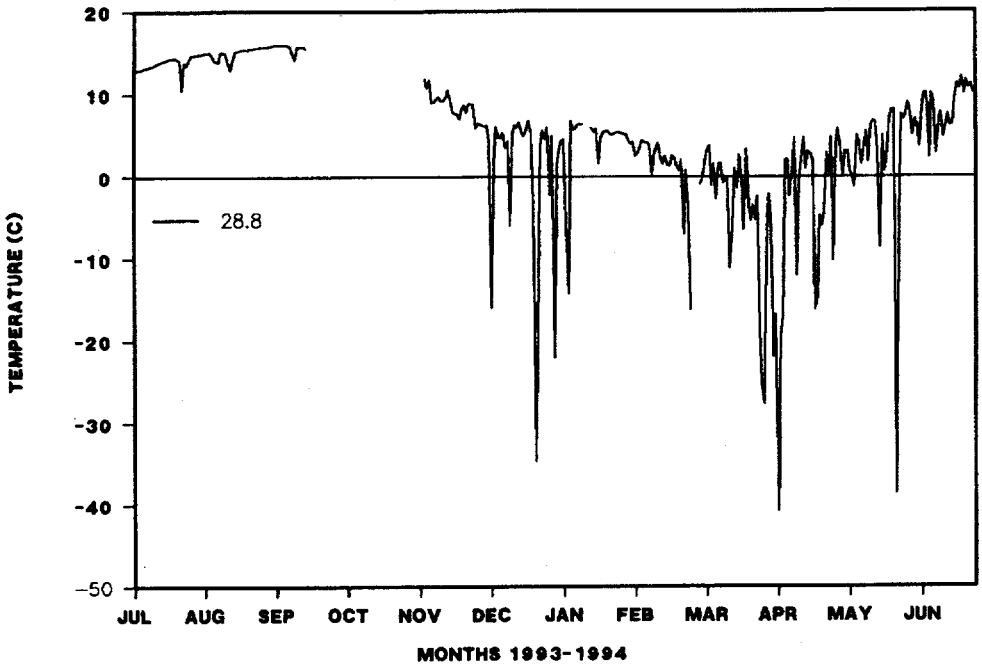

Figure B-5. ANL-E lysimeter 3 soil temperatures for 1993-94.

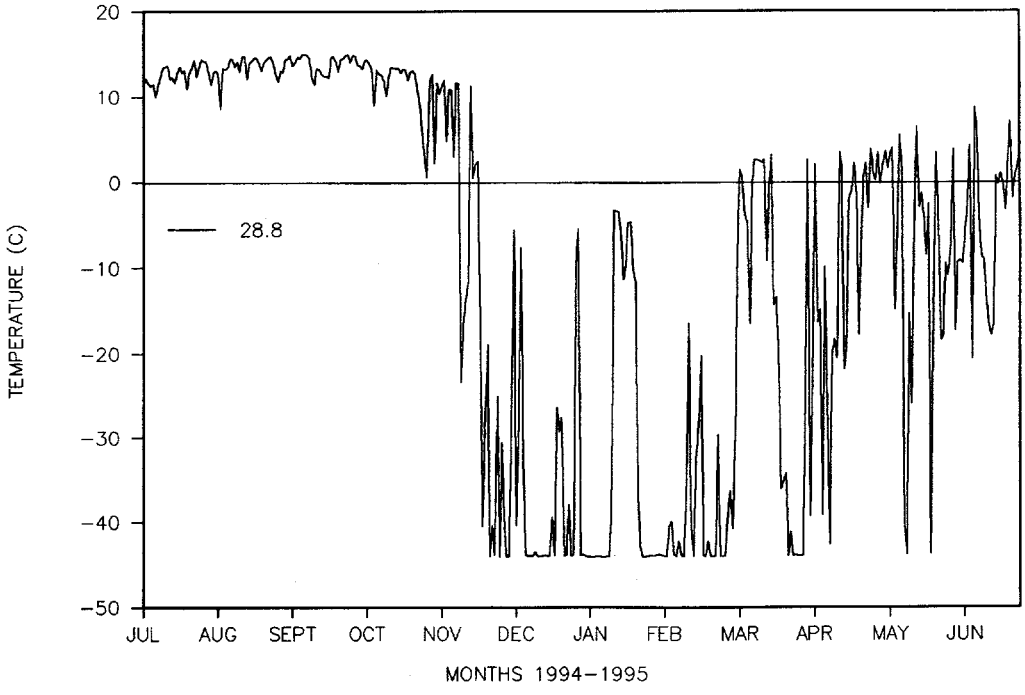

Figure B-6. ANL-E lysimeter 3 soil temperatures for 1994-95.

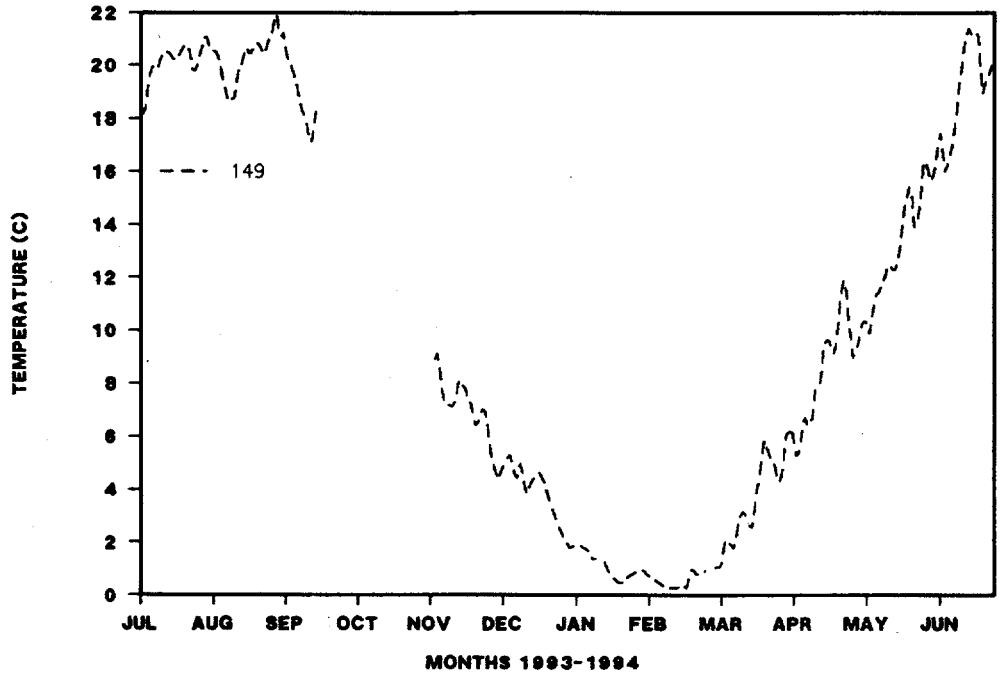

Figure B-7. ANL-E lysimeter 5 soil temperatures for 1993-94.

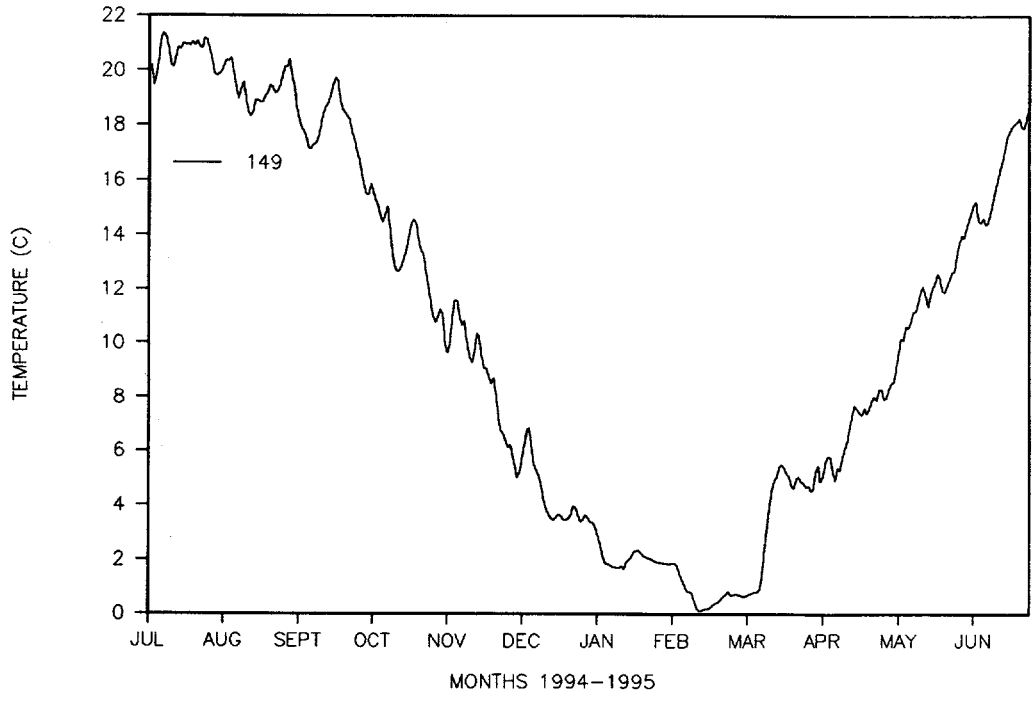

Figure B-8. ANL-E lysimeter 5 soil temperatures for 1994-95.

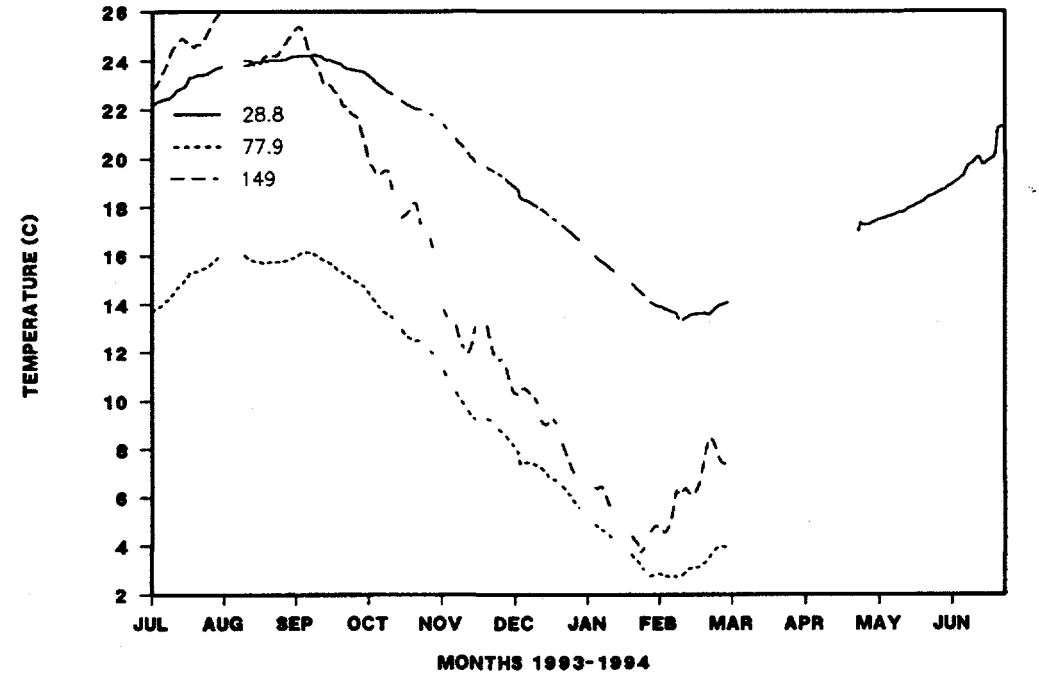

Figure B-9. ORNL lysimeter 1 soil temperatures for 1993-94.

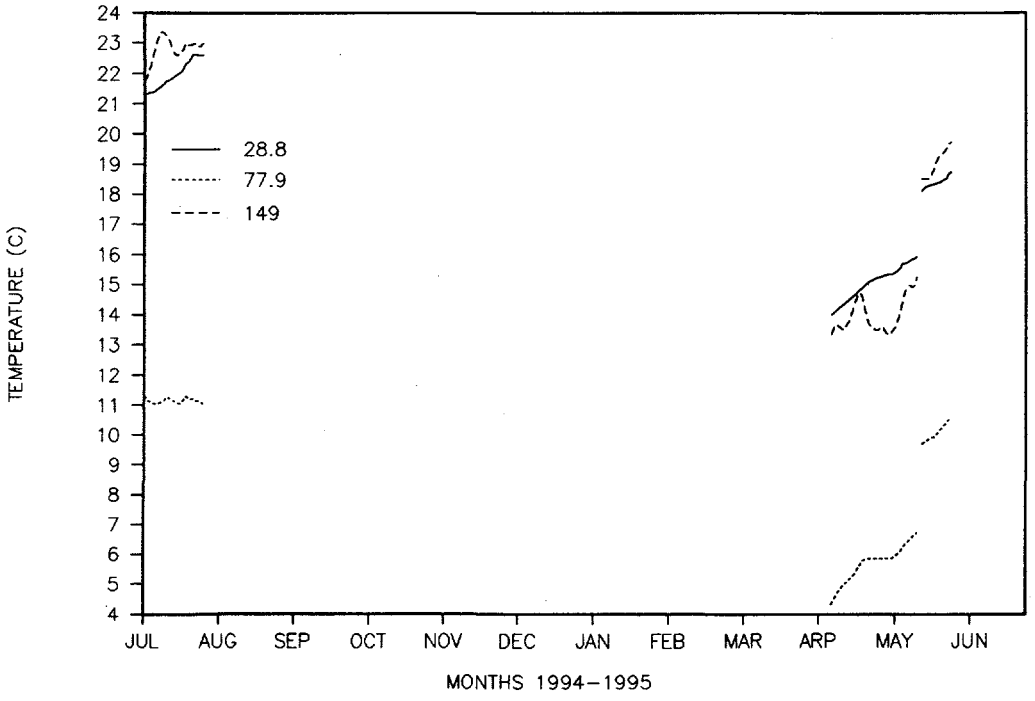

Figure B-10. ORNL lysimeter 1 soil temperatures for 1994-95.

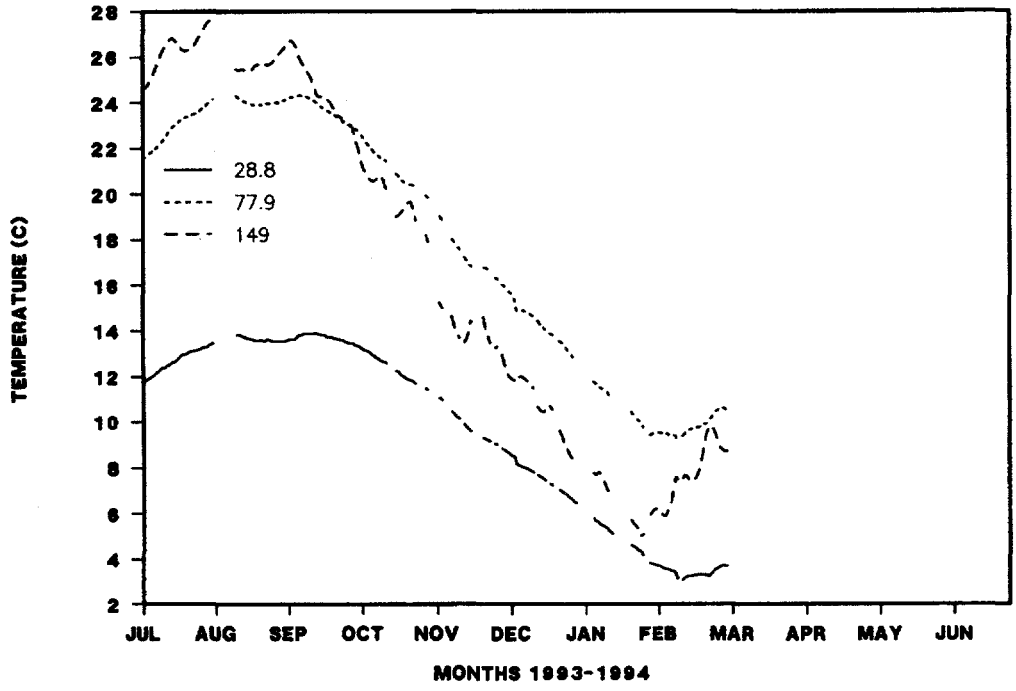

Figure B-11. ORNL lysimeter 2 soil temperatures for 1993-94.

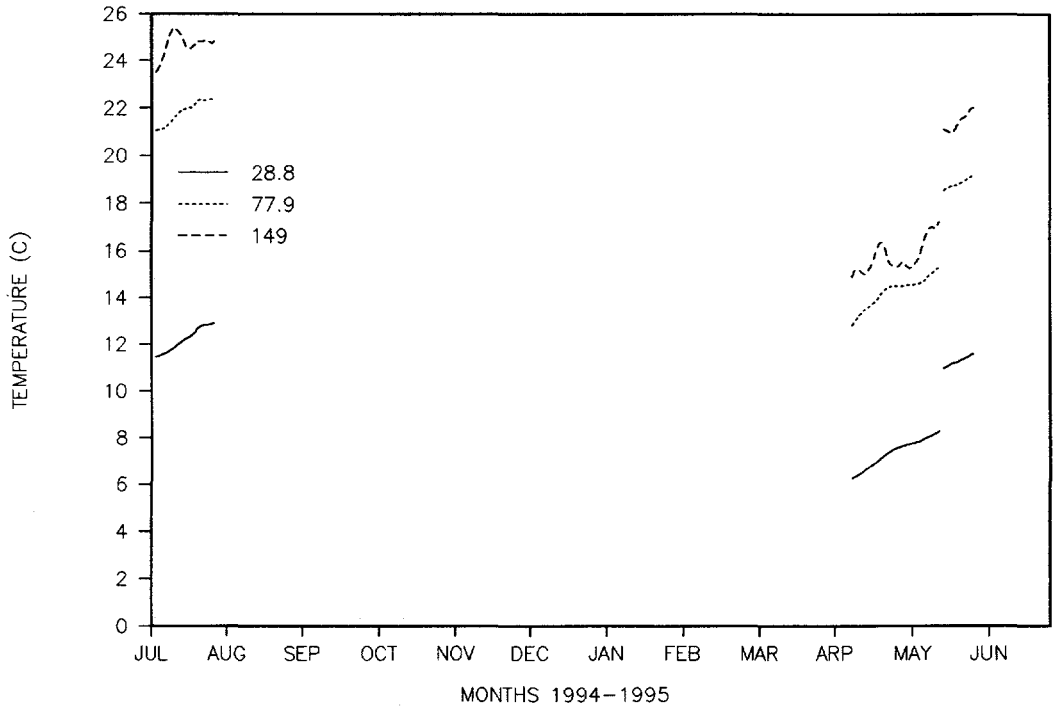

Figure B-12. ORNL lysimeter 2 soil temperatures for 1994-95.

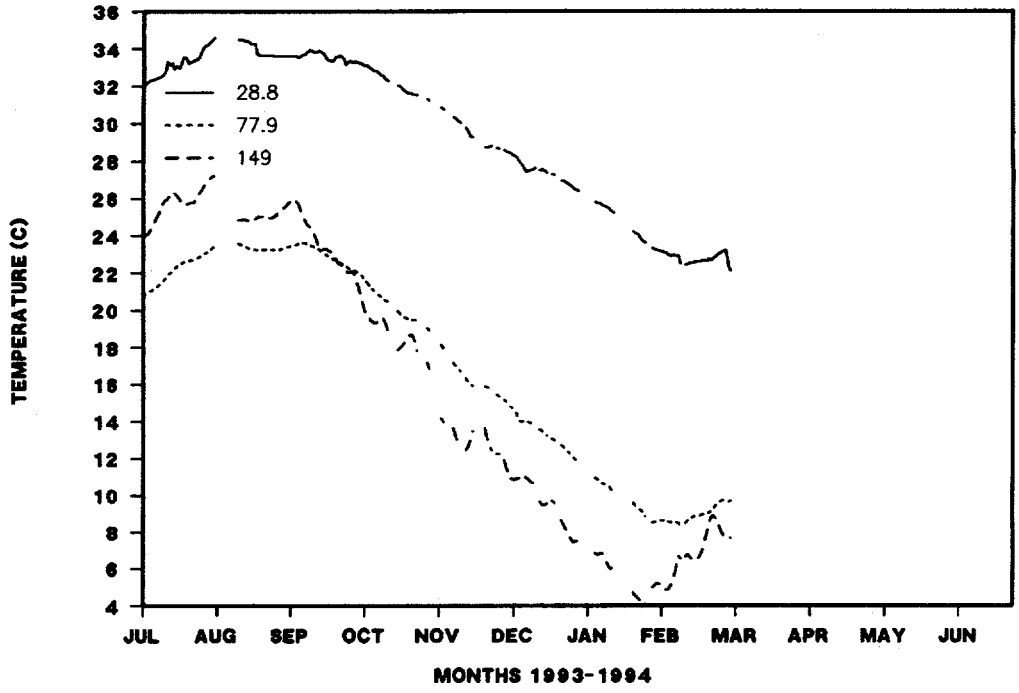

Figure B-13. ORNL lysimeter 3 soil temperatures for 1993-94.

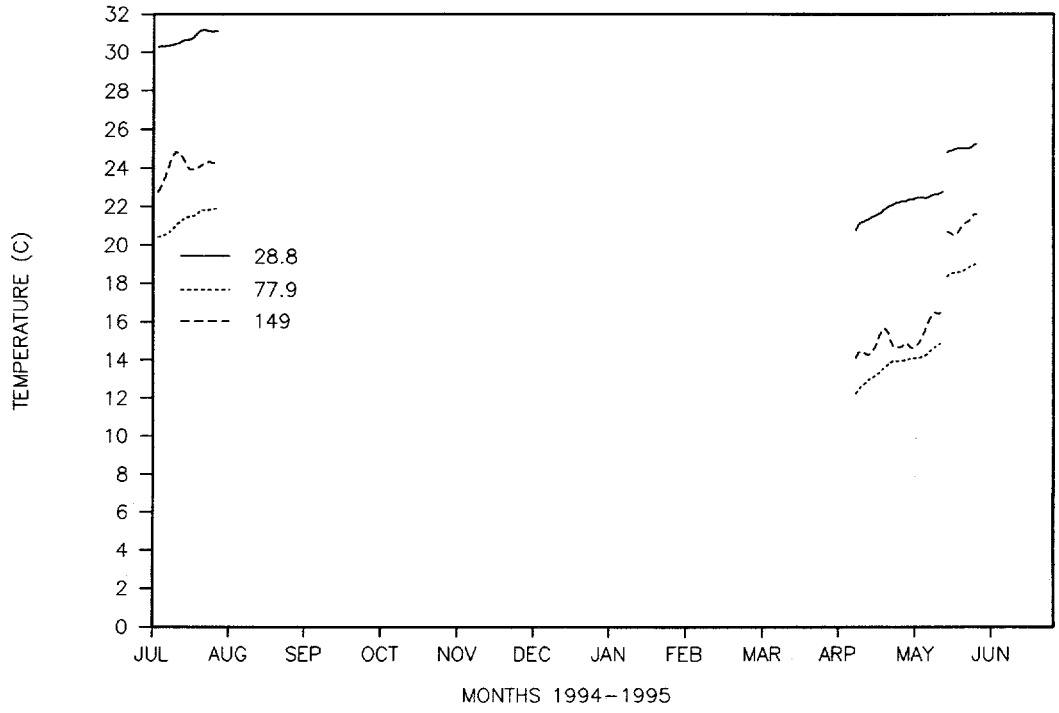

Figure B-14. ORNL lysimeter 3 soil temperatures for 1994-95.

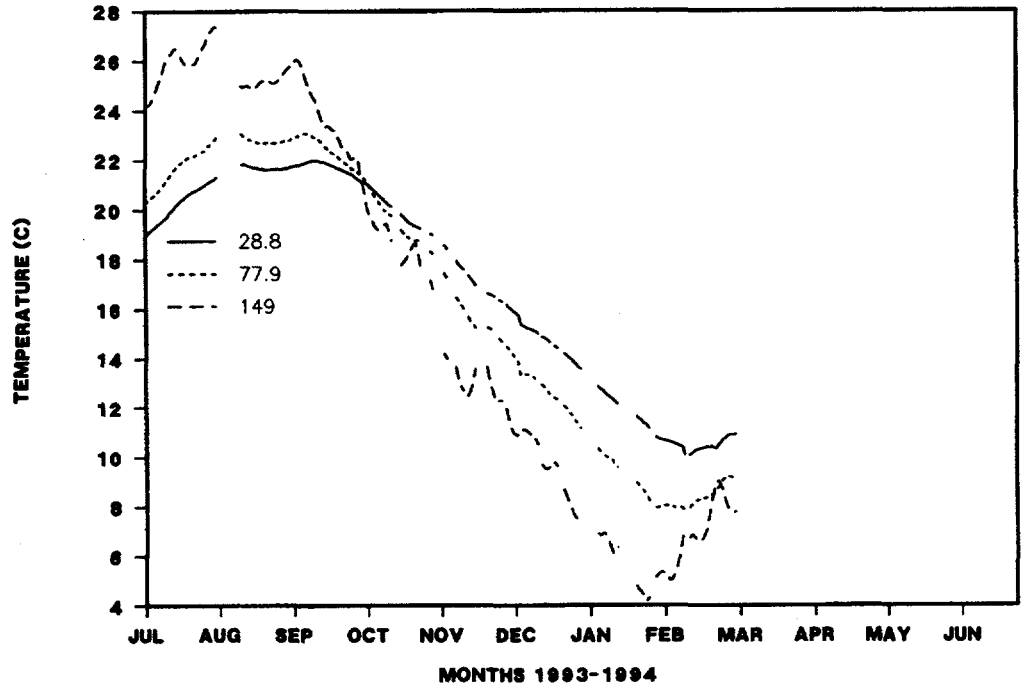

Figure B-15. ORNL lysimeter 4 soil temperatures for 1993-94.

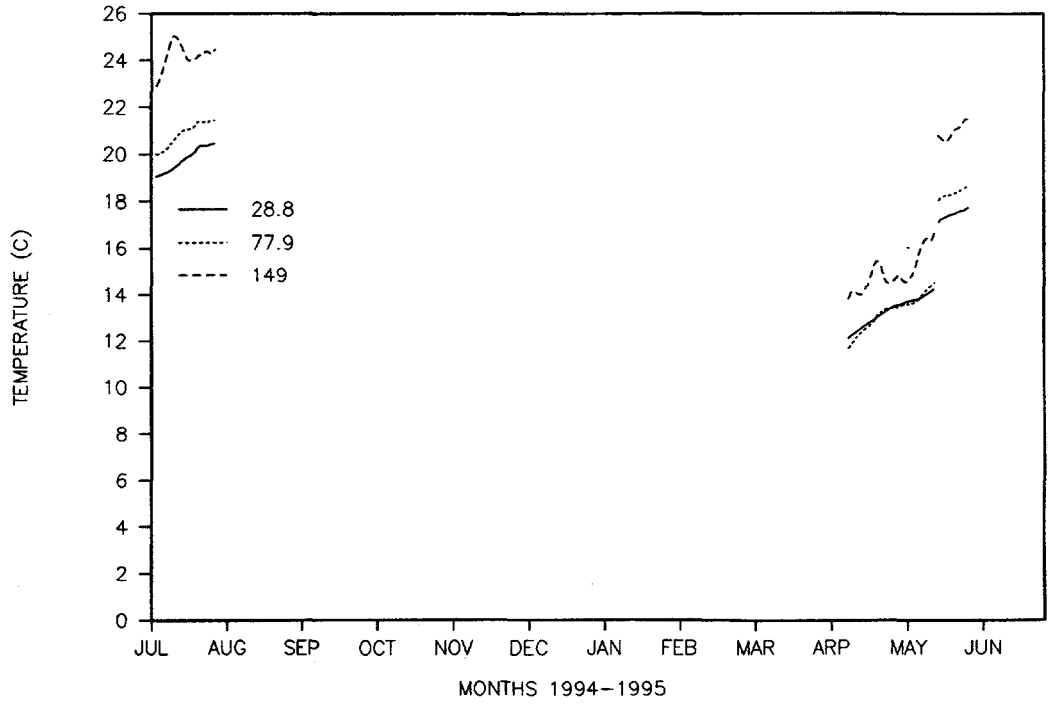

Figure B-16. ORNL lysimeter 4 soil temperatures for 1994-95.

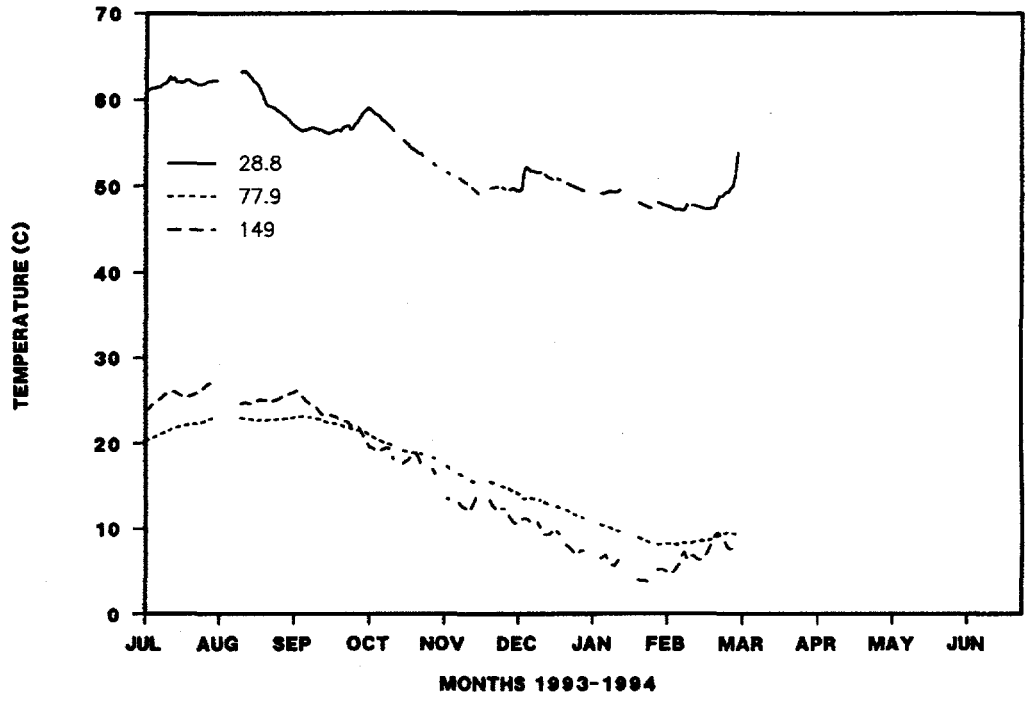

Figure B-17. ORNL lysimeter 5 soil temperatures for 1993-94.

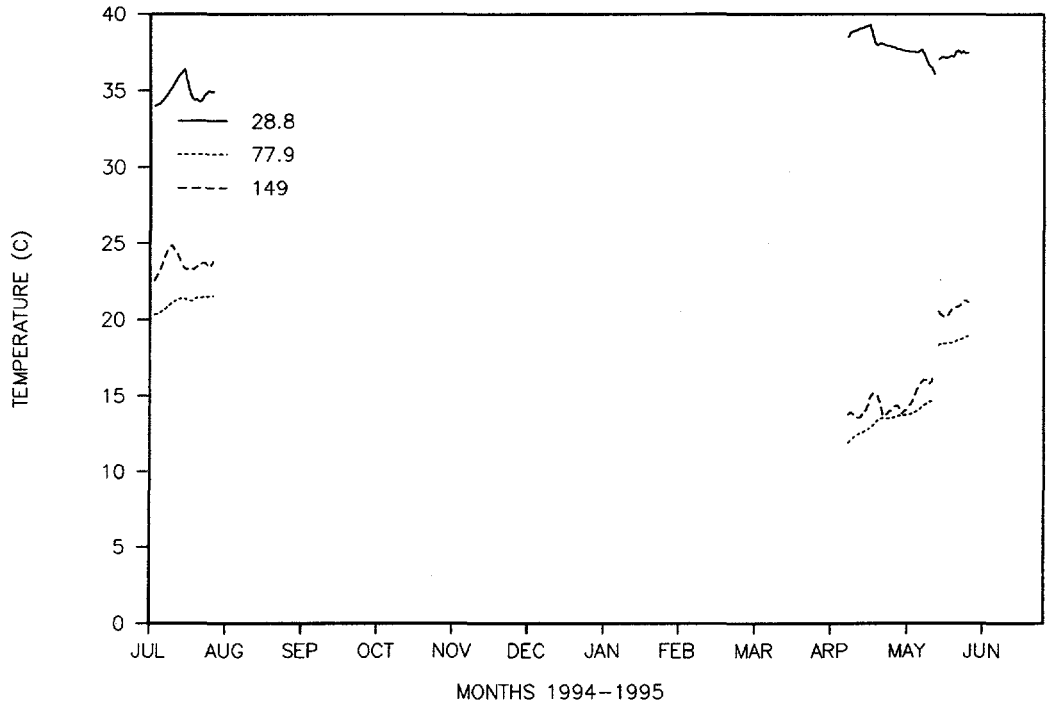

Figure B-18. ORNL lysimeter 5 soil temperatures for 1994-95.

Appendix C

Soil Moisture Data—Resistance Probes

Appendix C
Soil Moisture Data—Resistance Probes

List of Figures

Site	Lysimeter number	Year 1993–94	Year 1994–95
ANL-E	1	C-1	C-2
	2	C-3	C-4
	3	C-5	C-6
	4	C-7	C-8
	5	C-9	C-10
ORNL	1	C-11	C-12
	2	C-13	C-14
	3	C-15	C-16
	4	C-17	C-18
	5	C-19	C-20

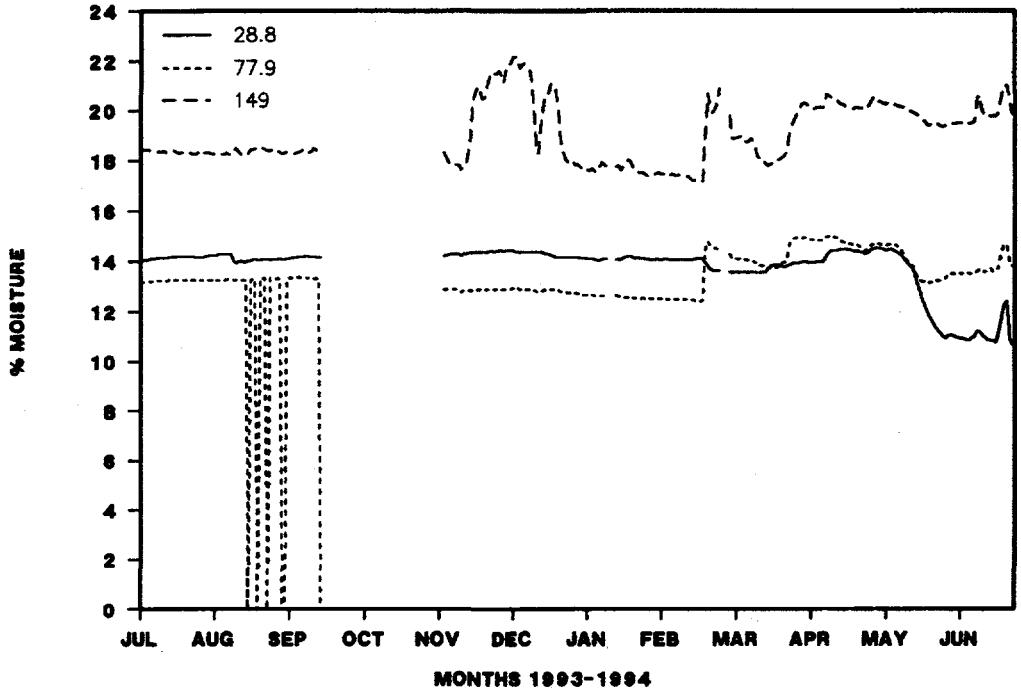

Figure C-1. ANL-E lysimeter 1 soil moisture for 1993-94.

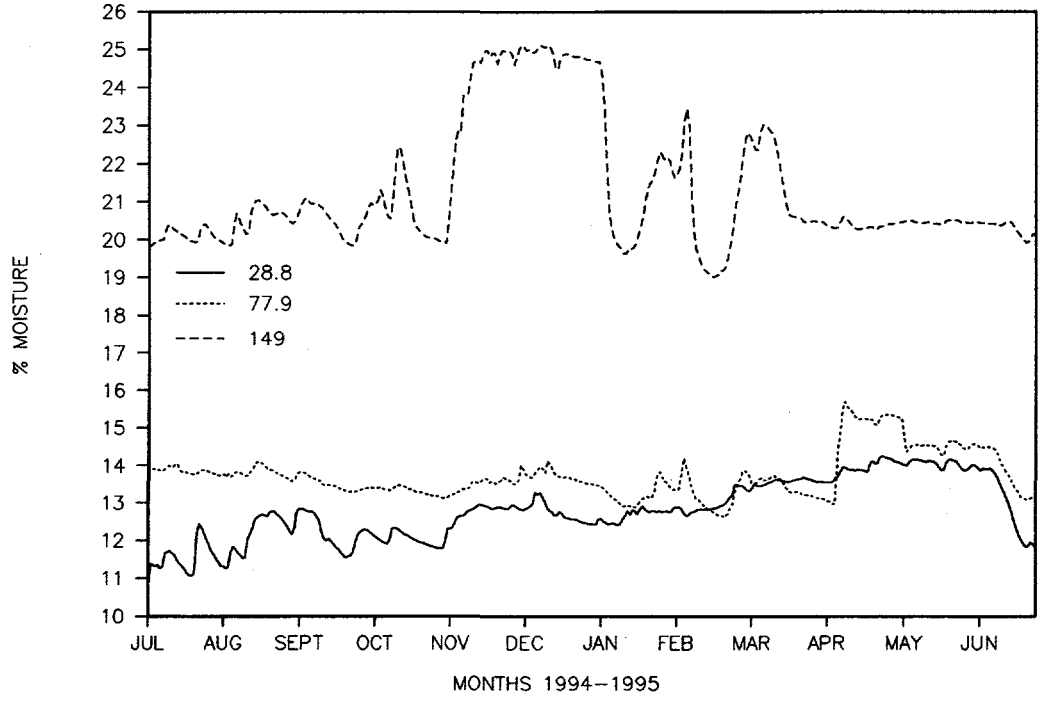

Figure C-2. ANL-E lysimeter 1 soil moisture for 1994-95.

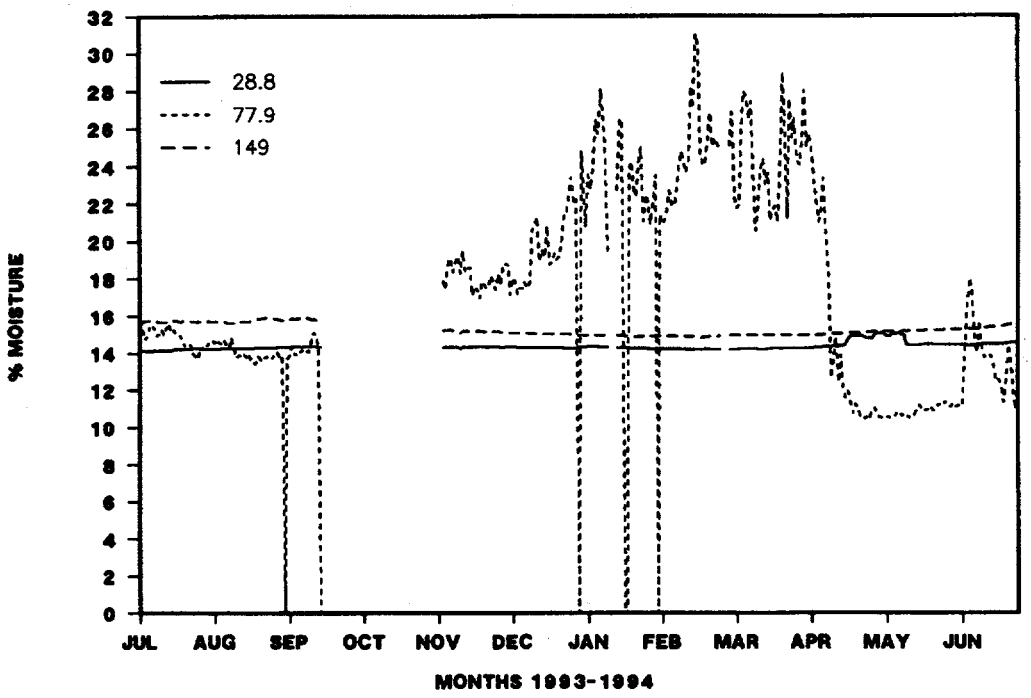

Figure C-3. ANL-E lysimeter 2 soil moisture for 1993-94.

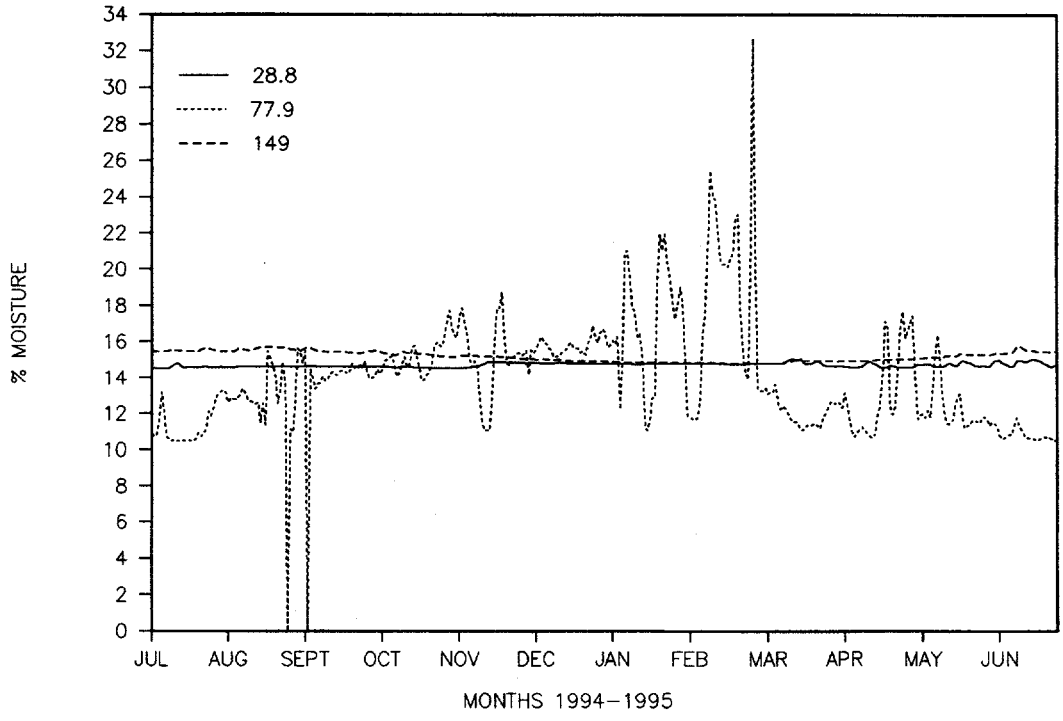

Figure C-4. ANL-E lysimeter 2 soil moisture for 1994-95.

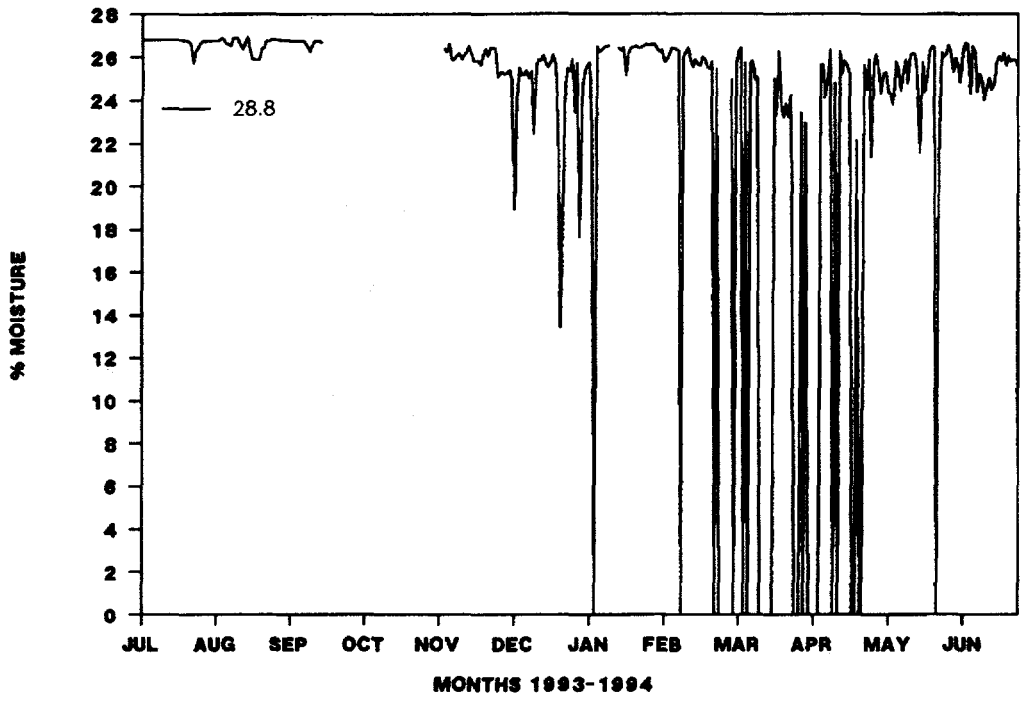

Figure C-5. ANL-E lysimeter 3 soil moisture for 1993-94.

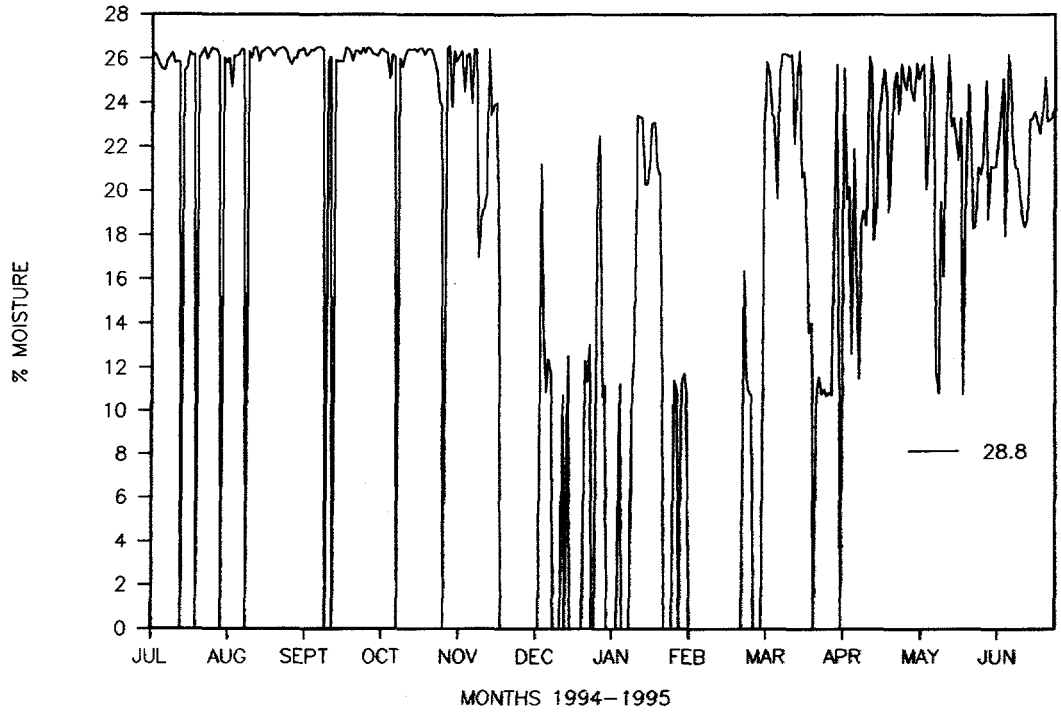

Figure C-6. ANL-E lysimeter 3 soil moisture for 1994-95.

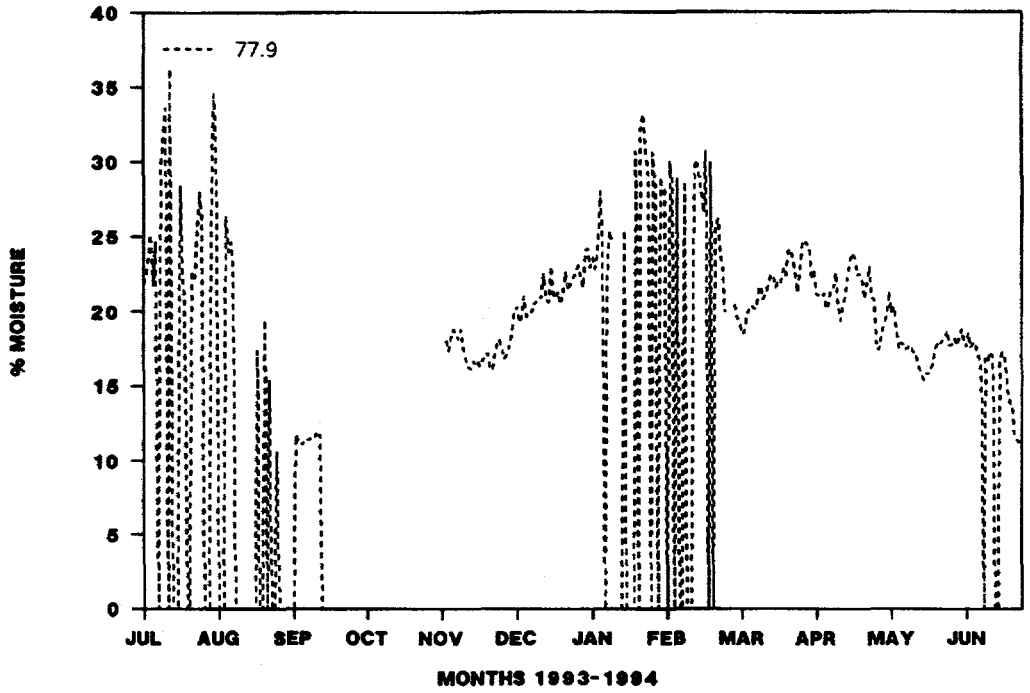

Figure C-7. ANL-E lysimeter 4 soil moisture for 1993-94.

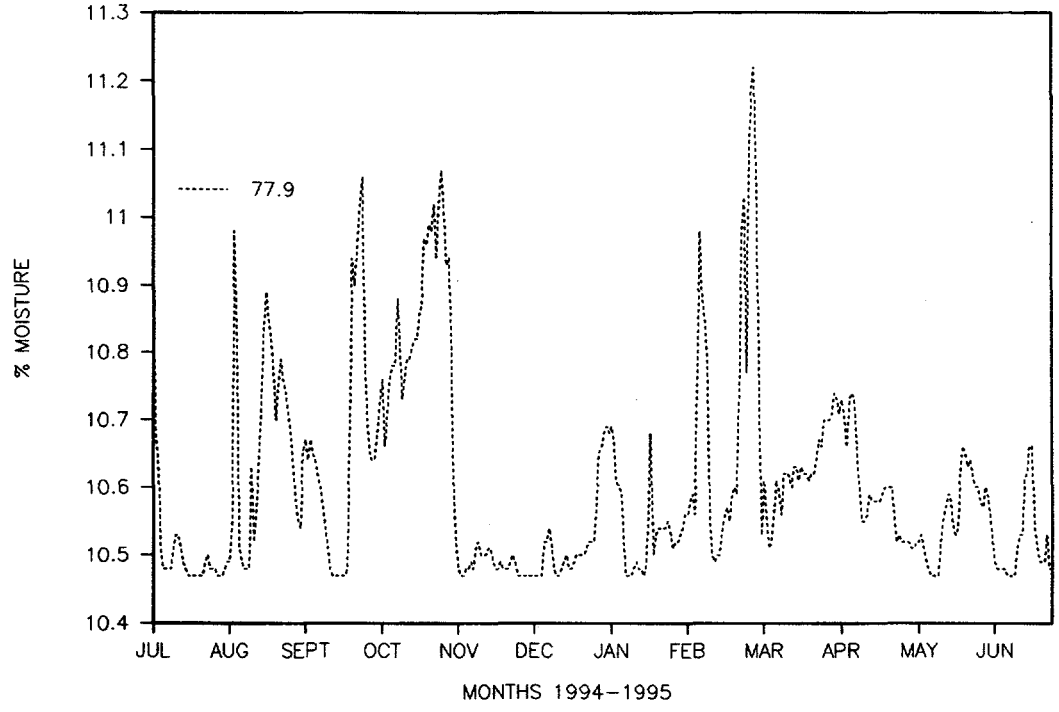

Figure C-8. ANL-E lysimeter 4 soil moisture for 1994-95.

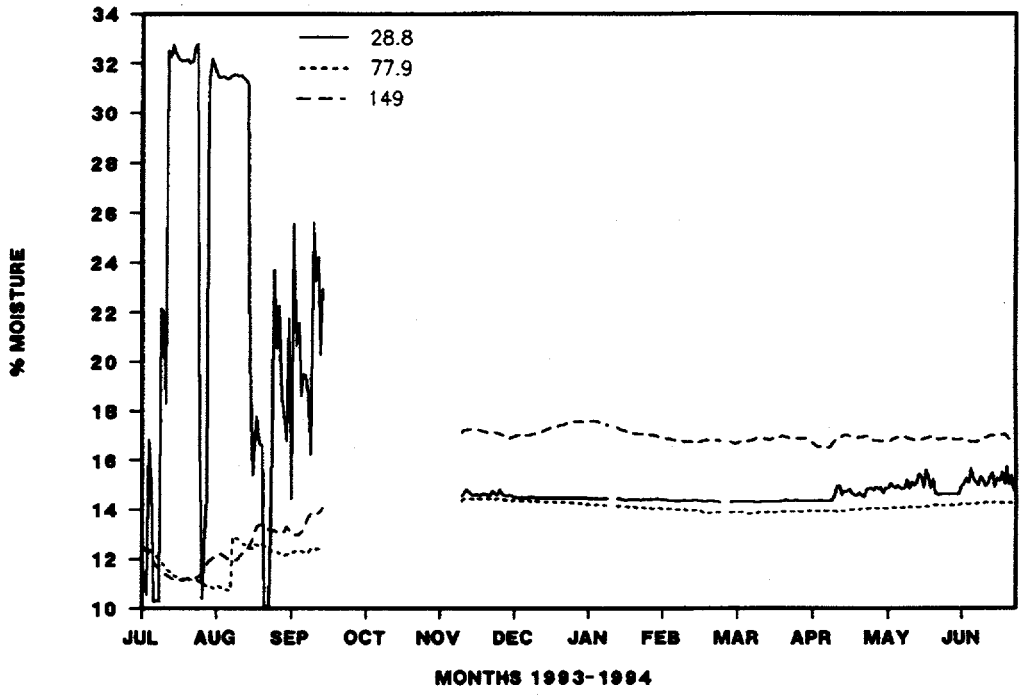

Figure C-9. ANL-E lysimeter 5 soil moisture for 1993-94.

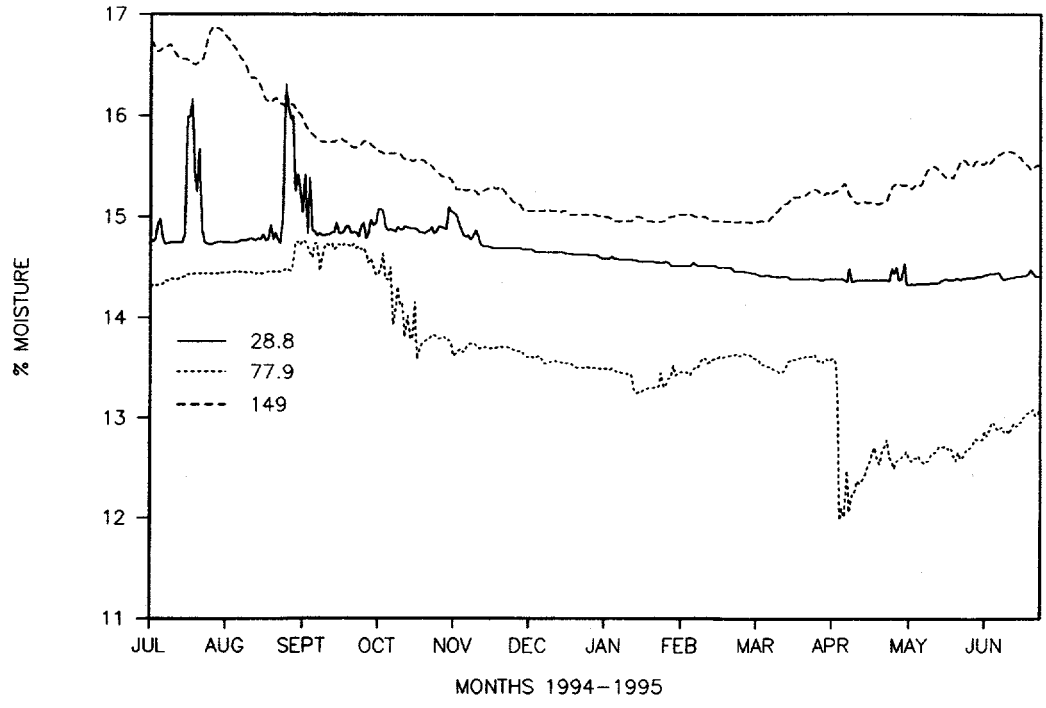

Figure C-10. ANL-E lysimeter 5 soil moisture for 1994-95.

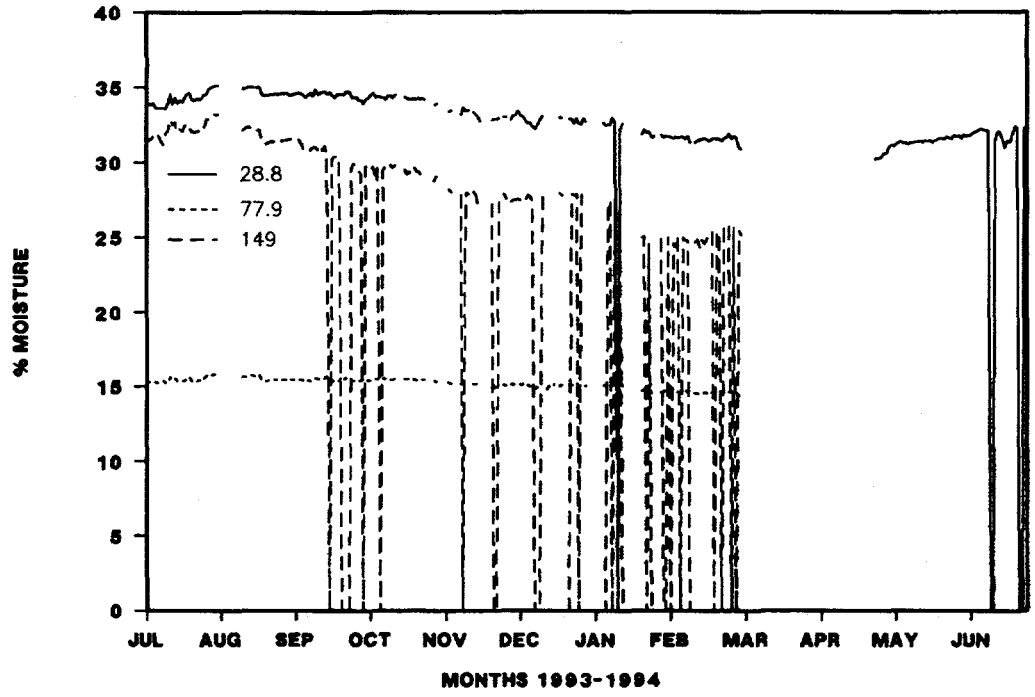

Figure C-11. ORNL lysimeter 1 soil moisture for 1993-94.

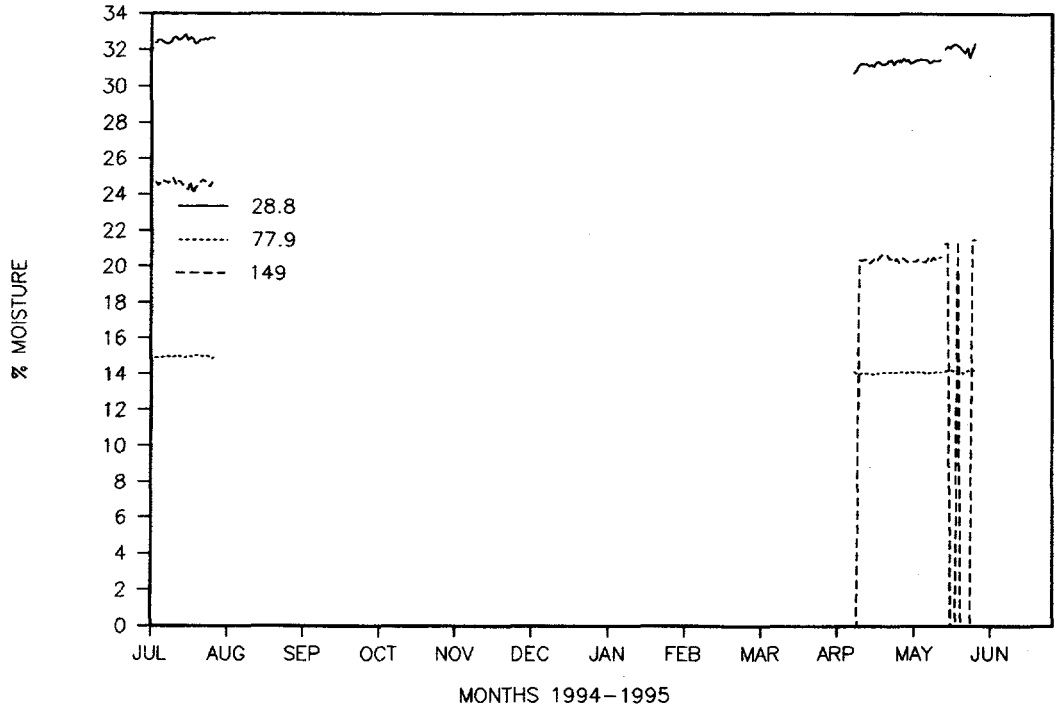

Figure C-12. ORNL lysimeter 1 soil moisture for 1994-95.

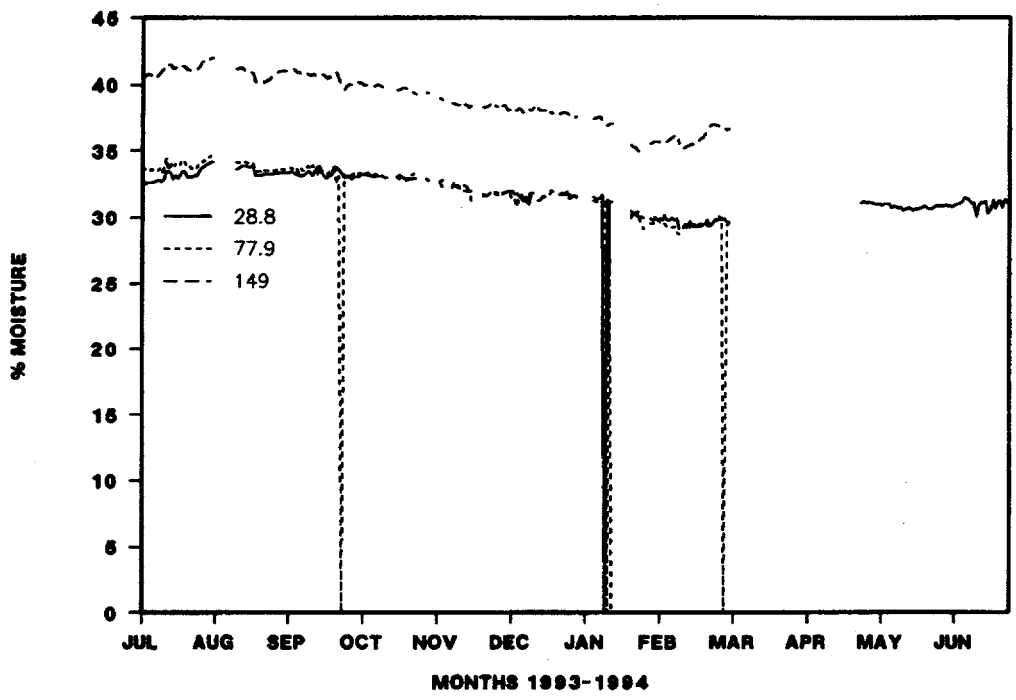

Figure C-13. ORNL lysimeter 2 soil moisture for 1993-94.

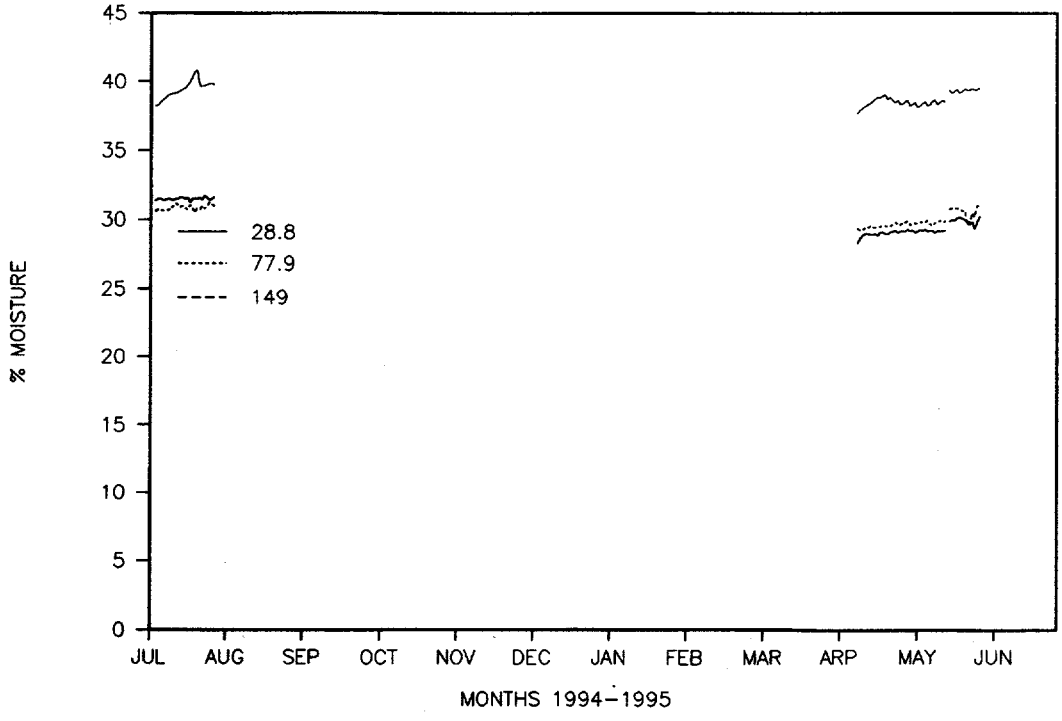

Figure C-14. ORNL lysimeter 2 soil moisture for 1994-95.

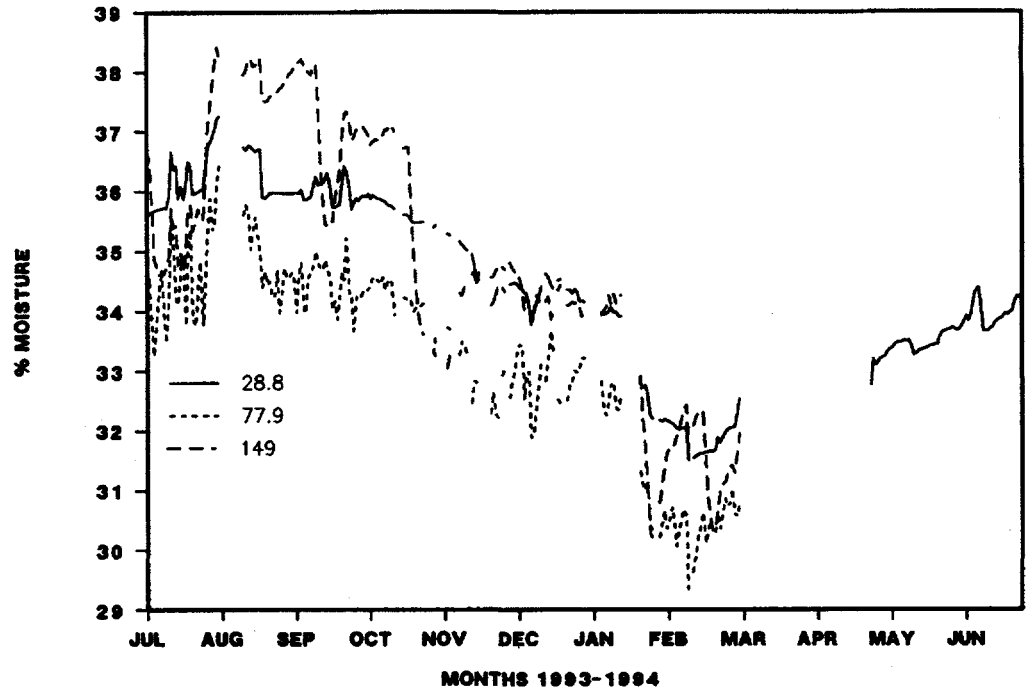

Figure C-15. ORNL lysimeter 3 soil moisture for 1993-94.

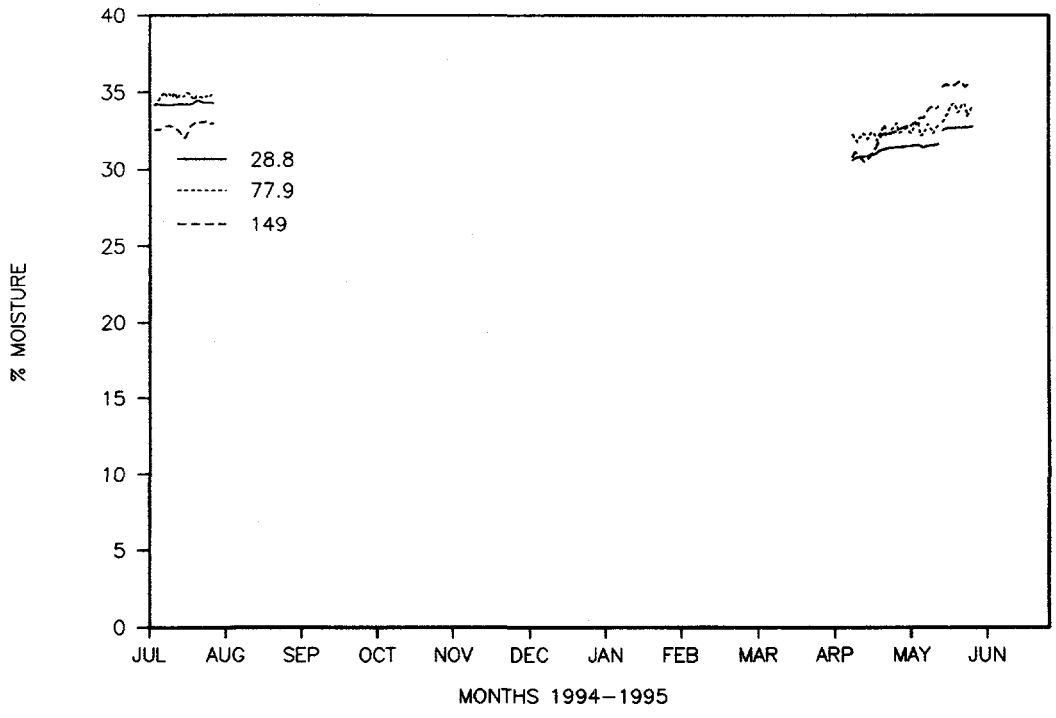

Figure C-16. ORNL lysimeter 3 soil moisture for 1994-95.

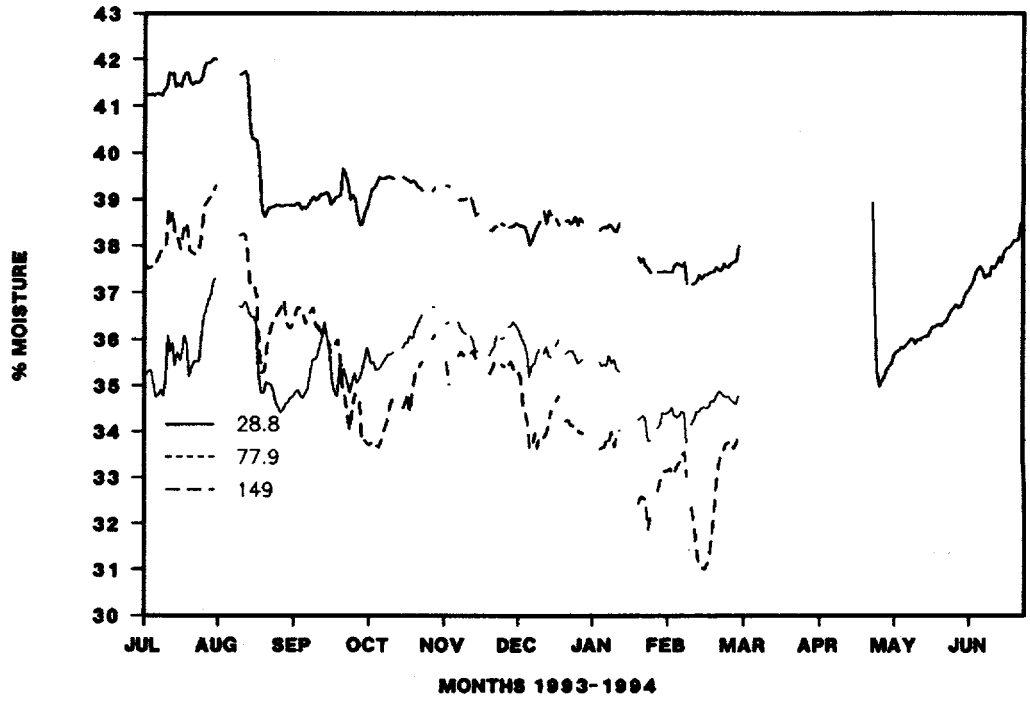

Figure C-17. ORNL lysimeter 4 soil moisture for 1993-94.

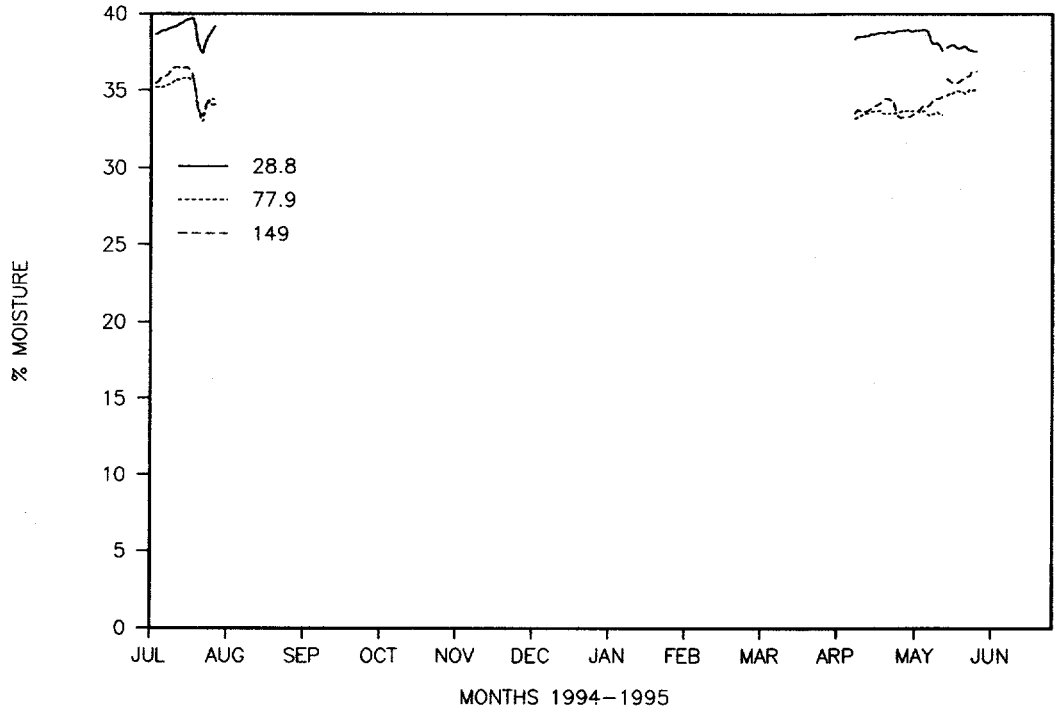

Figure C-18. ORNL lysimeter 4 soil moisture for 1994-95.

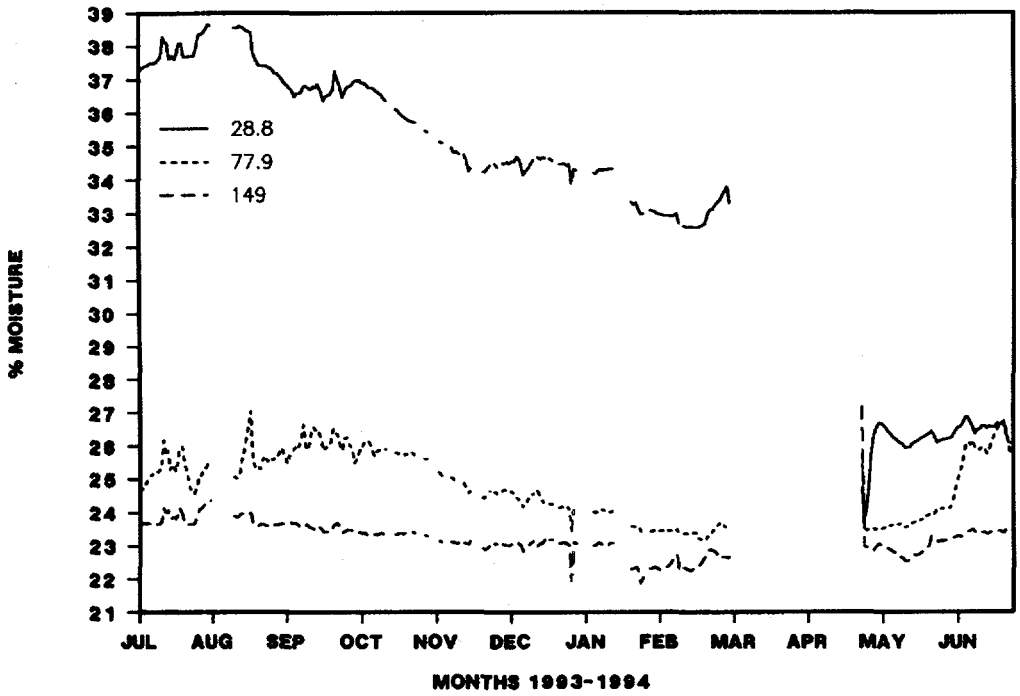

Figure C-19. ORNL lysimeter 5 soil moisture for 1993-94.

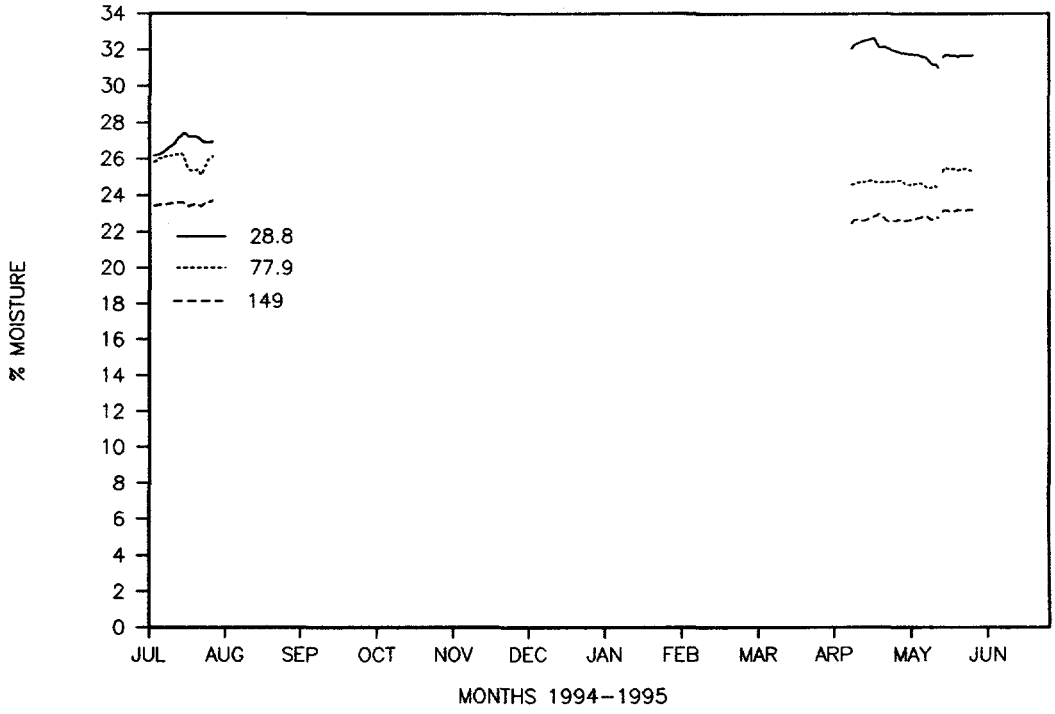

Figure C-20. ORNL lysimeter 5 soil moisture for 1994-95.

Appendix D

Soil Moisture Data—Gravimetric

Appendix D
Soil Moisture Data—Gravimetric

List of Tables

Site	Year 1993–94	Year 1994–95
ANL-E	D-1	D-2
ORNL	D-3	D-4

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

Lysimeter	Depth (cm)	% moisture (dry weight) Gravimetric	Neutron probe
1	0–41	15.3	—
1	41–62	19.6	19.2
1	62–85	23.1	20.1
1	82–107	23.3	—
1	107–133	23.3	—
1	133–153	22.6	20.7
1	153–182	23.1	—
1	182–202	23.4	20.6
2	0–41	15.7	—
2	41–62	18.6	16.4
2	62–82	20.4	18.4
2	82–107	23.1	—
2	107–133	24.3	—
2	133–153	24.5	18.8
2	153–182	24.0	—
2	182–202	24.4	19.6
3	0–41	16.3	—
3	41–62	20.0	17.3
3	62–82	23.4	21.2
3	82–107	23.3	—
3	107–133	22.7	—
3	133–153	24.0	19.9
3	153–182	23.8	—
3	182–202	24.5	20.7
4	0–41	17.4	—
4	41–62	22.1	19.4
4	62–82	22.0	21.4
4	82–107	24.1	—
4	107–133	23.6	—
4	133–153	24.2	21.5
4	153–182	24.2	—
4	182–202	23.9	21.6

a. Samples were collected on July 21, 1994.

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

Lysimeter	Depth (cm)	% moisture (dry weight) Gravimetric	Neutron probe
1	0–41	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 D-3. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

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

a. Samples were collected on June 13, 1994.

Table D-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.

Appendix E

Results of Beta and Gamma Analysis

Appendix E
Results of Beta and Gamma Analysis

List of Tables

Site	Year	
	1993–94	1994–95
ANL-E	E-1	E-2
ORNL	E-3	E-4

Table E-1. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 9 (1993–1994).

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

a. Concentration ±2 sigma.

b. One-L subsample from leachate collector.

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

d. None detected.

Table E-2. 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 E-3. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 9 (1993–1994).

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

Table E-3. (continued).

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

a. Concentration ± 2 sigma.

b. One-L subsample from leachate collector.

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

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

e. None detected.

Table E-4. 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 E-4. (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.

Appendix F

Results of Chemical Speciation

Appendix F
Results of Chemical Speciation

List of Tables

Site	Year	
	1993-94	1994-95
ANL-E	F-1	F-2
ORNL	F-3	F-4

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

		Cation					Anion			
Sample	Solidification agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	100	9.5	13	1.8	54	2.4	0.6	0.7	37
Lys 1-3		24	3.0	3.2	0.6	13	8.5	7.3	<0.5	57
Lys 1-5		37	1.6	6.4	0.4	16	1.9	<0.1	<0.5	28
Lys 2-2	Cement	85	6.7	11	0.3	46	4.3	<0.1	<0.5	23
Lys 2-3		145	13.0	14	0.6	58	3.6	<0.1	<0.5	36
Lys 2-4		115	7.0	14	0.3	61	11	<0.1	<0.5	56
Lys 3-1	VES	119	4.8	15	0.9	45	1.6	1.1	<0.5	21
Lys 3-3		86	6.1	14	0.3	47	6.6	<0.1	<0.5	22
Lys 3-5		93	2.6	15	0.2	45	1.5	0.4	<0.5	24
Lys 4-1	VES	NA	NA	NA	NA	NA	13	<0.1	<0.5	36
Lys 4-3		88	5.4	10	0.2	43	3.5	<0.1	<0.5	34
Lys 4-5		67	2.4	11	0.5	27	2.4	<0.1	<0.5	17
Lys 5-1	Cement	9.4	2.5	8.4	0.6	3.9	1.6	1.5	<0.5	4.6
Lys 5-3		9.8	8.6	25	4.6	4.4	1.7	1.4	<0.5	5.6
Lys 5-5		8.3	1.1	108	1.4	3.3	1.6	2.3	<0.5	6.0

NA = no sample.

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

		Cation					Anion			
Sample	Solidification agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	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 F-3. ORNL chemical speciation results from lysimeter moisture cups 1, 3, and 5, June 1994.

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

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

Table F-4. 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.

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11. ABSTRACT (200 words or less)
The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program, funded by the U.S. Nuclear Regulatory Commission (NRC), is (a) studying the degradation effects in EPICOR-II organic ion-exchange resins caused by radiation, (b) examining the adequacy of test procedures recommended in the Branch Technical Position on Waste Form to meet the requirements of 10 CFR 61 using solidified EPICOR-II resins, (c) obtaining performance information on solidified EPICOR-II ion-exchange resins in a disposal environment, and (d) determining the condition of EPICOR-II liners.

Results of the final 2 (10 total) years of data acquisition from operation of 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 experimental equipment is described and results of waste form characterization using tests recommended by the NRC's "Technical Position on Waste Form" are presented. The study is designed to provide continuous data on nuclide release and movement, as well as environmental conditions, over a 20-year period. At the end of the tenth year, the experiment was closed down. Examination of soil and waste forms is planned to be conducted next and will be reported later.

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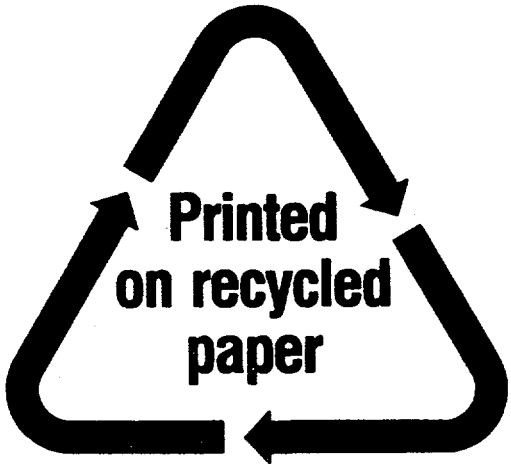

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