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Field Lysimeter Investigations–Test Results

Low-Level Waste Data Base Development Program: Test Results for Fiscal Years 1986, 1987, 1988, and 1989

Prepared by
J. W. McConnell, Jr., R. D. Rogers, M. W. Findlay, E. C. Davis, J. D. Jastrow,
R. M. Neilson, Jr., L. D. Hilton

Idaho National Engineering Laboratory
Lockheed Idaho Technologies Company

Prepared for
U.S. Nuclear Regulatory Commission

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Prepared by
J. W. McConnell, Jr., R. D. Rogers, M. W. Findlay,[1] E. C. Davis,[2] J. D. Jastrow,[3]
R. M. Neilson, Jr., L. D. Hilton

Idaho National Engineering Laboratory
Managed by the U.S. Department of Energy

Lockheed Idaho Technologies Company
Idaho Falls, ID 83415

Prepared for
Division of Regulatory Applications
Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, DC 20555-0001
NRC Job Code A6876

[1]Formerly with Argonne National Laboratory, Argonne, IL
[2]Formerly with Oak Ridge National Laboratory, Oak Ridge, TN
[3]Argonne National Laboratory, Argonne, IL

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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 first 4 years of data acquisition from the field testing are presented and discussed. During the continuing field testing, both Portland type I–II cement and Dow vinyl ester-styrene waste forms are being tested in lysimeter arrays located at Argonne National Laboratory-East in Illinois and at Oak Ridge National Laboratory. The 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.

FIN No. 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 the 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 in Illinois, is to expose samples of ion-exchange resin (which were solidified in task two) 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 first 4 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.

Field Lysimeter Investigations: Low-Level Waste Data Base Development Program Lysimeter Test Results for Fiscal Years 1986, 87, 88, and 89

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 subsequent 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 revised BTP,[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 this study, EPICOR-II waste forms were subjected to the recommended NRC test procedures to ensure compliance with the BTP stability requirements

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and to characterize the waste forms. The solidification studies were completed and reported. Results are briefly reviewed in this report.

In the Field Testing work, 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 objectives of the Field Testing task are to (a) examine the performance of the waste forms in typical low-level waste disposal environments, (b) compare field results with bench leach studies and with Department of Energy Special Waste Program field test results, and (c) develop a low-level radioactive waste field leach-rate data base for use in performance assessment source term calculations. This report discusses the results obtained during the first 4 years of operation of the experiment.

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, will be obtained over a 20-year period.

This report contains data from the first 4 years of lysimeter operation,[6–9] 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 10.

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.

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. 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.

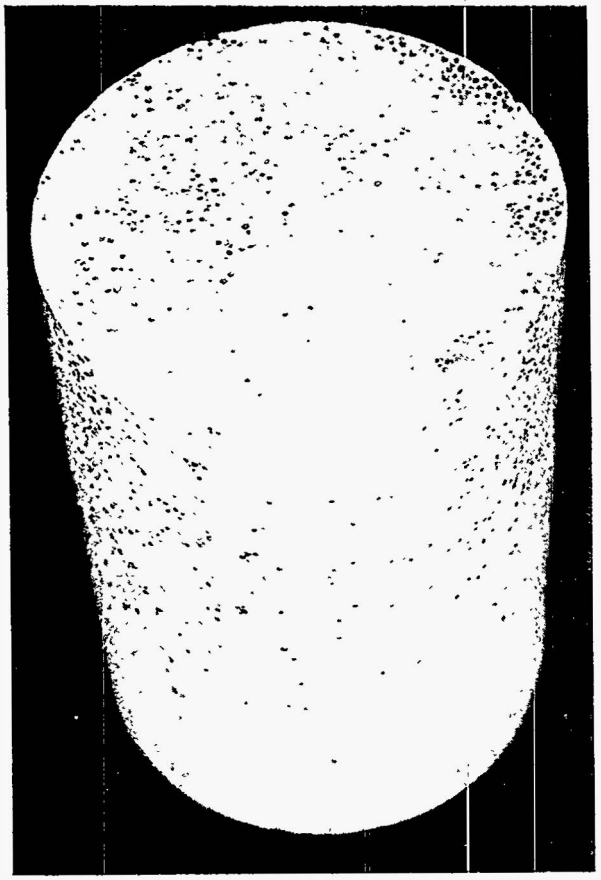

Figure 1. An example of an EPICOR-II prefilter waste form.

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 activities are given in Table 2.

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. 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. 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-05 ± 2.83E-07
	Cs-137	1.17E-03 ± 9.90E-05
	Sr-90	6.92E-05 ± 7.21E-06
PF-24	Cs-134	3.30E-04 ± 5.80E-05
	Cs-137	4.99E-03 ± 3.04E-04
	Sr-90	1.18E-05 ± 6.36E-07

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.

Binder	Waste type	Compressive strength ± 1σ (psi) 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.

Binder	Waste type	Leachant	Gamma dose (rad)	Leachability index Cs-134	Cs-137	Sr-90
PC	PF-7	DI	0	10.3	10.3	—
PC	PF-7	DI	5.3E+08	9.4	9.3	—
PC	PF-24	DI	0	10.6	10.4	—
PC	PF-24	DI	5.4E+08	10.0	9.9	—
PC	PF-7	SW	0	9.6	9.5	—
PC	PF-7	SW	5.3E+08	10.0	9.9	—
PC	PF-24	SW	0	10.4	10.3	—
PC	PF-24	SW	5.4E+08	10.9	10.8	—
PC	PF-7	DI	5.3E+08	—	—	9.0
VES	PF-7	DI	0	12.4	12.2	—
VES	PF-7	DI	5.7E+08	9.8	9.7	—
VES	PF-24	DI	0	14.0	13.8	—
VES	PF-24	DI	4.9E+08	12.3	12.2	—
VES	PF-7	SW	0	9.4	9.3	—
VES	PF-7	SW	5.7E+08	8.8	8.7	—
VES	PF-24	SW	0	10.9	10.7	—
VES	PF-24	SW	4.9E+08	10.0	9.8	—
VES	PF-7	DI	5.7E+08	—	—	9.7

PC = Portland type I–II cement.

VES = Vinyl ester-styrene.

DI = Demineralized water.

SW = Synthetic seawater.

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.

VES and cement waste forms were placed in nutrient-rich media to test the growth of the applied species of fungi and bacteria. 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 11; bench testing of those EPICOR-II waste forms, according to the recommendations of the BTP, is further described in References 7, 12, and 13.

Description of Test Sites

Field testing is being conducted at Argonne National Laboratory (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 2 and 3. Both sites offer unobstructed exposure to prevailing environmental conditions while providing security from inadvertent personnel exposure to irradiation or contamination.

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.[14]

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.[15]

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

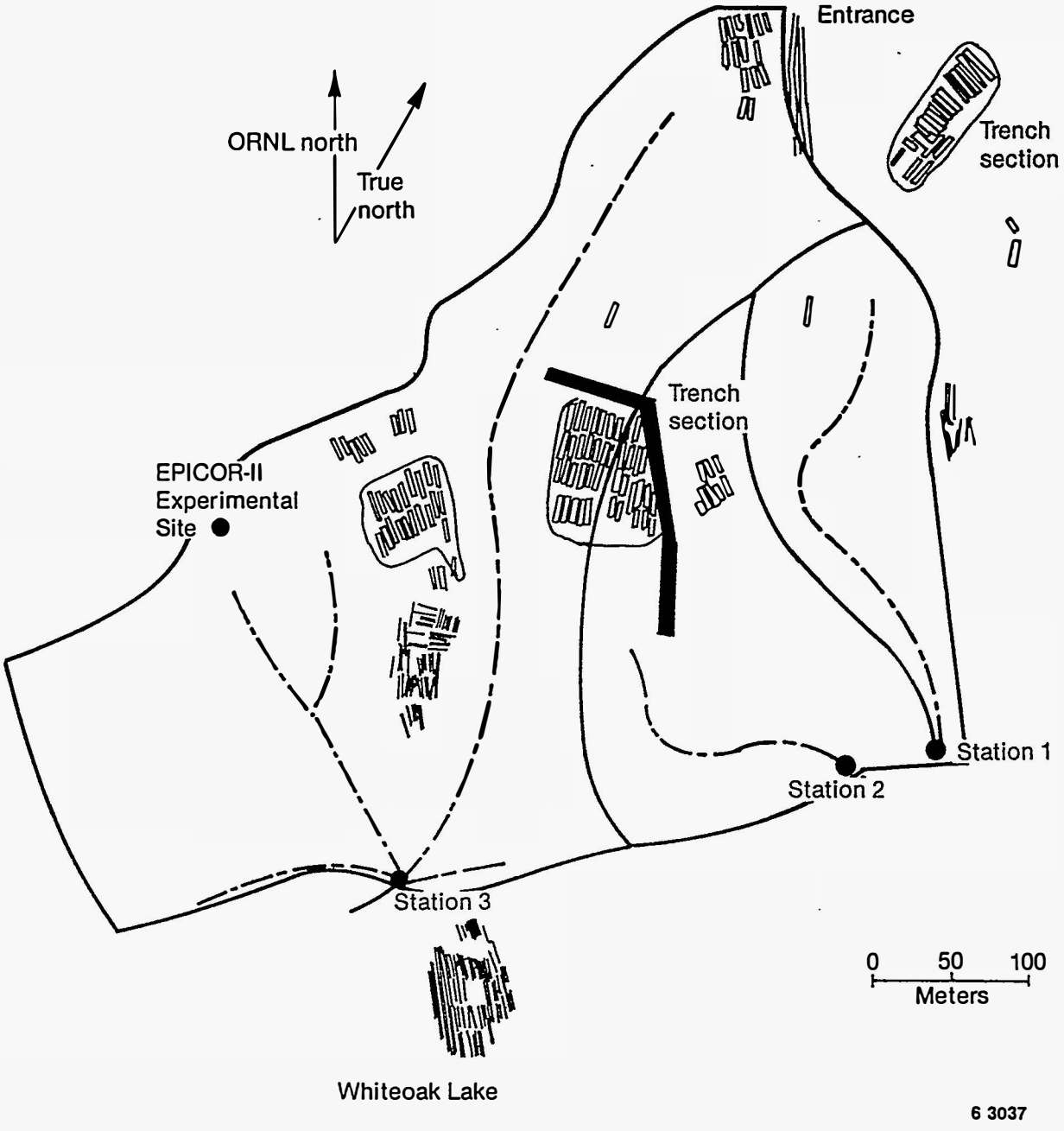

Figure 2. Location of the EPICOR-II lysimeter experiment at ORNL.

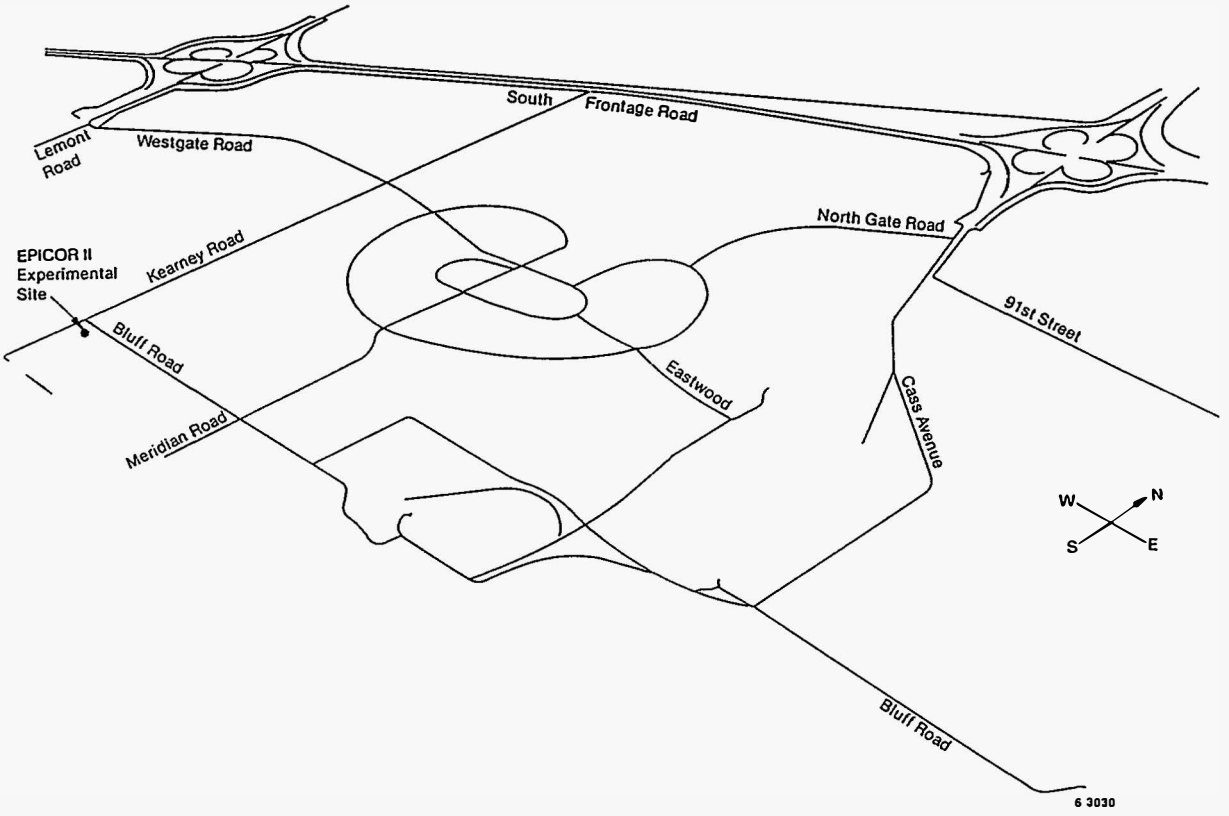

Figure 3. Location of the EPICOR-II lysimeter experiment at ANL-E.

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 4). Internally, the lysimeter is divided into two sections, the upper being 1,532 L in volume and the lower being 396 L (Figure 5). 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.[10] 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.

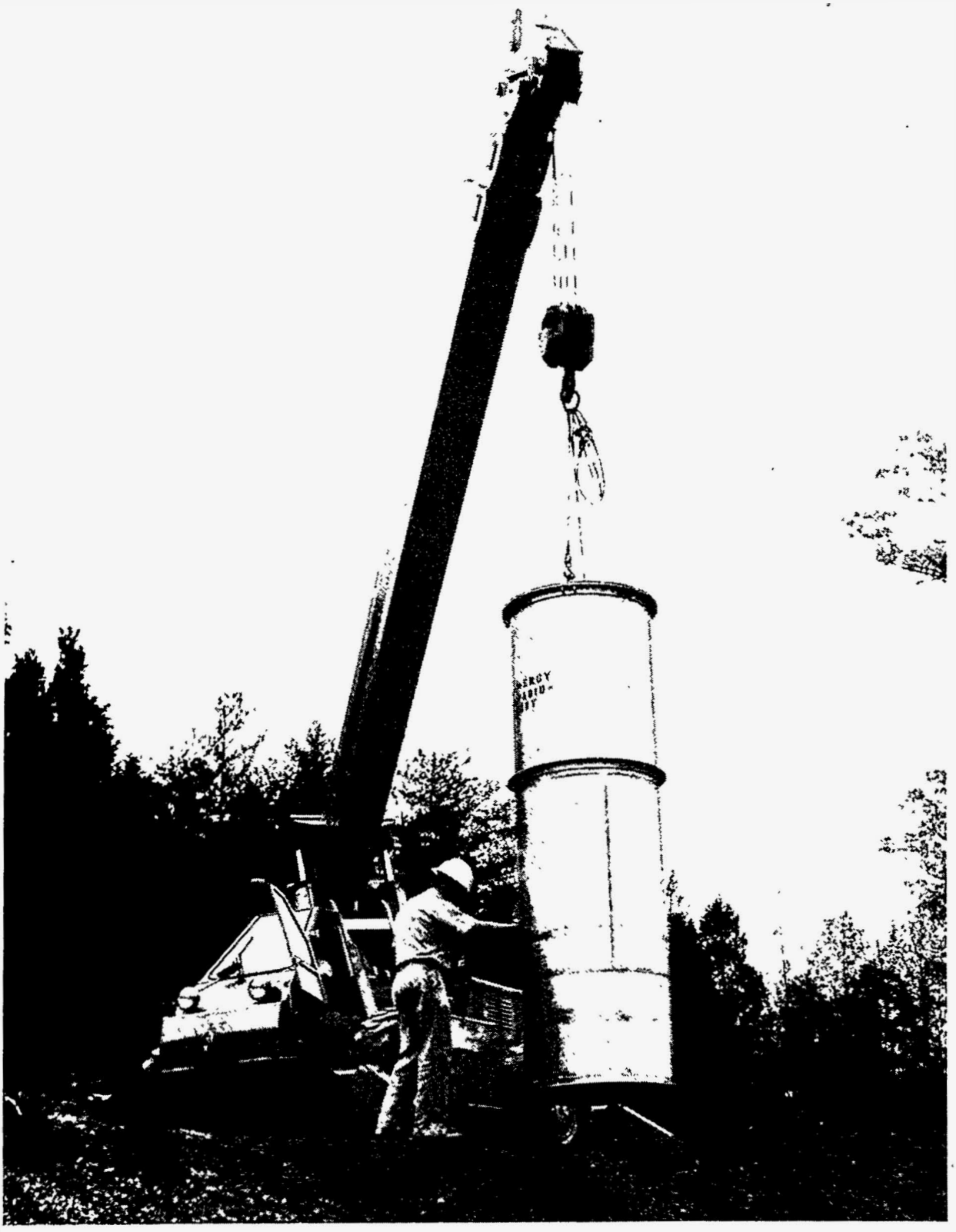

Figure 4. Unfilled lysimeter vessel being lowered into position at ORNL.

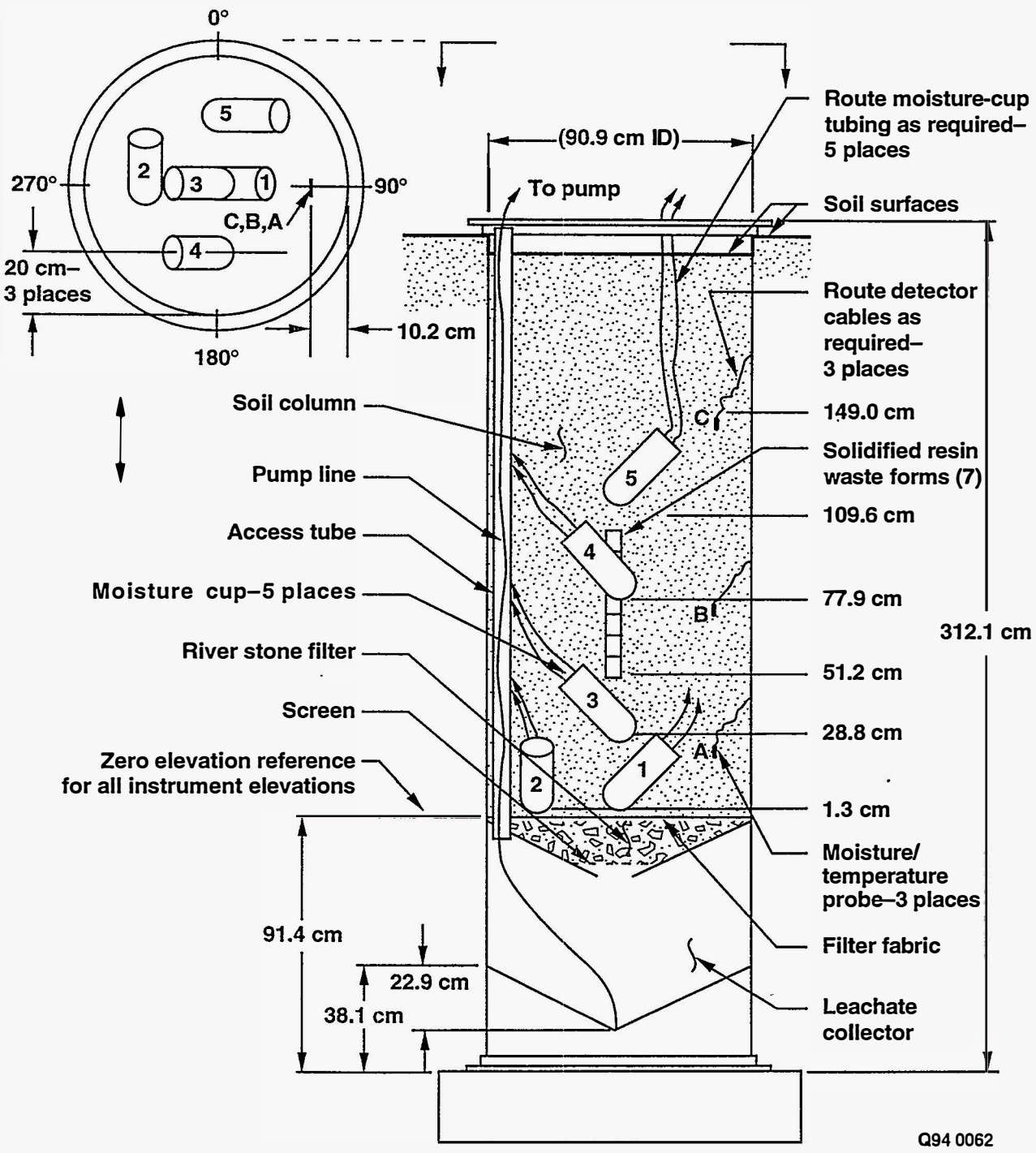

Figure 5. Lysimeter vessel component locations.

Table 5. Lysimeter waste form composition.

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

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

	Soil		
		ORNL	
Characteristic	ANL-E	Savanah 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.

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 5). 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 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 5). 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 weighes 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 (0000 h), the system processes the data from 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.

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 display 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 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

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

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 1989. In addition, information on water balance, nuclide, and cation/anion content in soil water and leachate is presented. Much of the data is displayed in graphic format so that information can easily be correlated with time.

Each DAS functioned fairly well during the first 4 years. However, from late August 1985 into November 1985, the ORNL system was at the manufacturer for repair. Also, data for the last week of September and first 3 weeks of October 1986 were lost from the ANL-E DAS system due to a malfunction of a microprocessor chip on the control board. The malfunction was discovered during a routine data transfer and was repaired at that time.

Weather Data

Precipitation, air temperature, wind speed, and relative humidity, as recorded by the ANL-E and ORNL systems during the 48-month reporting period, are presented in Appendix A. Average annual precipitation for the period was ~~338.2~~ 342.6 cm at ANL-E and ~~460.2~~ 445.1 cm at ORNL. ANL-E was near the normal annual rainfall[14] of ~~340.8~~ 85.6 cm, while ORNL had 83% of the normal annual rainfall[15] of ~~555.2~~ 538.8 cm. FY-89 was the first year in the experiment that ORNL reached the normal amount of yearly precipitation. The monthly precipitation pattern for each site can be seen from the histograms in Figures A-1 through A-4 and Figures A-17 through A-20 in Appendix A. Figure 6 shows the cumulative precipitation for both sites since the initiation of field work.

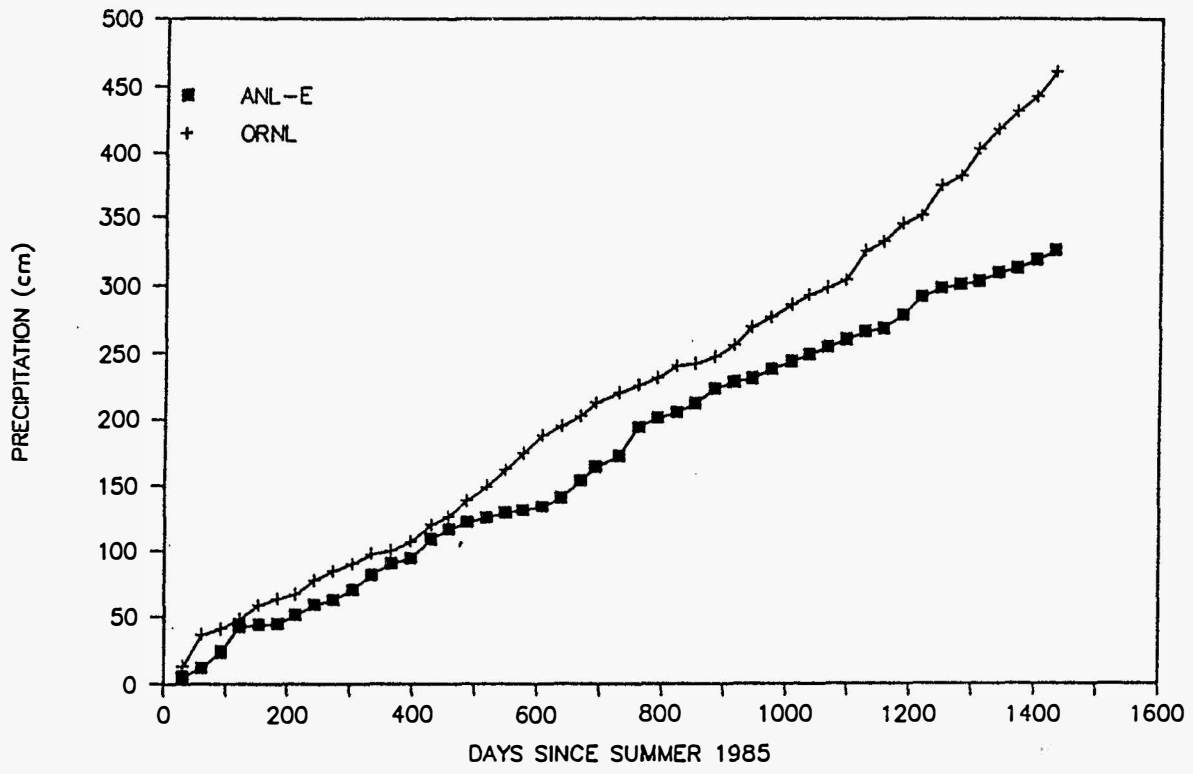

Figure 6. ANL-E and ORNL cumulative precipitation.

Rainfall events from the ORNL site appear greatly exaggerated. This trend became apparent during December 1985, and early indications were that the Weather Measure tipping-bucket rain gauge supplied with the DAS was not capable of accurately responding to periods of intense rainfall. In June 1986, this 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 rain gauge 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.

Corrective measures for determining the source of these spurious data are ongoing. They include monitoring the rain gauge with a separate, single-channel data collection system and testing the circuits within the DAS that are responsible for processing rainfall data. This malfunction has 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 were used to calculate the total quantities of precipitation received by each site.

Air temperature data from ANL-E show that there were periods of freezing temperatures from mid-November 1988 until near mid-March each year, except 1987, when low temperatures extended into mid-April. ORNL experienced some days in which there was an average air temperature 0°C or lower (Figures A-5 through A-8 and Figures A-21 through A-24).

Lysimeter Soil Temperature Data

Soil temperature and moisture sensors are physically located within a common housing or probe. These probes are located at three elevations: 149, 77.9, and 28.8 cm, as measured from the bottom of the soil column within each lysimeter (Figure 5). 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-39 of Appendix B. At no time during the reporting period was a valid freezing temperature recorded at any depth within a lysimeter. A direct correspondence can be seen between air temperature and soil temperatures at both locations.

Some abnormally low soil temperature readings were observed from the intermediate and bottom probes in lysimeter ANL-3 in January 1986 and in ANL-4 by June 1986 (Figures B-9 and B-13). There were no such occurrences with near-surface probes. One possible explanation for the malfunction is related to an average soil subsidence of 30 cm in all ANL-E lysimeters except the sand-filled control. It is hypothesized that subsiding soil may have caused damage to the lead wires connecting the probes to the soil surface. These probes are now being replaced with new ones, and data from the replacements shows that they are functioning normally. An example of how closely temperature data from the ANL-E lysimeters tracked each other can be seen in Figures B-1, B-5, and B-16.

The bottom temperature probes in ORNL-3 and -5 have consistently indicated elevated soil temperature (Figures B-28 and B-36). 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. All of the other temperature

probes at ORNL are functioning, including the probes at the 77.9-cm elevation, which are close to the waste forms.

It was found during the first year of operation that five of the 15 moisture and temperature probes at ANL-E failed. In August of 1986, two new probes were placed in ANL-3 to replace failed probes located at 28.8 and 77.9 cm. The replacement probes provided satisfactory temperature data during the 1986–87 reporting period.

In April of 1987, the two failed ANL-3 probes were retrieved and returned to Soil Test, the vendor that had supplied them. During examination of the probes, the inside was found to be extensively corroded. In addition, coating surrounding the temperature-sensing thermistor was degraded in each of the probes. It was obvious that some environmental condition at ANL-E decreased the useful life of the probes. Soil Test representatives indicated that less frequent reading of the probes (perhaps once a day or even once a week) should reduce corrosion and extend the life of the probes.

By July of 1988, all of the probes in ANL-4 had failed; therefore, the 1988–89 data for ANL-4 are not included in this report. Also during 1988, the replacement probe at 77.9 cm in ANL-3 failed, and by 1989, none of the probes in ANL-3 were functioning properly. A replacement probe is being sought. Arrangements are also being made to test the potential for using a neutron probe to measure moisture within the lysimeters.

Lysimeter Soil Moisture Data

Data from the moisture probes at both ANL-E and ORNL, shown in Figures C-1 through C-40 in Appendix C, indicate that the lysimeter soil columns at both sites have remained moist during the reporting period after an initial wetting period of 2 months (ORNL) or 3 months (ANL-E). The moisture content of the soil column of each lysimeter over time (as determined by averaging the outputs of the three probes in each lysimeter) shows that the variation in moisture data for the lysimeters at each site is relatively small and not excessive (Table 12). The probes continue to serve their original purpose of providing some indication of the status of lysimeter soil moisture. As was mentioned earlier in the Lysimeter Soil Temperature Data section, some of the probes at ANL-E are no longer functioning.

The actual moisture of the soil column in each lysimeter at each site has been determined gravimetrically once each year (see Tables D-1 through D-8 in 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 probe data recorded near the time of the gravimetric determination (Table 12). Percent differences between the gravimetric data and probe data for ANL-E lysimeters range between a low of 2.9% in 1986–87 to a high of 43.7% in that same year. While these values increased in the fourth year, they were still within a reasonable range given the use of the information. Data from ORNL probes overestimate the actual percent soil moisture from a low of 120.1 in 1988–89 to a high of 168.8 in 1986–87. Corrective action first consisted of recalculation of the polynomial equation that transforms probe input into percent moisture. Data for recalculation of the equation came from laboratory recalibration of several soil moisture probes using lysimeter soils. Later, information was collected and a second polynomial equation was formulated. This was done following the vendor's procedures for field calibration, which required the use of gravimetrically obtained moisture data collected over a period of several months.

Soil moisture (as gravimetrically determined) at each sampling depth has remained uniformly consistent between intrasite lysimeters during the past several years (Figures 7 and 8). The uniformity of soil moisture in the ANL-E lysimeters (Figure 7) is somewhat surprising given the long-term decrease in water infiltration in ANL-1, -2, and now -3. Action to improve drainage of these two lysimeters was begun in July 1987. Soil from the top of those lysimeters was removed (136 kg) and replaced with similar soil. However, by the spring of 1988, it had become obvious that replacement had not solved the

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

Lysimeter number	Period	Probe data for June 30, 1989	Gravimetric data for July 1989	Percent difference between actual and probe
ANL-1	1986–87	13.4 ± 3.9	23.8 ± 0.3	43.7
ANL-2		19.7 ± 4.5	24.1 ± 0.9	18.3
ANL-3		23.6 ± 2.1	24.3 ± 0.6	2.9
ANL-4		14.5 ± 3.4	24.5 ± 0.4	40.8
ORNL-1		42.4 ± 0.9	17.5 ± 0.9	142.3
ORNL-2		41.6 ± 4.4	16.6 ± 1.2	150.6
ORNL-3		45.7 ± 1.2	17.0 ± 1.0	168.8
ORNL-4		45.4 ± 1.3	17.0 ± 1.0	167.1
ANL-1	1987–88	17.2 ± 7.0	22.0 ± 0.7	22.7
ANL-2		15.5 ± 5.0	21.4 ± 1.6	27.6
ANL-3		16.6 ± 4.2	22.7 ± 2.5	26.9
ANL-4		18.8 ± 1.6	21.6 ± 2.6	13.0
ORNL-1		39.0 ± 3.5	16.8 ± 1.7	142.3
ORNL-2		40.5 ± 1.8	16.8 ± 1.2	141.1
ORNL-3		43.5 ± 1.2	16.6 ± 1.1	162.0
ORNL-4		44.5 ± 1.6	17.6 ± 1.3	166.5
ANL-1	1988–89	15.3 ± 3.3	21.7 ± 1.1	29.5
ANL-2		14.2 ± 3.0	22.1 ± 0.8	35.8
ANL-3		20.1 ± 9.1	23.3 ± 2.0	13.7
ANL-4		13.9 ± 3.4	23.4 ± 2.0	40.6
ORNL-1		40.3 ± 1.4	16.7 ± 1.4	141.3
ORNL-2		41.6 ± 0.2	15.3 ± 0.9	171.9
ORNL-3		36.1 ± 2.9	16.4 ± 1.5	120.1
ORNL-4		38.7 ± 3.8	15.5 ± 1.8	149.7

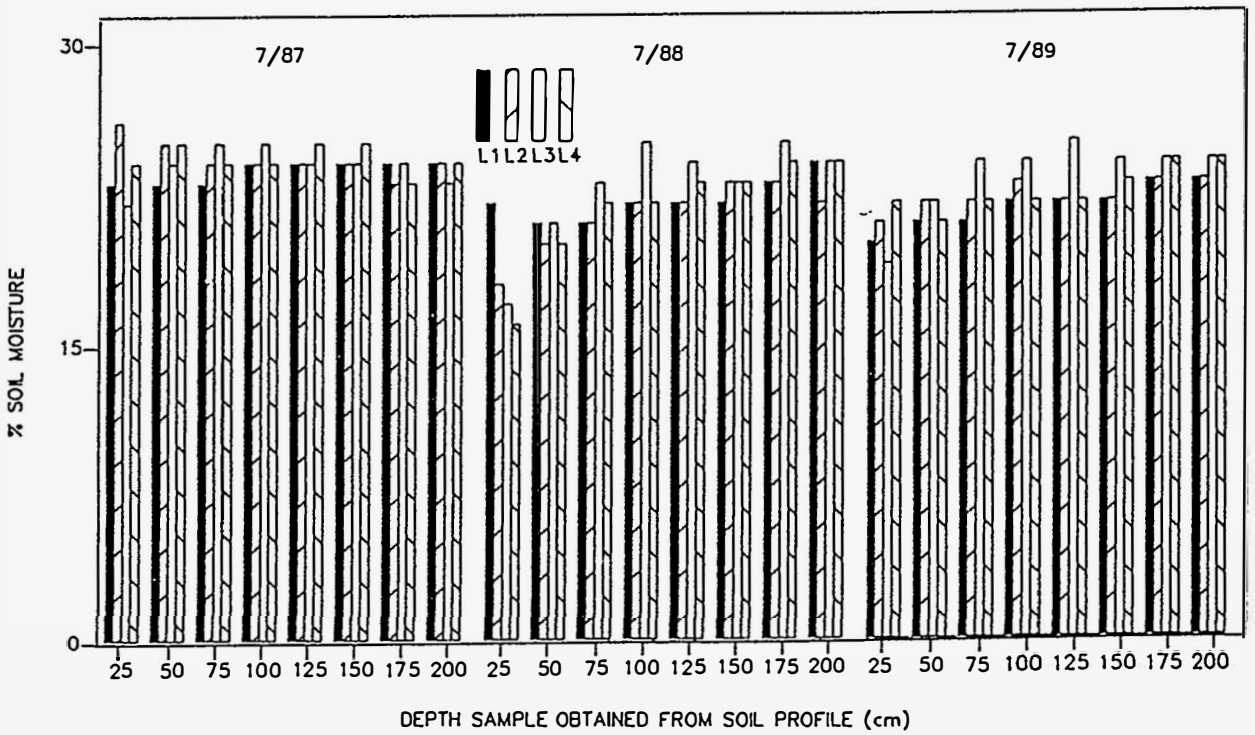

Figure 7. Moisture profile of ANL-E lysimeters 1 through 4 by year based on gravimetric measurement of water content.

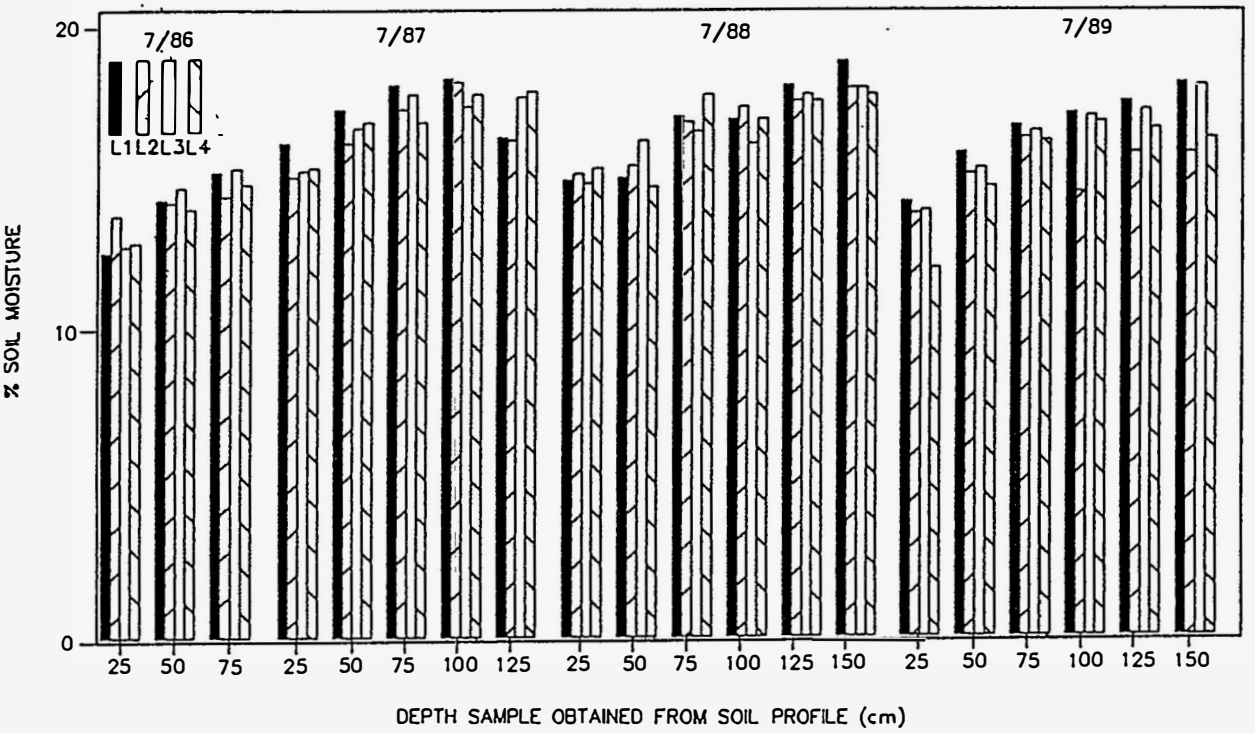

Figure 8. Moisture profile of ORNL lysimeters 1 through 4 by year based on gravimetric measurement of water content.

drainage problems. Apparently, the surface structure of those soils was being degraded by rain impact to the point of interferring with water infiltration. In an effort to improve the situration, about 15 cm of soil (159 kg) was removed from the affected lysimeters in June 1988 and spread on plastic sheets to dry. Soil clods were broken up into pieces 1.5 cm in diameter or less. That structured soil was then placed into the two lysimeters. The exposed soil surface was then covered with a polyester mesh and a 2.5-cm layer of pea gravel. It is thought that this treatment would stop the effects of rain impact and allow for improved water infiltration. Results after significant rainfall events suggested that this method is effective. However, it has now become obvious that the initial rate of drainage cannot be restored, probably because of the soil's low permeability, which is an inherent property of the soil. The present conditions are now thought to be indicative of what would be found if a disposal trench were constructed in this soil. No further effort to improve drainage of these lysimeters is anticipated. Instead, water will no longer be allowed to pond on the soil surface. After each rainfall, water in excess of 2 to 3 cm in depth will be removed. Records of the amounts of water removed will be maintained for use in the water balance calculations.

As is apparent from data presented in Figures 7 and 8, after initial wetting, the vertical moisture content within the lysimeter soil columns at each of the sites appears to have remained relatively uniform (Tables D-1 through D-8). At the time of the last sampling, the average soil moisture of ANL-E soils was 55.7% of the soil moisture holding capacity, while at ORNL, this value was 35.9%. The latter value is lower than expected given the higher precipitation at ORNL.

Measurement of Leachate

By using the cumulative rainfall data from each site since the time the lysimeters were placed in operation (Figure 6), it is possible to calculate the approximate volume of water that has been received by the exposed lysimeter surfaces (6,489.5 cm^2). The cumulative volume of precipitation received by each ANL-E lysimeter was 2,111.7 L; at ORNL, this value was 2,991.6 L. Precipitation per year is listed in Table 13 as well as average volume of leachate through the lysimeters. The volume of the precipitation that has

Table 13. Precipitation received and leachate passing through lysimeters at ANL-E and ORNL.

		ANL-E cumulative		ORNL cumulative	
	Test period	Volume (L)	Total (%)	Volume (L)	Total (%)
Precipitation received	1985–86	607.0	—	643.0	—
	1986–87	1,085.8	—	1,424.1	—
	1987–88	1,639.8	—	1,965.0	—
	1988–89	2,111.7	—	2,991.6	—
Average leachate passed through soil–filled lysimeters	1985–86	128.7 ± 22.6	21.2	441.7 ± 20.9	68.7
	1986–87	441.5 ± 81.2	40.6	1,135.0 ± 22.5	79.7
	1987–88	783.5 ± 182.9	47.8	1,598.0 ± 21.2	82.7
	1988–89	998.0 ± 255.6	47.3	2,558.0 ± 28.5	85.5
Leachate passed through sand–filled lysimeters	1985–86	337.9	55.7	528.0	82.1
	1986–87	945.0	87.0	1,342.0	94.2
	1987–88	1,567.0	95.6	1,931.0	99.9
	1988–89	2,080.0	98.5	2,991.0	100.0

passed through the lysimeters can be seen graphically in Figures 9 and 10. The throughput of precipitation is dependent on site conditions and lysimeter fill material. At ANL-E, an average of 998.0 ± 255.6 L or 47.3% of total precipitation passed through the soil lysimeters, while for the control, this value was 2,080.0 L or 98.5% of available precipitation. For ORNL, the values were 2,558.0 ± 28.5 L (85.5%) for the soil-filled lysimeters and 2,991.0 L (100%) for the control. These data are comparable year to year and reflect a high percentage of precipitation throughput. 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).

The total volumes of precipitation that have moved through the lysimeters represent an average 1.41 pore volumes for the ANL-E soil lysimeters and 3.60 pore volumes for soil lysimeters at ORNL, while the controls at ANL-E and ORNL were 3.54 and 5.34 pore volumes, respectively. Theoretically, then, by this time, all of the water held in pore spaces of the soil column in the ANL-E lysimeters has been replaced, while at ORNL, more than three times the original amount of water held in the soil porespace has passed through the soil lysimeters. 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 these differences in soil texture and to weather conditions (Figures 9 and 10).

Soil used at ANL-E is heavier (contains more fine material such as silts and swelling clay) than the soil used at ORNL.[14,15] 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 ANL-E, 98.5% of the volume of precipitation passed through that lysimeter versus 100% for the sand-filled lysimeter at ORNL. During the first year, 42% of the 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. (Wind speed and relative humidity for ANL-E and ORNL are shown in Appendix A.) Also as noted earlier, ANL-E lysimeters 1 and 2 have experienced water ponding during periods of heavy rainfall. To prevent loss of precipitation, some water was drained from the surface of these lysimeters in 1987 and was later replaced.

Therefore, if nuclides were in 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 the 12-month period. At each sampling, only 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 5 for cup placement) should be analyzed. Because of funding levels, however, it has not been possible to follow this protocol since the study was initiated. Rather, only water samples from number 3 cups are routinely analyzed at ANL-E, and number 1 and 3 cups at ORNL.

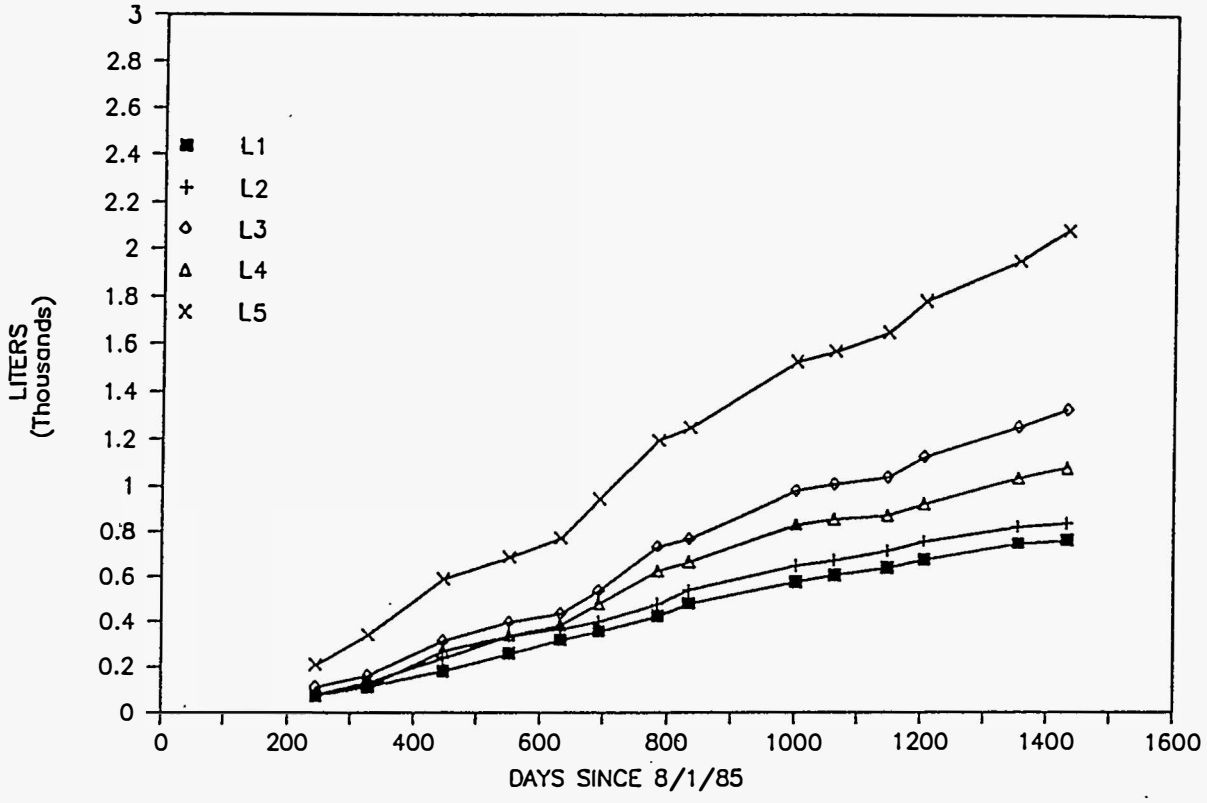

Figure 9. ANL-E cumulative volume of leachate from lysimeters.

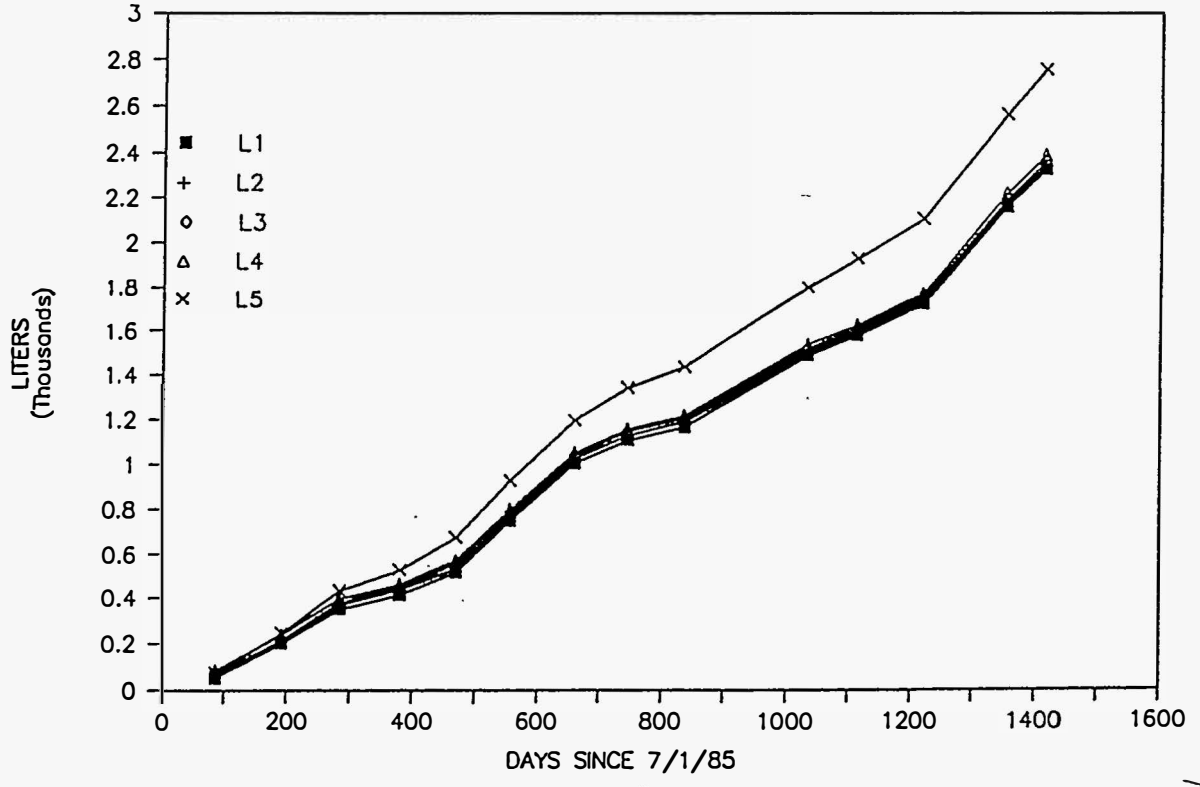

Figure 10. ORNL cumulative volume of leachate from lysimeters.

During June 1986, in addition to obtaining water samples from leachate collectors and moisture cup 3, water samples were taken from moisture cup 5 (the one nearest the soil surface) of each soil lysimeter. Those samples were then combined for use as a composite sample. Because moisture cup 5 is located above the waste forms, the composite water sample serves as a control to detect nuclides that might originate from sources other than the waste forms, such as incoming precipitation.

Tabulated results of beta and gamma analysis for the samples taken during the period are found in Tables 14 and 15 and in Tables E-1 through E-8 in Appendix E. Four samples were taken at each site during each 12-month period, except only two samples were taken in FY-86 at both sites and three samples were taken at ORNL in FY-88. The cumulative amounts of nuclides found in water samples obtained from lysimeter number 3 cups and leachate collectors for all sampling periods are displayed graphically in Figures 11 through 19.

As has been reported in the past,[6–9] not all nuclides are appearing consistently in either the water obtained from the cups or leachate collectors. The nuclide that appears with the most regularity at both sites is Sr-90 (Tables 14 and 15; Appendix E). This nuclide consistently occurs in significant amounts in all the number 3 cups at ANL-E and the number 3 cups at ORNL, though it was found in only small amounts in cup 4-3 during the last five samplings at ORNL, and the number 5 leachate collectors at both sites (Figures 11 through 14). There are standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a cumulative total of 270,090 pCi from 3-3 at ANL-E (Table 14 and Figure 11) and 17,285 pCi from 1-3 at ORNL as well as ORNL 2-3 and 3-3 (Table 15 and Figure 12). The releases into ANL 3-3 and ORNL 1-3 appear to be steady and almost linear.

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 total in the highest release cup, ANL 3-3, represents only about 0.001% of the waste form inventory (Table 16).

At ANL-E, Sr-90 retrieved from number 3 cups of the soil lysimeters during the fourth year ranges from 74% to 3,200% of that found in the leachate collectors (Table 14), while at ORNL, these values are between 0.1% and 70% (Table 15). 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.

During the first year, only the leachate-collector water from the control lysimeters and no water from the soil lysimeters at either site contained significant amounts of Sr-90 (Tables 14 and 15; Figures 13 and 14). This was comparable from year to year[6–9] (except in 1987) and is in sharp contrast to the number 3 cup data, which demonstrate that substantial amounts of Sr-90 are continuously being released from the waste forms in the soil lysimeters. In the fourth year, the percent of total Sr-90 measured in the leachate water and number 3 cups was somewhat inconsistent between the two sites (Table 16). This could indicate differences in waste form performance at the two sites. However, there was still a comparable percent of total Sr-90 in the leachate water of the control lysimeters for the two sites (Table 16). The general conclusion about Sr-90 movement into the leachate collectors is that the limiting factor is not the release of the nuclide from the waste forms; rather, it is the soil characteristics and possibly the amounts of water that have passed through each of the soil profiles (1.41 pour volumes at ANL-E and 3.60 at ORNL), which are dictating Sr-90 movement.

Gamma-producing nuclides were not found in the first two samplings in 1985. However, in April 1986, Co-60 was discovered in water samples from the moisture cup of ANL-3, Cs-137 was found in the leachate of ANL-5 (the sand-filled

Table 14. 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
1985–86	245	0	0	0	0.3	10	36.6	40.5	45	50.4	960.3	0	0
	328	0	0	1.6	0.3	22.8	36.6	40.5	45	50.4	11,125.3	0	0
1986--87	446	22.6	0	20.8	0.3	74.6	36.6	40.5	45	50.4	18,812.3	13	0
	551	22.6	2.2	1,370.8	0.3	169.6	3,937.6	40.5	—	—	—	931	0
	632	76.1	299.2	4,970.8	5.5	189.4	3,937.6	826.5	45	50.4	26,122.3	1,028	0
	692	210.1	436.2	12,890.8	5.5	418.4	4,969.6	1,617.5	7,665	50.4	52,451.3	1,100	2
1987–88	784	470.1	584.2	29,790.8	16.5	622.4	4,969.6	1,617.5	7,665	50.4	83,573.3	1,173	6
	833	884.1	743.2	44,990.8	30.5	908.4	4,969.6	1,617.5	7,665	50.4	88,820.3	1,205	13
	1,001	1,269.1	884.2	74,190.8	45.5	917.4	4,969.6	1,617.5	7,665	50.4	140,993.3	1,225	33
	1,062	1,737.1	1,026.2	100,190.8	159.5	1,420.4	4,969.6	1,617.5	7,665	50.4	146,970.3	1,256	75
1988–89	1,146	2,350.1	1,145.2	125,190.8	359.5	1,908.4	4,969.6	1,617.5	7,665	50.4	168,698.3	1,451	183
	1,204	2,421.1	1,345.2	165,190.8	492.5	2,264.4	4,969.6	1,617.5	7,665	50.4	220,362.3	1,580	428
	1,354	2,930.1	1,517.2	224,190.8	728.5	3,068.4	4,969.6	1,617.5	7,665	50.4	26,6619	1,642	685
	1,431	3,693.1	1,699.2	270,090.8	973.5	4,028.4	4,969.6	1,617.5	8,345	50.4	321,579.3	1,766	1,389

Table 15. ORNL 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 leachate collector (pCi)				
		1-3	2-3	3-3	4-3	5-3	1	2	3	4	5	1	2	3	4	5
	284	0	7	0	0	0	9,392	4,374	797	8,640	8,629	0	0	0	0	0
	380	0	7	3.2	0	1.8	9,980	4,374	797	9,405	8,629	0	0	0	0	0
1986–87	470	6.5	13.5	5.7	0.6	3.4	12,979	6,391	12,461	9,405	9,143	0	0	0	0	0
	556	36.2	40.5	10.3	0.6	3.4	12,979	6,391	12,974	9,405	9,143	0	0	0	0	0
	660	130.8	132.4	26.8	2.5	3.4	12,979	8,519	12,974	9,405	10,876	0	0	0	0	0
	744	284.8	278.4	67.3	2.5	3.4	22,077	9,201	13,793	9,865	17,577	0	0	0	0	0
1987–88	834	608.8	446.4	105.3	19.5	4.4	22,077	9,201	14,199	9,865	18,385	0	0	0	0	0
	1,033	1,743.8	743.4	321.3	19.5	7.4	22,077	9,201	14,199	9,865	22,801	0	0	0	0	0
	1,114	3,555.8	1,149.4	619.3	38.5	7.4	22,077	9,201	14,199	9,865	27,123	0	0	0	0	0
1988–89	1,220	6,555.8	1,659.4	1,399.3	38.5	14.4	22,077	9,201	14,199	9,865	32,723	0	0	1,898	1,204	2,626
	1,352	6,685.8	1,681.4	1,456.3	38.5	14.4	24,629	9,201	16,735	9,865	47,443	0	0	1,898	1,204	2,626
	1,414	10,785.8	2,411.4	1,556.3	41.5	28.4	24,629	9,201	16,735	9,865	54,128	0	0	1,898	1,204	74,824
	1,466	17,285	3,221.4	4,756.3	48.5	49.4	24,629	9,201	16,735	9,865	71,210	4,040	2,040	3,098	2,020	100,096

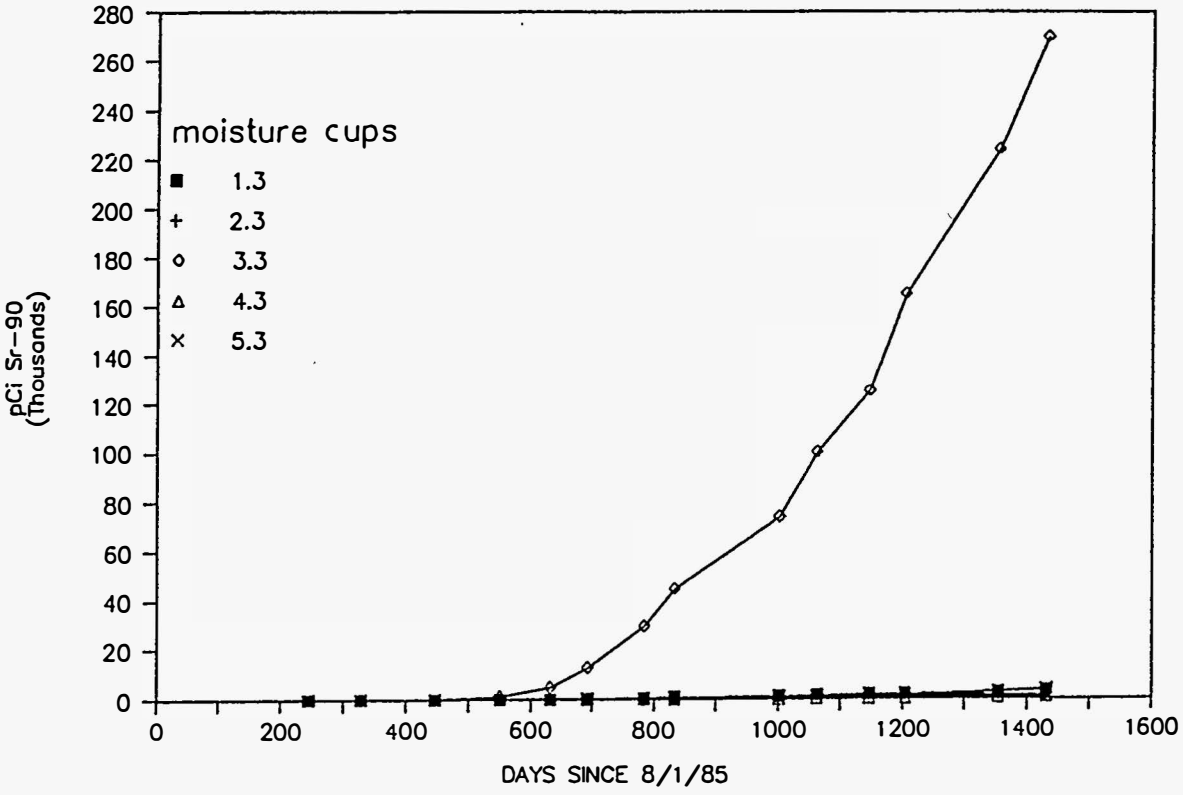

Figure 11. ANL-E cumulative Sr-90 collected in moisture cup number 3.

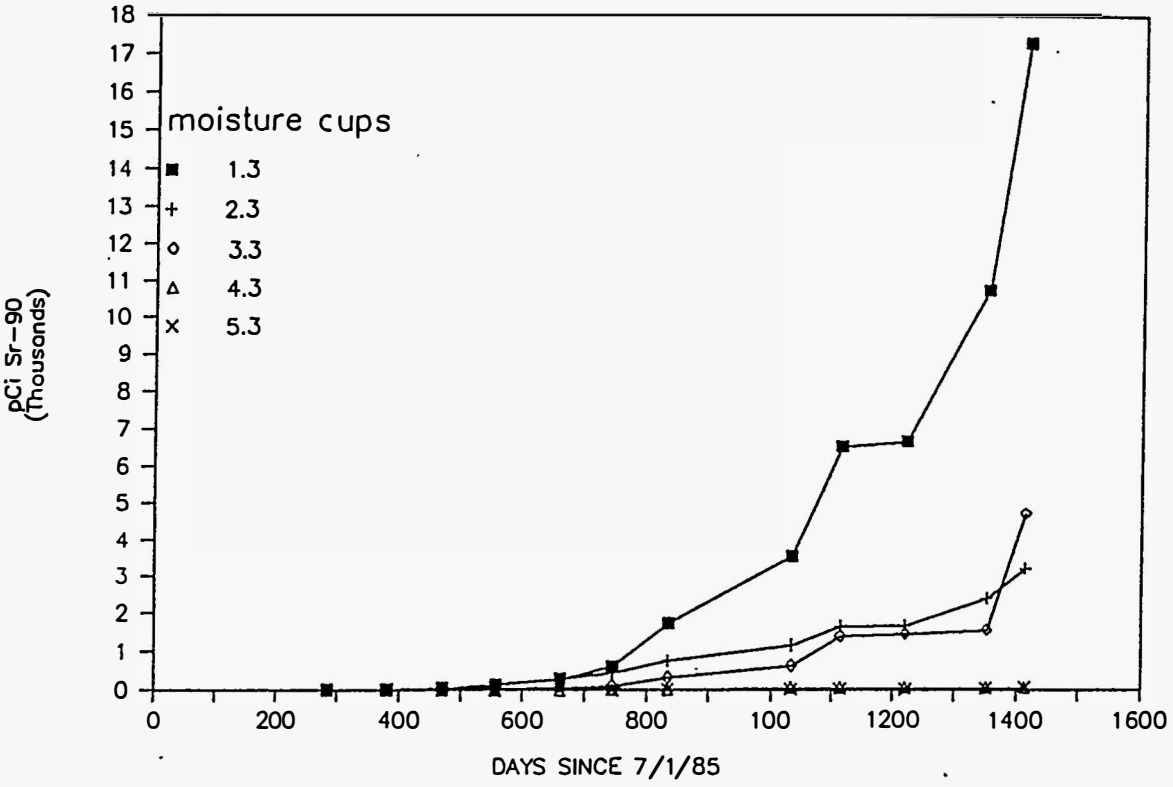

Figure 12. ORNL cumulative Sr-90 collected in moisture cup number 3.

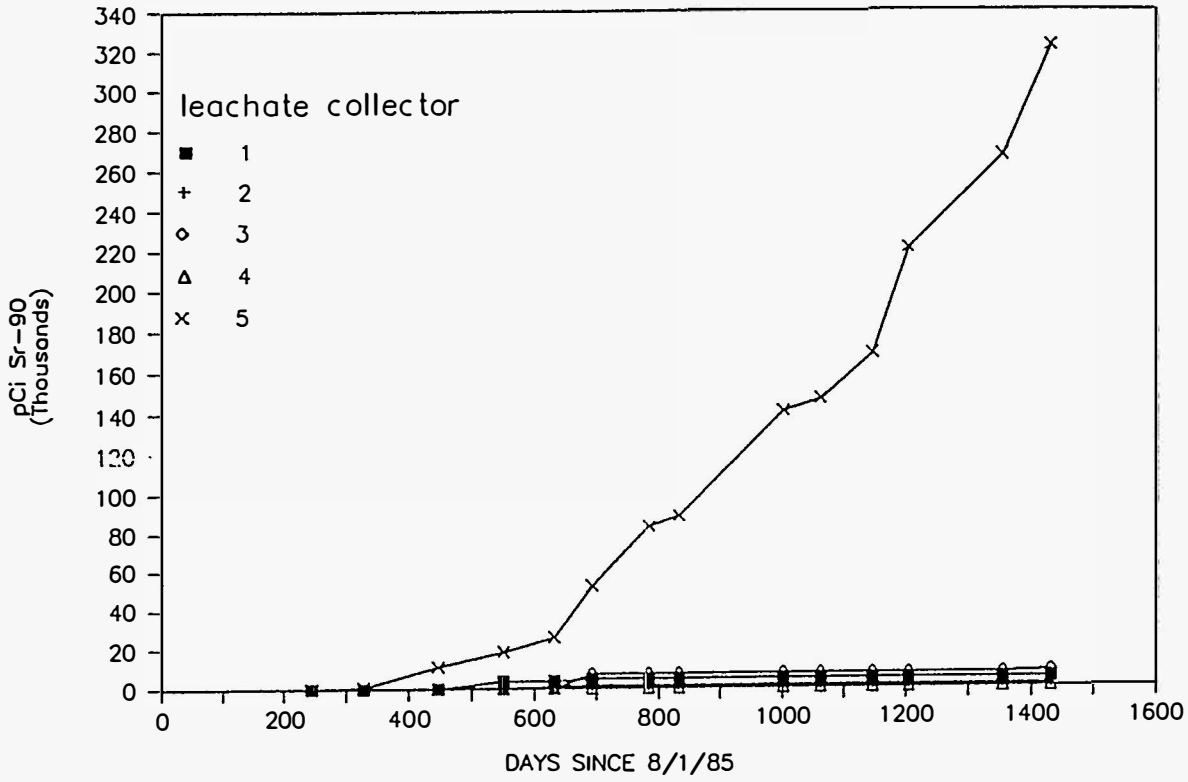

Figure 13. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

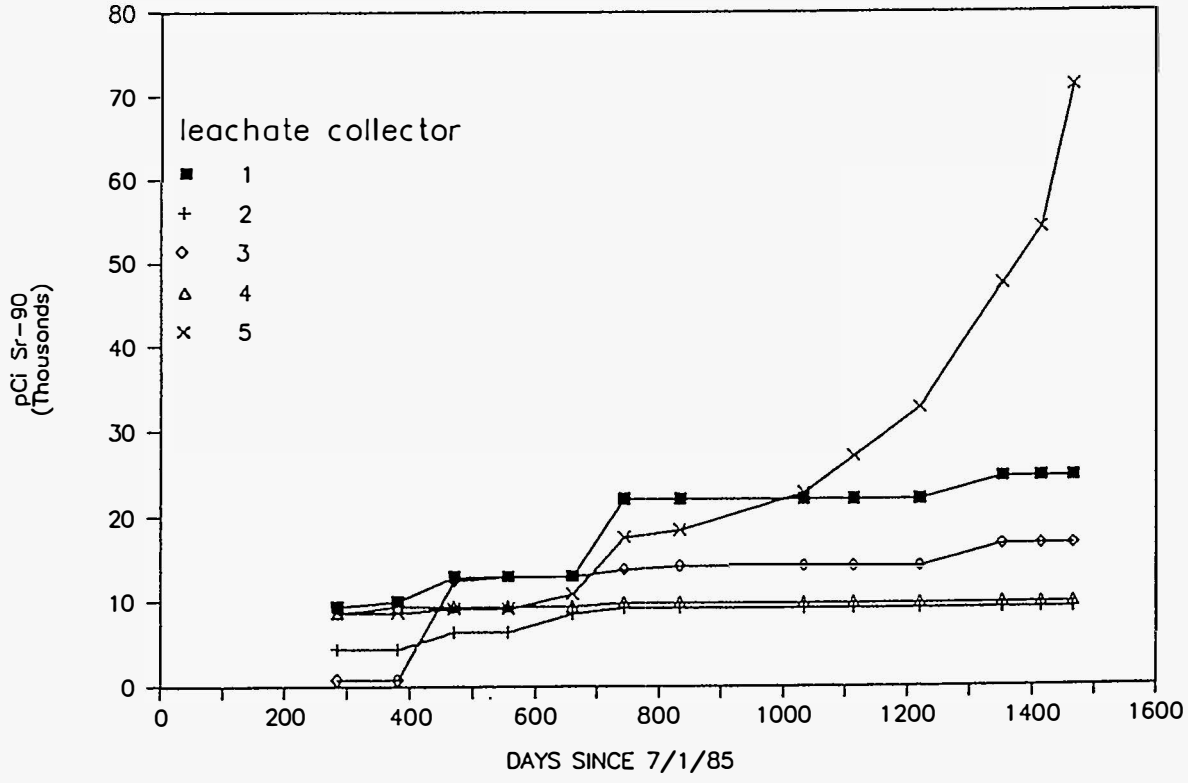

Figure 14. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

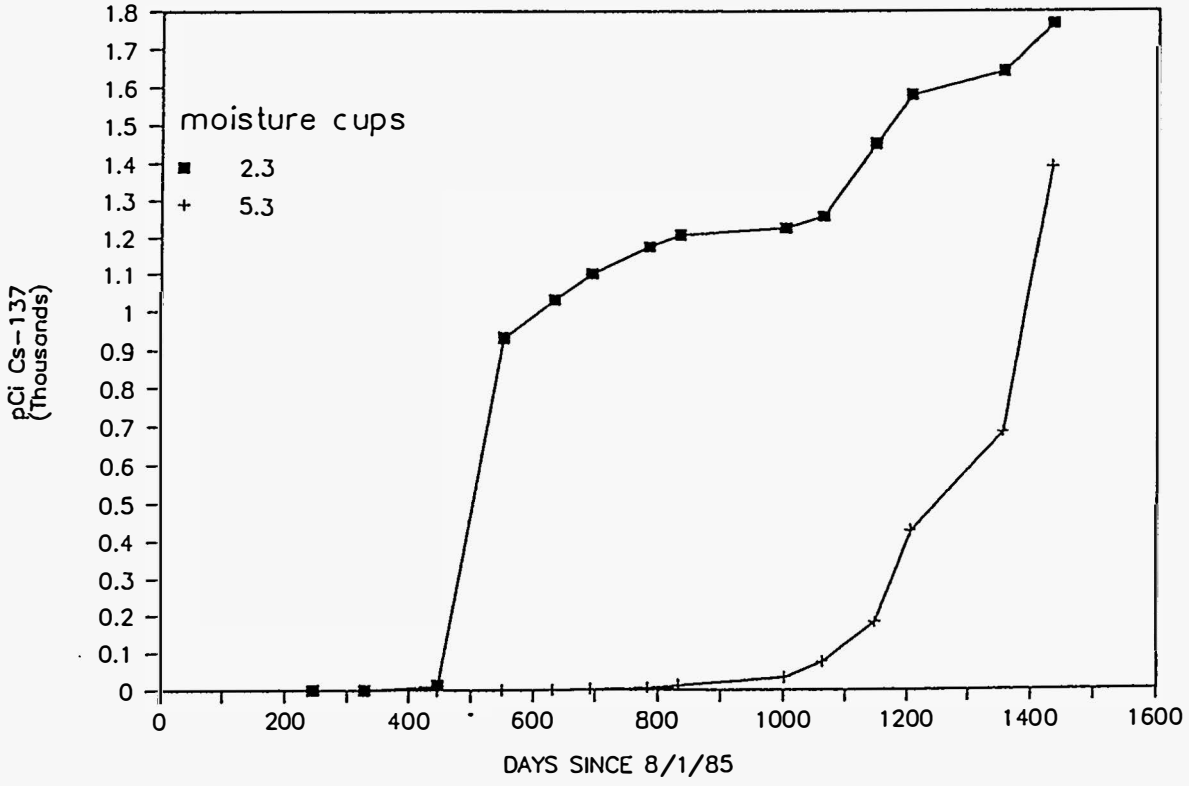

Figure 15. ANL-E cumulative Cs-137 collected in moisture cup number 3.

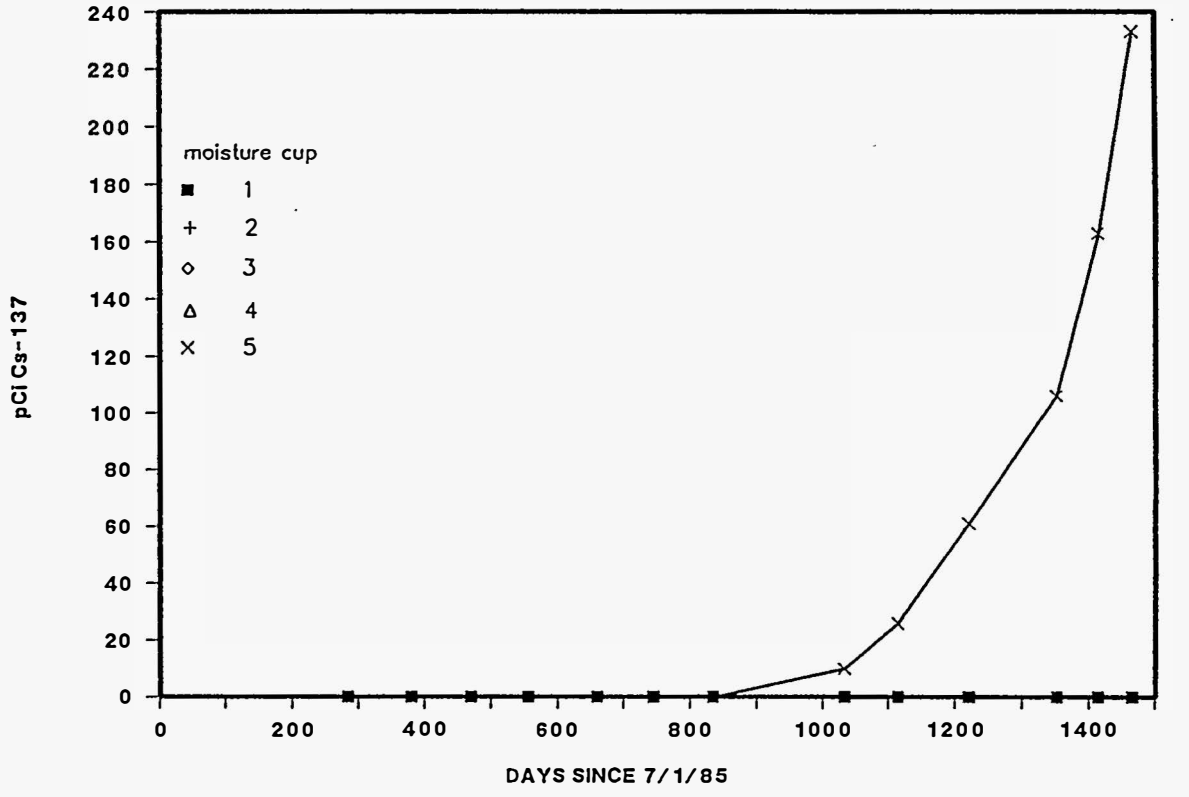

Figure 16. ORNL cumulative Cs-137 collected in moisture cup number 3.

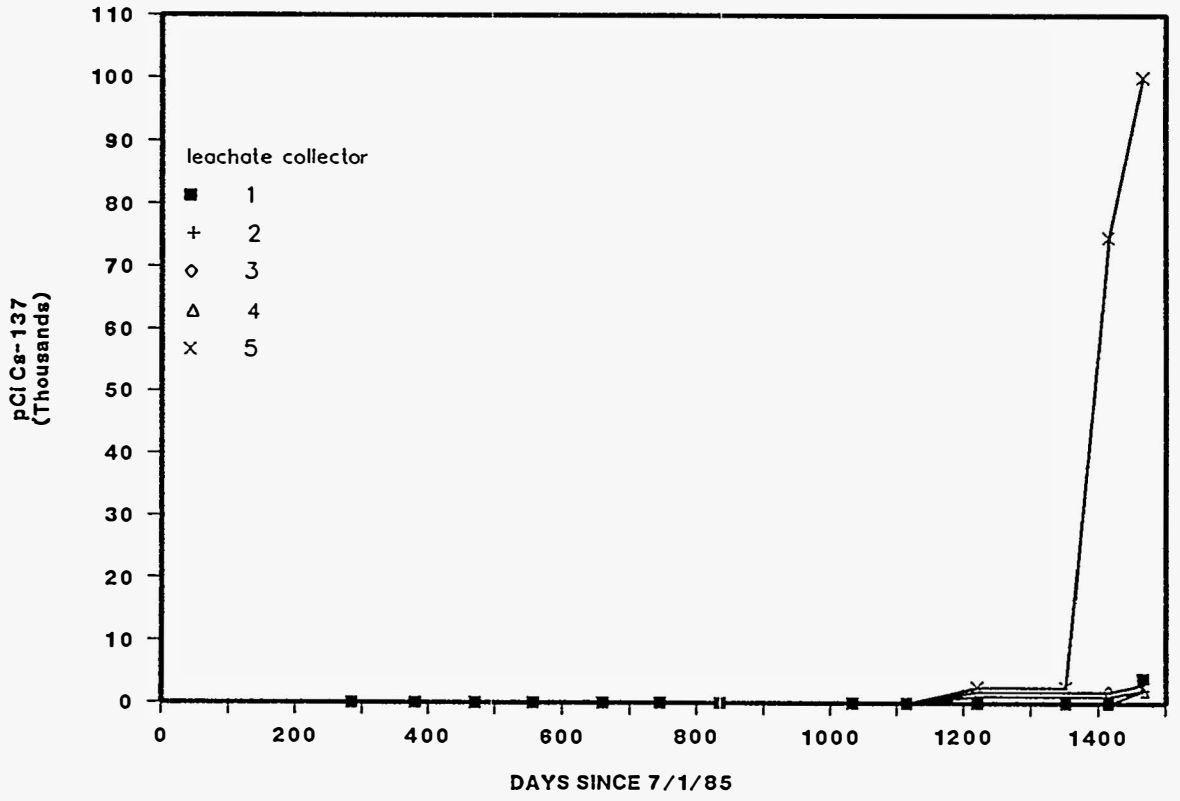

Figure 17. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

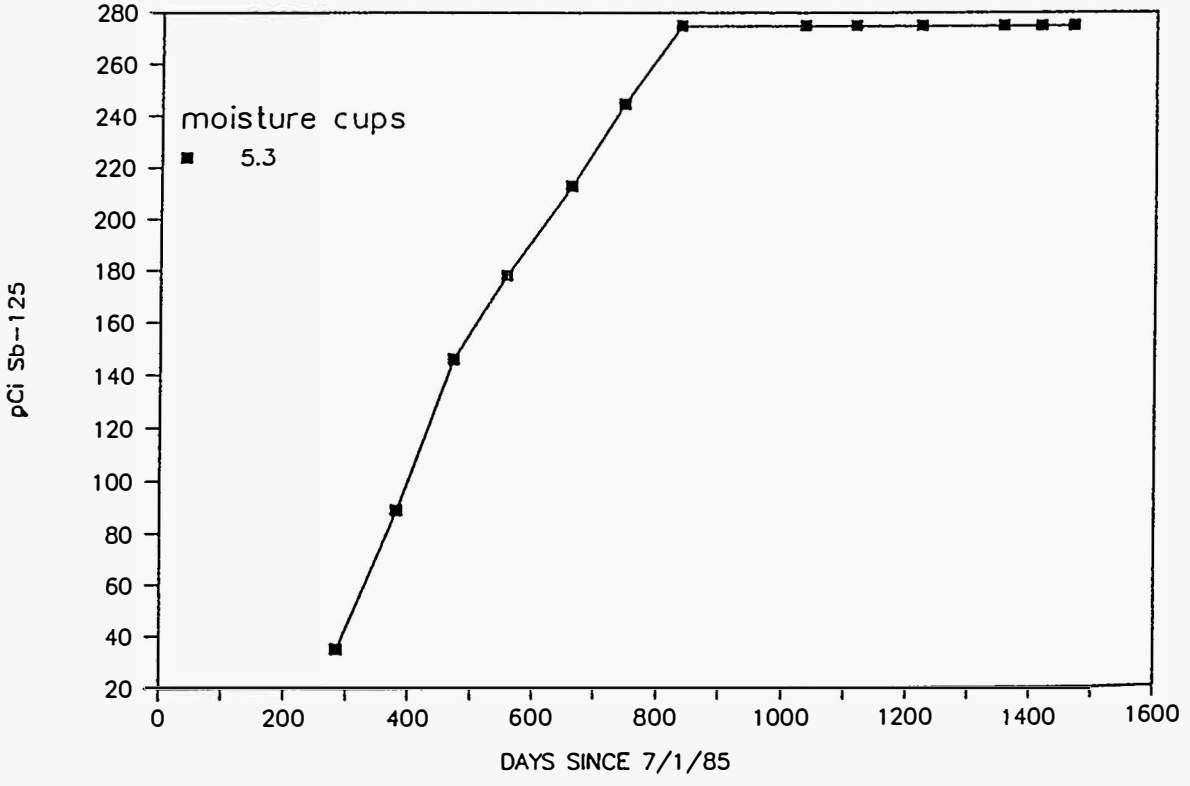

Figure 18. ORNL cumulative Sb-125 collected in moisture cup number 3.

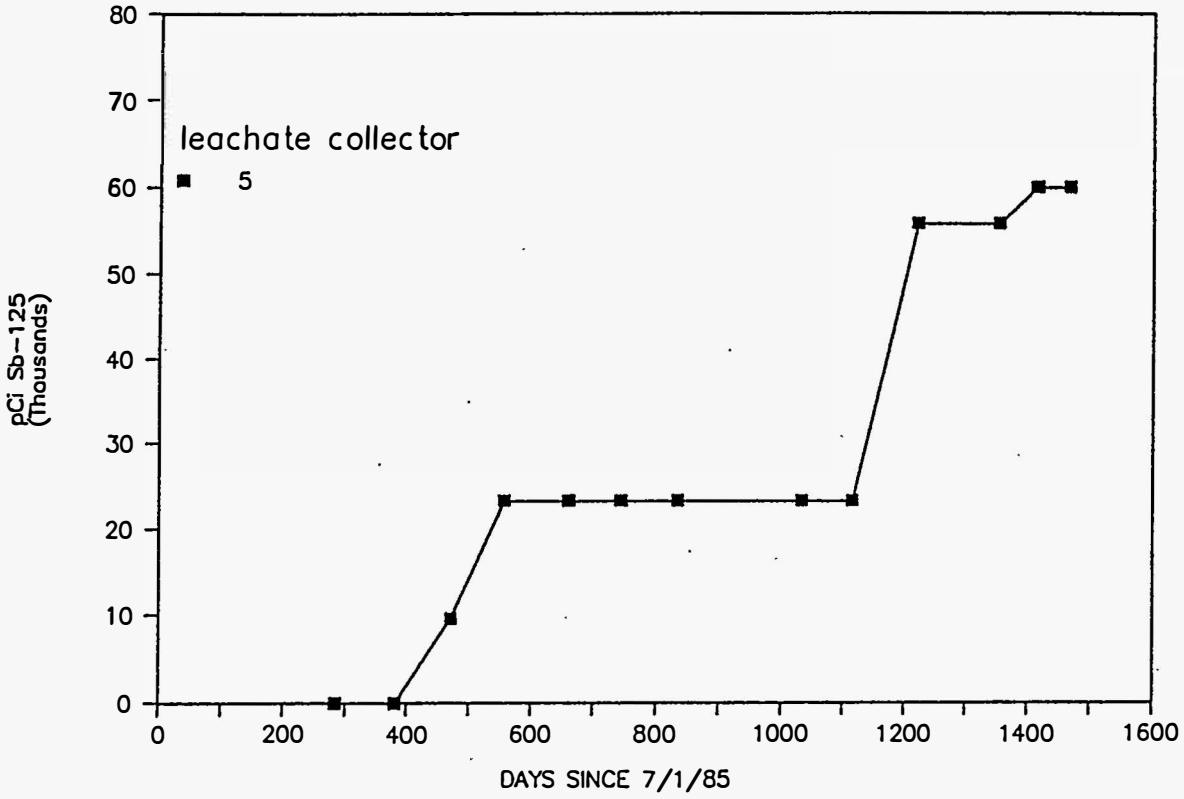

Figure 19. ORNL cumulative Sb-125 collected in lysimeter leachate collectors.

Table 16. Percent of total Sr-90 inventory per lysimeter extracted from moisture cups and leachate water through July 1989

Lysimeter number	Solidification agent	Total Sr-90 lysimeter (pCi)	Percent of total Sr-90	
			Moisture cups	Leachate water
ANL-1	Cement	18.5E+09	0.000020	0.000027
ANL-2	Cement	3.3E+09	0.000051	0.000049
ANL-3	VES	27.4E+09	0.000986	0.000030
ANL-4	VES	4.5E+09	0.000022	0.000001
ANL-5	Cement	18.5E+09	0.000022	0.001800
ORNL-1	Cement	18.5E+09	0.000094	0.000140
ORNL-2	Cement	3.3E+09	0.000096	0.000279
ORNL-3	VES	27.4E+09	0.000017	0.000060
ORNL-4	VES	4.5E+09	0.000001	0.000220
ORNL-5	Cement	3.3E+09	0.000001	0.002160

lysimeter), and Sb-125 was found in the moisture cup of ORNL-5 (also a sand lysimeter). The origin of Sb-125 is not known but is assumed to be the waste forms. Original evaluation of radionuclide content of the prefilters from which this resin was taken identified Sb-125 in quantities of 0.1% of total nuclide content, although resin analysis has not found any.

Analysis of June 1986 water samples showed that Co-60 still persisted in the moisture cup of ANL-3, with a substantial increase in Sb-125 in ORNL 5-3. Cobalt-60 was also found for the first time in June 1986 in ORNL 5-3.

Gamma-producing nuclides have since occurred with regularity at ANL-E and are present at ORNL. ANL 2-3, below a cement waste form containing large amounts of Cs-137, continues to receive Cs-137 (Table 14 and Figure 15). A significant increase in the quantities of this nuclide appeared in 1988–89 after initially peaking in the February 1987 sample (Figure 15). Since June 1987, Cs-137 has begun appearing in ANL 5-3 (Table 14). The quantity of this nuclide increased in each of the sampling periods during the year, with an abrupt increase during the last sampling period (Figure 15). There continues to be no sustained occurrence of Cs-137 in any of the ANL-E leachate water. In sharp contrast to 1987–88, Cs-137 has been detected consistently in water from the ORNL lysimeters in 1988–89. Measurable amounts of Cs-137 began to occur in ORNL 5-3 during the April 1988 sample and have continued in subsequent samplings for a total of 233 pCi (Table 15 and Figure 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; thus far, a total of 100,096 pCi has passed through the lysimeter (Table 15 and Figure 17). In addition, both ORNL-3 and -4 have had Cs-137 occur in leachate water. Thus far, the larger releases of Cs-137 have been from cement waste forms.

Although Sb-125 has not been found in water samples from ORNL-5 after October 1987, it again appeared in ORNL-5 leachate collector samples beginning with the November 1988 sampling (Table E-8 and Figure 19). It is now calculated that approximately 0.004% of the Sb-125 inventory from the ORNL-5 waste form has been recovered, with 0.003% of that occurring during the last year. The release curve for Sb-125 into cup ORNL 5-3 (Figure 18) appears to resemble the bench leach results for Sr-90 and Cs-137 (Reference 8), indicating that the limiting factor on movement of Sb-125 in this lysimeter could be release from the waste form.

From an intrasite comparison (Figures 13 and 14; Table 16), it is concluded that the VES waste forms (lysimeters 3 and 4; see Table 16) have released quantities of Sr-90 to the leachate comparable to those lysimeters containing cement waste forms (lysimeters 1 and 2) since any movement of Sr-90, or lack thereof, does not appear to correlate to either type of waste form. Data from the number 3 cups tends to support the evidence that VES is no better at retaining Sr-90 than cement (Figures 11 and 12; Table 16). Based on percent released to the number 3 cups, ANL-1 and -2 (cement) have received 2 to 5E-5% of the Sr-90, while ANL-3 and -4 (VES) have received 2 to 98E-5%. Comparable data at ORNL for the cement are 9.4 to 9.6E-5% and 0.1 to 1.7E-5% for VES. These data are only initial results, and as such only suggest what may be occurring.

Occurrence of nuclides in water samples from both the soil and inert-sand lysimeters in such a short period of time (months rather than years) was unexpected. While Sr-90 is known to be soluble in soil solution and does move through the soil column almost unhindered by the soil matrix, it appears that leaching and movement of the nuclides is occurring at a more accelerated rate in the soil than was thought possible.

The data for Cs-137 in ANL 2-3 and 5-3, and ORNL 5-3, as well as Sb-125 from ORNL-5 are of interest, but the continued lack of occurrence of these nuclides in the other lysimeters with the same type of waste forms makes it difficult to draw conclusions. The continued appearance of Cs-137 in ANL 2-3 and ORNL-5, as well as its reappearance in ANL 5-3, would indicate that this occurrence is not an artifact. These data, as well

as those for Sr-90, demonstrate the continued need for long-term field testing of the present waste forms.

Finally, because it is apparent that the soil in the lysimeters has subsided (very evident at ANL-E), it was decided to determine if the movement had caused a shift in the position of the waste forms. This was accomplished by lowering a radiation-detecting probe down the access tube that leads into the leachate holding tank. Readings were taken every 15.2 cm in all lysimeters, and radiation intensity with depth was recorded. Readings of the soil lysimeters were then compared with readings from the sand-filled controls. At ORNL, the intensity of radiation readings for each lysimeter approximated the known depth of the waste forms (Table 17). However, at ANL-E, some settling has occurred; readings in the soil-filled lysimeters (1–4) were still high at the 182.9-cm depth, while the activity in the inert control had moderated by that depth, indicating a downward movement of about 7.5 cm in the soil-filled lysimeters. There is no evidence that this movement has impacted the experiment except for minor damage to some moisture/temperature probes.

Field Versus Laboratory Results

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. A comparison of releases from those waste forms that were bench tested was presented in the FY-88 annual report of this project.[8] Total fractional release of the radionuclide Sr-90 from irradiated waste forms varied from 4 to 8% of inventory after being immersed for 90 days in deionized water. Comparing the maximum observed percent of total inventory of Sr-90 released from the lysimeter waste forms of about 0.002% (Table 16) with that from the leach tests indicates that the field testing experiment is still in the initial stages of developing releases of radionuclides. The percent of total inventory of gamma-emitting nuclides released in the lysimeters is too small to examine at this time (Tables 14 and 15 and Appendix E).

Major Cation and Anion Analysis

A clear understanding of the factors that influence movement of radionuclides through the lysimeter soils has not been developed. A preliminary effort was initiated at ORNL to analyze for some major cation and anion species in water samples obtained from the moisture cups. It is anticipated that such data could prove useful as a first indication of deterioration of waste form solidifying material, as well as an indication of 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 cause movement as a result of competition with radionuclides for the limited number of soil exchange sites (e.g., K^+ vs 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 1988 and 1989 was analyzed for the major ionic species listed in Table 18. The justification for the choice of ions is also provided in the table. In 1988, four water samples from each lysimeter (cups 1, 3, and 5, and a randomly chosen split of one of those samples) were analyzed, as were appropriated standards and precipitation samples. Cups 1 and 3 water samples were analyzed in 1989. Data from 1988 showed that ionic concentrations in the soil water were not introduced by the precipitation, which had very low ion content (Table 19). It appeared that the waste forms could have been an influencing factor either as the source of ions or

Table 17. Radiation intensity with depth in EPICOR-II field lysimeters.

Depth from soil surface (cm)	Radiation intensity (mR/h)									
	ANL-E lysimeter number					ORNL lysimeter number				
	1	2	3	4	5	1	2	3	4	5
15.2	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]
30.5	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]
45.7	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	0.02	0.04	0.01	0.04
61.0	—[a]	—[a]	—[a]	0.3	1.0	0.005	0.04	0.18	0.29	0.21
76.2	—[a]	—[a]	—[a]	1.0	3.0	0.16	0.24	0.65	1.6	1.4
91.4	0.4	—[a]	0.5	1.3	6.0	0.81	1.0	2.7	1.6	1.4
106.7	0.7	2.0	2.5	7.0	10.0	2.9	3.5	13.7	25.8	20.8
121.9[b]	3.5	18.0	5.0	35.0	12.0	6.1	11.2	29.0	52.4	40.3
137.2	6.0	28.0	20.0	43.0	12.0	11.2	16.1	39.5	70.2	48.3
152.4	7.5	39.0	18.0	67.0	12.0	11.2	17.7	40.3	70.9	36.3
167.6[b]	7.0	32.0	17.0	65.0	8.0	8.1	12.9	30.6	54.8	20.9
182.9	3.6	21.0	10.0	38.0	6.0	2.4	4.6	14.5	25.8	5.9
198.1	1.8	10.0	2.5	18.0	5.0	0.73	1.5	3.7	9.7	1.1
213.4	—[a]	—[a]	—[a]	—[a]	—[a]	0.16	0.31	0.89	1.4	0.16

a. Readings were not above background.

b. Location of waste form is indicated by sets of bars.

Table 18. Ionic species analyzed for ORNL 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.

possibly by causing replacement of ions from the surrounding soil (Table 19; Figures 20 and 21). The cement and VES waste forms performed similarly. With a few exceptions, the 1989 cation data (Table 20 and Figure 21) closely resemble those of 1988. However, the 1989 anion concentrations (Table 20 and Figure 21) were considerably decreased from 1988 and actually showed little of the cup-to-cup variability found in 1988. While these early data are interesting, no correlation can be made with nuclide movement as yet. Further analysis and data interpretation will depend on the collection of additional data.

Use of Lysimeter Data for Performance Assessment

It is becoming apparent, through operational experience and cumulative data provided by the NRC lysimeter array during the past 4 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) that directly relate to waste form stability. Information that can be obtained from the data includes the mass balance of released constituents, solubility of radionuclides in a site-specific geochemical system, as well as the retardation or dispersion of released constituents during transport to the far-field. Also, soil-pore water chemistry (inorganic and radioactive constituents), soil mineralogy, soil water/mineral mass ratio, net infiltration rate, soil profile moisture and temperature, porosity, hydraulic conductivity, and dispersiveness are being or could be extracted from 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 21. These parameters have been calculated using data collected during the 48-month operation of the ANL-E and ORNL lysimeters (Table 22). The data could be used in such codes at PATHRAE,[16] PRESTO,[17] and others to predict the stability of waste forms for a 300-year period of time.

Table 19. ORNL results of chemical speciation, lysimeter moisture cups 1, 3, and 5, July 1988.

		Cation					Anion			
Sample[a]	Waste form binder type	Ca^{+2} (mg/L)	Na^{+} (mg/L)	Si^{+4} (mg/L)	K^{+} (mg/L)	Mg^{+2} (mg/L)	Cl^{-} (mg/L)	NO_3^{-} (mg/L)	PO_4^{-3} (mg/L)	SO_4^{-2} (mg/L)
Lab Stk		10.1	1.01	—	1.68	2.06	3.08	31.2	—	8.14
Lab Std		9.74	0.92	0.009	1.59	1.78	3.33	31.9	0.015	7.03
Fld Std		9.44	0.95	0.010	1.58	1.76	3.13	33.7	0.006	7.19
Fld Std		9.92	0.99	0.020	1.61	1.80	3.13	37.8	<0.005	7.19
Lab Blk		0.04	<0.01	0.010	<0.01	<0.02	35.1	<0.10	0.015	0.10
Fld Blk		0.02	0.03	0.006	<0.01	<0.02	<0.05	7.53	<0.005	<0.05
Rain		0.20	0.1	0.018	0.49	0.10	1.115	1.48	0.126	2.71
Lys 1-1	Cement	75.5	19.1	22.5	0.15	3.10	1.04	5.54	1.677	142
Lys 1-3		59.0	22.9	23.6	0.46	3.24	0.99	9.18	0.806	135
Lys 1-3		60.5	23.2	23.5	0.47	3.27	1.25	8.72	1.800	129
Lys 1-5		41.6	1.46	19.3	0.07	1.38	0.63	7.47	1.539	33.7
Lys 2-1	Cement	49.0	11.6	21.4	0.15	1.82	0.91	16.4	1.634	43.5
Lys 2-3		34.2	18.6	32.3	0.80	1.48	1.16	8.82	2.824	26.9
Lys 2-5		3.88	1.20	8.40	0.07	0.62	0.86	7.30	0.742	5.81
Lys 2-5		3.92	1.23	8.40	0.07	0.64	0.86	7.32	0.745	5.76
Lys 3-1	VES	39.8	9.4	20.6	0.12	1.32	6.71	10.5	1.588	16.1
Lys 3-1		39.6	9.2	20.6	0.14	1.32	6.71	9.70	1.640	16.3
Lys 3-3		NS[b]	NS	NS	NS	NS	NS	NS	NS	NS
Lys 3-5		0.90	1.26	12.2	0.11	0.32	0.82	4.89	0.886	0.74
Lys 4-1	VES	8.56	10.3	11.8	0.12	1.24	11.5	12.8	0.978	12.7
Lys 4-3		2.74	9.20	14.9	0.14	0.62	9.38	24.4	1.076	9.27
Lys 4-5		1.20	1.38	12.2	0.10	0.28	0.56	6.58	1.033	1.448
Lys 5-1	Cement	10.5	1.00	15.5	1.15	3.84	0.82	29.3	1.383	6.60
Lys 5-3		13.3	9.10	40.3	4.98	4.80	1.83	3.64	3.759	11.4
Lys 5-5		15.1	0.52	25.4	1.48	4.44	1.78	29.9	1.876	18.9

a. Lab Stk = Standard stock solution made in the laboratory containing known concentration of Ca, K, Mg, Na, Cl, NO_3 and SO_4.
Lab Std = A sample of the LAB STK submitted for analysis.
Fld Std = A sample of the LAB STK run through the collection tubing at the lysimeter site on two different dates.
Lab Blk = A sample of laboratory-distilled water used to make LAB STK.
Fld Blk = A sample of laboratory-distilled water run through the collection tubing on the lysimeter site.

b. NS = No sample.

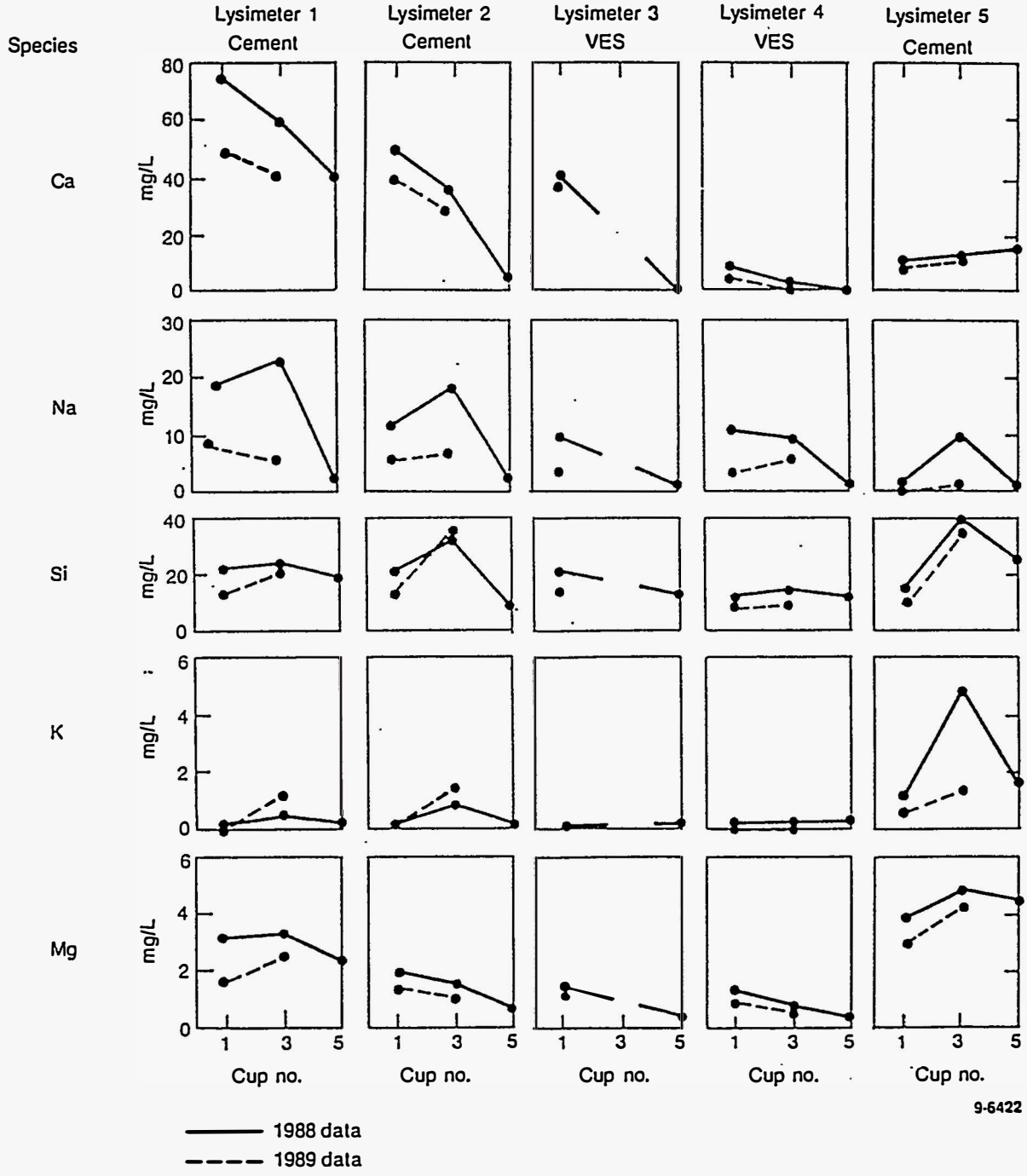

Figure 20. Results of chemical speciation at ORNL—cations.

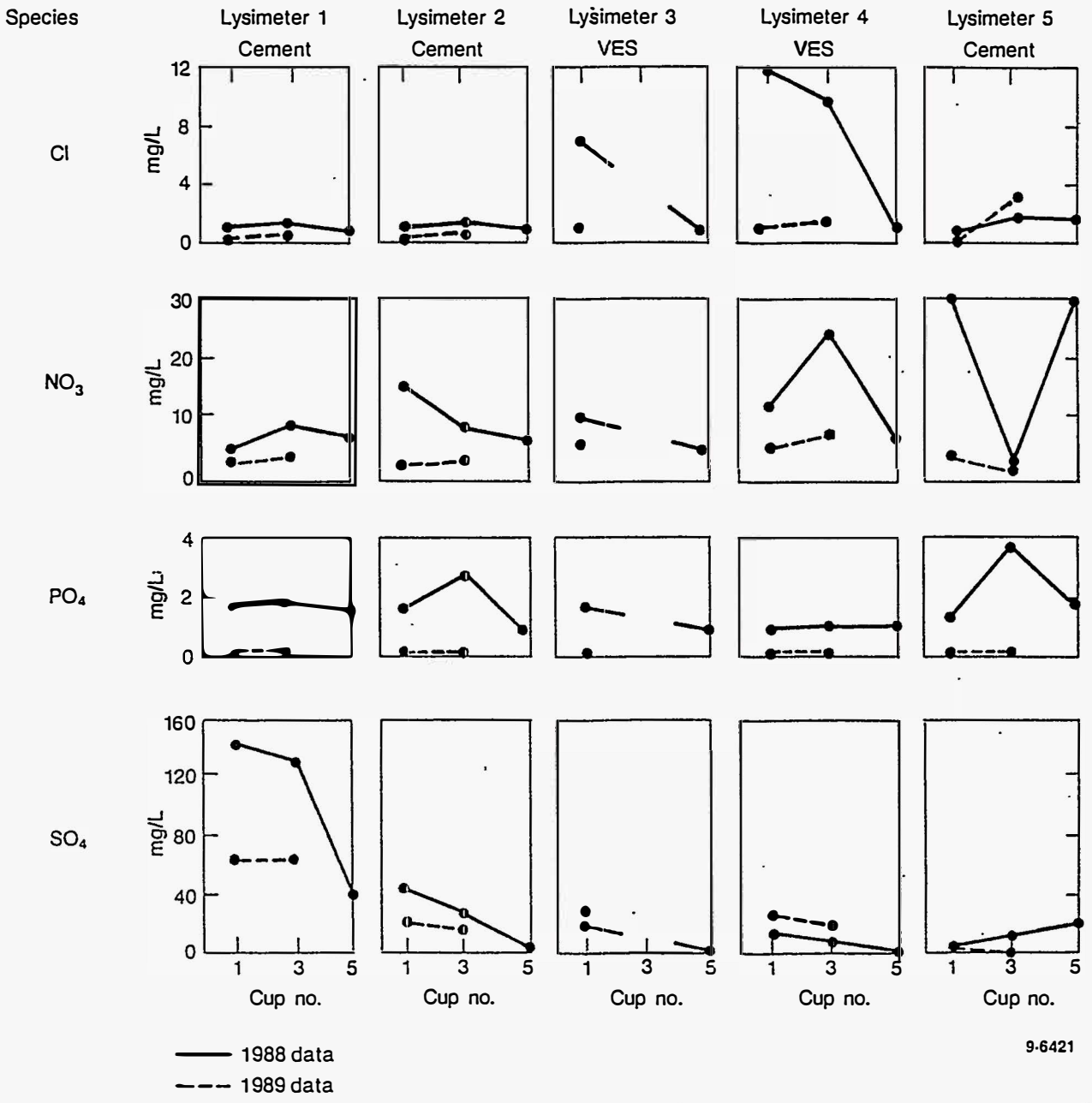

Figure 21. Results of chemical speciation at ORNL—anions.

Table 20. ORNL results of chemical speciation for lysimeter moisture cups 1 and 3, July 1989.

Sample	Waste form binder type	Cation					Anion			
		Ca^{+2} (mg/L)	Na^{+} (mg/L)	Si^{+4} (mg/L)	K^{+} (mg/L)	Mg^{+2} (mg/L)	Cl^{-} (mg/L)	NO_3^{-} (mg/L)	PO_4^{-3} (mg/L)	SO_4^{-2} (mg/L)
Lys 1-1	Cement	45	9.2	16	<0.1	1.9	0.35	3.74	<0.05	56.5
Lys 1-3		43	5.0	20	1.3	2.4	0.33	4.06	<0.05	53.3
Lys 2-1	Cement	38	6.1	17	<0.1	1.5	0.43	1.44	<0.05	16.8
Lys 2-3		30	6.7	33	1.2	1.2	0.66	2.26	<0.05	11.7
Lys 3-1	VES	37	2.4	16	<0.1	1.3	0.98	5.51	<0.05	20.9
Lys 3-3		NS[a]	NS	NS	NS	NS	NS	NS	NS	NS
Lys 4-1	VES	6.9	3.7	8.7	<0.1	0.98	0.81	5.84	<0.05	16.3
Lys 4-3		3.1	5.4	11	<0.1	0.61	0.92	6.14	<0.05	14.1
Lys 5-1	Cement	9.6	<2.0	10	0.5	3.2	0.79	3.21	<0.05	1.81
Lys 5-3		13	<2.0	39	1.6	4.5	3.54	3.23	0.16	3.93

a. NS = No sample.

Table 21. 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

Table 22. Performance assessment code parameters derived from ANL-E and ORNL data.

	ANL-E					ORNL				
Code parameters	1	2	3	4	5	1	2	3	4	5
Annual percolation (P) M/yr	0.297	0.346	0.520	0.422	0.817	0.969	0.977	0.983	0.995	1.148
Vertical water velocity (V_v) M/yr	1.14	1.33	2.00	1.62	3.89	5.21	5.25	5.28	5.35	5.74
Inventory (Q) pCi Sr-90	18.2E+09	3.3E+09	27.4E+09	4.5E+09	18.2E+09	18.2E+09	3.3E+09	27.4E+09	4.5E+09	3.3E+09
Fraction of saturation (S) (ave. of past 3 years)	56.4	56.5	56.4	56.4	50	37.2	37.2	37.2	37.2	50
Soil bulk density (d_s) G/cm^3	1.42	1.39	1.42	1.48	1.55	1.30	1.34	1.30	1.30	1.60
Effective soil porosity (P_s)	0.46	0.48	0.46	0.44	0.42	0.51	0.49	0.51	0.51	0.42
Inventory release (I_r) % Sr-90	27E-06	49E-06	29E-06	1E-06	1,500E-06	140E-06	279E-06	60E-06	220E-06	2,160E-06
Radionuclide concentration (C_w) ave pCi Sr-90/L leachate	6.6	1.9	5.8	0.1	128.2	10.6	3.9	7.1	4.1	25.8

CONCLUSIONS

Lysimeter operation during the fourth year at ANL-E and ORNL was successful. Analyses of data collected during the 48 months show a pattern in nuclide availability and movement such that the cumulative data could provide significant insight into waste form performance.

Strontium-90 is the most prevalent radionuclide in collected liquid samples. It appears that waste form performances are similar with respect to release of Sr-90 (except for a very high release from the VES waste form of ANL-3). It is also apparent that Sr-90 is able to move more freely through the Savannah River Laboratory (SRL) soil at ORNL. During the last 24 months of the period, Sr-90 continued to be found in leachate water in the control lysimeters at both sites. It appears then that the primary factor in receiving Sr-90 in the leachate is not the release of the nuclide from the waste forms (since Sr-90 is found in all number 3 cups directly below the waste form); rather, it is the soil characteristics (including soil and quantity of soil water) that limit movement. This conclusion is supported by data from lysimeter work at SRL and the Pacific Northwest Laboratory at Hanford.

SRL has found that Sr-90 will move from buried waste forms, migrate through the soil column, and appear in collected leachate water.[18] It is not surprising, then, that Sr-90 moves through soil in the ORNL lysimeters, since that soil originated at SRL.[10] On the other hand, lysimeter work with waste forms at Pacific Northwest Laboratory has shown that Sr-90 does not move in those soils.[19]

Data on waste form performance presented in this report continue to suggest that VES is comparable to cement in its ability to retain Sr-90. These data differ from those obtained at SRL. SRL data show that cement minimizes the release of Sr-90.[18] Both data reported herein and data reported by SRL and Hanford agree that Cs-137 is more readily released from cement than from VES.

During two consecutive sampling years at ORNL, no correlation was made between major ions present and the movement of radionuclides on ORNL lysimeters. Too few data have been obtained to allow for a conclusive statement.

It is fortunate that the NRC has such a data base at a time when the concept of site performance assessment of buried radioactive waste is being developed for implementation. It appears that the data generated from properly designed lysimeter arrays could be sold as a tool for performance assessment of solidified radioactive waste, thus assisting the NRC in its development of methods to verify the 300-year stability of waste forms. The preliminary conclusion that Sr-90 movement is soil-dependent indicates that using laboratory leach-test results in performance assessment does not represent actual conditions, which, in the context of waste form testing, is perhaps the most interesting and significant conclusion of this work. These results can serve the NRC as guidelines for developing new protocols for testing waste forms.

The data provided by these lysimeter experiments have been shown to be useful as input parameters for performance assessments codes. The utility of this reliable source of data will be demonstrated through continued operation of the lysimeters for a minimum of 20 years. This is thought to be the minimum time required to produce sufficient data for use in verifying the codes used to provide predictions on waste form stability for 300 years and beyond.

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Appendix A

Weather Data

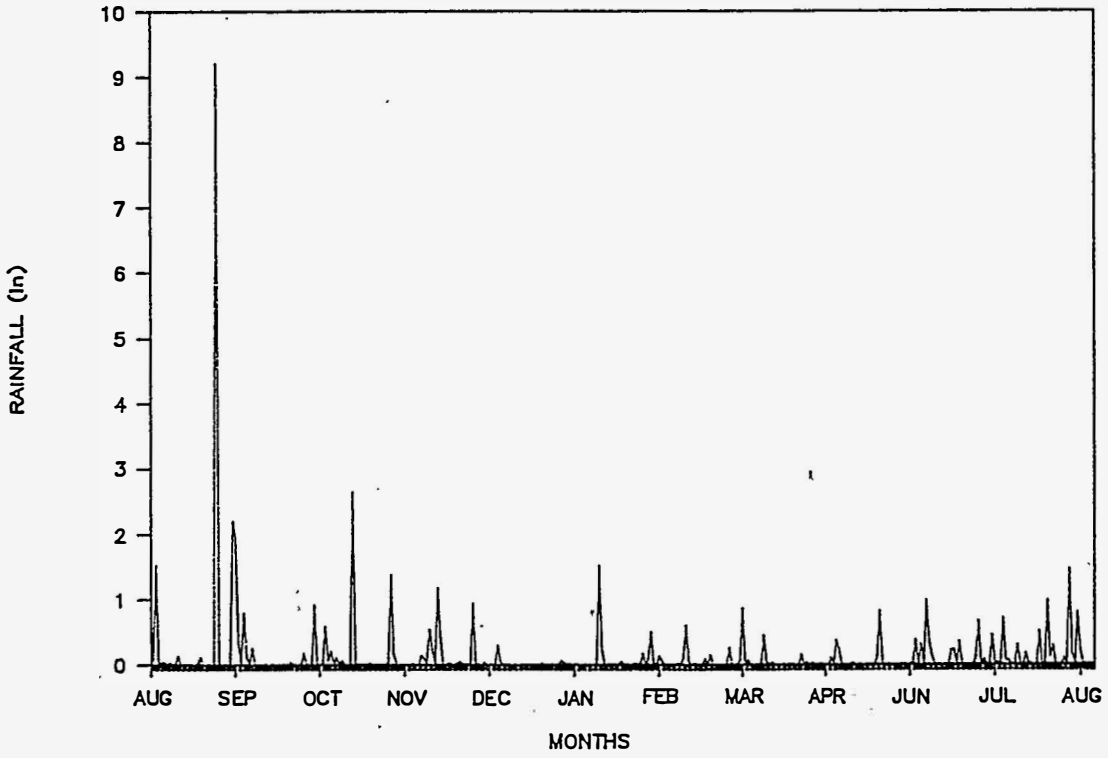

Figure A-1. ANL-E weather data for 1985-86—precipitation.

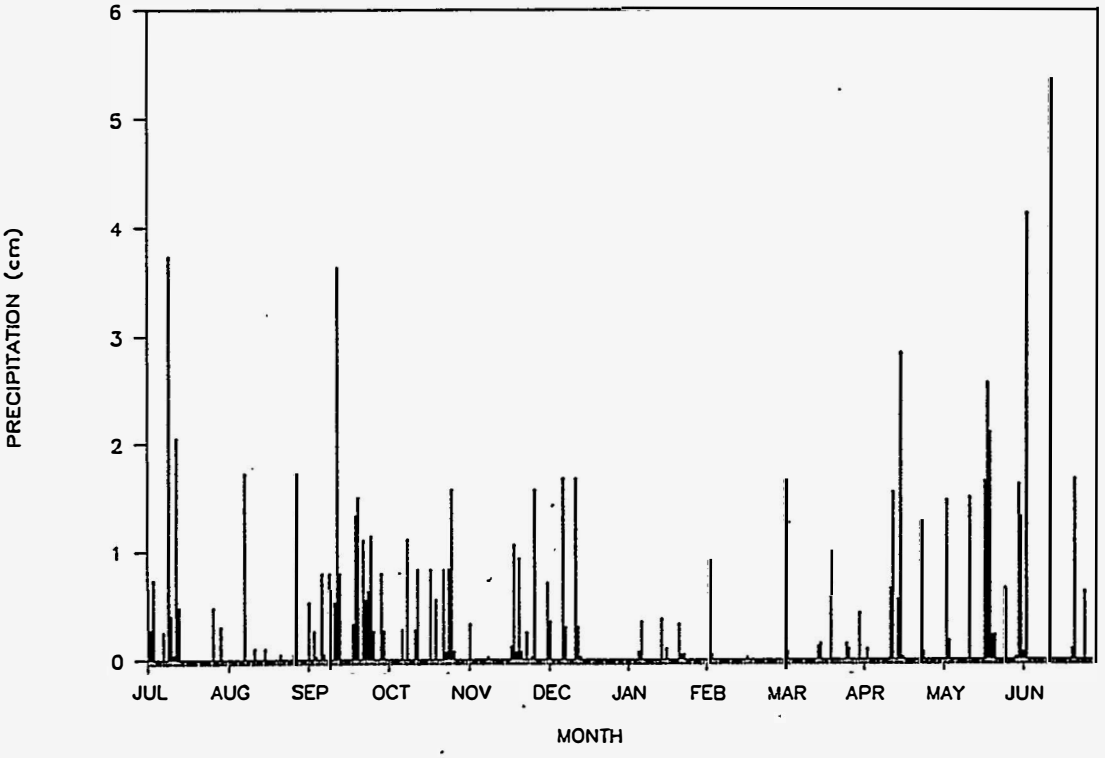

Figure A-2. ANL-E weather data for 1986-87—precipitation.

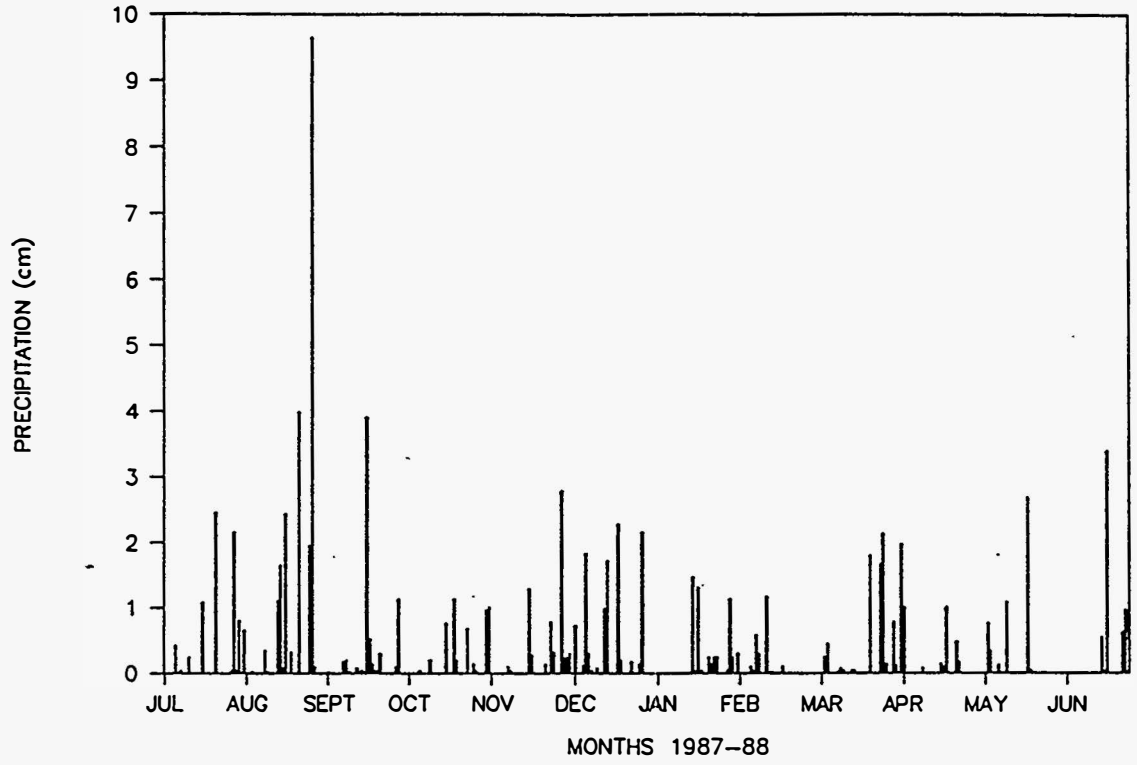

Figure A-3. ANL-E weather data for 1987-88—precipitation.

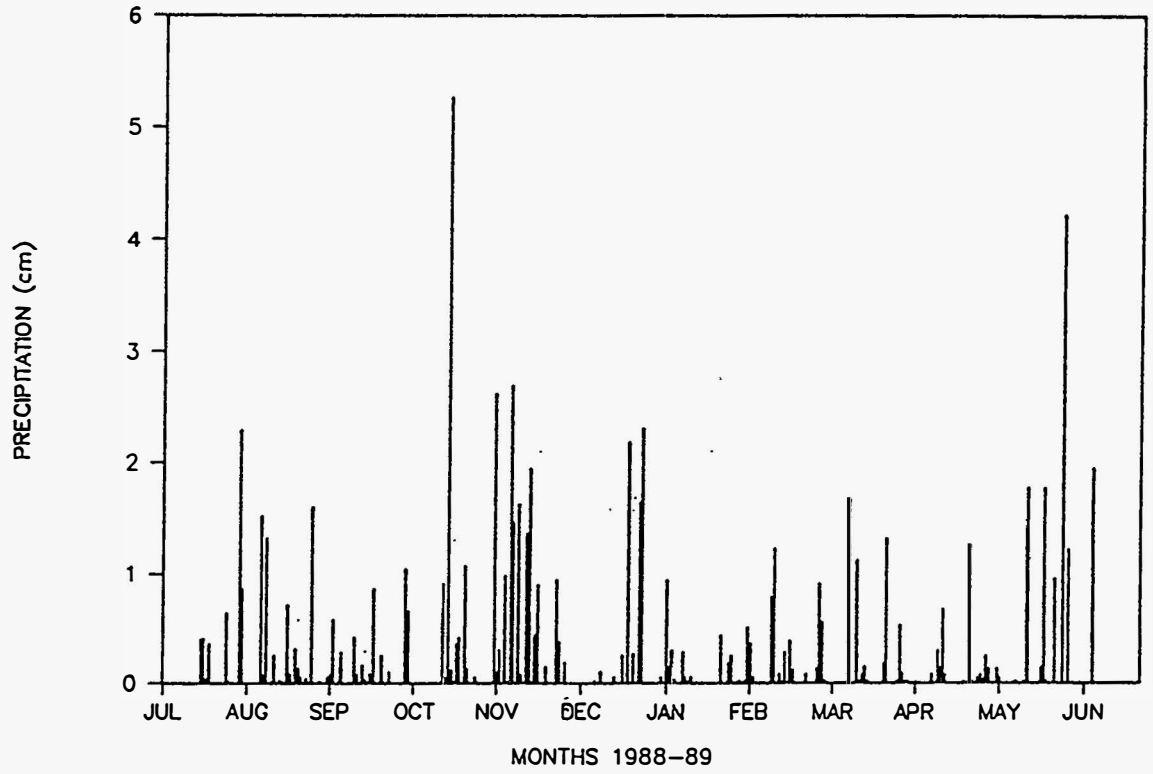

Figure A-4. ANL-E weather data for 1988-89—precipitation.

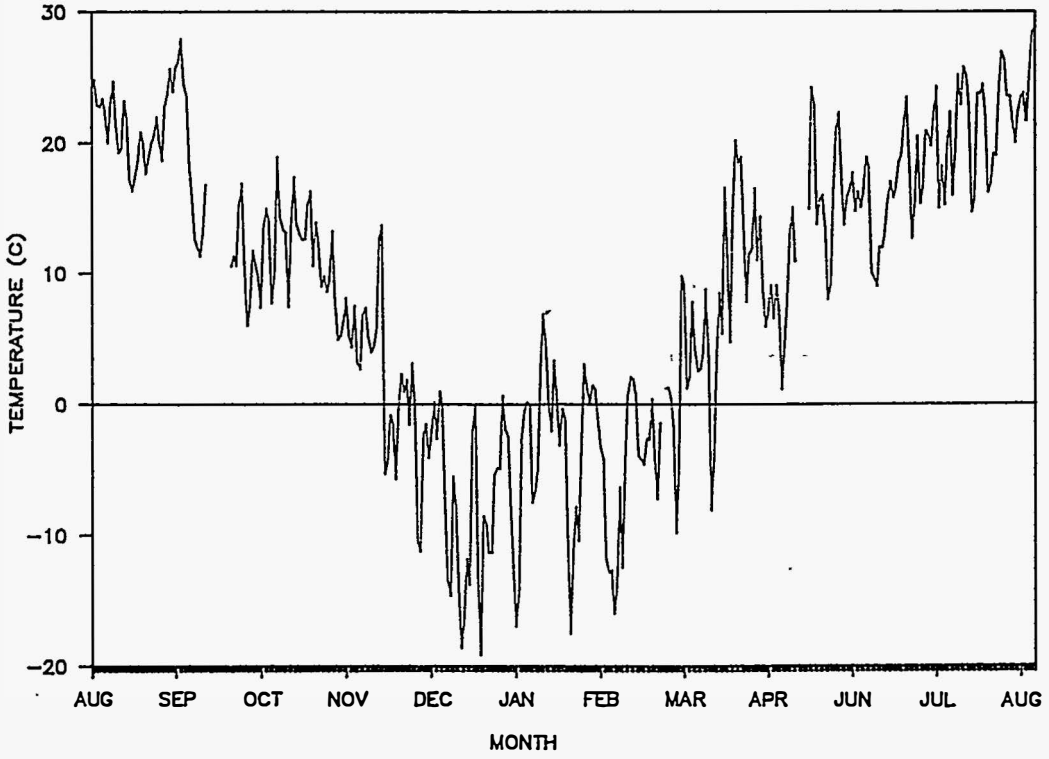

Figure A-5. ANL-E weather data for 1985-86—air temperature

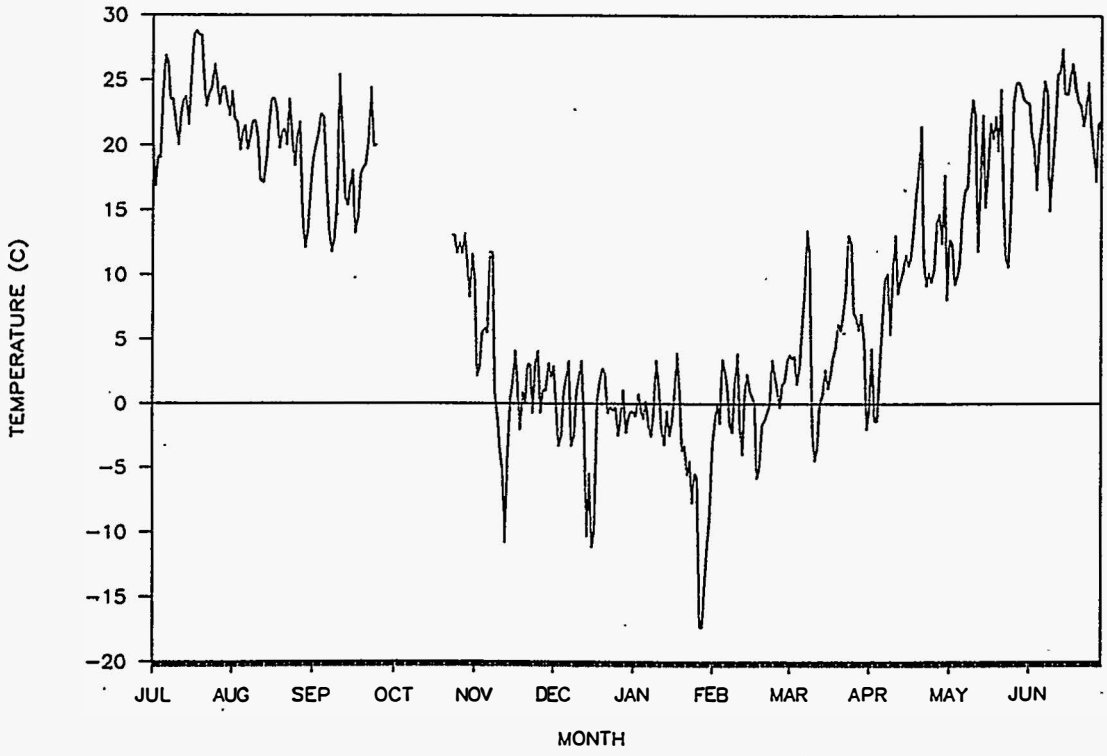

Figure A-6. ANL-E weather data for 1986-87—air temperature.

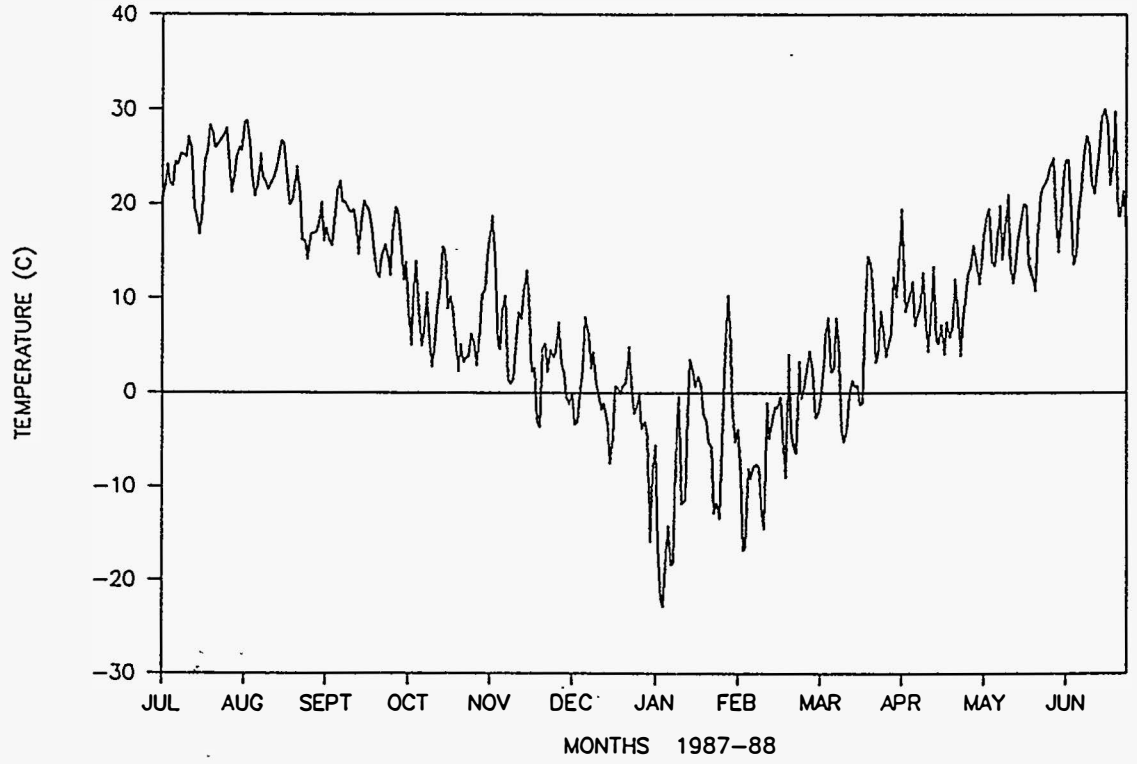

Figure A-7. ANL-E weather data for 1987-88—air temperature.

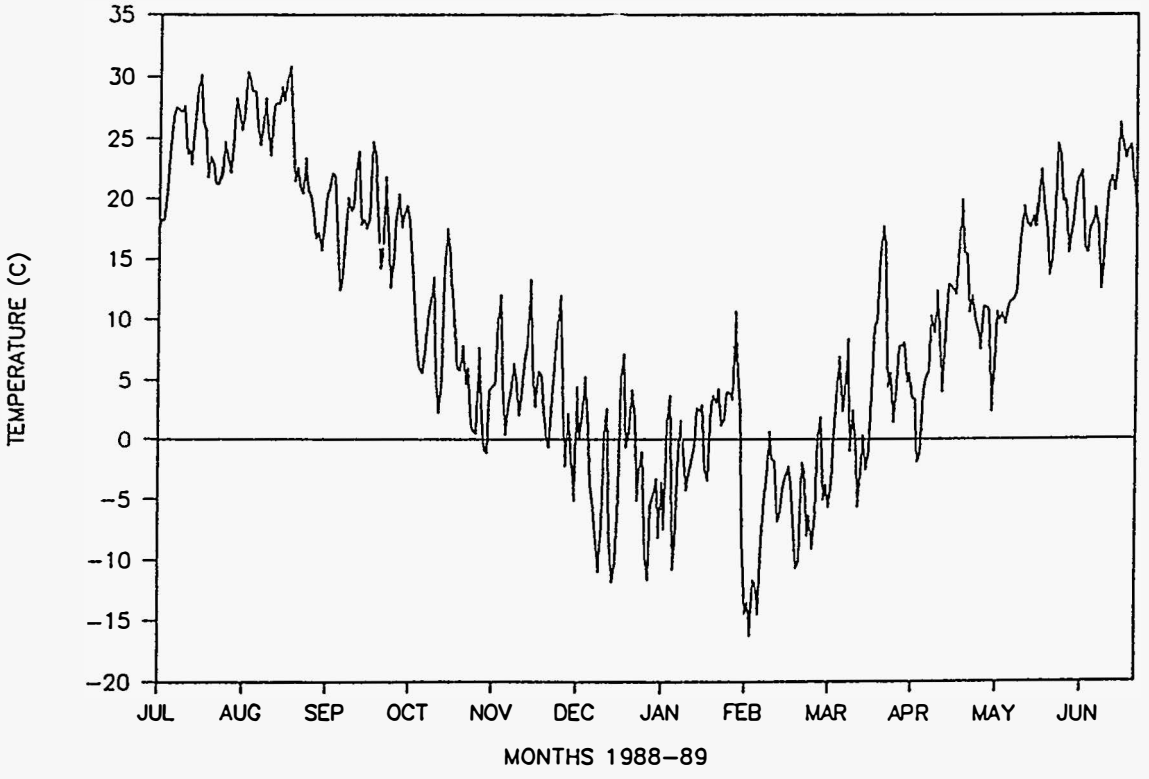

Figure A-8. ANL-E weather data for 1988-89—air temperature.

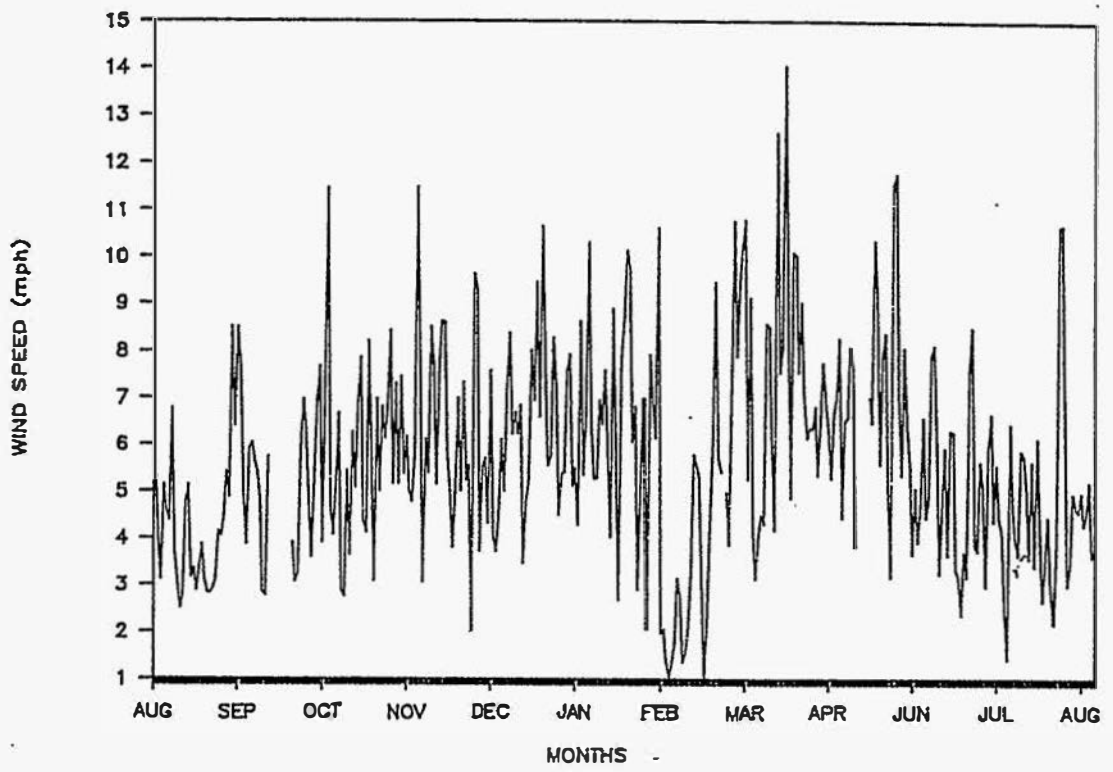

Figure A-9. ANL-E weather data for 1985-86—wind speed.

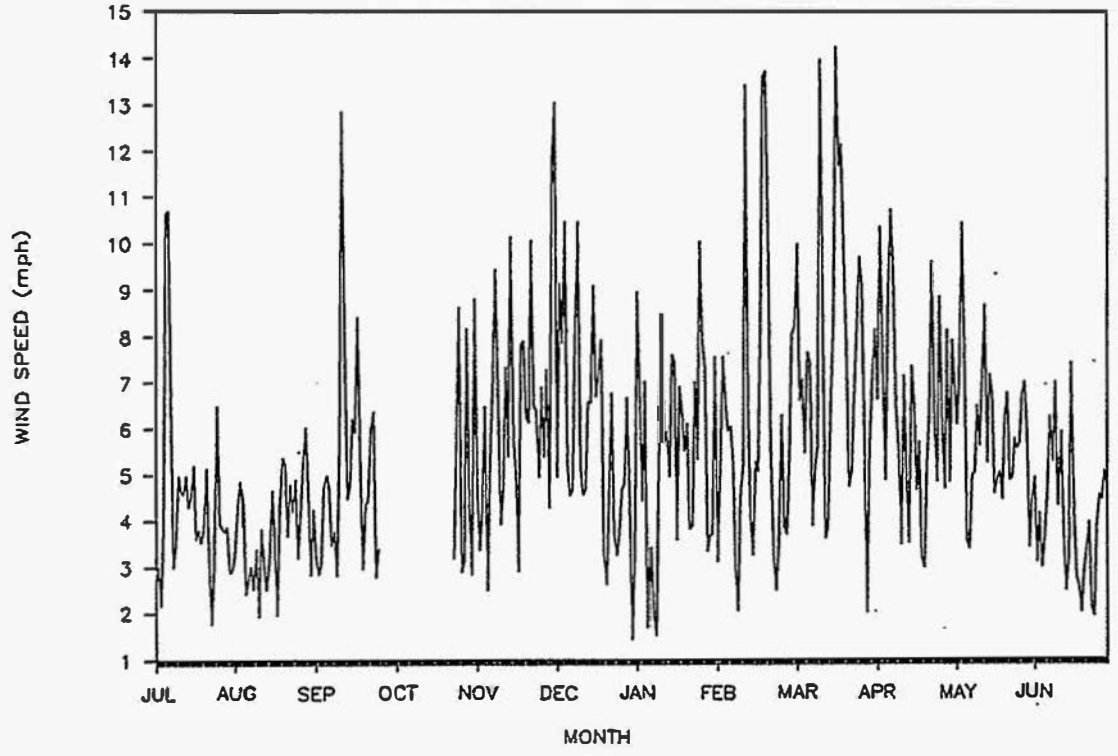

Figure A-10. ANL-E weather data for 1986-87—wind speed.

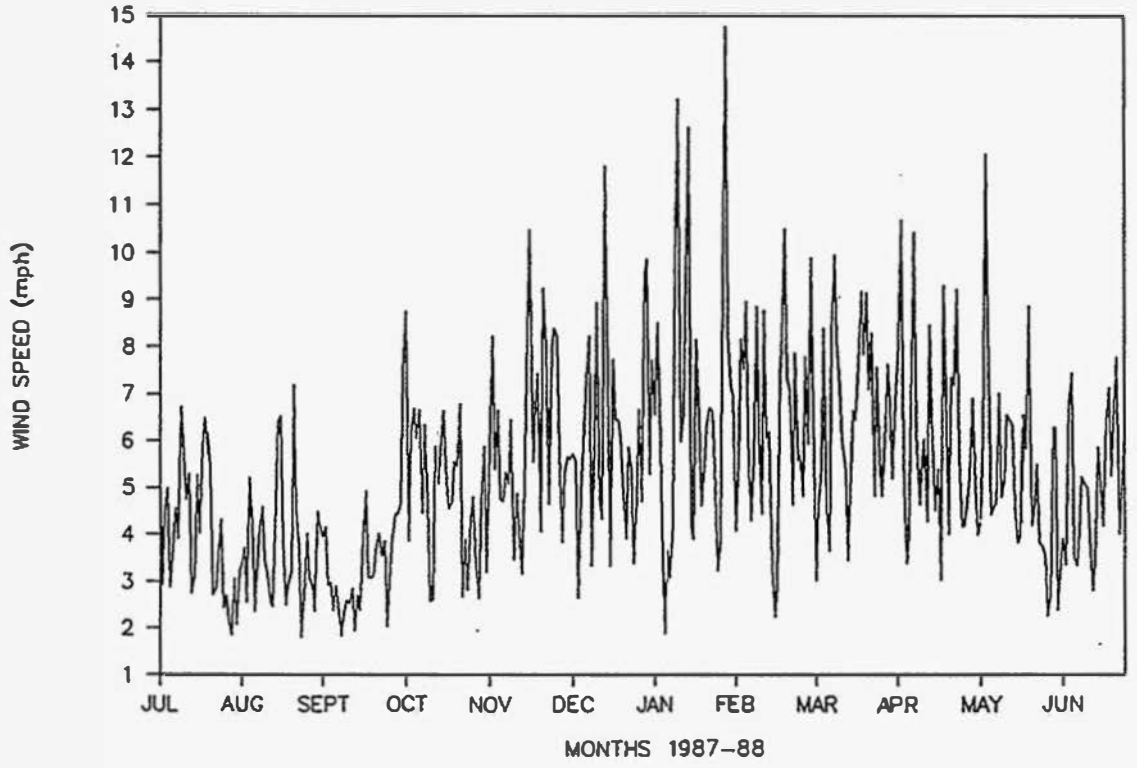

Figure A-11. ANL-E weather data for 1987-88—wind speed.

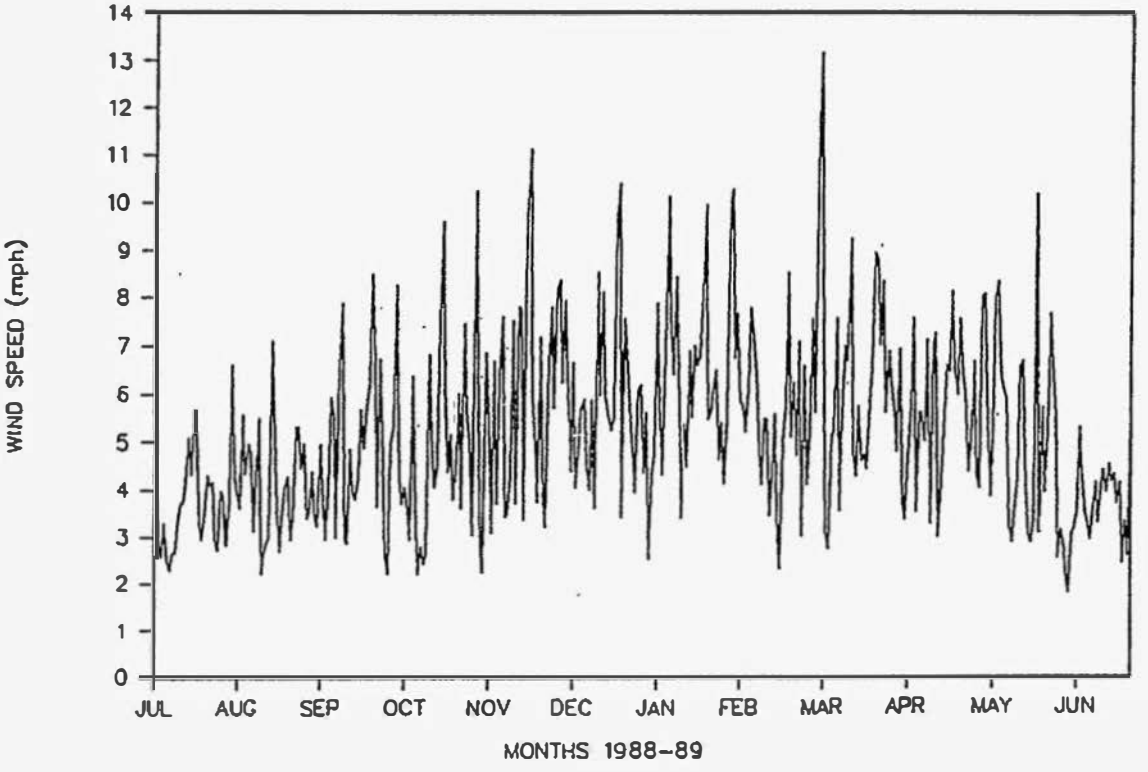

Figure A-12. ANL-E weather data for 1988-89—wind speed.

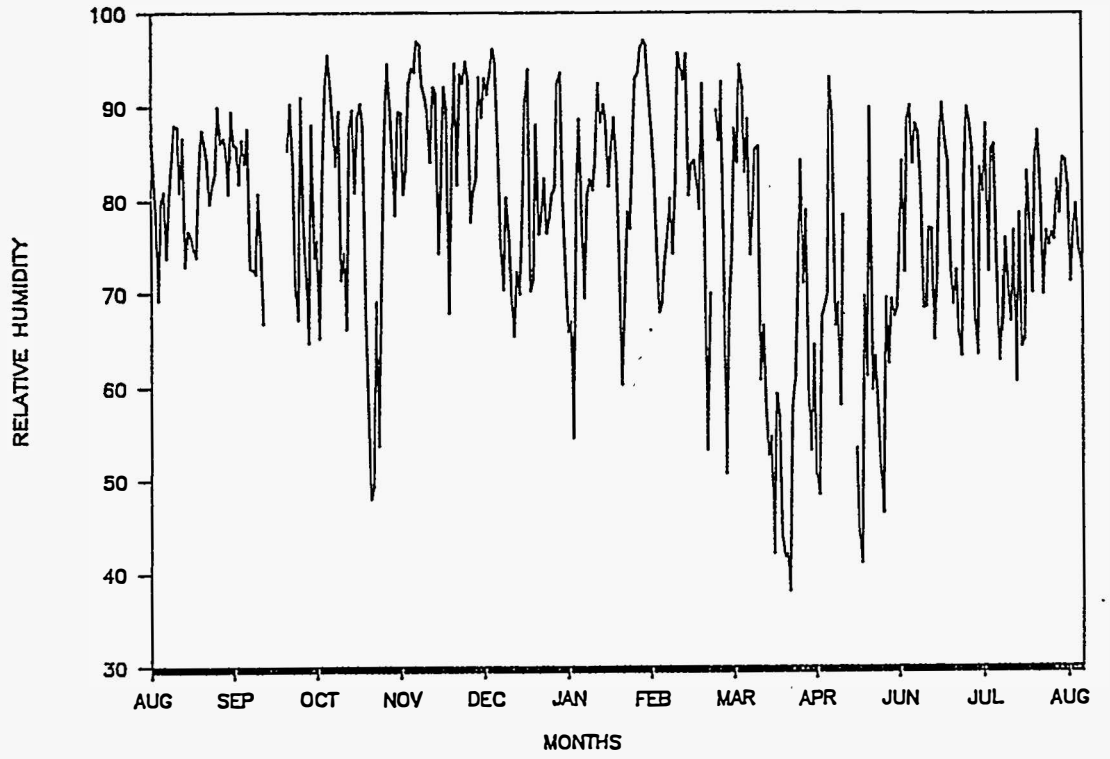

Figure A-13. ANL-E weather data for 1985-86—relative humidity

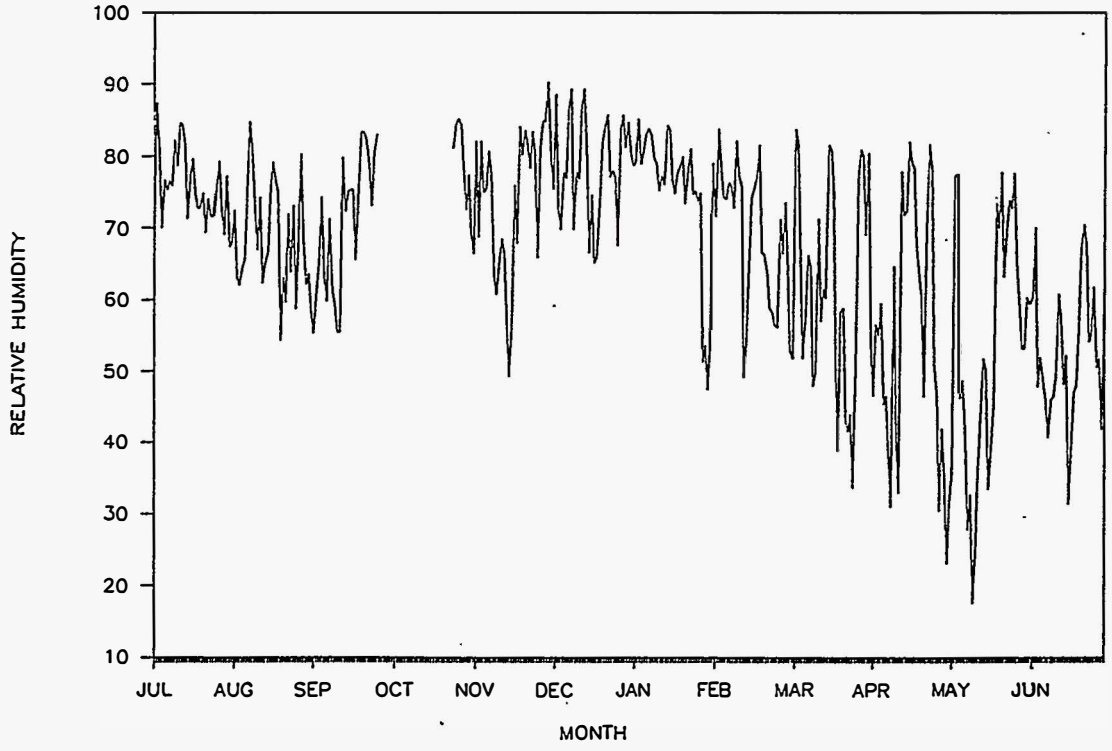

Figure A-14. ANL-E weather data for 1986-87—relative humidity.

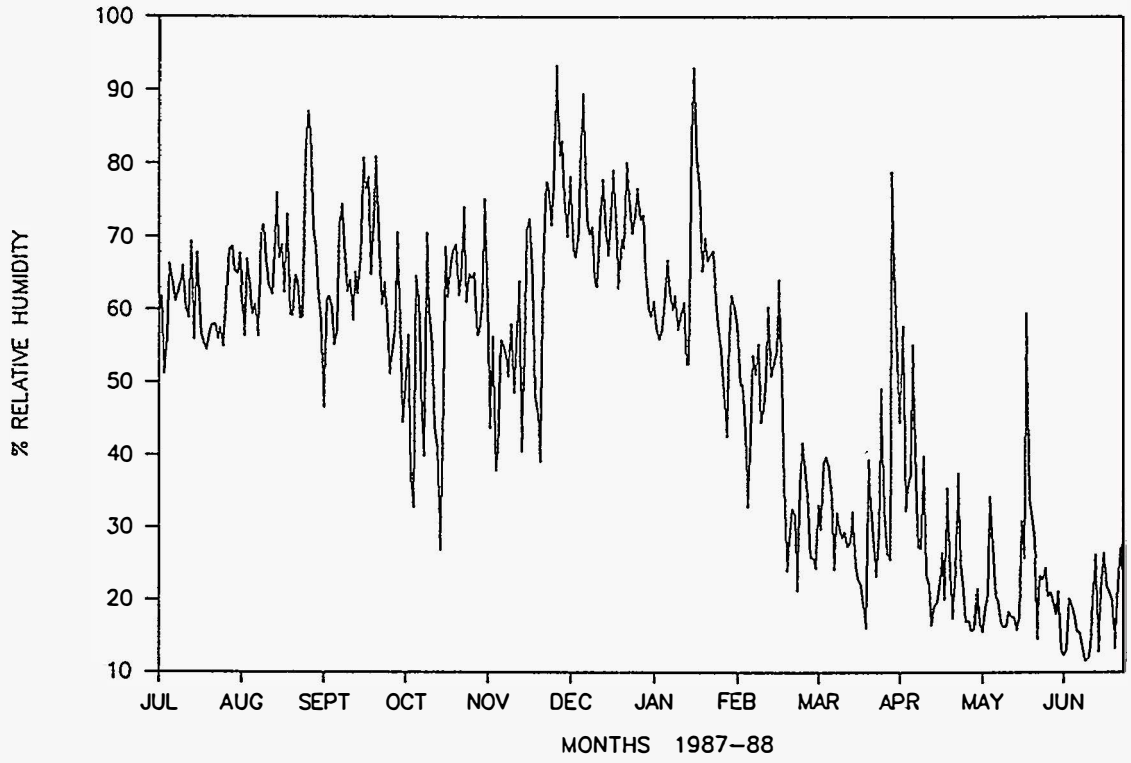

Figure A-15. ANL-E weather data for 1987-88—relative humidity.

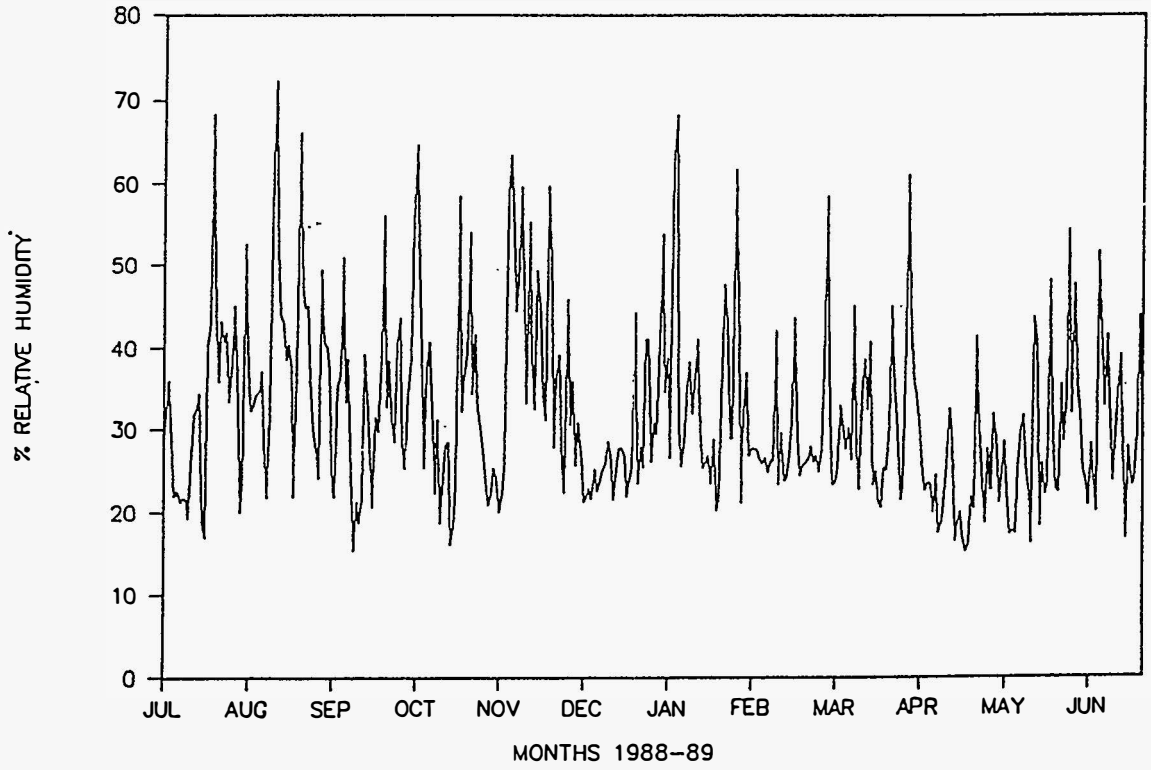

Figure A-16. ANL-E weather data for 1988-89—relative humidity.

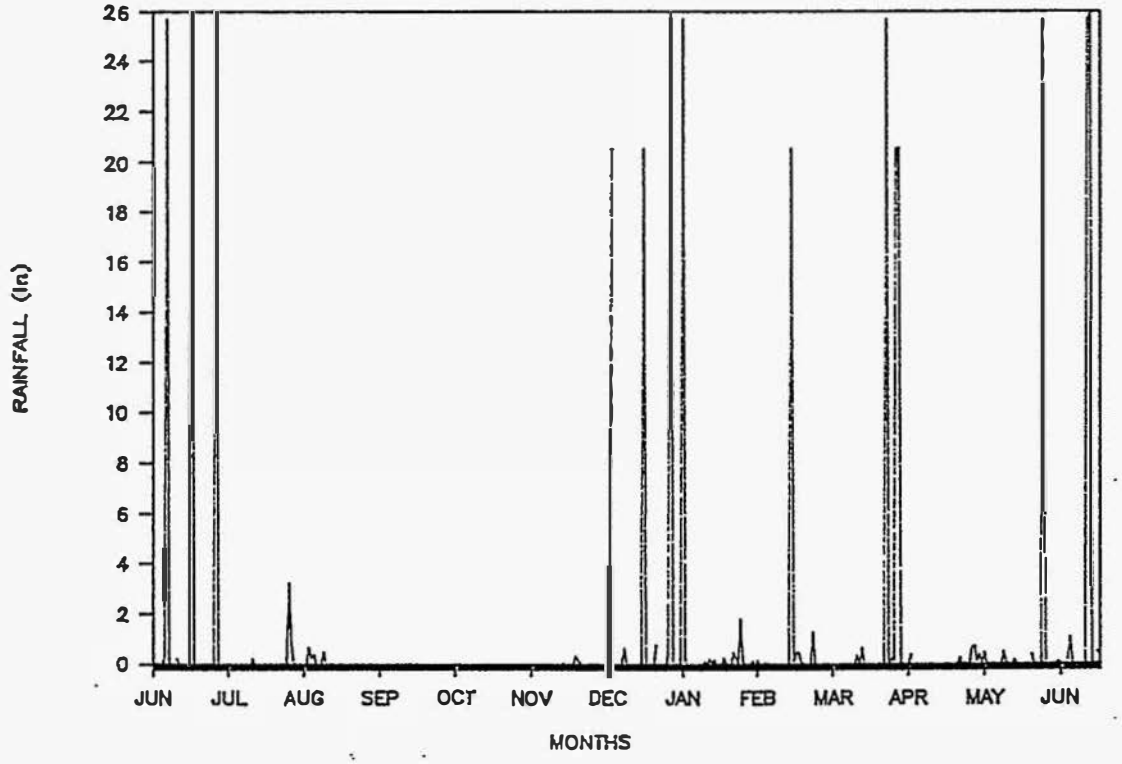

Figure A-17. ORNL weather data for 1985-86—precipitation.

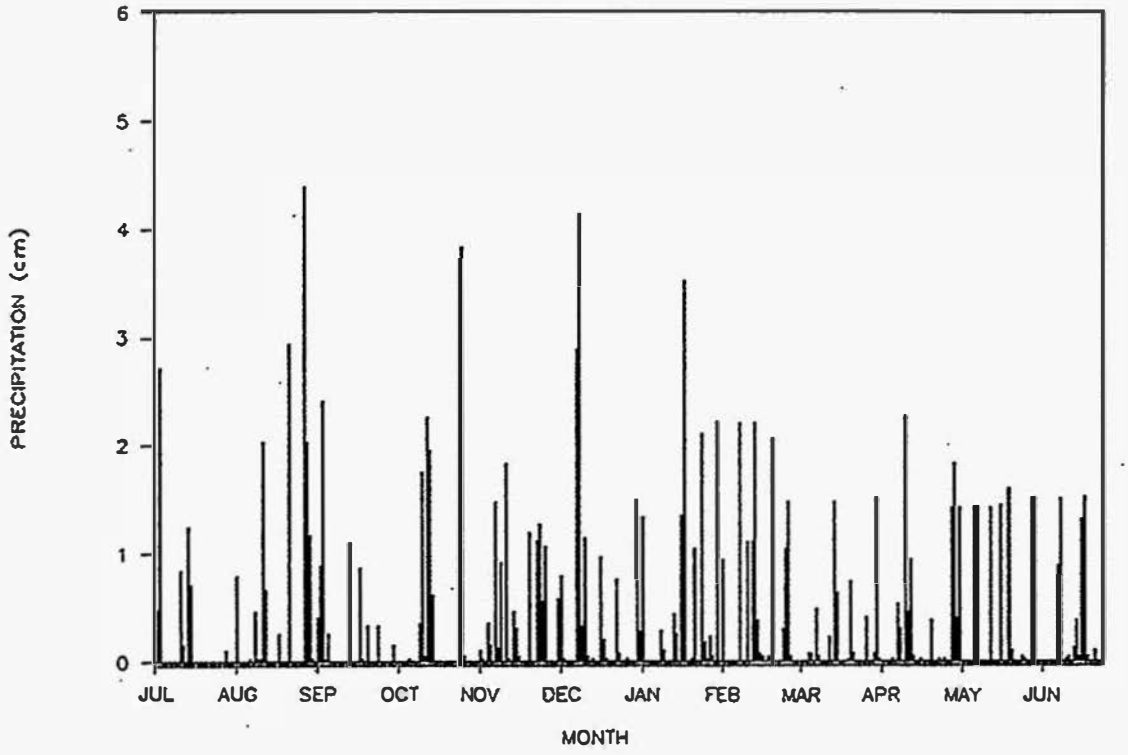

Figure A-18. ORNL weather data for 1986-87—precipitation.

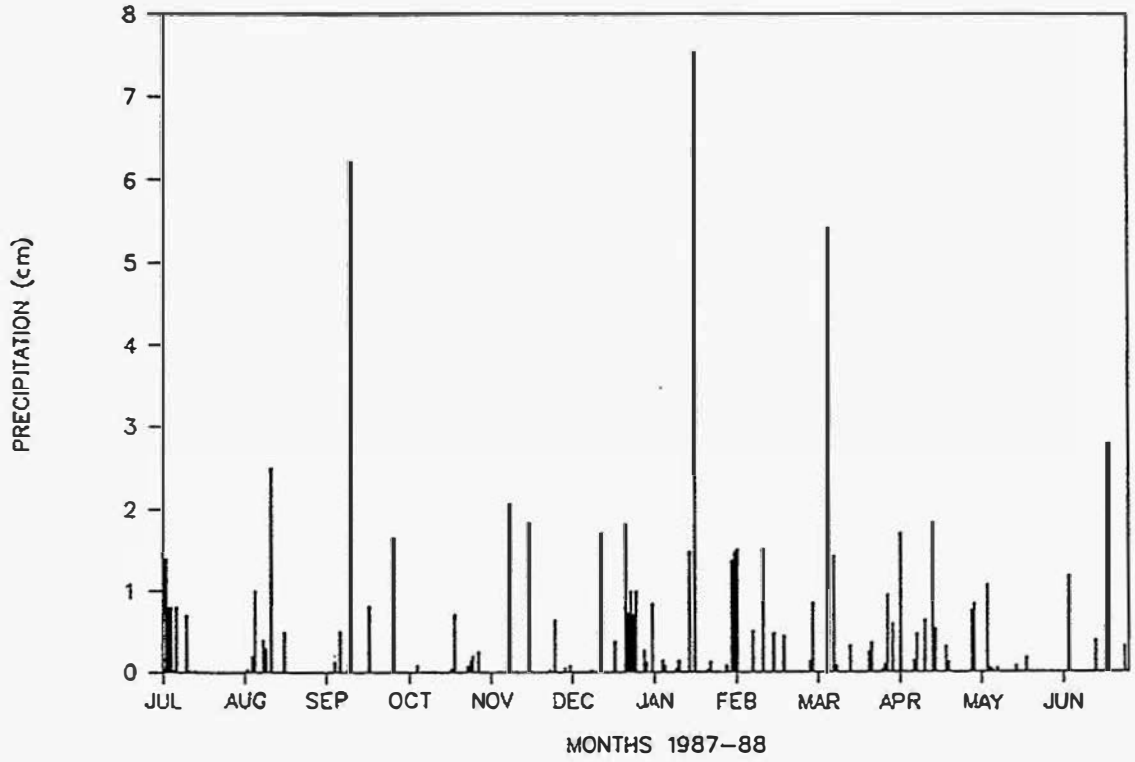

Figure A-19. ORNL weather data for 1987-88—precipitation.

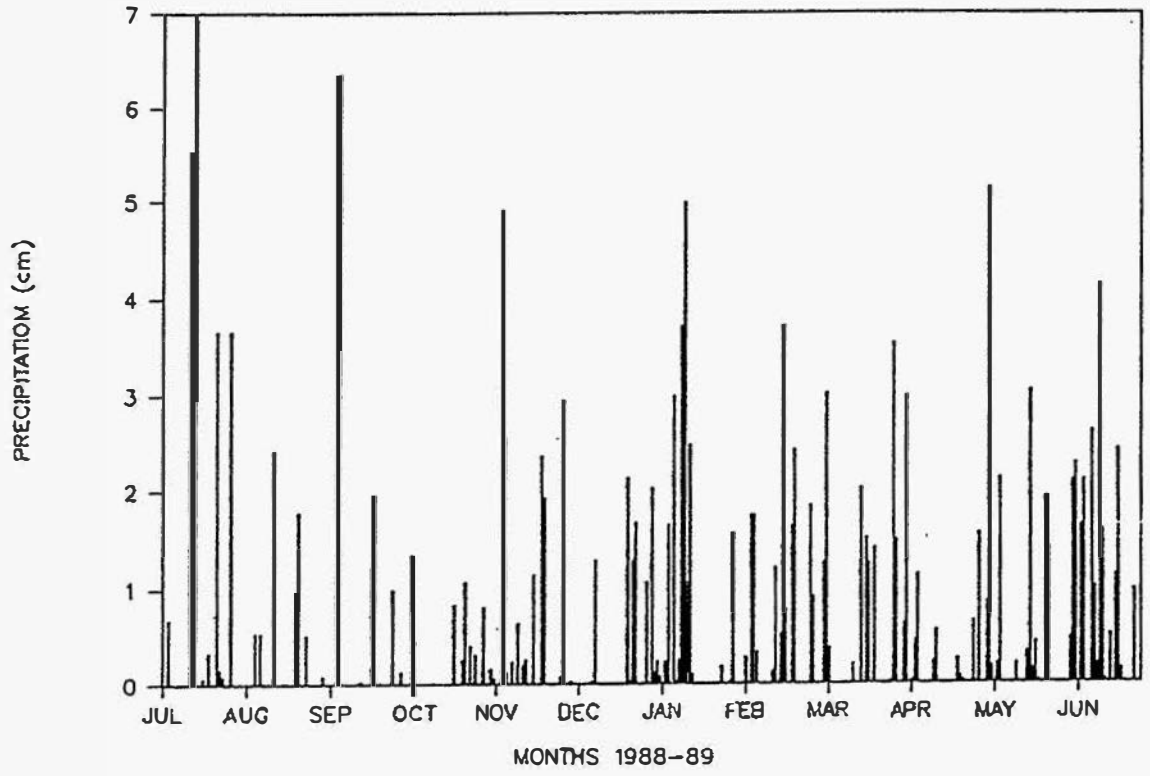

Figure A-20. ORNL weather data for 1988-89--precipitation.

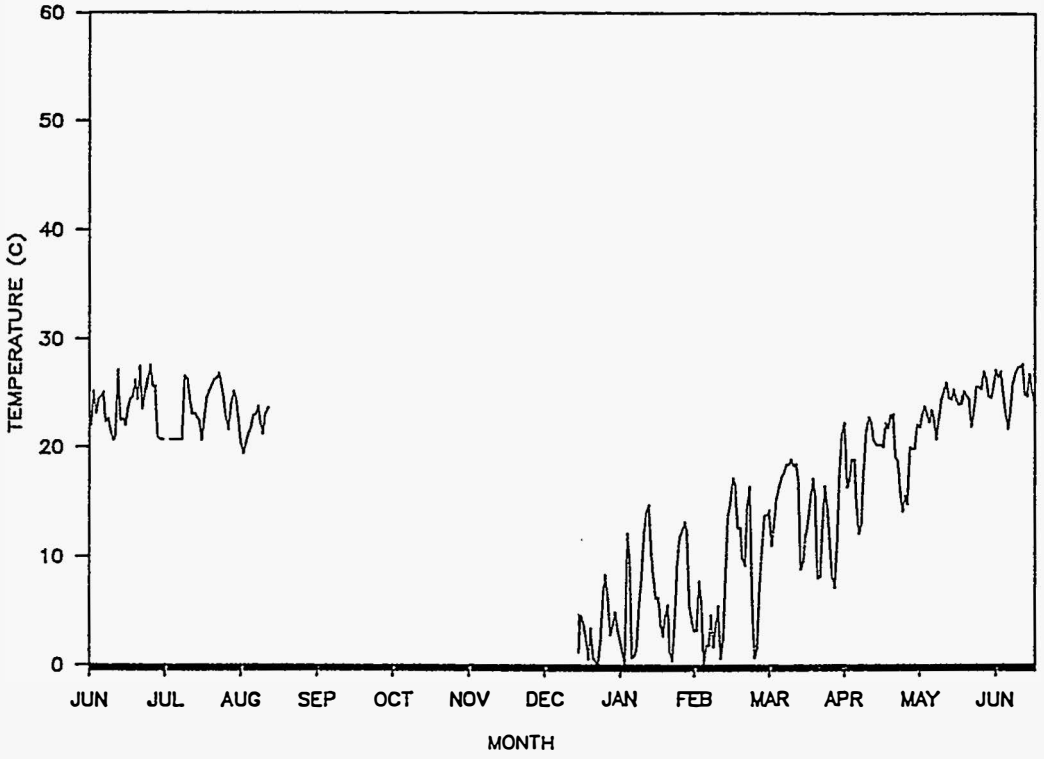

Figure A-21. ORNL weather data for 1985-86—-air temperature.

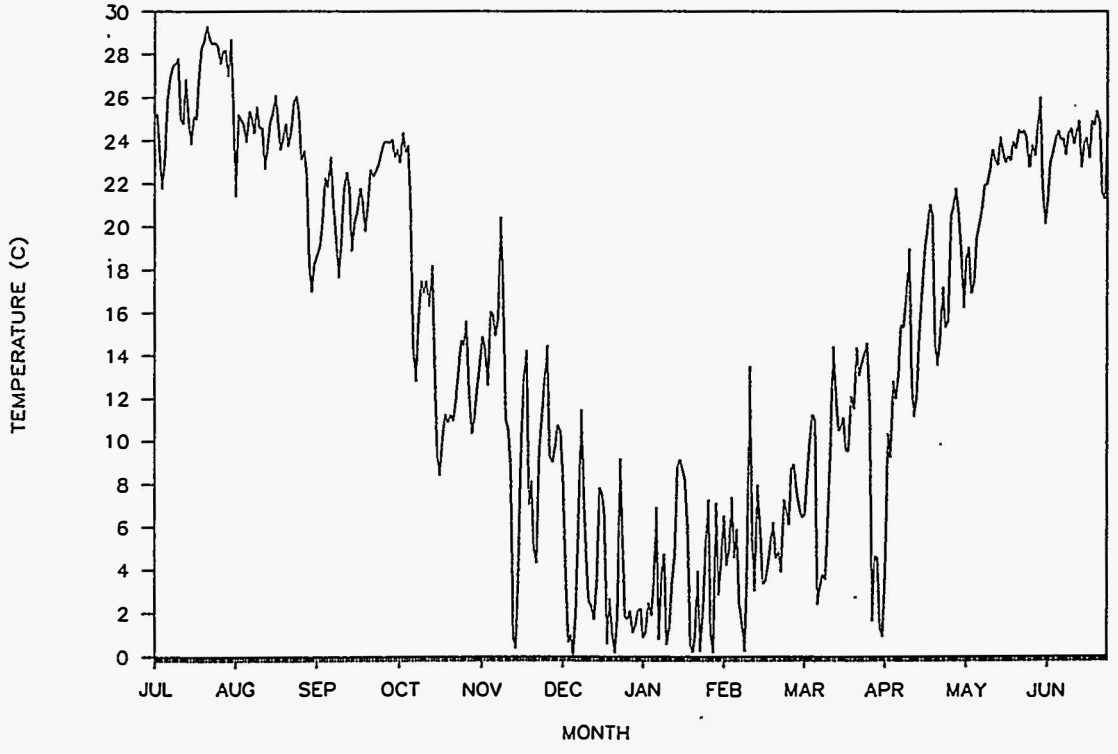

Figure A-22. ORNL weather data for 1986-87—air temperature.

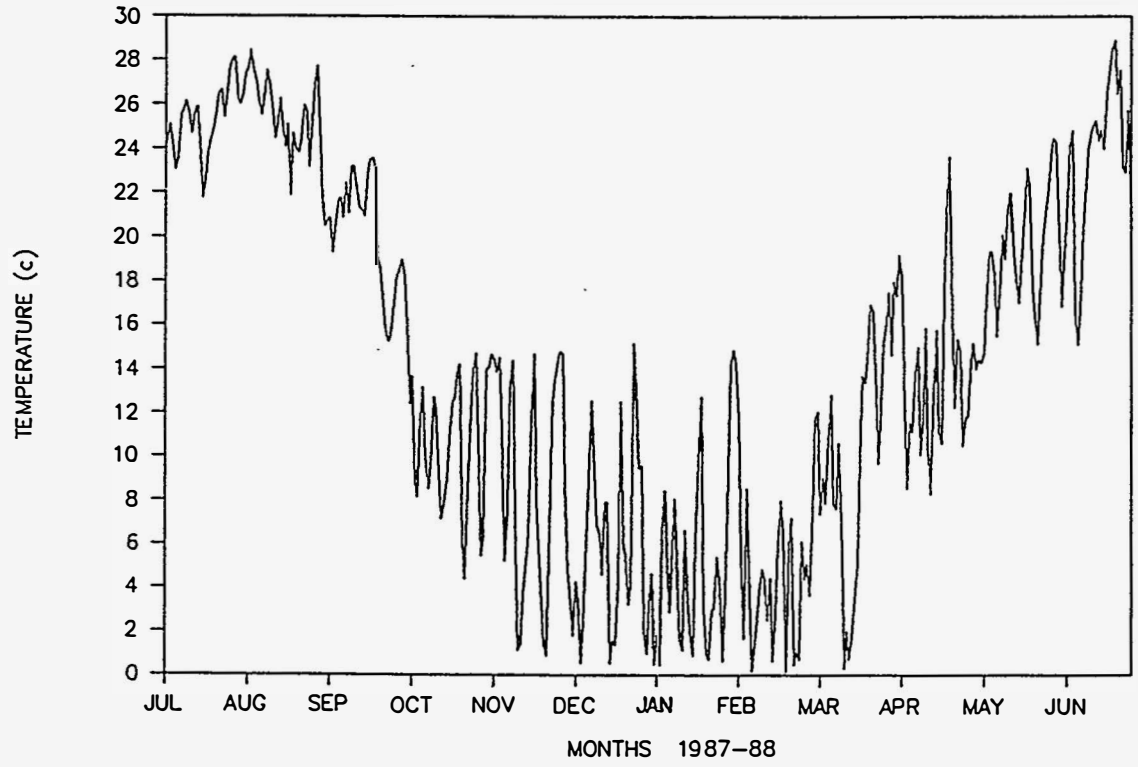

Figure A-23. ORNL weather data for 1987-88—air temperature.

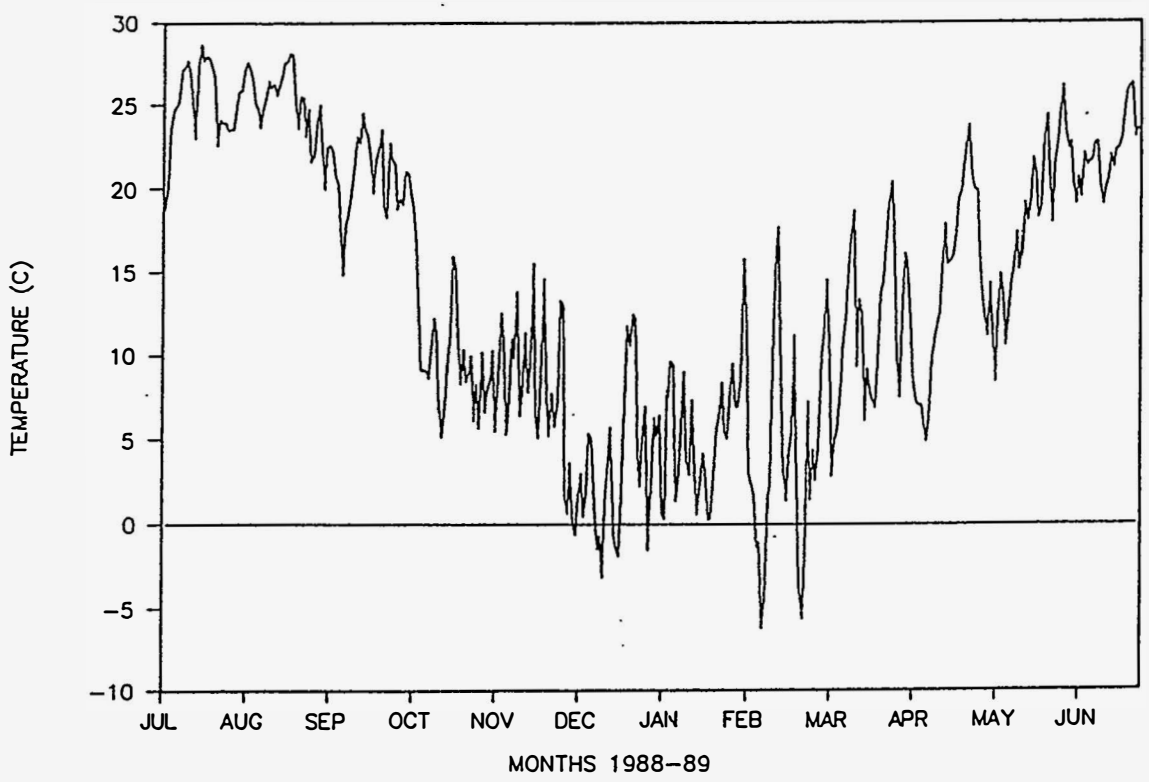

Figure A-24. ORNL weather data for 1988-89—air temperature.

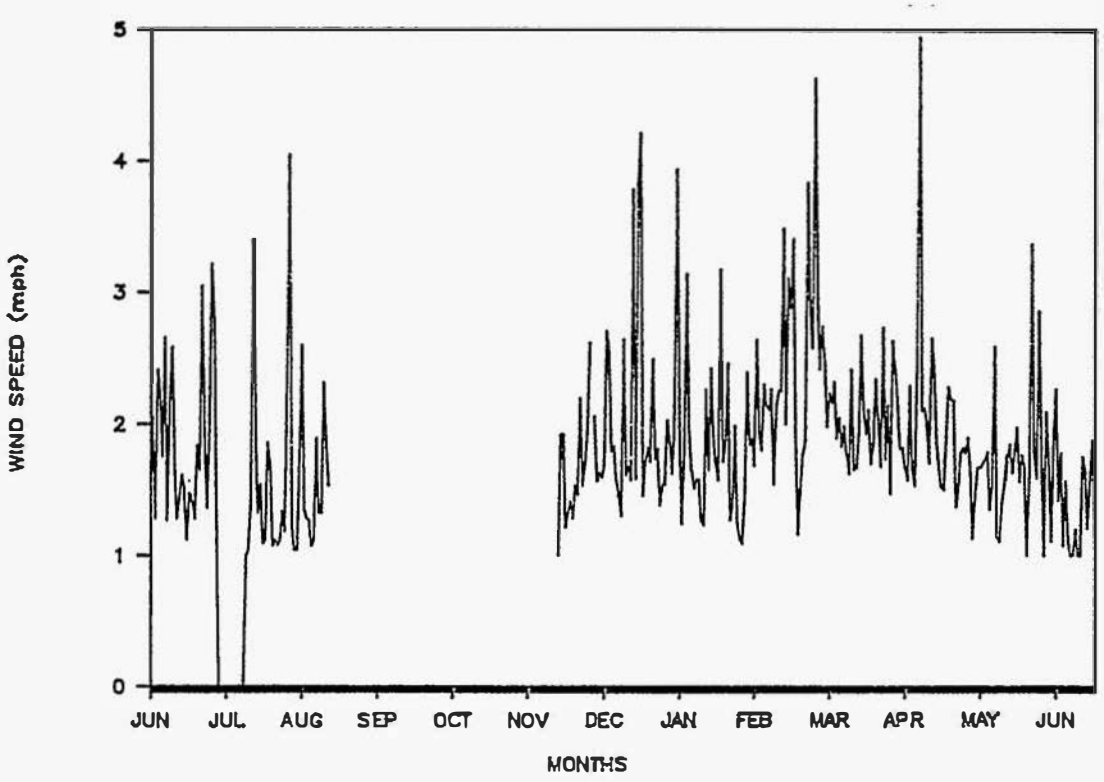

Figure A-25. ORNL weather data for 1985-86—wind speed.

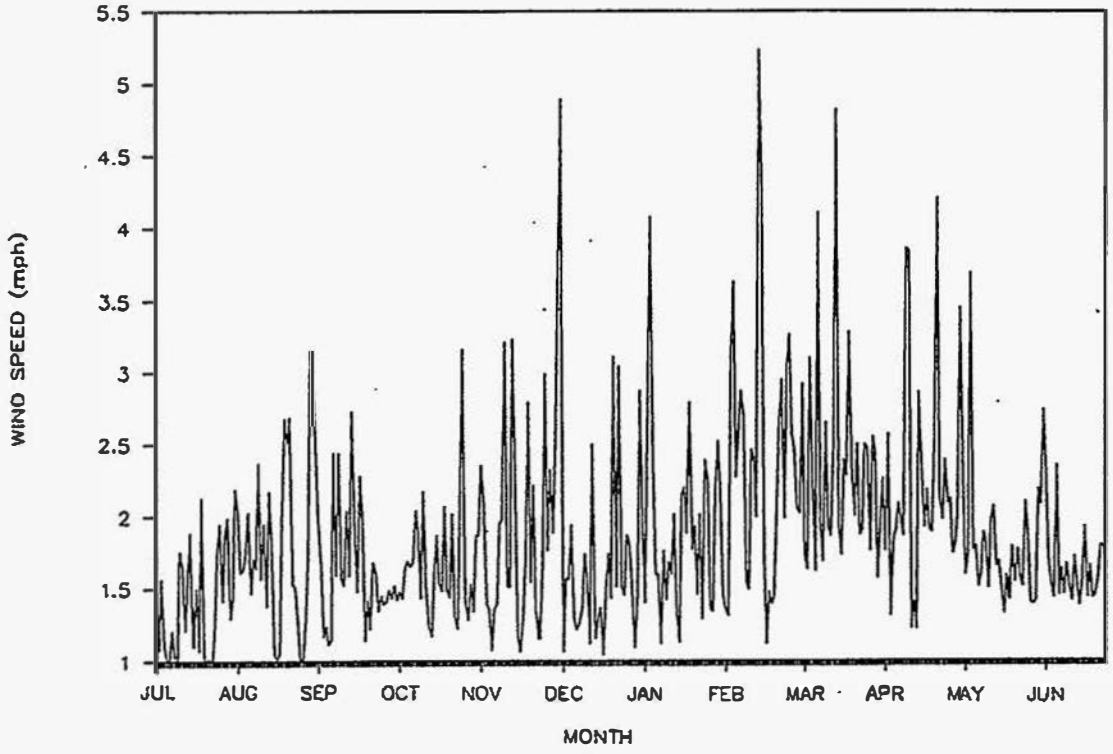

Figure A-26. ORNL weather data for 1986-87—wind speed.

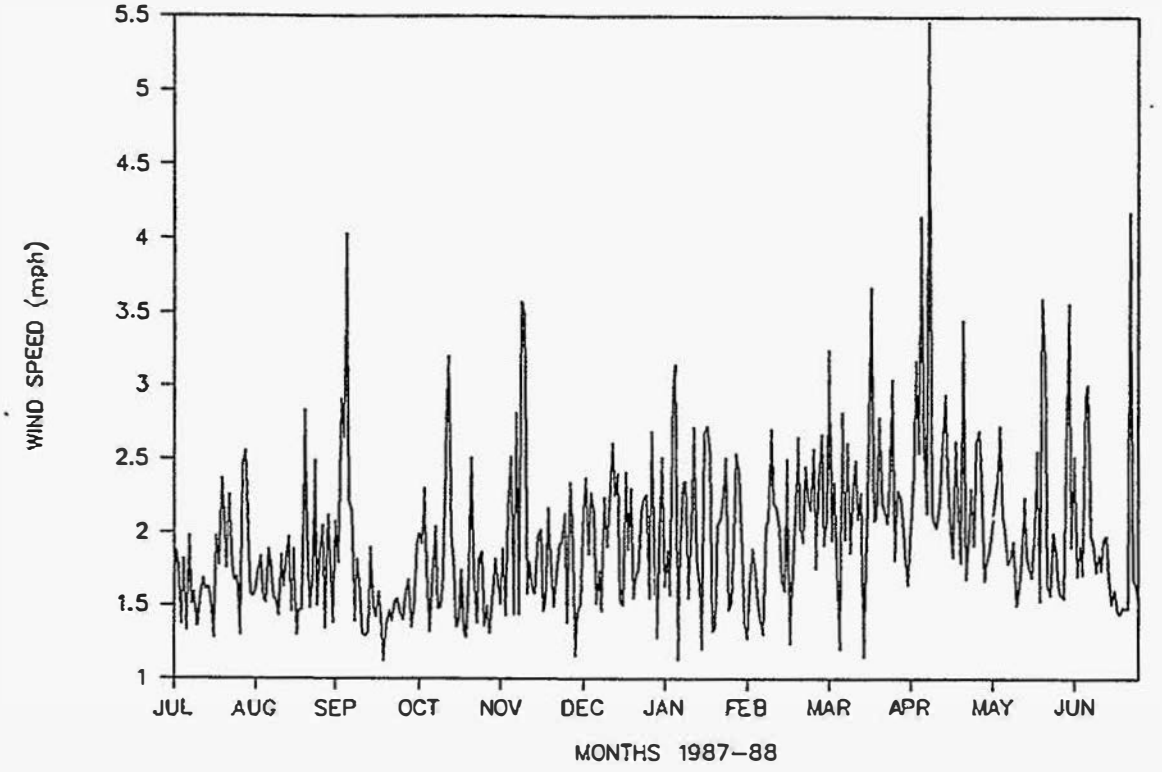

Figure A-27. ORNL weather data for 1987-88—wind speed.

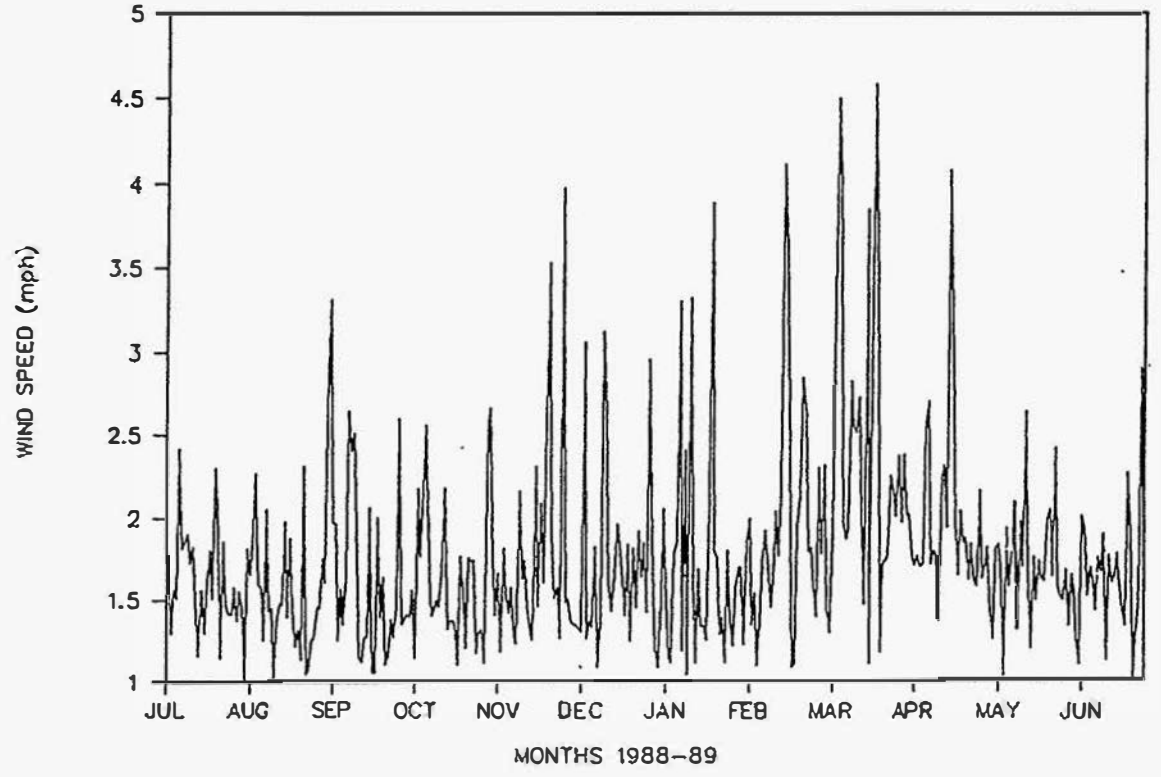

Figure A-28. ORNL weather data for 1988-89—wind speed.

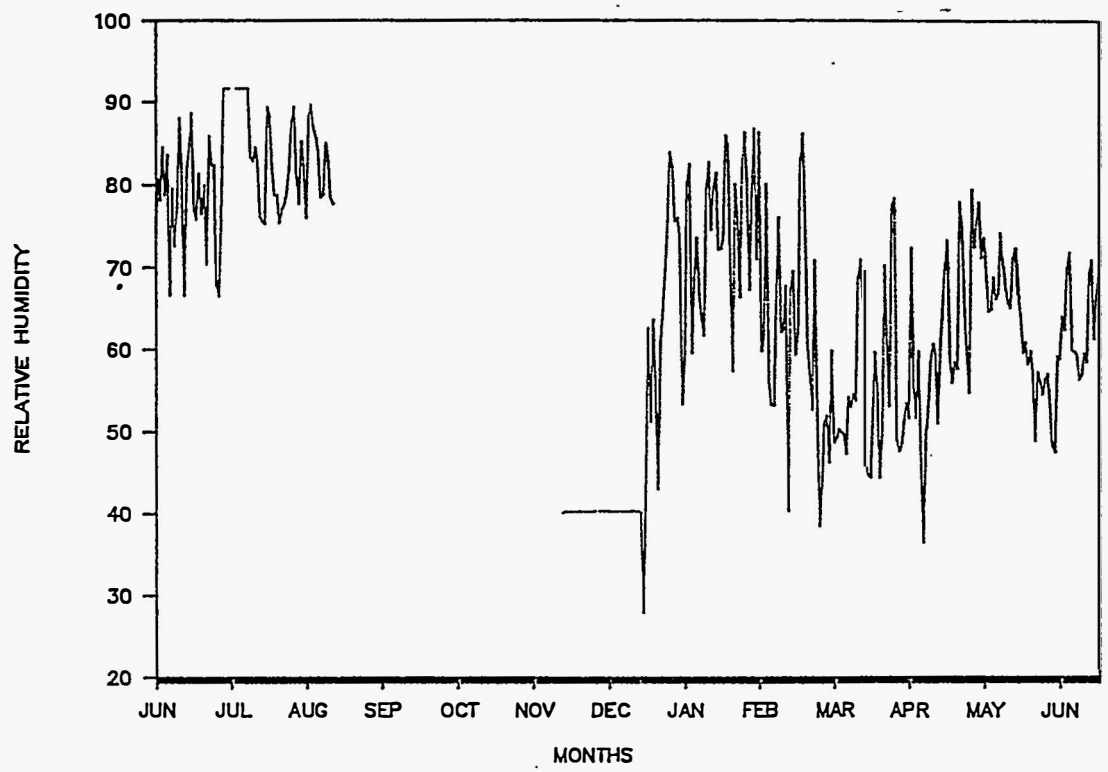

Figure A-29. ORNL weather data for 1985-86—relative humidity.

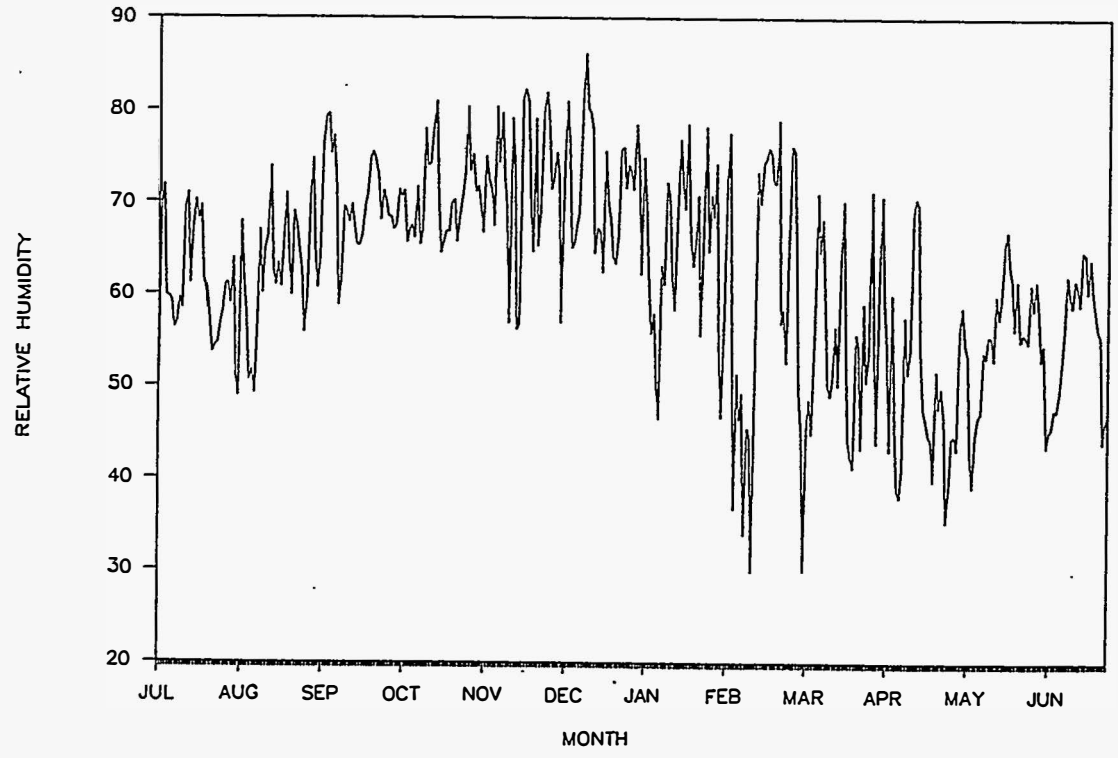

Figure A-30. ORNL weather data for 1986-87—relative humidity.

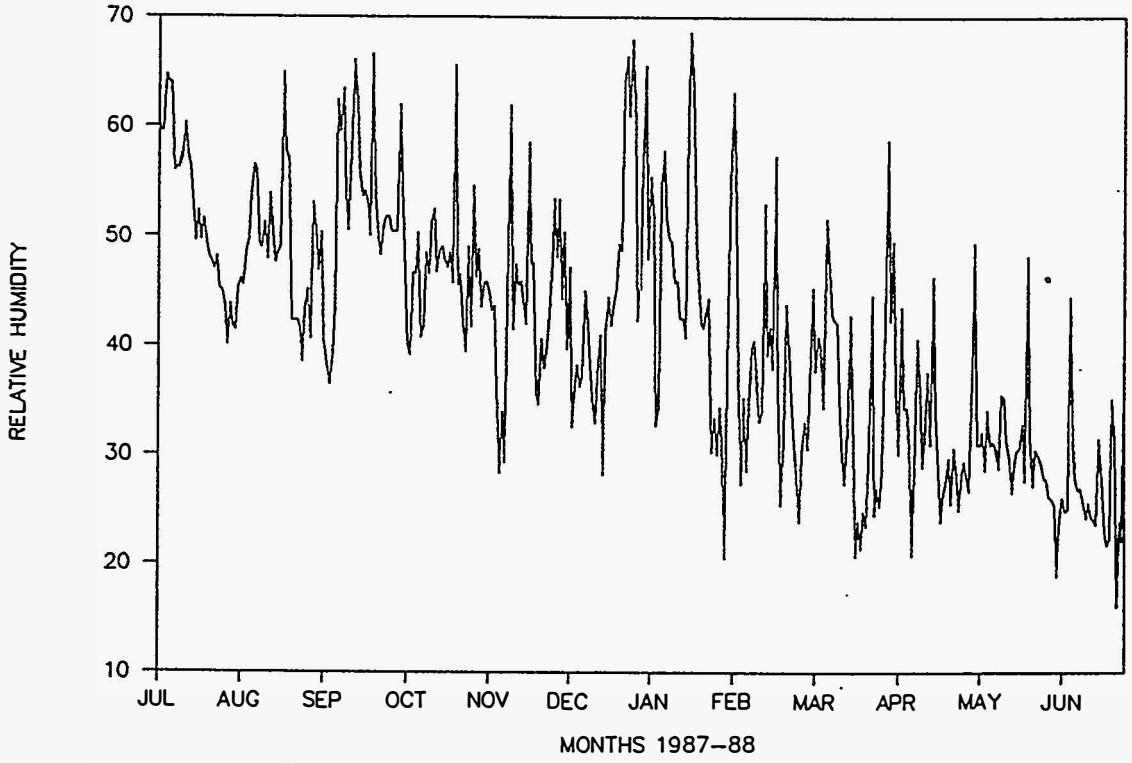

Figure A-31. ORNL weather data for 1987-88—relative humidity

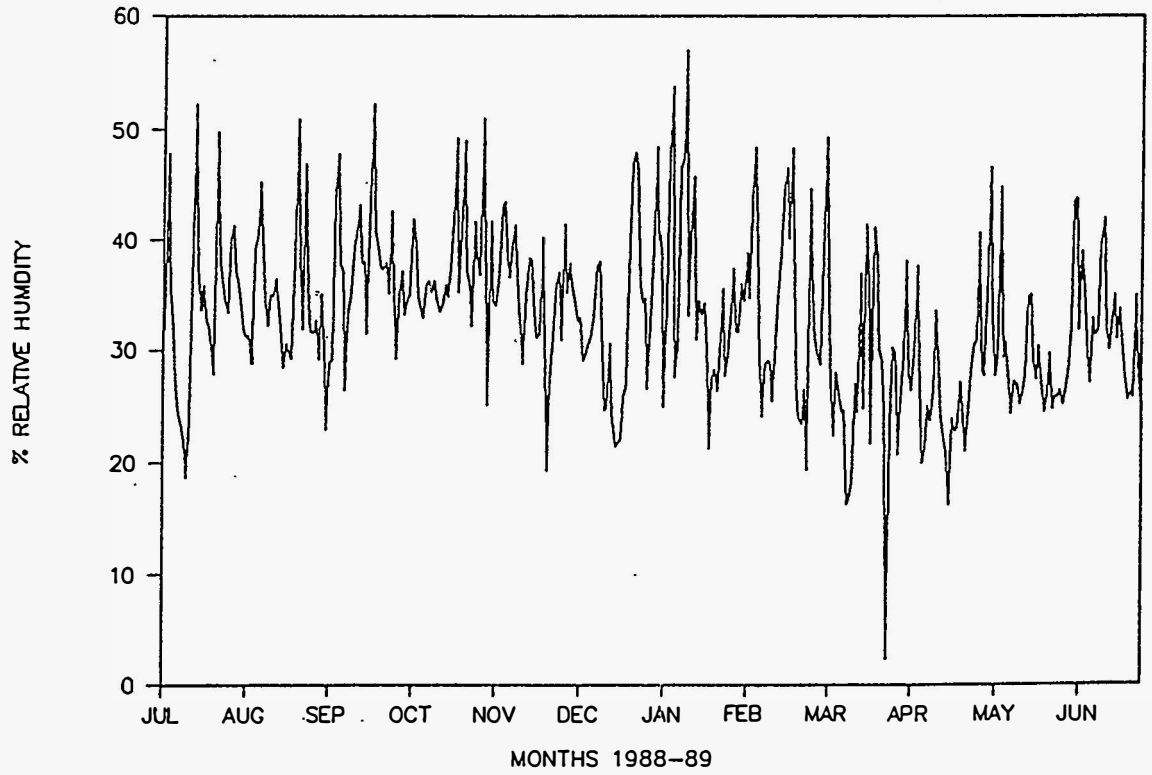

Figure A-32. ORNL weather data for 1988-89—relative humidity.

Appendix B

Soil Temperature Data—Resistance Probes

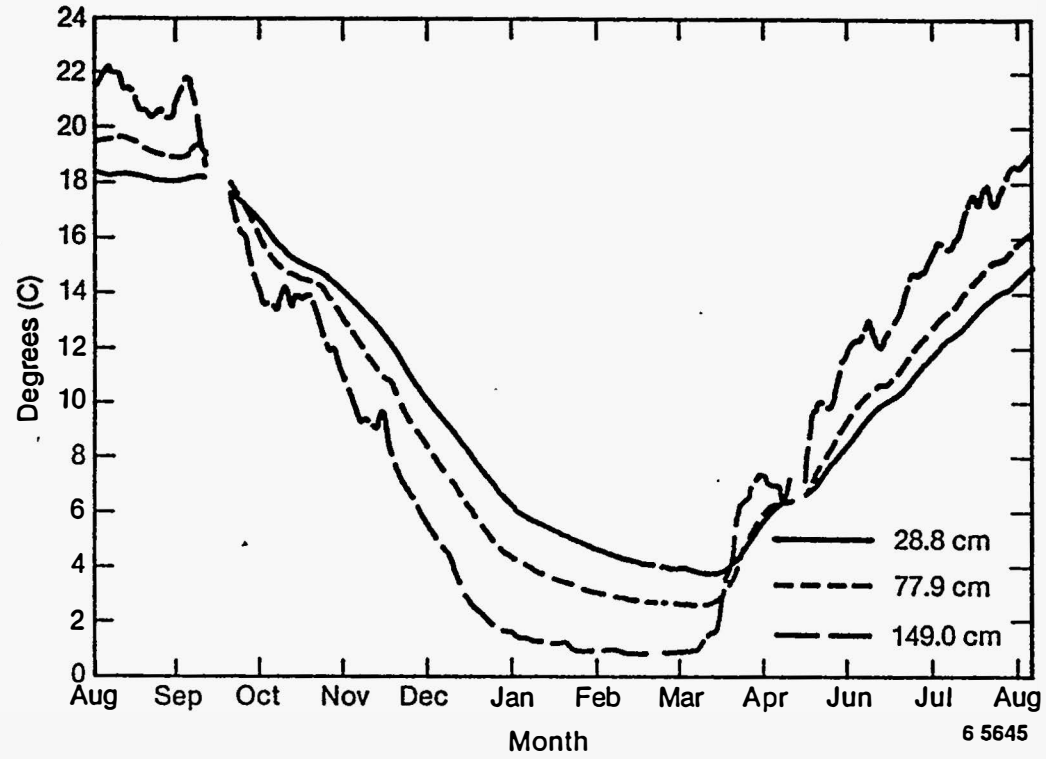

Figure B-1. ANL-E lysimeter 1 soil temperatures for 1985-86.

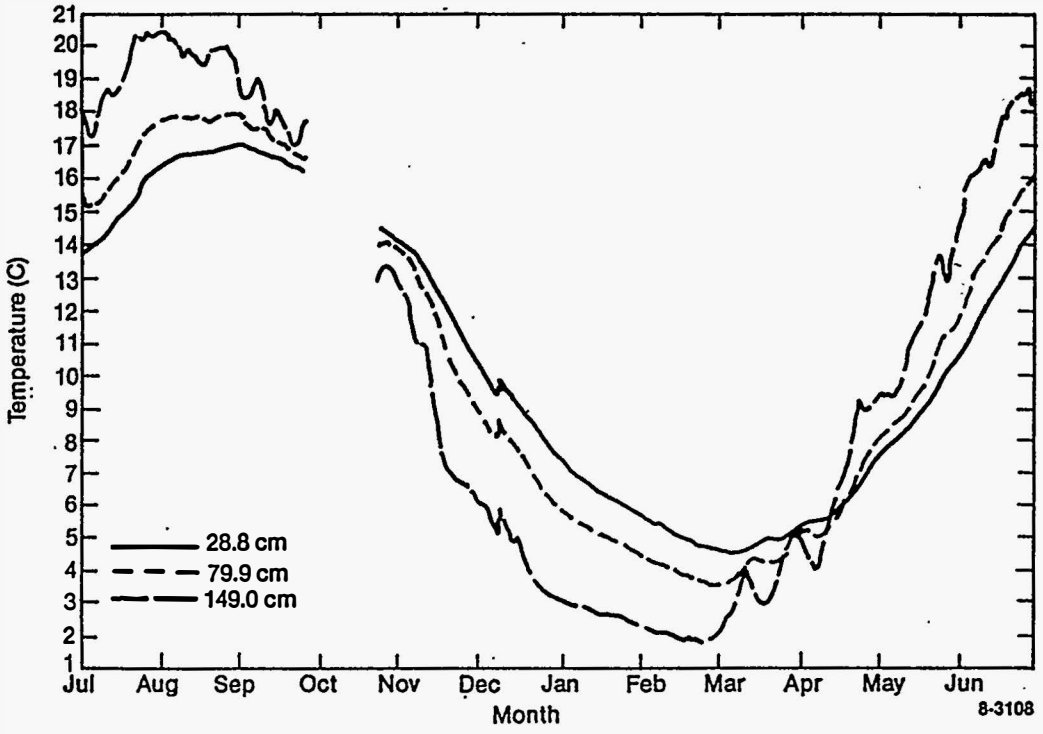

Figure B-2. ANL-E lysimeter 1 soil temperatures for 1986-87.

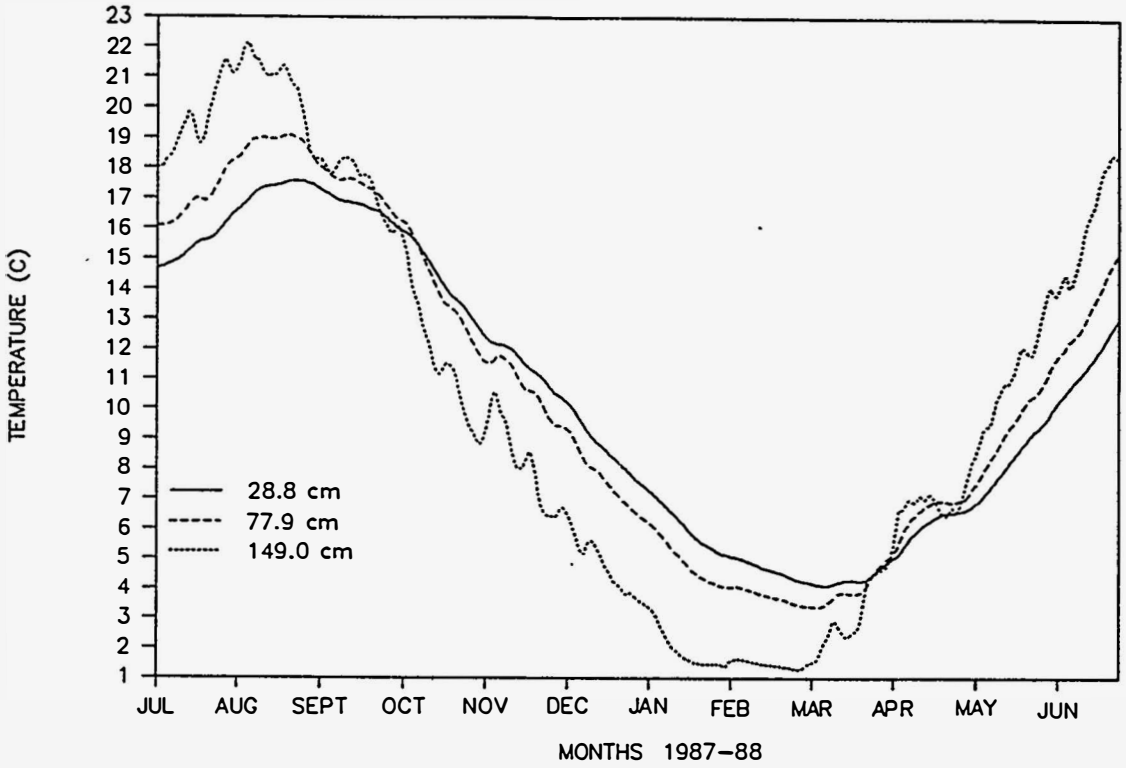

Figure B-3. ANL-E lysimeter 1 soil temperatures for 1987-88.

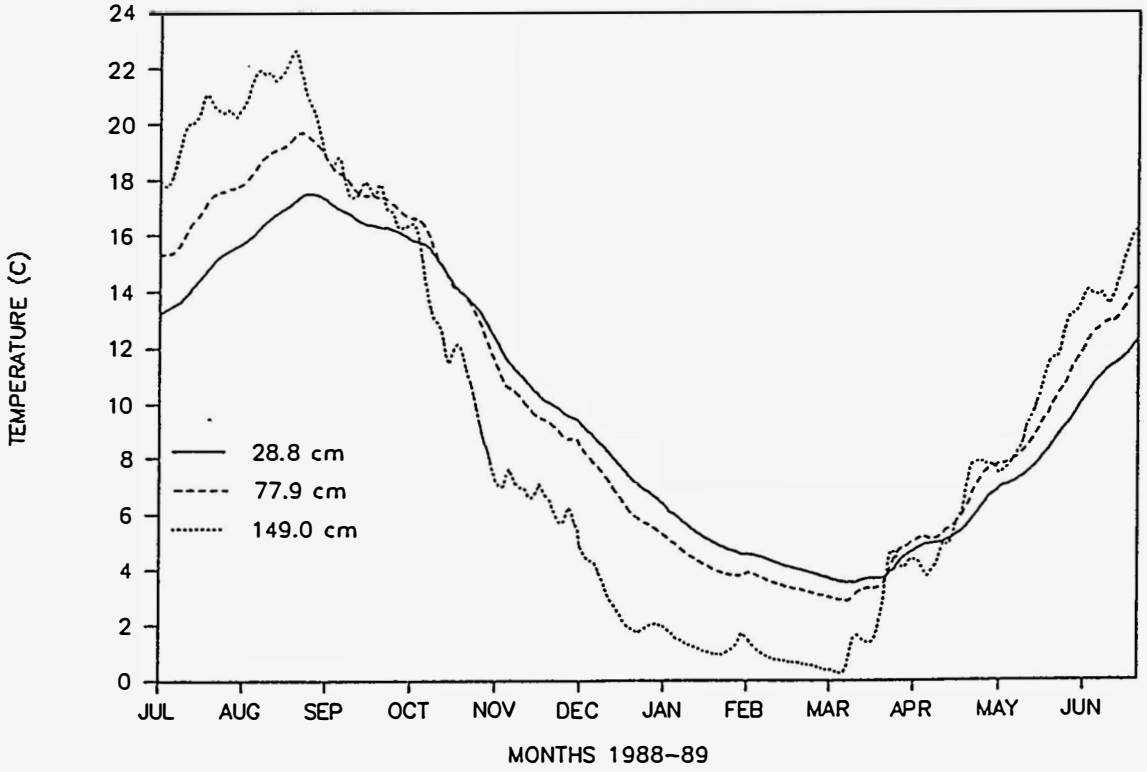

Figure B-4. ANL-E lysimeter 1 soil temperatures for 1988-89.

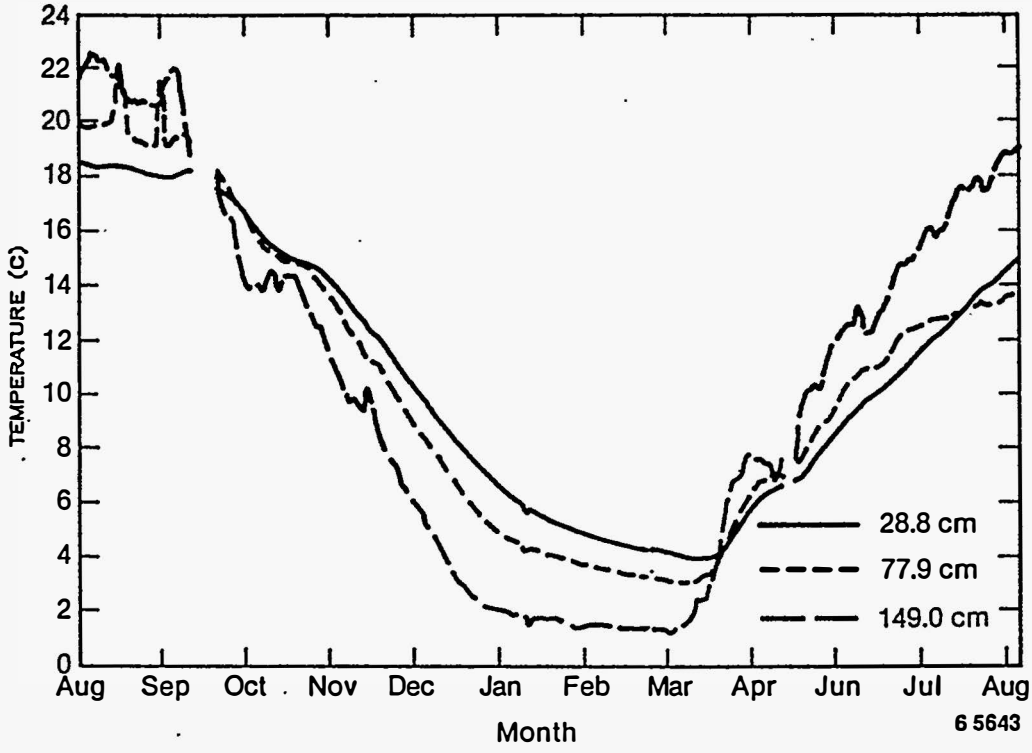

Figure B-5. ANL-E lysimeter 2 soil temperatures for 1985-86.

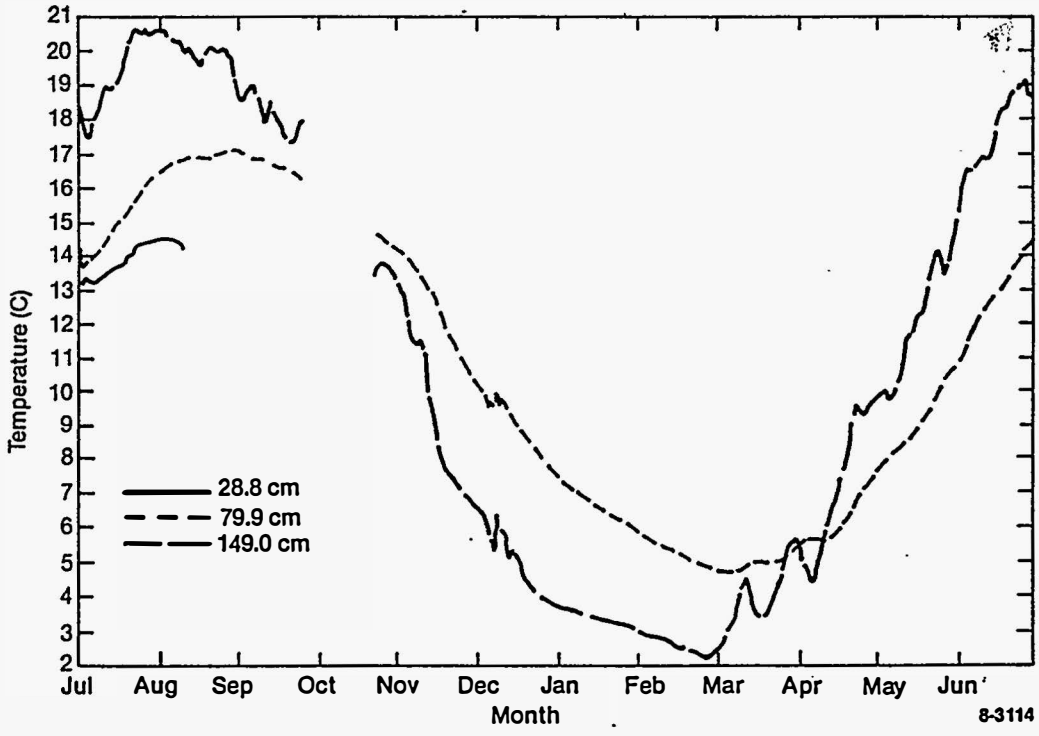

Figure B-6. ANL-E lysimeter 2 soil temperatures for 1986-87.

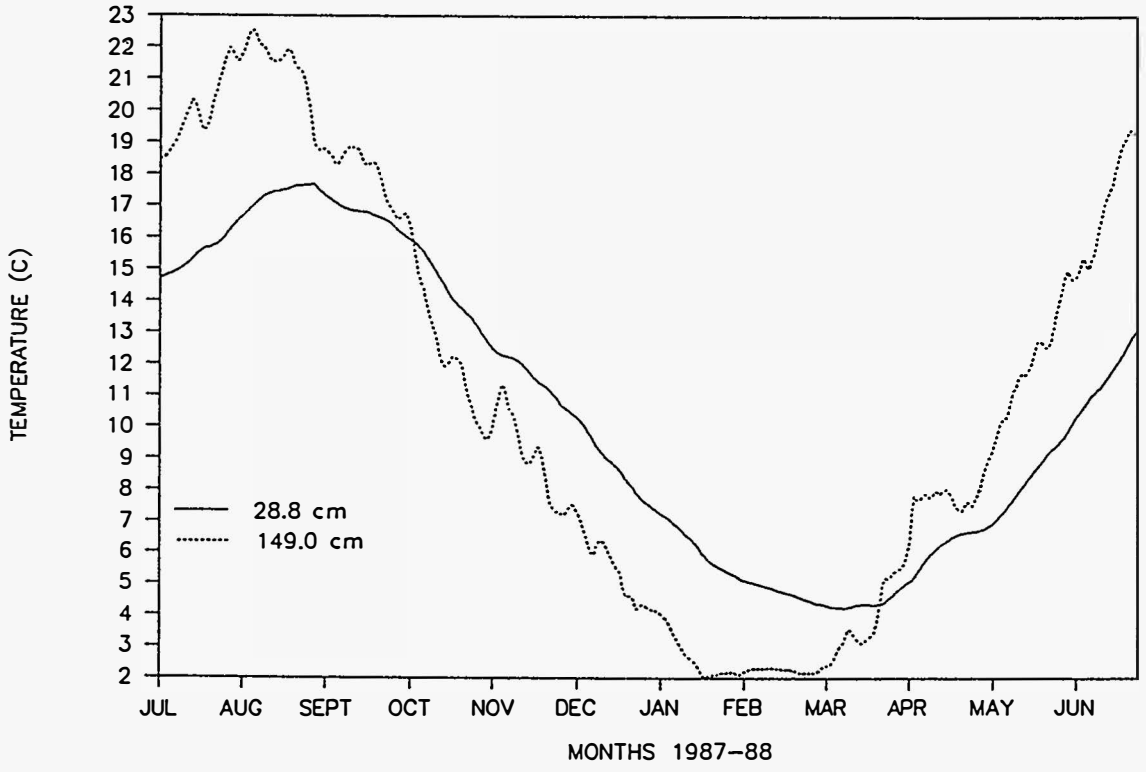

Figure B-7. ANL-E lysimeter 2 soil temperatures for 1987-88.

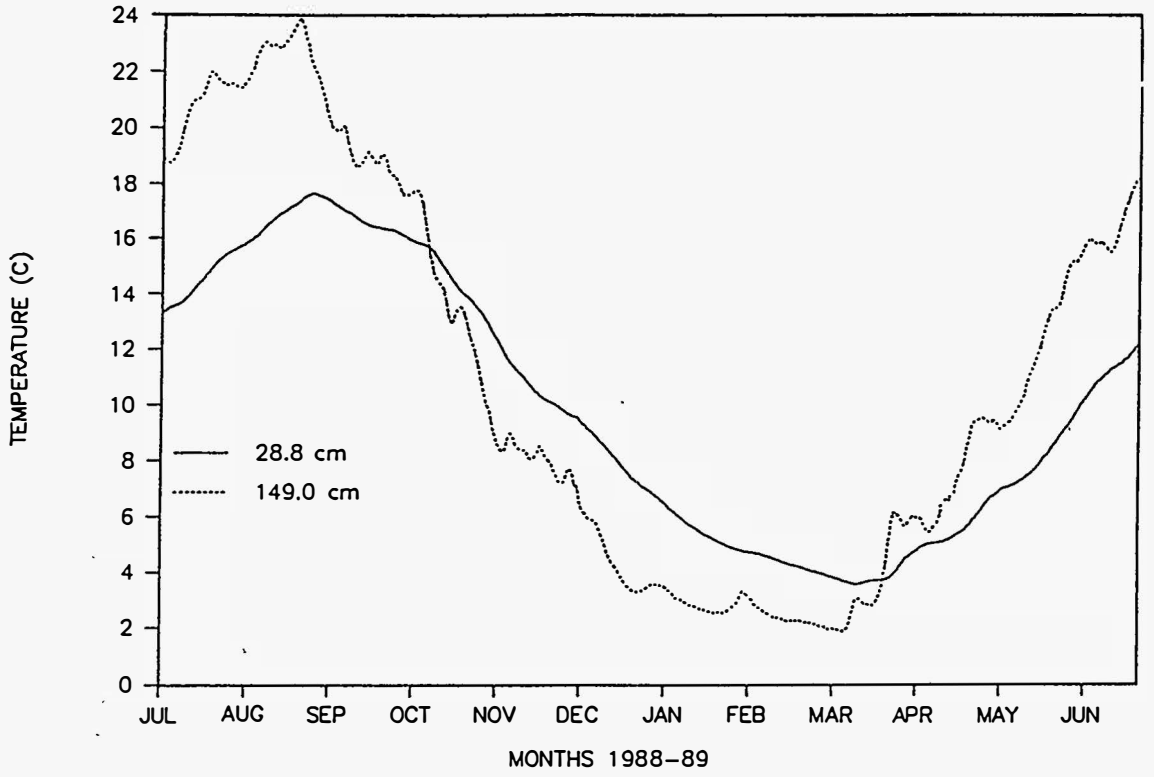

Figure B-8. ANL-E lysimeter 2 soil temperatures for 1988-89.

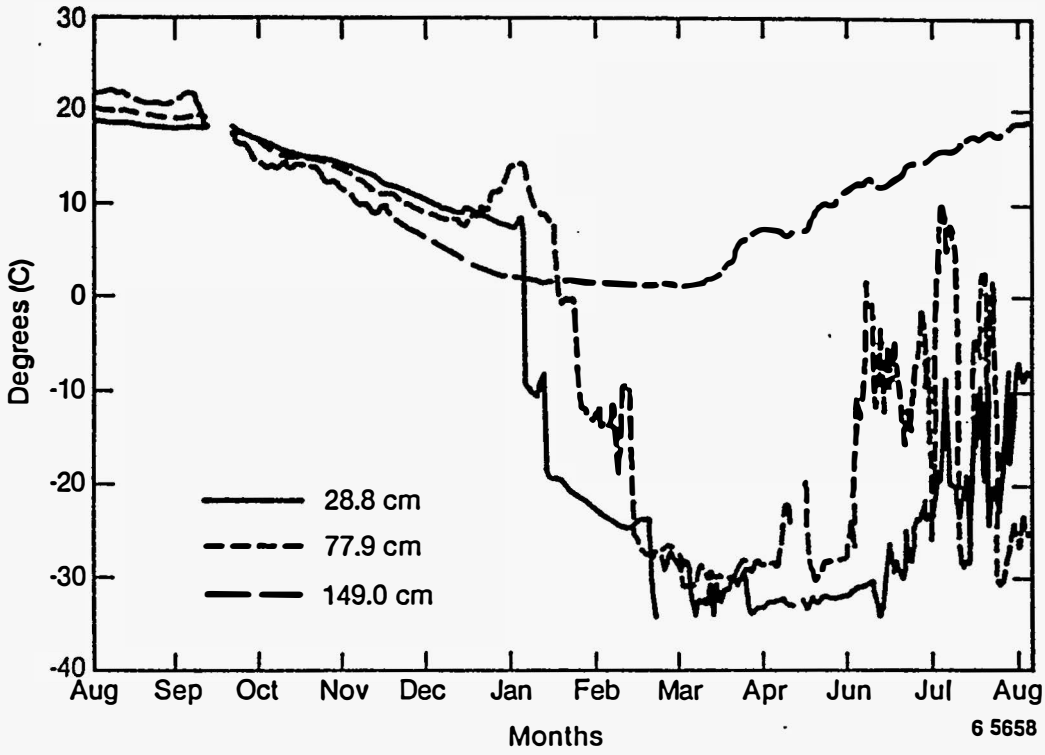

Figure B-9. ANL-E lysimeter 3 soil temperatures for 1985-86.

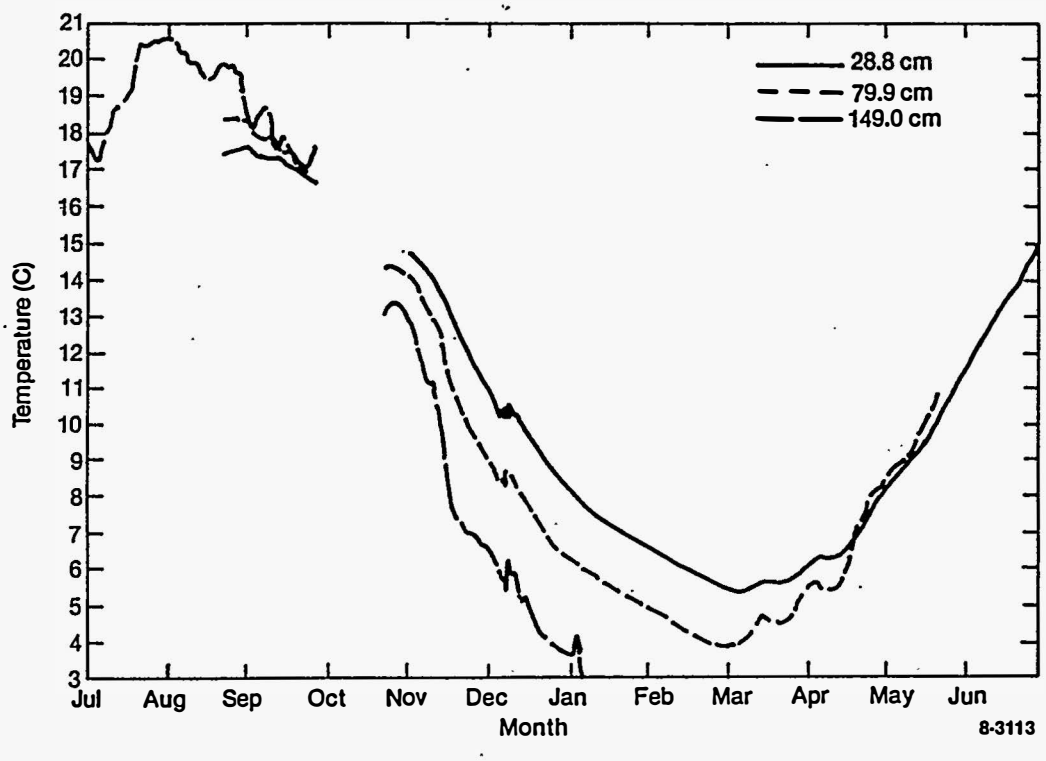

Figure B-10. ANL-E lysimeter 3 soil temperatures for 1986-87.

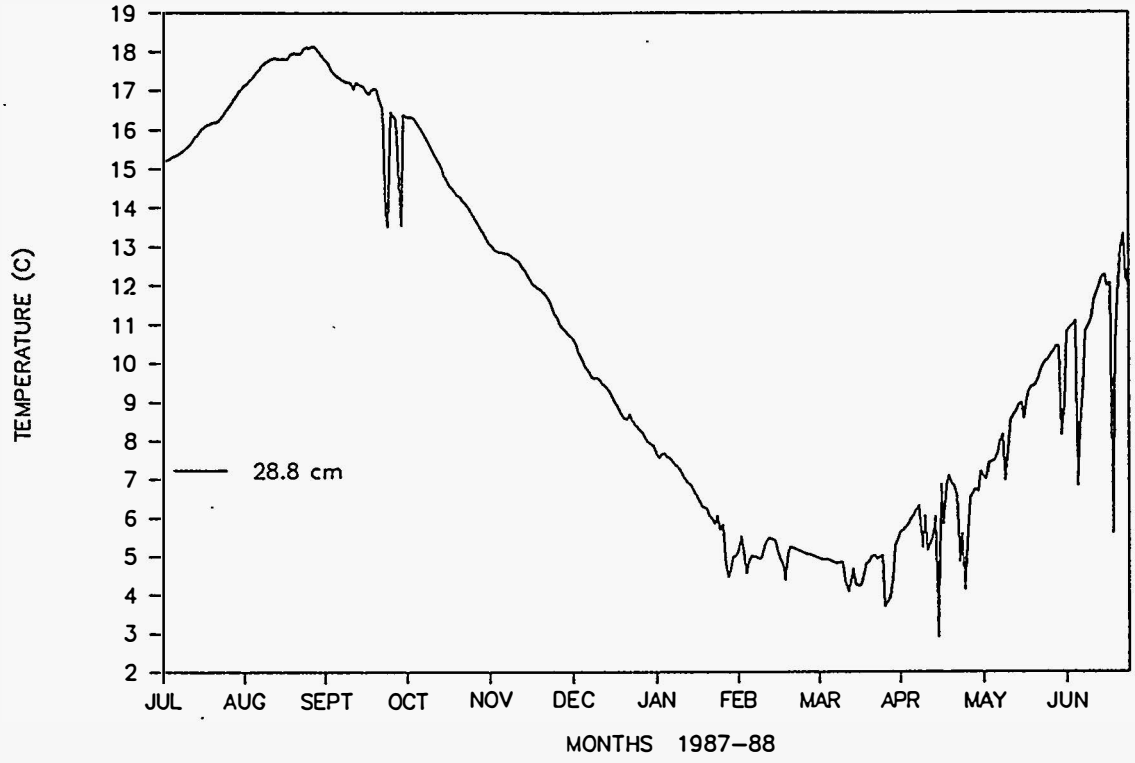

Figure B-11. ANL-E lysimeter 3 soil temperatures for 1987-88.

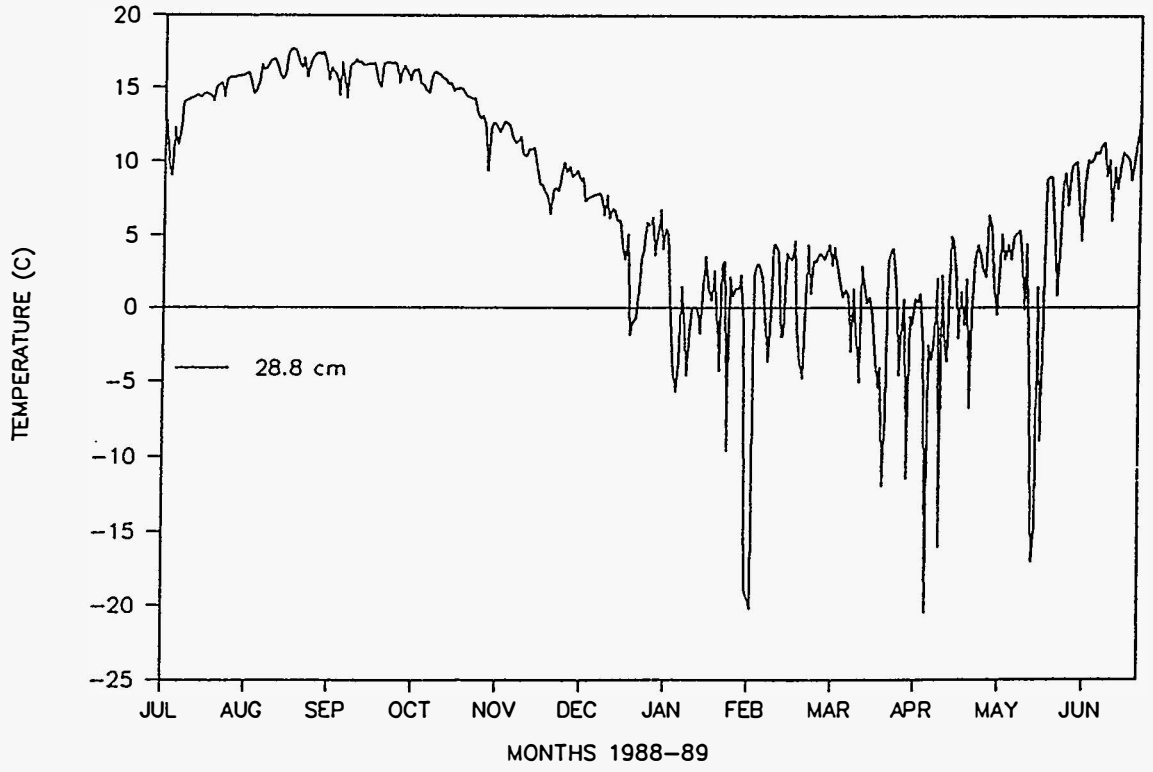

Figure B-12. ANL-E lysimeter 3 soil temperatures for 1988-89.

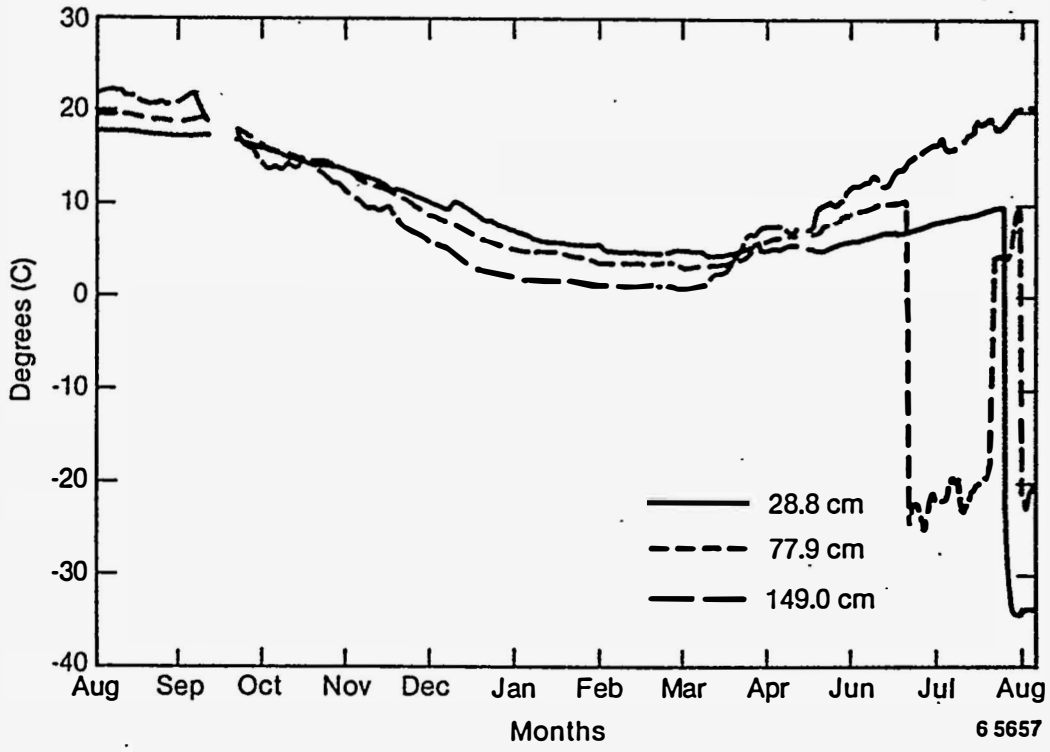

Figure B-13. ANL-E lysimeter 4 soil temperatures for 1985-86.

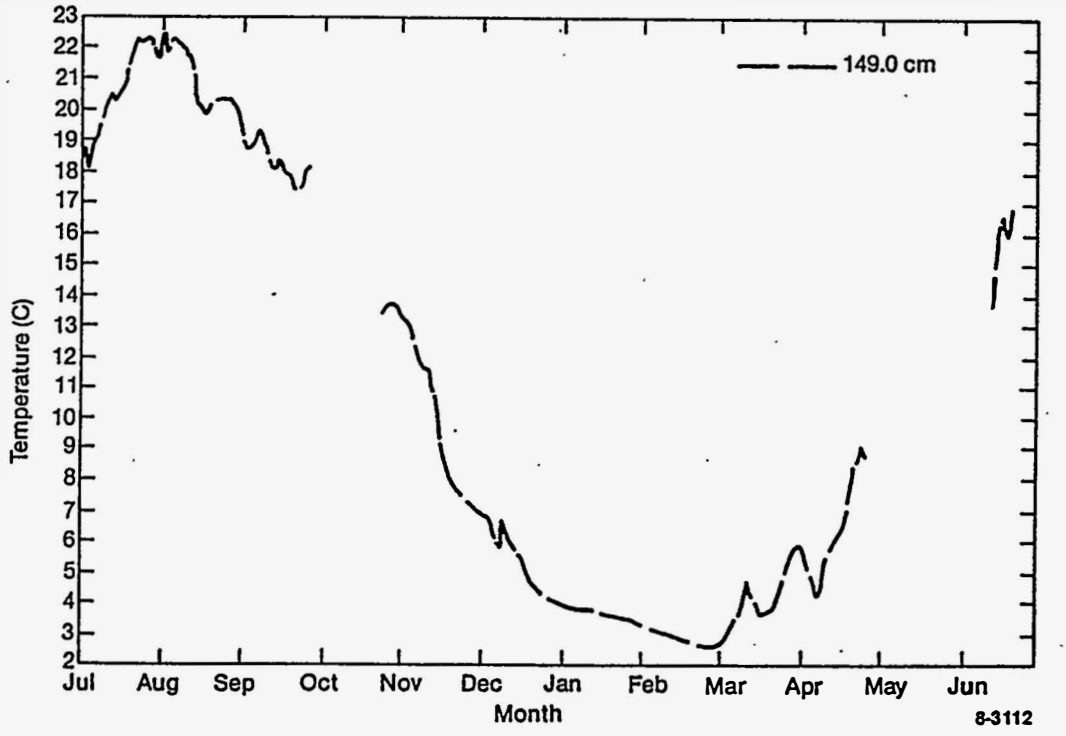

Figure B-14. ANL-E lysimeter 4 soil temperatures for 1986-87.

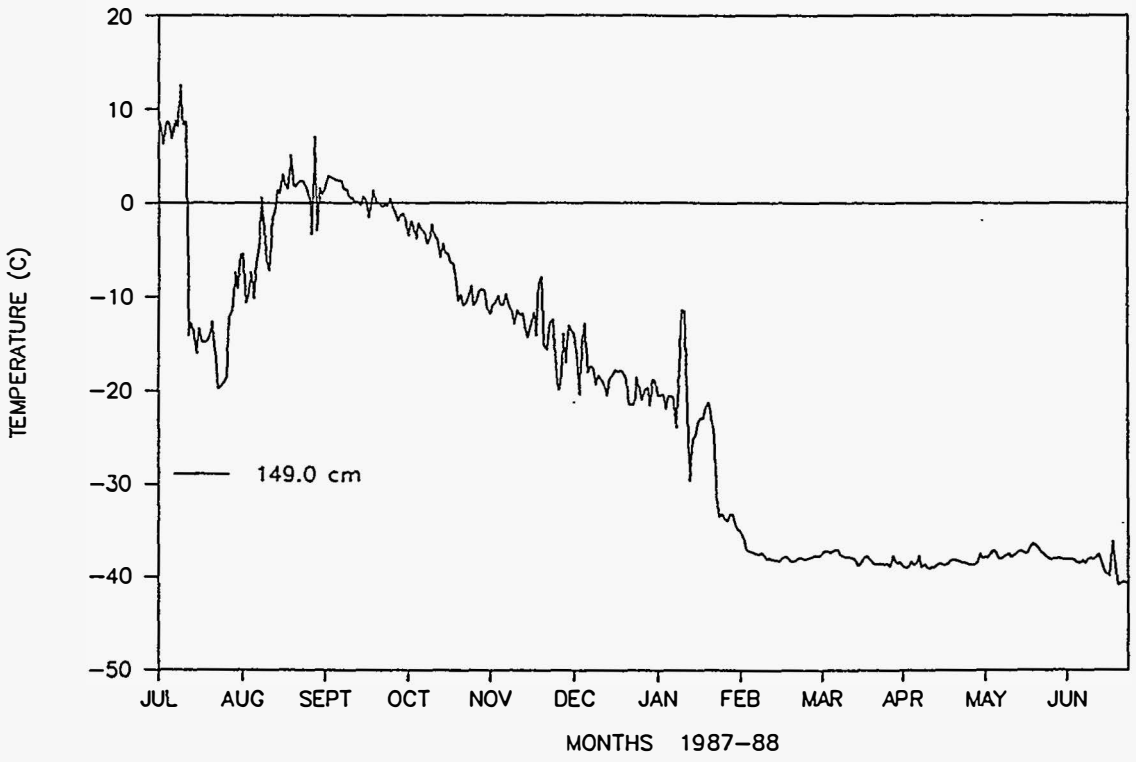

Figure B-15. ANL-E lysimeter 4 soil temperatures for 1987-88.

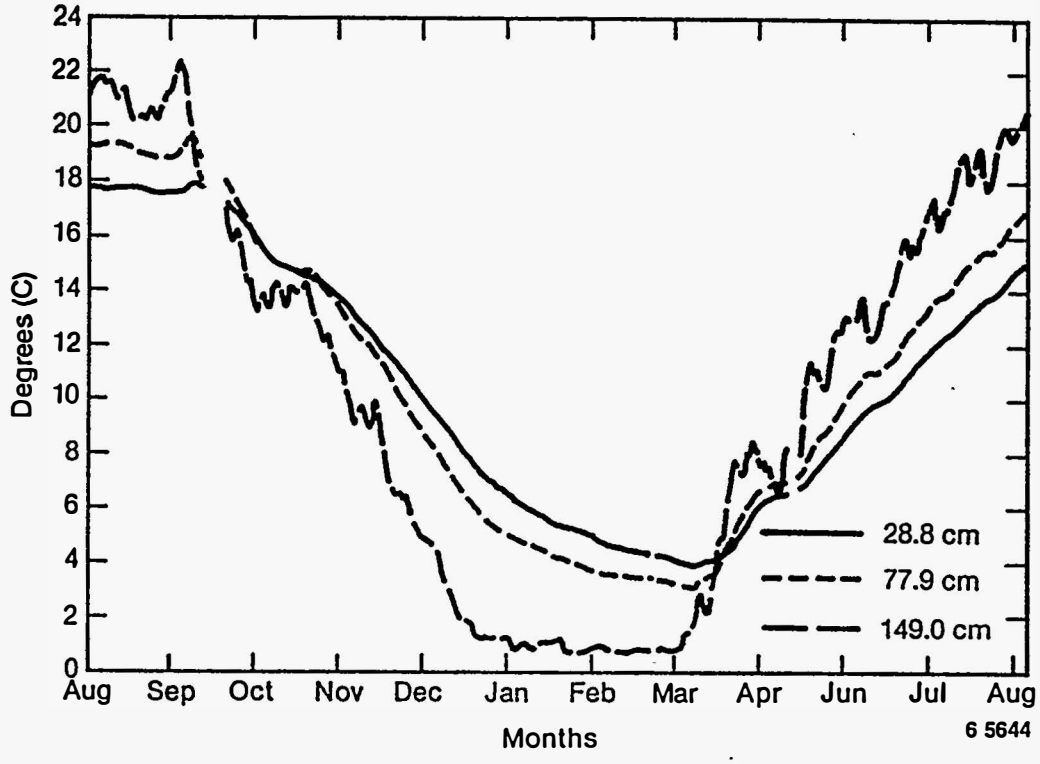

Figure B-16. ANL-E lysimeter 5 soil temperatures for 1985-86.

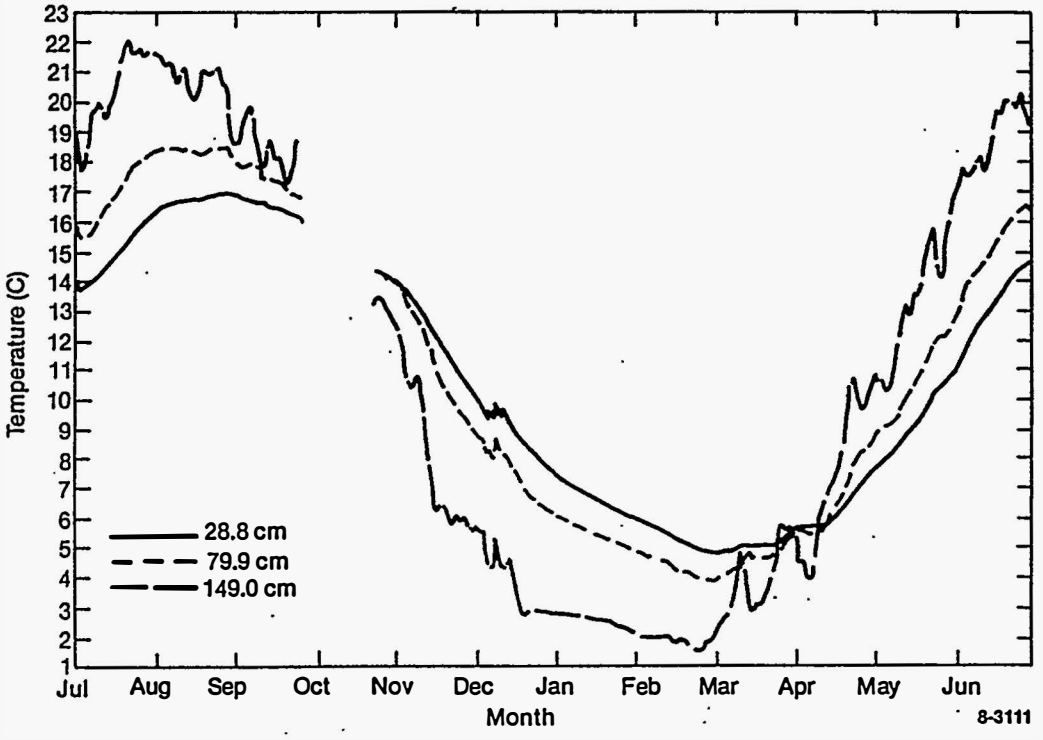

Figure B-17. ANL-E lysimeter 5 soil temperatures for 1986-87.

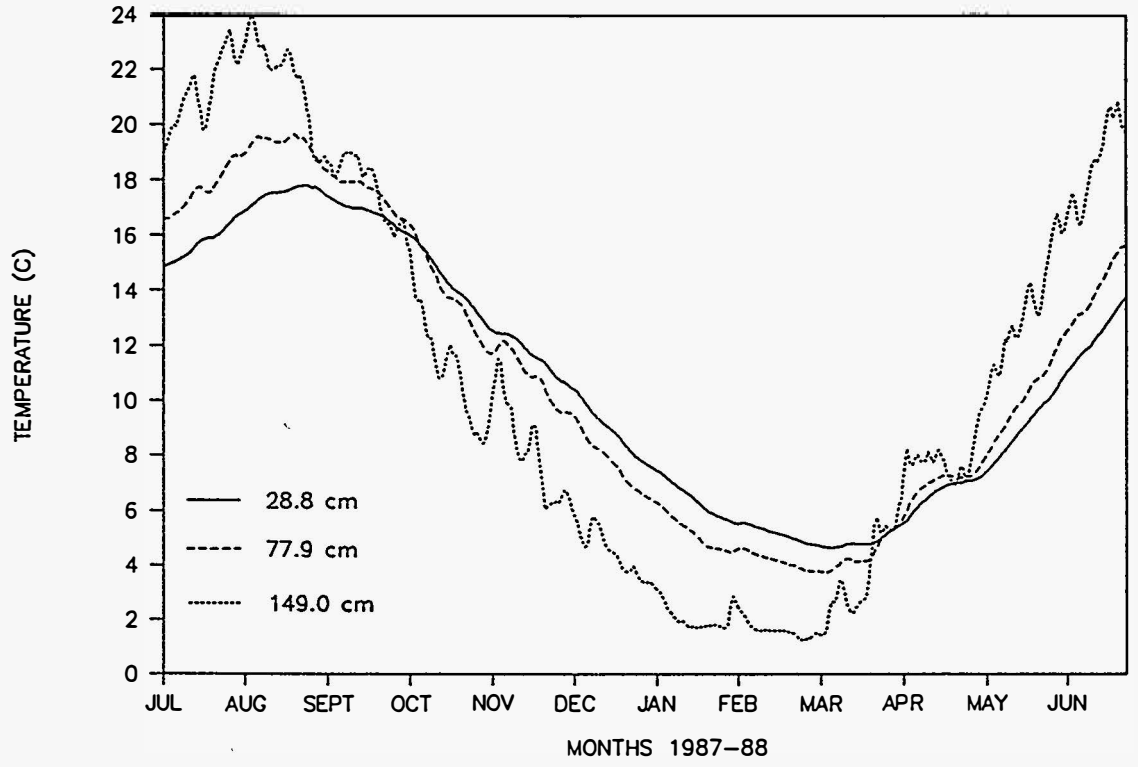

Figure B-18. ANL-E lysimeter 5 soil temperatures for 1987-88.

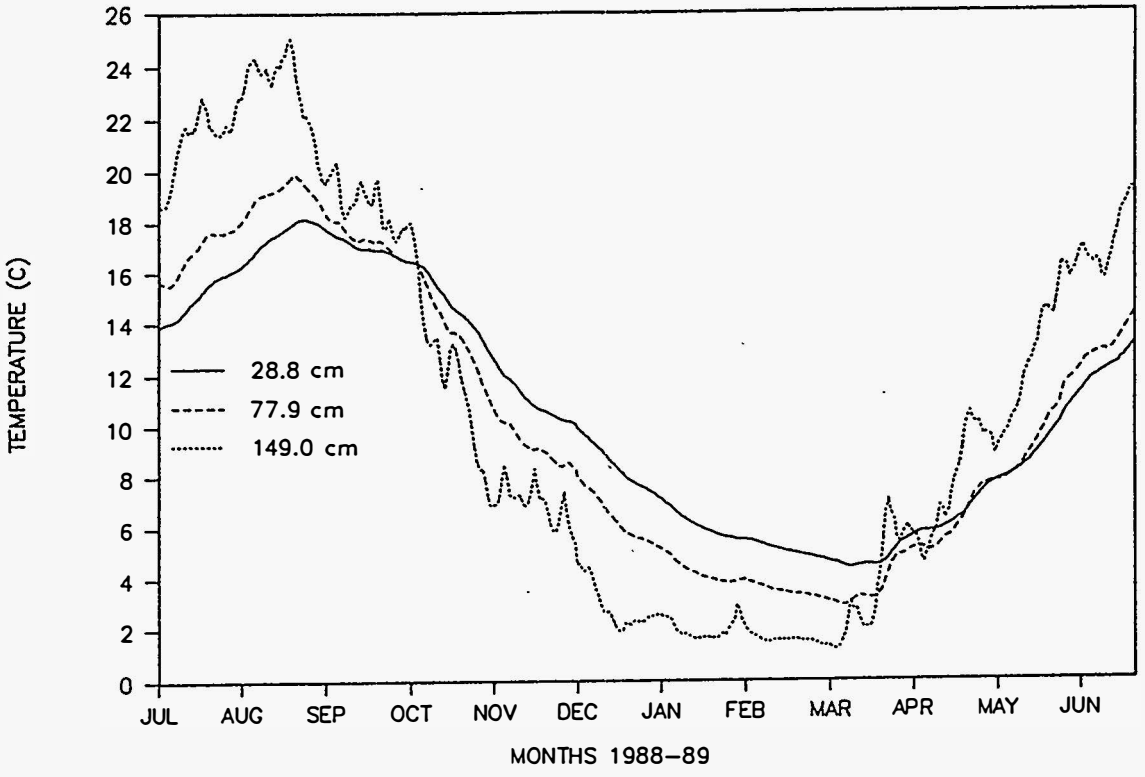

Figure B-19. ANL-E lysimeter 5 soil temperatures for 1988-89.

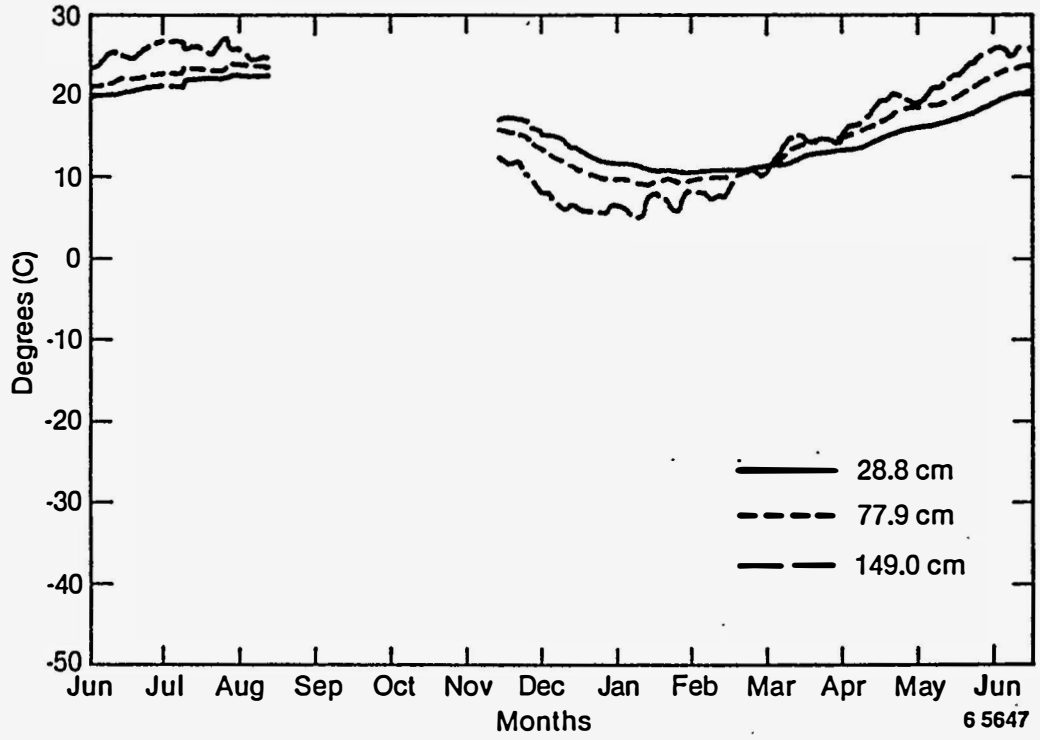

Figure B-20. ORNL lysimeter 1 soil temperatures for 1985-86.

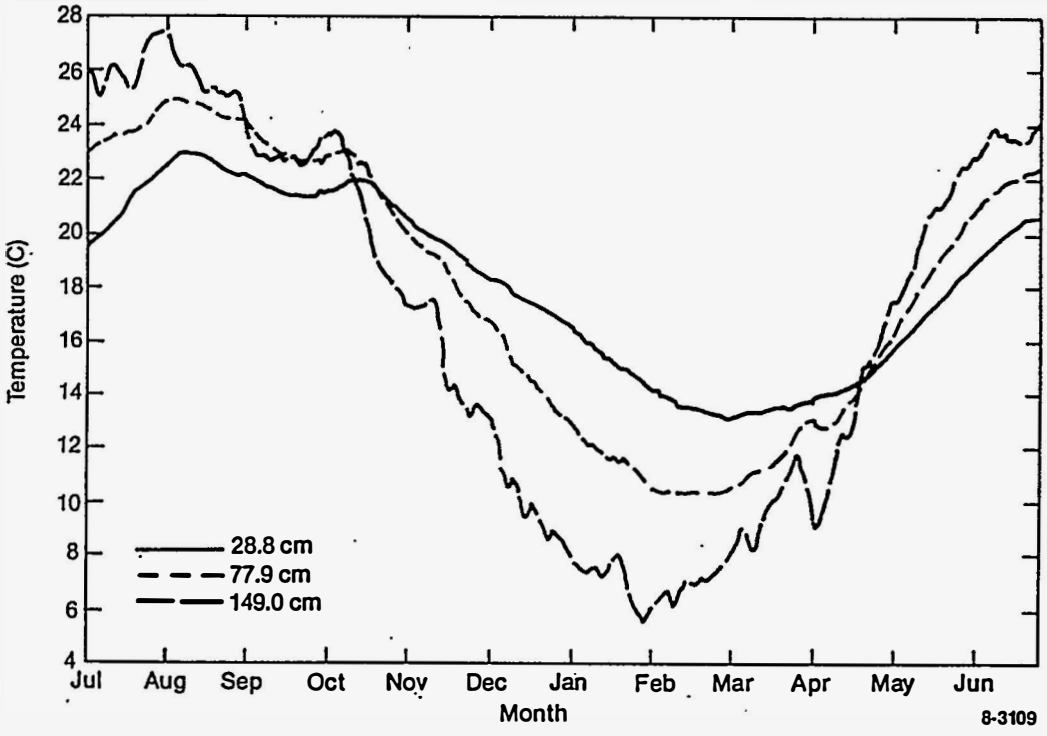

Figure B-21. ORNL lysimeter 1 soil temperatures for 1986-87.

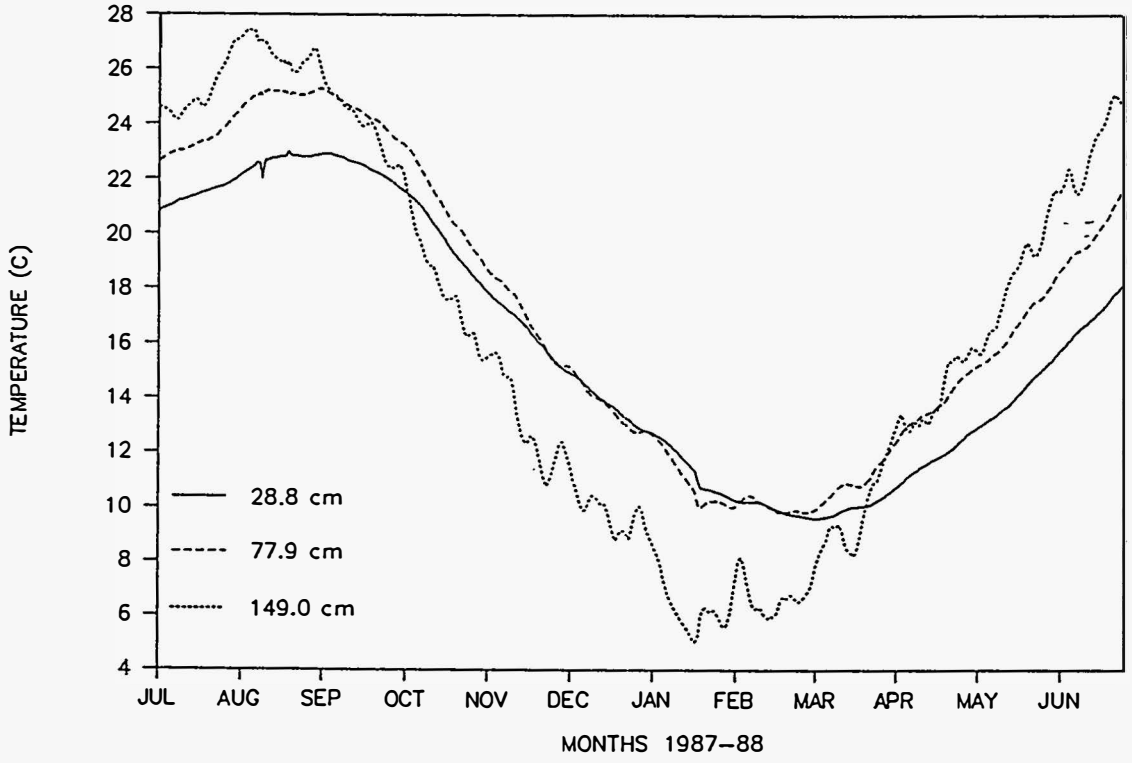

Figure B-22. ORNL lysimeter 1 soil temperatures for 1987-88.

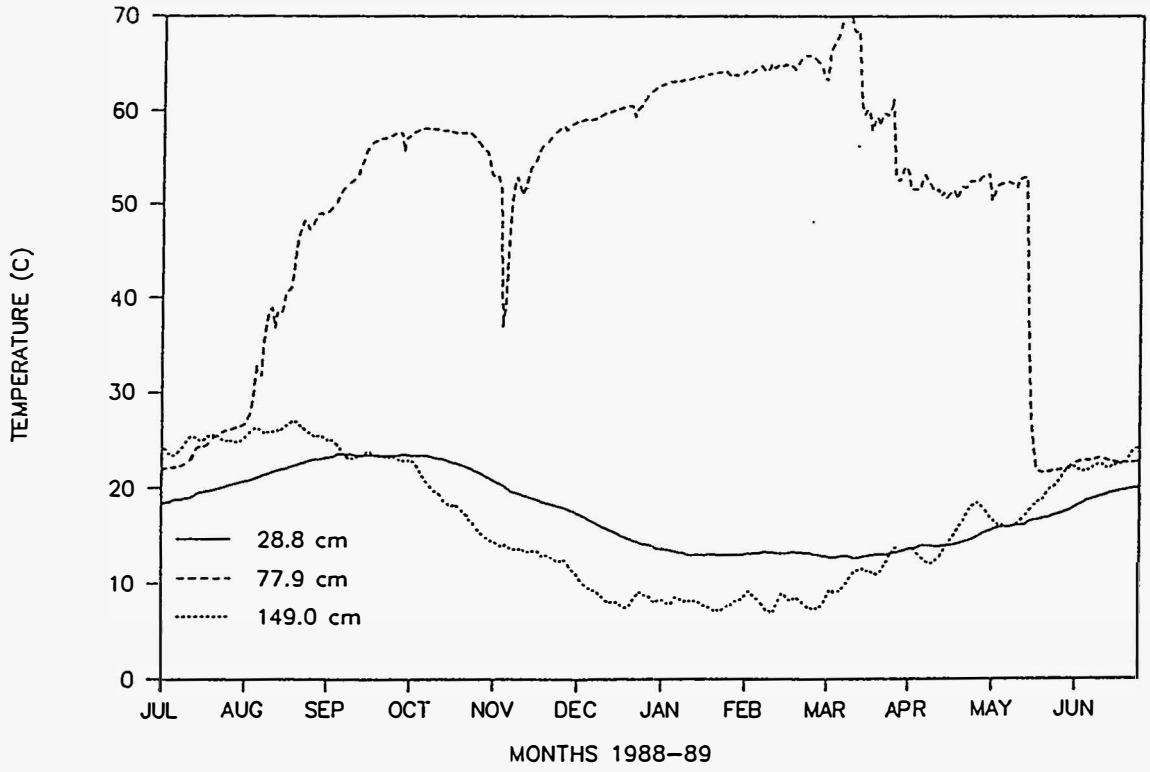

Figure B-23. ORNL lysimeter 1 soil temperatures for 1988-89.

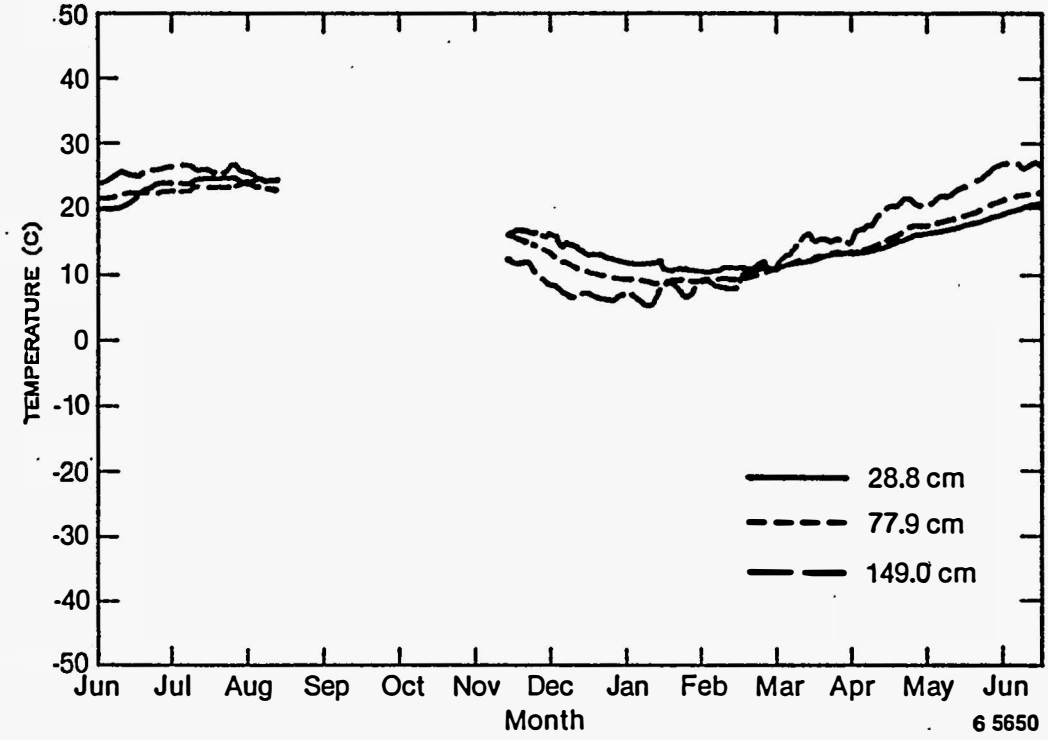

Figure B-24. ORNL lysimeter 2 soil temperatures for 1985-86.

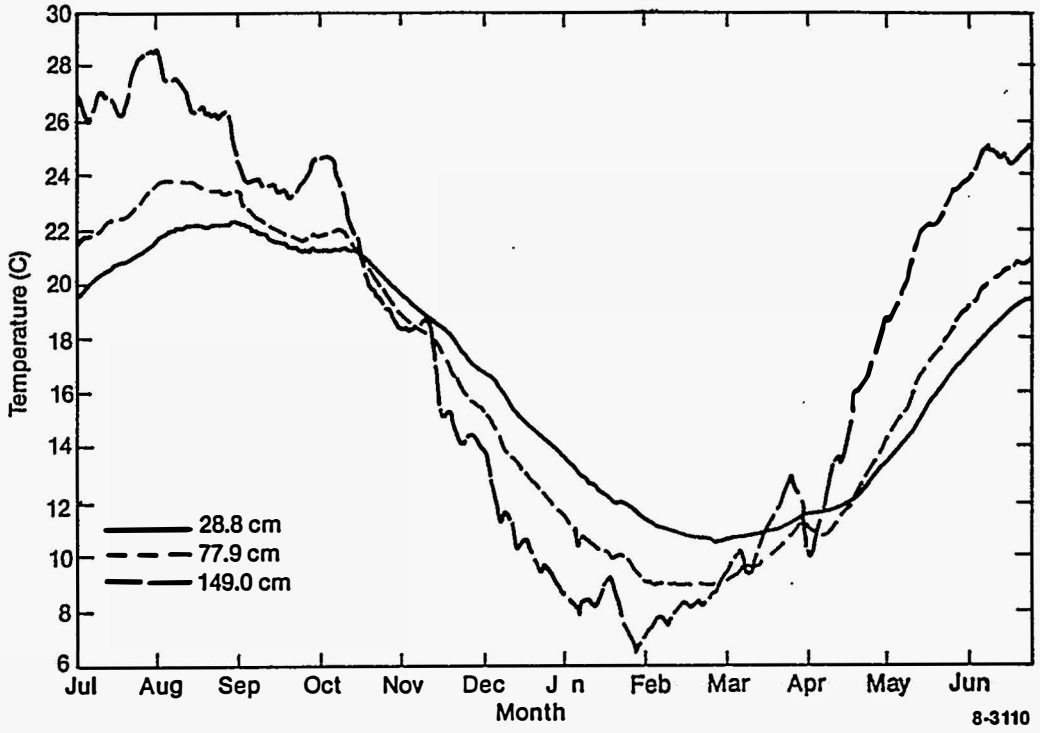

Figure B-25. ORNL lysimeter 2 soil temperatures for 1986-87.

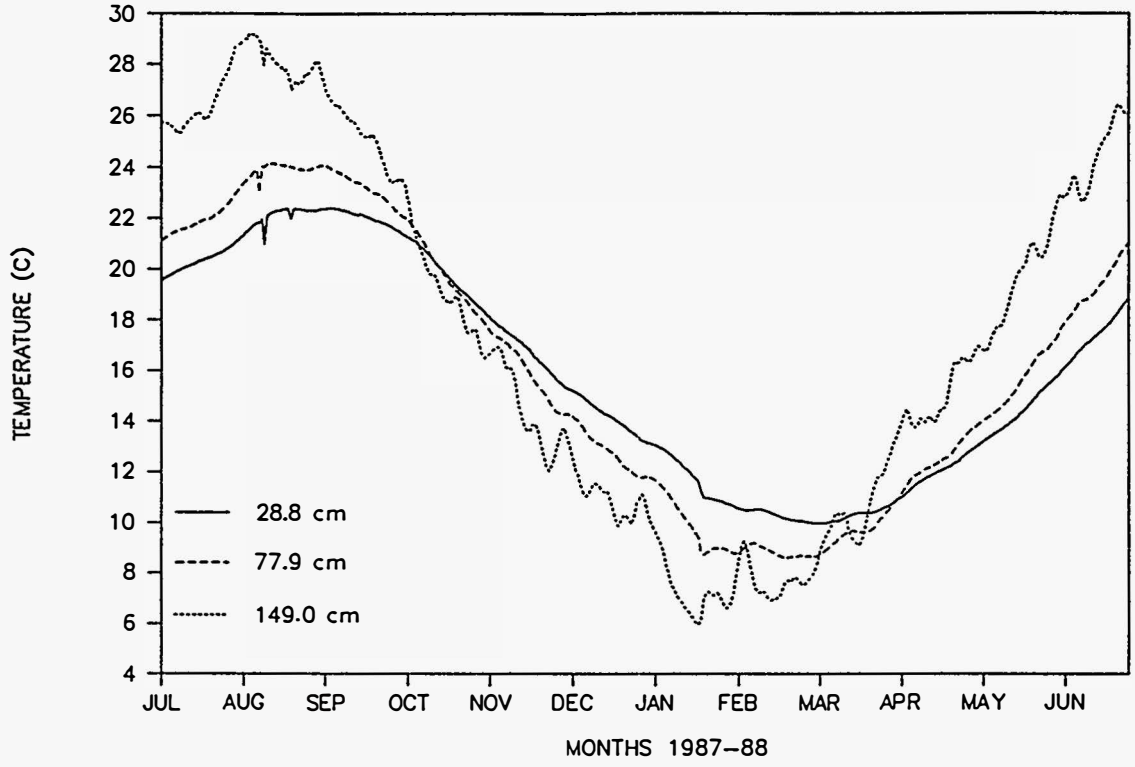

Figure B-26. ORNL lysimeter 2 soil temperatures for 1987-88.

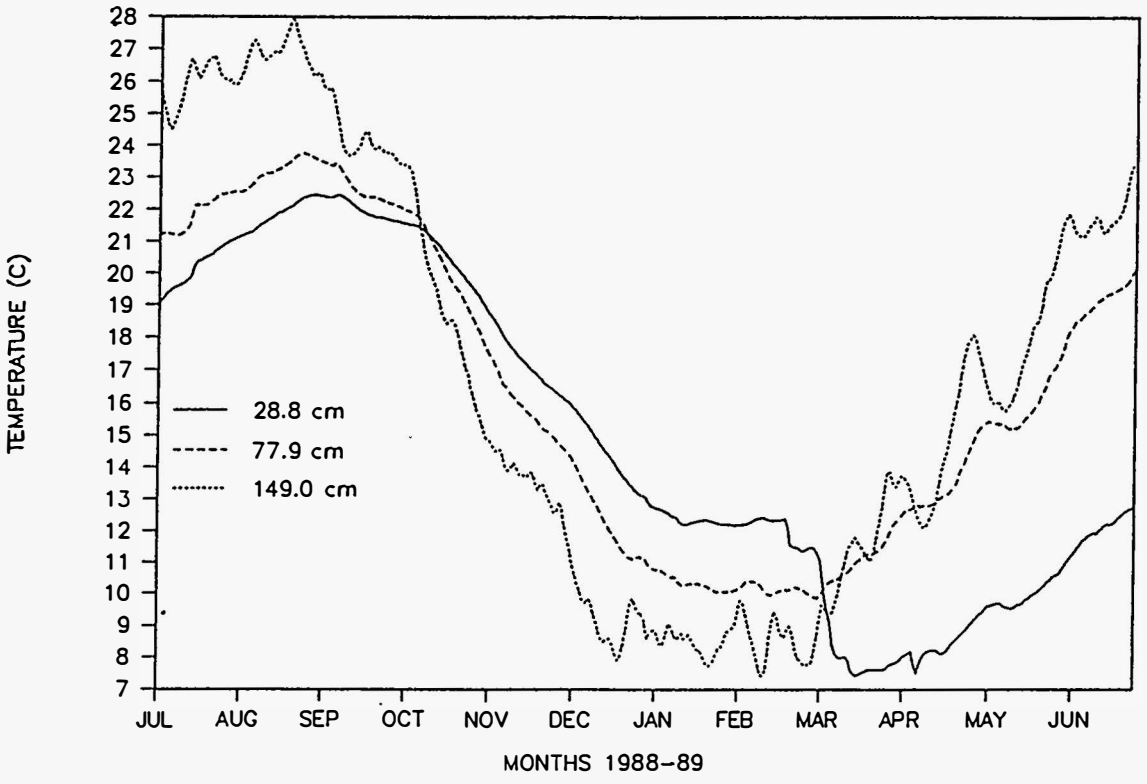

Figure B-27. ORNL lysimeter 2 soil temperatures for 1988-89.

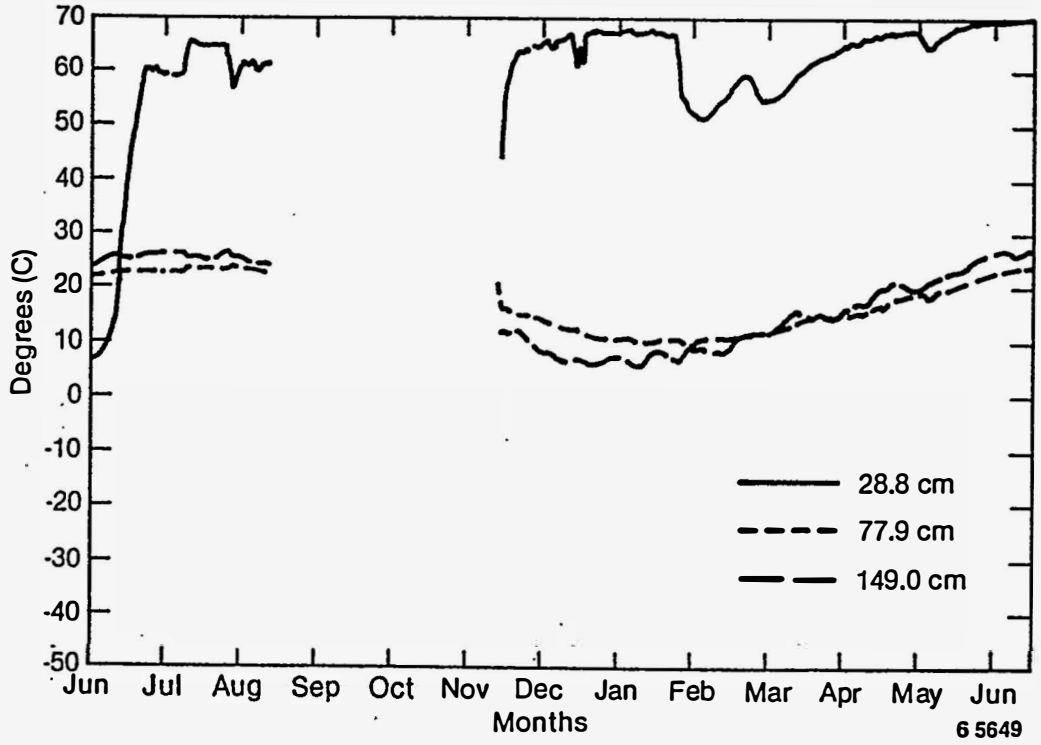

Figure B-28. ORNL lysimeter 3 soil temperatures for 1985-86.

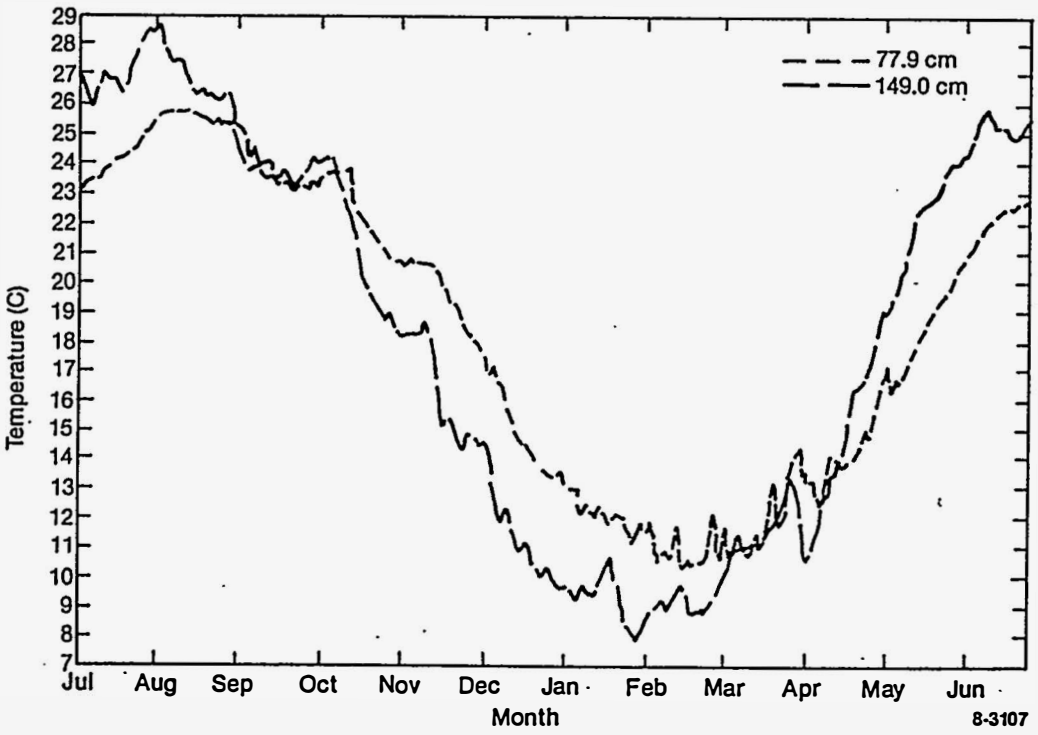

Figure B-29. ORNL lysimeter 3 soil temperatures for 1986-87.

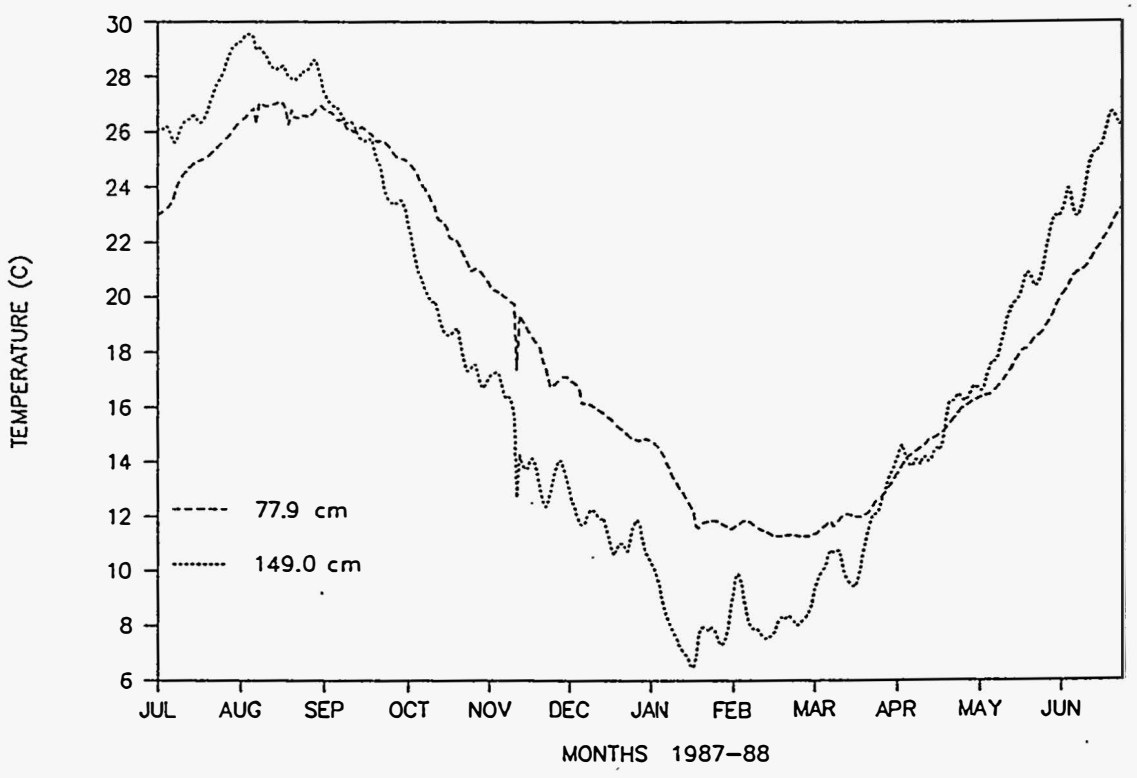

Figure B-30. ORNL lysimeter 3 soil temperatures for 1987-88.

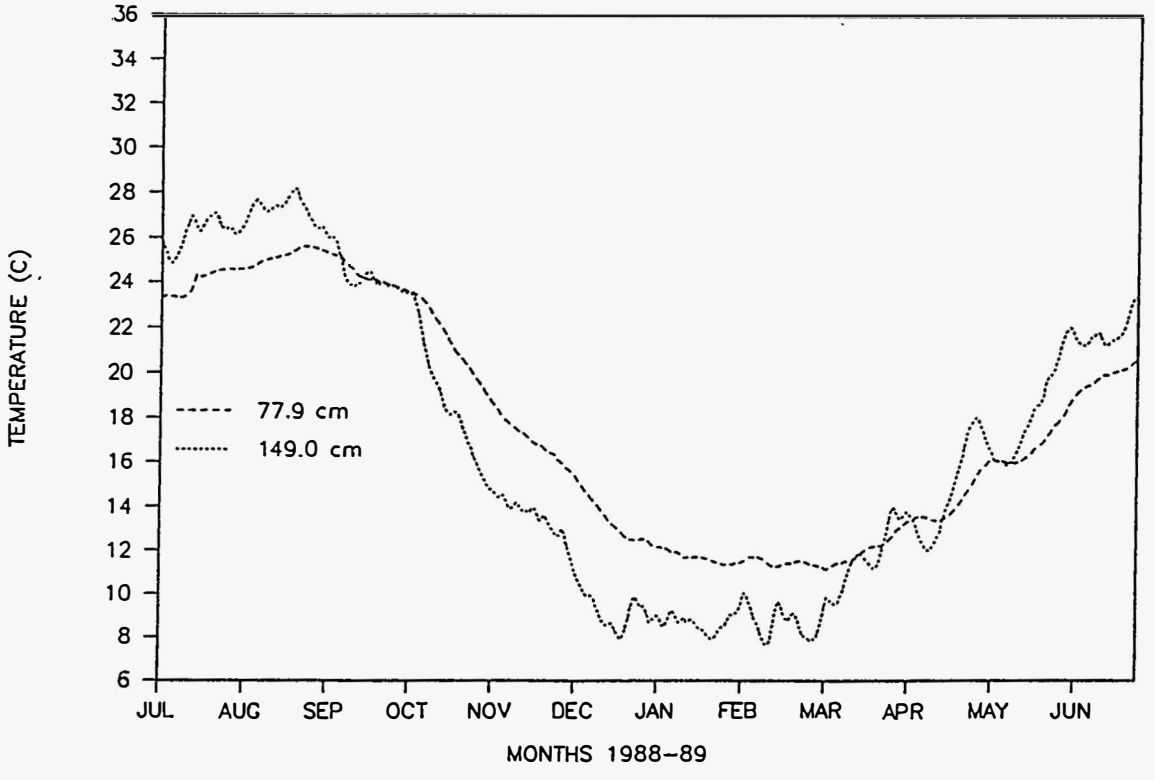

Figure B-31. ORNL lysimeter 3 soil temperatures for 1988-89.

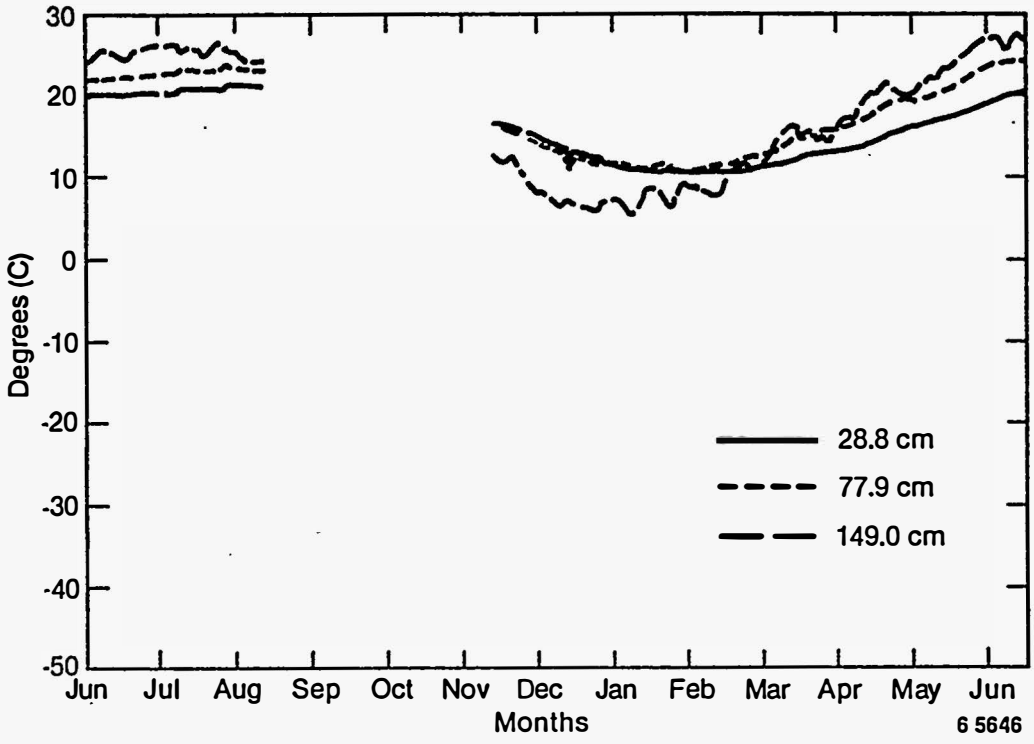

Figure B-32. ORNL lysimeter 4 soil temperatures for 1985-86.

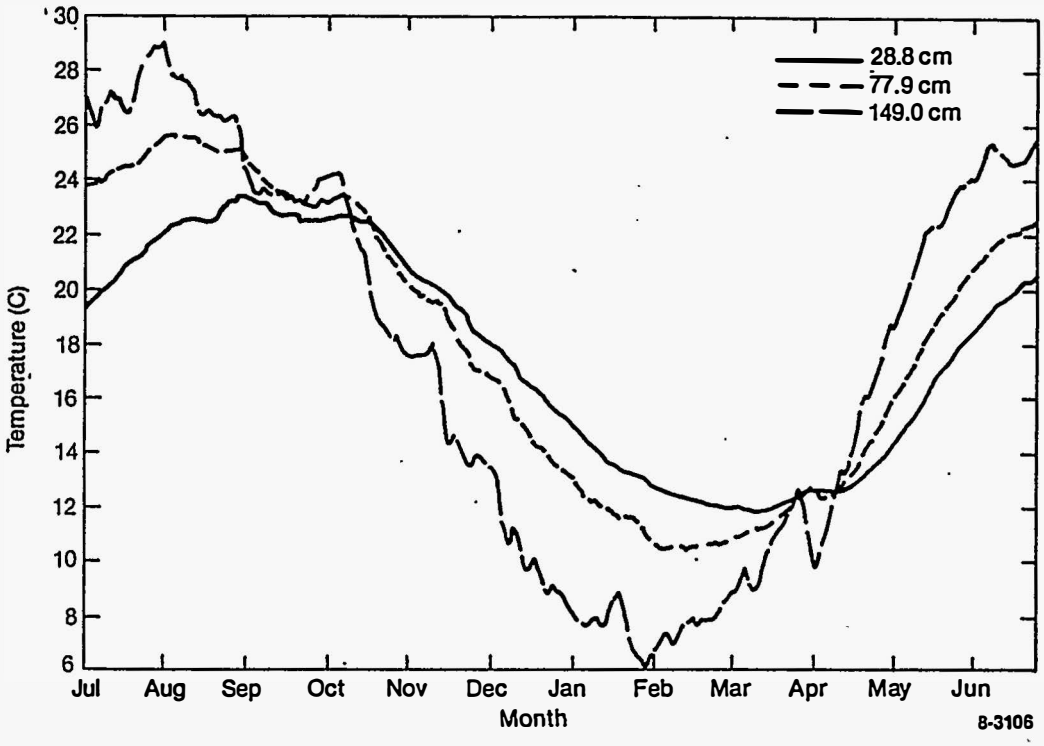

Figure B-33. ORNL lysimeter 4 soil temperatures for 1986-87.

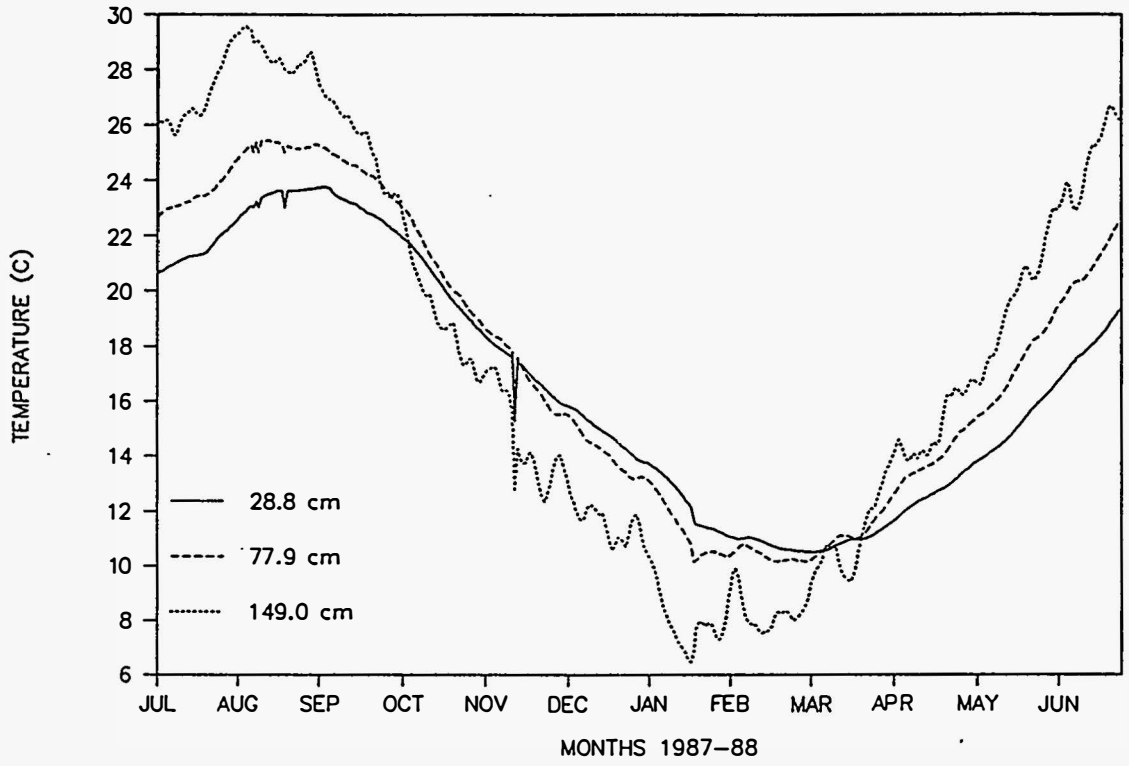

Figure B-34. ORNL lysimeter 4 soil temperatures for 1987-88.

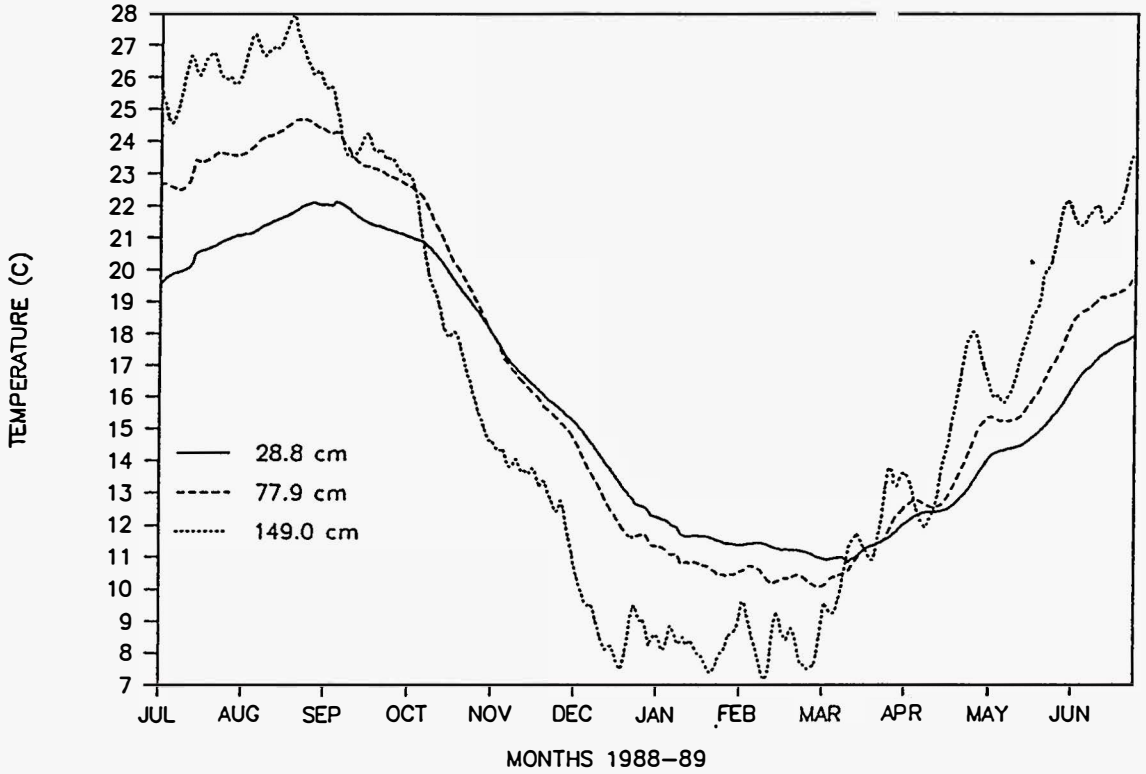

Figure B-35. ORNL lysimeter 4 soil temperatures for 1988-89.

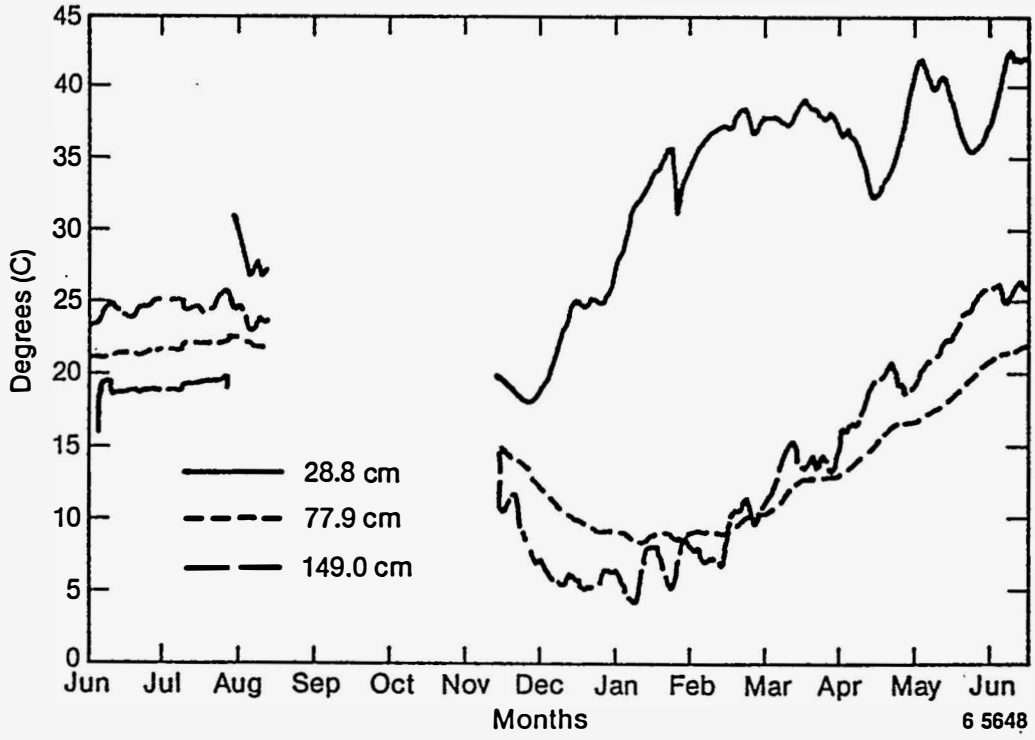

Figure B-36. ORNL lysimeter 5 soil temperatures for 1985-86.

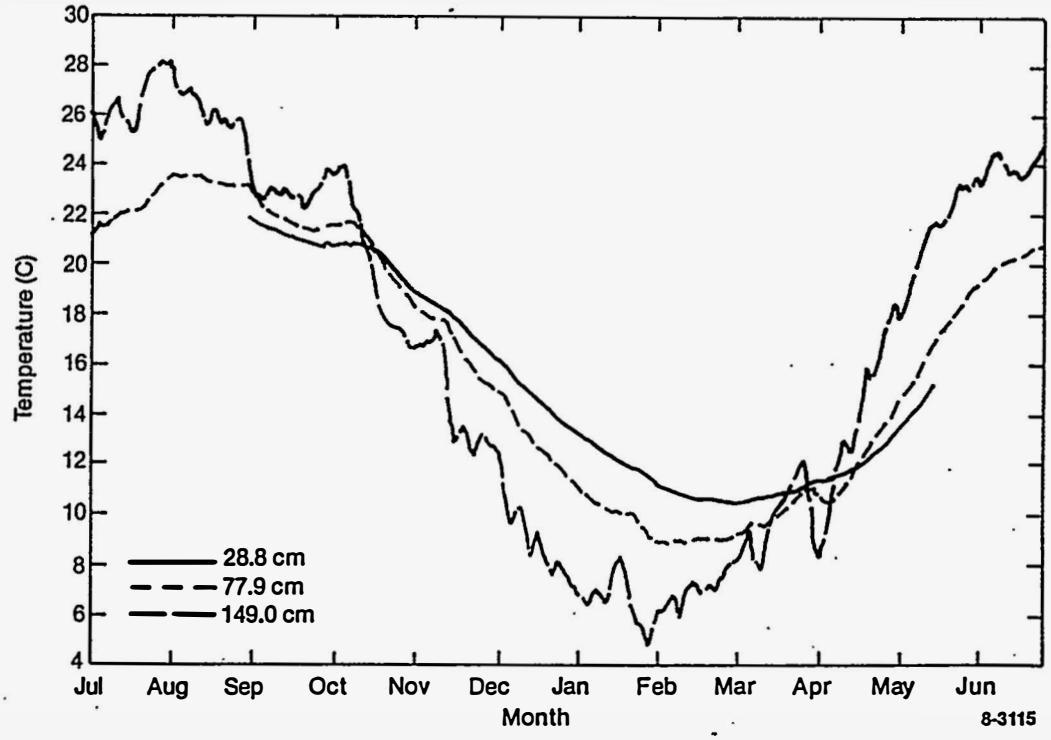

Figure B-37. ORNL lysimeter 5 soil temperatures for 1986-87.

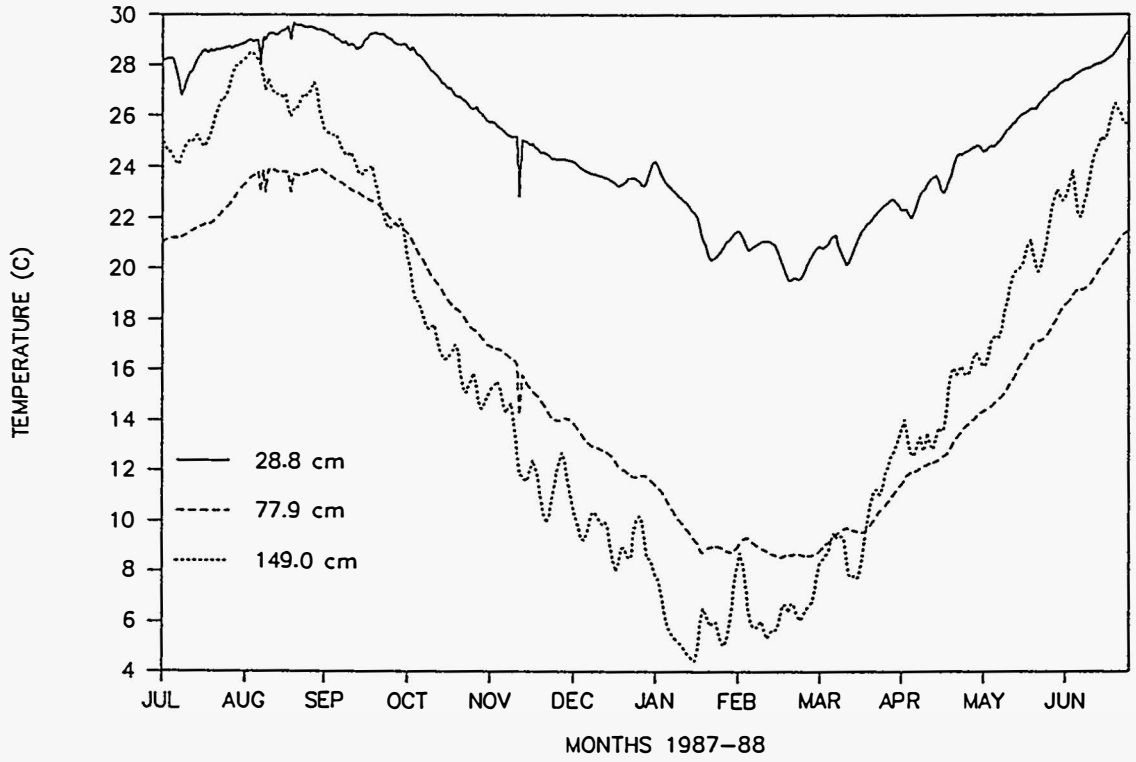

Figure B-38. ORNL lysimeter 5 soil temperatures for 1987-88.

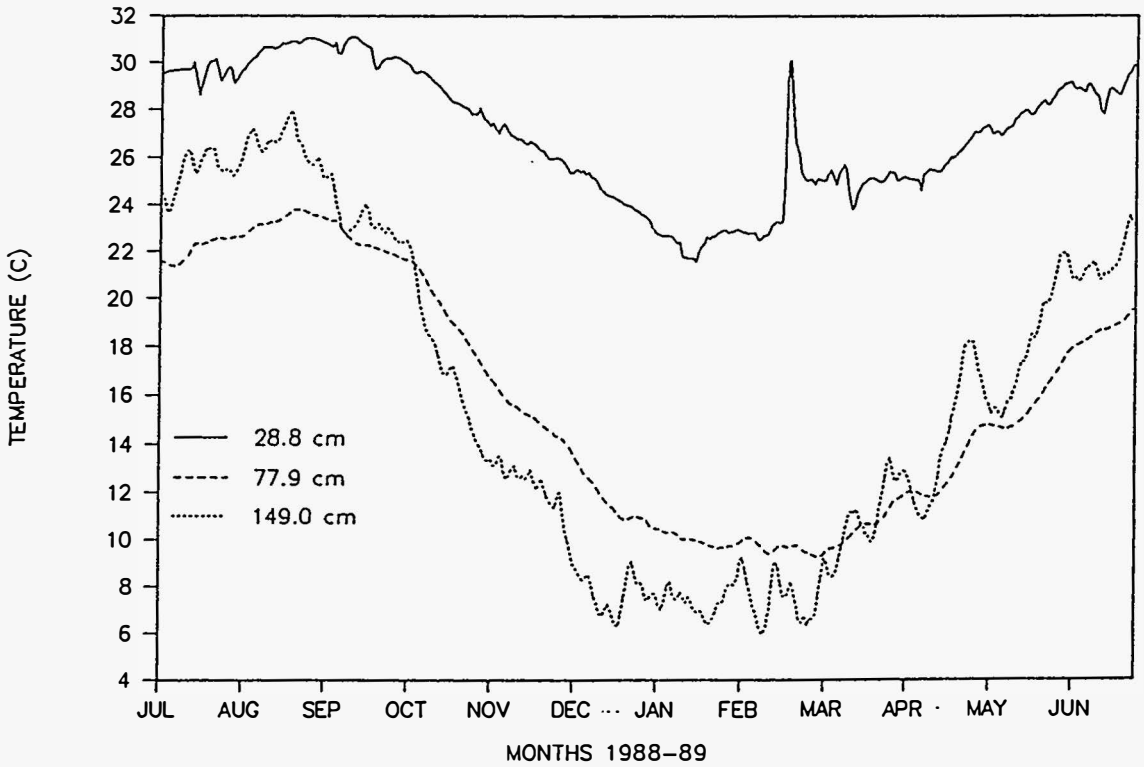

Figure B-39. ORNL lysimeter 5 soil temperatures for 1988-89.

Appendix C

Soil Moisture Data—Resistance Probes

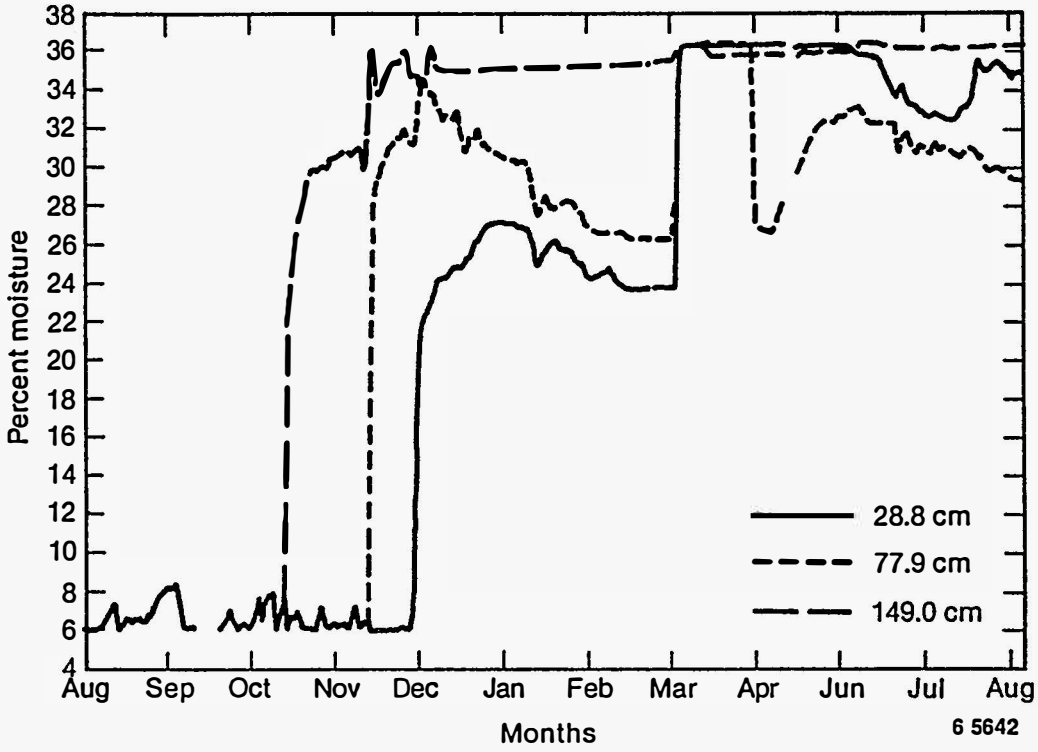

Figure C-1. ANL-E lysimeter 1 soil moisture for 1985-86.

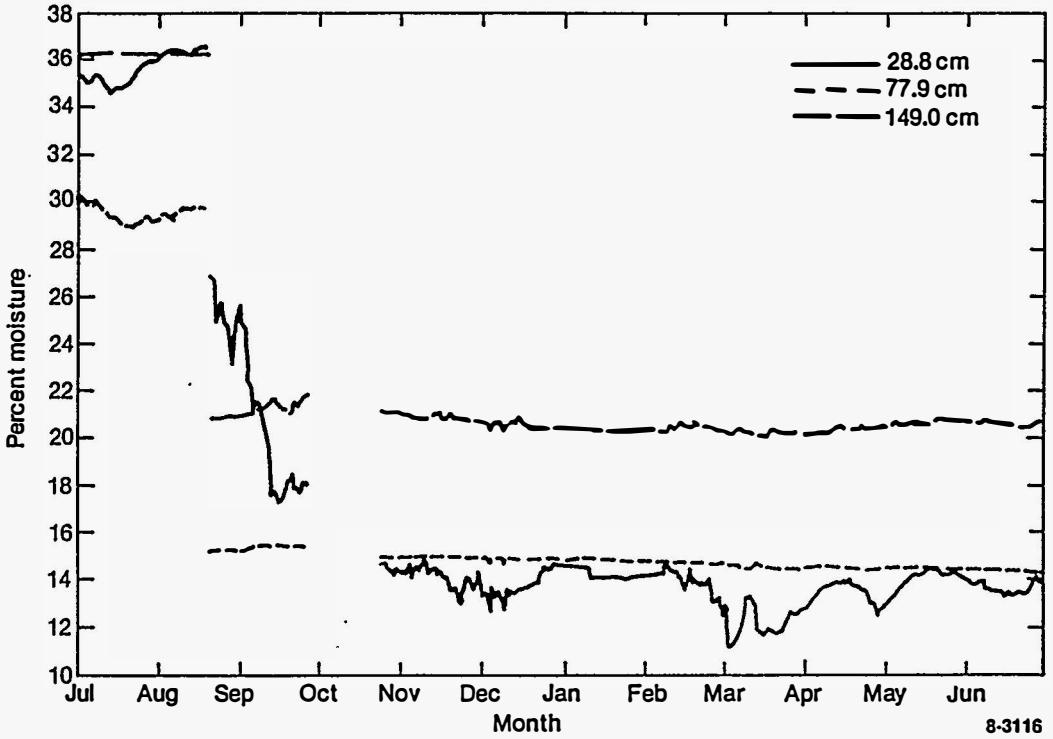

Figure C-2. ANL-E lysimeter 1 soil moisture for 1986-87.

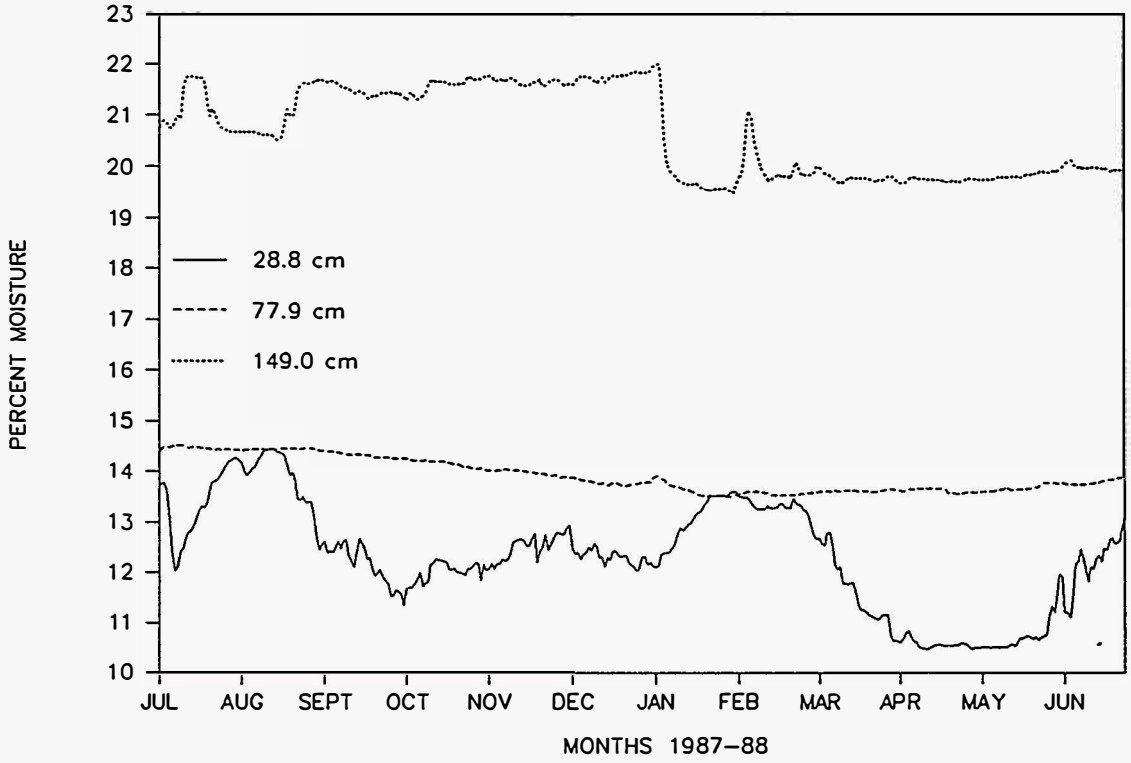

Figure C-3. ANL-E lysimeter 1 soil moisture for 1987-88.

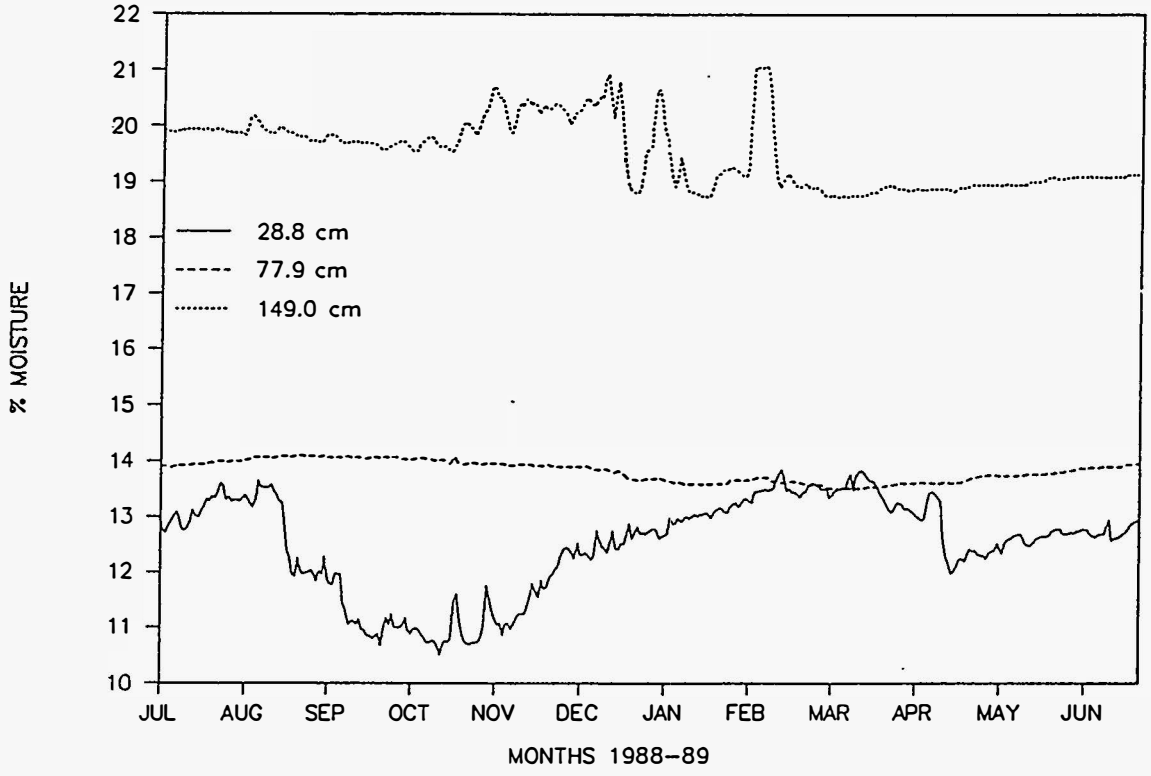

Figure C-4. ANL-E lysimeter 1 soil moisture for 1988-89.

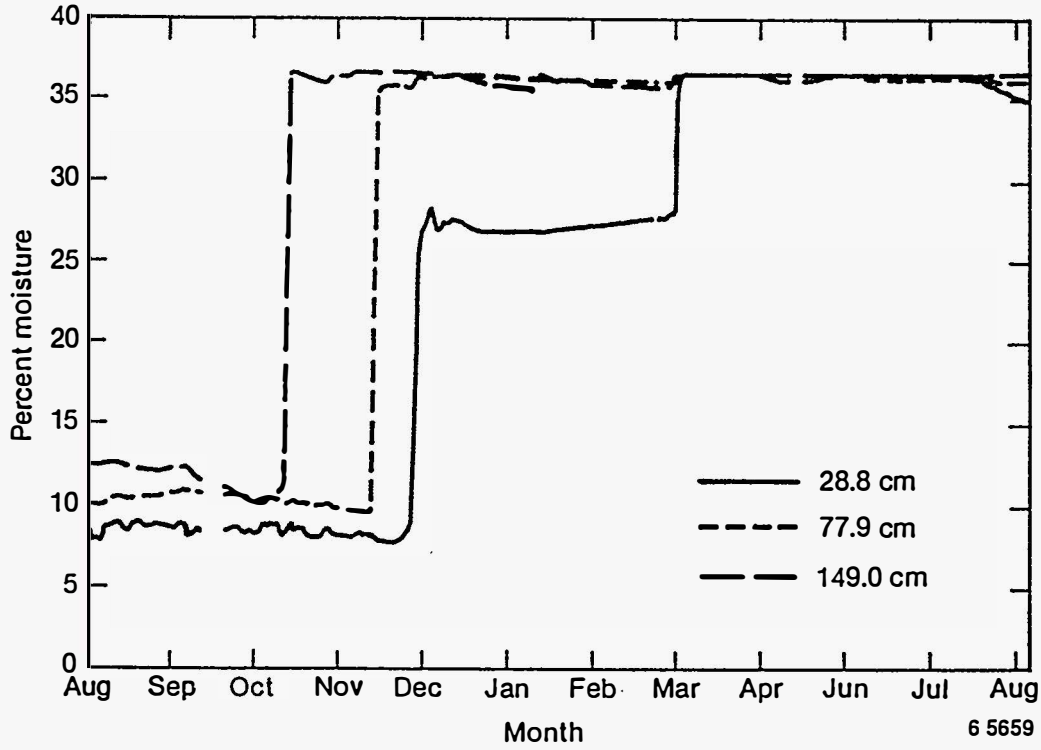

Figure C-5. ANL-E lysimeter 2 soil moisture for 1985-86.

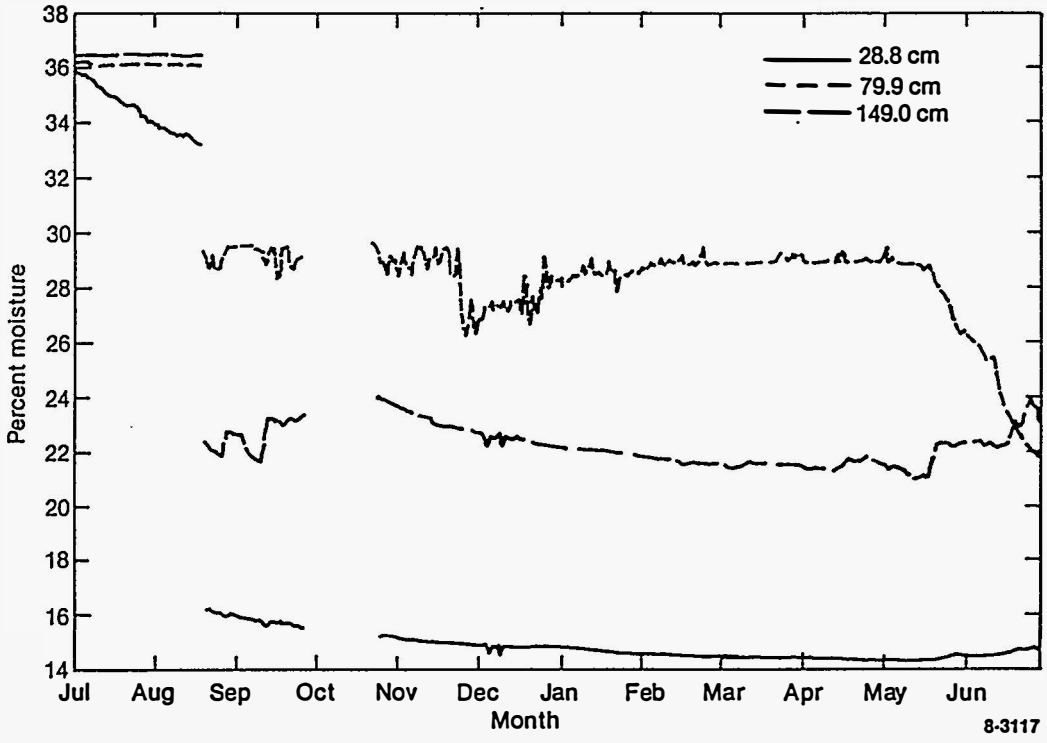

Figure C-6. ANL-E lysimeter 2 soil moisture for 1986-87.

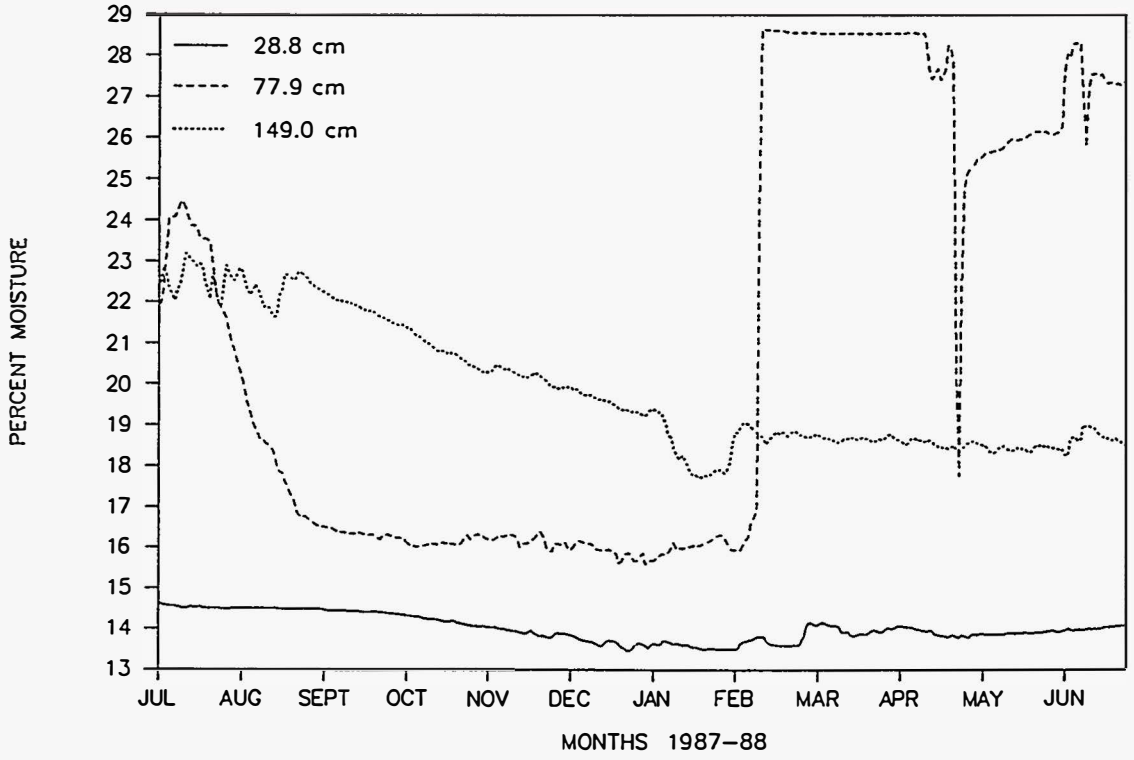

Figure C-7. ANL-E lysimeter 2 soil moisture for 1987-88.

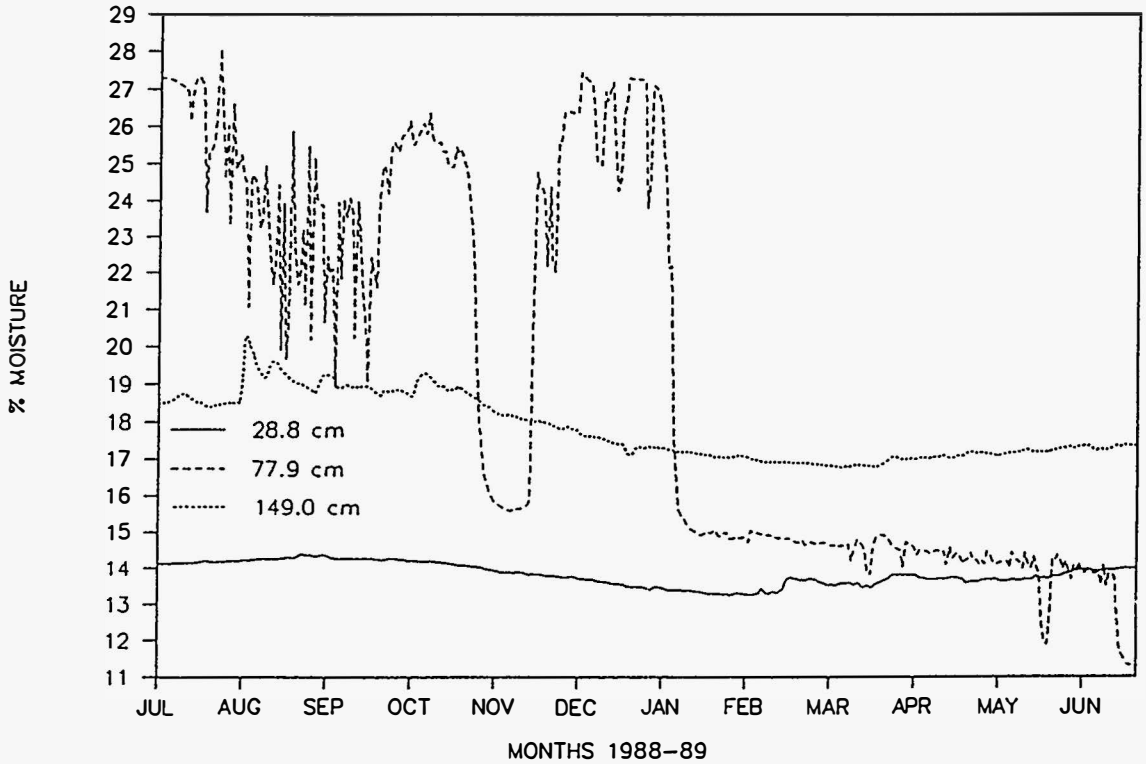

Figure C-8. ANL-E lysimeter 2 soil moisture for 1988-89.

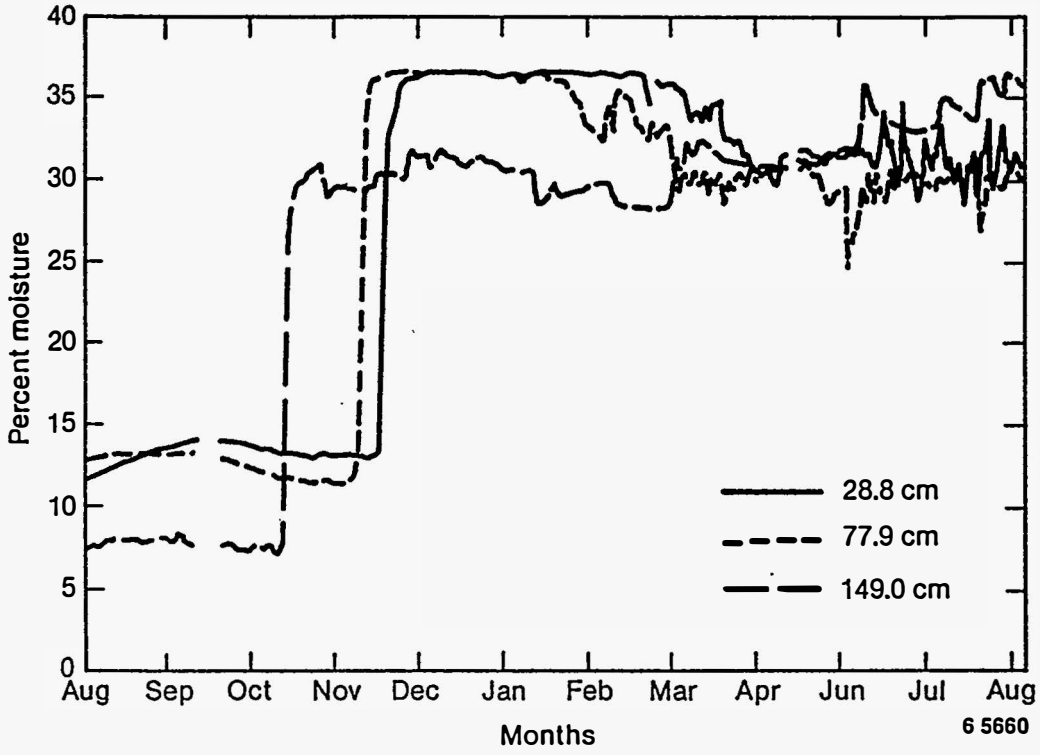

Figure C-9. ANL-E lysimeter 3 soil moisture for 1985-86.

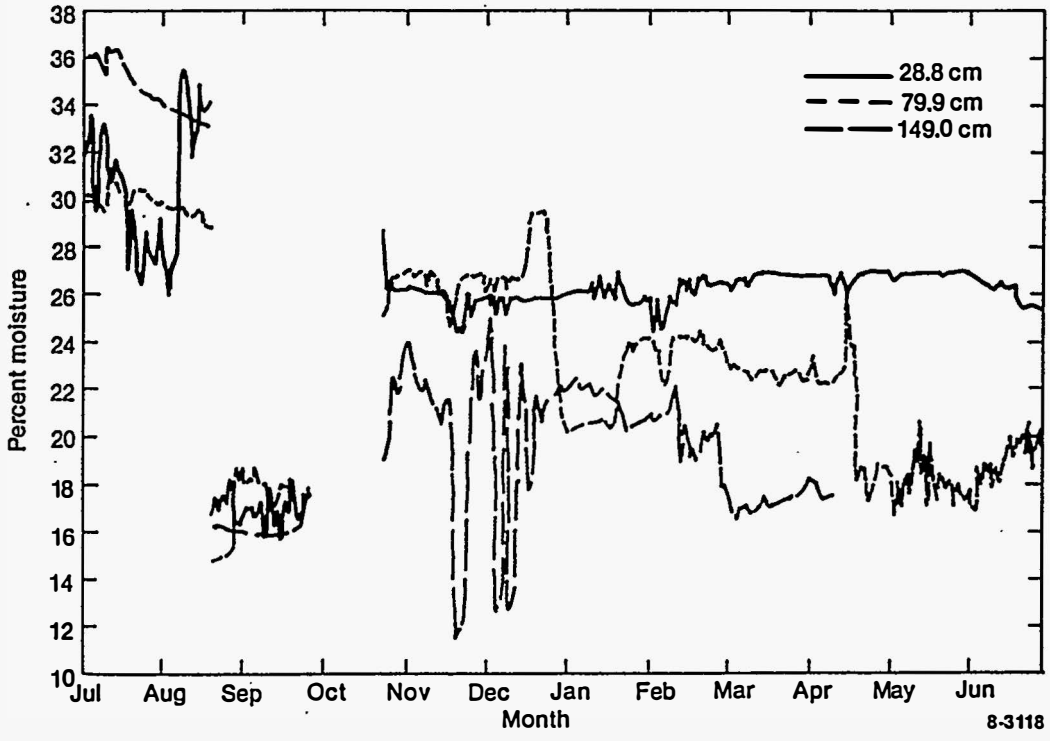

Figure C-10. ANL-E lysimeter 3 soil moisture for 1986-87.

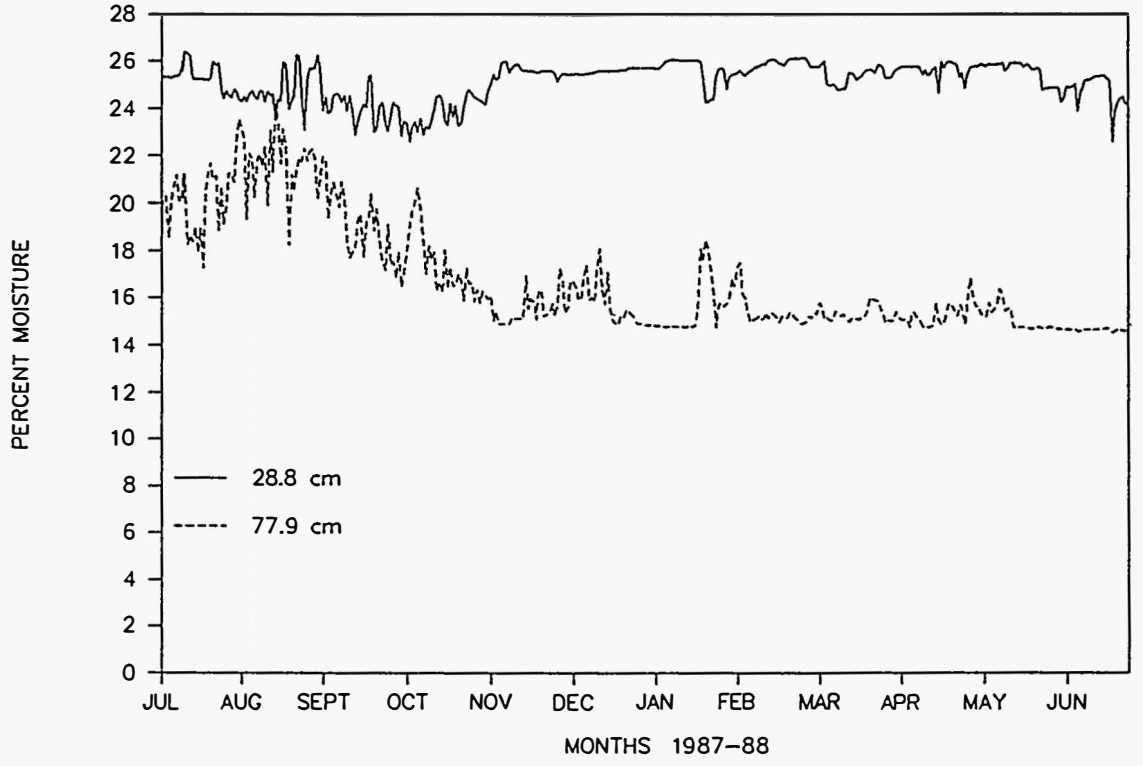

Figure C-11. ANL-E lysimeter 3 soil moisture for 1987-88.

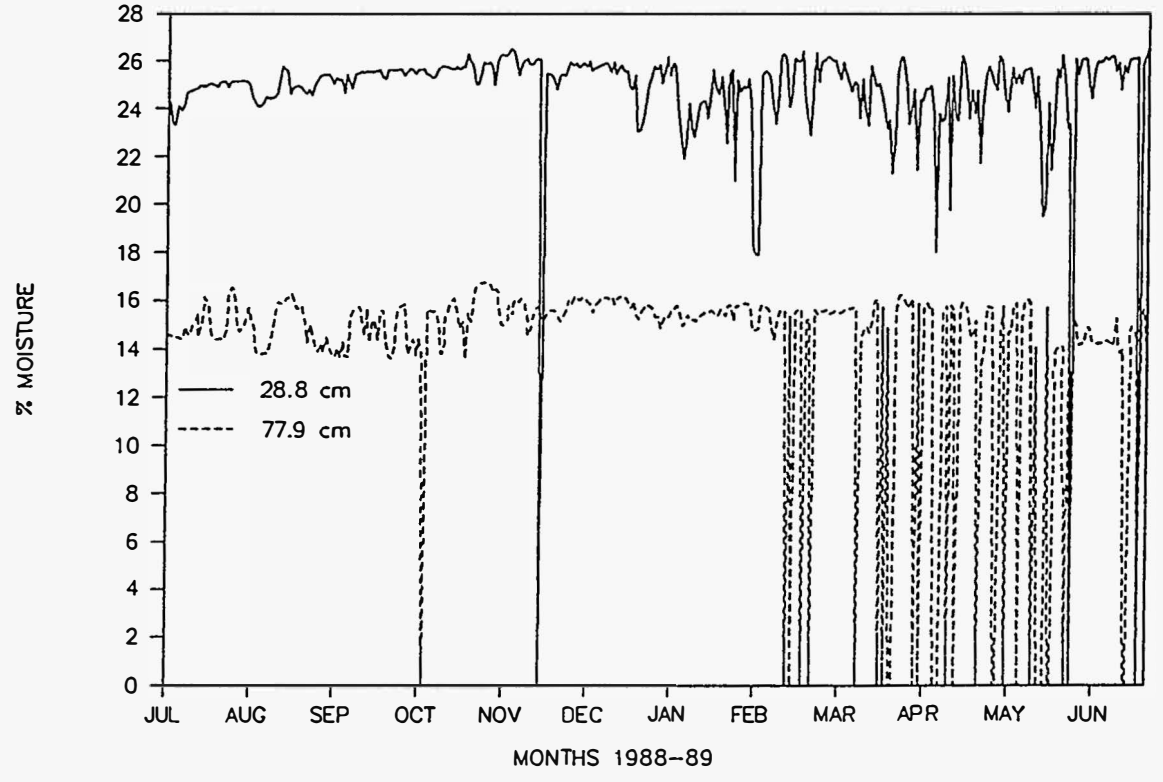

Figure C-12. ANL-E lysimeter 3 soil moisture for 1988-89.

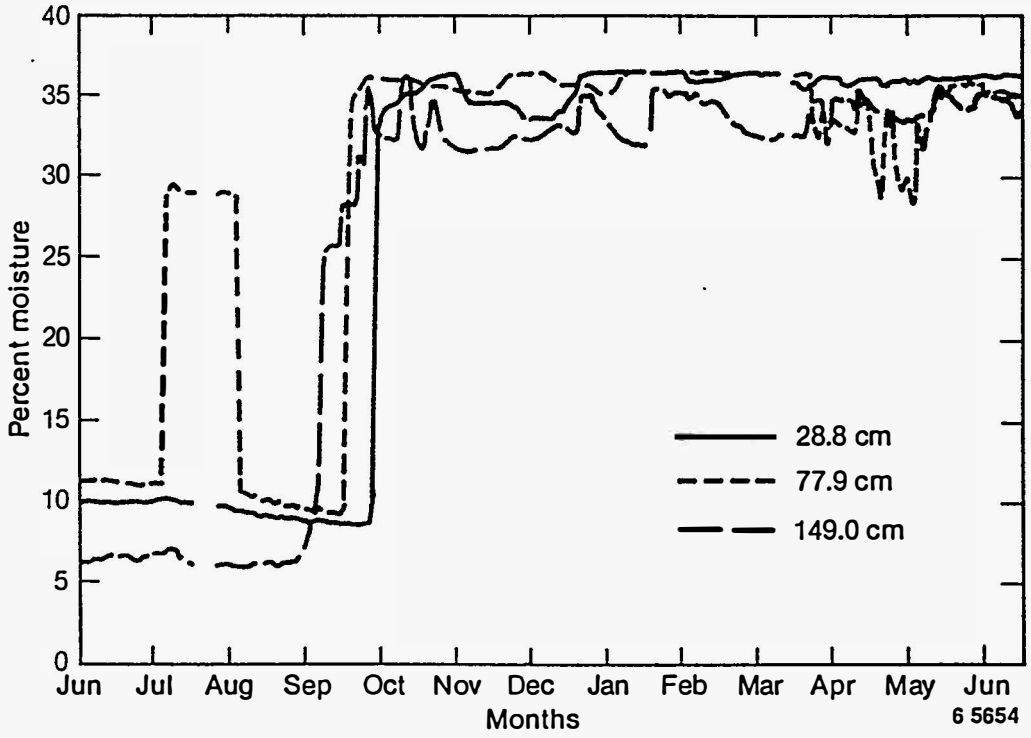

Figure C-13. ANL-E lysimeter 4 soil moisture for 1985-86.

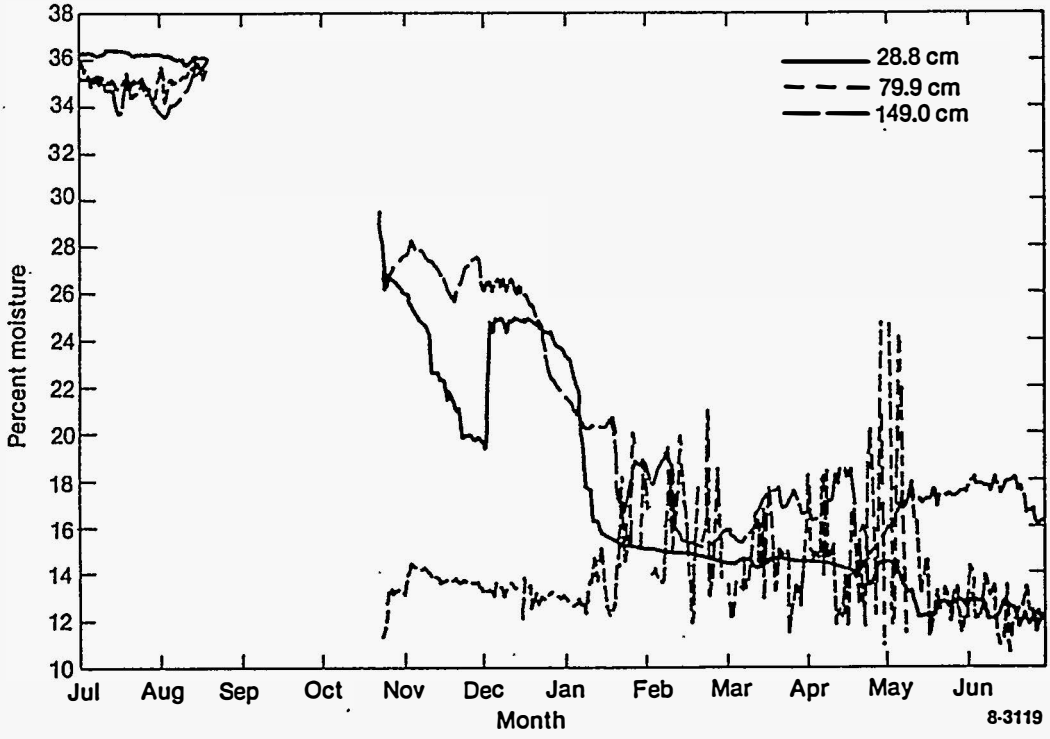

Figure C-14. ANL-E lysimeter 4 soil moisture for 1986-87.

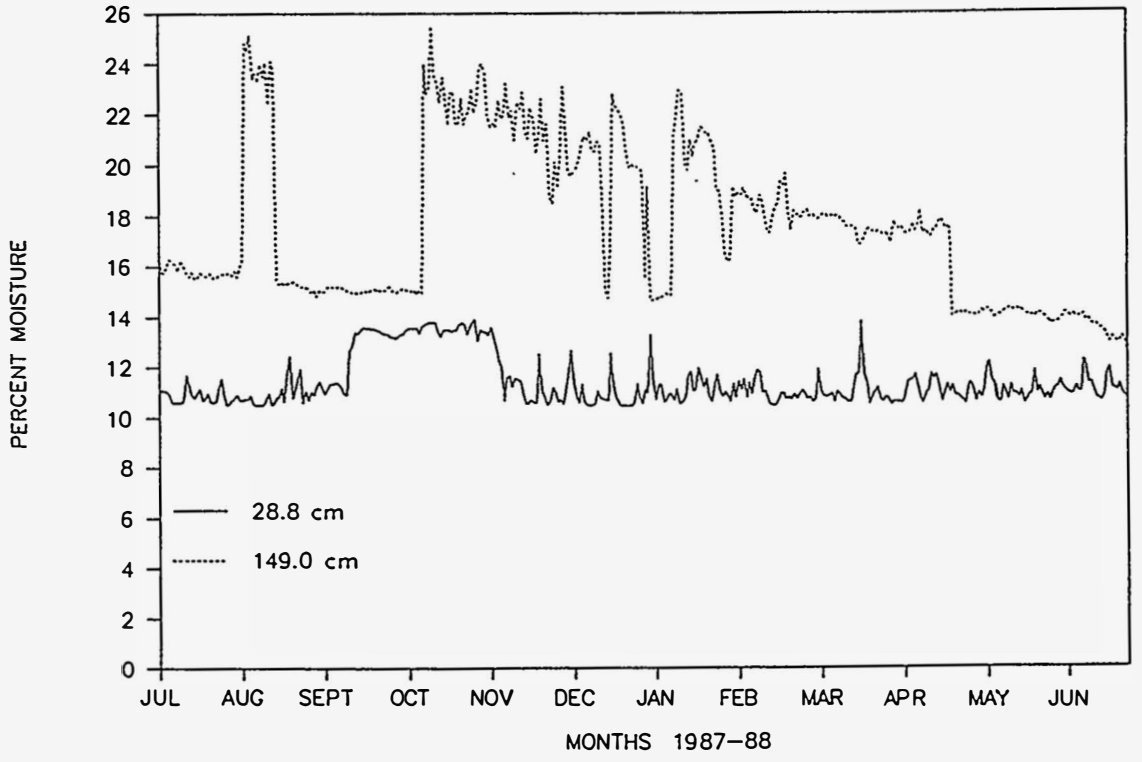

Figure C-15. ANL-E lysimeter 4 soil moisture for 1987-88.

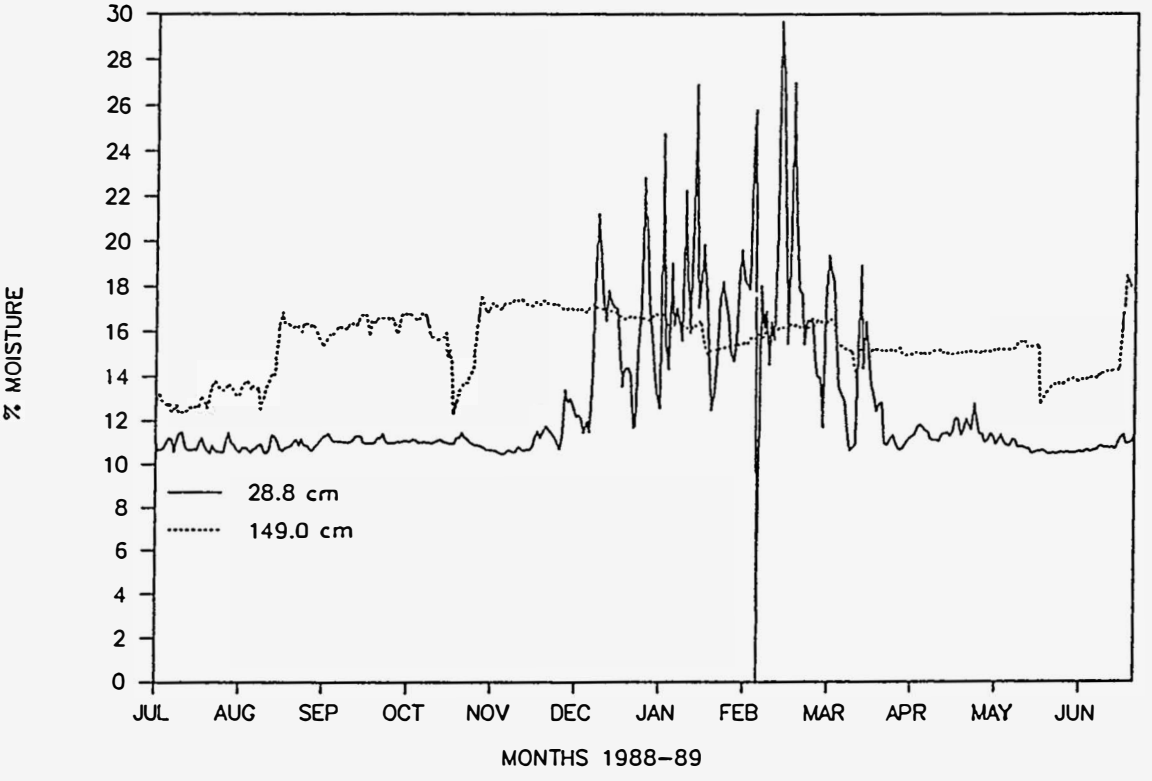

Figure C-16. ANL-E lysimeter 4 soil moisture for 1988-89.

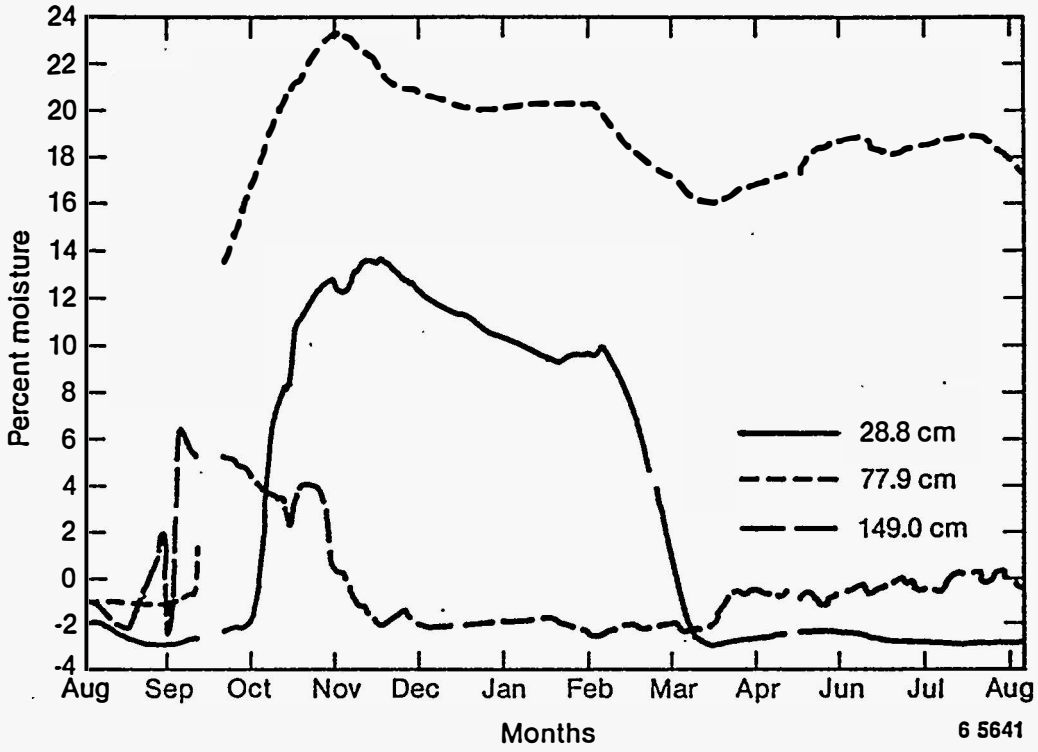

Figure C-17. ANL-E lysimeter 5 soil moisture for 1985-86.

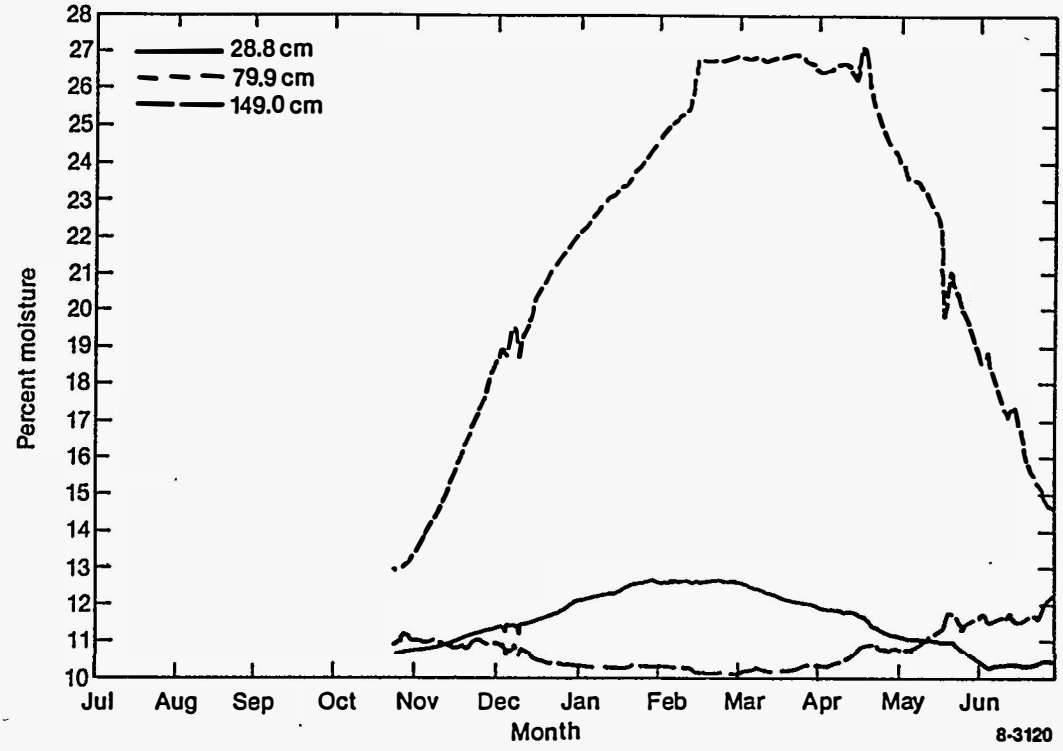

Figure C-18. ANL-E lysimeter 5 soil moisture for 1986-87.

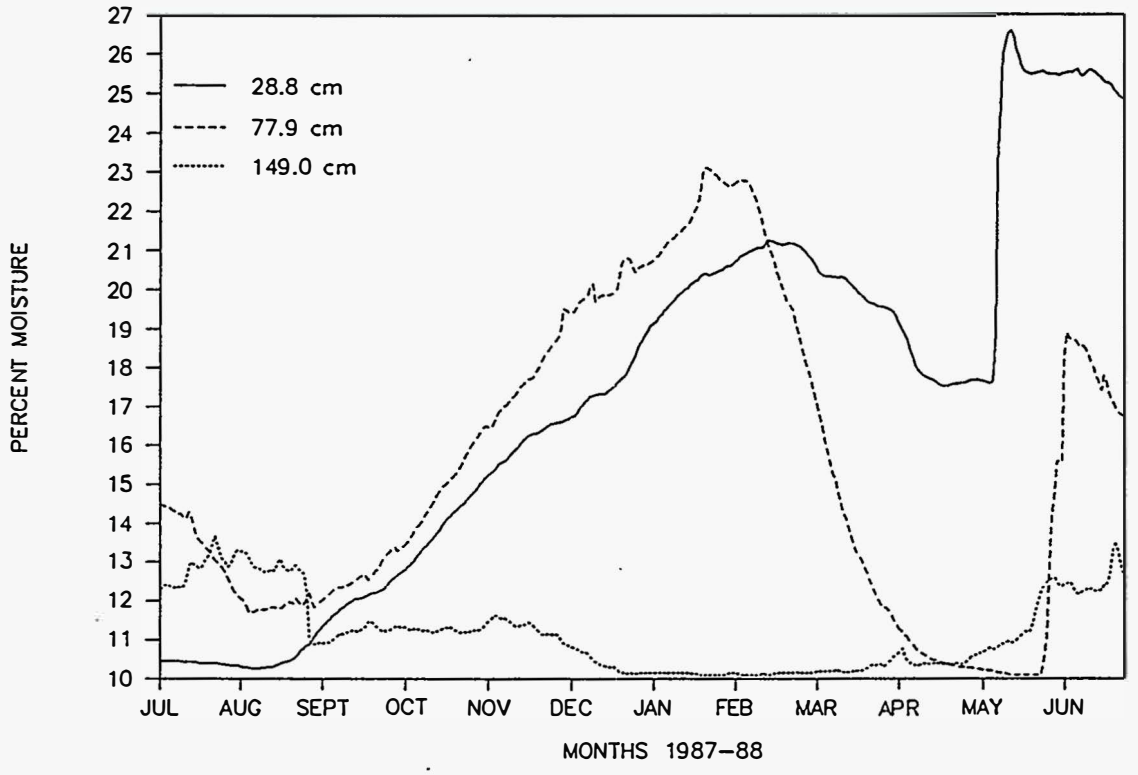

Figure C-19. ANL-E lysimeter 5 soil moisture for 1987-88.

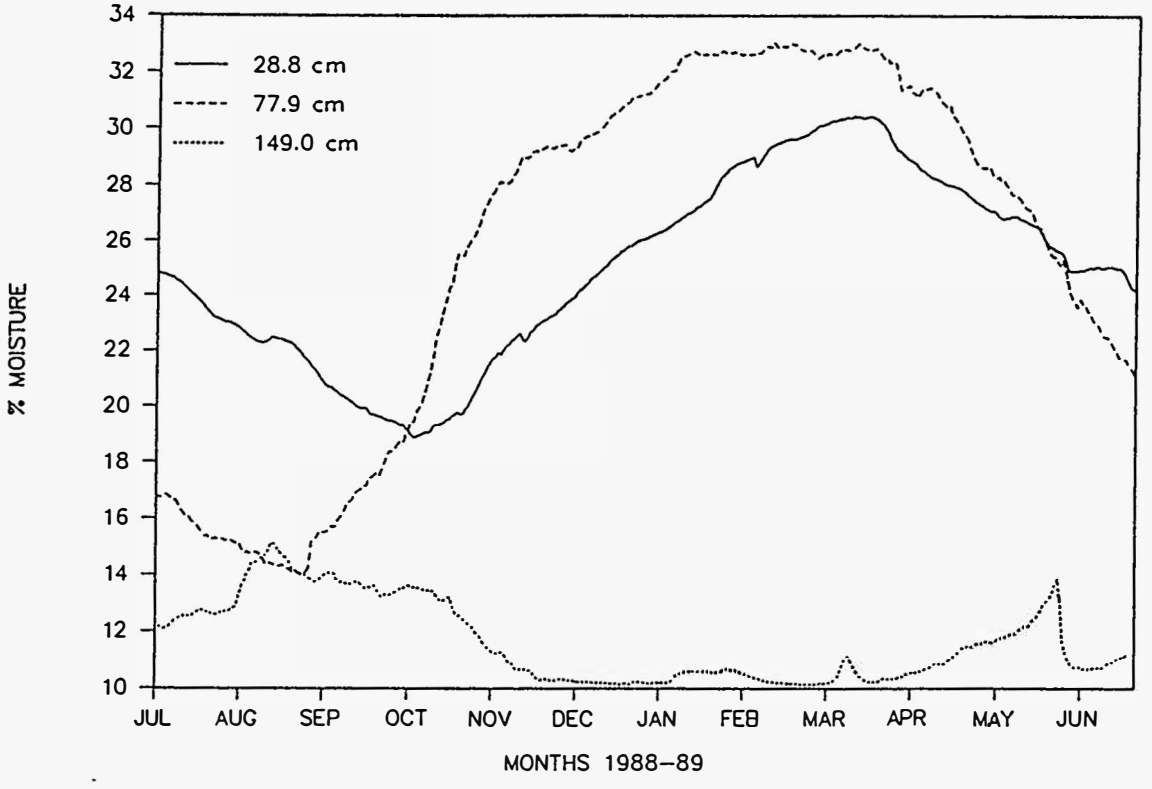

Figure C-20. ANL-E lysimeter 5 soil moisture for 1988-89.

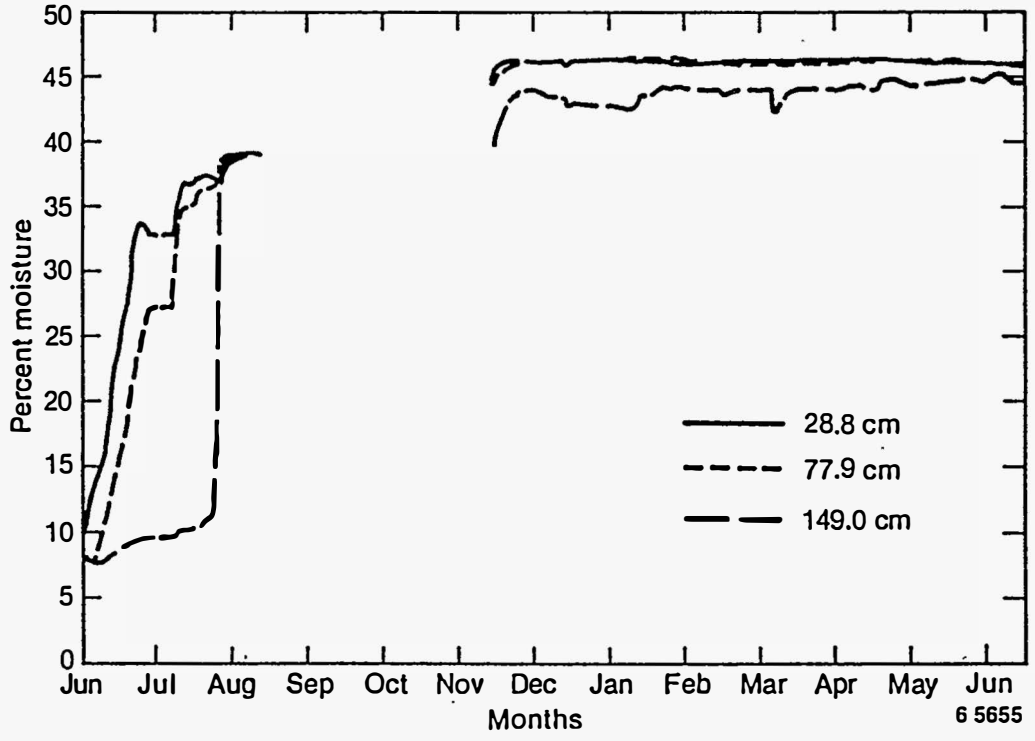

Figure C-21. ORNL lysimeter 1 soil moisture for 1985-86.

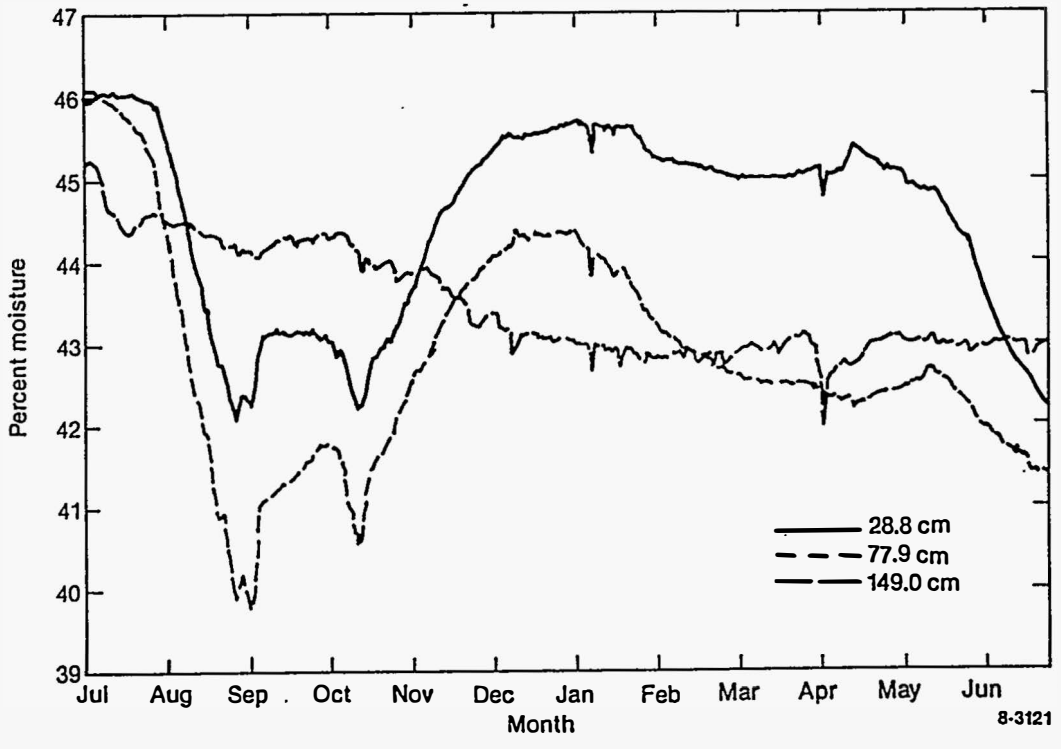

Figure C-22. ORNL lysimeter 1 soil moisture for 1986-87.

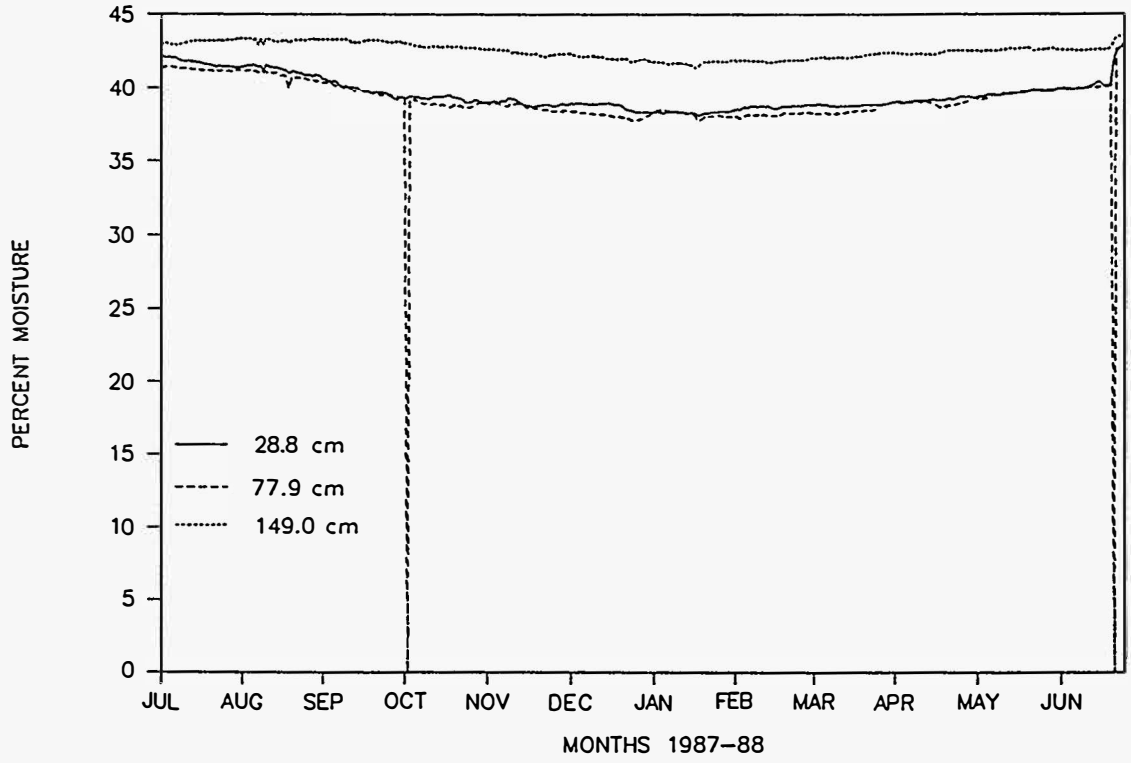

Figure C-23. ORNL lysimeter 1 soil moisture for 1987-88.

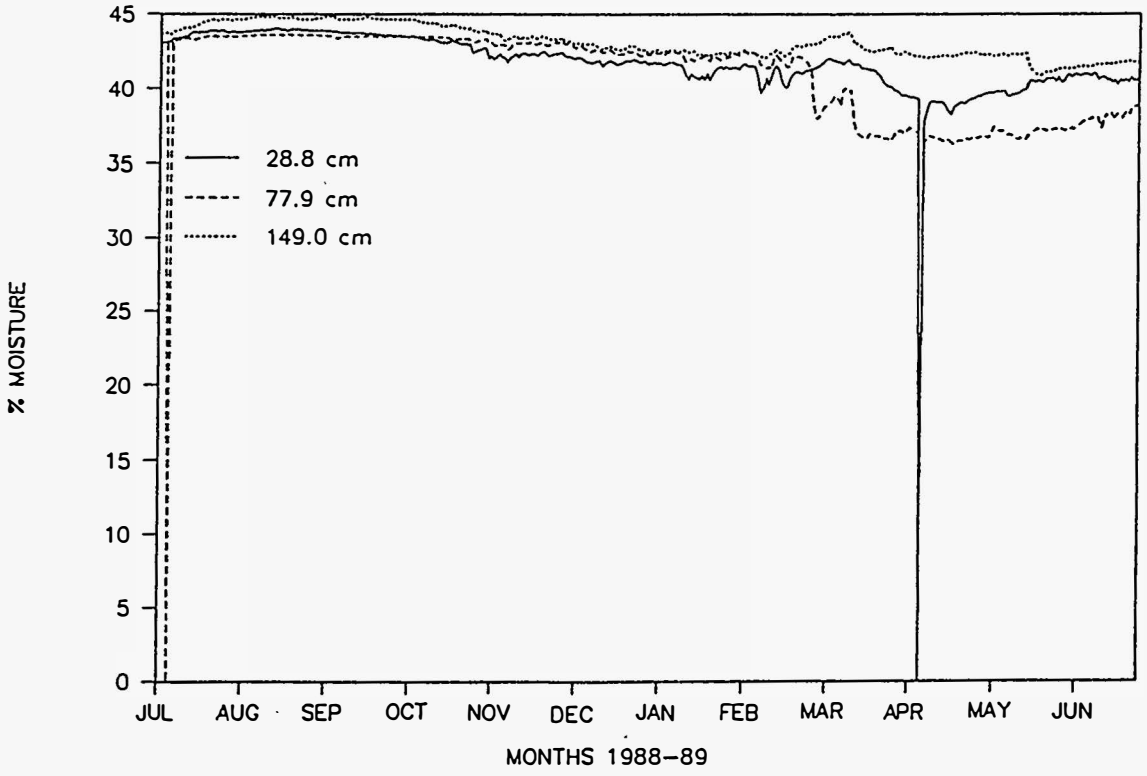

Figure C-24. ORNL lysimeter 1 soil moisture for 1988-89.

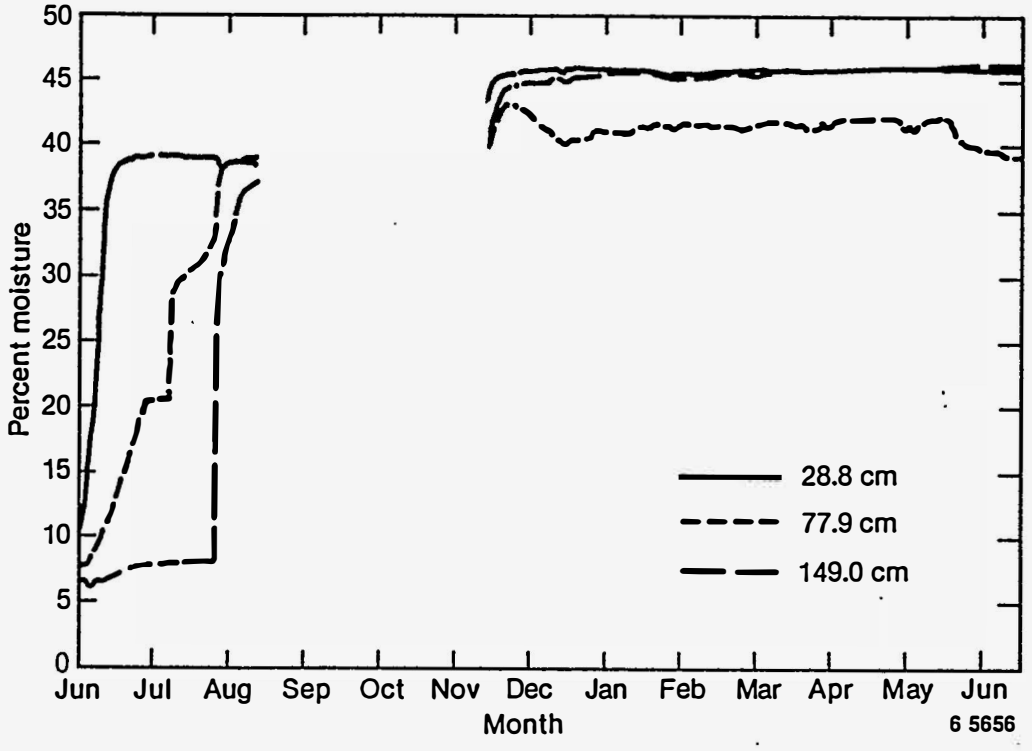

Figure C-25. ORNL lysimeter 2 soil moisture for 1985-86.

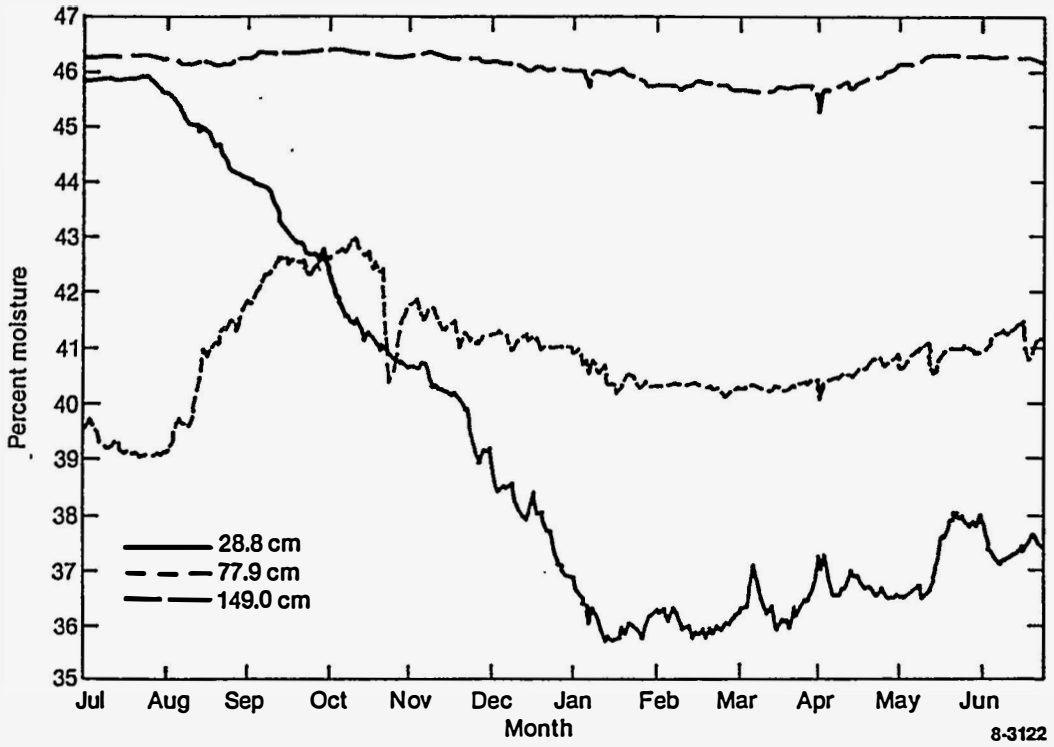

Figure C-26. ORNL lysimeter 2 soil moisture for 1986-87.

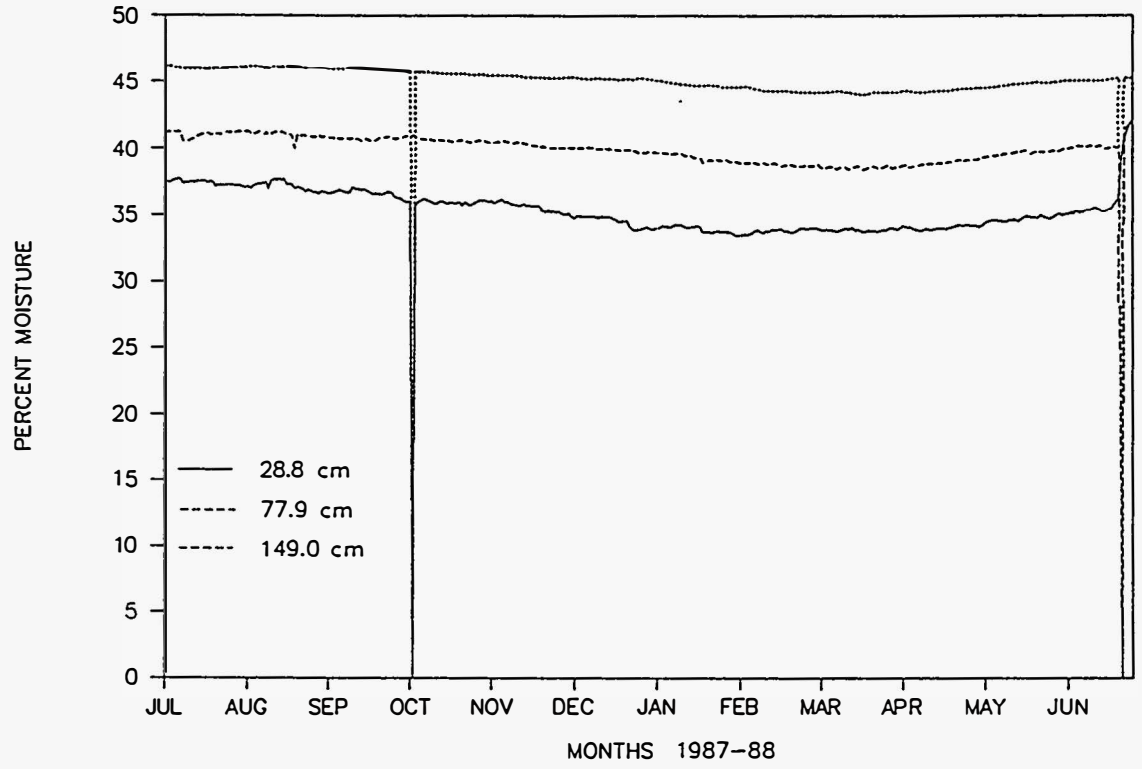

Figure C-27. ORNL lysimeter 2 soil moisture for 1987-88.

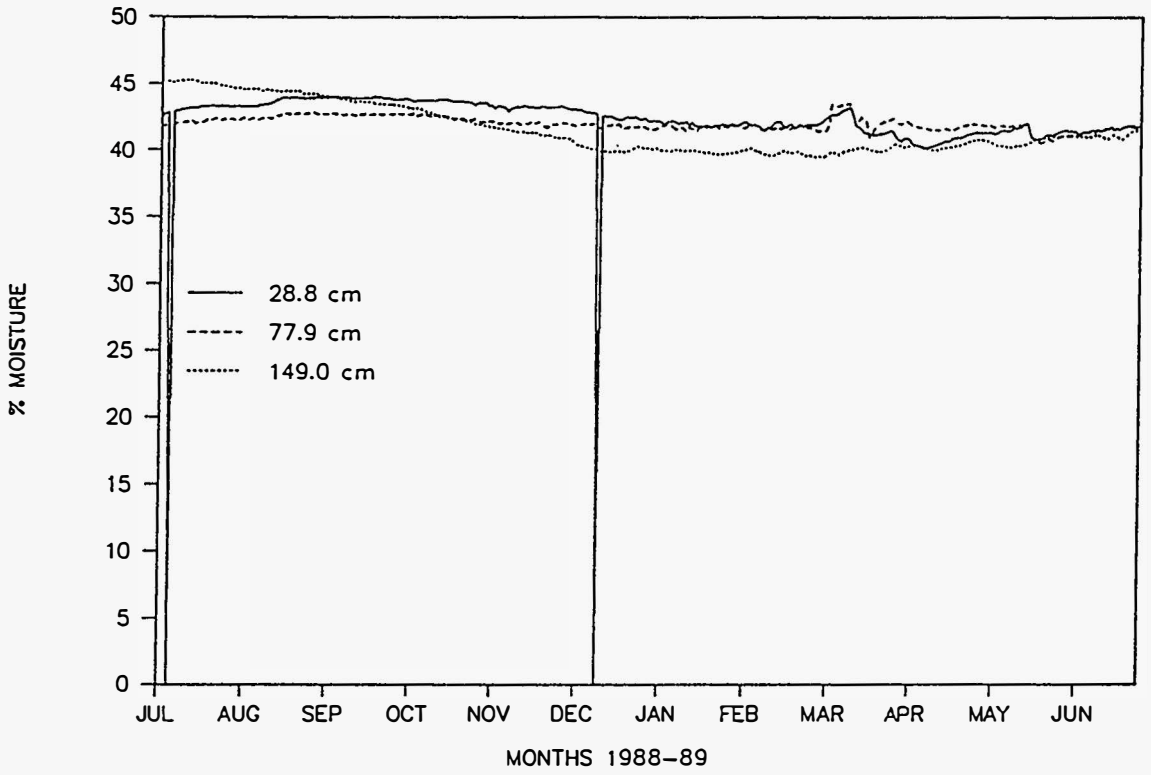

Figure C-28. ORNL lysimeter 2 soil moisture for 1988-89.

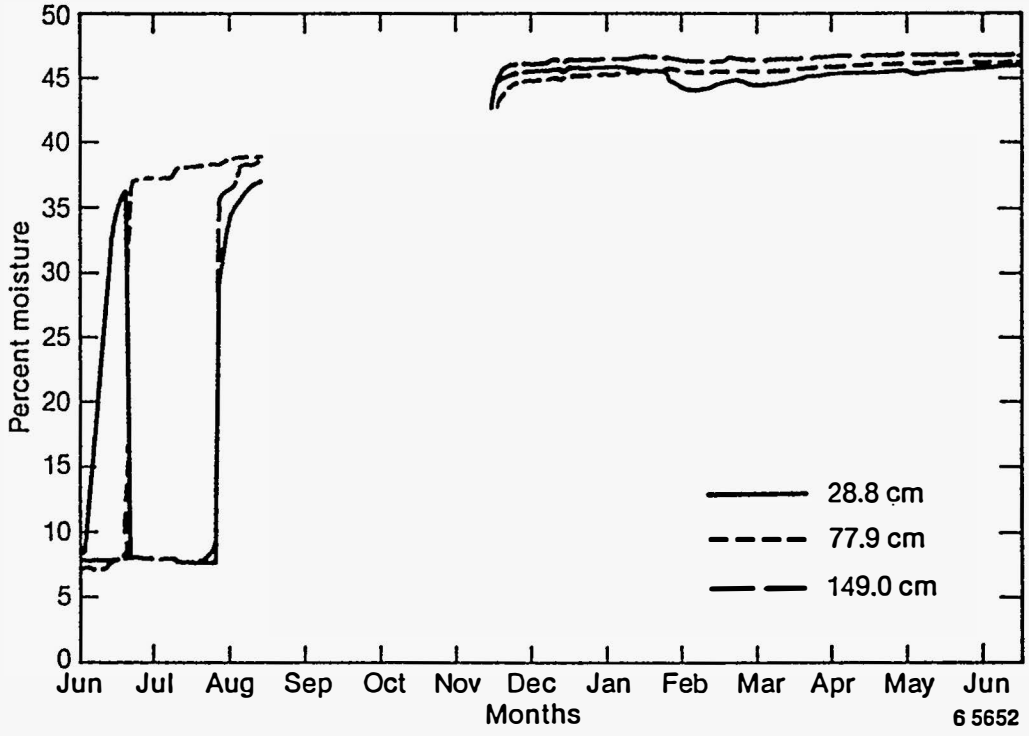

Figure C-29. ORNL lysimeter 3 soil moisture for 1985-86.

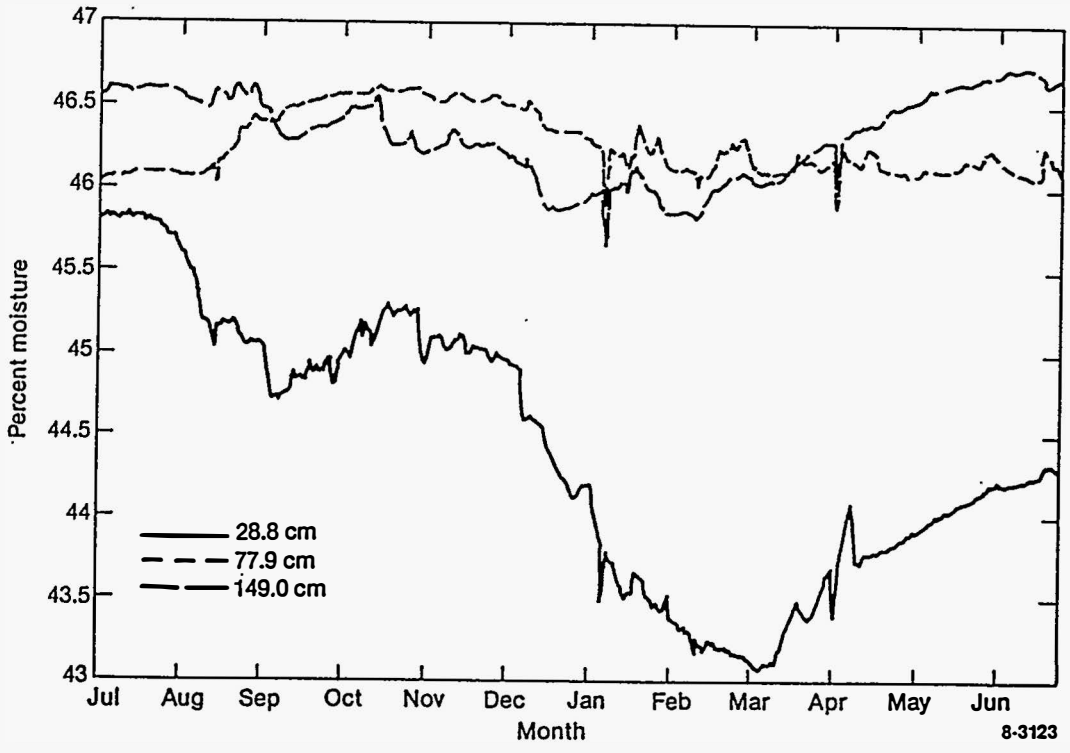

Figure C-30. ORNL lysimeter 3 soil moisture for 1986-87.

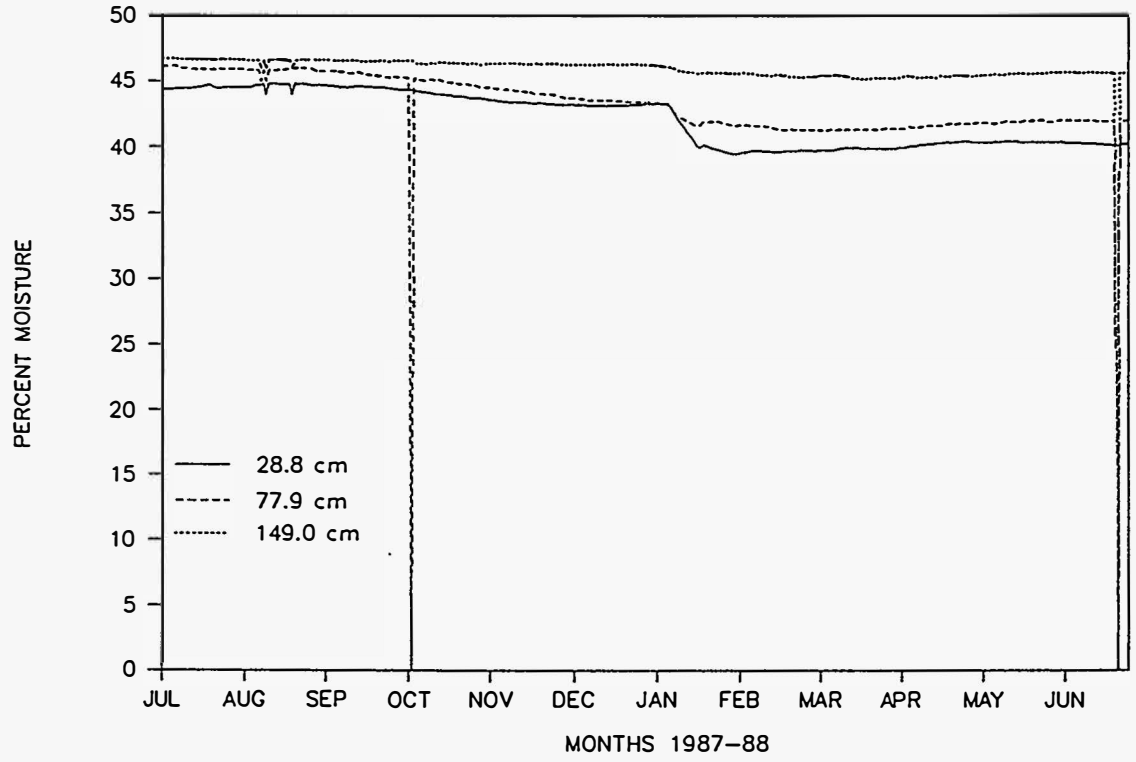

Figure C-31. ORNL lysimeter 3 soil moisture for 1987-88.

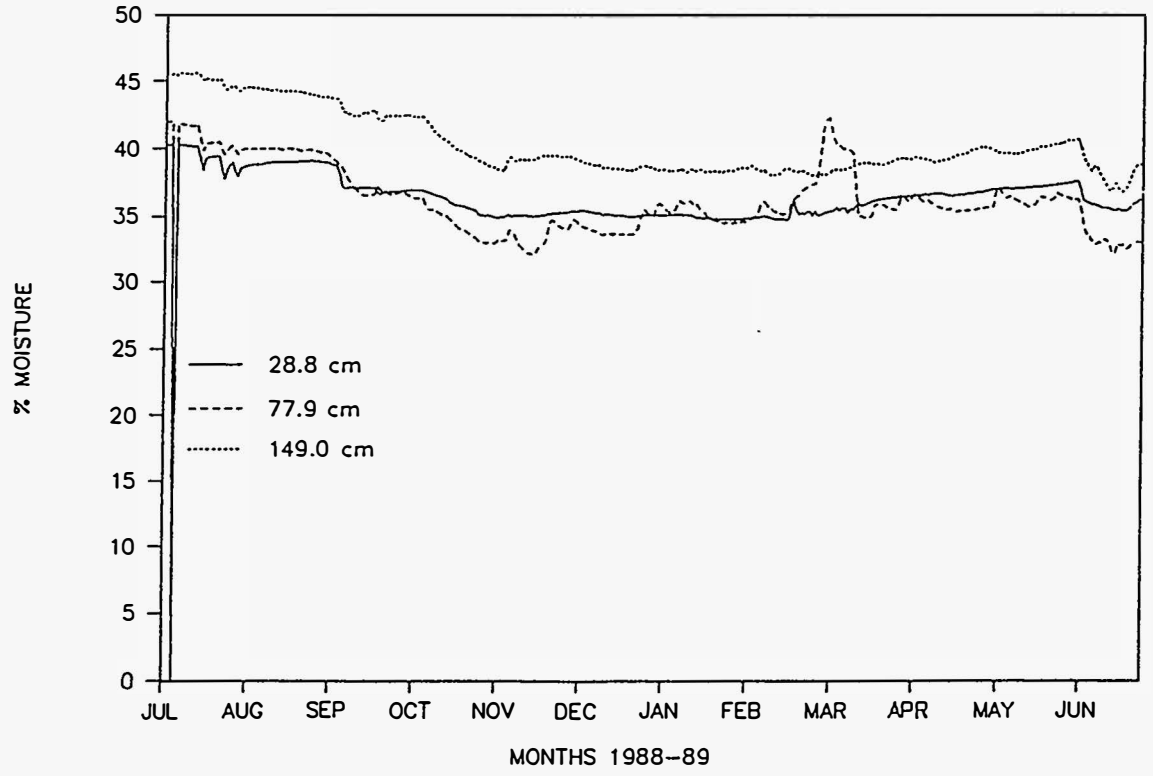

Figure C-32. ORNL lysimeter 3 soil moisture for 1988-89.

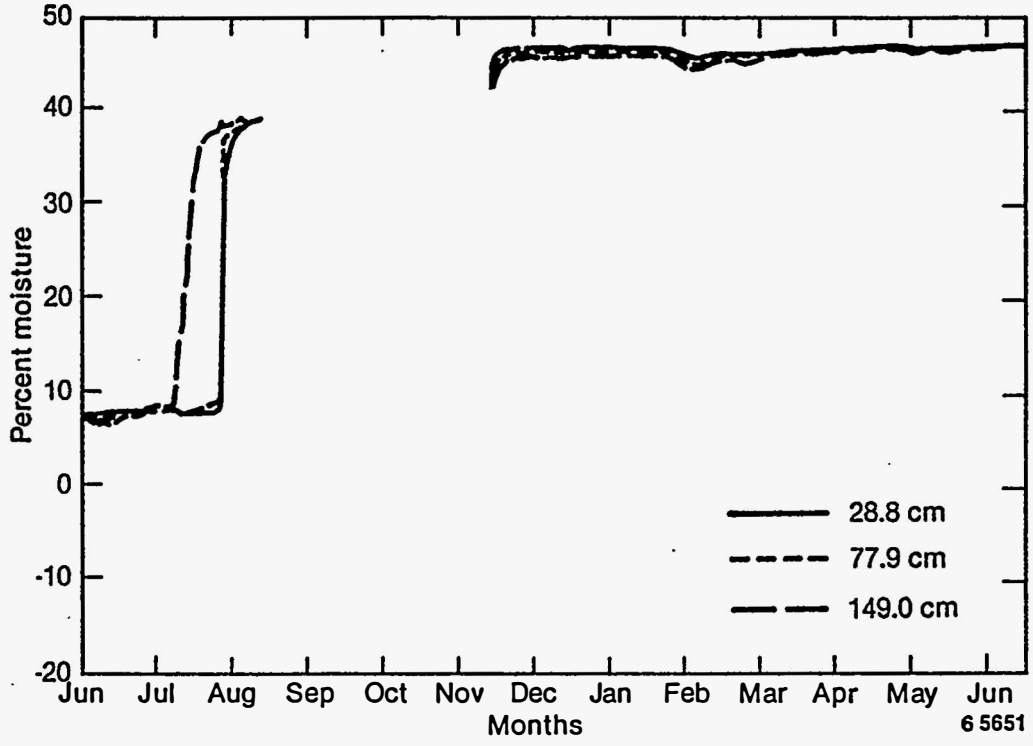

Figure C-33. ORNL lysimeter 4 soil moisture for 1985-86.

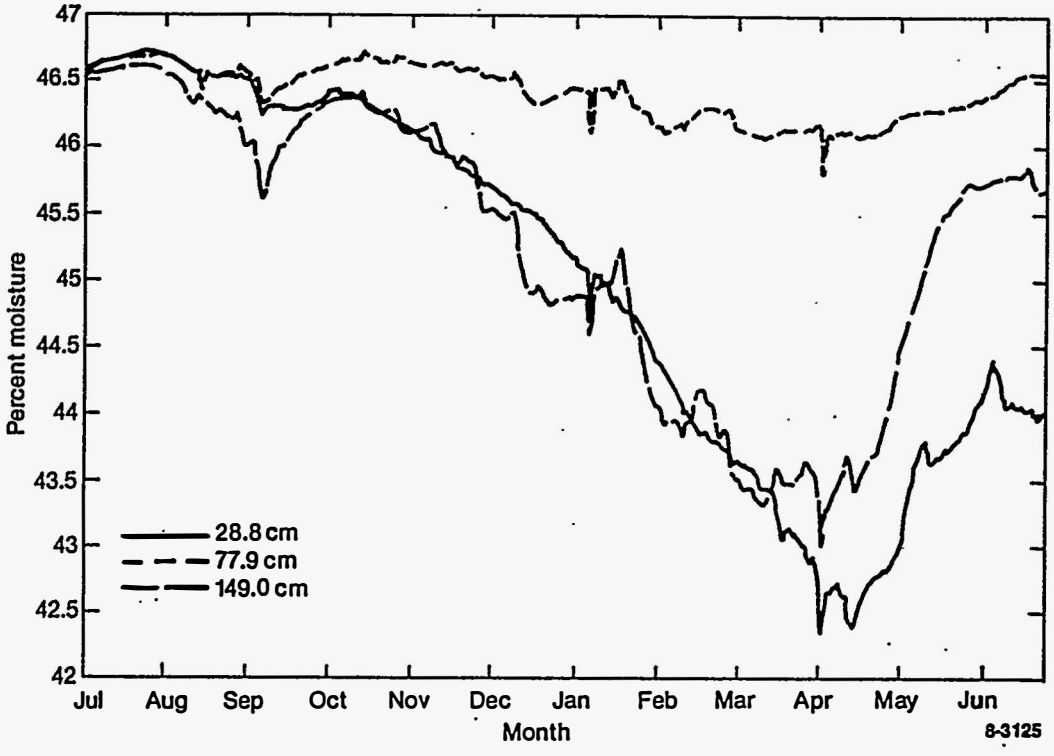

Figure C-34. ORNL lysimeter 4 soil moisture for 1986-87.

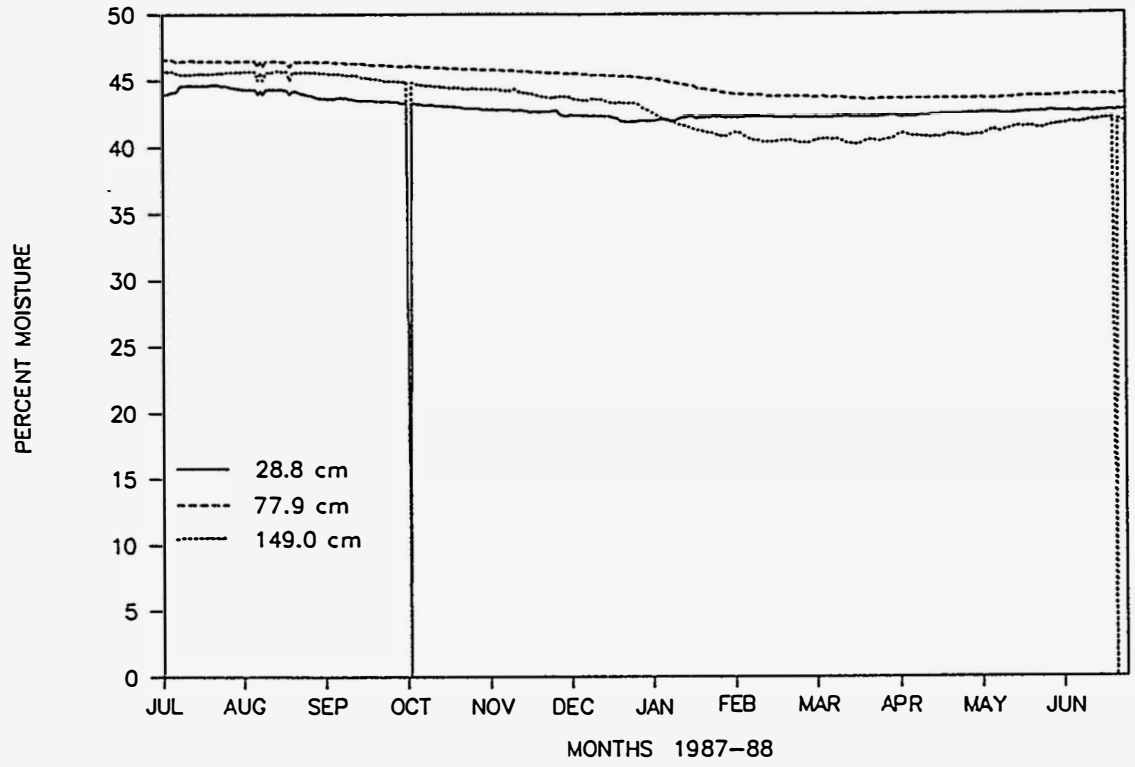

Figure C-35. ORNL lysimeter 4 soil moisture for 1987-88.

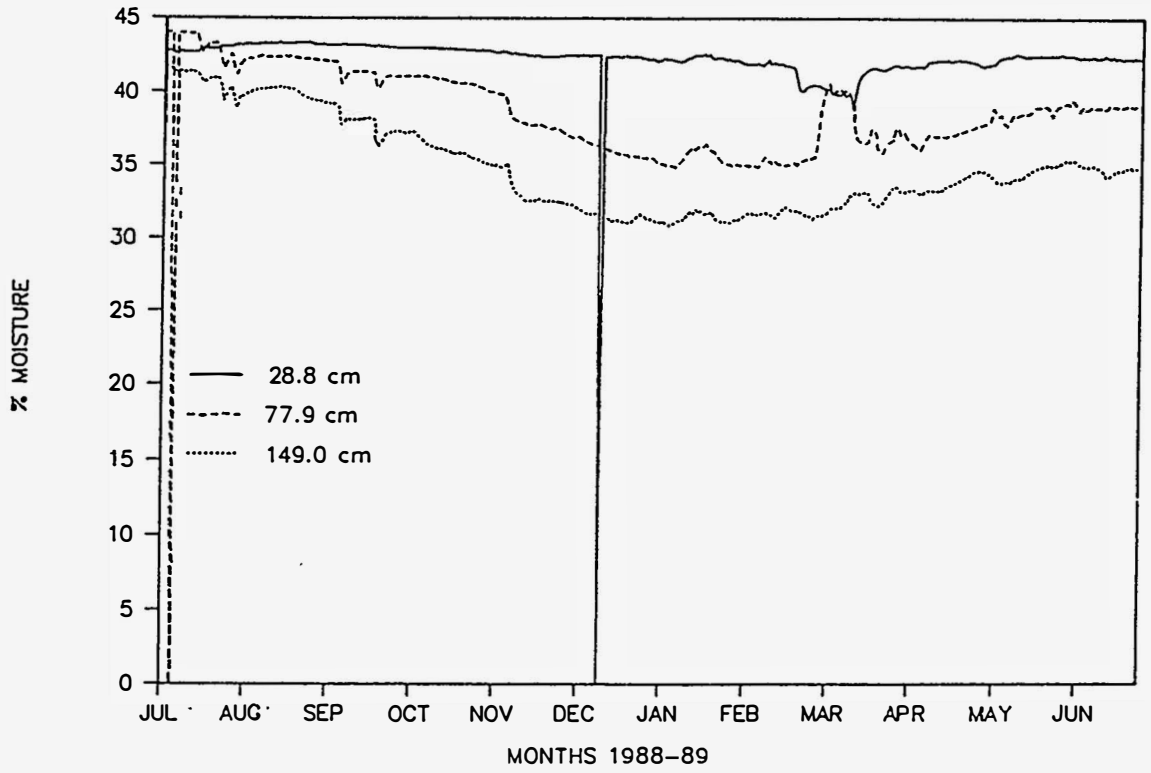

Figure C-36. ORNL lysimeter 4 soil moisture for 1988-89.

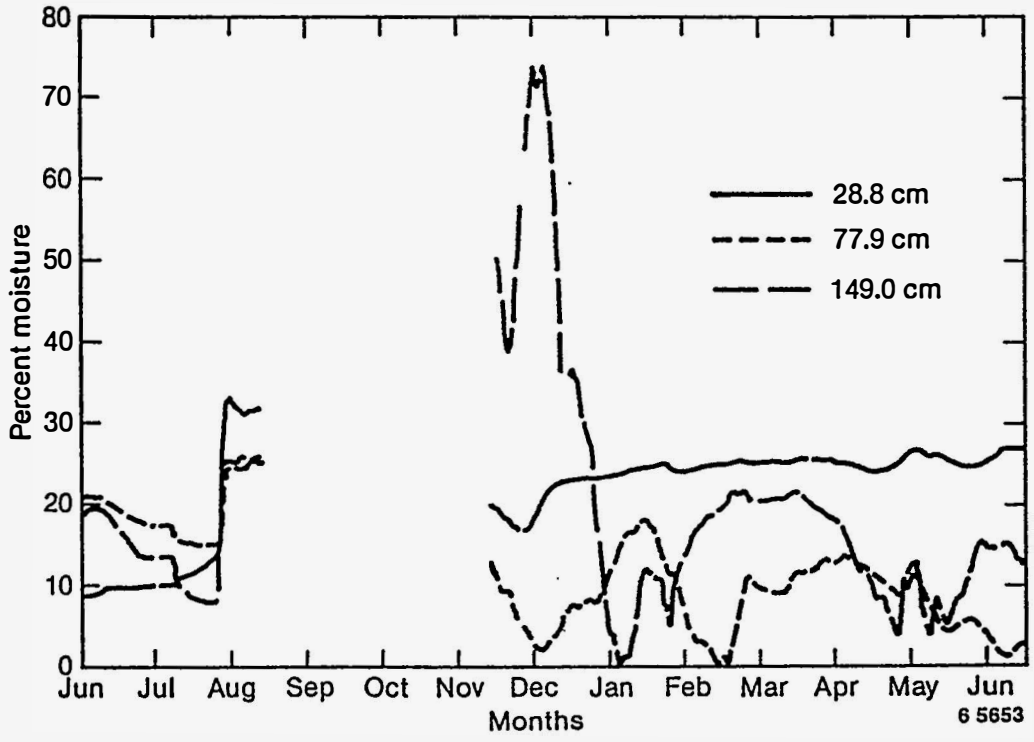

Figure C-37. ORNL lysimeter 5 soil moisture for 1985-86.

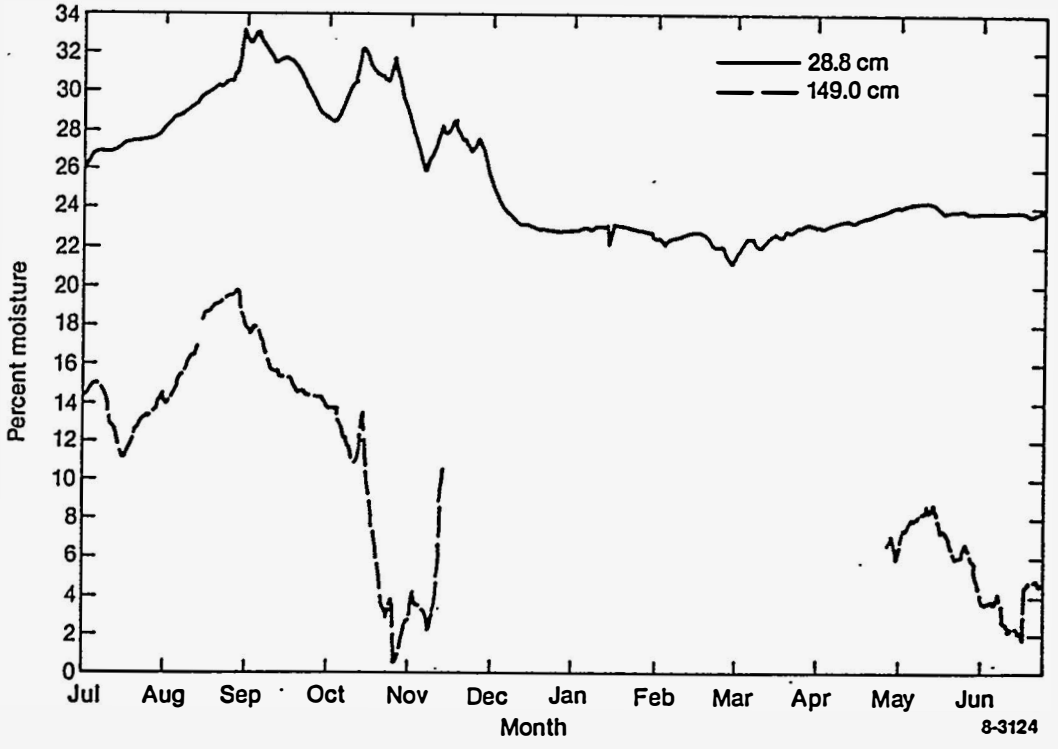

Figure C-38. ORNL lysimeter 5 soil moisture for 1986-87.

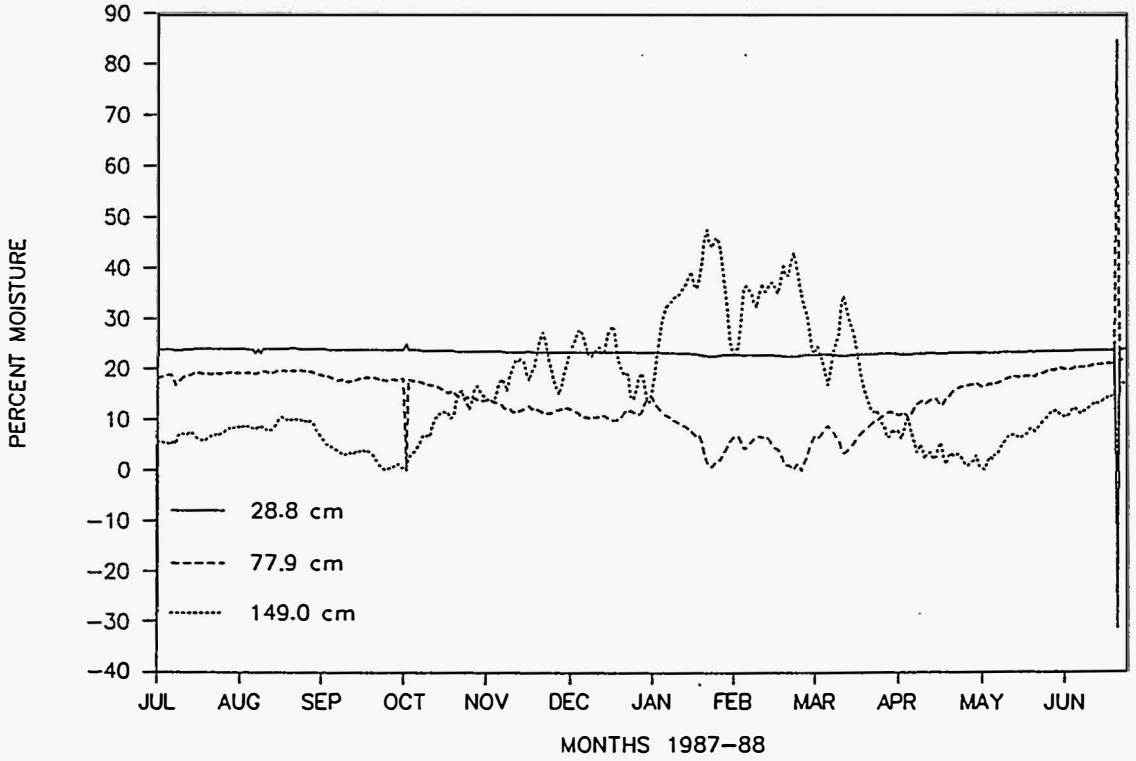

Figure C-39. ORNL lysimeter 5 soil moisture for 1987-88.

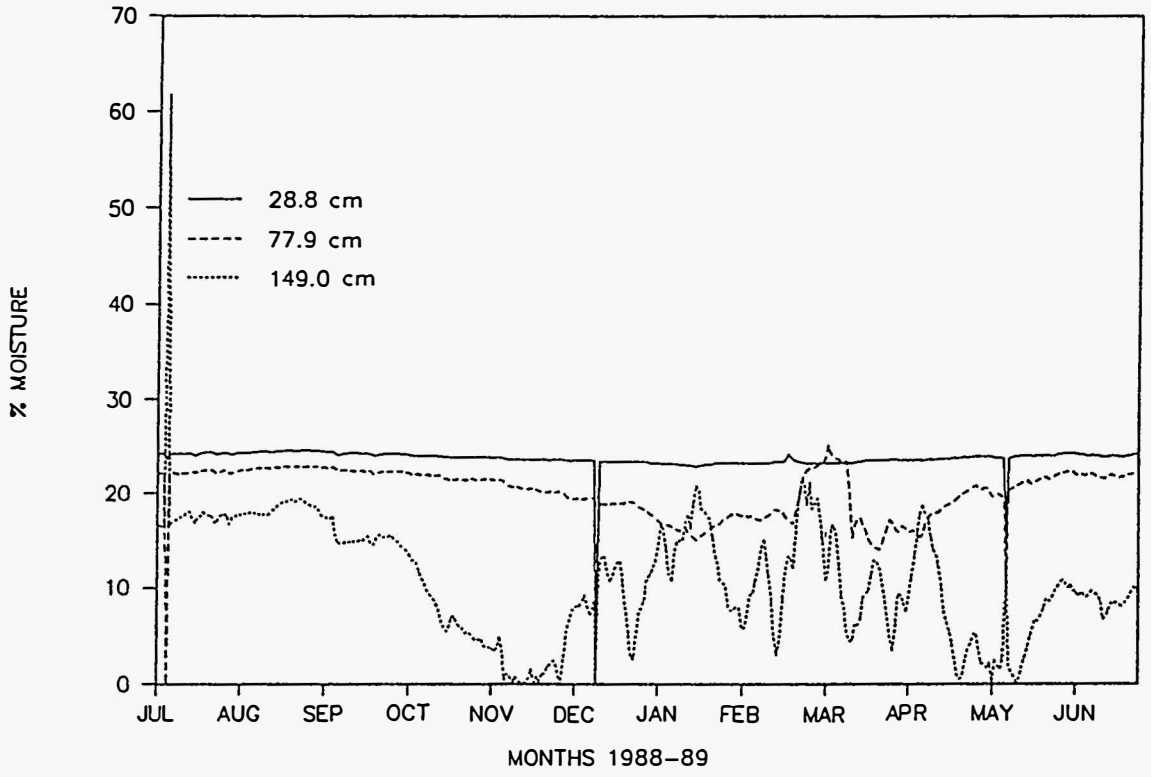

Figure C-40. ORNL lysimeter 5 soil moisture for 1988-89.

Appendix D

Soil Moisture Data—Gravimetric

Table D-1. Moisture profile of ANL-E lysimeter 3 based on gravimetric measurement of water content for 1985–86.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
3	0–20	17.4
3	20–41	19.9
3	41–61	21.0
3	61–81	22.9
3	81–102	22.7
3	102–122	23.4
3	122–142	23.1
3	142–162	22.7
3	162–183	24.3

a. Samples were collected on July 28, 1986.

Table D-2. Moisture profile of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content for 1986–87.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–20	25.0
1	20–41	23.0
1	41–62	23.4
1	62–82	23.4
1	82–107	23.6
1	107–133	24.0
1	133–153	24.1
1	153–182	23.7
1	182–202	23.8
2	0–20	25.7
2	20–41	26.2
2	41–62	25.1
2	62–82	24.5
2	82–107	24.0
2	107–133	24.4
2	133–153	24.3
2	153–182	23.4
2	182–202	24.0
3	0–20	13.9
3	20–41	22.3
3	41–62	23.9
3	62–82	25.2
3	82–107	24.6
3	107–133	24.2
3	133–153	23.8
3	153–182	24.5
3	182–202	23.4
4	0–20	16.4
4	20–41	24.4
4	41–62	24.7
4	62–82	24.2
4	82–107	24.5
4	107–133	25.1
4	133–153	24.8
4	153–182	24.2
4	182–202	24.1

a. Samples were collected on July 22, 1987.

Table D-3. Moisture profile of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content for 1987–88.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–41	22.3
1	41–62	21.4
1	62–82	21.3
1	82–107	21.5
1	107–133	21.8
1	133–153	22.0
1	153–182	22.9
1	182–202	23.9
2	0–41	17.8
2	41–62	20.4
2	62–82	21.4
2	82–107	22.1
2	107–133	22.3
2	133–153	22.7
2	153–182	22.9
2	182–202	21.8
3	0–41	17.3
3	41–62	20.7
3	62–82	23.2
3	82–107	24.7
3	107–133	23.6
3	133–153	23.3
3	153–182	24.6
3	182–202	24.3
4	0–41	16.2
4	41–62	19.8
4	62–82	21.8
4	82–107	22.4
4	107–133	22.9
4	133–153	22.8
4	153–182	23.8
4	182–202	23.8

a. Samples were collected on July 22, 1988.

Table D-4. Moisture profile of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content for 1988–89.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–41	19.8
1	41–62	21.1
1	62–82	21.0
1	82–107	21.8
1	107–133	21.9
1	133–153	22.3
1	153–182	22.9
1	182–202	23.1
2	0–41	20.7
2	41–62	21.5
2	62–82	21.6
2	82–107	22.6
2	107–133	22.5
2	133–153	22.3
2	153–182	22.6
2	182–202	23.1
3	0–41	18.7
3	41–62	22.1
3	62–82	24.3
3	82–107	24.3
3	107–133	24.6
3	133–153	24.3
3	153–182	24.2
3	182–202	24.2
4	0–41	22.4
4	41–62	20.4
4	62–82	21.8
4	82–107	21.6
4	107–133	21.9
4	133–153	23.0
4	153–182	23.5
4	182–202	24.2

a. Samples were collected on July 28, 1989.

Table D-5. Moisture profile of ORNL lysimeters 1 thorugh 4 based on gravimetric measurement of water content for 1985–86.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–36	12.5
1	36–71	14.2
1	71–107	15.1
2	0–36	13.7
2	36–71	14.1
2	71–107	14.3
3	0–36	12.7
3	36–71	14.6
3	71–107	15.2
4	0–36	12.8
4	36–71	13.9
4	71–107	14.7

a. Samples were collected in July, 1986.

Table D-6. Moisture profile of ORNL lysimeters 1 thorugh 4 based on gravimetric measurement of water content for 1986–87.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–68	16.2
1	68–136	17.3
1	136–204	18.1
1	204–272	18.3
1	272–325	16.4
2	0–68	15.1
2	68–136	16.2
2	136–204	17.3
2	204–272	18.2
2	272–325	16.3
3	0–68	15.3
3	68–136	16.7
3	136–204	17.8
3	204–272	17.4
3	272–325	17.7
4	0–68	15.4
4	68–136	16.9
4	136–204	16.9
4	204–272	17.8
4	272–325	17.9

a. Samples were collected on July 14, 1987.

Table D-7. Moisture profile of ORNL lysimeters 1 thorugh 4 based on gravimetric measurement of water content for 1987–88.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–25	15.0
1	25–50	15.1
1	50–75	07.1
1	75–100	—
1	100–125	18.1
1	125–150	18.9
2	0–25	15.2
2	25–50	15.5
2	50–75	16.9
2	75–100	17.4
2	100–125	17.6
2	125–150	18.0
3	0–25	14.9
3	25–50	16.3
3	50–75	16.6
3	75–100	16.2
3	100–125	17.8
3	125–150	18.0
4	0–25	15.4
4	25–50	14.8
4	50–75	17.8
4	75–100	17.0
4	100–125	17.6
4	125–150	17.8

a. Samples were collected on July 18, 1988.

Table D-8. Moisture profile of ORNL lysimeters 1 thorugh 4 based on gravimetric measurement of water content for 1988–89.[a]

Lysimeter	Depth (cm)	Moisture (dry wt) (%)
1	0–25	14.3
1	25–50	15.9
1	50–75	16.8
1	75–100	17.2
1	100–125	17.6
1	125–150	18.2
2	0–25	13.9
2	25–50	15.2
2	50–75	16.4
2	75–100	14.6
2	100–125	15.9
2	125–150	15.9
3	0–25	14.0
3	25–50	15.4
3	50–75	16.6
3	75–100	17.1
3	100–125	17.3
3	125–150	18.1
4	0–25	12.4
4	25–50	14.8
4	50–75	16.3
4	75–100	16.9
4	100–125	16.7
4	125–150	16.4

a. Samples were collected on July 28, 1989.

Appendix E

Results of Beta and Gamma Analysis

Table E-1a. Results of gamma-ray and strontium analyses of ANL-E soil moisture and leachate samples for 1985–86.[a]

Sample identification	Concentration[b] (pCi/L)		
	Co-60	Cs-137	Sr-90
Lys 1-3[c]	<5	<5	1.0 ± 1.8
Lys 2-3	<5	<5	1.1 ± 1.2
Lys 3-3	11 ± 7	<5	1.1 ± 1.0
Lys 4-3	<5	<5	2.7 ± 1.8
Lys 5-3	<5	<5	55.6 ± 3.1
Lys 1[d]	<5	<5	0.5 ± 0.3
Lys 2	<5	<5	0.5 ± 0.2
Lys 3	<5	<5	0.4 ± 0.1
Lys 4	<5	<5	0.6 ± 0.3
Lys 5	<5	5.4 ± 1.1	1.0 ± 0.4

a. April 1986.

b. Concentration ± 2 sigma.

c. Moisture cup identity number.

d. Leachate collector identity number.

Table E-1b. Results of gamma-ray analysis of ANL-E soil moisture and leachate samples for 1985–86.

Sample identification	Concentration[b] (pCi/L)		
	Co-60	Cs-137	Sr-90
Composite[c]	<5	<5	<1
Lys 1-1[d]	<5	<5	<1
Lys 1-3	<5	<5	<1
Lys 2-3	<5	<5	<1
Lys 3-3	13 ± 7	<5	11.3 ± 1.4
Lys 4-3	<5	<5	<1
Lys 5-1	<5	<5	349.6 ± 11.3
Lys 5-3	<5	<5	127.6 ± 6.7
Lys 1[e]	<1	<1	<1
Lys 2	<1	<1	<1
Lys 3	<1	<1	<1
Lys 4	<1	<1	<1
Lys 5	<1	<1	5.8 ± 0.3

a. June 1986.

b. Concentration ± 2 sigma.

c. Composite of water from the number 5 moisture cups of lysimeters 1 through 4.

d. Moisture cup identity number.

e. Leachate collector identity number.

Table E-2. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples for 1986–87.

Sample identification	Concentration (pCi/L)[a] Co-60 October 86	Co-60 February 87	Co-60 April 87	Co-60 June 87	Cs-137 October 86	Cs-137 February 87	Cs-137 April 87	Cs-137 June 87	Sr-90 October 86	Sr-90 February 87	Sr-90 April 87	Sr-90 June 87
Lys 1[b]	<5	<5	<5	<1	<5	<5	3 ± 2	<1	3.0 ± 18.6	51.3 ± 27.0	<1	26.2 ± 21.3
Lys 2	<5	<5	<5	<1	<5	<5	<5	<1	8.1 ± 18.1	20.3 ± 25.4	25.7 ± 21.1	22.1 ± 20.5
Lys 3	<5	<5	<5	<1	<5	18 ± 1	<5	<1	7.0 ± 17.0	0.4 ± 16.6	<1	75.6 ± 32.4
Lys 4	<5	<5	<5	<1	<5	<5	<5	<1	11.9 ± 16.2	<27	<1	8.6 ± 19.4
Lys 5	<5	<5	<5	<1	<5	<5	<5	<1	40.5 ± 21.6	82 ± 2	85.8 ± 0.5	148.5 ± 40.5
Lys 1-3[c]	<5	<5	<5	<5	<5	<5	<5	<5	118.8 ± 37.8	13.5 ± 51.3	535 ± 18	1,341 ± 15
Lys 2-3	<5	<5	<5	<5	109 ± 19	9,183 ± 116	969 ± 28	723 ± 27	11.9 ± 23.6	22.1 ± 22.1	2,970 ± 54	1,370 ± 16
Lys 3-3	<5	<5	<5	<5	<5	<5	<5	<5	1,198 ± 135	13,500 ± 270	36,100 ± 471	79,200 ± 672
Lys 4-3	<5	<5	<5	<5	<5	<5	<5	<5	NA	16.2 ± 24.8	52 ± 7	15.7 ± 21.9
Lys 5-3	<5	<5	<5	<5	<5	<5	<5	21 ± 11	647 ± 10	950 ± 54	198 ± 34	2,293 ± 35
Lys 2-1	—	FL[d]	<5	<5	—	FL	<5	<5	—	FL	<5	—
Lys 3-1	<5	NA[e]	—	—	<5	NA	—	—	NA	—	—	NA
Lys 5-1	<5	<5	<5	<5	<5	<5	<5	<5	99 ± 2	190 ± 2	198 ± 11	NA
Lys 2-4	—	—	<5	—	—	—	<5	—	—	—	<1	—
Lys 5-4	<5	NS[f]	<5	—	<5	NS	<5	—	6 ± 5	NA	<1	NA
Lys 2-2	—	—	—	<5	—	—	<5	—	—	—	—	NA
Lys 5-2	NA	—	—	<5	<5	—	—	<5	<5	—	24 ± 3	NA

a. Concentration ±2 sigma.

b. Leachate sample from 1-L sample size.

c. Moisture cup sample from ~0.1-L sample size.

d. Sample not taken due to frozen lines.

e. Sample not analyzed.

f. No sample taken.

Table E-3. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples for 1987–88.

Sample identification	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
	September 1987	November 1987	April 1988	June 1988	September 1987	November 1987	April 1988	June 1988	September 1987	November 1987	April 1988	June 1988
Lys 1[b]	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Lys 2	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Lys 3	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Lys 4	<1	<1	<1	<1	<1	2 ± 1	<1	<1	<1	<1	<1	<1
Lys 5	<1	<1	<1	<1	39 ± 3	2 ± 1	<1	3 ± 1	126 ± 1	99 ± 4	187 ± 7	139 ± 2
Lys 1-3[c]	<5	<5	<5	<5	6 ± 5	<5	9 ± 7	8 ± 6	2,886 ± 68	5,173 ± 195	6,423 ± 259	6,680 ± 135
Lys 2-3	<5	<5	<5	<5	666 ± 46	317 ± 35	191 ± 25	255 ± 15	1,350 ± 18	1,588 ± 26	1,281 ± 30	1,182 ± 26
Lys 3-3	<5	<5	<5	<5	<5	<5	9 ± 12	<5	1.7E5 ± 1,648	1.9E5 ± 2,894	3.6E5 ± 601	2.6E5 ± 3720
Lys 4-3	<5	<5	<5	<5	<5	<5	<5	33 ± 8	99 ± 24	142 ± 25	133 ± 9	1,140 ± 38
Lys 5-3	<5	<5	<5	<5	45 ± 24	69 ± 14	200 ± 27	521 ± 27	2,870 ± 318	4,089 ± 502	187 ± 7	6,283 ± 151

a. Concentration ±2 sigma.

b. Leachate sample from 1-L sample size.

c. Moisture cup sample from ~0.1-L sample size.

Table E-4. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples for 1988–89.

Sample identification	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
	September 1988	November 1988	April 1989	July 1989	September 1988	November 1988	April 1989	July 1989	September 1988	November 1988	April 1989	July 1989
Lys 1[b]	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Lys 2	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Lys 3	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	9.1 ± 1.3
Lys 4	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1
Lys 5	<1	<1	<1	<1	2 ± 1	2 ± 1	<1	<1	274 ± 25	387 ± 10	276 ± 5	417 ± 17
Lys 1-3[c]	<5	<5	<5	<5	<5	20 ± 18	<5	<5	10E4 ± 115	1.2E4 ± 75	8,479 ± 90	1.09E4 ± 171
Lys 2-3	<5	<5	<5	<5	2,171 ± 59	1,294 ± 51	625 ± 36	1,309 ± 48	1,324 ± 32	2,000 ± 31	1,720 ± 154	2,031 ± 36
Lys 3-3	<5	<5	<5	760 ± 45	<5	<5	<5	3,638 ± 86	3.1E5 ± 4,466	5.4E5 ± 9,740	5.8E5 ± 5,017	6.12E5 ± 3,554
Lys 4-3	<5	<5	<5	<5	<5	<5	<5	<5	1,818 ± 38	1,108 ± 28	1,968 ± 65	2,578 ± 49
Lys 5-3	<5	<5	<5	<5	1,539 ± 58	3,501 ± 94	2,572 ± 70	9,389 ± 924	6,972 ± 73	5,088 ± 119	8,042 ± 55	1.28E4 ± 305

a. Concentration ±2 sigma.

b. Leachate sample from 1-L sample size.

c. Moisture cup sample from ~0.1-L sample size.

Table E-5a. Results of gamma-ray and strontium analyses of ORNL soil moisture and leachate samples for 1985--86.[a]

Sample identification	Concentration[b] (pCi/L)			
	Co-60	Cs-137	Sb-125	Sr-90
Lys 1-3[c]	<16	<14	<27	2.7 ± 4.9
Lys 2-3	<16	<14	<27	7.0 ± 13.5
Lys 3-3	<16	<11	<27	5.9 ± 12.9
Lys 4-3	<14	<11	<27	<11
Lys 5-3	<14	<14	351 ± 5	6.2 ± 18.1
Lys 1[d]	<2	<1	<3	62.2 ± 8.1
Lys 2	<1	<1	<3	27.0 ± 5.4
Lys 3	<3	<2	<5	4.9 ± 2.7
Lys 4	<2	<2	<3	54.1 ± 8.1
Lys 5	<2	<1	<5	45.9 ± 8.1

a. April 1986.

b. Concentration ± 2 sigma.

c. Moisture cup identity number.

d. Leachate collector identity number.

Table E-5b. Results of gamma-ray and strontium analyses of ORNL soil moisture and leachate samples for 1985–86.[a]

Sample identification	Concentration[b] (pCi/L)			
	Co-60	Cs-137	Sb-125	Sr-90
Composite[c]	<19	<19	<27	8.6 ± 10.2
Lys 1-3[d]	<27	<22	<27	0.5 ± 7.6
Lys 2-3	<81	<54	<27	15.4 ± 18.1
Lys 3-3	<54	<54	<27	32.4 ± 18.9
Lys 4-3	<27	<16	<27	<5
Lys 5-3	89.2 ± 32.4	<3	540 ± 81	17.6 ± 12.2
Lys 1[e]	<5	<5	<3	9.2 ± 3.5
Lys 2	<8	<6	<0.3	2.4 ± 2.7
Lys 3	<8	<6	<0.3	1.1 ± 2.7
Lys 4	<8	<5	<0.3	11.6 ± 4.6
Lys 5	<8	<5	<37	2.4 ± 2.9

a. June 1986.

b. Concentration ± 2 sigma.

c. Composite of water from the number 5 moisture cups of lysimeters 1 through 4.

d. Moisture cup identity number.

e. Leachate collector identity number.

Table E-6. Results of beta and gamma analysis of ORNL soil moisture and leachate samples for 1986–87.

Sample identification	Concentration (pCi/L)[a]															
	Co-60				Cs-137				Sb-125				Sr-90			
	October 1986	January 1987	April 1987	July 1987	October 1986	January 1987	April 1987	July 1987	October 1986	January 1987	April 1987	July 1987	October 1986	January 1987	April 1987	July 1987
Lys 1[b]	<5	<5	<5	<11	<8	<5	<5	<5	—	—	—	—	29.7 ± 5.4	1.3 ± 1.6	2.5 ± 4.3	89.2 ± 13.5
Lys 2	<8	<8	<0.5	<8	<8	<5	<0.5	<8	—	—	—	—	20.8 ± 5.1	1.3 ± 1.6	8.4 ± 5.1	6.49 ± 4.05
Lys 3	<8	<8	<8	<11	<5	<5	<8	<11	—	—	—	—	108 ± 10.8	2.2 ± 1.8	3.0 ± 4.8	7.84 ± 3.78
Lys 4	<5	<8	<5	<11	<5	<5	<5	<8	—	—	—	—	1.9 ± 2.5	1.4 ± 1.5	3.2 ± 4.0	4.32 ± 3.78
Lys 5	<5	<8	<5	<11	<5	<5	<5	<8	64.9 ± 24.3	54.0 ± 18.9	ND[d]	ND	3.5 ± 2.7	0.7 ± 1.4	6.5 ± 4.9	45.9 ± 8.11
Lys 1-3[c]	<54	<22	<22	<27	<27	<19	<19	<54	—	—	—	—	64.9 ± 10.8	297 ± 27	946 ± 54	1,540 ± 108
Lys 2-3	<27	<24	<22	<54	<27	<22	<19	<54	—	—	—	—	64.9 ± 27.0	270 ± 27	919 ± 81	1,459 ± 108
Lys 3-3	<54	<24	<27	<54	<54	<19	<19	<27	—	—	—	—	24.9 ± 7.8	45.9 ± 13.5	165 ± 27	405 ± 54.0
Lys 4-3	<54	<27	<22	<54	<54	<24	<19	<54	—	—	—	—	6.2 ± 3.2	<5	18.9 ± 11.3	10.0 ± 14.3
Lys 5-3	<54	<24	<16	<27	<27	<27	<16	<54	568 ± 108	324 ± 81	351 ± 108	ND	16.5 ± 10.3	5.1 ± 6.5	3.8 ± 7.6	11.3 ± 18.6
Lys 1-1	<24	<27	<3	<81	<22	<19	<3	<81	—	—	—	—	16.2 ± 10.8	1.9 ± 6.2	<5.4	9.19 ± 12.4
Lys 2-1	<81	<27	<27	<54	<54	<22	<27	<54	—	—	—	—	0.5 ± 5.7	4.3 ± 7.3	67.6 ± 27.0	8.65 ± 11.6
Lys 3-1	<19	<22	<14	<54	<19	<22	<16	<54	—	—	—	—	5.1 ± 6.8	16.2 ± 14.9	2.2 ± 7.3	4.86 ± 10.8
Lys 4-1	<19	<22	<19	<81	<19	<22	<19	<81	—	—	—	—	3.0 ± 6.2	2.4 ± 5.9	4.0 ± 7.8	26.7 ± 23.8
Lys 5-1	<54	<27	<22	<54	<27	<22	<19	<54	—	—	—	—	70.3 ± 16.2	10.0 ± 86	<8.1	8.65 ± 13.8

a. Concentration ±2 sigma.

b. Leachate sample from 1-L sample size.

c. Moisture cup sample from ~0.1-L sample size.

d. No isotope detected.

Table E-7. Results of beta and gamma analysis of ORNL soil moisture and leachate samples for 1987–88.

Sample identification	Concentration (pCi/L)[a]											
	Co-60			Cs-137			Sb-125			Sr-90		
	October 1987	April 1988	July 1988	October 1987	April 1988	July 1988	October 1987	April 1988	July 1988	October 1987	April 1988	July 1988
Lys 1[b]	<27	<8	<8	<27	<8	<8	—	—	—	0.08 ± 4.0	2.97 ± 3.5	<2.7
Lys 2	<16	<8	<5	<22	<8	<5	—	—	—	0.73 ± 2.7	1.35 ± 2.7	<2.7
Lys 3	<19	<8	<5	<19	<5	<5	—	—	—	7.3 ± 5.1	0.00 ± 2.7	2.7 ± 3.2
Lys 4	<19	<8	<5	<24	<8	<5	—	—	—	0.57 ± 2.8	3.24 ± 4.8	<2.7
Lys 5	<19	<8	<5	<22	<5	<5	<54	<24	<13	8.65 ± 4.3	12.2 ± 4.8	18.9 ± 5.4
Lys 1-3[c]	<19	<54	23.5 ± 18.7	<24	<54	<54	—	—	—	3,243 ± 270	11,350 ± 270	18,123 ± 541
Lys 2-3	<16	<54	<27	<27	<54	<54	—	—	—	1,675 ± 54	2,970 ± 270	4,057 ± 270
Lys 3-3	35 ± 32	<54	<27	<27	<54	<19	—	—	—	378 ± 27	2,160 ± 270	2,975 ± 270
Lys 4-3	<16	<54	<27	<16	<27	<21	—	—	—	167 ± 205	2.16 ± 14.1	186 ± 78
Lys 5-3	51 ± 24	<54	<27	<24	100 ± 40	162 ± 32	297 ± 81	<108	<81	13.5 ± 5.5	32.4 ± 21.6	8.3 ± 19.2
Lys 1-1	<19	<54	<57	24 ± 24	<81	<27	—	—	—	3.5 ± 3.8	<13	<2.7
Lys 2-1	<27	<54	<27	<27	<54	<27	—	—	—	1.1 ± 3.3	20.3 ± 18.4	<2.7
Lys 3-1	<19	<54	<27	<22	<27	<27	—	—	—	2.9 ± 3.2	10.8 ± 17.0	<2.7
Lys 4-1	<22	<54	<24	<22	<54	<21	—	—	—	5.9 ± 6.2	<13	4.8 ± 15.7
Lys 5-1	<19	<54	<21	<22	<27	<21	—	—	—	4.0 ± 5.4	16.8 ± 15.1	32.5 ± 18.9

a. Concentration ± 2 sigma.

b. Leachate sample from 1-L sample size.

c. Moisture cup sample from ~0.1-L sample size.

Table E-8. Results of beta and gamma analysis of ORNL soil moisture and leachate samples for 1988–89.

Sample identification	Concentration (pCi/L)[a]															
	Co-60				Cs-137				Sb-125				Sr-90			
	Nov 1988	Mar 1989	May 1989	June 1989	Nov 1988	Mar 1989	May 1989	June 1989	Nov 1988	Mar 1989	May 1989	June 1989	Nov 1988	Mar 1989	May 1989	June 1989
Lys 1[b]	<3	<3	<3	<5	<3	<3	<5	20 ± 4	<8	<8	<8	<8	<1	5.8 ± 5.1	<3	<3
Lys 2	<3	<3	<3	<5	<3	<3	<3	10 ± 4	<5	<8	<8	<8	<1	<5	1.6 ± 3.8	<3
Lys 3	<3	<3	<5	<3	8.6 ± 4.5	<5	<5	6 ± 4	<8	<11	<8	<8	1.6 ± 3.2	5.7 ± 4.1	1.9 ± 3.8	0.3 ± 3.5
Lys 4	<3	<3	<5	<3	13 ± 2.7	<3	<5	4 ± 2	<3	<8	<11	<8	<1	<3	<3	2.4 ± 3.5
Lys 5	2.2 ± 4.3	<3	<3	<5	15 ± 3.5	<3	378 ± 27	108 ± 5	18 ± 5.1	<8	22 ± 14	<11	32 ± 8.1	32 ± 8.1	35 ± 5.4	73 ± 10.8
Lys 1-3[c]	<24	<27	<27	<27	<24	<22	<24	<27	<54	<54	<54	<54	3E4 ± 3,000	1,300 ± 54	4.1E4 ± 2,700	6.5E4 ± 2,700
Lys 2-3	<22	<22	<27	30 ± 22	<19	<25	<24	<22	<54	<54	<54	<54	5,100 ± 540	220 ± 19	7,300 ± 2,700	8,100 ± 540
Lys 3-3	<22	<27	NA[d]	<27	<16	<22	NA	20 ± 20	<54	<54	NA	<54	7,800 ± 540	570 ± 27	NA	3.2E4 ± 2,700
Lys 4-3	38 ± 35	<22	<19	<27	32 ± 24	<22	<19	<24	<54	<54	<54	<54	<11	<3	29.7 ± 21.6	67.6 ± 32.4
Lys 5-3	<27	<19	<19	<22	351 ± 27	405 ± 27	568 ± 27	703 ± 27	<54	<54	<54	<54	68 ± 22	4.3 ± 20	135 ± 32.4	205 ± 43.4
Lys 1-1	<27	<27	<54	<27	<24	35 ± 41	<24	<54	<54	<54	<54	<54	14 ± 17	<3	13.8 ± 16	9.5 ± 17.6
Lys 2-1	<19	<27	<27	<24	<19	<22	<19	<54	<54	<54	<54	<54	<1	1.7 ± 2.2	6.5 ± 13.8	11.9 ± 19.2
Lys 3-1	<22	<19	<24	<27	<19	<24	<24	<54	<54	<54	<54	<54	<1	<3	3 ± 14.1	<13
Lys 4-1	<19	<22	<22	<27	<27	<27	<27	<54	<54	<54	<54	<54	<1	3.0 ± 4.3	<14	2.4 ± 17.3
Lys 5-1	<27	<22	<22	<27	<22	<27	<27	<54	<54	<54	<54	<54	70.3 ± 21.6	<3	43.2 ± 21.6	23.8 ± 24.6

a. Concentration ±2 sigma.

b. 1-L subsample from leachate collection.

c. Total moisture cup sample ~0.1-L sample size.

d. Sample not analyzed.

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J.W. McConnell, Jr., R.D. Rogers, M.W. Findlay, E.C. Davis, J.D. Jastrow, R.M. Neilson, Jr., L.D. Hilton

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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 first 4 years off data acquisition from the field testing are presented and discussed. During the continuing field testing, both Portland type I-II cement and Dow vinyl ester-styrene waste forms are being tested in lysimeter arrays located at Argonne National Laboratory-East in Illinois and at Oak Ridge National Laboratory. The 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.

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