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Field Lysimeter Investigations–Test Results

Low-Level Waste Data Base Development Program: Test Results for Fiscal Years 1990, 1991, 1992, and 1993

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Prepared by
J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow[1], D. S. Wickliff Hicks[2],
W. E. Sanford[2], R. R. Brey[3], T. M. Sullivan[4], 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

P. Reed, NRC Project Manager

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[1]Argonne National Laboratory, Argonne, Il
[2]Oak Ridge National Laboratory, Oak Ridge, TN
[3]Formerly with Purdue University
[4]Brookhaven National Laboratory, Upton, NY

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ABSTRACT

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program, funded by the U.S. Nuclear Regulatory Commission (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 second 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 a commercial disposal facility in the State of Washington. Research is being conducted on materials from four of those EPICOR-II prefilters under three tasks of the TMI-2 EPICOR-II Resin/Liner Investigation: Low-Level Waste Data Base Development Program.

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

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

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

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

Results of the second 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 1990, 91, 92, and 93

INTRODUCTION

The March 28, 1979 accident at Three Mile Island Unit 2 released approximately 560,000 gal of contaminated water to the auxiliary and fuel handling buildings. The water was decontaminated using a demineralization system called EPICOR-II developed by Epicor, Inc.[a] The contaminated water was cycled through three stages of organic and inorganic ion-exchange media. The first stage of the system was designated the prefilter, and the second and third stages were called demineralizers. After the filtration process, the ion-exchange media in 50 of the prefilters contained radionuclides in concentrations greater than the U.S. Nuclear Regulatory Commission (NRC) recommended limits for low-level wastes. Those prefilters were transported to the Idaho National Engineering Laboratory for interim storage before final disposal. A special overpack (high-integrity container) was developed during that storage period to dispose the prefilters at a commercial disposal facility in the State of Washington. As part of the EPICOR and Waste Research and Disposition Program funded by the U.S. Department of Energy, 46 prefilters were disposed, while four were retained for research purposes. Those prefilters used in the research were stored in temporary storage casks and were later disposed at the Radioactive Waste Management Complex at the Idaho National Engineering Laboratory.

Under the EPICOR and Waste Research and Disposition Program, continuing research has been conducted by the INEL on materials from those four EPICOR-II prefilters.[1,2] That work is now funded and directed by the NRC as part of the Field Lysimeter Investigations: Low-Level Waste Data Base Development Program. Three studies were initiated on organic ion-exchange resins from selected prefilters: (a) the resins were examined to measure radiation degradation, (b) tests were performed to characterize solidified ion-exchange resin waste forms, and (c) experiments are being conducted to field test solidified wastes using lysimeters.

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

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

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

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

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

The Field Lysimeter Investigations: Low-Level Waste Data Base Development Program is exposing waste forms to the physical, chemical, and microbiological environment of typical disposal sites; monitoring release and movement of radionuclides from those waste forms; and comparing the results with short-term laboratory leach test results. This program has been operating lysimeters for 8 years to obtain information on the performance of radioactive waste forms in a disposal environment and investigate waste form stability per requirements of 10 CFR 61. The experiment measures the releases of radionuclides from the waste forms and subsequent transport through soil columns to sampling locations within the lysimeters. This study was developed to field test waste forms composed of solidified ion-exchange resins from EPICOR-II. The resins used in the study are significant because they have high loadings of radionuclides and are the commercial types used by the nuclear industry.

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

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

The objectives of the Field Testing program are to (a) examine the performance of the waste forms in typical low-level waste disposal environments, (b) compare field results with short-term laboratory leach studies, (c) compare field results with Department of Energy Special Waste Program field test results, (d) develop a low-level radioactive waste field leach-rate data base for use in performance assessment source term calculations, and (e) apply a source term code to model the radionuclide releases from the lysimeter waste forms.

The results of the first 4 years of operation were presented in the annual reports (References 6 through 9) and were discussed in the topical report (Reference 10). This report discusses the results obtained during the second 4 years of operation of the experiment, which were presented in References 11 through 14.

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 second 4 years of lysimeter operation,[11-14] 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 15.

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.

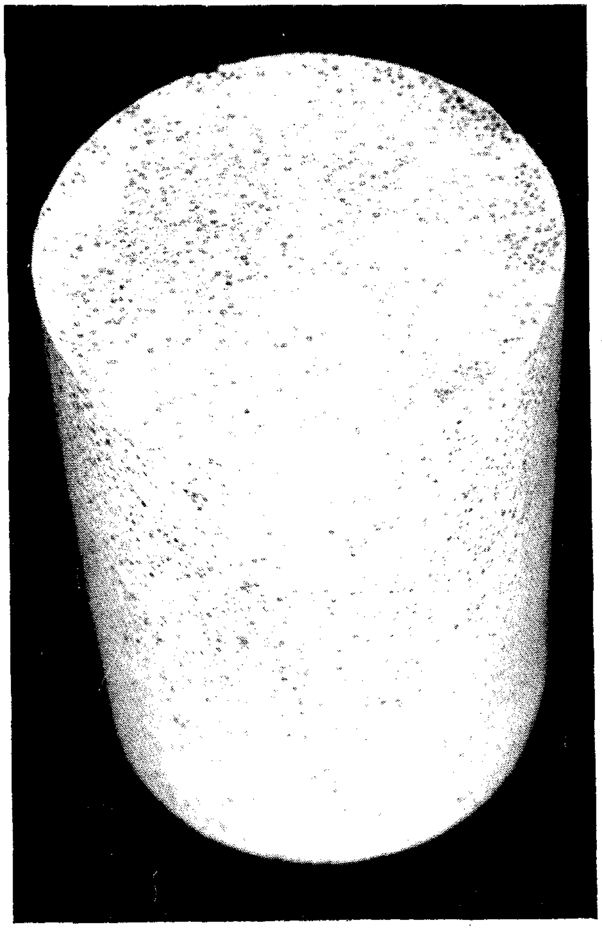


Figure 1. An example of an EPICOR-II prefilter waste form.

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] $\pm 1\sigma$ (Ci/g dry resin)
PF-7	Cs-134	7.73E-5 ± 2.83E-7
	Cs-137	1.17E-3 ± 9.90E-5
	Sr-90	6.92E-5 ± 7.21E-6
PF-24	Cs-134	3.30E-4 ± 5.80E-5
	Cs-137	4.99E-3 ± 3.04E-4
	Sr-90	1.18E-5 ± 6.36E-7

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

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

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.

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

PC = Portland type I–II cement.

VES = Vinyl ester-styrene.

DI = Demineralized water.

SW = Synthetic seawater.

CFR = Cumulative fractional release.

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

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

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

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

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

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

VES and cement waste forms were placed in nutrient-rich media to test the growth of the applied species of fungi and bacteria. 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 16; bench testing of those EPICOR-II waste forms, according to the recommendations of the BTP, is further described in References 8, 17, and 18.

Description of Test Sites

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

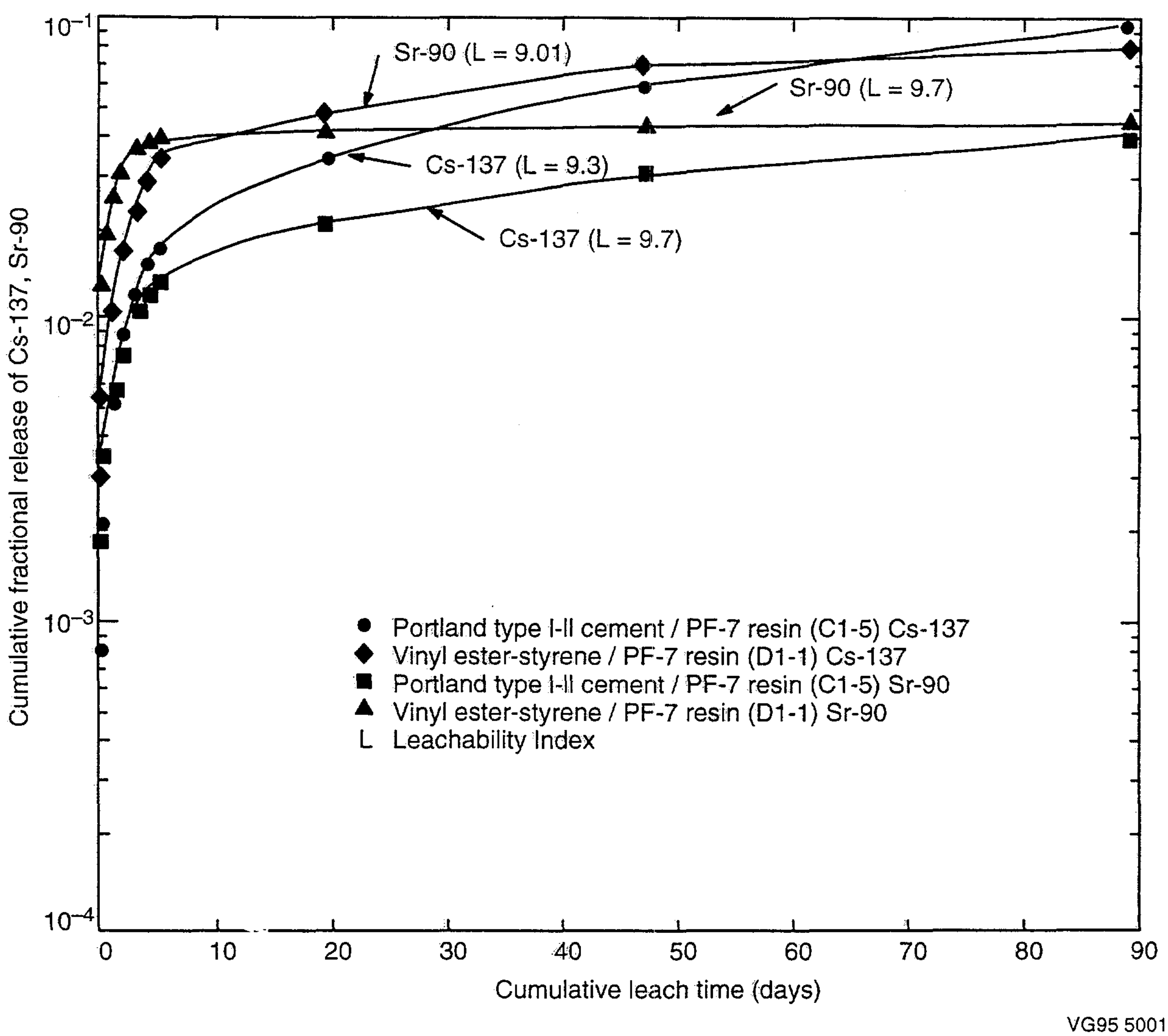


Figure 2. Irradiated EPICOR-II waste form radionuclide cumulative fractional release of Cs-137 and Sr-90 with demineralized water leachant.

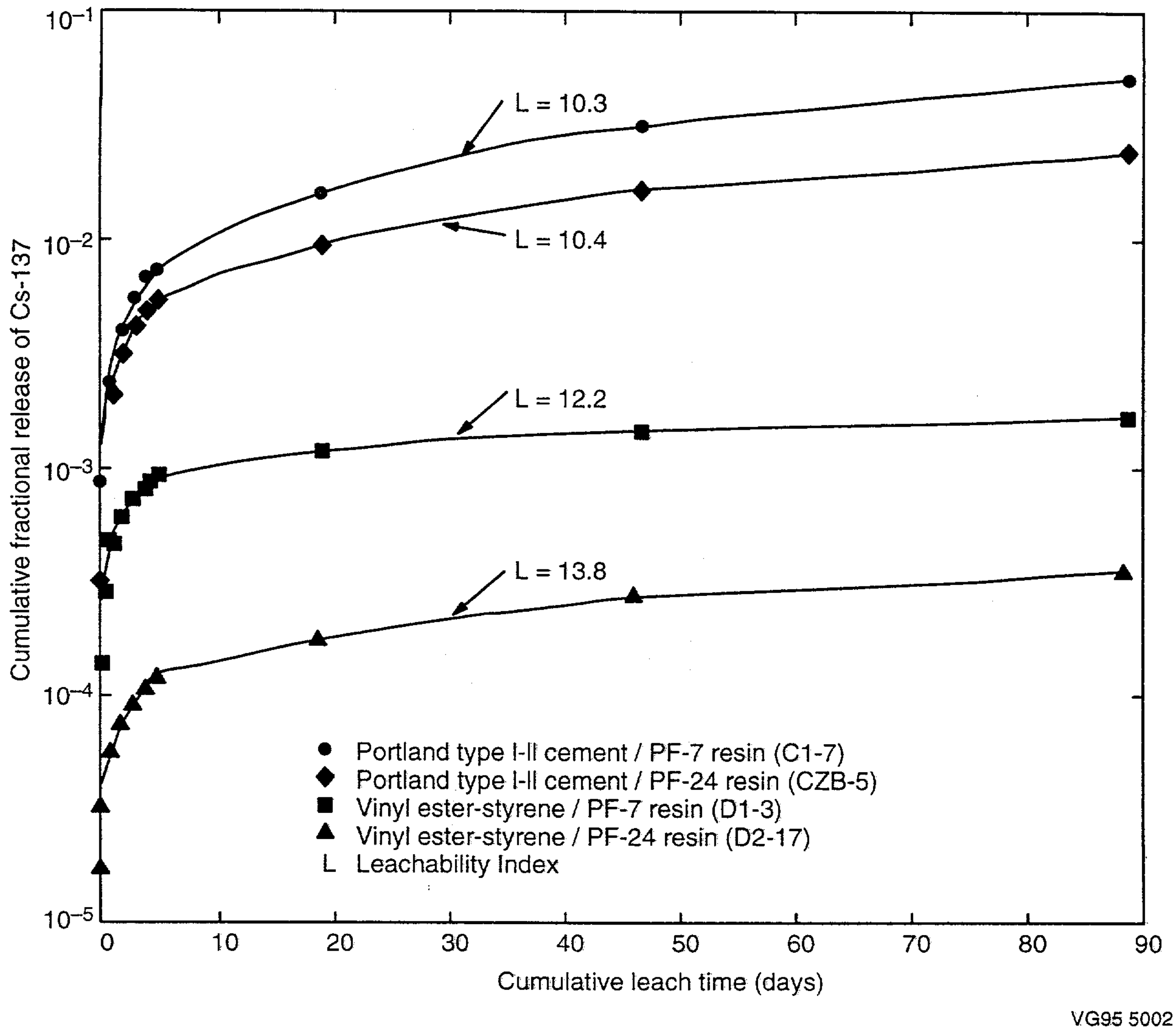


Figure 3. Cumulative fractional release of Cs-137 from unirradiated EPICOR-II waste forms with demineralized water leachant.

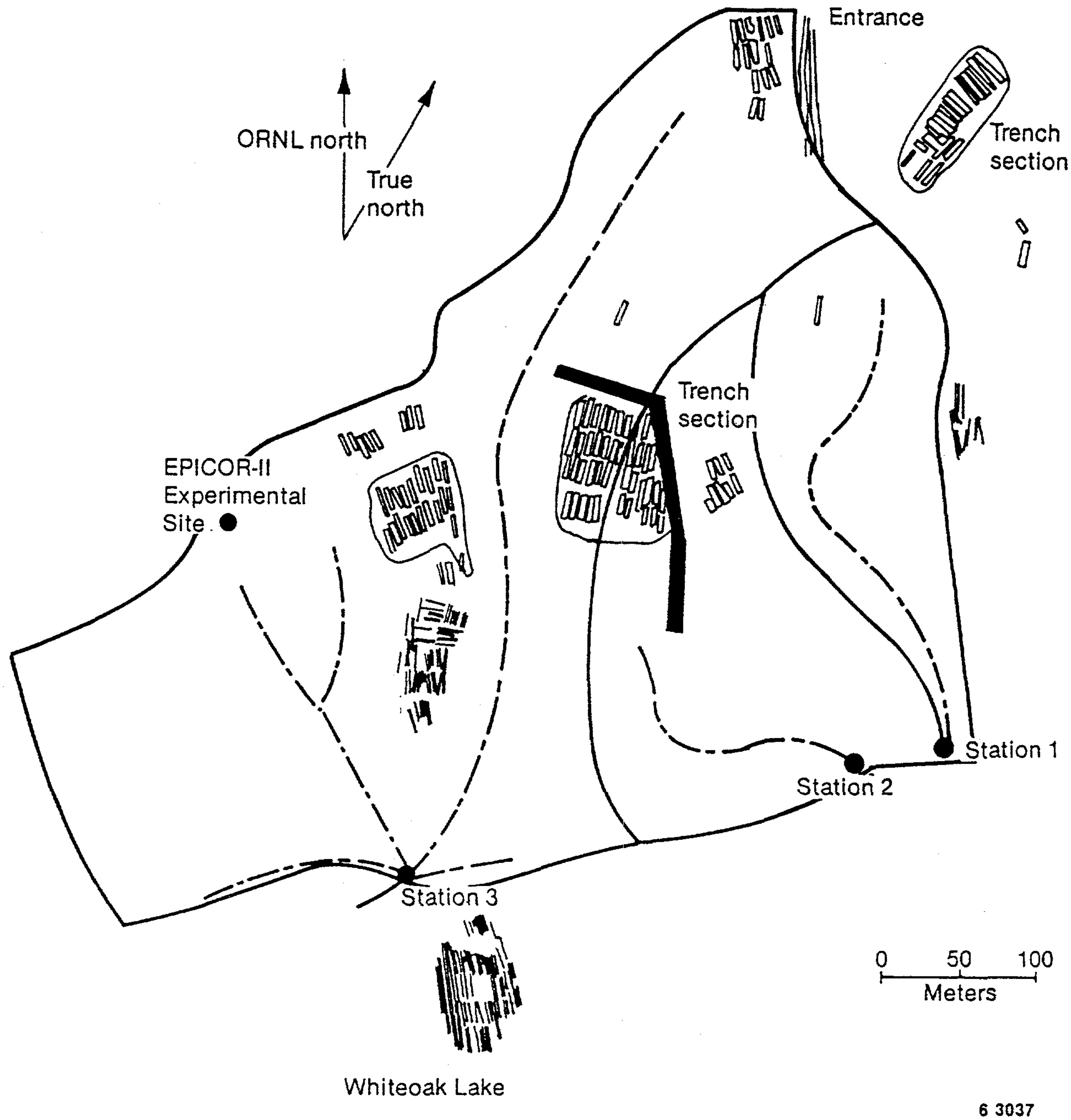


Figure 4. Location of the EPICOR-II lysimeter experiment at ORNL.

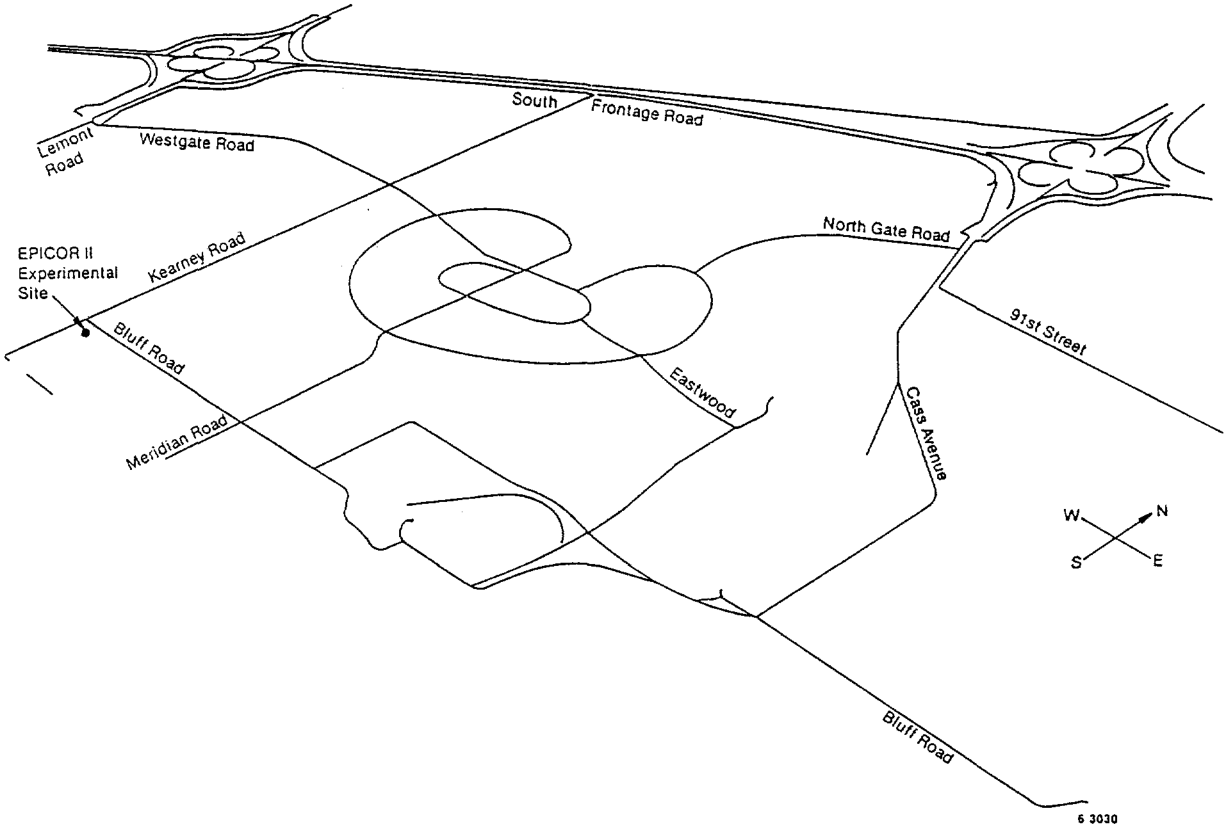


Figure 5. Location of the EPICOR-II lysimeter experiment at ANL-E.

ANL-E is located 43 km southwest of Chicago, Illinois, and 39 km due west of Lake Michigan. It has terrain that is gently rolling and partially wooded, which was formerly prairie and farm land. The area around the testing site has been allowed to return to natural vegetation, while the soil surface of each lysimeter has been weeded frequently to prevent the growth of any vegetative cover. The climate is that of the upper Mississippi Valley, as moderated by Lake Michigan. On average, temperatures of 0°C or colder prevail during the months of December through February, with temperatures near or slightly above 20°C during June through August. The average frost line in soil is 89 cm during the cold months. Precipitation (an average of 85.2 cm) appears to be uniformly distributed during the year, with May through September being the wettest months.[19]

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.[20]

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

Description of Lysimeters

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

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

The lysimeters at each site are consecutively numbered 1 through 5; lysimeters 1 through 4 contain soil, and number 5 is used as a control and is filled with an inert silica oxide sand.[15] Each lysimeter contains seven waste forms stacked end to end vertically. Table 5 shows which type of waste form was placed in each lysimeter.

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

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

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

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

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

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

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

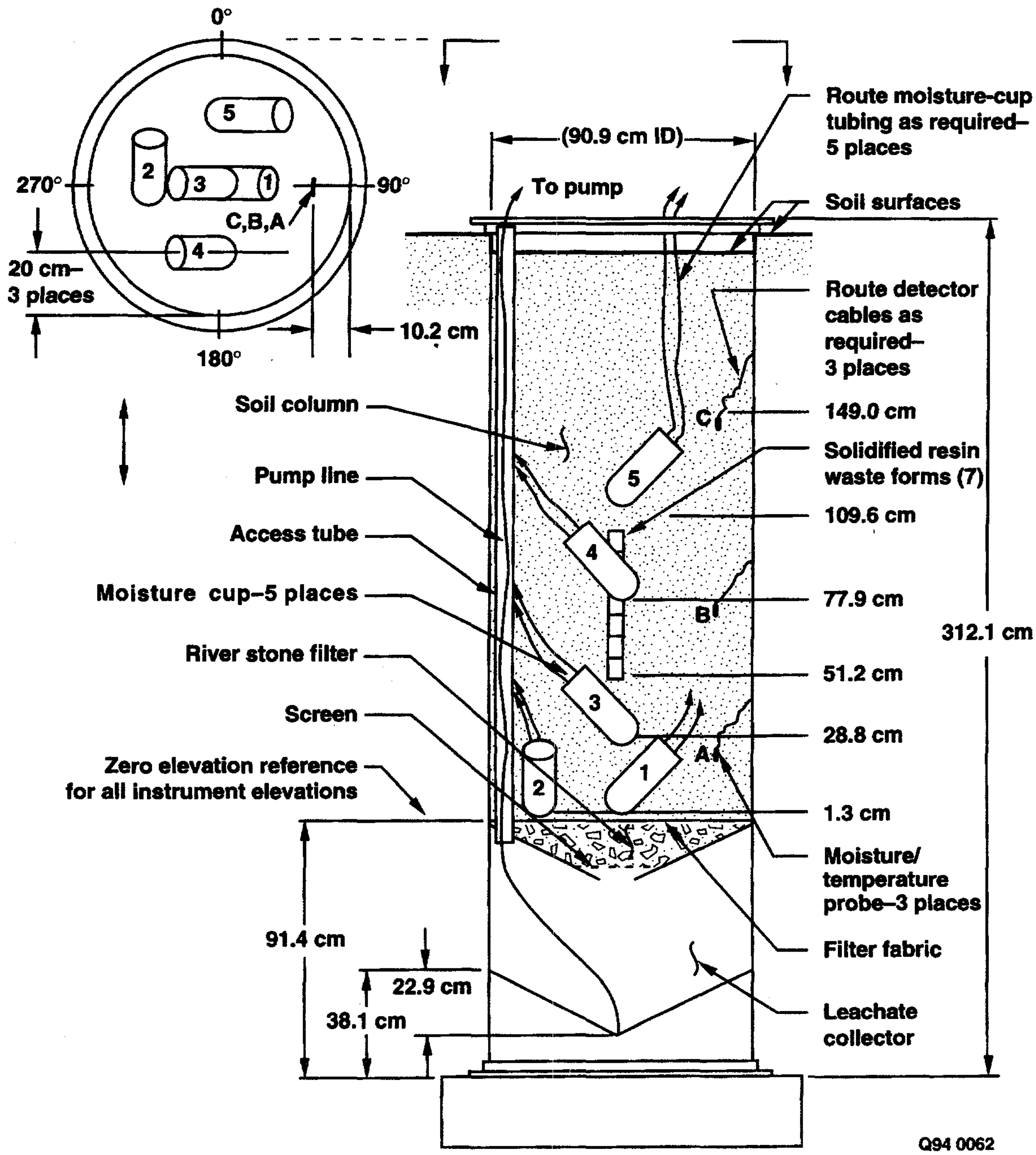


Figure 7. Lysimeter vessel component locations.

Table 5. Lysimeter waste form composition.

Lysimeter number	Fill material	Waste form description	Waste form inventory (pCi)
1	Soil	Cement with PF-7 resin waste	3.1E+11
2	Soil	Cement with PF-24 resin waste	14.3E+11
3	Soil	VES with PF-7 resin waste	4.6E+11
4	Soil	VES with PF-24 resin waste	19.3E+11
5 ANL-E	Silica oxide	Cement with PF-7 resin waste	3.1E+11
5 ORNL	Silica oxide	Cement with PF-24 resin waste	14.3E+11

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

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

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

b. Not available.

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

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

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

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

Table 8. Properties of Unimin silica oxide sand.

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

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

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

Data Collection and Analysis

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

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

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

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

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

The cassette tape is retrieved from the DAS each month and translated to an IBM PC compatible disk file using a Campbell Scientific C20 cassette interface. Once transferred to disk, the data are arranged in tables (see Table 11 for example). These files are printed in either text or graphic format. The graphic 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 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 1993. In addition, information on water balance, nuclide, and cation/anion content in soil water and leachate is presented. Many of the data are displayed in graphic format so that information can easily be correlated with time. This information has been presented on an annual basis in References 6, 7, 8, 9, 11, 12, 13, and 14.

Each DAS functioned fairly well during the second 4 years. However, there were three periods of time when the DAS was not in operation at ORNL. They were from July through September 1, 1991, from mid-June through August 17, and again in September of 1992. All were equipment failures requiring repair. There was another period of time during the month of January 1992 when data recorded by the DAS appeared to be incorrect.

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 75.7 cm at ANL-E and 140.2 cm at ORNL. ANL-E was at 89% of the normal annual rainfall[19] of 85.2 cm, while ORNL was near the normal annual rainfall[20] of 138.8 cm. The monthly precipitation pattern for each site can be seen from the histograms in Figures A-1 through A-4 and Figures A-14 through A-17 in Appendix A. Figure 8 shows the cumulative precipitation for both sites since the initiation of field work.

In 1990, ANL-E, for the first time since 1985, was well above the normal annual rainfall while ORNL was 110% of the normal annual rainfall. This was the second time in 4 years that ORNL

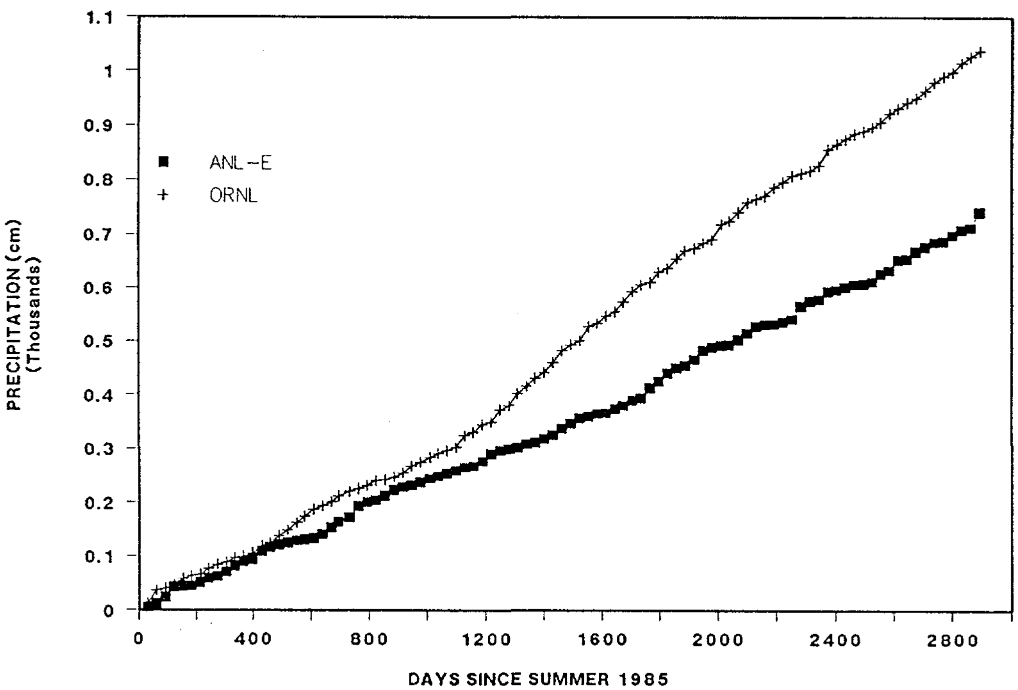


Figure 8. ANL-E and ORNL cumulative precipitation.

equalled or exceeded the normal amount of yearly precipitation. In 1991, for the third time in 5 years, ORNL equalled or exceeded the normal amount of yearly precipitation. In 1992, both sites were below the normal annual rainfall. This was the third time in 6 years that ORNL had not equalled or exceeded the normal amount of yearly precipitation. In 1993, ANL-E was above the normal annual rainfall while ORNL was nearly equal to the norm. This is the sixth time in the past 7 years that ORNL has equalled or exceeded the normal amount of yearly precipitation. By the end of this reporting period, there was a cumulative precipitation total of 742.1 cm at ANL-E, while ORNL received a total of 1,038.6 cm.

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

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

Air temperature data from ANL-E show that periods of freezing temperatures occurred each year from late October until mid-April. ORNL experienced periods of freezing temperatures from early November until early March during the reporting period (Figures A-5 through A-8 and Figures A-18 through A-21).

Lysimeter Soil Temperature Data

Soil temperature and moisture sensors (probes) are physically located within a common housing or probe. These probes are located at three elevations: 149, 77.9, and 28.8 cm, as measured from the bottom of the soil column within each lysimeter (Figure 7). The function of these probes is to provide data on the physical environment experienced by the buried waste forms, specifically, whether or not they experience freezing temperatures and if the surrounding soil is moist. Because all of the soil lysimeters at each site are exposed to the same environment, the current placement of probes provides a planned redundancy of collected data. Therefore, as long as there are functioning probes in any of the soil lysimeters at each site, data sufficient to satisfy reporting criteria will be available. In addition, temperature data collected during the years of extended service life of the probes will serve as a useful climatological reference for assessing waste form performance in future years.

The lysimeter soil temperature data recorded at ANL-E and ORNL during the reporting period are shown in Figures B-1 through B-36 of Appendix B. The only probe to record a valid freezing temperature was at the 149-cm elevation in ANL-1 (Figures B-1 through B-4). The 28.8-cm probe data for 1993 are erroneous (Figure B-4). A direct correspondence can be seen between air temperature and soil temperatures at both sites.

As stated in past reports, a number of temperature probes at ANL-E have failed. During the last 5 years, all the temperature probes in ANL-4 and one in ANL-2 had failed to function; therefore, data from these probes were not included in this report. During the reporting period, it appeared that two of the probes in ANL-3 as well as one in ANL-5 were not functioning properly, and those data are not reported. Partial deterioration of the remaining ANL-3 probe was seen during that period. The probes have probably been damaged by corrosion of the metal parts (Reference 7). At the present time, a more damage-resistant replacement for these probes has not been found. Occasional erratic behavior of some ORNL probes seen during 1991 and 1992 has been reduced to a single spike on several outputs. The bottom temperature probes in ORNL-3 and -5 have consistently indicated elevated soil temperature (Figures B-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 but continues to read high. 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.

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.

The probe output from the soil column of each lysimeter over time (as determined by averaging the outputs of the three probes in each lysimeter) showed that the variation in detected moisture among the lysimeters at each site was relatively similar and not excessive (Table 12). There was a coefficient of variation maximum (CV) of 37.4% at ANL-E and 20.8% at ORNL. The probes continue to serve their original purpose of providing some indication of lysimeter soil moisture. As was mentioned in the section on soil temperature, some of the combined moisture/temperature probes at ANL-E are no longer functioning. This condition was discussed in the previous section.

Soil moisture in the soil column of the lysimeters at each site is quantified gravimetrically once each year (see Tables D-1 through D-8 of Appendix D). Some idea of the accuracy of the soil moisture probes can be calculated by comparing the once-a-year gravimetric soil moisture data of each soil lysimeter to yearly averaged probe data (Table 12). Percent differences between the gravimetric data and moisture probe data for ANL-E lysimeters range between a low of 3.2% in 1991 to a high of 43.8% in 1992. These values have decreased significantly during this reporting period, and are well within a reasonable range given the use of the information. As in the past, data from the ORNL probes continue to overestimate the actual percent soil moisture from a low of 56.3% in 1993 to a high of 133.1% in 1991.

In addition to using the moisture probe and gravimetric data to calculate soil moisture starting the summer of 1991, a neutron moisture-detecting probe was used at ANL-E. Operation of the neutron probe, using 1991 calibration curves, produced data that were comparable to gravimetric overall average values within 9.1%, but underestimated those values (see Tables D-1 through D-4 of Appendix D). A new calibration curve using 1992 data decreased those variations to 4.8% underestimated. The variability between actual and measured moisture may be caused by the neutron probe integrating moisture data that were simultaneously measured both inside and outside the lysimeter. It appears that these soils vary in moisture content, with the outside soil being drier. Neutron probe measurements were first made at ORNL in 1992. Those data are given in Tables D-5 through D-8 of Appendix D. Comparison of the ORNL neutron probe results to gravimetric results, in overall average values, shows that the probe underestimated by 1.7%. In spite of the difference between actual and measured soil moisture at ANL-E, the accuracy appears very good at ORNL. Therefore, it can be said that the use of the neutron probe provides a rapid, accurate estimate of moisture in the soil column.

Soil moisture (as gravimetrically determined) at each sampling depth has remained uniformly consistent between intrasite lysimeters during the past several years (Figures 9 and 10). The uniformity of soil moisture in the ANL-E lysimeters (Figure 9) continues to be of interest given the long-term, nonuniform decrease in water infiltration into the ANL-E soil lysimeters. Lysimeters 1 and 2 appear to have less stored water than 3 and 4 (Table 12). While action to improve drainage of the ANL-E lysimeters was taken early in the

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

Lysimeter number	Period	Average percent moisture for soil column probes[a]	Average percent moisture for soil column[b]	Percent difference
ANL-1	1989–90	15.4 ± 2.6	21.4 ± 1.9	28.0
ANL-2		14.5 ± 1.7	22.2 ± 1.8	34.7
ANL-3		16.2 ± 8.5[c]	24.1 ± 1.0	32.8
ANL-4		14.6 ± 1.7	23.4 ± 0.7	37.6
ORNL-1		39.0 ± 0.6	17.4 ± 1.0	124.1
ORNL-2		39.6 ± 0.2	17.4 ± 0.8	127.6
ORNL-3		34.0 ± 1.7	17.4 ± 1.4	95.4
ORNL-4		37.2 ± 3.9	17.6 ± 1.1	111.4
ANL-1	1990–91	14.7 ± 3.3	18.2 ± 3.4	19.2
ANL-2		15.0 ± 1.0	17.1 ± 4.2	12.3
ANL-3		19.6 ± 8.3[c]	19.0 ± 3.4	3.2
ANL-4		17.3 ± 6.4	20.2 ± 3.4	14.4
ORNL-1		38.0 ± 1.7	17.1 ± 1.3	122.2
ORNL-2		39.4 ± 1.1	16.9 ± 1.1	133.1
ORNL-3		34.2 ± 0.9	16.7 ± 1.0	104.8
ORNL-4		36.4 ± 2.7	16.8 ± 1.2	116.7
ANL-1	1991–92	15.1 ± 3.6	21.8 ± 1.8	30.7
ANL-2		16.7 ± 3.4	21.3 ± 1.4	21.6
ANL-3		13.1[c]	23.3 ± 1.7	43.8
ANL-4		14.2 ± 4.2	22.6 ± 1.0	37.2
ORNL-1		32.4 ± 1.5	16.2 ± 2.4	100.0
ORNL-2		35.7 ± 2.3	15.7 ± 2.8	127.4
ORNL-3		32.2 ± 1.3	18.6 ± 1.0	73.1
ORNL-4		34.9 ± 1.1	18.8 ± 2.4	85.6
ANL-1	1992–93	15.7 ± 3.7	22.8 ± 3.2	31.1
ANL-2		15.9 ± 2.1	21.0 ± 2.2	24.3
ANL-3		24.6[d]	23.0 ± 2.1	6.5
ANL-4		17.1 ± 7.0	24.8 ± 2.5	31.0
ORNL-1		26.1 ± 9.6	16.7 ± 2.3	56.3
ORNL-2		34.6 ± 3.2	15.9 ± 1.4	117.6
ORNL-3		33.8 ± 1.0	18.5 ± 0.6	82.7
ORNL-4		37.0 ± 2.6	19.1 ± 1.7	93.7

a. July through June.
b. Determined gravimetrically for July 1990.
c. Average from two probes.
d. Average from one probe.

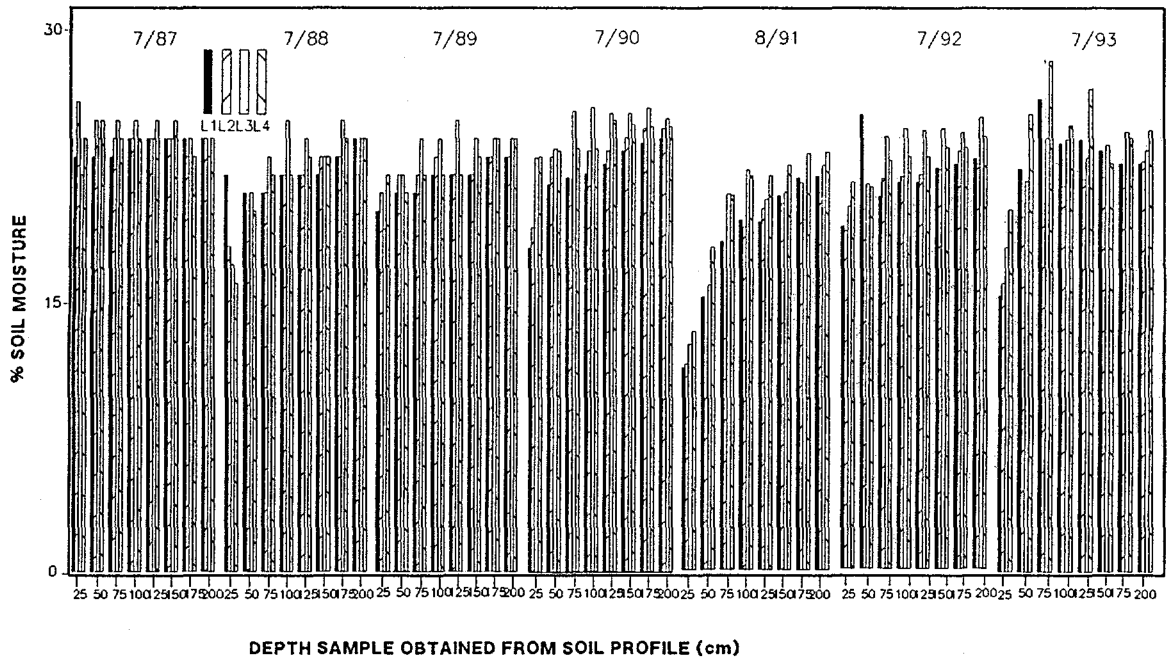


Figure 9. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.

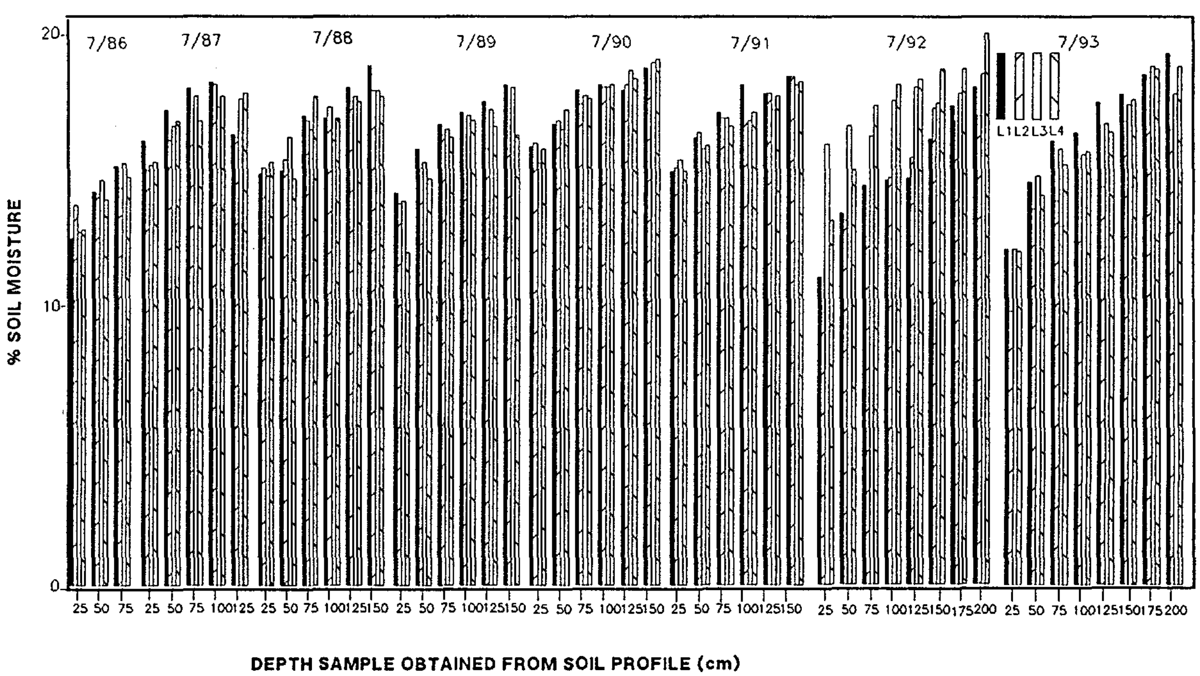


Figure 10. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.

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

As shown in Figures 9 and 10, the amount of moisture within the deeper horizons of the lysimeter soil columns at each site appears to have remained fairly constant (see Tables D-1 through D-8). At the time of the 1993 sampling, the average soil moisture of ANL-E soils had increased from a low of 45.1% to 56.3% of the soil moisture holding capacity in 1991, while at ORNL, this value remained approximately the same: a low of 38.0% for 1991 and a high of 39.4% for 1993. These values have remained fairly constant from year to year.

Measurement of Leachate

By using the cumulative rainfall data from each site since the time the lysimeters were placed in operation (Figure 8), it is possible to calculate the approximate volume of water that has been received by the exposed surface (6,489.5 cm^2) of each lysimeter. The cumulative volume of precipitation received by each ANL-E lysimeter was 4,815.8 L; at ORNL, this value was 6,739.9 L. Precipitation per year is listed in Table 14 as well as average volume of leachate through the lysimeters. The volume of the precipitation that has passed through the lysimeters can be seen graphically in Figures 11 and 12. The throughput of precipitation is dependent on site conditions and lysimeter fill material. At ANL-E, an average of 1,939.2 ± 872.8 L or 40.3% of total precipitation passed through the soil lysimeters, while for the control, this value was 4,829.0 L or 100.3% of the calculated available precipitation. For ORNL, the values were 6,050.5 ± 45.6 L (89.8%) for the soil-filled lysimeters and 6,910.0 L (102.5%) 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, which are shown in Figures 11 and 12). Also, the small deviation in total yearly leachate throughput with the ORNL soil lysimeters (0.8%) continues to demonstrate that these lysimeters perform as a unit as compared to the individual drainage activity of the ANL-E lysimeters.

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

	Water removed from lysimeter surfaces (L)			
Lysimeter number	1990	1991	1992	1993
ANL-1	431	395	154	495
ANL-2	428	363	110	452
ANL-3	—	74	—	76
ANL-4	210	273	82	393

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

		ANL-E		ORNL	
	Test period	Cumulative volume (L)	Total (%)	Cumulative volume (L)	Total (%)
Precipitation received	1989–90	2,769	—	4,138	—
	1990–91	3,451	—	5,009	—
	1991–92	3,886	—	5,791	—
	1992–93	4,816	—	6,740	—
Average leachate passed through soil–filled lysimeters	1989–90	1,214 ± 437	43.8	3,512 ± 21	84.8
	1990–91	1,469 ± 608	42.6	4,509 ± 34	90.0
	1991–92	1,665 ± 682	42.9	5,199 ± 39	89.8
	1992–93	1,939 ± 873	40.3	6,051 ± 46	89.8
Leachate passed through sand–filled lysimeters	1989–90	2,761	99.7	4,084	98.7
	1990–91	3,529	102.3	5,203	103.9
	1991–92	3,955	101.7	5,983	103.3
	1992–93	4,829	100.3	6,910	102.5

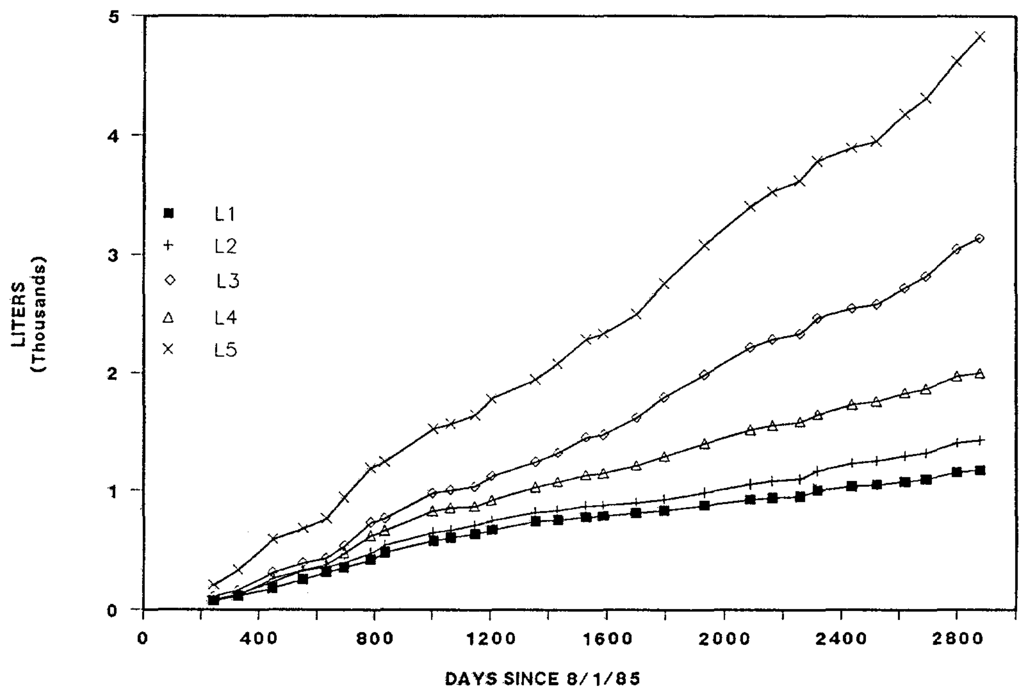


Figure 11. ANL-E cumulative volume of leachate from lysimeters.

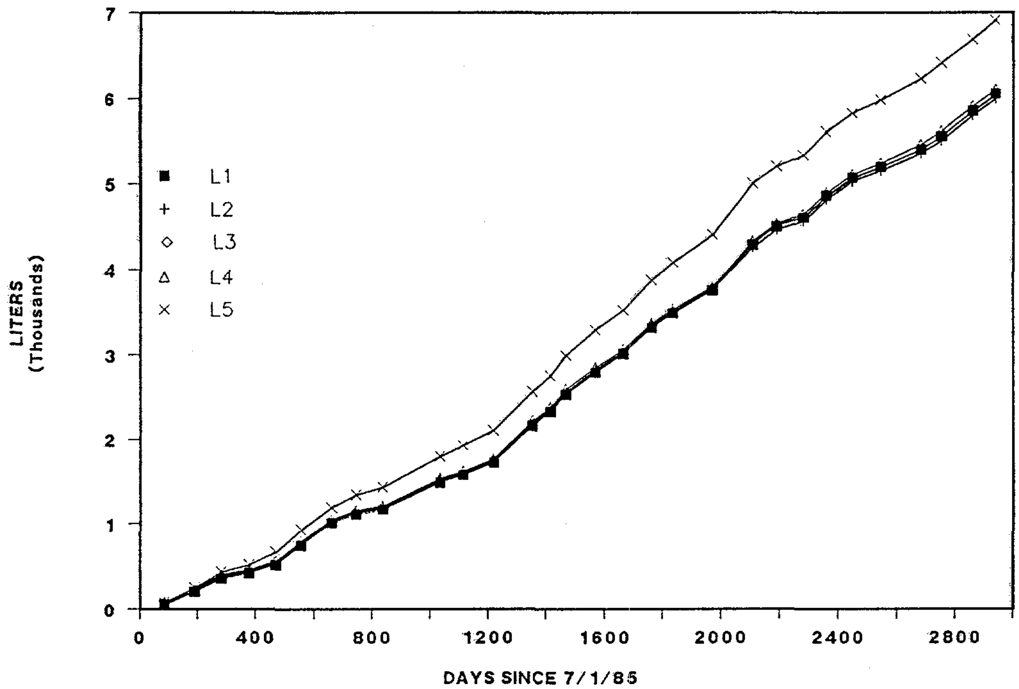


Figure 12. ORNL cumulative volume of leachate from lysimeters.

The total volumes of precipitation that have moved through the lysimeters represent an average 2.74 pore volumes for the ANL-E soil lysimeters and 8.52 pore volumes for soil lysimeters at ORNL, while the controls at ANL-E and ORNL were 10.5 and 10.72 pore volumes, respectively. These data show that the ORNL soil lysimeters have had an average of three times more water pass through them as those at ANL-E. The lysimeters at each site received comparable volumes of water; however, those quantities did not move through the lysimeters at each site in equal amounts due to these differences in soil texture and to weather conditions (Figures 11 and 12).

Soil used at ANL-E is heavier (contains more fine material such as silts and swelling clay) than the soil used at ORNL.[20] Therefore, infiltration and percolation of water through the ANL-E soil would be expected to be significantly reduced in comparison to ORNL soil. The effect of weather is not apparent when comparing the sand-filled control lysimeters at the two sites. At both ANL-E and ORNL, 100% of the volume of precipitation passed through those lysimeters. At ANL-E, precipitation came during the months of November through March when the average air temperature was below 0°C. This precipitation then was in the form of freezing rain or snow that would not penetrate the frozen soil surface and could have been blown off (in the case of snow) or lost due to sublimation. Other factors such as generally gustier winds and lower humidity at ANL-E indicate that evaporation of water from the ANL-E lysimeters could have been higher than at ORNL. (Wind speed and relative humidity for ANL-E and ORNL are shown in Appendix A.) Also as noted earlier, ANL-E lysimeters 1, 2, 3, and 4 have experienced water ponding during periods of heavy rainfall. To prevent loss of precipitation, that water was drained from the surface of those lysimeters.

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

Radionuclide Analysis

Water samples are normally collected on a quarterly basis from leachate collectors and moisture cups in each of the lysimeters during 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 7 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. During the first 5 years of operation, water samples from only cups 3 were routinely analyzed at the sites. However, starting in 1991, water from cups 1 has been analyzed and reported. In 1993, water from cups 2 has also been analyzed and reported, and ORNL analyzed cups 5 water in the last quarter of 1992.

Tabulated results of beta and gamma analysis for the samples taken during the period are found in Tables E-1 through E-8 in Appendix E. Four samples were taken at each site during each 12-month period, except only three samples were taken at ORNL in FY-91. The cumulative amounts of nuclides found in water samples obtained from lysimeter number 3 cups and leachate collectors for all sampling periods are given in Tables 15 and 16 and displayed graphically in Figures 13 through 21.

As has been reported in the past,[6–14] not all nuclides are appearing consistently in the water obtained from either the cups or leachate collectors. The nuclide that appears with the most regularity at both sites is Sr-90 (Tables 15 and 16 and Appendix E). This nuclide consistently occurs in significant amounts in all the number 3 cups at ANL-E and ORNL, and in the number 5 leachate collectors at both sites (Figures 13 through 16). There continues to be standout amounts of Sr-90 retrieved from cup 3 samples at both sites. Those include a cumulative total of 1,411,575 pCi from 3-3 at ANL-E (Table 15 and Figure 13) and 117,617 pCi from 3-3 at ORNL, which is now well above ORNL 1-3 (Table 16 and Figure 14). The releases into ANL 3-3, ORNL 1-3, and ORNL 3-3 are almost linear, indicating a continuance of an established rate of release. In addition, the increase in Sr-90 release continues in ORNL 5-3 as well as in ORNL 4-3 (Figure 14). The above data show that significant quantities of Sr-90 continue to be transported from the waste forms.

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

At ANL-E, Sr-90 retrieved from number 3 cups of the soil lysimeters during the fourth year ranges from 74% to 3,200% of that found in the leachate collectors (Table 15), while at ORNL, these values are between 0.1% and 70% (Table 16). These are increases over previous years and are the result of both an increased quantity of Sr-90 moving into the area near the moisture cups and a decrease in the movement of the nuclide through the entire soil profile into the leachate collectors.

During the past 3 years, amounts of Sr-90 in leachate water from the control (sand-filled) lysimeters at each site have remained similar and at least one order of magnitude larger than the largest cumulative release from a soil lysimeter (Figures 15 and 16). This is comparable to the previous year's findings (References 6, 7, 8, 9, 11, 12, 13, and 14). For leachates from soil lysimeters, intersite-comparable percentages of total inventory of Sr-90 were found in ANL-E 1, 2, 3, and 4 and ORNL 2, 3, and 4 (Table 17). There was a significant increase in the total cumulative

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

Test period	Operating days	Sr-90 in moisture cups (pCi)					Sr-90 in leachate collectors (pCi)					Cs-137 in moisture cups (pCi)	
		1-3	2-3	3-3	4-3	5-3	1	2	3	4	5	2	5
1989–90	1,526	4,953.1	1,913.2	329,340.8	1,255.5	4,591.4	4,969.6	1,617.5	9,416	50.4	401,566.3	1,872	2,529
	1,586	5,200.1	2,069.2	362,840.8	1,707.5	4,981.4	4,969.6	1,617.5	9,460	284.4	418,368.3	2,028	2,979
	1,698	5,338.1	2,317.2	452,840.8	2,105.5	5,881.4	4,969.6	1,617.5	11,087	284.4	492,767.3	2,284	4,269
1990–91	1,795	5,356.1	2,580.2	542,840.8	2,325.5	6,581.4	4,969.6	1,617.5	16,577	284.4	604,568.3	2,407	6,159
	1,931	5,665.1	2,922.2	594,347.8	3,039.5	7,016.4	4,969.6	1,617.5	24,065	284.4	785,933.3	2,439	7,088
	2,089	5,980.1	3,223.2	677,363.8	3,944.5	7,683.4	4,969.6	1,617.5	34,084	284.4	1,000,097.0	2,454	9,413
1991–92	2,166	6,102.1	3,723.2	747,622.8	5,387.5	8,900.4	4,969.6	1,617.5	36,775	2,408.4	1,071,521.0	2,515	13,347
	2,259	7,392.1	4,060.2	839,375.8	7,153.5	10,521.4	4,969.6	1,617.5	40,297	2,408.4	1,191,581.0	2,538	26,847
	2,319	8,960.1	4,628.2	985,375.8	9,753.5	12,621.4	4,969.6	1,617.5	47,393	2,736.4	1,214,342.0	2,611	33,047
	2,438	10,860.1	5,412.2	987,575.8	10,321.5	13,135.4	4,969.6	1,617.5	51,475	3,121.4	1,338,327.0	2,638	37,347
1992–93	2,522	10,860.1	5,980.2	1,095,575.0	13,021.5	16,135.4	4,969.6	1,755.5	53,102	3,121.4	1,379,815.0	2,689	46,547
	2,619	11,217.1	6,818.2	1,173,575.0	17,886.5	18,567.4	4,969.6	1,917.5	94,622	3,620.4	1,643,124.0	2,727	55,871
	2,692	11,721.1	7,415.2	1,243,575.0	21,397.5	20,932.4	6,624.6	1,955.5	116,464	3,839.4	1,813,050.0	2,778	62,021
	2,797	12,551.1	7,433.2	1,327,575.0	25,018.5	23,479.4	6,692.6	2,069.5	169,499	4,730.4	2,092,875.0	2,812	67,071
	2,876	14,332.1	7,923.2	1,411,575.0	29,799.5	24,607.4	6,692.6	2,069.5	189,413	4,826.4	2,283,525.0	2,855	67,071

Table 16. 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)				
		1-3	2-3	3-3	4-3	5-3	1	2	3	4	5
1989–90	1,568	21,339.8	3,653.4	6,973.3	54.5	67.4	24,629	10,351	16,735	9,865	130,186
	1,664	24,853.8	4,085.4	6,973.3	61.5	89.4	24,629	10,351	16,735	9,865	224,551
	1,762	28,907.8	4,869.4	6,973.3	79.5	135.4	39,770	10,351	16,735	9,865	329,095
	1,833	37,286.8	5,761.4	6,973.3	109.5	192.4	42,830	10,351	17,440	9,865	329,095
1990–91	1,971	46,476.8	6,788.4	16,974.3	171.5	267.4	50,069	10,351	17,440	9,865	506,543
	2,106	53,504.8	7,680.4	26,434.3	206.5	302.4	58,163	10,351	17,440	9,865	716,541
	2,188	61,604.8	8,706.4	39,934.3	354.5	1,382.4	72,905	13,078	17,440	20,524	810,339
1991–92	2,279	71,304.8	9,906.4	56,134.3	554.5	4,582.4	80,657	13,418	17,744	21,075	914,611
	2,357	81,304.8	11,206.4	68,534.3	778.5	7,282.4	131,680	13,418	17,744	21,075	1,108,363
	2,447	89,704.8	12,206.4	83,934.3	1,129.5	9,482.4	166,870	15,488	19,021	21,075	1,313,645
	2,544	99,204.8	13,406.4	104,434.3	1,588.5	12,682.4	196,102	16,263	19,021	21,075	1,496,751
1992–93	2,681	108,643.8	14,622.4	121,731.3	2,264.5	15,925.4	262,198	18,140	19,815	21,075	1,958,089
	2,752	116,211.8	15,460.4	136,055.3	2,859.5	15,931.4	337,528	21,143	21,287	21,075	2,301,590
	2,860	122,698.8	16,487.4	153,893.3	3,643.5	16,958.4	466,696	25,508	22,305	21,075	2,529,481
	2,939	131,076.8	17,622.4	177,677.3	4,427.5	18,390.4	563,410	29,776	23,803	21,075	2,716,321

Test period	Operating days	Cs-137 in moisture cups (pCi)					Cs-137 in leachate collectors (pCi)				
		1-3	2-3	3-3	4-3	5-3	1	2	3	4	5
1989–90	1,568	0	0	0	0	293	4,040	2,040	3,098	2,020	119,856
	1,664	0	0	0	0	336	4,040	2,040	3,098	2,020	128,710
	1,762	0	0	0	0	387	5,300	2,040	3,098	2,020	128,710
	1,833	0	0	0	0	482	5,300	2,040	3,098	2,020	133,198
1990–91	1,971	0	0	0	0	571	5,300	2,040	3,098	2,020	141,103
	2,106	0	0	0	0	798	5,300	2,040	3,098	2,020	173,454
	2,188	0	0	0	0	1,095	5,300	2,040	3,098	2,020	203,677
1991–92	2,279	0	0	0	0	1,392	5,300	2,040	3,098	2,020	213,386
	2,357	0	0	0	0	1,524	5,300	2,040	3,098	2,020	229,118
	2,447	0	0	0	0	1,686	5,300	2,040	3,098	2,020	229,118
	2,544	0	0	0	0	2,010	5,300	2,040	3,098	2,020	234,508
1992–93	2,681	0	0	0	0	2,686	5,300	2,040	3,098	2,020	254,086
	2,752	0	0	0	0	2,697	5,300	2,040	3,343	2,020	259,098
	2,860	0	0	0	0	3,129	5,300	2,040	3,343	2,020	266,404
	2,939	0	0	0	0	3,994	5,300	2,040	6,415	6,178	292,324

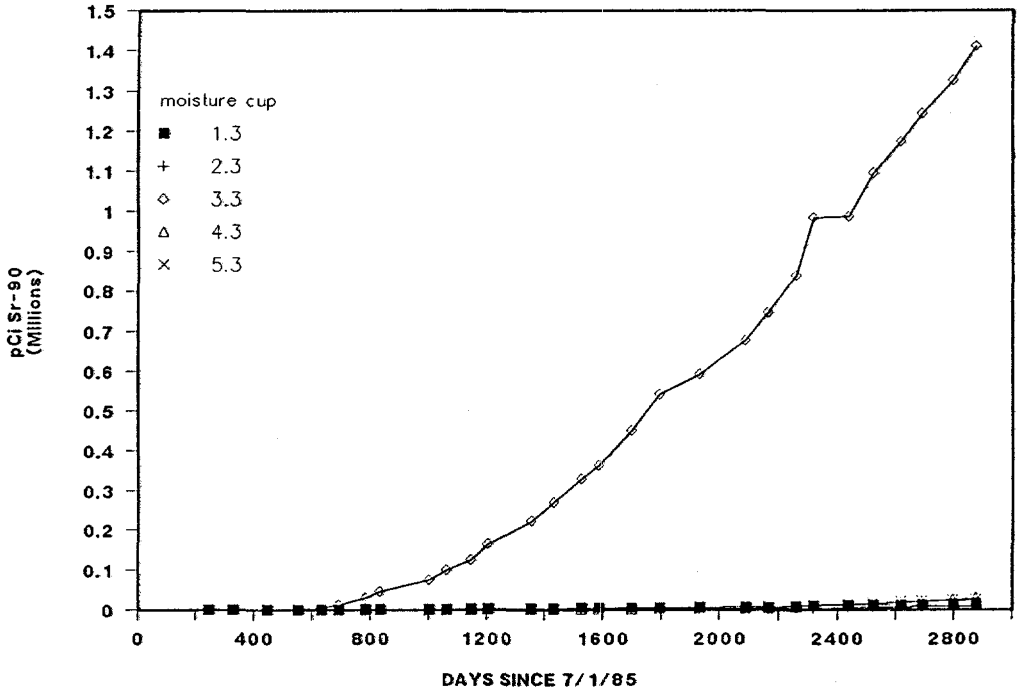


Figure 13. ANL-E cumulative Sr-90 collected in moisture cups number 3.

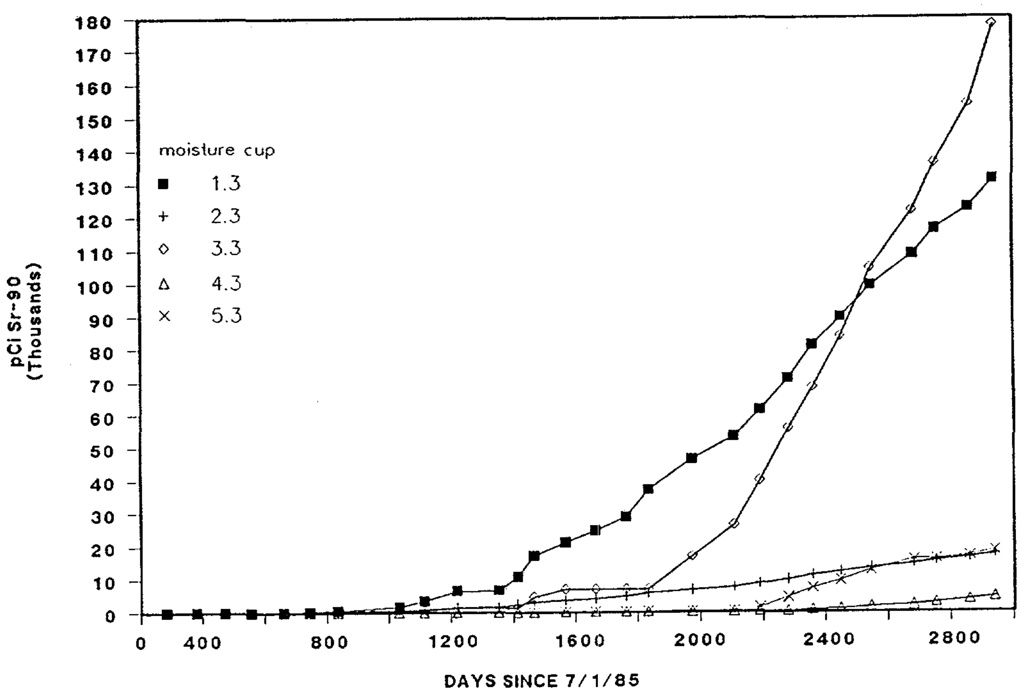


Figure 14. ORNL cumulative Sr-90 collected in moisture cups number 3.

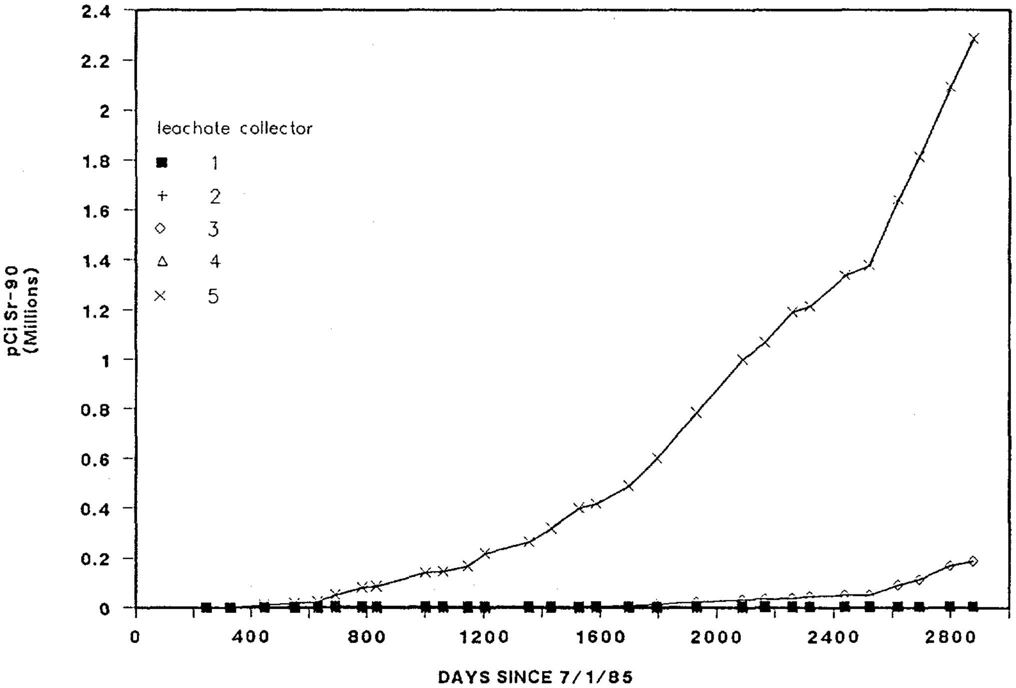


Figure 15. ANL-E cumulative Sr-90 collected in lysimeter leachate collectors.

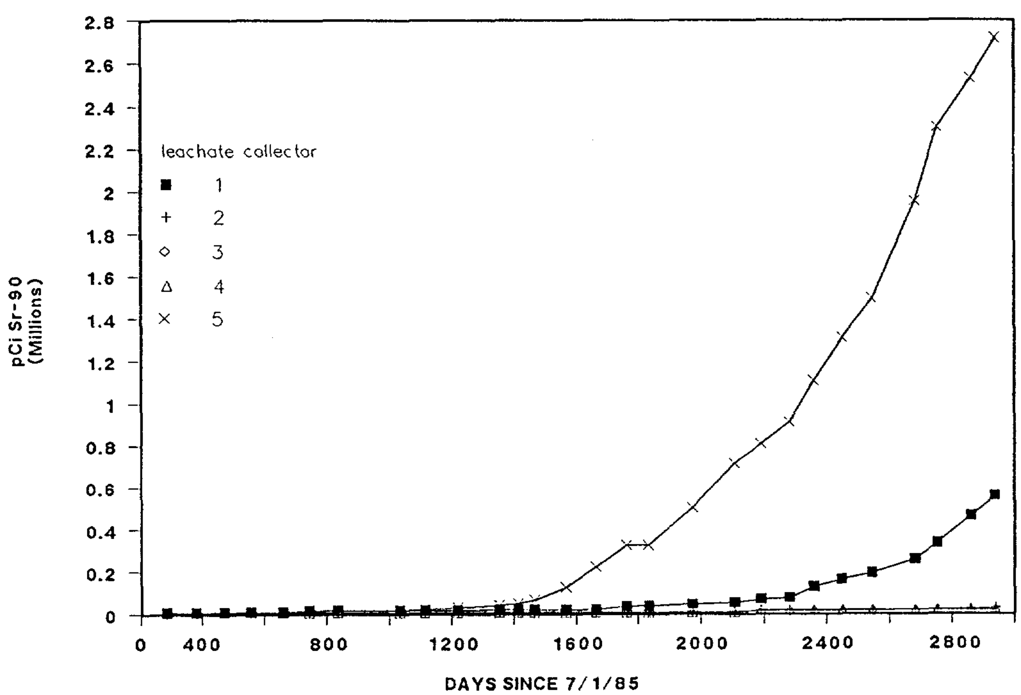


Figure 16. ORNL cumulative Sr-90 collected in lysimeter leachate collectors.

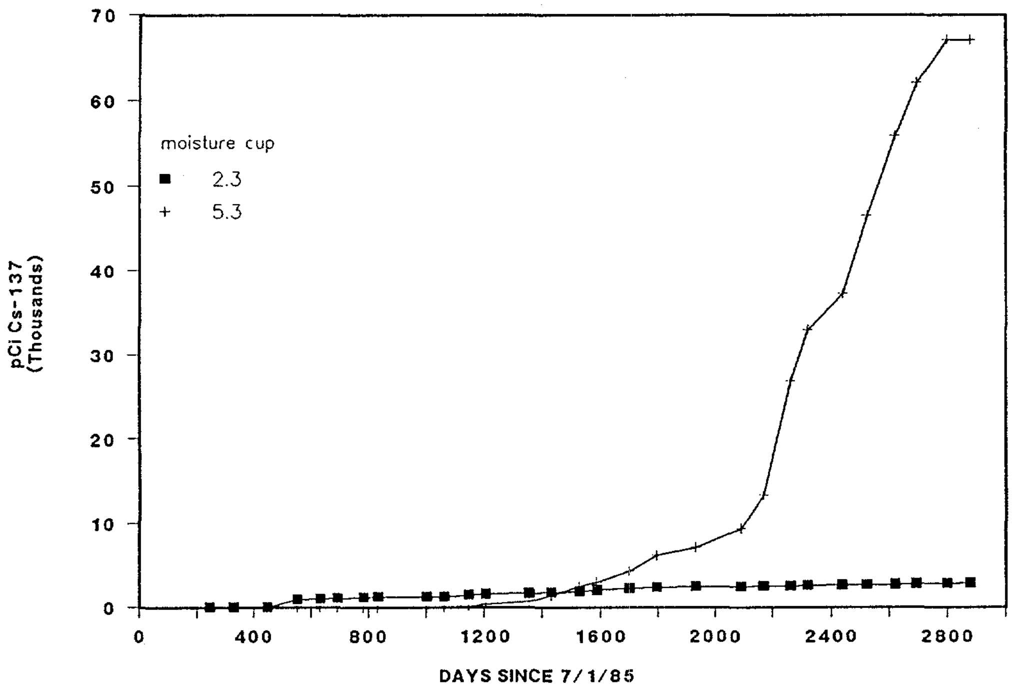


Figure 17. ANL-E cumulative Cs-137 collected in moisture cups number 3.

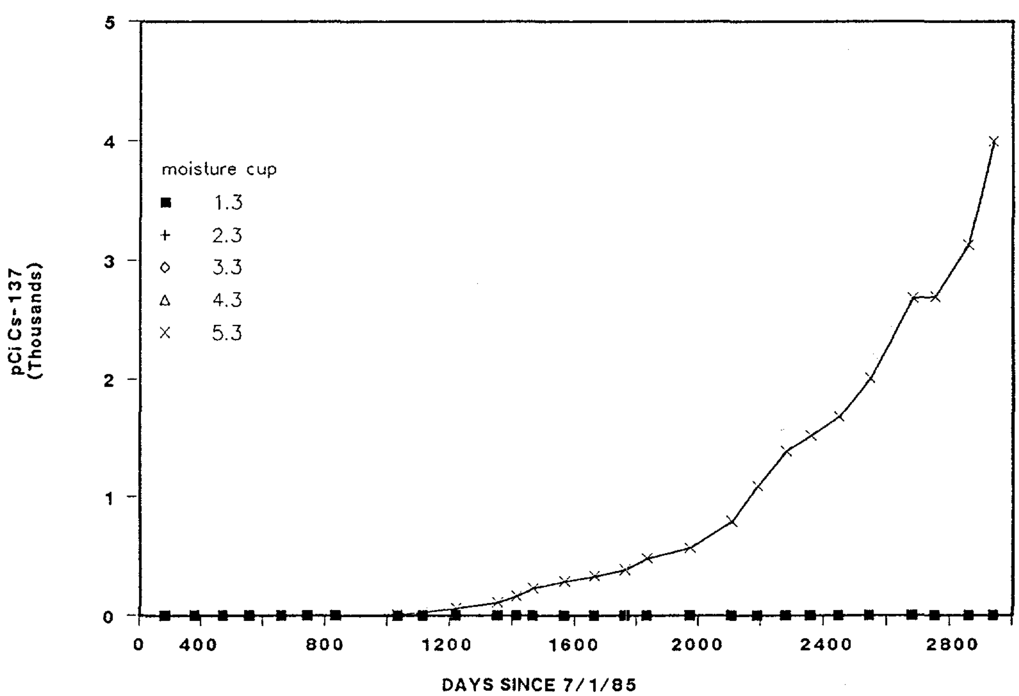


Figure 18. ORNL cumulative Cs-137 collected in moisture cups number 3.

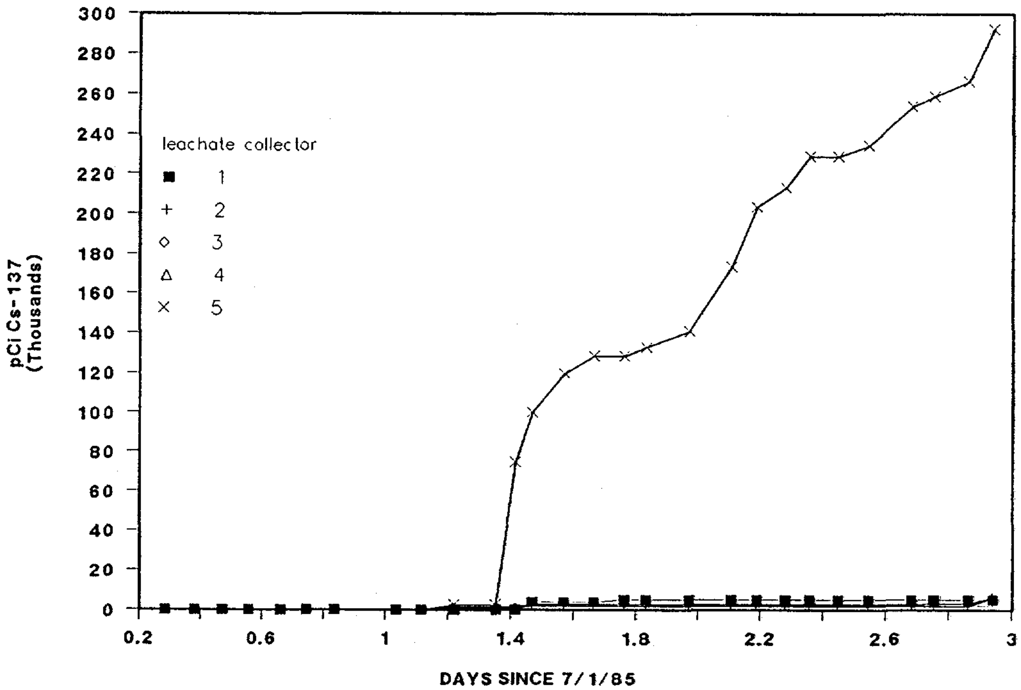


Figure 19. ORNL cumulative Cs-137 collected in lysimeter leachate collectors.

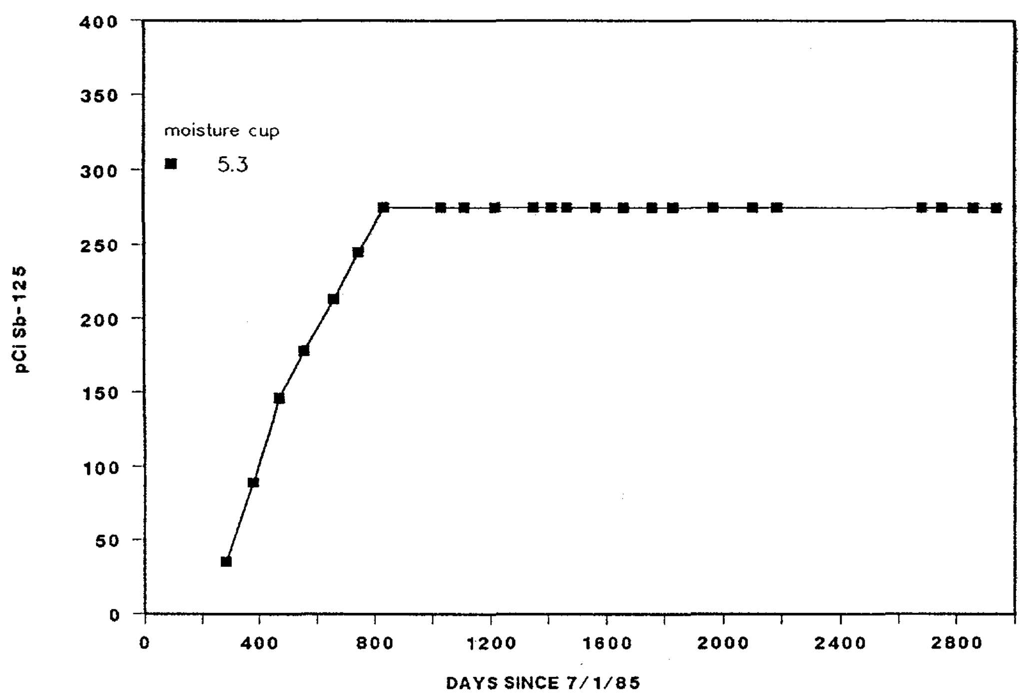


Figure 20. ORNL cumulative Sb-125 collected in moisture cup number 3.

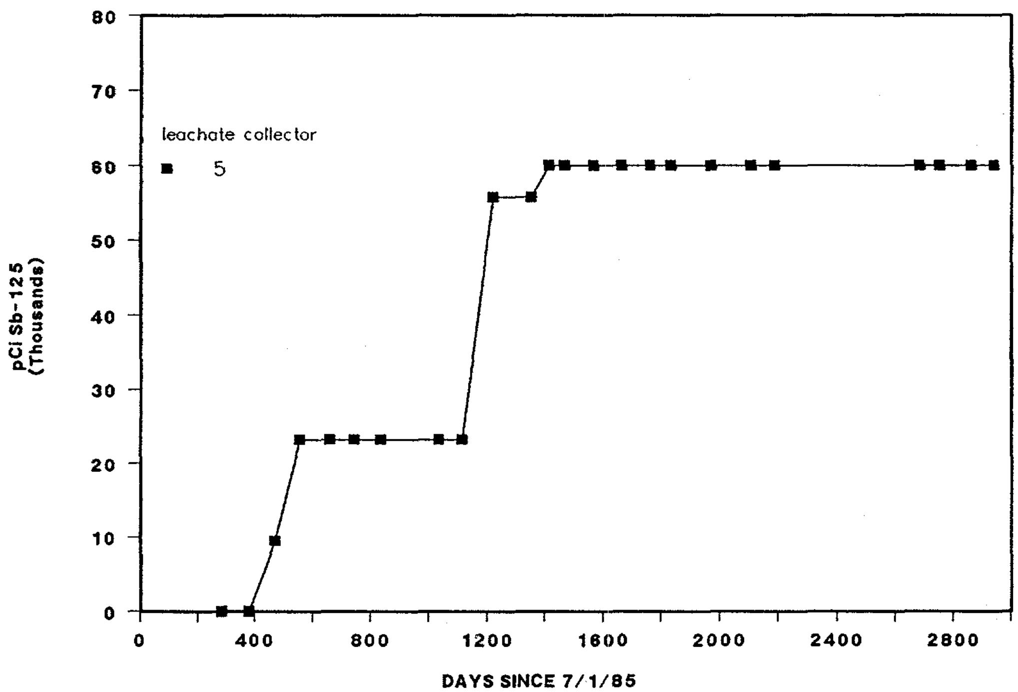


Figure 21. ORNL cumulative Sb-125 collected in lysimeter leachate collectors.

quantity of Sr-90 released in the leachate water in all lysimeters at both sites this period (Tables 15 and 16). For ORNL lysimeters 1, 2, and 4, the percent of total inventory of the nuclide released in leachate water was comparable to or greater than that in the cups. These data follow a trend seen over the past 30 months and make it appear that a pulse of Sr-90 could be moving through the soil columns of the ORNL lysimeters. For the control lysimeters at both sites, there was substantially more Sr-90 in the leachate than in cups 3 (two orders of magnitude for ANL-E and ORNL).

The percent of total Sr-90 being measured in the leachate water and cups 3 continues to be somewhat inconsistent between the two sites (Table 17). Perhaps this represents a difference in how the environment at the two sites affects the movement of Sr-90 being released from the waste forms. This difference is also seen when the percent of total Sr-90 found in the leachate water from the two control lysimeters is examined. The percent passing through the ORNL control was 6.3 times that of ANL-E (Table 17).

Gamma-producing nuclides continue to occur with regularity at both sites. ANL 2-3, below a cement waste form containing large amounts of Cs-137, continues to receive significant quantities of Cs-137 (Table 15; Figure 17). Since Cs-137 began appearing in ANL 5-3, the quantity of this nuclide has dramatically increased in each of the sampling periods with significant increases (45% in the eighth) during the last 4 years (Figure 17). However, no cesium was recovered from the water of this cup during the last sampling. Leachate water from ANL-5 has received sporadic releases this year. There continues to be no sustained occurrence of Cs-137 in any ANL-E leachate water.

Measurable amounts of Cs-137 began to occur in ORNL 5-3 during the May 1988 sample (Figure 18) and have continued in subsequent samplings for a total of 3,994 pCi (100% increase

Table 17. Percent of total Sr-90 and Cs-137 inventory per lysimeter extracted from moisture cups and leachate water through July 1993.

			Total inventory[15,16,17] per lysimeter (pCi)[a]		Percent total inventory Sr-90		Percent total inventory Cs-137	
Lysimeter number	Solidification agent	Liner number	Sr-90	Cs-137	Moisture cups	Leachate water	Moisture cups	Leachate water
ANL-1	Cement	PF-7	18.5E+9	3.1E+11	7.9E-5	3.7E-5	—[b]	—
ANL-2	Cement	PF-24	3.3E+9	14.3E+11	2.4E-4	6.2E-5	9.0E-7	—
ANL-3	VES	PF-7	27.4E+9	4.6E+11	5.2E-3	6.9E-4	—	—
ANL-4	VES	PF-24	4.5E+9	19.3E+11	6.6E-4	1.1E-4	—	—
ANL-5	Cement	PF-7	18.5E+9	3.1E+11	1.4E-4	1.3E-2	2.2E-5	—
ORNL-1	Cement	PF-7	18.5E+9	3.1E+11	7.2E-4	3.1E-3	—	2.0E-6
ORNL-2	Cement	PF-24	3.3E+9	14.3E+11	5.3E-4	9.0E-4	—	1.0E-7
ORNL-3	VES	PF-7	27.4E+9	4.6E+11	6.5E-4	8.7E-5	—	1.5E–6
ORNL-4	VES	PF-24	4.5E+9	19.3E+11	9.8E-5	4.7E-4	—	3.0E-7
ORNL-5	Cement	PF-24	3.3E+9	14.3E+11	5.6E-4	8.2E-2	2.0E-7	1.1E-4

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

b. Percent release is essentially equal to zero.

in the last year). Detectable amounts of Cs-137 have been consistently found in leachate water from ORNL-5 and sporadically in the other ORNL waters, though none have been found during the past 3 years (Figure 19 and Table 16). Breakthrough of Cs-137 into the ORNL-5 leachate collector occurred in November 1988, some 7 months after its occurrence in moisture cup ORNL 5-3 (Figures 18 and 19). Thus far, a total of 292,324 pCi has passed through to that collector.

For 4 years in a row, Sb-125 has not been found in ORNL-5 leachate water. Also, this is the fifth year of its absence in ORNL cup 5-3.

By using a matrix (as in Table 17), several comparisons can be made based on the intra- and intersite data. Overall, of the nuclides contained in the waste forms (see Table 17), a greater recovery of Sr-90 has occurred in terms of quantity and percent of inventory than of other nuclides. Next is Cs-137, followed by Sb-125 and Co-60 (not listed in Table 7). Compared to Sr-90, the recovery of Cs-137 appears insignificant. There have been significant occurrences of Cs-137 in cups 3 of the ORNL soil lysimeters during past years, and there was evidence of its reoccurrence in ORNL 1-3 (Table 16). On the other hand, this nuclide has been consistently occurring in ORNL 5-3 (Figure 18) and in the leachate collector of the ORNL-5 lysimeter (Figure 19). Cesium-137 has also occurred in the moisture cups of ANL-E lysimeters 2 and 5 but not in the leachate water. More Cs-137 has passed through the ORNL lysimeters than those at ANL-E.

At ANL-E, a comparison of Sr-90 occurrence in cups 3 and the leachate collectors (Table 17) contrasts the difference between movement of the nuclide away from the waste form into the bulk water solution versus its transport with the water through the soil column. This behavior might be influenced by the amount of water passing through the ANL-E lysimeters (Figure 11). However, a lack in uniformity is also seen with the ORNL data (Table 17), and these lysimeters have larger quantities of water (up to five times as much), with more uniform unit-to-unit movement (Figure 12).

As seen from Tables 2, 5, and 17, the lysimeters at both sites have been loaded with waste forms based on solidification agent and total nuclide content. Numbers 1, 2, and 5 were solidified with cement; numbers 3 and 4 with VES. ANL-1, -3, and -5, and ORNL-1 and -3 contain 5% of activity as Sr-90; the others contain 1% of activity as Sr-90 (Reference 15). This provides a total of five matched sets for the sites (ANL-1 and -2, ANL-3 and -4, ORNL-1 and -2, ORNL-3 and -4, and ANL-5 and ORNL-5). It could be assumed that nuclide leaching from these waste forms would be proportional to content (i.e., those with the higher loading would have proportionally larger Sr-90 releases, but the total percent of release should be close to the same). The first part of this assumption appears to be correct in the case of Sr-90 movement into cups 3 for both sites when compared to other cups at that site (Table 17). Figures 13 and 14 show that cumulative total quantities of Sr-90 in water retrieved from cups 3 are higher from the lysimeters with the higher loaded waste forms (range of 34 to 4,637% more) (Figures 13 and 14). The same was also true for the four soil lysimeters when the quantity of Sr-90 in leachate water is compared (13 to 3,825%). So it appears that there is a general trend for more Sr-90 to be removed from the higher loaded waste forms with a subsequent movement through the soil column. The assumption of a uniform percent release of Sr-90 from the waste forms, however, is not supported by the data (Table 17). For the moisture cup soil water collection, only three of the five sets have a higher total percent released to the cup water from those lysimeters containing the higher loaded waste forms (35 to 678%), and only two of the five have the higher Sr-90 released to the leachate water (243 and 546%).

A greater percentage of Sr-90 continues to be found in ANL 3-3 and ANL 4-3 (which both contain VES waste forms) than in the other ANL-E cups 3 (Table 17). As has been noted, the length of the soil column appears to moderate the quantity of the nuclide that travels from the waste form to the leachate collector. The leachate collectors in those same ANL-3 and -4 lysimeters also receive a higher percentage of Sr-90 than the other ANL-E soil lysimeter collectors, but a significant amount less than the cups 3 (754 and 600%). The percent of available nuclide that continues to move into the leachate of ANL-5 is much greater than that of the other ANL-E lysimeters (1,716 to 33,819%), thus providing further evidence of the moderating effect of soil.

Greater quantities of Sr-90 are moving through the ORNL lysimeters in comparison to the ANL-E lysimeters. Once again, there appears to be no correlation between the type of waste form and the amount of nuclide recovered in the leachate collector. About 0.082% of the Sr-90 contained in ORNL-5 has now been recovered in leachate from that lysimeter. The percent of available Sr-90 that has moved into the ORNL-5 leachate collector remains significantly higher than the other ORNL collectors (2,559 to 94,510%).

Recovery of Sr-90 in the ORNL cups is comparable for those lysimeters containing the cement waste forms and one of the two containing VES waste forms. These data together with those from ANL-E continue to indicate that cement and VES have comparable releases.

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

Upward Migration of Radionuclides

During previous samplings, the presence of both Cs-137 and Sr-90 were discovered at the surface of lysimeter ORNL-5, which is the sand-filled control. Radionuclide activity was first detected during a routine gamma survey of the lysimeter's surface in 1991. At that time, more activity was found near the center than at the edges. Core samples were obtained from the center of the lysimeter at depths from 0 to 2.5 cm and from 2.5 to 5 cm for analysis of cesium and Sr-90. Analysis detected 1,760 pCi Cs-137, 10 pCi Cs-134, and 0.5 pCi Sr-90 per gram of sand in the 0 to 2.5-cm core, and 306 pCi Cs-137, 3 pCi Cs-134, and 0.1 pCi Sr-90 in the 2.5 to 5-cm core material. These data showed that more nuclides were at the surface, suggesting some type of an active deposition mechanism. There remained a question, however, concerning the source of the nuclides. In August of 1992, samples were again taken from the lysimeter and analyzed for Cs-137 and Cs-134. The results were similar to the previous sampling, with 1,533 pCi Cs-137 and 6 pCi Cs-134 being found per gram in the surface, and 574 pCi Cs-137 and 2.4 pCi Cs-134 per gram in the 2.5 to 5.0-cm sample. A comparison was made between the ratio of Cs-137 and Cs-134 in the surface material and the ratio in the buried waste form. The ratio of the two types of cesium at the surface was 264, and the ratio at 5 cm was 242. Within the analysis uncertainty, the similarity of the two ratios suggests that the source of the nuclides was the same. To determine if the waste form was the source of the nuclides, the present ratio of these nuclides in the waste was calculated by using the standard radioactive decay equation. Based on waste history, the calculated cesium ratio in the waste form was 252. The ratio of cesium in the waste form (which would change only due to time or if there were an alternate source of cesium) is for all practical purposes the same as that of the cesium detected on the surface material. Therefore, it was concluded that the surface contamination of cesium came from the waste form. Measurement of Cs-137 in cup 5, the upper cup (Figure 7), shows a presence at that location in June (not shown in Table 16).

If the cesium at the surface migrated from the waste form, and it appears that it did, then it is important to find out how this nuclide migrated more than 1 m upward. Cesium tends to be sorbed much like potassium to clays or other sorptive material. Therefore, it would be expected that both the free unassociated cesium ions and the particles to which they could sorb would be washed downward away from the waste form during periods of water infiltration. Data on the occurrence of cesium in the leachate from lysimeter ORNL-5 seems to confirm that assumption (Table 16; Figure 19). However, since the fill material in the lysimeter is a fine-to-medium-grained silica sand with a very low cation-exchange capacity, a case can be made for cesium migrating as a solute in the pore water, which could move upward due to a wicking effect caused by evaporation. It is not likely that extensive evaporation is a regular occurrence, since the quantity of water moving through this lysimeter accounts for ~100% of the amount of precipitation that falls on the lysimeter surface. However, ORNL has experienced extended periods (three or more weeks) of hot weather with no rainfall during the summer months. Evaporation from the surface, enhanced by increased temperature, could result in an upward flux of water. Of course, any solute carried by this water would be left behind as a residue on the surface. The presence of wind-accumulated clays and organic matter on the sand surface could then fix the cesium and prevent its reentry. Planning to determine the mechanism of this unexpected cesium movement is underway. A sand core will be extracted and examined in FY-94.

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. Table 18 is a comparison of the cumulative

Table 18. Cumulative fractional releases from lysimeter field testing compared to those from bench leach testing (8,16).

				Cumulative fractional release			
						Leachate collectors	
Test type	Prefilter number	Solidification agent	Radio-nuclide	Demineralized water	Seawater	Soil	Sand
Bench,[a] INEL	7	Cement	Sr-90	7.8E-2	—	—	—
Bench,[a] INEL	7	VES	Sr-90	4.5E-2	—	—	—
Bench,[a] INEL	7	Cement	Cs-137	9.4E-2	—	—	—
Bench,[a] INEL	7	VES	Cs-137	4.6E-2	—	—	—
Bench, INEL	7	Cement	Cs-137	4.8E-2	9.0E-2	—	—
Bench, INEL	24	Cement	Cs-137	2.3E-2	2.6E-2	—	—
Bench, INEL	7	VES	Cs-137	2.1E-3	6.4E-2	—	—
Bench, INEL	24	VES	Cs-137	3.4E-4	1.3E-2	—	—
Field, ANL-E	7	Cement	Sr-90	—	—	3.7E-7	1.3E-4
Field, ANL-E	24	Cement	Sr-90	—	—	6.2E-7	—
Field, ANL-E	7	VES	Sr-90	—	—	6.9E-6	—
Field, ANL-E	24	VES	Sr-90	—	—	1.1E-6	—
Field, ORNL	7	Cement	Sr-90	—	—	3.1E-5	—
Field, ORNL	24	Cement	Sr-90	—	—	9.0E-6	8.2E-4
Field, ORNL	7	VES	Sr-90	—	—	8.7E-7	—
Field, ORNL	24	VES	Sr-90	—	—	4.7E-6	—
Field, ORNL	7	Cement	Cs-137	—	—	2.0E-8	—
Field, ORNL	24	VES	Cs-137	—	—	1.0E-9	1.1E-6
Field, ORNL	7	VES	Cs-137	—	—	1.5E-8	—
Field, ORNL	24	Cement	Cs-137	—	—	3.0E-9	—

a. Waste forms were irradiated before test.

fractional releases in to leachate collectors from field testing EPICOR-II waste forms in lysimeters to releases from bench-leach-testing similar waste forms in demineralized and seawaters as reported in References 8 and 17. Releases observed in the lysimeters are at least four orders of magnitude less for Sr-90 in soil and at least five orders of magnitude less for Cs-137 in soil. It is interesting to note that release of Sr-90 in the sand-filled lysimeter is only one or two orders of magnitude less than bench-test results with demineralized water. At the present rate of increase (Figures 15 and 16), these cumulative fractional releases will be of similar magnitude in a couple of years.

Use of Lysimeter Data for Performance Assessment and Source Term Calculations

It is becoming apparent, through operational experience and cumulative data provided by the NRC lysimeter array during the past 8 years, that lysimeters are a valuable source of data used in the development of site-specific performance assessments. The operational lysimeters are providing continuous data from the near-field (that area comprised of the waste form and surrounding soil). These data directly relate to waste form stability. Information that can be obtained from the data includes the mass balance of released constituents, 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 dispersivity 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 19. These parameters have been calculated using data collected during the first 48-month operation of the ANL-E and ORNL lysimeters (Table 20). The data could be used in such codes at PATHRAE,[21] PRESTO,[22] and others to predict the stability of waste forms for a 300-year period of time.

Table 19. 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 20. Performance assessment code parameters derived from the first 4 years of ANL-E and ORNL data.

Code parameters	ANL-E					ORNL				
	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+9	3.3E+9	27.4E+9	4.5E+9	18.2E+9	18.2E+9	3.3E+9	27.4E+9	4.5E+9	3.3E+9
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-6	49E-6	29E-6	1E-6	1,500E-6	140E-6	279E-6	60E-6	220E-6	2,160E-6
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

During this reporting period, the collected lysimeter data were used as inputs for the computer code MIXBATH.[25] Use of this model is intended to predict the release of nuclides from a waste form in a failed container surrounded by a porous medium containing a solute. The solute is treated as a well-stirred fluid (i.e., a mixing bath), and solute concentration is calculated using a mass balance that depends on the solute flow rate, the amount of partitioning between the porous medium and solute, the size of the mixing bath, the radioactive decay rate, and the rate of nuclide release from the waste form. Modeling of the waste form is accomplished using a one-dimensional finite difference model. MIXBATH has the capability to simultaneously consider three waste form release mechanisms: diffusion, dissolution, and surface rinse limited by partitioning.

Releases of Cs-137 and Sr-90 from the waste forms were modeled. The most appropriate release process was considered to be diffusion from a cylinder (the shape of the waste forms). The waste form diffusion coefficients for Cs-137 were available from data in Reference 17 while those for Sr-90 were obtained based on measurements of similar waste forms of equal size.[26] Calculations for the mass balance of the solute concentration required a Darcy velocity (volumetric flow rate per area), which could not be calculated from the available data. These data were estimated from lysimeter leachate collector analytical data.[11] Soil/water distribution coefficients were estimated from previous published work.[27] Tables 21, 22, and 23 list the values used for the most important parameters. These include the soil/water partition coefficients (K_d) and decay constants, the diffusion coefficients (D) for each waste form and isotope, and the Darcy velocities of the soils. The K_d values used were assumed to fall between the upper and lower boundaries for the model parameters in soils (Table 21). With the Unimin sand, the best curve fit was obtained using an assumed $K_d = 0$. It should be noted that the VES waste form diffusion coefficient for Sr-90 listed in Table 22 is approximately six orders of magnitude larger than that for Cs-137. The cause for this discrepancy is the use of a literature value for the Sr-90 and a bench-leach-test value for Cs-137. This highlights the necessity of using waste-form-specific parametric values.

Results of this preliminary lysimeter performance assessment modeling produced data for which the parametric information available was broad enough for accurate predictions. Of course, there were also data in which predicted and measured values were in poor agreement. Such differences appeared to be the result of a lack of waste-form-specific diffusion coefficient data, together with the low cumulative concentration of nuclides in some of the lysimeter leachate waters. Figure 22 shows plots of predicted and measured Sr-90 cumulative activity versus time for ORNL 5. Two predictions are shown using diffusion coefficients of 4E-10 cm^2/s and 5E-11 cm^2/s. With the latter, the MIXBATH prediction and measured values agree within one order of magnitude. Releases of this magnitude appear to be consistent with those measured during other work using these waste forms.[16] Use of the diffusion coefficient of 4E-10 cm^2/s gave results that were five orders of magnitude greater than actual values. These data indicate that the determination and use of waste-form-specific diffusion coefficients for model input is important.

The results as shown in Figure 22 indicate that there was insufficient cumulative radionuclide activity as of this reporting period for code validation. However, there were sufficient data to show similarities between the predicted and measured curves. These plots appear to be typical of the predictions made about Sr-90 release from both cement and VES waste forms. Strontium-90 diffusion coefficients used for prediction are probably much greater than actual values. Data from the lysimeter project have indicated that for VES, Cs-137 and Sr-90 diffusion coefficients are probably of the same order of magnitude.

Data from a comparison of cumulative Cs-137 activity from ORNL-3 appears to give a reasonable prediction (Figure 23). This demonstrates how the measured value of the diffusion coefficient and close approximations of the partition coefficient (Table 21) can significantly increase the accuracy of the prediction. From these data

Table 21. Partition coefficients (cm^3/g) of three soils used in lysimeters.

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

a. The value assumed for essentially inert material.

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

Table 22. Diffusion coefficients of waste forms and radionuclides used in lysimeters (cm^2/s).

Waste form	Radionuclide: Cs-137[a]	Radionuclide: Sr-90[b]
Vinyl ester-styrene	3.30E-14	1.35E-8
portland type I-II cement	5E-11	4E-10

a. See Reference 17.

b. See Reference 26.

Table 23. Darcy velocities of soils used in lysimeters.

Lysimeter number	Darcy velocity (cm/s) ANL-E	Darcy velocity (cm/s) ORNL
1	9.42E-7	3.07E-6
2	1.10E-6	3.10E-6
3	1.65E-6	3.12E-6
4	1.34E-6	3.16E-6
5	2.59E-6	3.60E-6

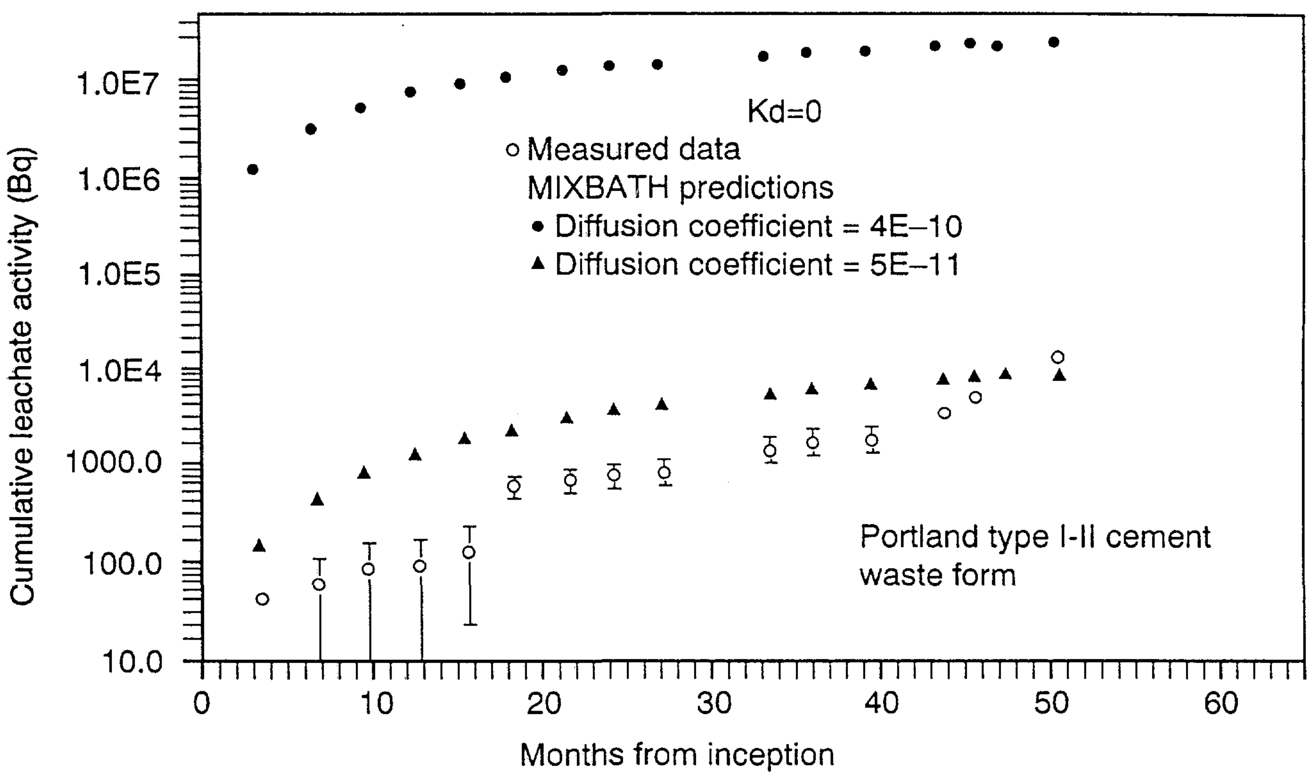


Figure 22. Comparison of Sr-90 cumulative activities for measured data from ORNL lysimeter 5 leachate collector MIXBATH predicted results.

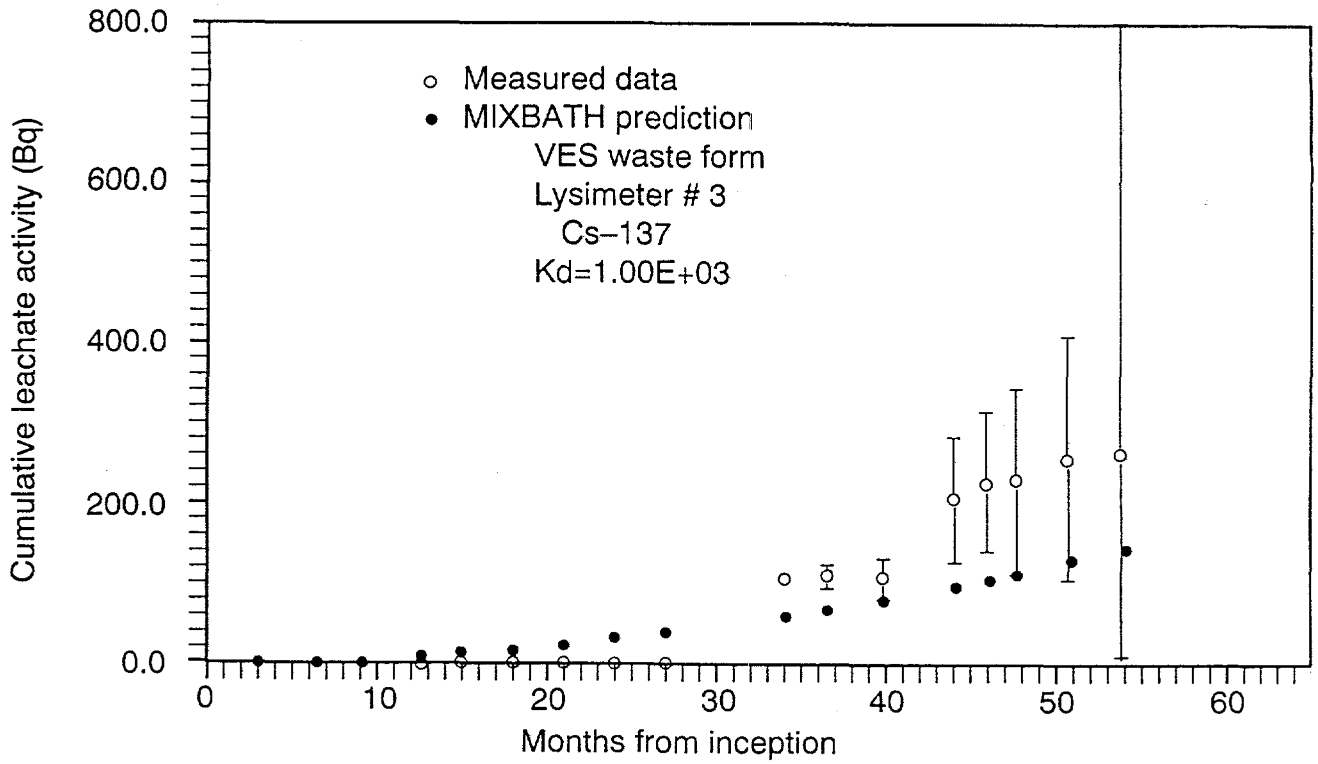


Figure 23. Comparison of Cs-137 cumulative activities for measured data at ORNL lysimeter 3 leachate collector MIXBATH prediction.

and those from Sr-90, it appears that MIXBATH performed adequately for the purposes of this preliminary performance assessment. It helped identify those areas in which additional data (diffusivity values, soil K_d values, and soil hydraulic properties) will be required in order to use the lysimeter data effectively in performance assessment modeling.

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

Source term code studies were performed using the data produced through FY-93 by the ANL-E and ORNL field experiments. A brief summary of the pertinent characteristics of the lysimeters is in order. At each site, four of the lysimeters are filled with soil while the fifth lysimeter (a control) is filled with Unimin silica oxide sand. At ORNL, the soil used is from the C horizon of a Fuquay sandy loam from the Savannah River Plant adjacent to the Barnwell facility in South Carolina. ANL-E lysimeters are filled with a local soil that represents a typical Midwestern type. It is a morley silt loam with the surface layer removed. Each lysimeter is filled with seven cylindrical waste forms measuring 4.8 cm in diameter and 7.5 cm in height. They are stacked one on top of the other in the lysimeters forming a height of 53.2 cm and a volume of 1 L. The waste forms were solidified in either vinyl ester-styrene or portland type I-II cement. The waste streams included two resin types. Type I was a mixture of synthetic organic ion-exchange resins (phenolic cation, strong acid cation, and strong base anion). Type II resin was a mixture of synthetic ion-exchange resins (strong acid cation and strong base anion resins) with inorganic zeolite. Each lysimeter is equipped with five moisture collecting cups and three soil moisture/temperature probes, which are located at various elevations in the lysimeter (Figure 5) along with a leachate container located at the bottom of the lysimeter (Reference 15). Below the fill material, a layer of filter fabric was placed between the soil or sand and the gravel bed. A gravel bed is located below the filter fabric. The height of the gravel bed was set to 10 cm in these modeling studies. The data used in this study were collected from moisture cup 3, located approximately 23 cm from the bottom of the waste forms, and from the lysimeter leachate collector, located 61 cm below the bottom of the waste forms. The radionuclides found to date in the leachate waters have been primarily Cs-137 and Sr-90.

The Disposal Unit Source Term (DUST) code was used to model the release of Cs-137 and Sr-90 from the lysimeter waste forms. DUST is a one-dimensional code that can model release by a finite difference method or by a mixing cell cascade approach, and has the ability to simultaneously model three different types of release mechanisms: diffusion, dissolution, and surface rinse. The mixing cell model is limited in that it does not take diffusional release into consideration. Therefore, for these simulations, the finite difference model was selected because it is more flexible and capable of handling a variety of different parameters. A further description of the models in the code is given in Reference 28.

Lysimeters 5 at ORNL and ANL-E were chosen for study of the release of Cs-137 and Sr-90 from portland type I-II cement because releases from other lysimeters were substantially lower and the data were not sufficient to model. At ANL-E, lysimeter 5 contained resin waste type I solidified in cement; at ORNL, lysimeter 5 contained resin waste type II, which was also solidified in portland type I-II cement (see Table 5). Diffusional release is believed to be the controlling mechanism for a cement-solidified waste. The waste form diffusion coefficients for portland type I-II cement were presented in Reference 17. Measured values were 9.6E-10 cm^2/s for Sr-90 and 5E-11 cm^2/s for Cs-137. The Darcy velocities ranged from 2.59E-6 cm/s at ANL-E to 3.6E-6 cm/s at ORNL (Reference 12). The soil bulk density values were 1.55 g/cm^3 at ANL-E and 1.60 g/cm^3 at ORNL (Reference 15). Moisture content values were calculated using the effective soil porosity and the fraction of saturation values found in Reference 8. In lysimeter 5 at both sites, the moisture content was calculated as 21%. The distribution coefficients have not been measured for Sr-90 or Cs-137; therefore, they were estimated by fitting the model predictions to the data. The cumulative leachate activity collected from the lysimeters over the first 7 years of the experiment, which was used to make comparisons to the DUST code predictions, represented 0.045% and 0.008% of the total inventory of Sr-90 in lysimeters 5 at ORNL and ANL-E, respectively. At ORNL, the collected amount represented less than 8.6E-5% of the Cs-137 inventory in lysimeter 5, while nothing has been collected in ANL-E lysimeter 5 (Table 24).

Concentrations and predicted releases were matched to moisture cup 3 and the lysimeter leachate collector. The concentrations and releases were taken at 23 and 51 cm below the waste forms. In this report, the cumulative leachate activity collected 51 cm beneath the waste form is used as the performance measure. Initial amounts of Cs-137 and Sr-90 varied at ORNL and ANL-E because the control lysimeters contained different resin types. In ORNL lysimeter 5, the type I waste form had a total initial inventory of 3.29E-3 Ci of Sr-90 and 1.432 Ci of Cs-137 (Reference 8). The type II waste form at ANL-E had a total initial inventory of 1.84E-2 Ci of Sr-90 (Table 17 and Reference 15). Cesium-137 was not modeled at ANL-E for lack of sufficient releases.

The cumulative activity collected from the lysimeters is less than 5E-2% in comparison to the total inventory for Sr-90 and less than 9E-5% for Cs-137 (Table 24). Therefore, either the waste form release rates are much lower than anticipated or transport processes are controlling release through the soil column. At that level, it is possible that random fluctuations (noise) are being seen, and release patterns may not develop for several more years.

Three parameters are known to strongly influence release through the soil column. They are distribution coefficient (K_d) and dispersivity, which together control transport from the waste form through the soil column, and waste form diffusion, which controls waste form release rates. Several cases were modeled where either K_d, dispersivity, or waste form diffusion coefficients were varied to best match the actual release data from the lysimeters.

An exponentially decaying waste form release rate of 1.75E-6 exp (-lt) Ci/yr was chosen, where (l) is the decay constant for Sr-90 and (t) is the time; also chosen were a dispersivity of 10.5 cm and K_d values of between 4.5 and 4.8 (Figure 24). In doing so, a very good fit to the data was obtained, although the parameters used are highly unlikely. The waste form is releasing approximately 0.01% of inventory per year, i.e., 0.07% over 7 years. The experimentally measured release from lysimeter 5 at ANL-E was 0.007%.

The domain of the model was extended to 52 cm below the waste form. This ensures that boundary conditions (BCs) will not significantly affect the predicted concentrations. Therefore, the results in Figures 25 and 26 are obtained using a bottom BC of zero dispersive flux. A concentration trace continued to be taken at the location of the filter fabric, which is 51 cm below the waste form.

As shown in Figure 25, the actual data for Sr-90 from ORNL lysimeter 5 for 8 years are compared with the DUST code predicted releases using zero dispersive flux BC, K_d = 24, and dispersivity = 8.5 cm. Also shown are predicted releases using zero concentration flux BC, K_d = 10, and dispersivity = 0.6 cm. The measured waste form diffusion coefficient of 9.6E-10 cm^2/s was used. The predicted releases of zero dispersive flux BC show a very good fit to the actual data after 3 years. The DUST curve that is generated with the zero dispersive flux BC is rising at a much more shallow slope than the zero concentration BC curve, indicating lower predicted releases over 20 years.

Figure 26 shows the actual data for Sr-90 at ANL-E lysimeter 5, which covers a period of 8 years. In addition, the DUST predictions of 20 years of cumulative leachate activity is plotted in two cases, using dispersive flux BCs. The measured waste form diffusion coefficient of 9.6E-10 cm^2/s was used. Case 1 has a dispersivity of 8.5 cm and a K_d of 24.5. Case 2 has a dispersivity of 0.6 cm and a K_d of 10. Case 2 releases less activity over 8 years than Case 1; however, at 20 years, the amount of activity released by case 2 is an order of magnitude higher than the amount in case 1. Over 20 years, case 2 will have released 33% of the total Sr-90 inventory, whereas case 1 will have released 3.3% of the total Sr-90 inventory. Case 1, also, is a better fit to the actual data at 8 years, indicating a predicted higher dispersivity and K_d than previously thought.

Table 24. Total and collected Ci amounts of Sr-90 and Cs-137 in lysimeter 5 through July 1992.

	Total amount (Ci)	Amount collected (Ci)	Percent collected
ORNL Cs-137	1.432	0.23E-6	8.6E-5
ORNL Sr-90	3.39E-3	1.6E-6	4.5E-2
ANL-E Sr-90	1.84E-2	1.4E-6	7.6E-3

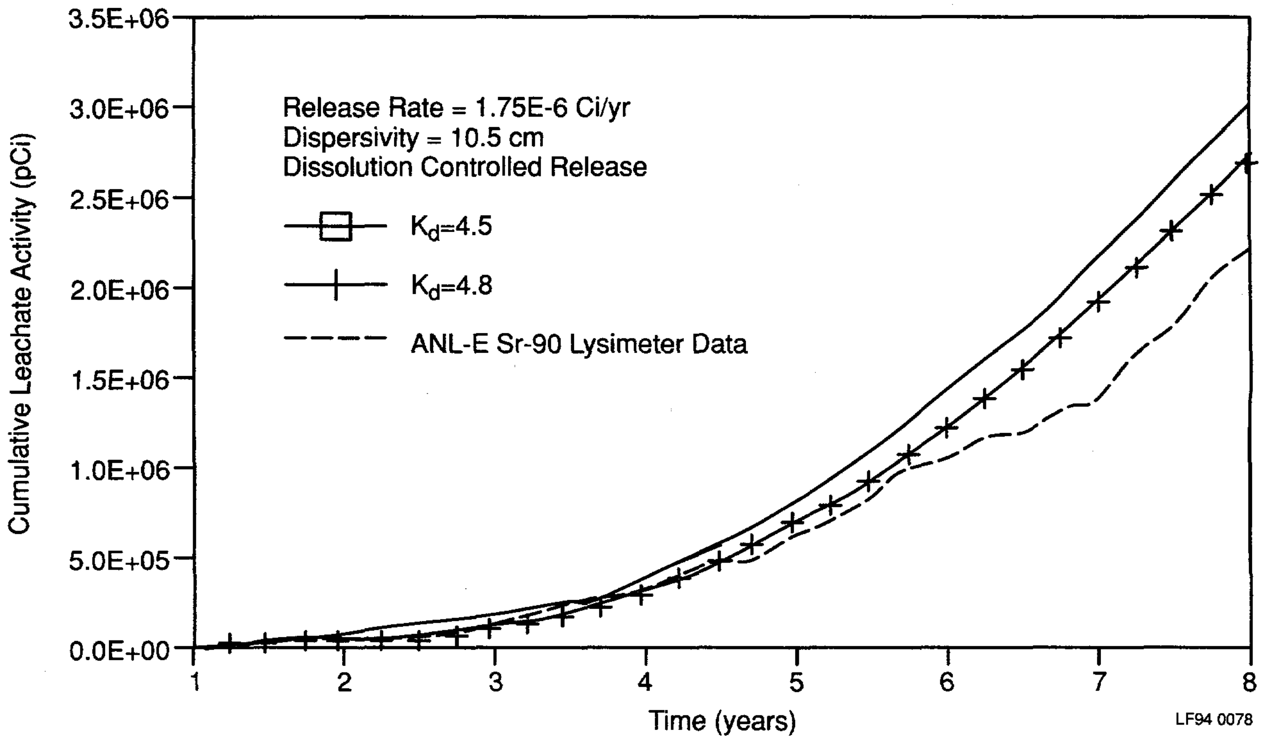


Figure 24. Data for Sr-90 at ANL-E lysimeter 5, compared with the effects of K_d values on predicted releases with an exponentially decaying waste form release rate.

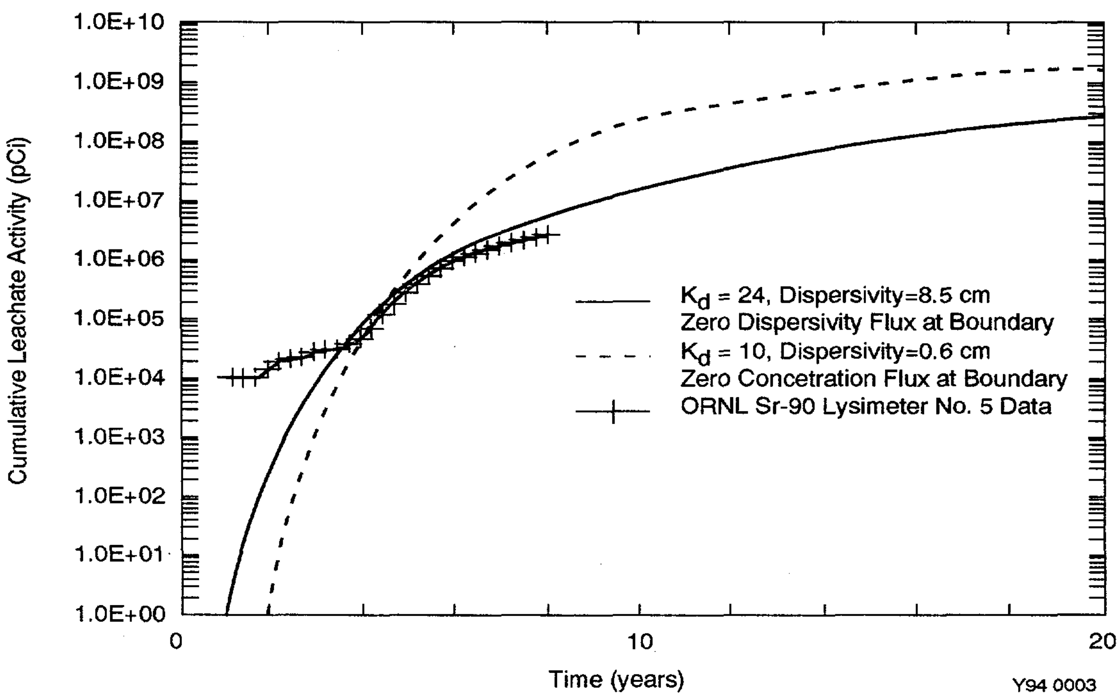


Figure 25. Data for Sr-90 at ORNL lysimeter 5, compared with two sets of estimated K_d and dispersivity values.

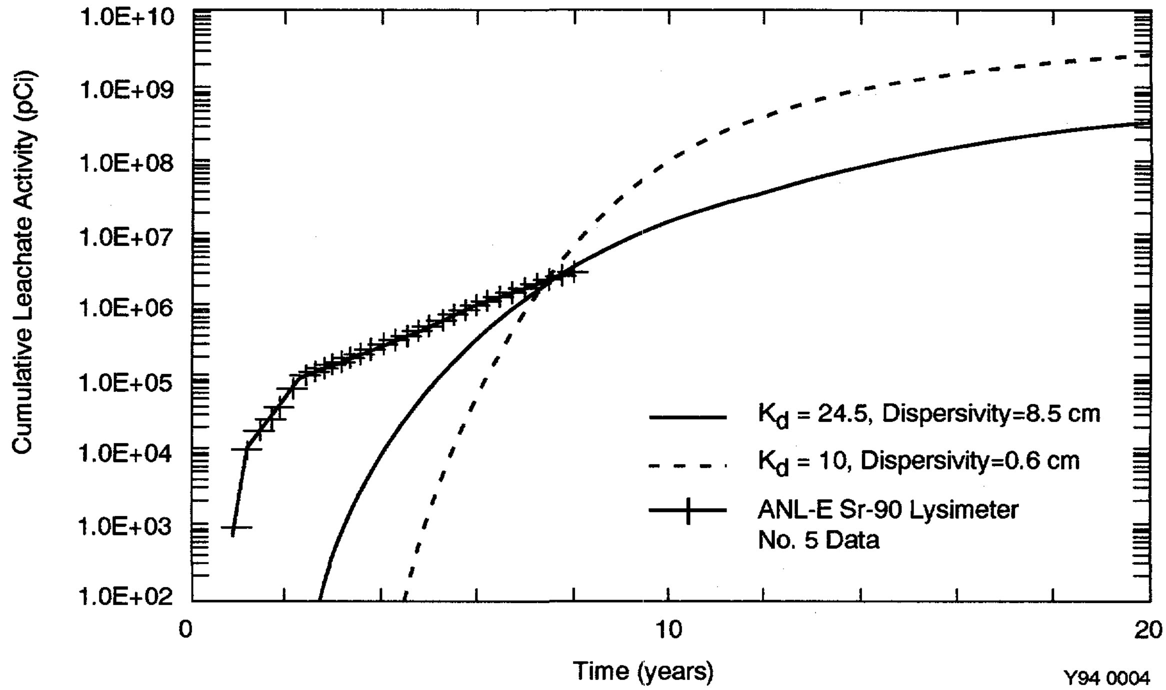


Figure 26. Eight years of data for Sr-90 at ANL-E lysimeter 5, compared with two sets of estimated K_d and dispersivity values for 20 years.

Major Cation and Anion Analysis

A clear understanding of the factors that influence movement of radionuclides through the lysimeter soils is not available in the literature. A preliminary effort was initiated at ORNL in 1988 and at ANL-E in 1991 to analyze water samples obtained from the moisture cups 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. It could also indicate the presence of major ions, which could enhance radionuclide transport by either forming soluble complex formations with radionuclides [e.g., Sr-90 $(HCO_3)_2$—an electrically neutral dissolved species] or by causing movement as a result of competition with radionuclides for the limited number of soil exchange sites (e.g., K^+ versus Cs^+). These data, together with a future analysis of the mineralogical composition of the lysimeter soil, could be used to develop equilibrium geochemical modeling, which could in turn be used to calculate the concentration of various radionuclide complexes in the soil solution.

A portion of the water obtained during the summer sampling periods in 1991, 1992, and 1993 was analyzed for the major ionic species listed in Table 25. The justification for the choice of ions is also provided in the table. At ANL-E, cups 1, 3, and 5 were sampled on lysimeters 1, 3, 4, and 5; and cups 2, 3, and 4 on lysimeter 2. Cups 1, 3, and 5 water samples were sampled in 1993 at ORNL. Data from precipitation samples at ORNL in 1989 and ANL-E in 1991 showed that ionic concentrations in the soil water were not introduced by the precipitation (References 9 and 12). It appears that the waste forms could be an influencing factor either as the source of ions or possibly by causing replacement of ions from the surrounding soil (Tables F-1 through F-6 of

Table 25. Ionic species analyzed from lysimeter moisture cup water samples.

Ionic species	Justification
Na^{+}	Indicator of weathering reactions if Na-feldspars are present.
Mg^{+2}	Forms complexes with bicarbonate and carbonate.
Ca^{+2}	In the absence of calcium minerals, this may be an indicator of cement breakdown. Forms complexes with bicarbonate and carbonate. An indicator of Sr behavior.
K^{+}	Indicator of weathering reactions if K-feldspars or illite are present. Competes with Cs for exchange sites.
H_4SiO_4	Indicator of weathering reactions. Concentrations of dissolved silica above saturation with quartz may indicate weathering of the zeolite.
Alkalinity	Bicarbonate and carbonate form complexes with Ca, Mg, and Sr. Typically the major anion in soil solutions.
SO_4^{-2}	Second most abundant anion in soil waters. Forms complexes with most cations.
PO_4^{-3}	Complex forming anion. Sorbs on iron oxide surfaces. Indicator of Sb behavior.
NO_3^{-}	Needed for charge balance calculation.
Cl^{-}	Needed for charge balance calculation.

Appendix F and Figures 27, 28, 29, and 30). It appears that the cement and VES waste forms performed similarly at both sites. With a few exceptions, the ORNL soil lysimeter cation and anion data (Tables F-4 through F-6 and Figures 29 and 30) closely resemble each other over the reporting period and actually showed little of the cup-to-cup variability found in 1988. ANL-E 1993 data are similar, in most cases, to ORNL 1993 data when compared in Figures 27, 28, 29, and 30. While these early data are interesting, no correlation can be made with nuclide movement as yet.

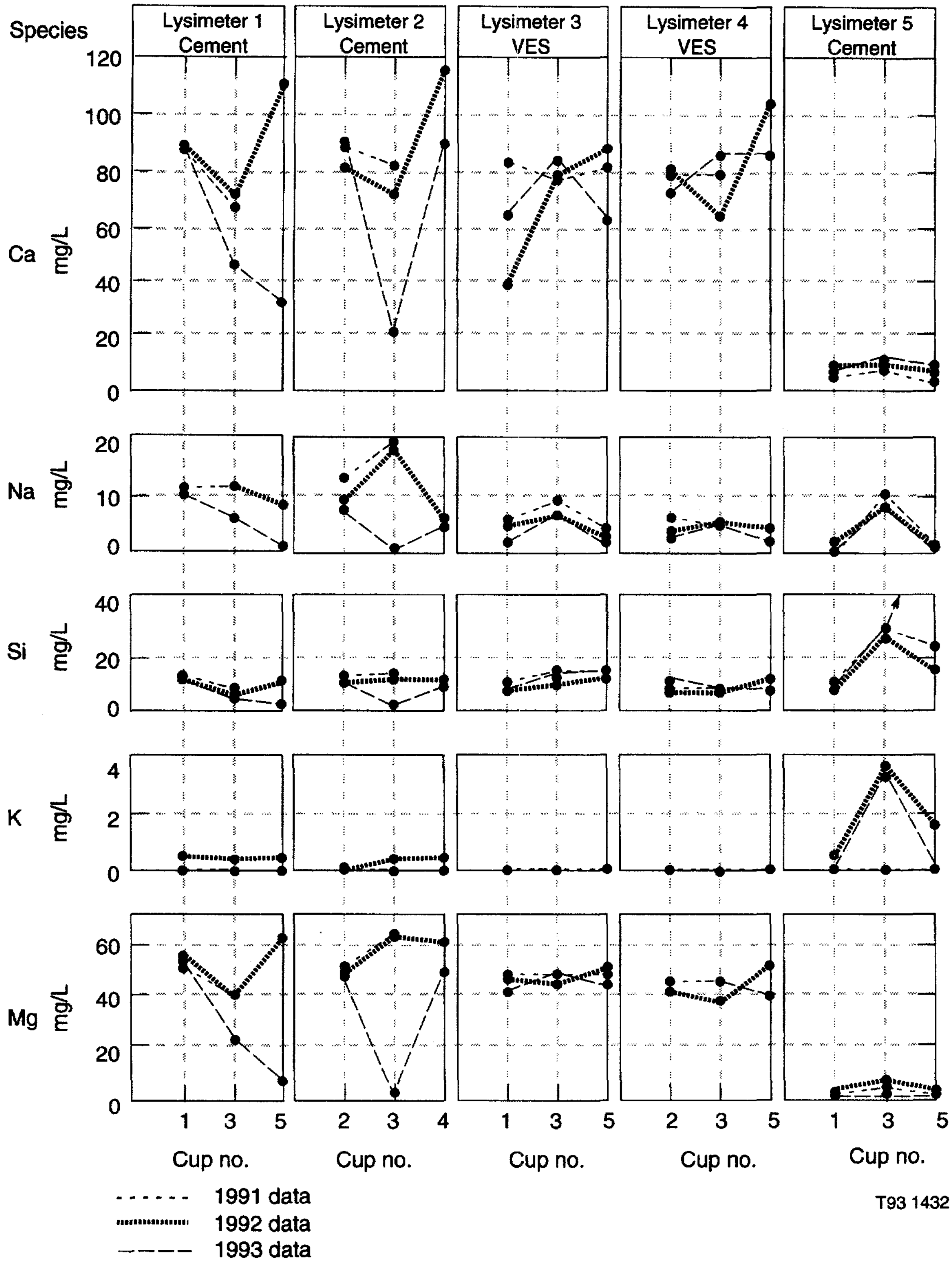


Figure 27. Results of chemical speciation at ANL-E cations.

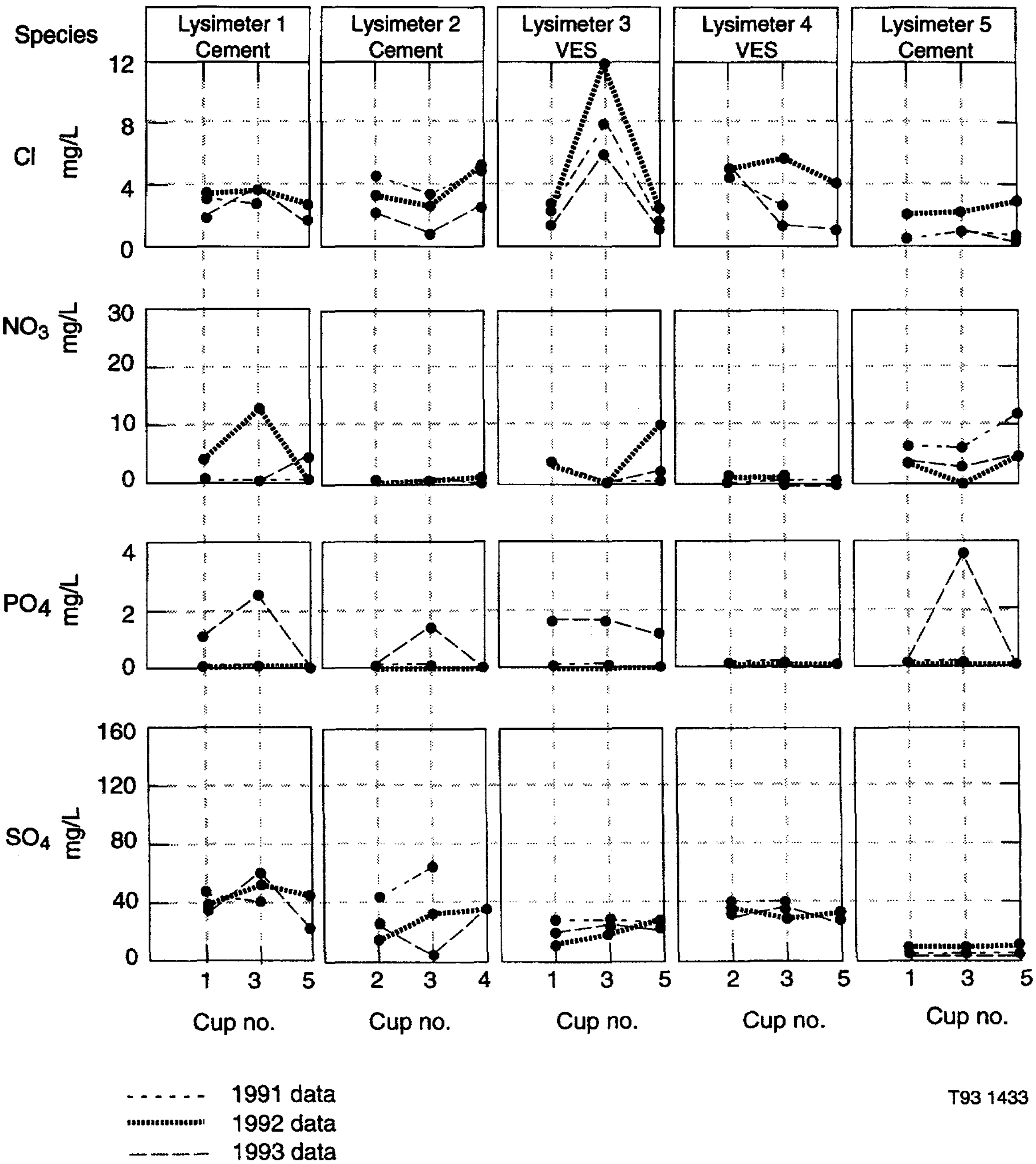


Figure 28. Results of chemical speciation at ANL-E anions.

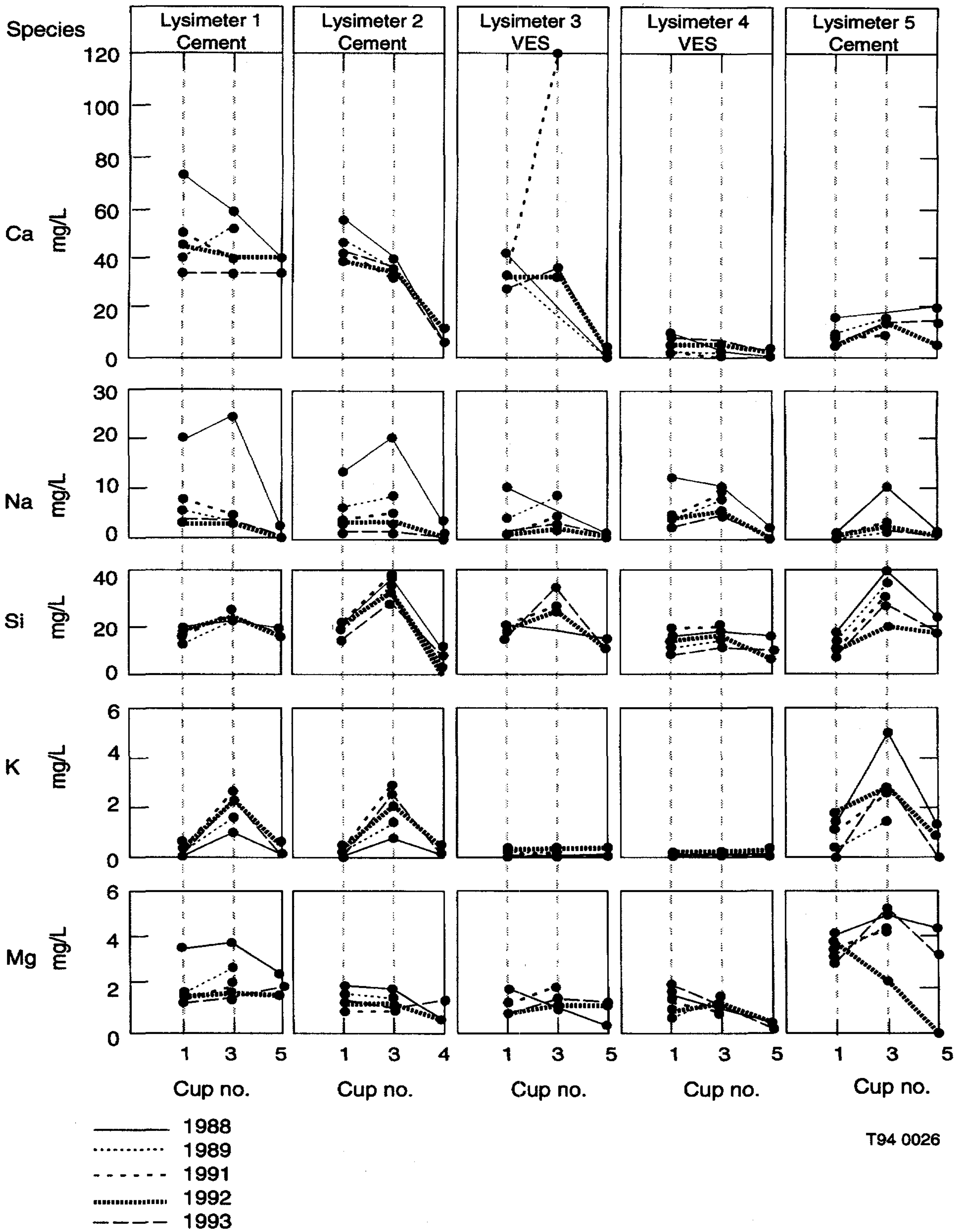


Figure 29. Results of chemical speciation at ORNL cations.

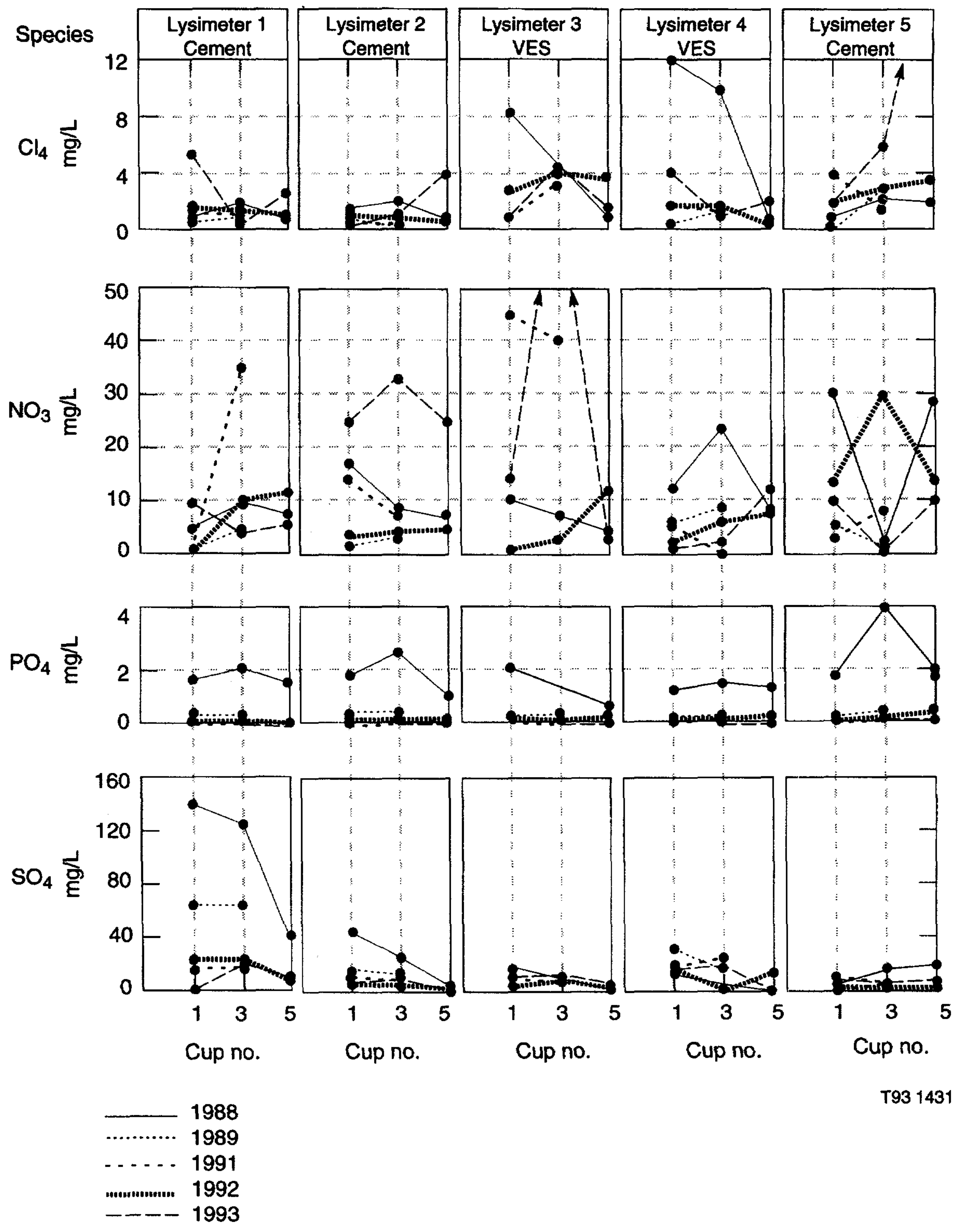


Figure 30. Results of chemical speciation at ORNL anions.

CONCLUSIONS

The lysimeter experiment during the 8 years of operation has been successful. Analyses of data collected during the past 96 months continue to show a pattern in nuclide availability and movement such that the cumulative results are beginning to provide an insight on waste form performance.

There continues to be a greater recovery of Sr-90 in terms of quantity and percent of inventory than other nuclides. Next in abundance is Cs-137, followed by Sb-125 (this nuclide has not been detected for the past 48 months) and Co-60. Compared to Sr-90, the occurrence of Cs-137 appears insignificant.

On a cumulative basis, a larger amount of Sr-90 is being removed in leachate water from the ORNL soil lysimeters. This is thought to be a result of the difference in soils as well as in environmental conditions between the two sites. During the past 72 months, Sr-90 continues to be found in equal concentrations in leachate water from the sand-filled control lysimeters at both sites, with a slightly more rapid accumulation at ORNL, which now has had six-and-one-half times more of the available source of Sr-90 released than the control lysimeter at ANL-E. Such data continue to reinforce the assumption that the limiting step in receiving Sr-90 in leachate water is not release of the nuclide from the waste forms (since Sr-90 is found in larger quantities in leachate water at ORNL rather than in cups), but rather, the movement is limited by environmental characteristics (including soil and quantity of soil water). This conclusion is supported by data from lysimeter work at Savannah River Laboratory (SRL) and Pacific Northwest Laboratory (PNL).[23,24] SRL has found that Sr-90 will move from buried waste forms, migrate through the soil column, and appear in collected leachate water.[23] It is not surprising, then, that Sr-90 moves through soil in the ORNL lysimeters, since that soil originated at SRL.[14] On the other hand, lysimeter work with waste forms at PNL has shown that Sr-90 does not move in those soils.[24]

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

In the past, Cs-137 has been found with consistency in leachate water from the sand-filled lysimeters only at ORNL. In 1992 and 1993, Cs-137 was found in leachate water from the sand-filled control lysimeters at both sites. It is also interesting to note that cesium was found to have migrated from the waste form to the surface sand of the ORNL control lysimeter.

As a conclusion, data from the two sites have not yet demonstrated which type of solidification product is preferable for nuclide retention. It appears at this time that releases of Sr-90 and Cs-137 from cement and VES are comparable but dependent on environmental influences. These data still differ from those obtained at SRL. Those data show that cement minimizes the release of Sr-90.[23] This interesting difference should be studied further. Both data reported herein and data reported by SRL and PNL agree that Cs-137 is more readily released from cement than from VES.

On two occasions, lysimeter data have been reviewed to determine the possibility of using these data to initiate limited performance assessment modeling. The results from a preliminary evaluation using the computer code MIXBATH that was carried out in FY-91 indicated that in lysimeters with experimentally determined diffusion coefficients, where there were high enough leachate concentrations of nuclides for comparison between predicted and experimental results, a computer code could be tested. In 1992 and again in 1993, refinements made it possible to model some of the lysimeter Sr-90 release data using the DUST computer code. Once again, as has been the case of others using these data, it was strongly recommended that the lysimeter experiments be continued. Rapidly increasing radionuclide release showed that data from future years could be used to obtain a reliable, quantitative understanding of nuclide movement through the use of numerical codes.

The numerical studies have been hampered by the lack of soil data. It is important to know the site-specific soil distribution coefficient (K_d) and dispersivity values to better predict the release characteristics in the lysimeters.

Boundary conditions have little effect on predicted cumulative activity release; however, they play an important role in predicted concentrations. Concentration profiles are developing slowly, and further releases should, therefore, continue to be monitored. Further numerical studies are planned.

REFERENCES

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

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

3. Code of Federal Regulations, 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Wastes," Office of the Federal Register, December 1982.

4. U.S. Nuclear Regulatory Commission, "Technical Position on Waste Form," Low-Level Waste Management Branch, Washington, D.C., May 1983.

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

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

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

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

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

10. J. W. McConnell, Jr., R. D. Rogers, M. W. Findley, E. C. Davis, J. D. Jastrow, R. M. Neilson, L. D. Hilton, *Field Lysimeter Investigations: Low-Level Waste Data Base Development, Program Lysimeter Test Results for Fiscal Years 1986, 87, 88, and 89*, NUREG/CR-6256, INEL-95/0073, Vol. 1, Idaho National Engineering Laboratory, Lockheed Idaho Technologies Co., May 1995.

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

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

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

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

15. R. D. Rogers, J. W. McConnell, Jr., E. C. Davis, M. W. Findlay, *Field Testing of Waste Forms Containing EPICOR-II Ion Exchange Resins Using Lysimeters*, NUREG/CR-4498, EGG-2438, June 1986.

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

17. R. M. Nielson, Jr. and J. W. McConnell, Jr., *EPICOR-II Resin Waste Form Testing*, NUREG/CR-4637, EGG-2457, October 1986.

18. R. D. Rogers and J. W. McConnell, Jr., *Biodegradation Testing of TMI-2 EPICOR-II Waste Forms*, NUREG/CR-5137, EGG-2450, June 1988.

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

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

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

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

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

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

25. T. M. Sullivan and C. J. Suen, *Low-Level Waste Source Term Model Development and Testing*, NUREG/CR-5681, May 1991.

26. M. Fuhrmann, R. F. Pietrzak, J. Heiser, B. M. Franz, and P. Colombo, *Accelerated Leach Test Development Program*, BNL-52270, November 1990.

27. T. M. Sullivan and C. J. Suen, *Low-Level Waste Shallow Land Disposal Source Term Model: Data Input Guides*, NUREG/CR-5387, BNL-NUREG-52206, July 1989.

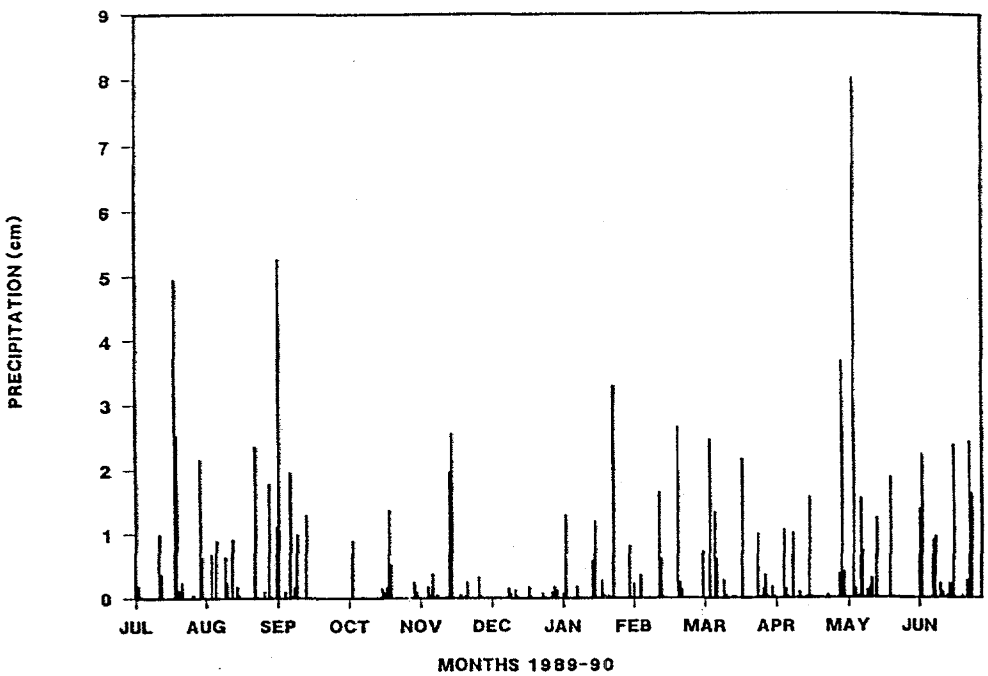
28. T. M. Sullivan, *Selection of Models to Calculate the LLW Source Term*, NUREG/CR-5773, BNL-NUREG-52295, Brookhaven National Laboratory, October 1991.

Appendix A

Weather Data

Appendix A
Weather Data

List of Figures



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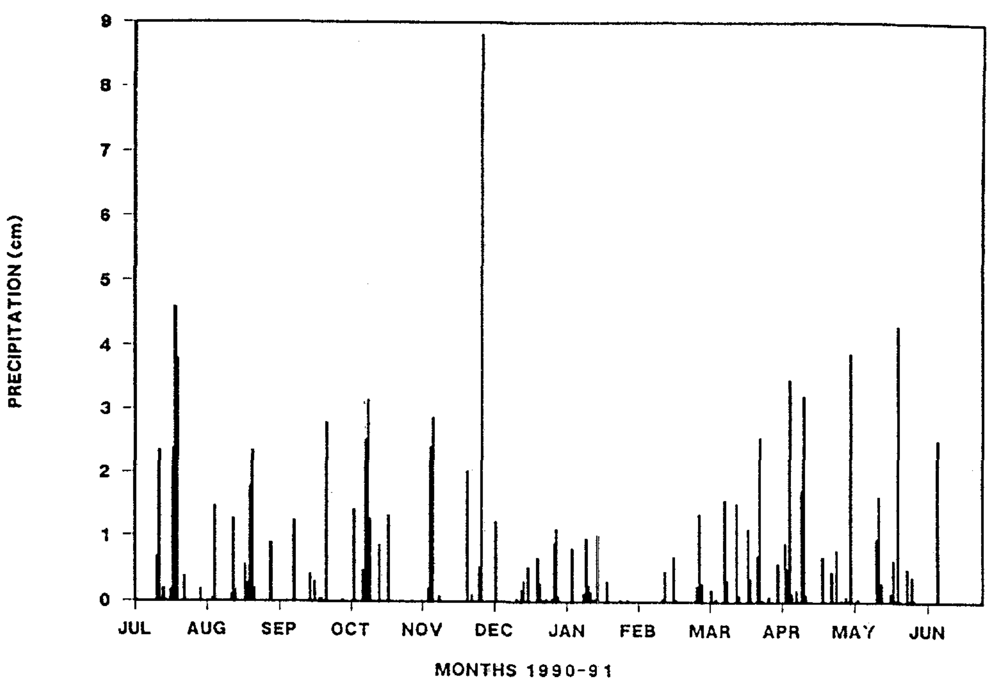

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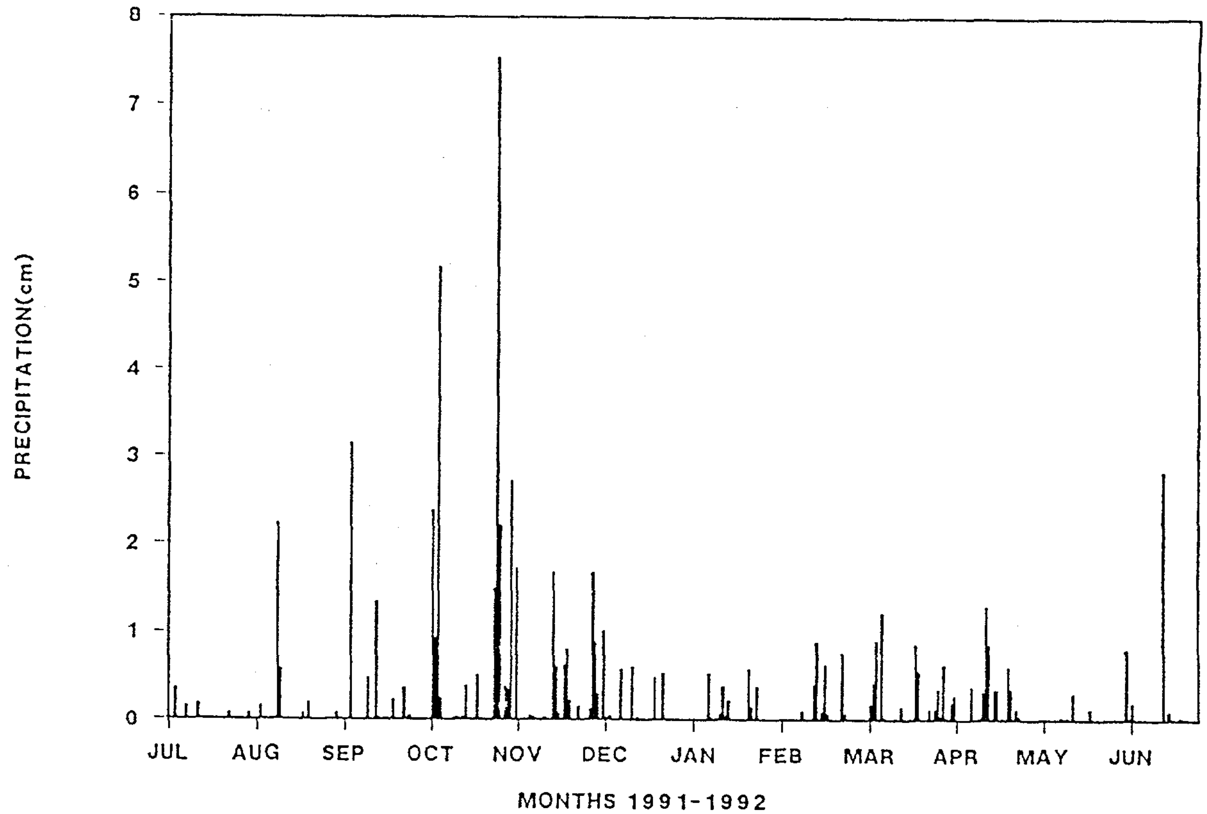

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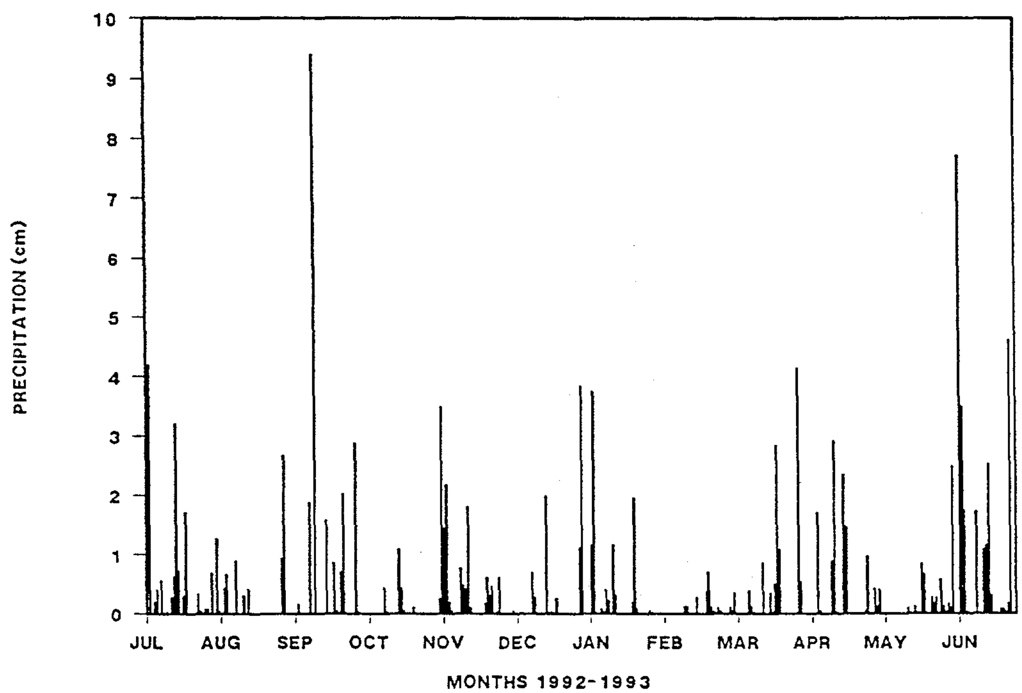

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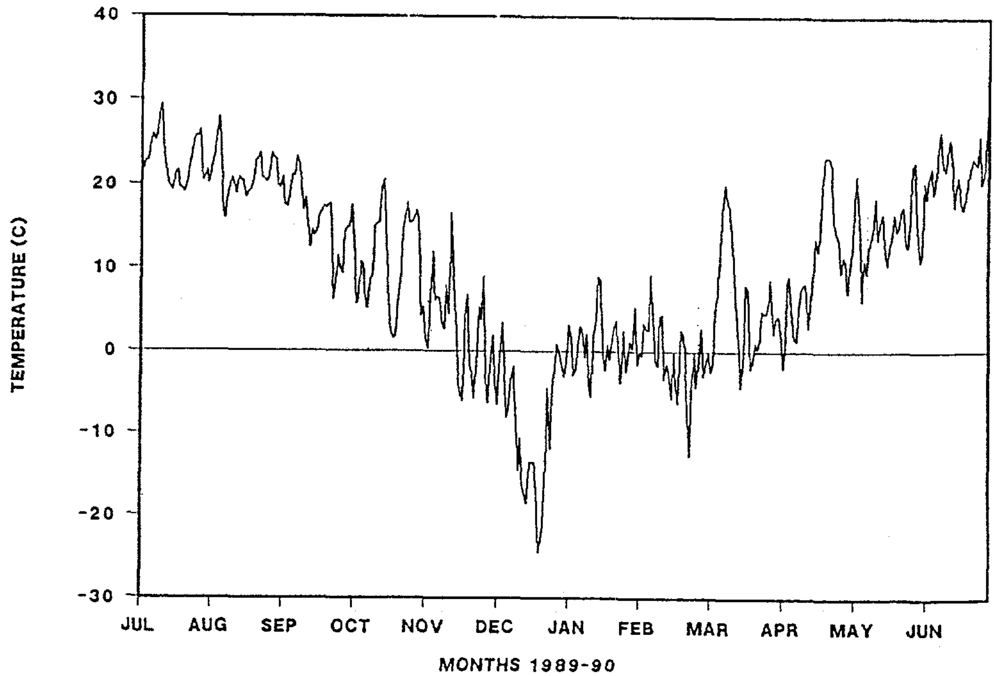

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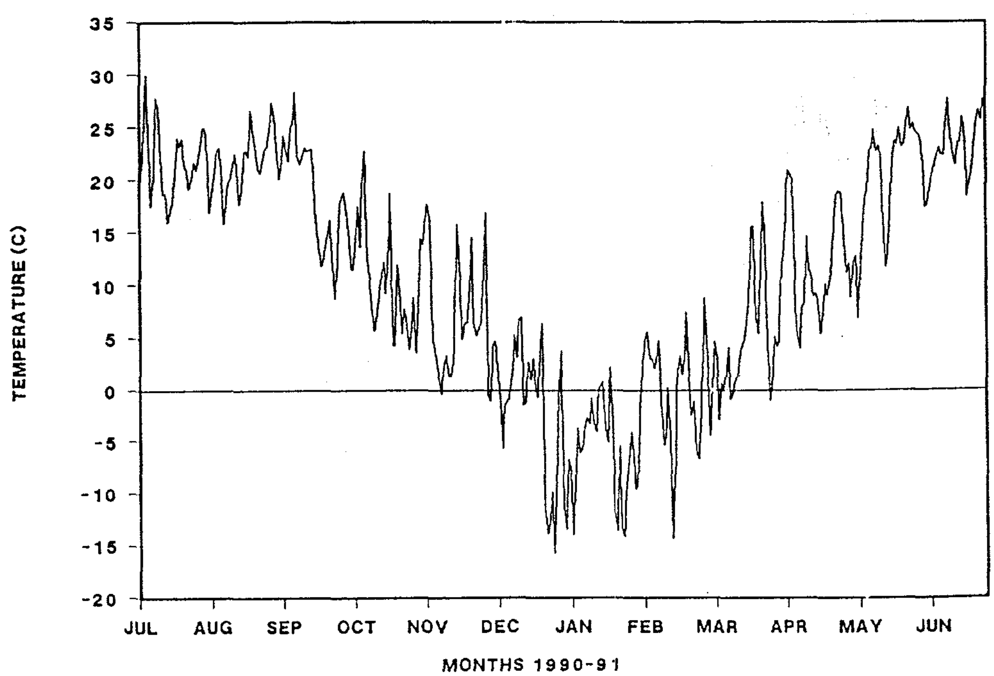

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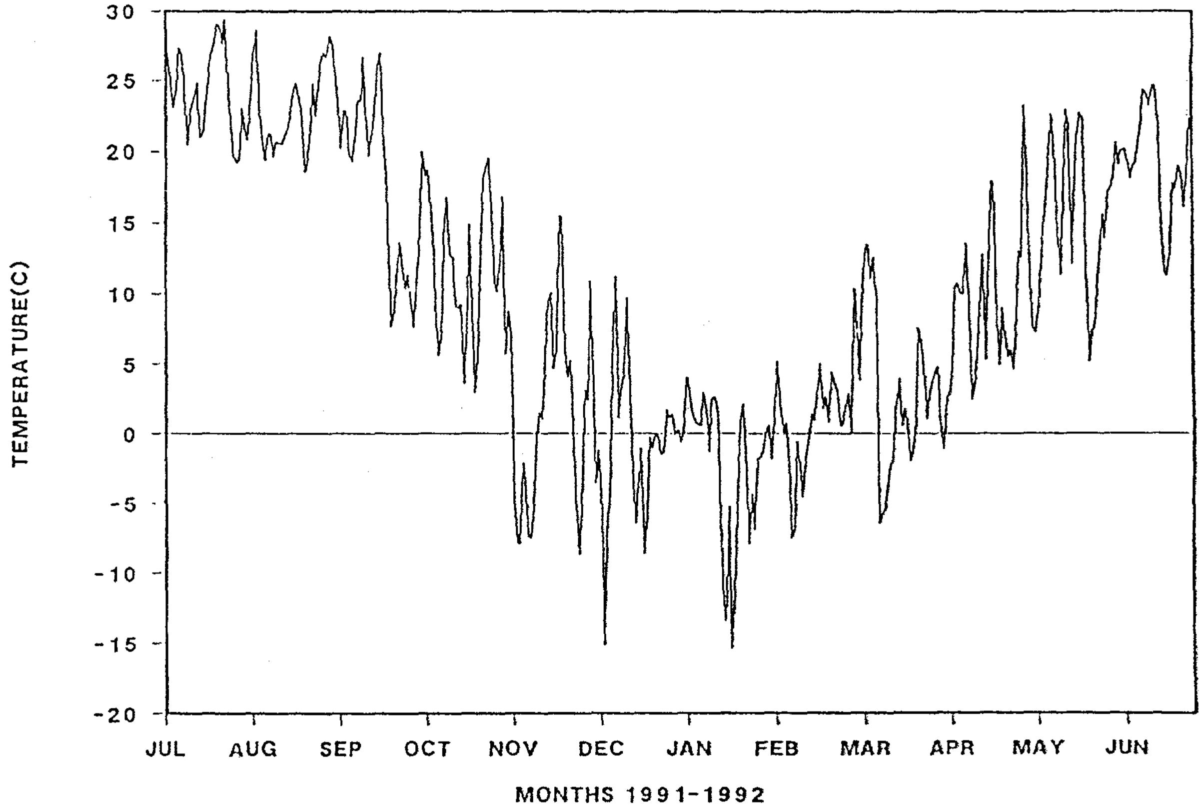

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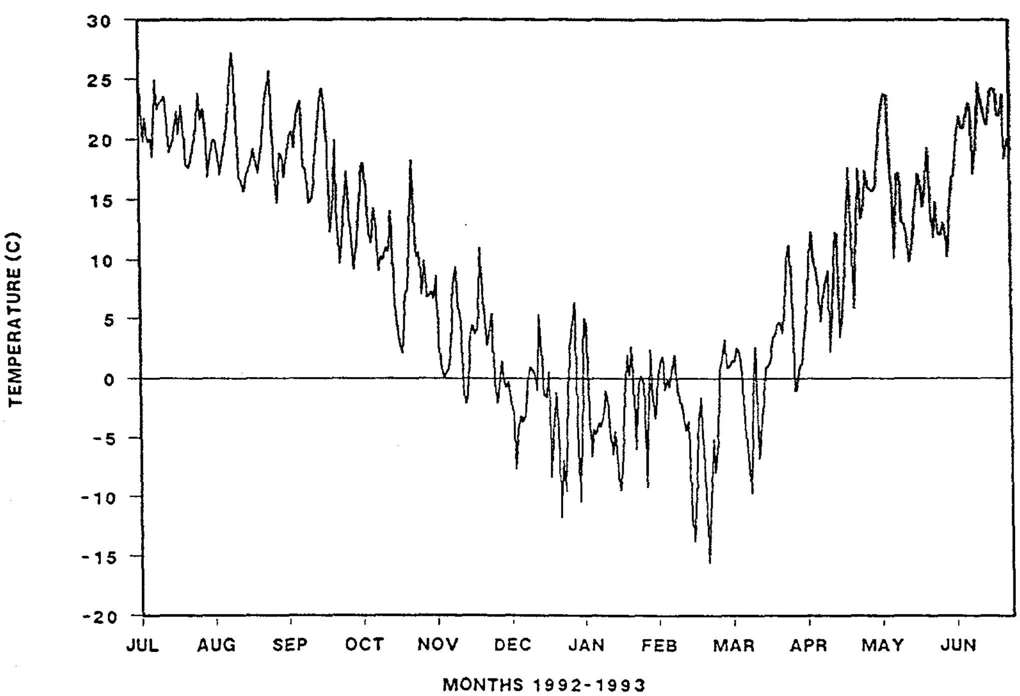

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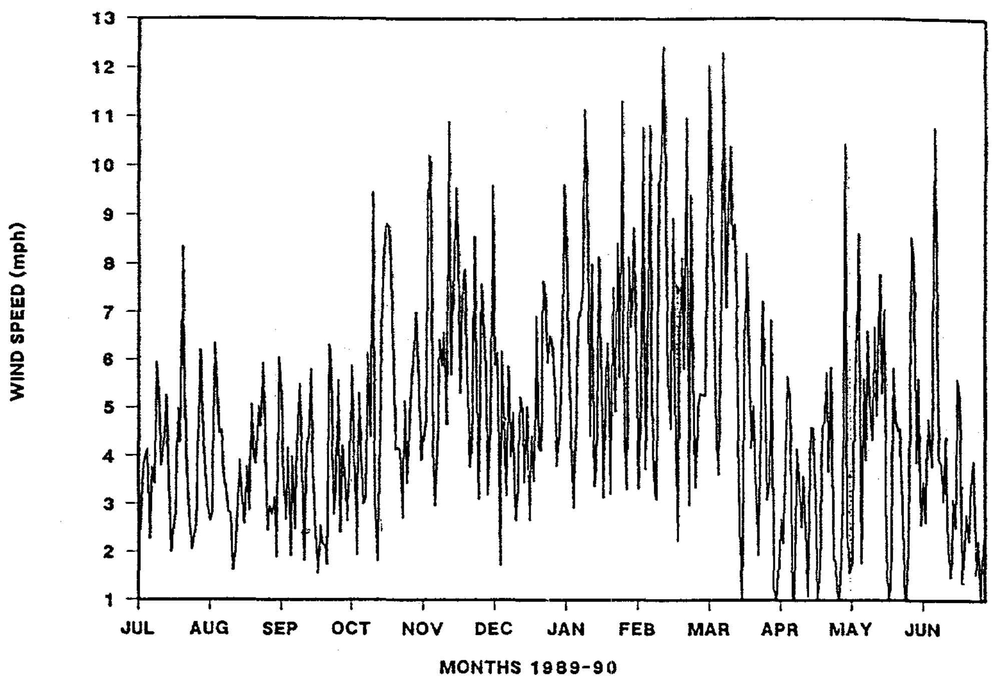

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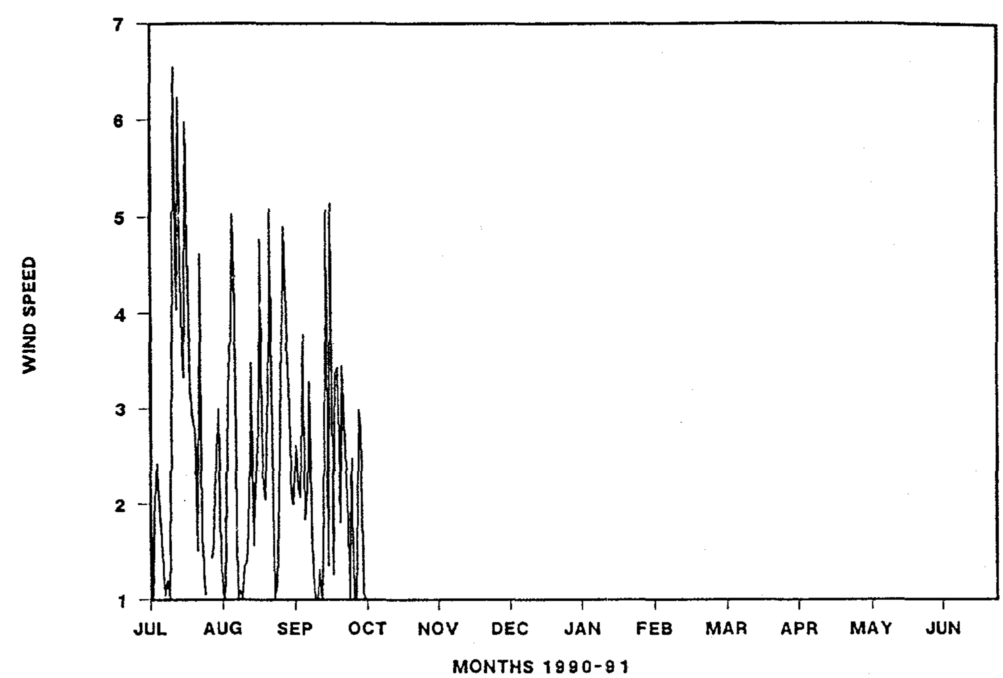

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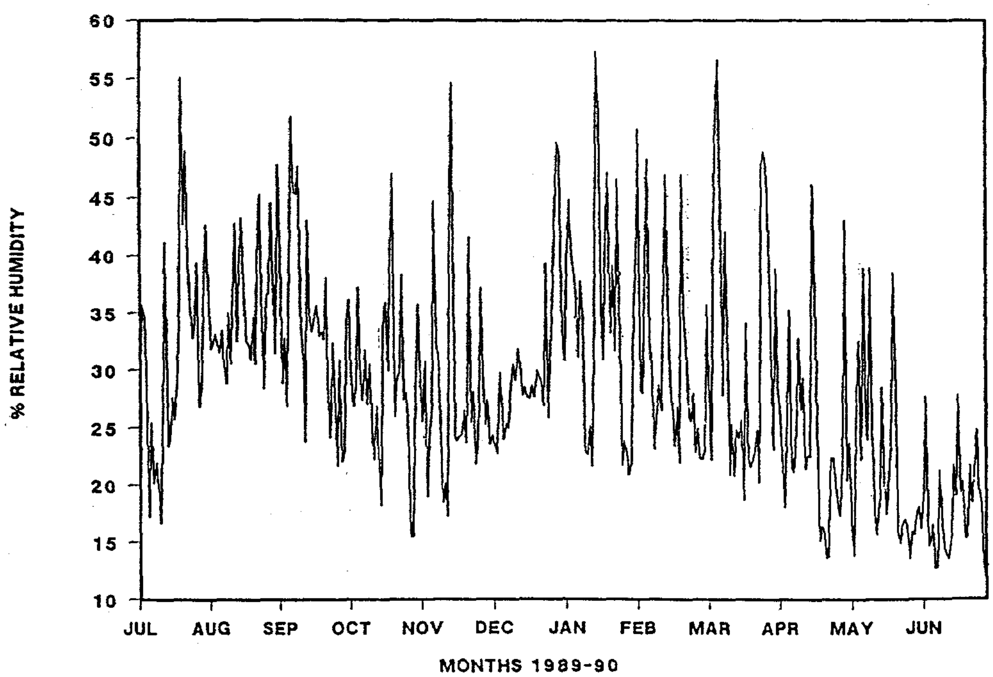

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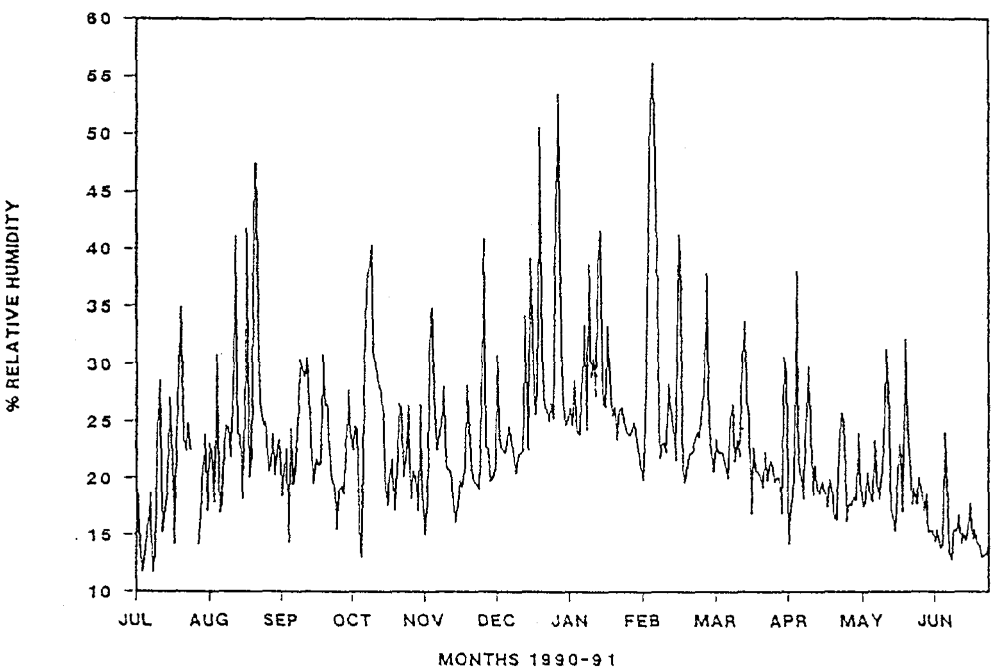

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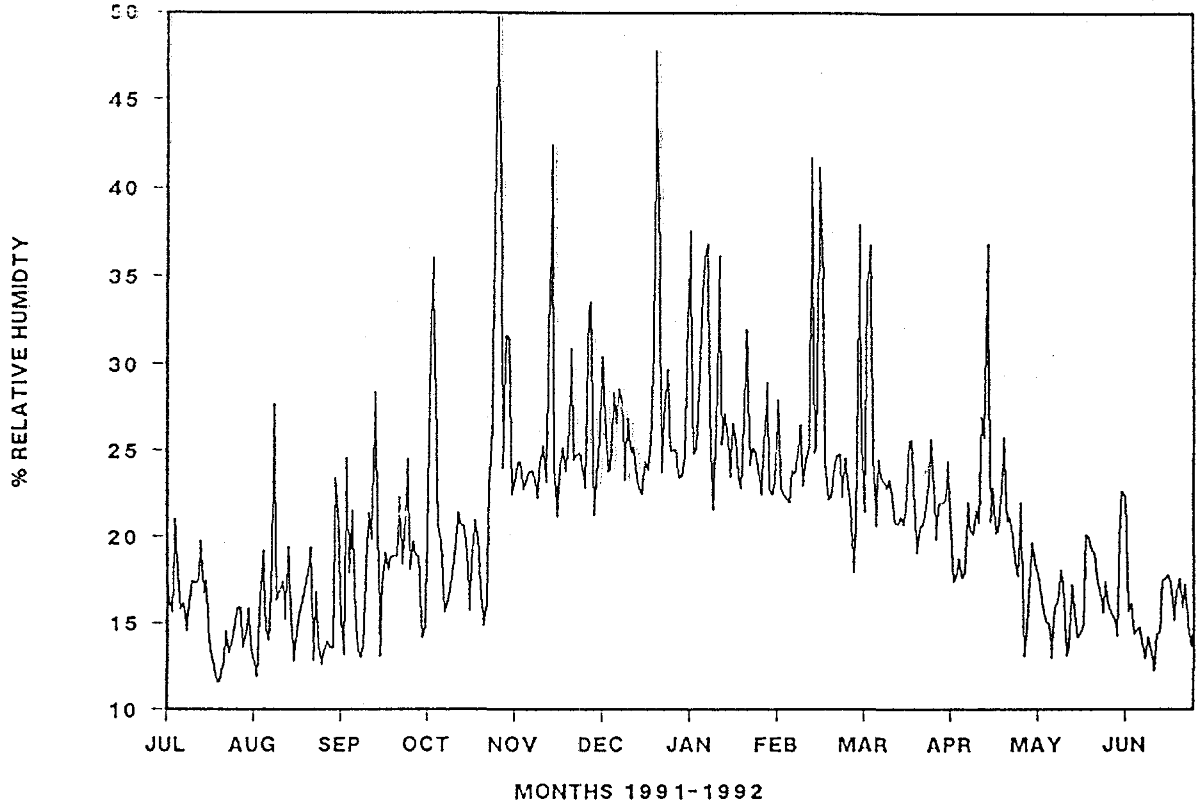

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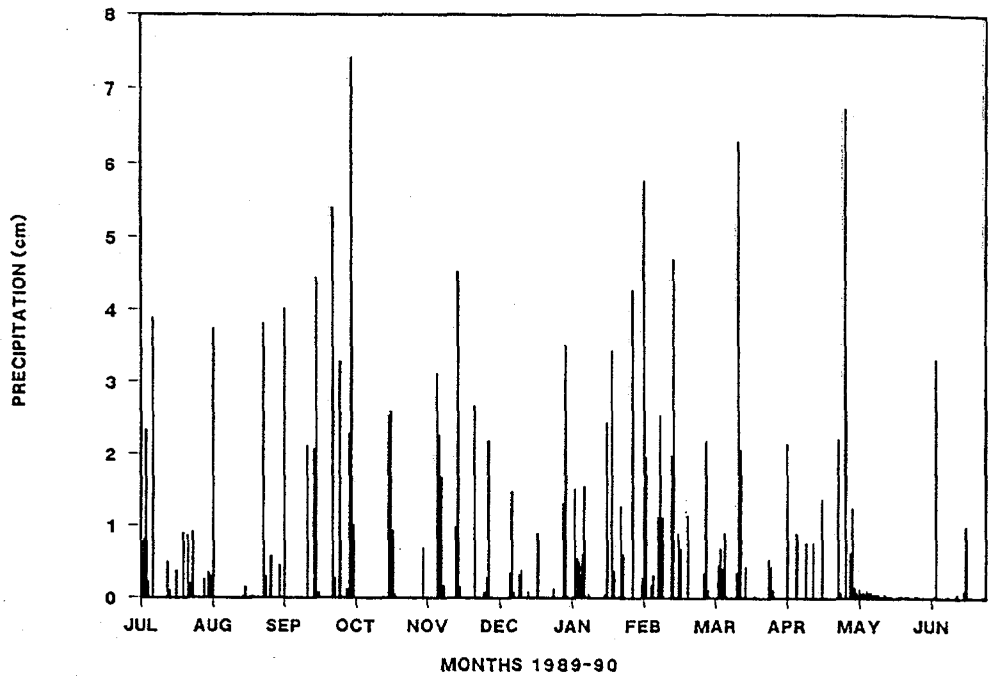

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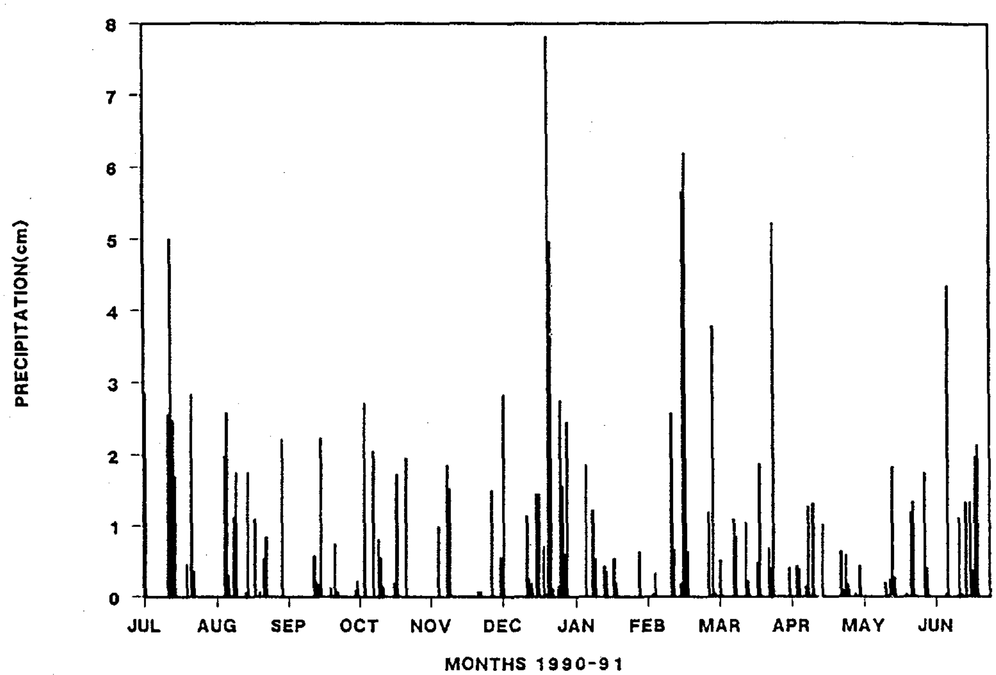

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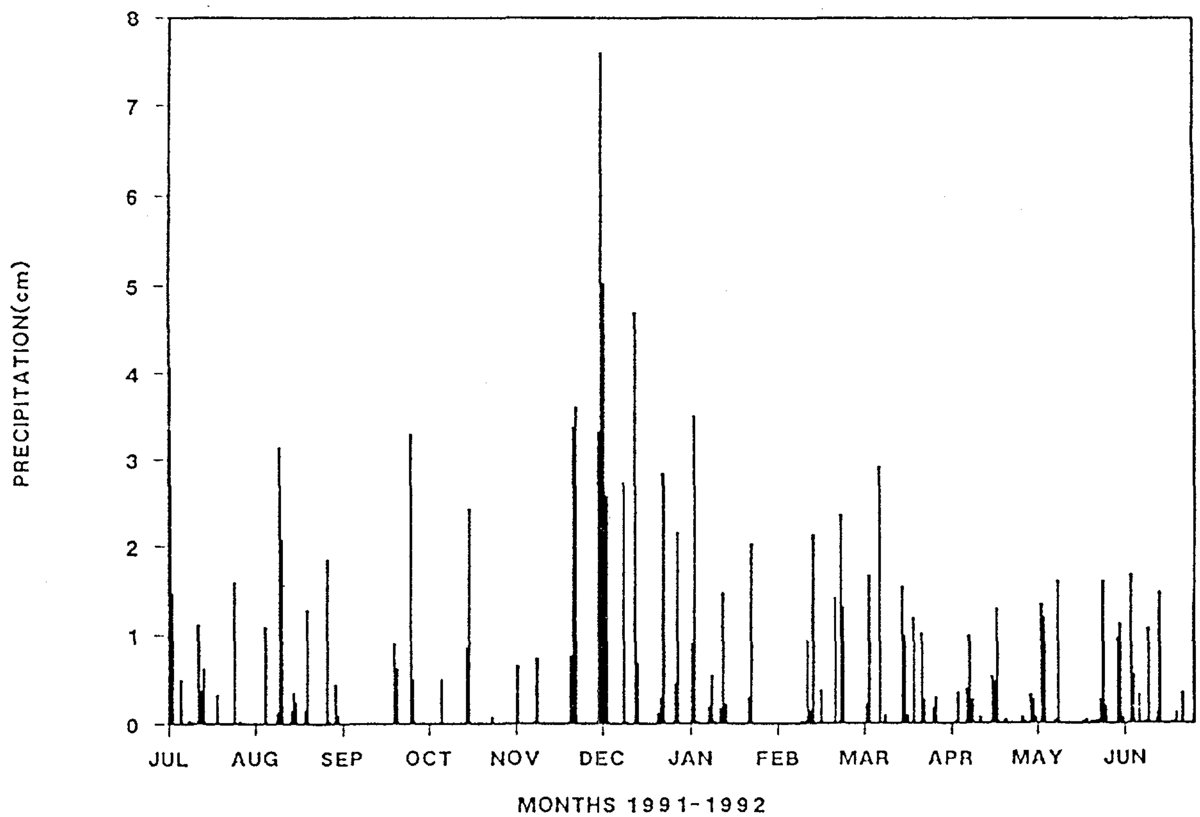

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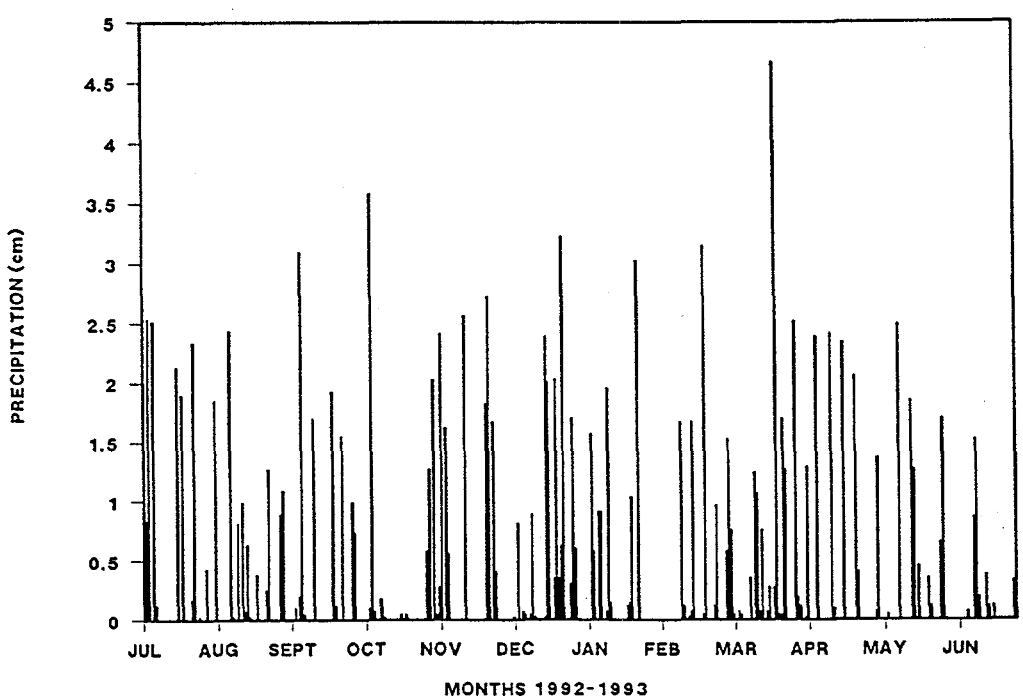

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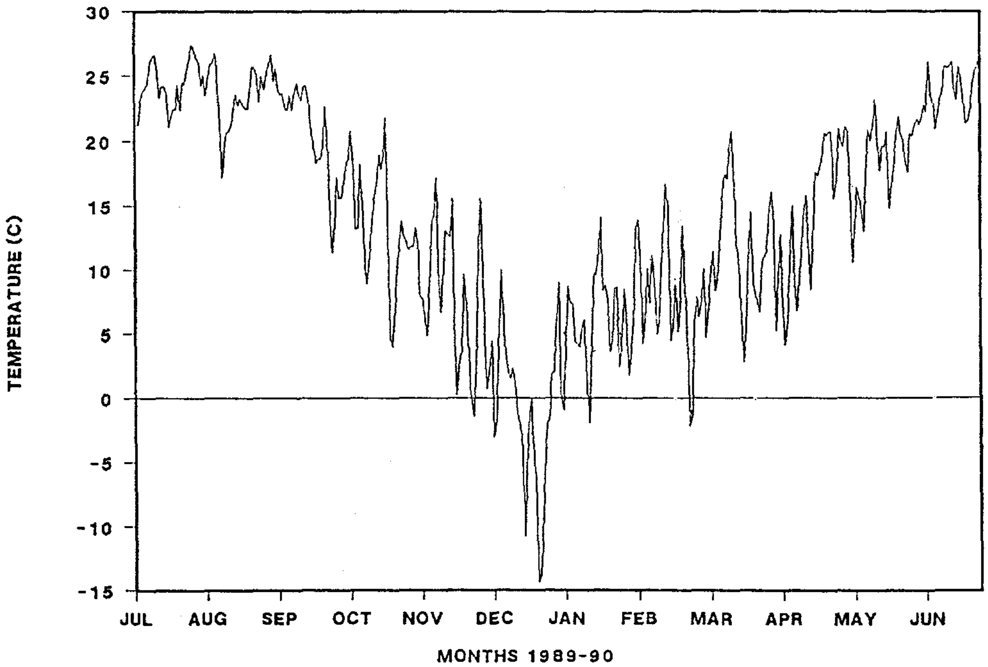

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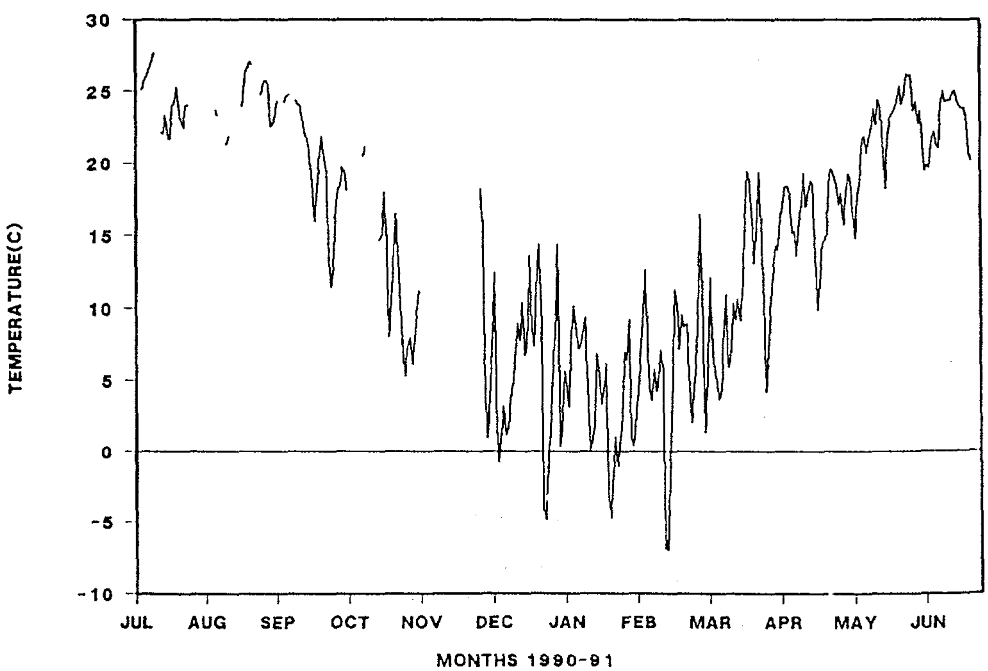

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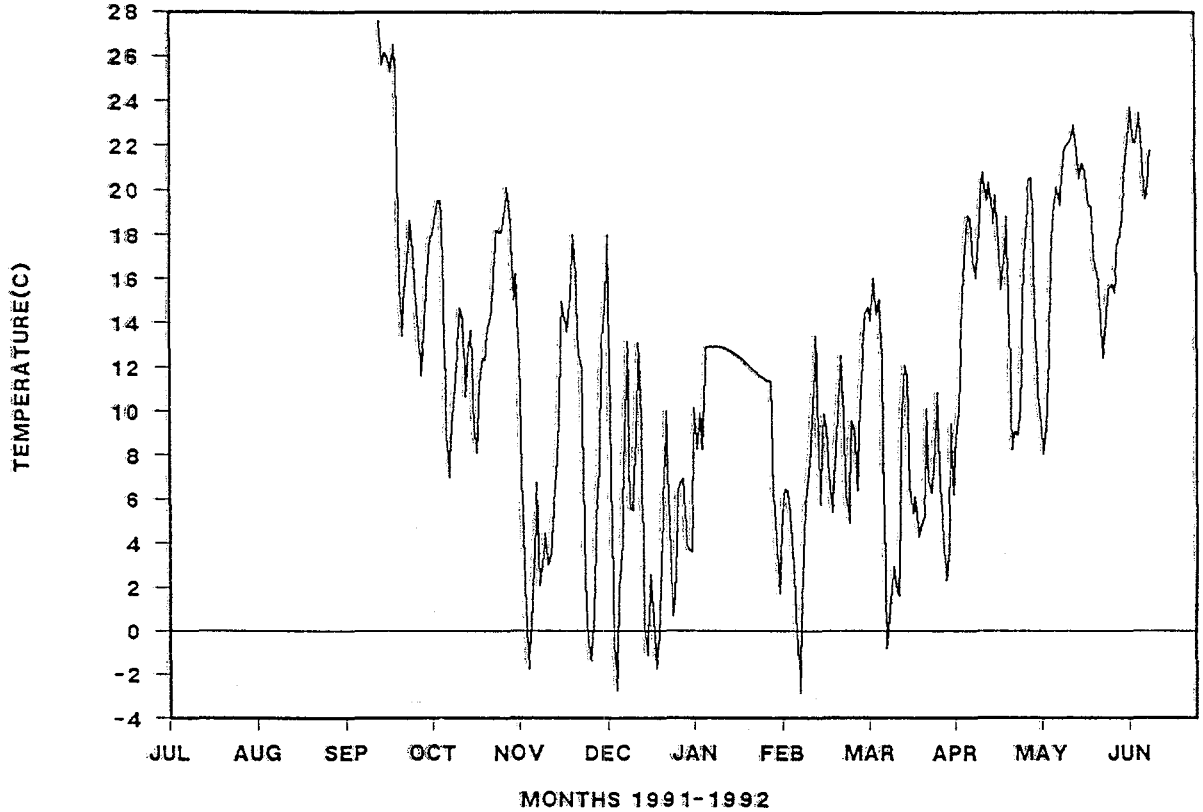

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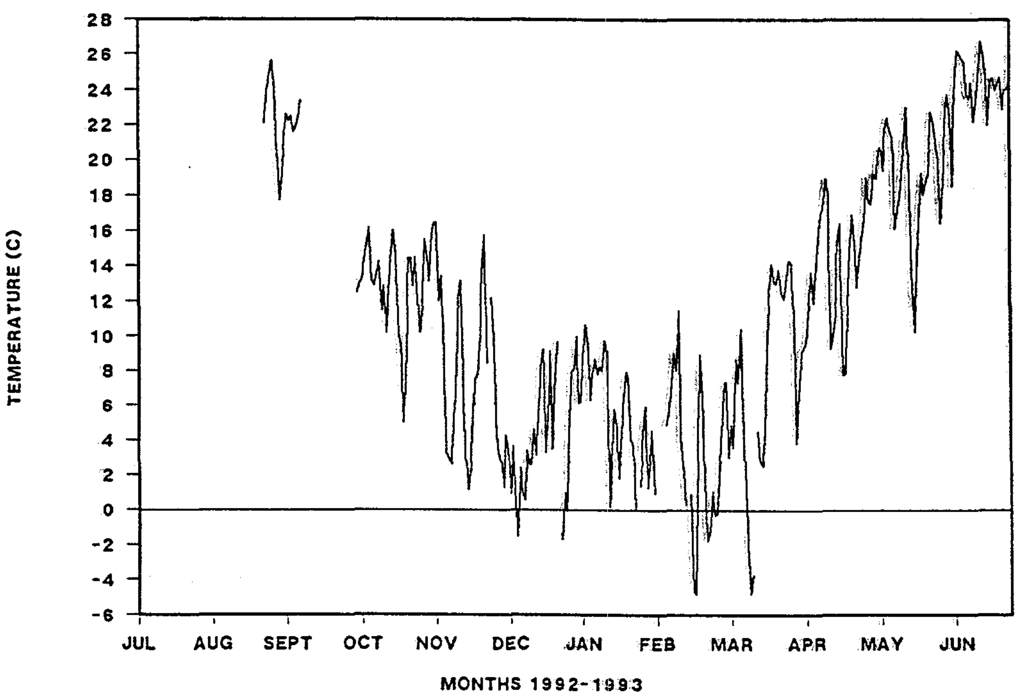

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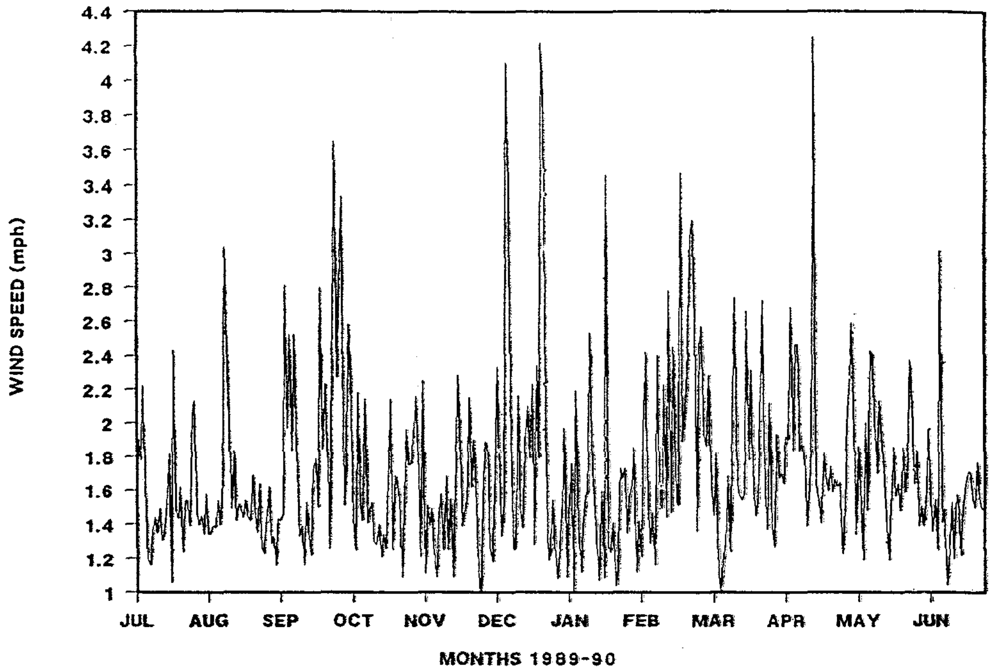

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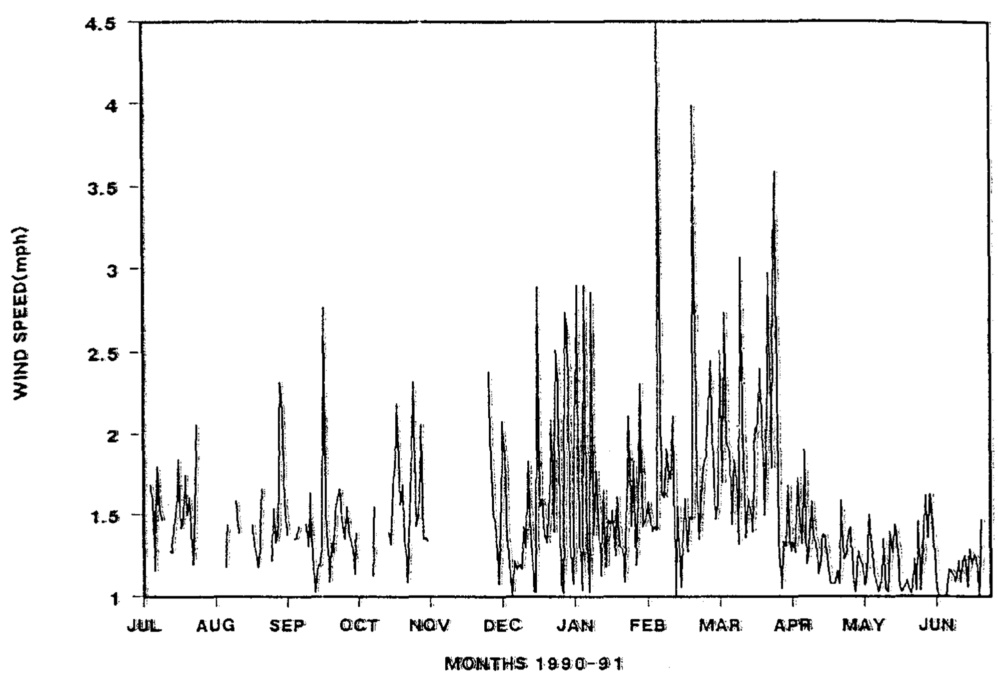

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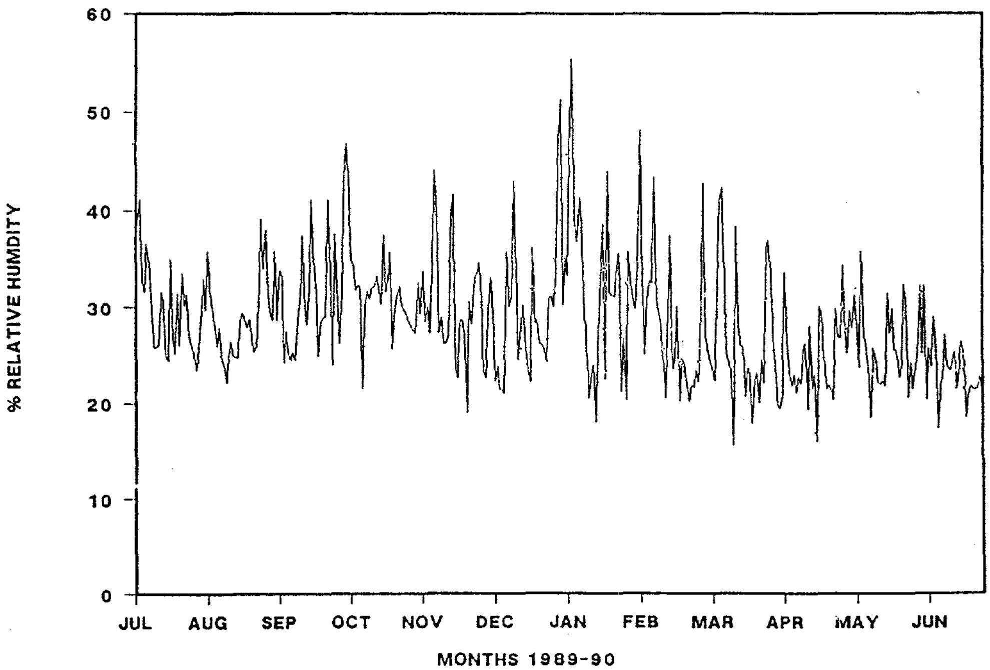

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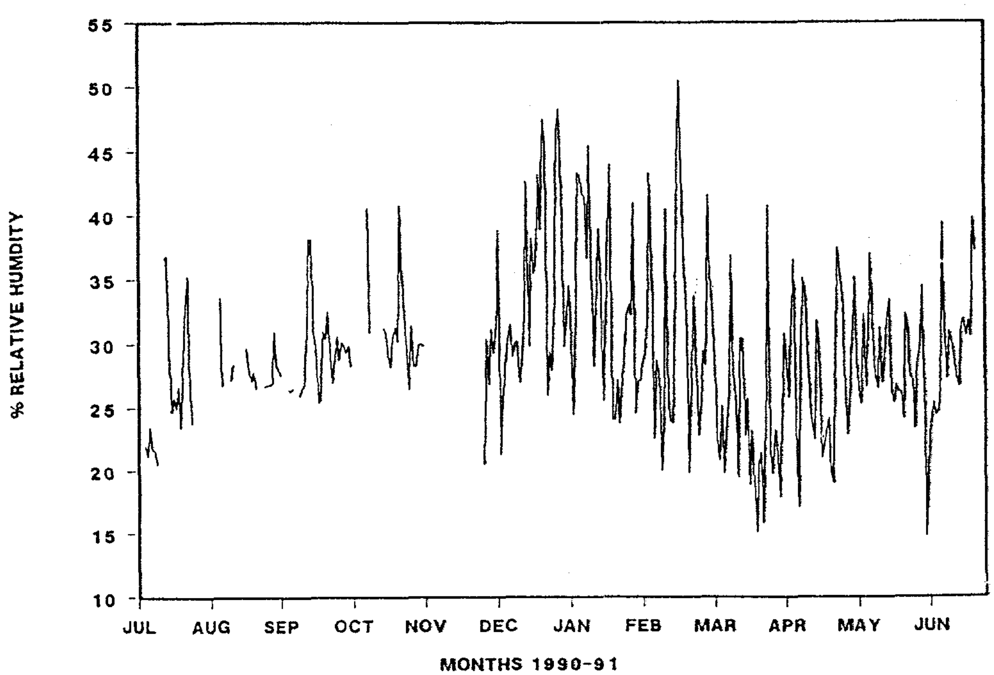

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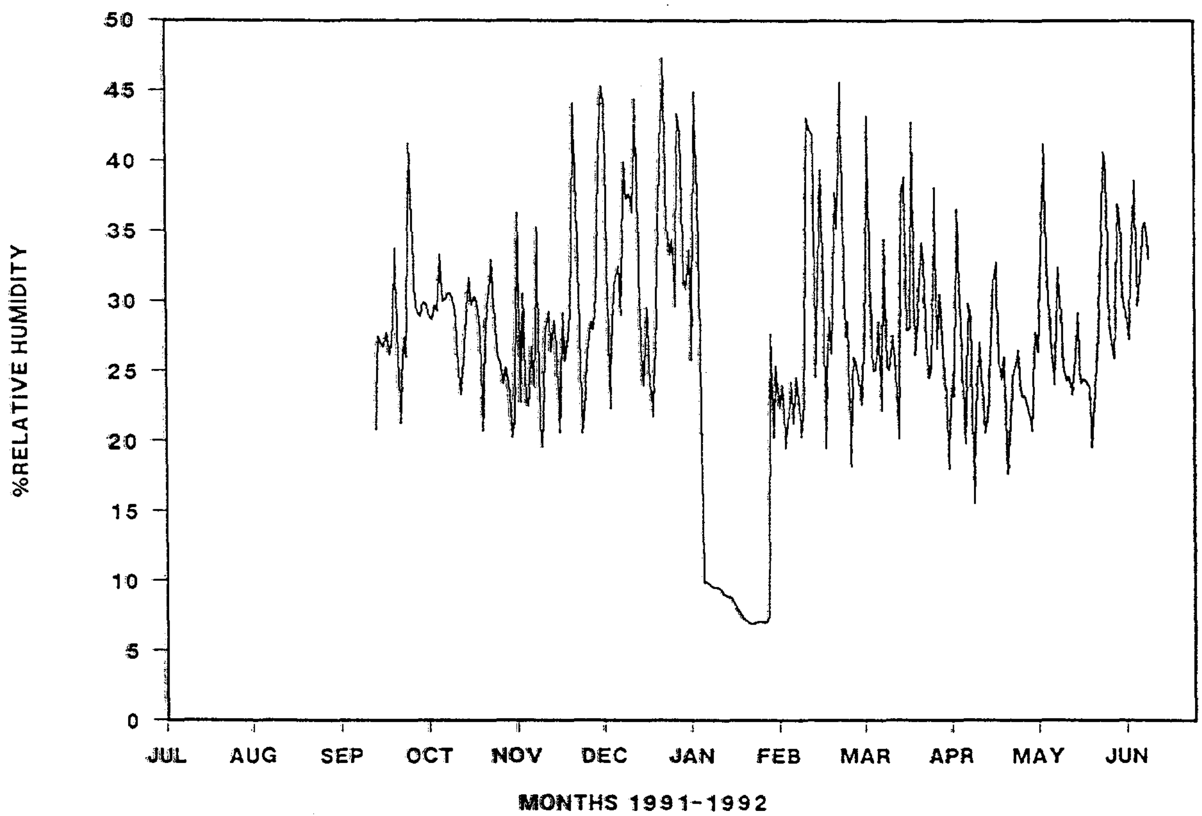

Figure A-26. ORNL weather data for 1991-92—relative humidity.

Appendix B

Soil Temperature Data—Resistance Probes

Appendix B
Soil Temperature Data—Resistance Probes

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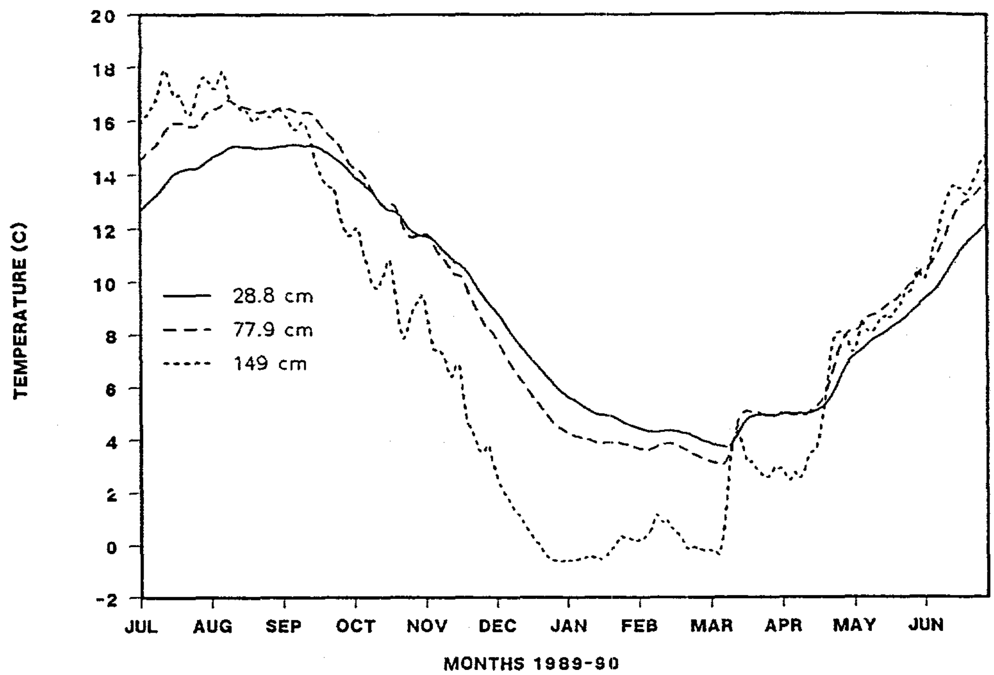
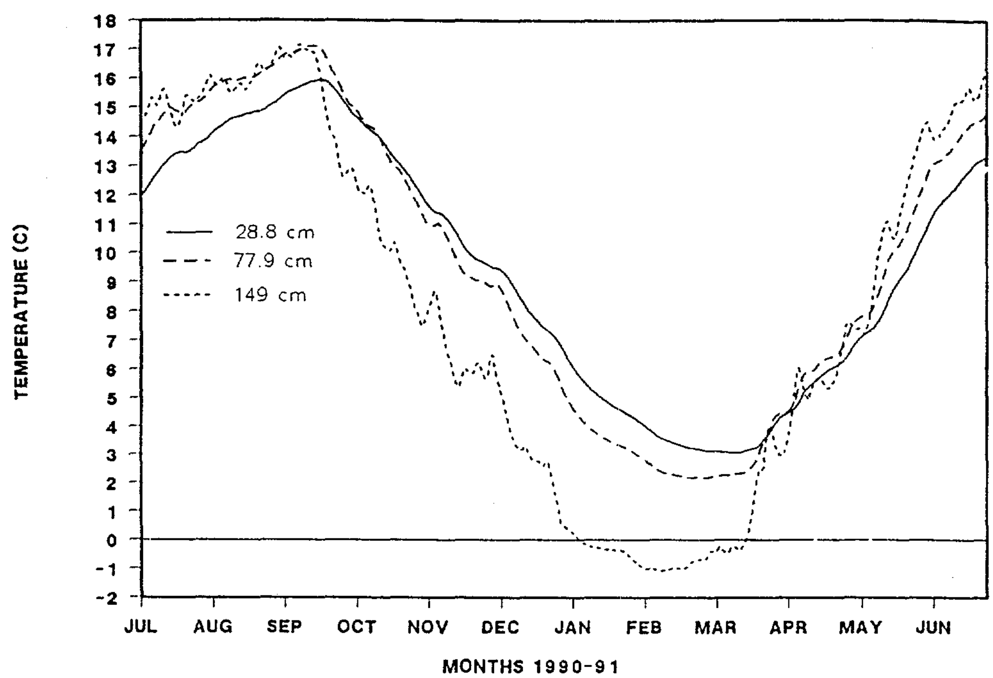


Figure B-1. ANL-E lysimeter 1 soil temperatures for 1989-90.



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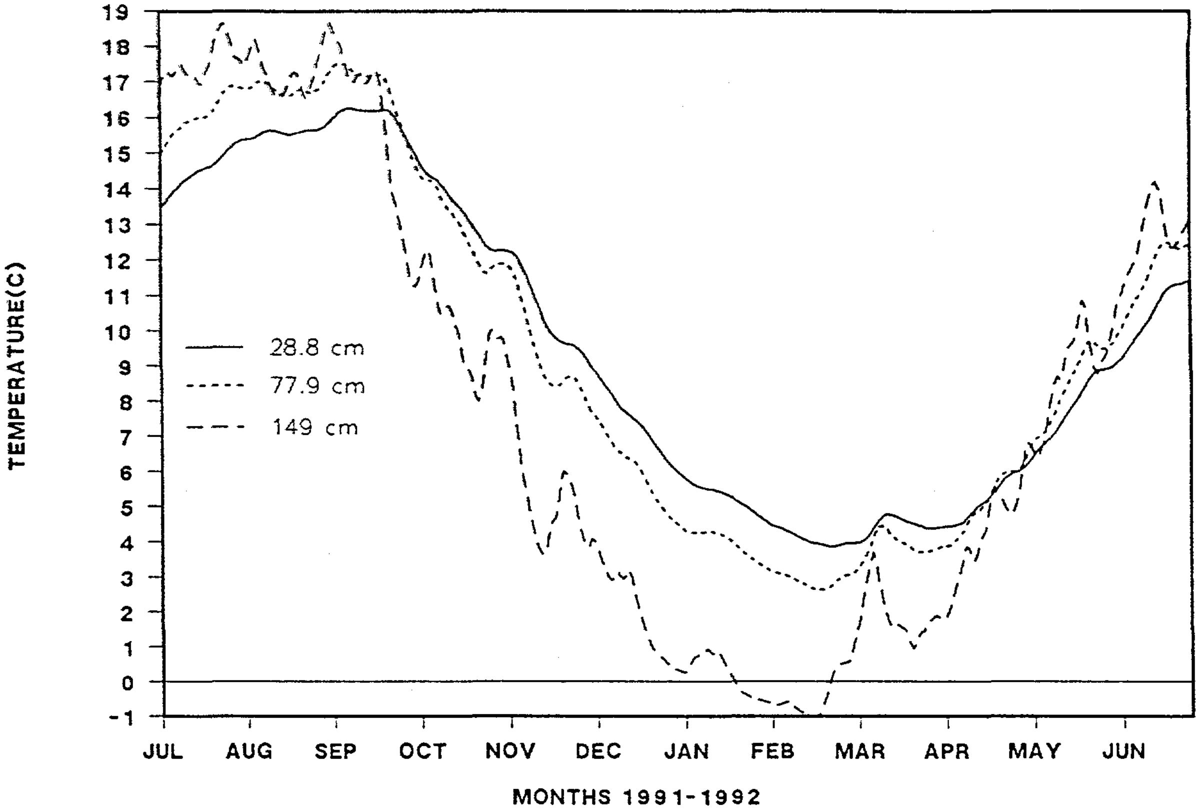
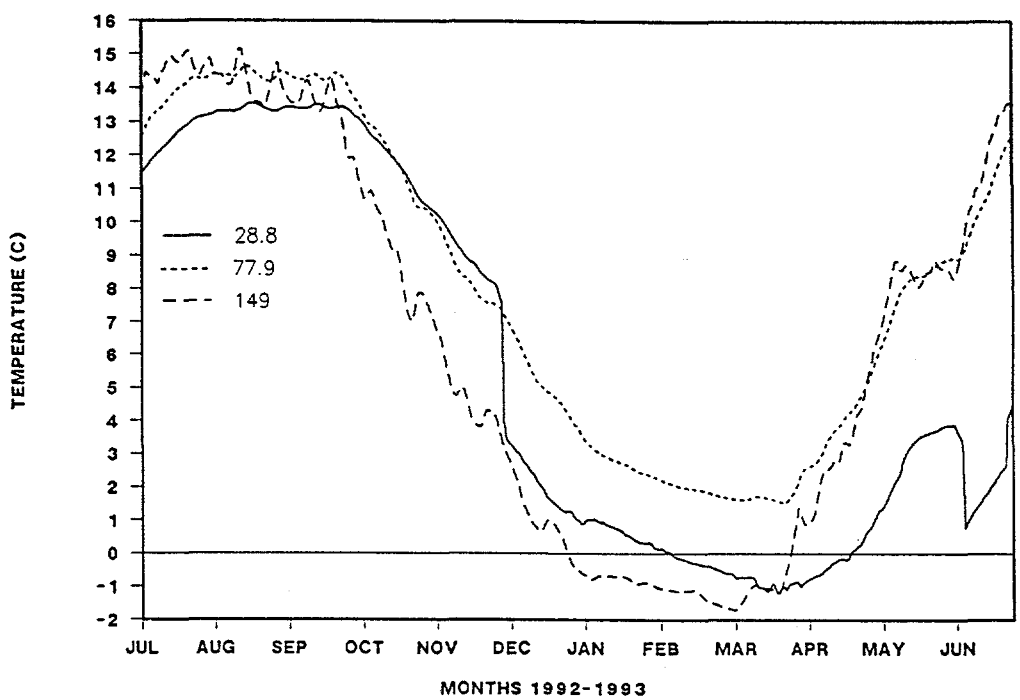


Figure B-3. ANL-E lysimeter 1 soil temperatures for 1991-92.



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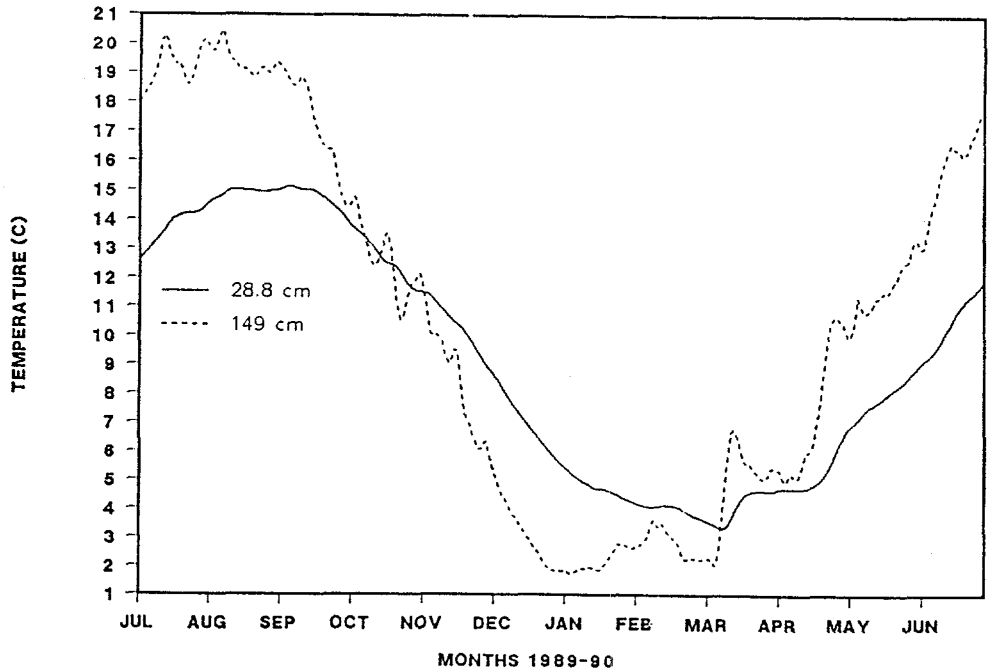

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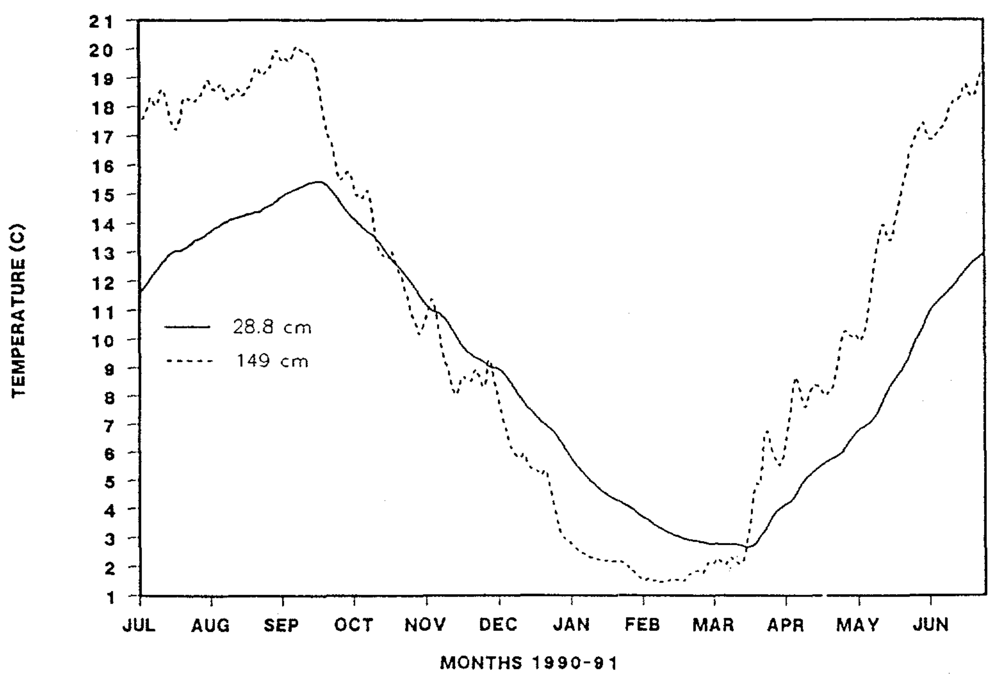

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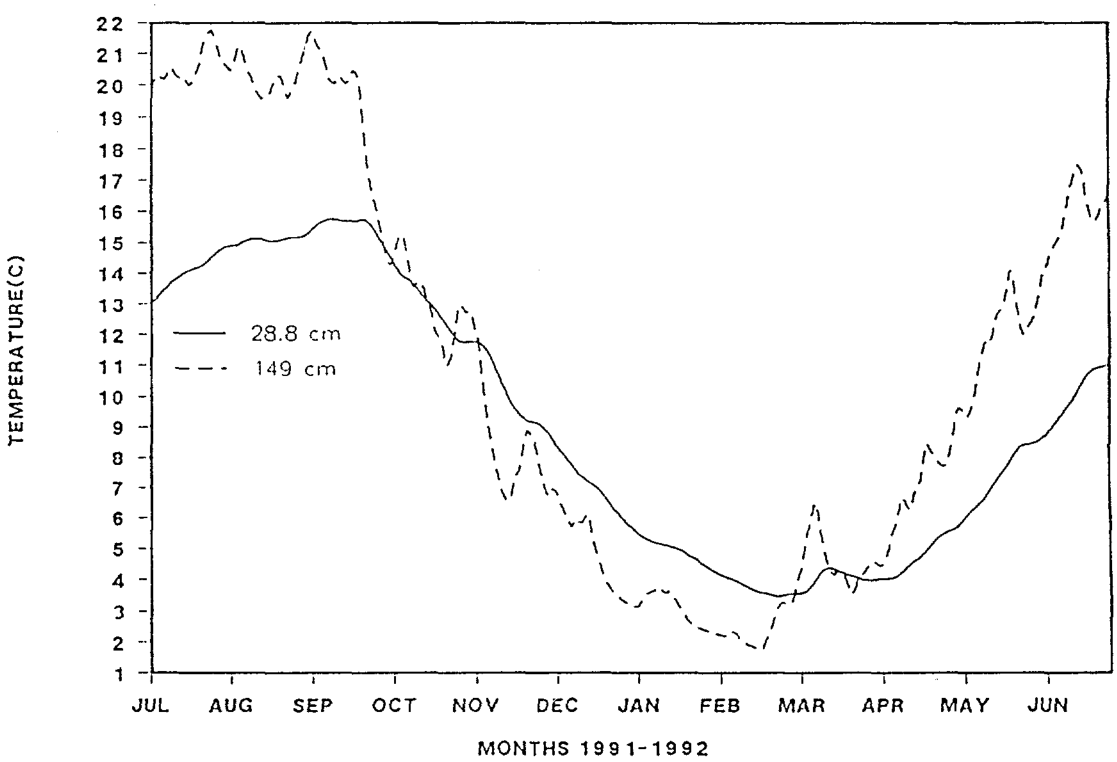

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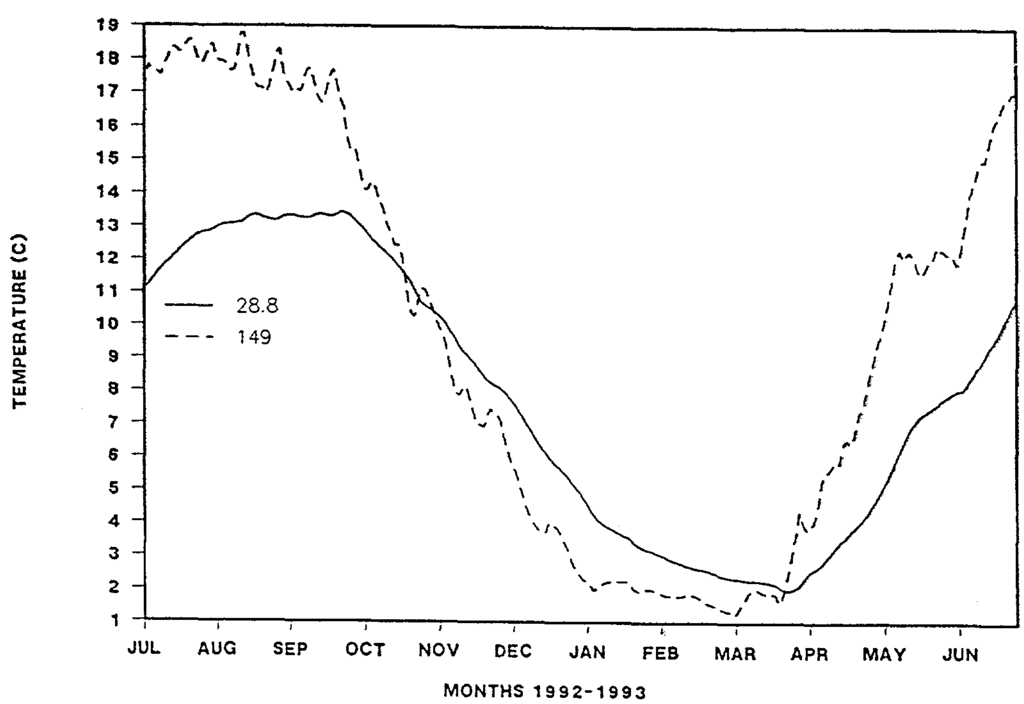

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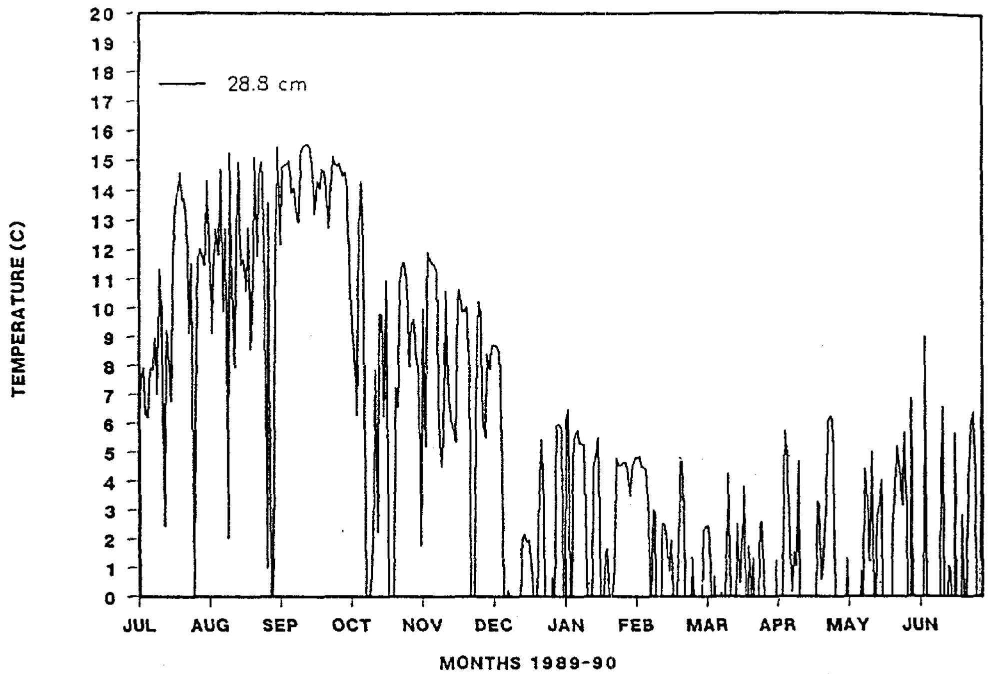

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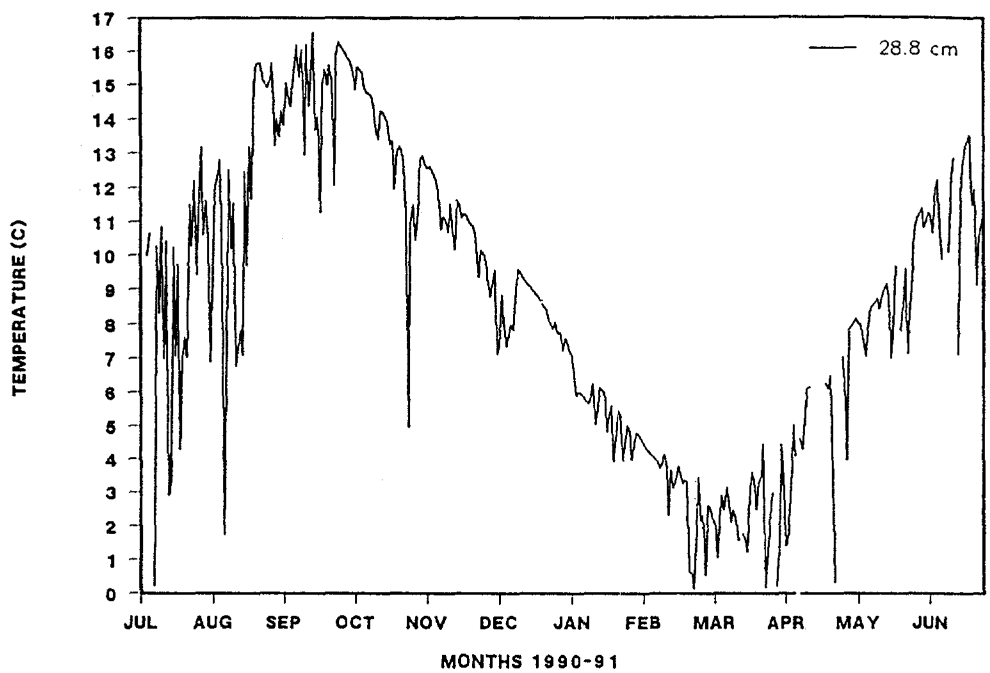

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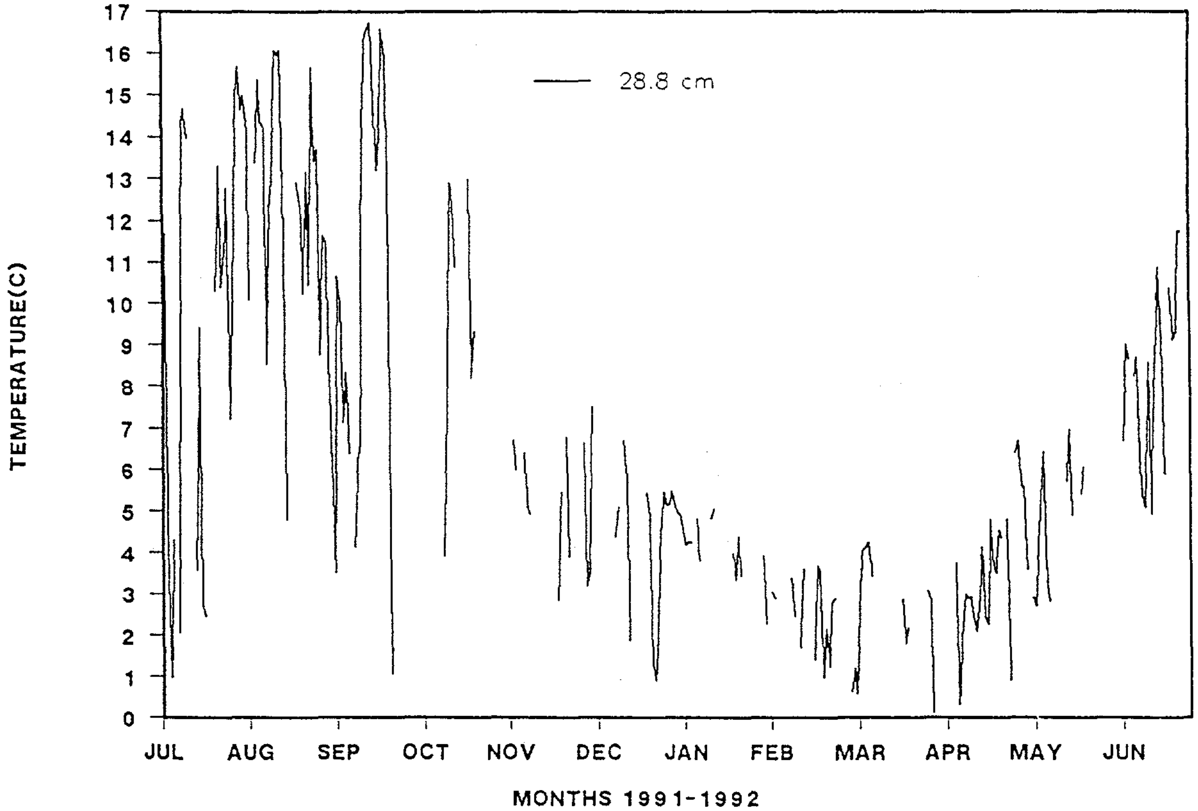

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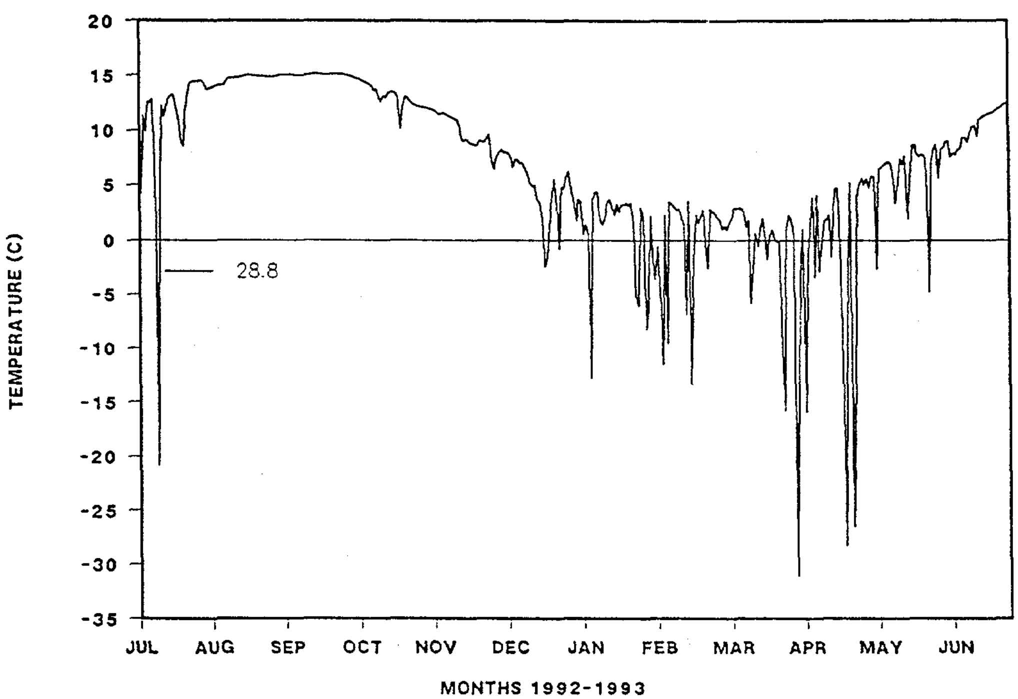

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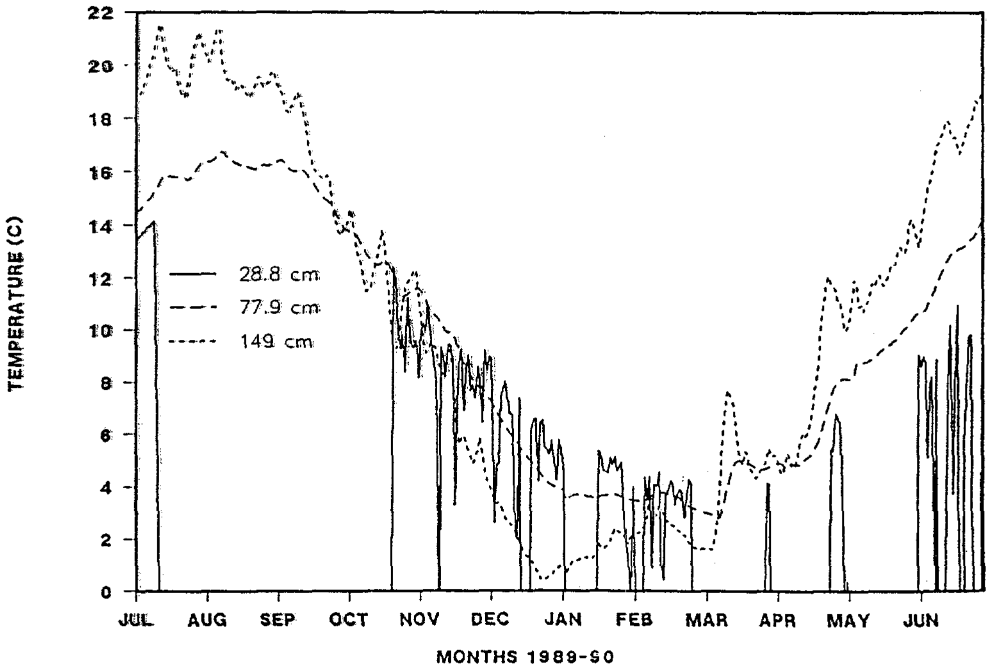

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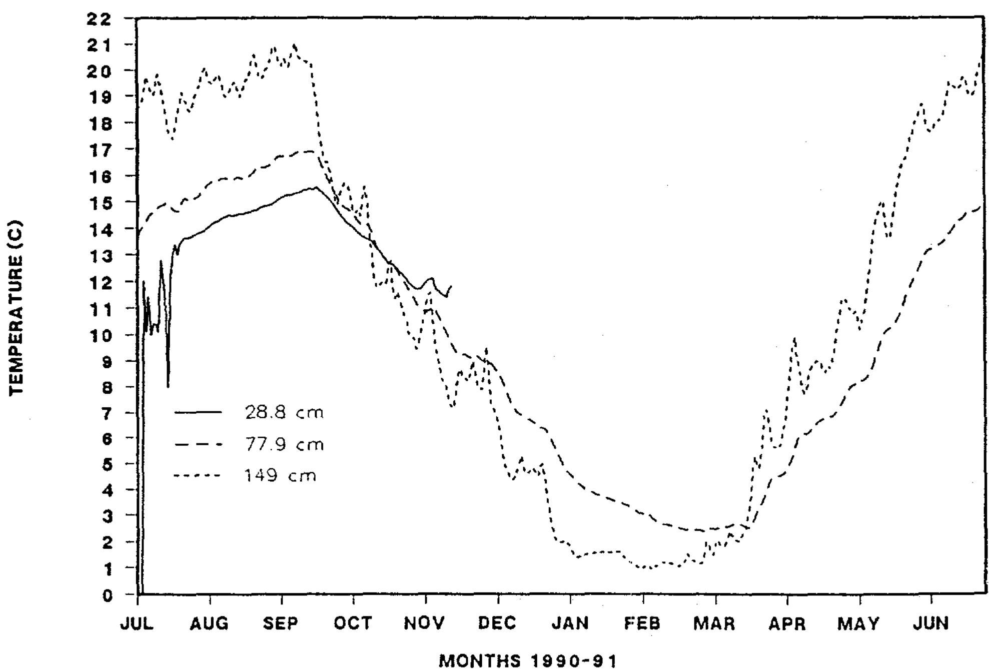

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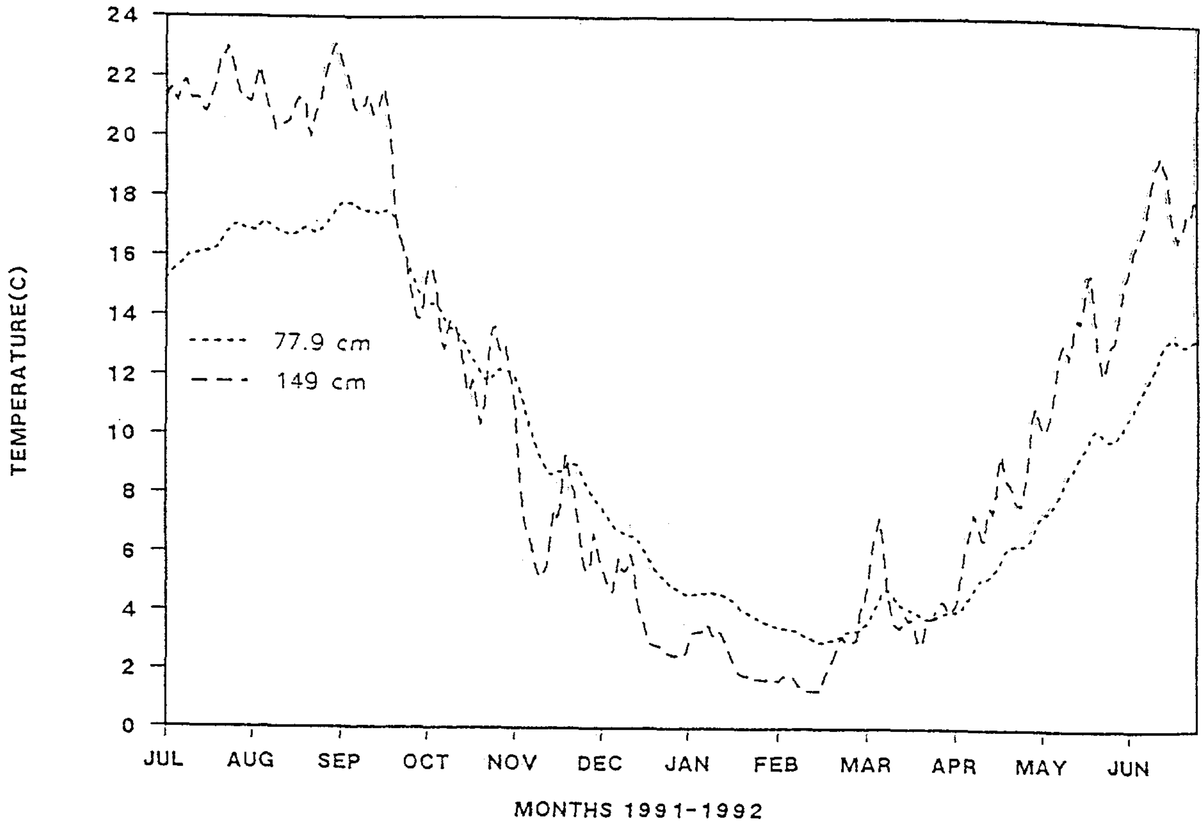

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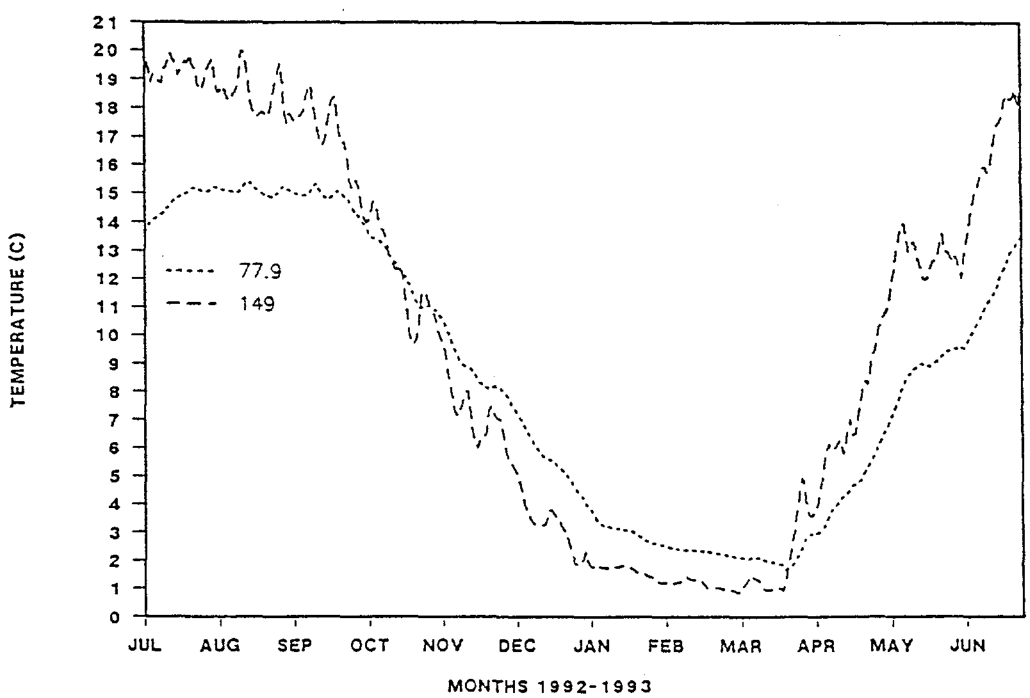

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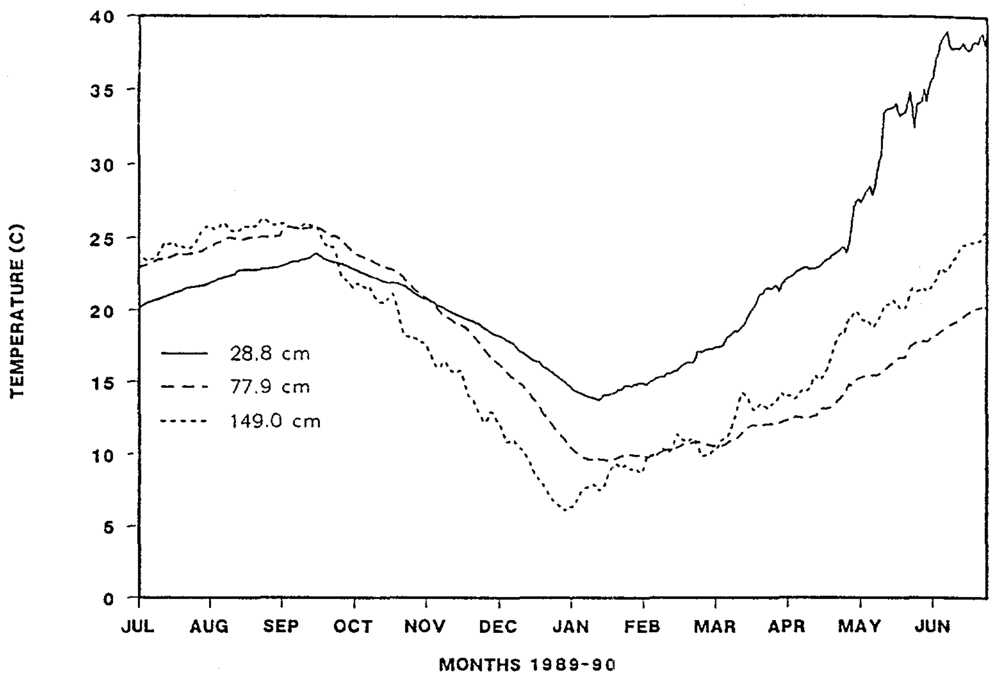

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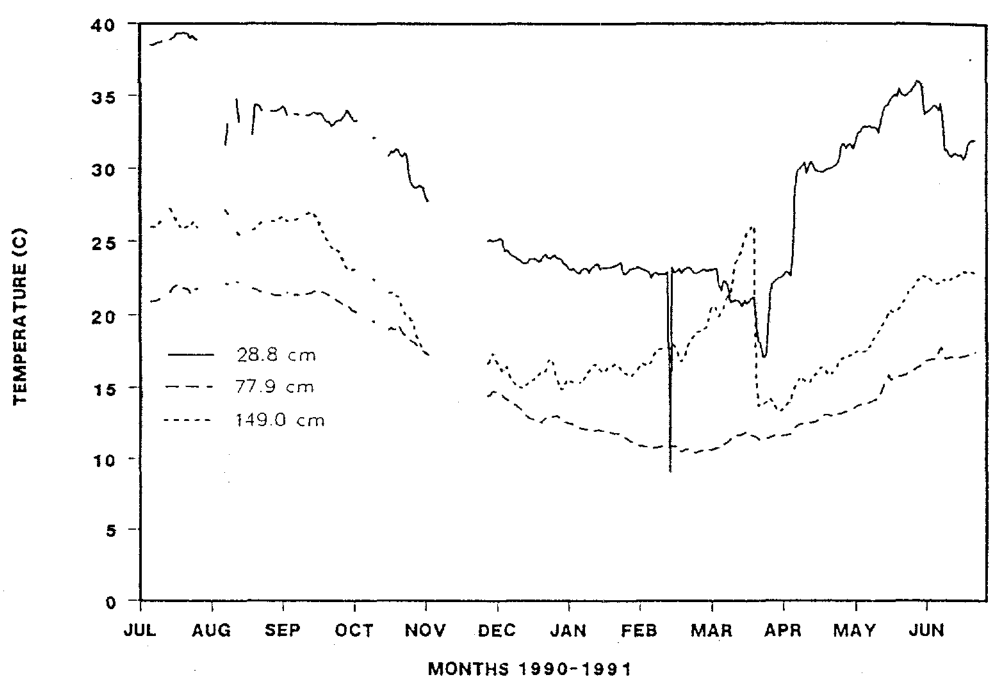

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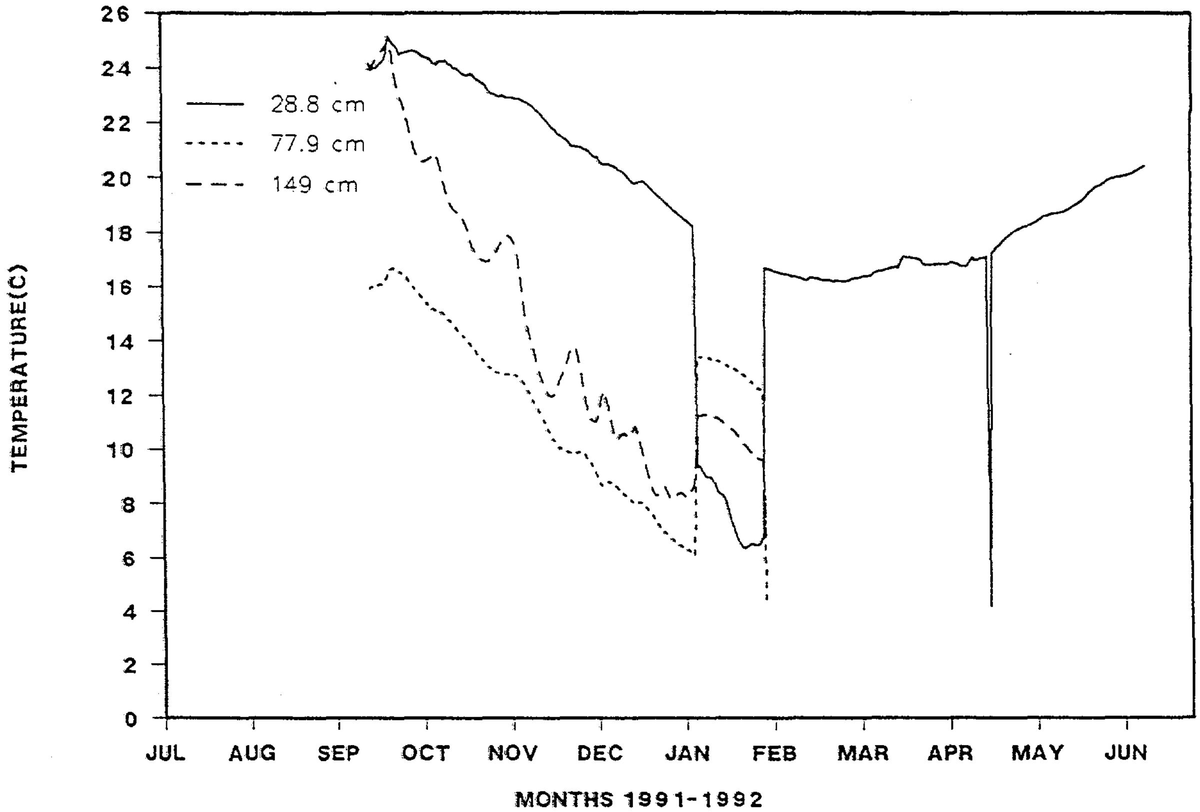

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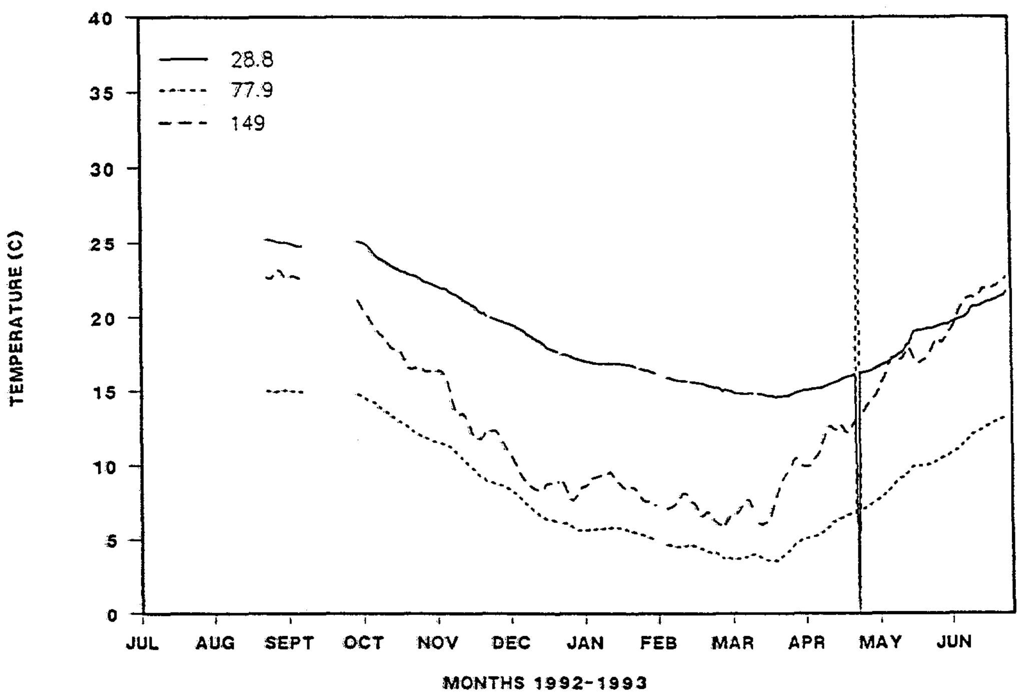

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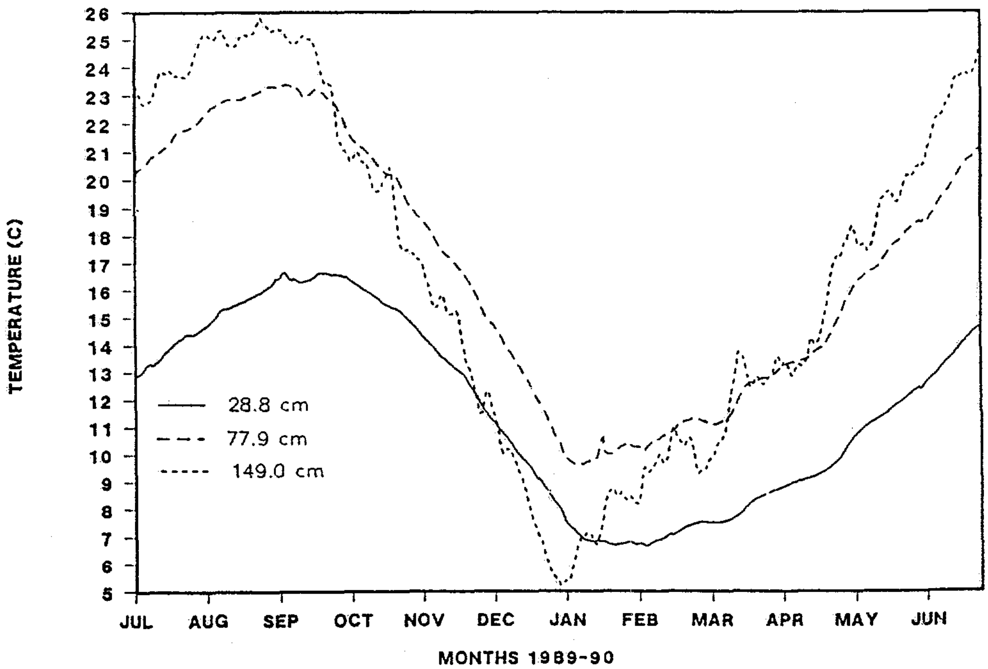

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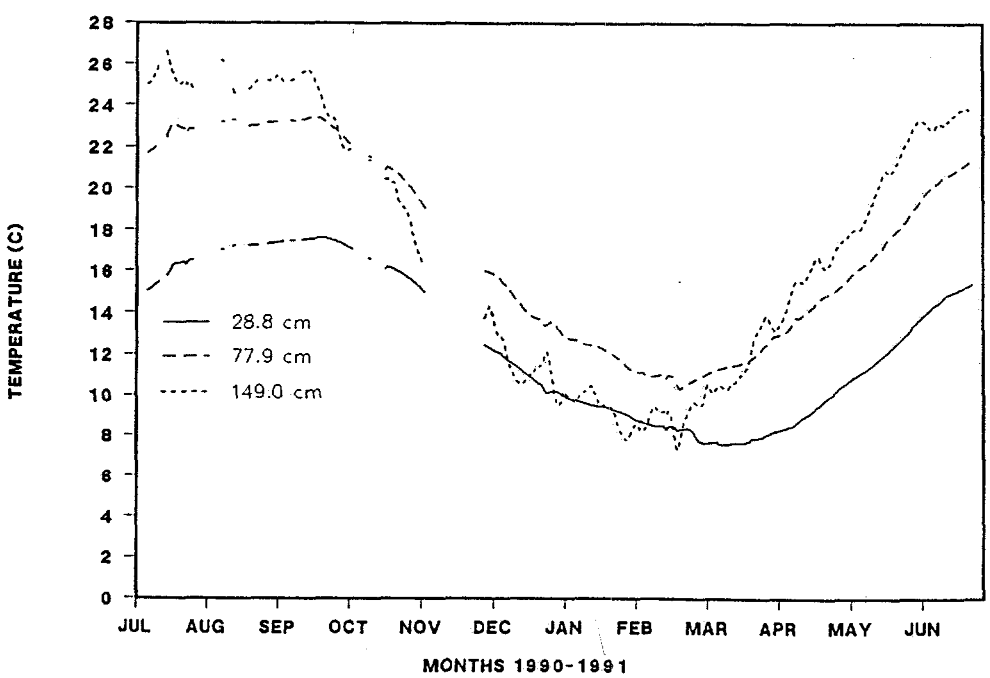

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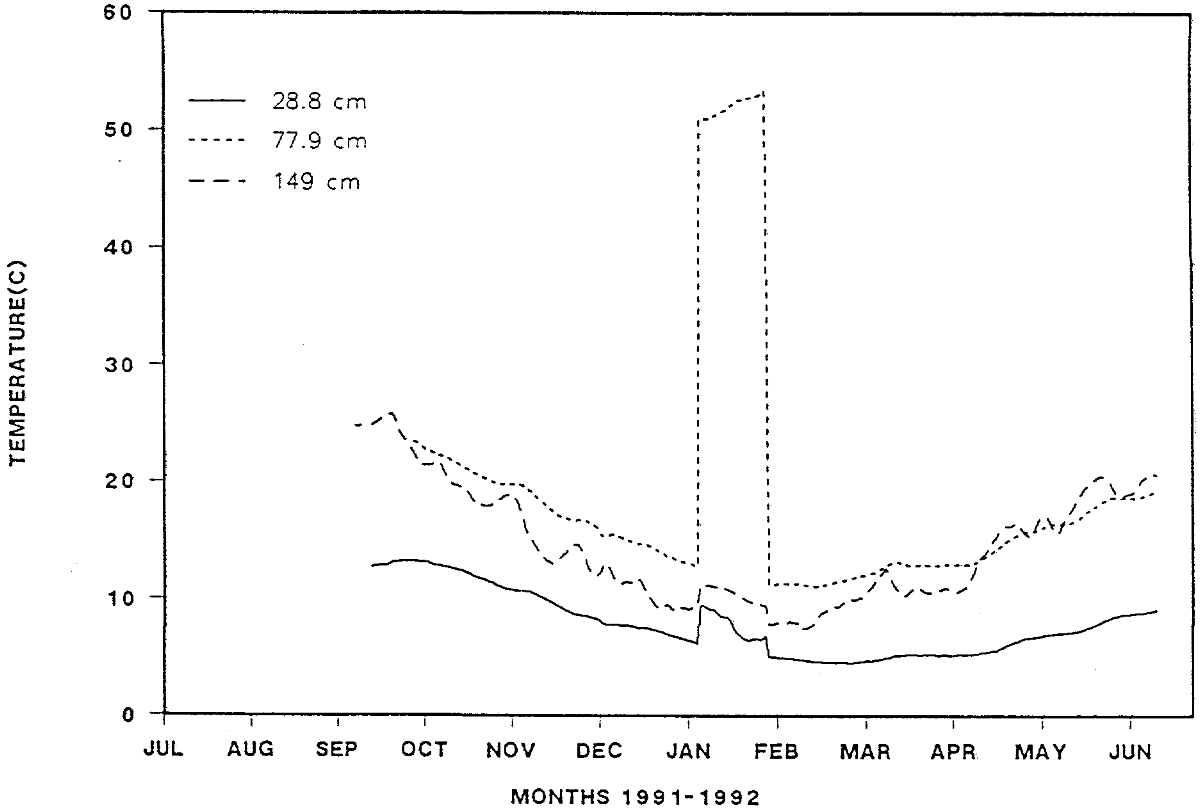

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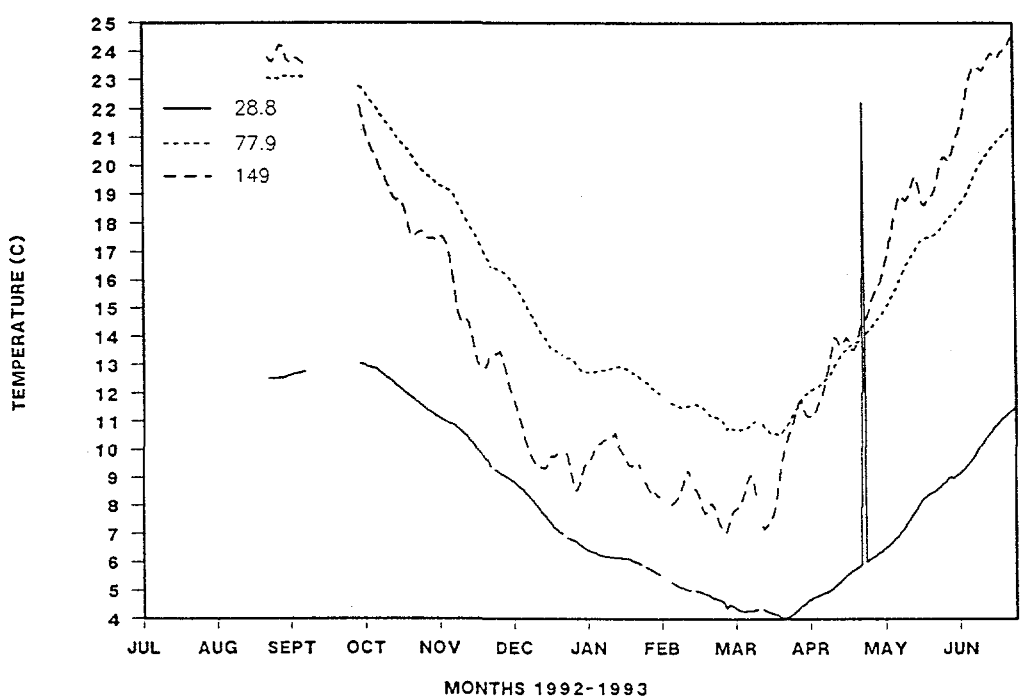

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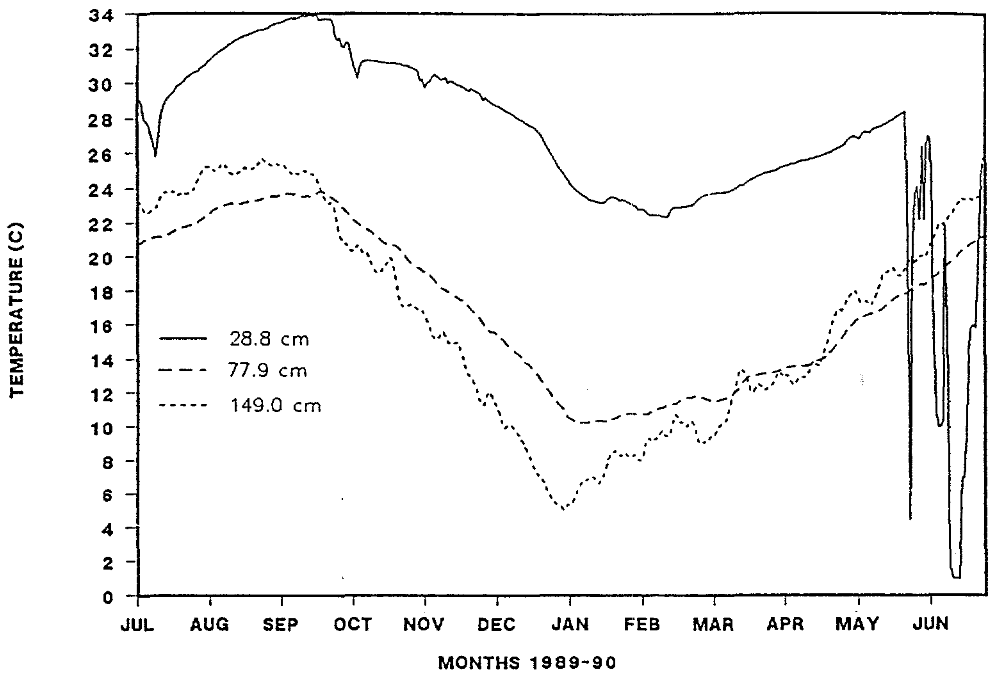

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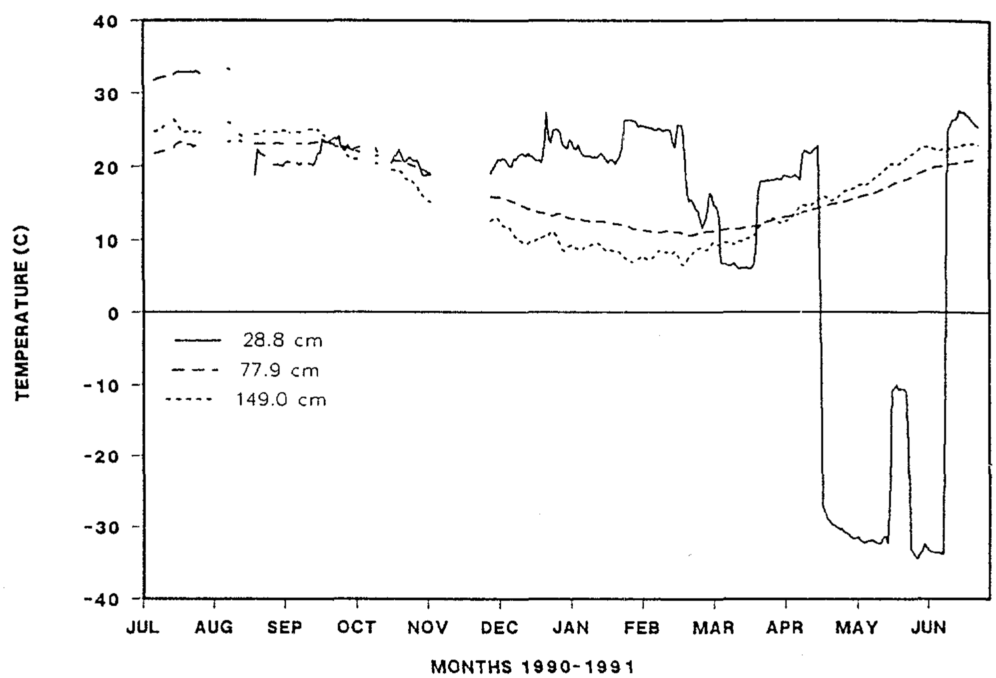

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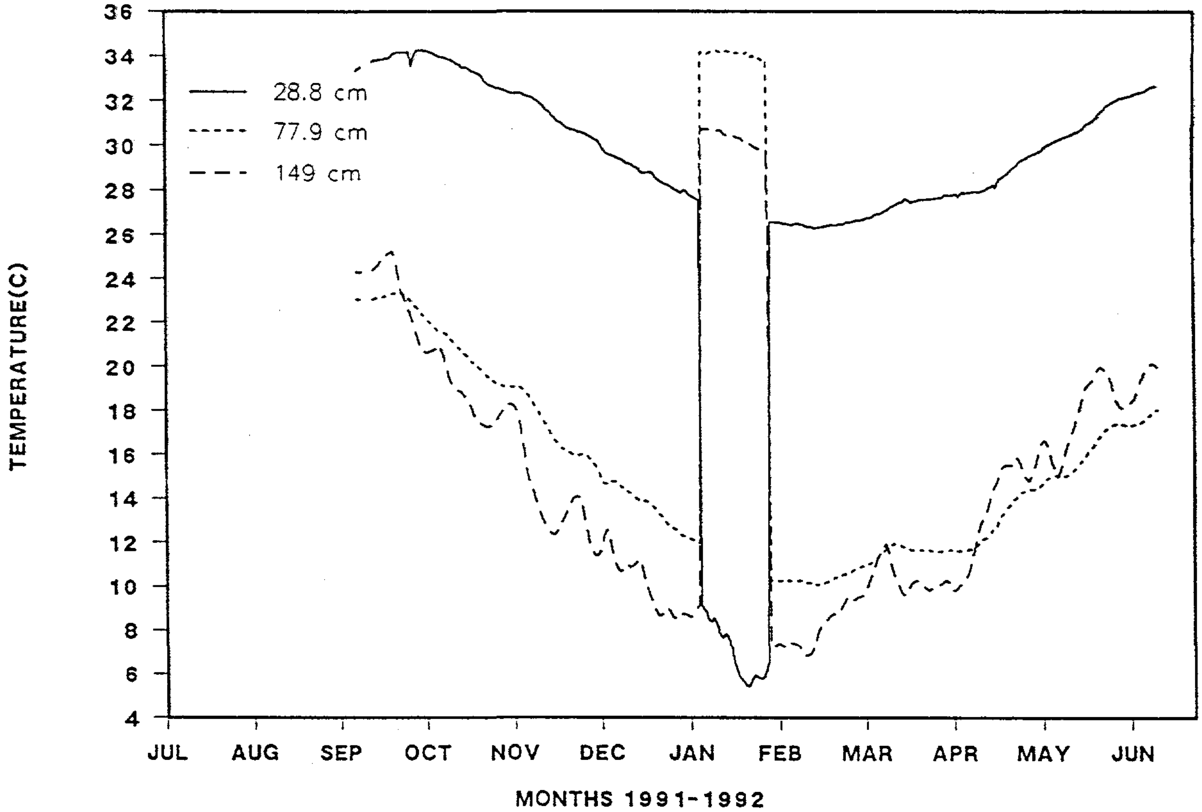

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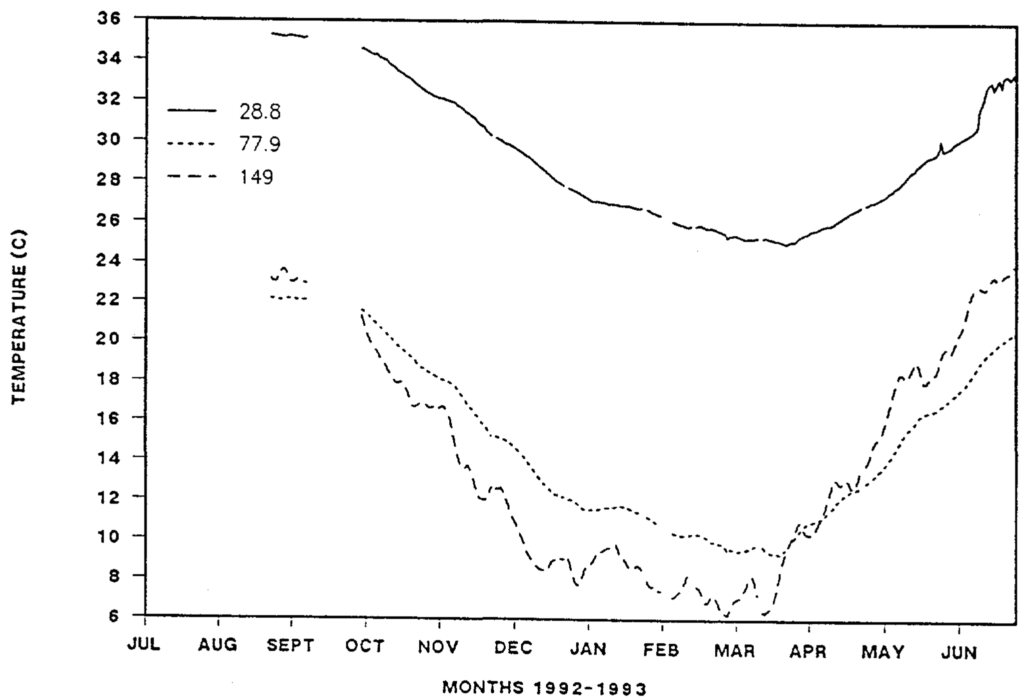

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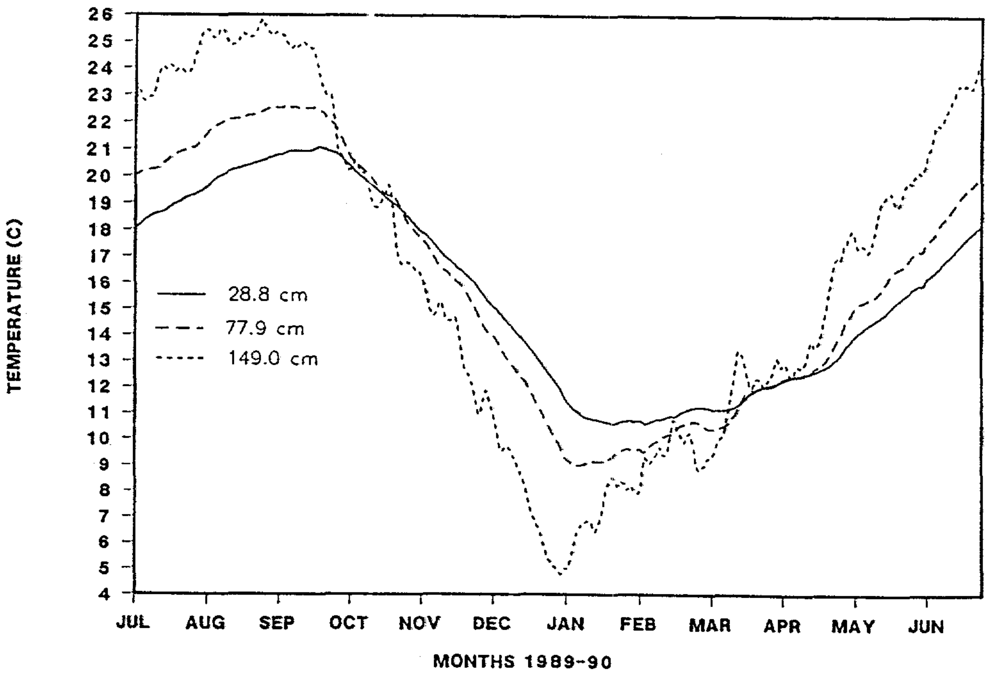

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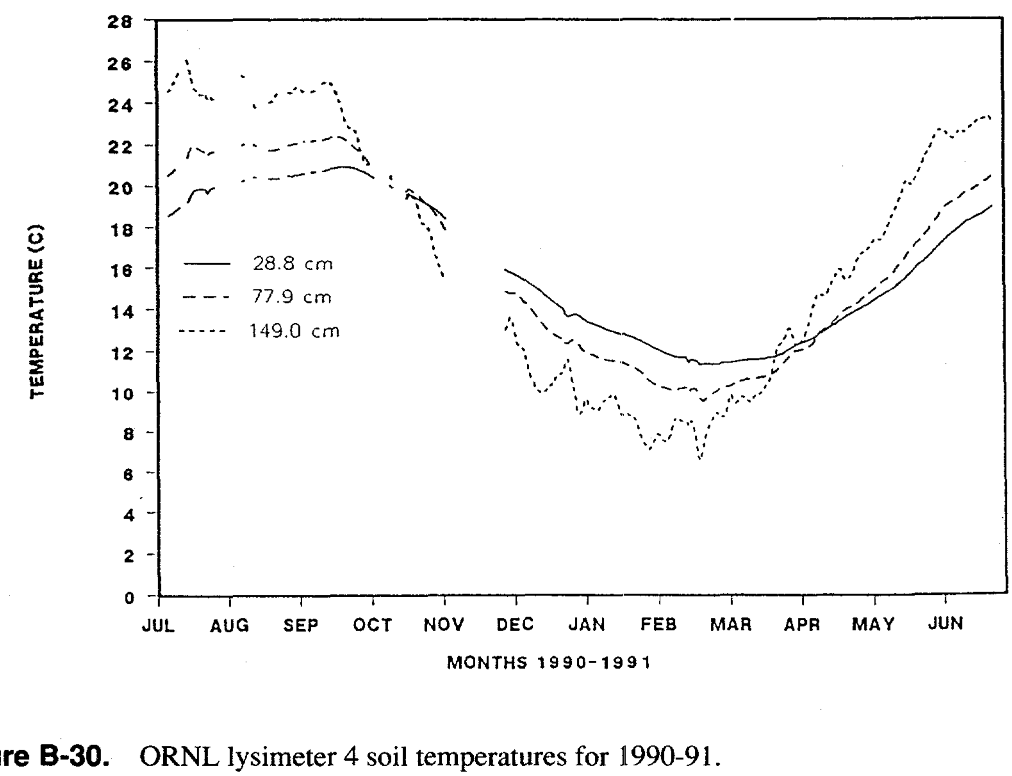

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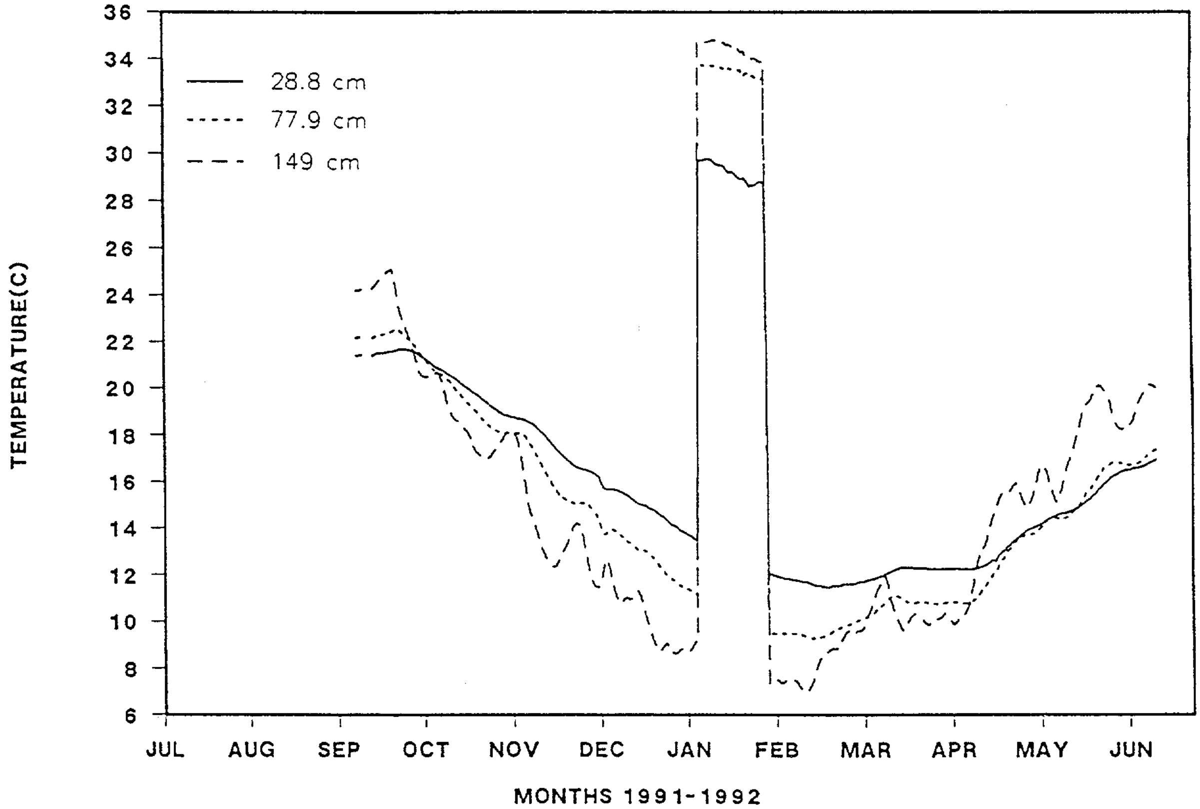

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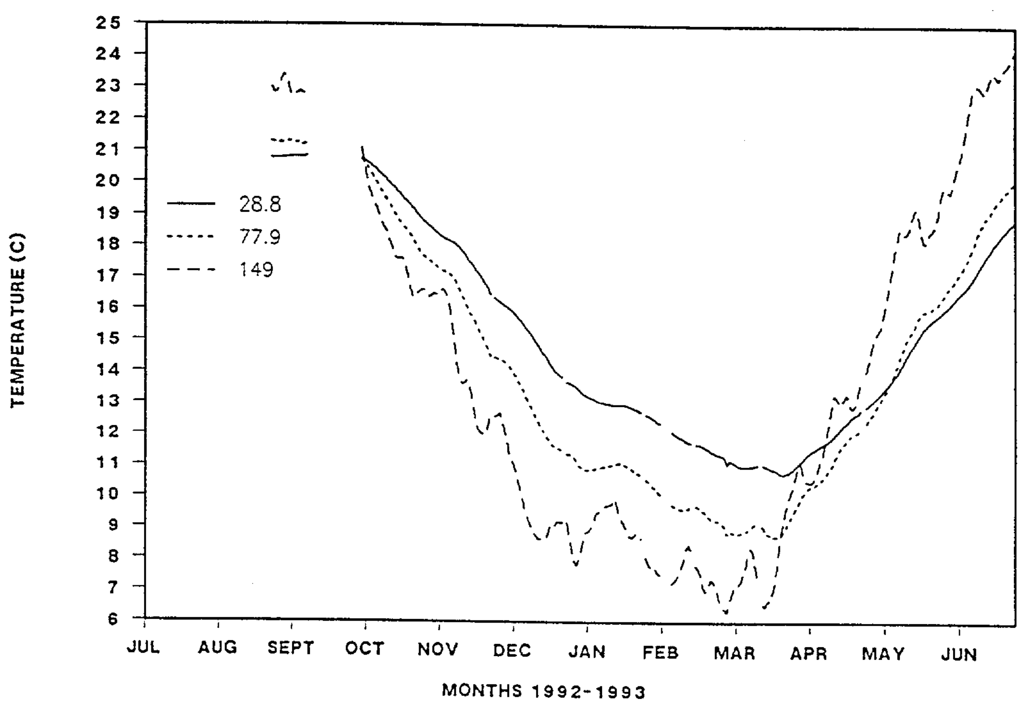

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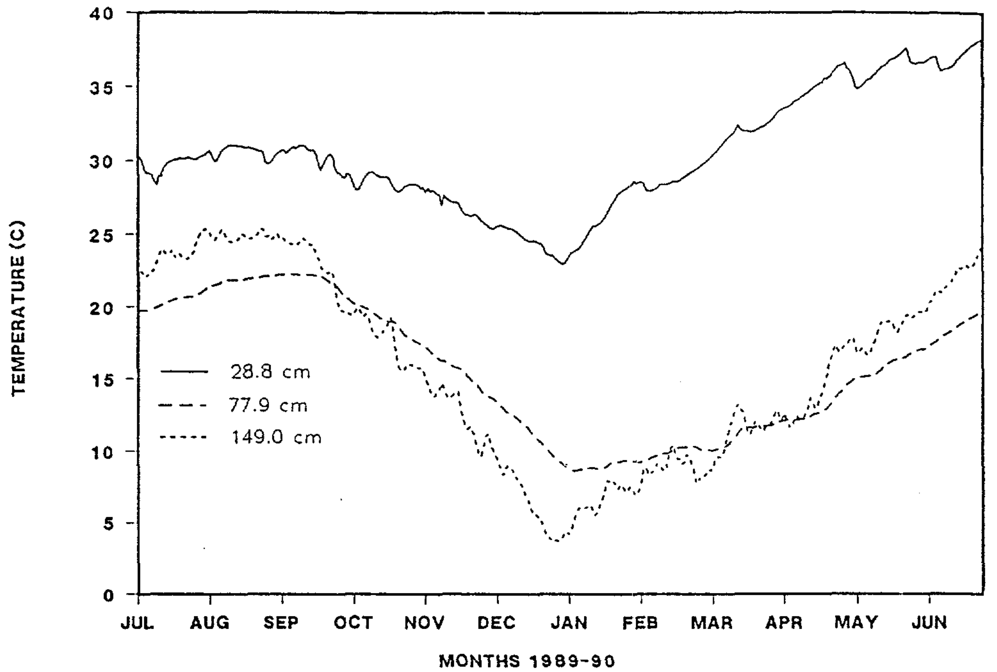

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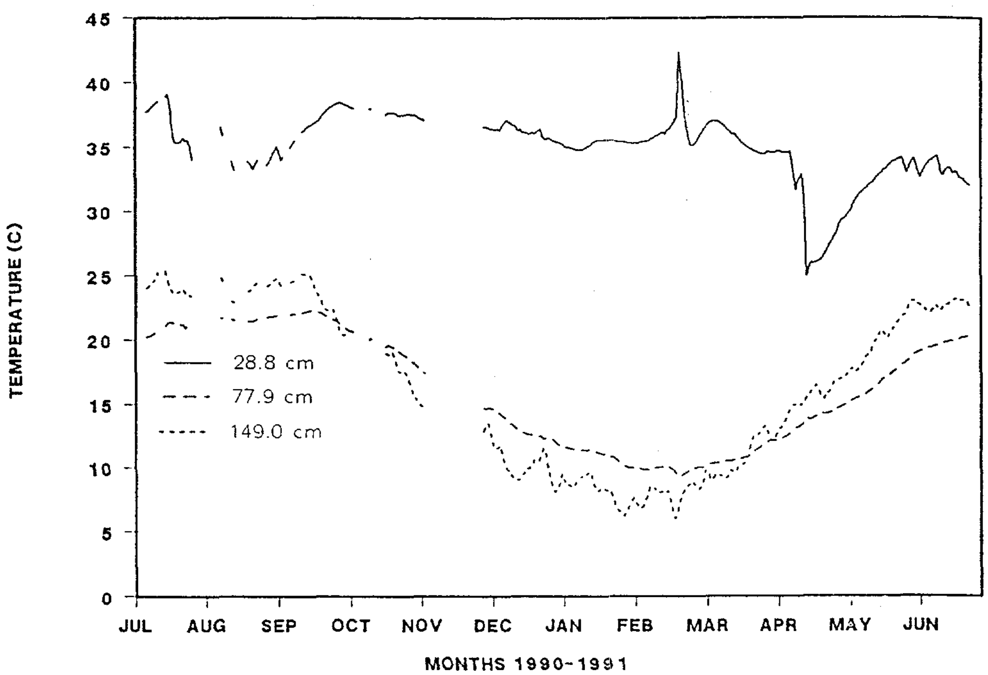

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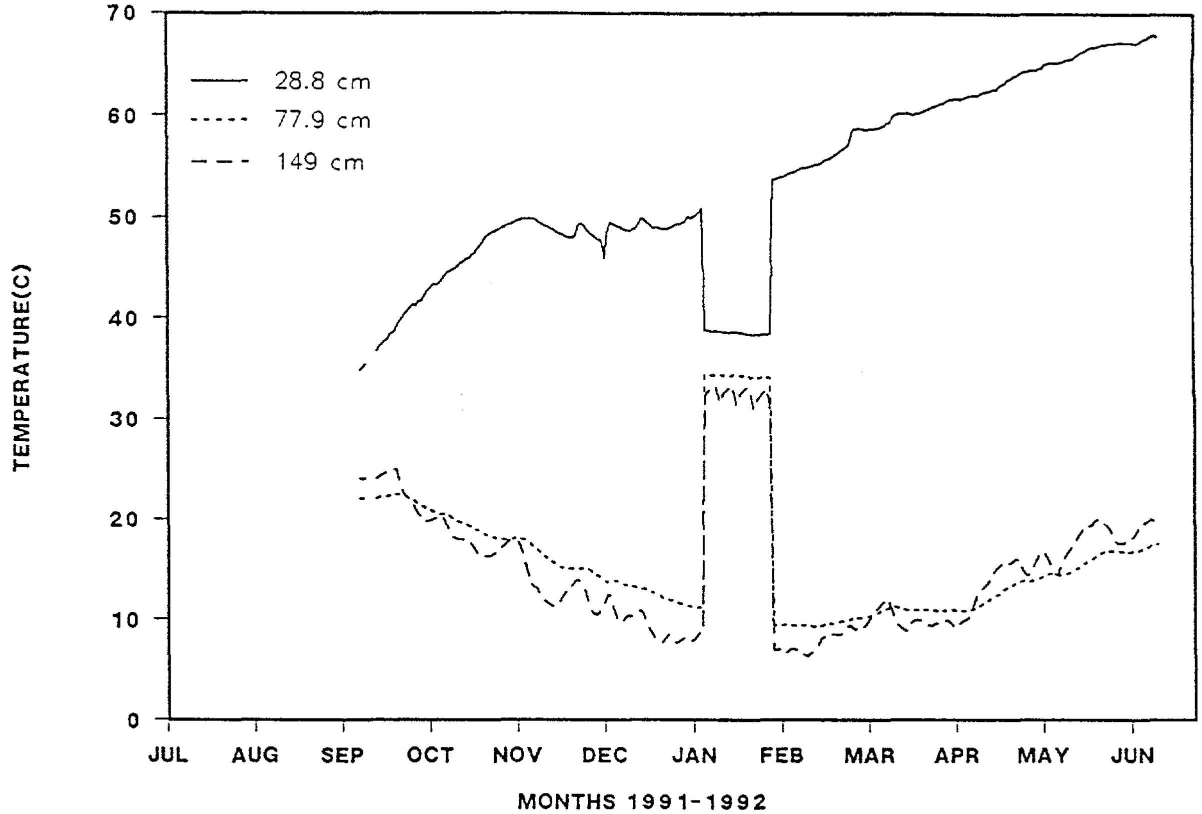

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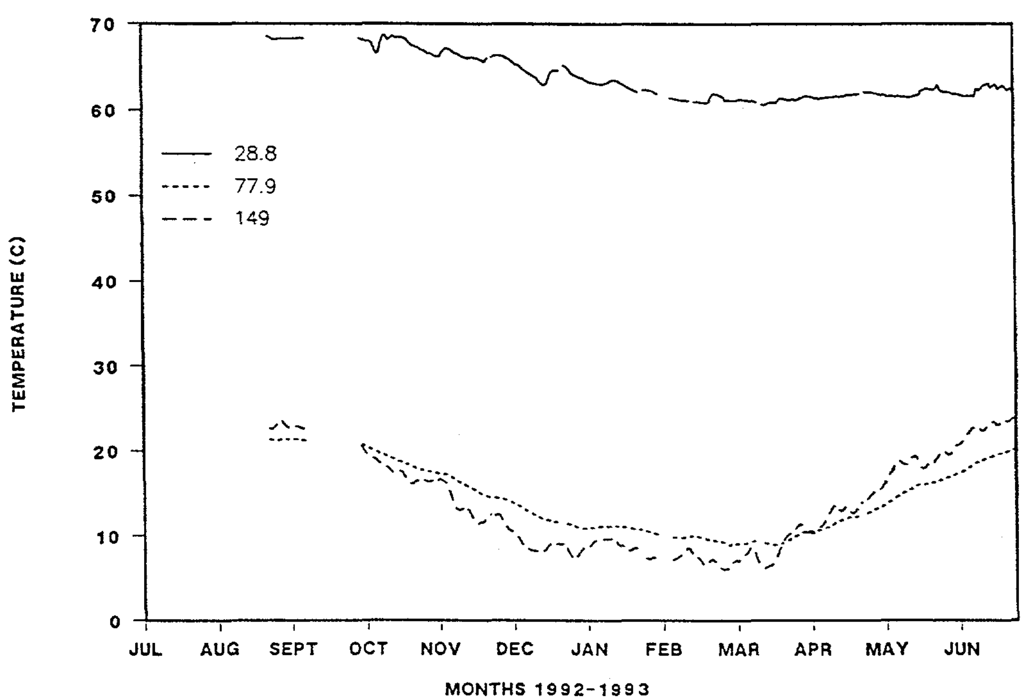

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Appendix C

Soil Moisture Data—Resistance Probes

Appendix C
Soil Moisture Data—Resistance Probes

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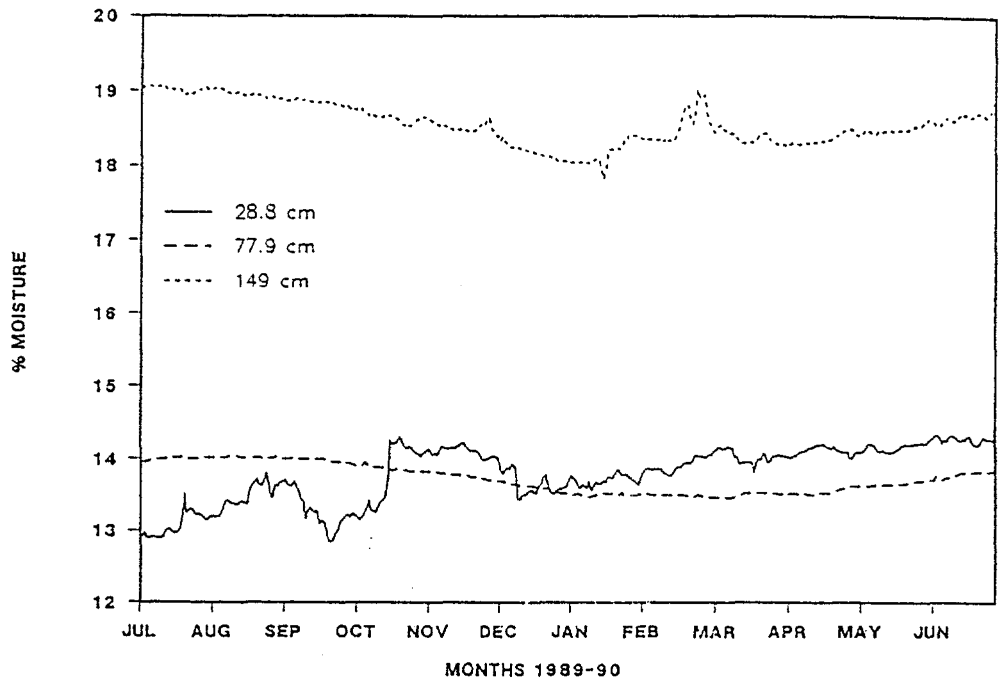

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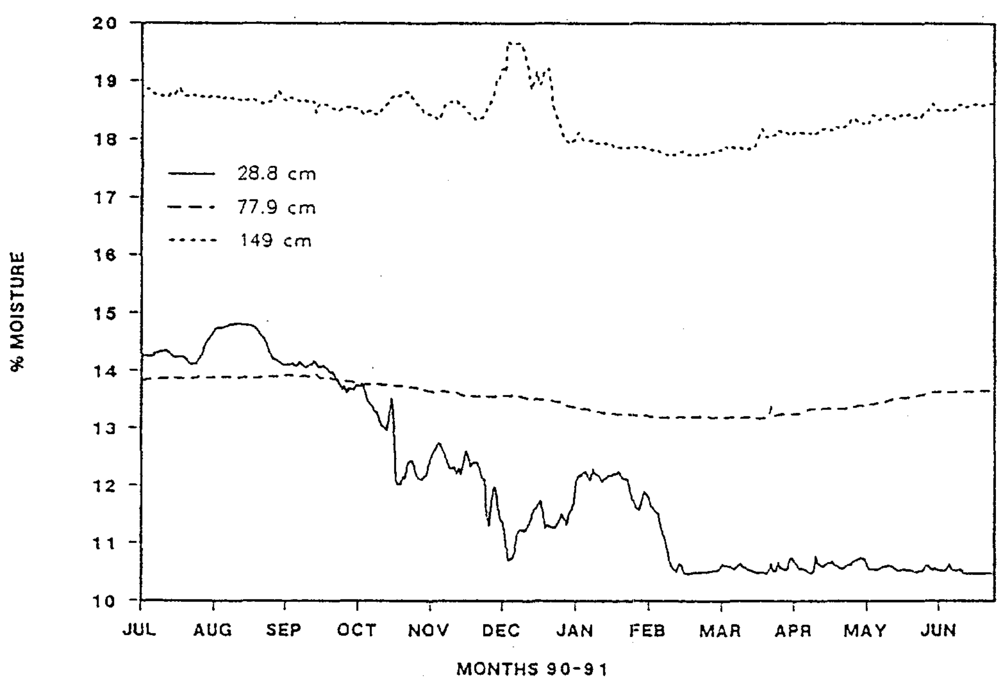

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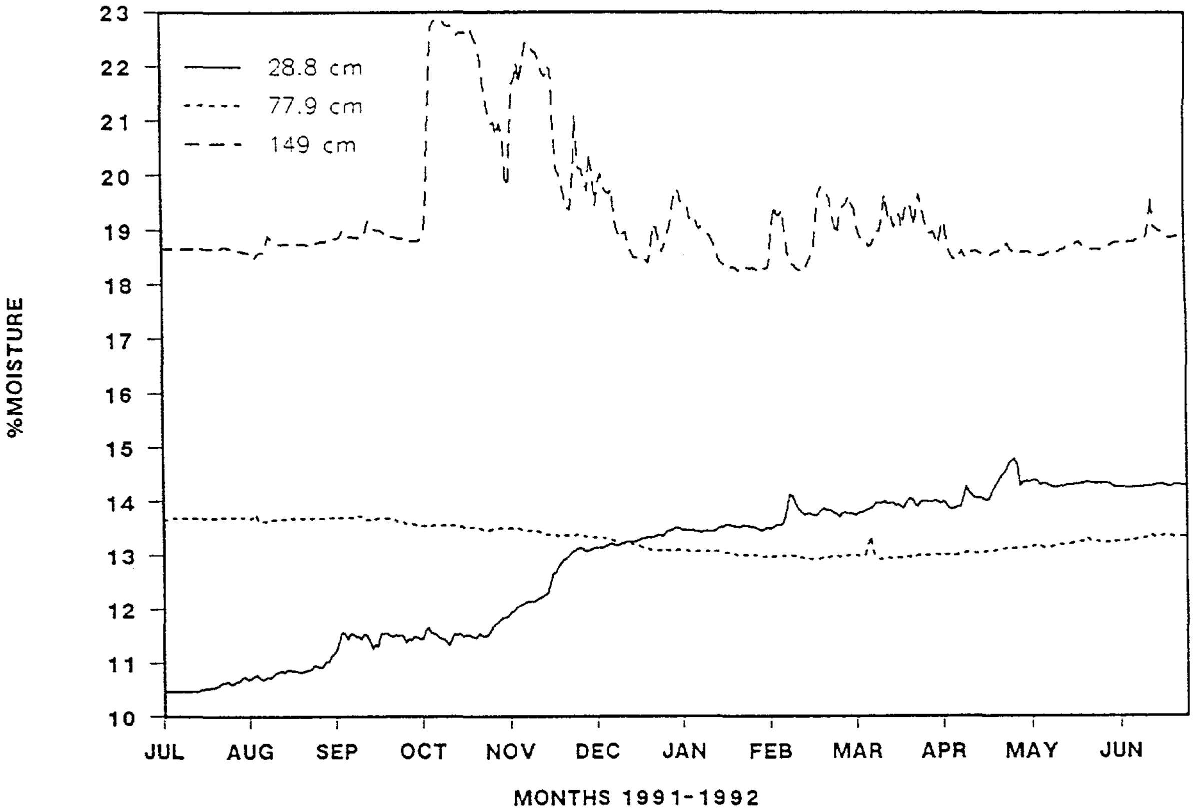

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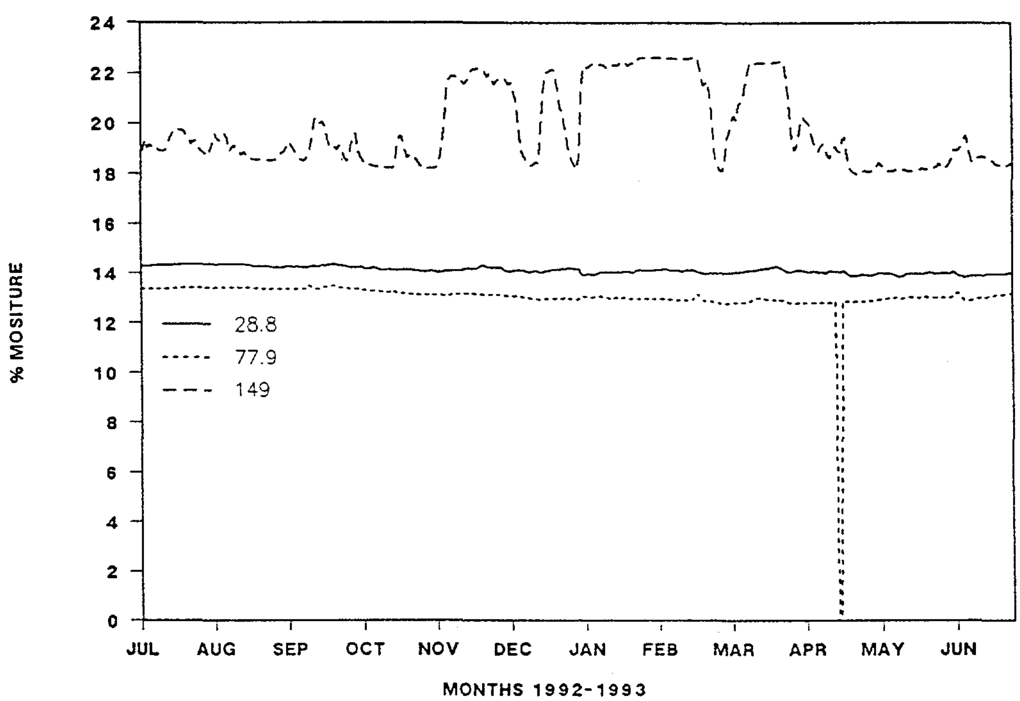

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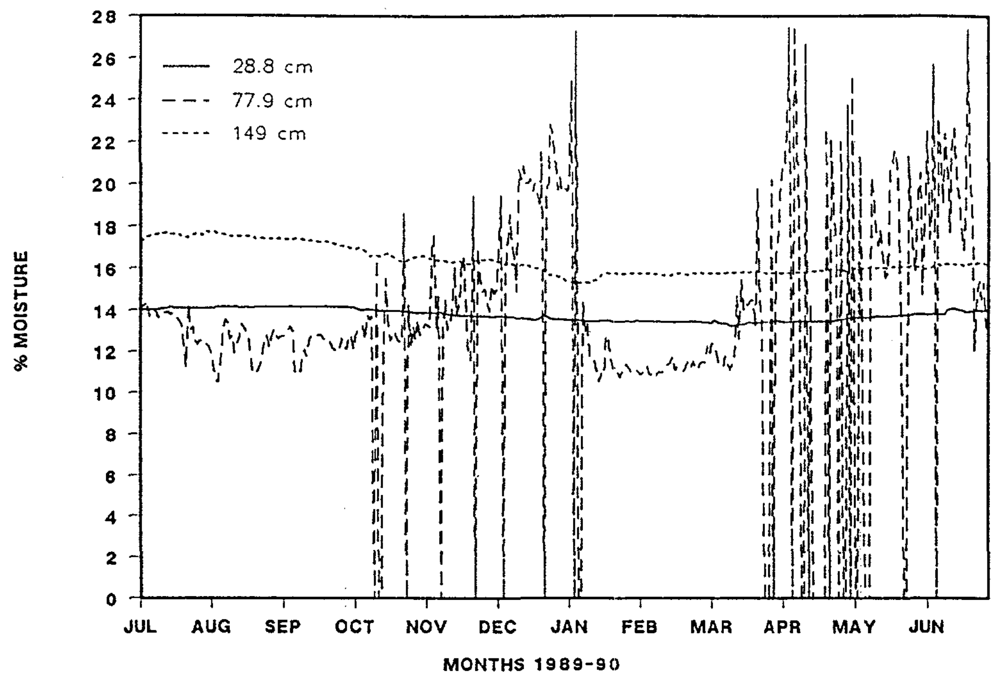

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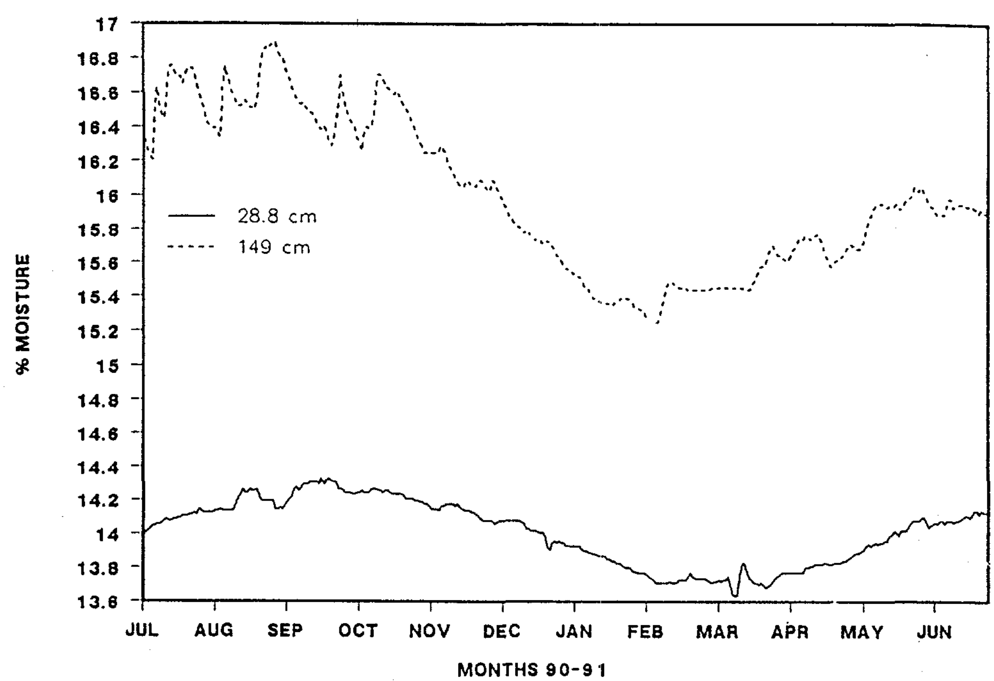

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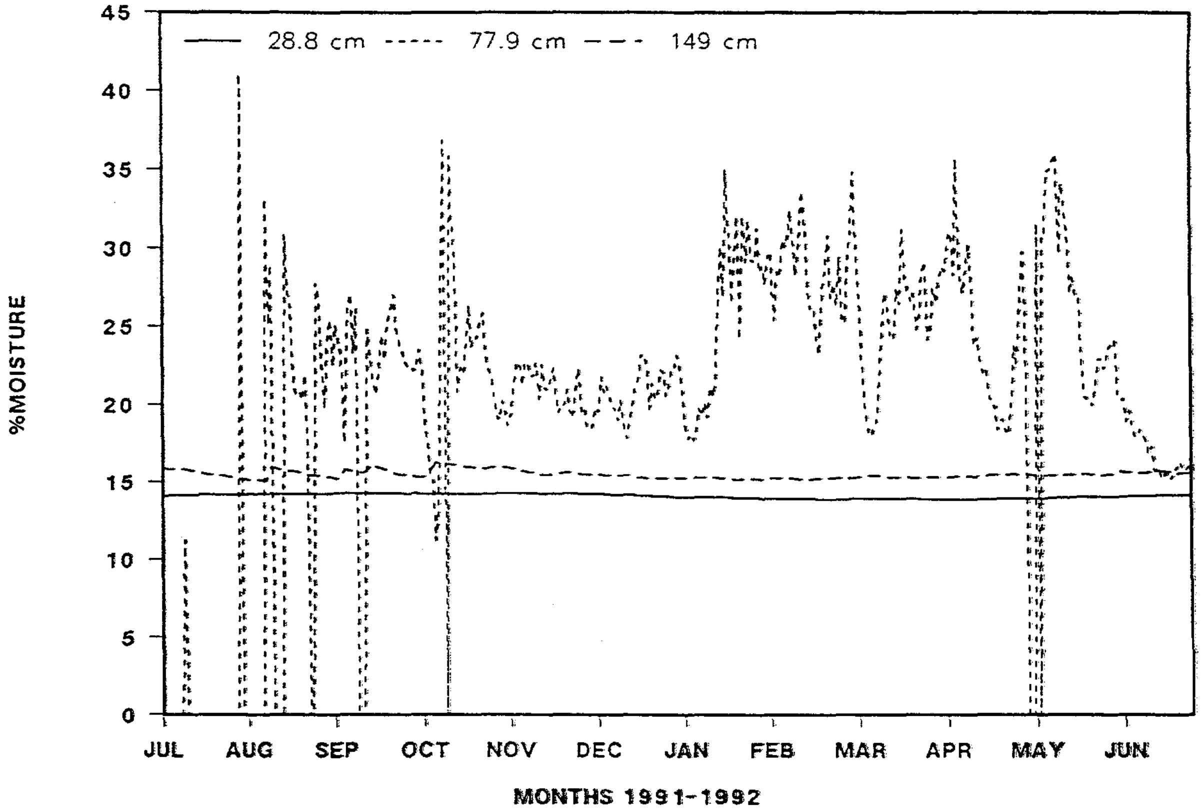

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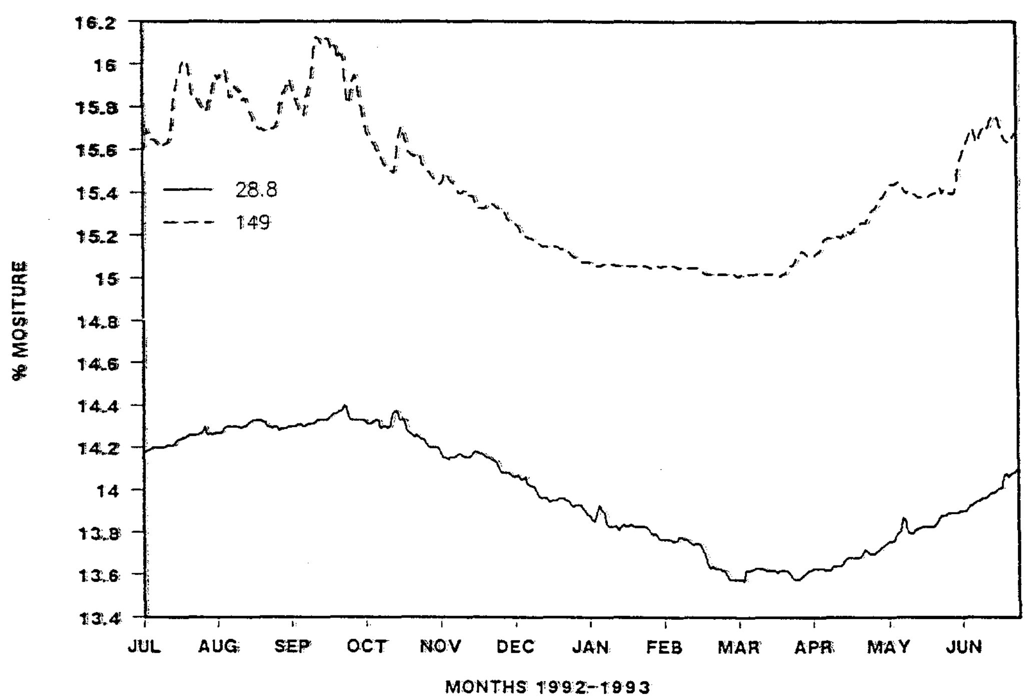

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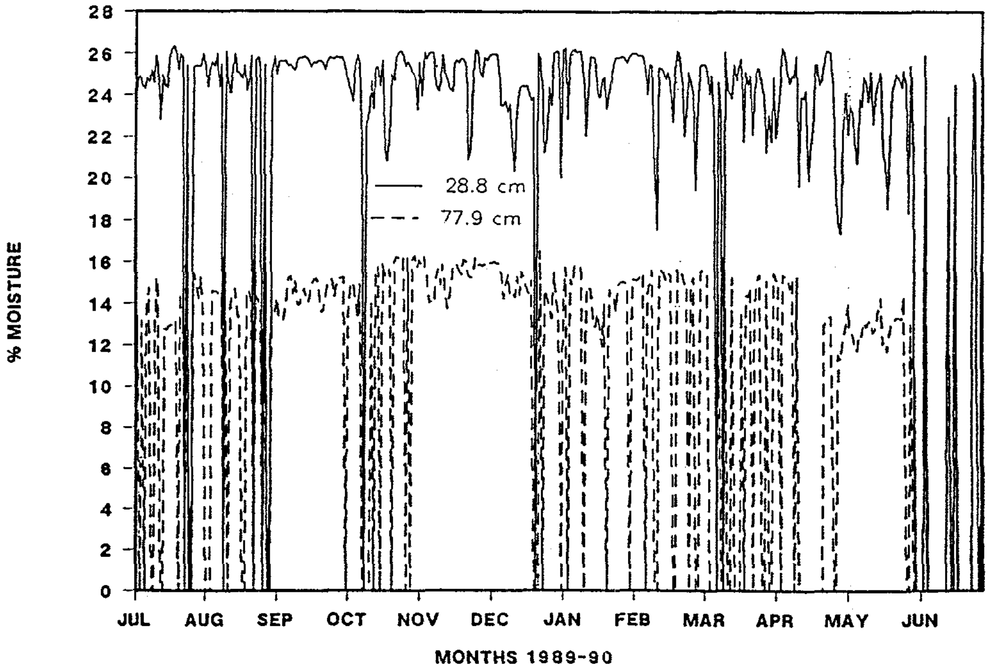

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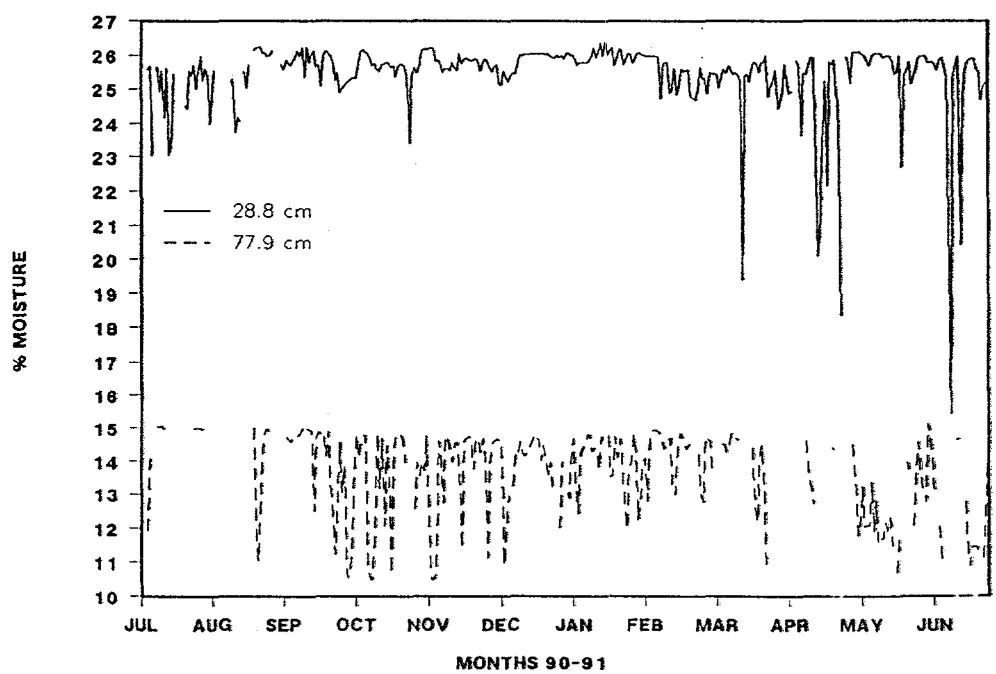

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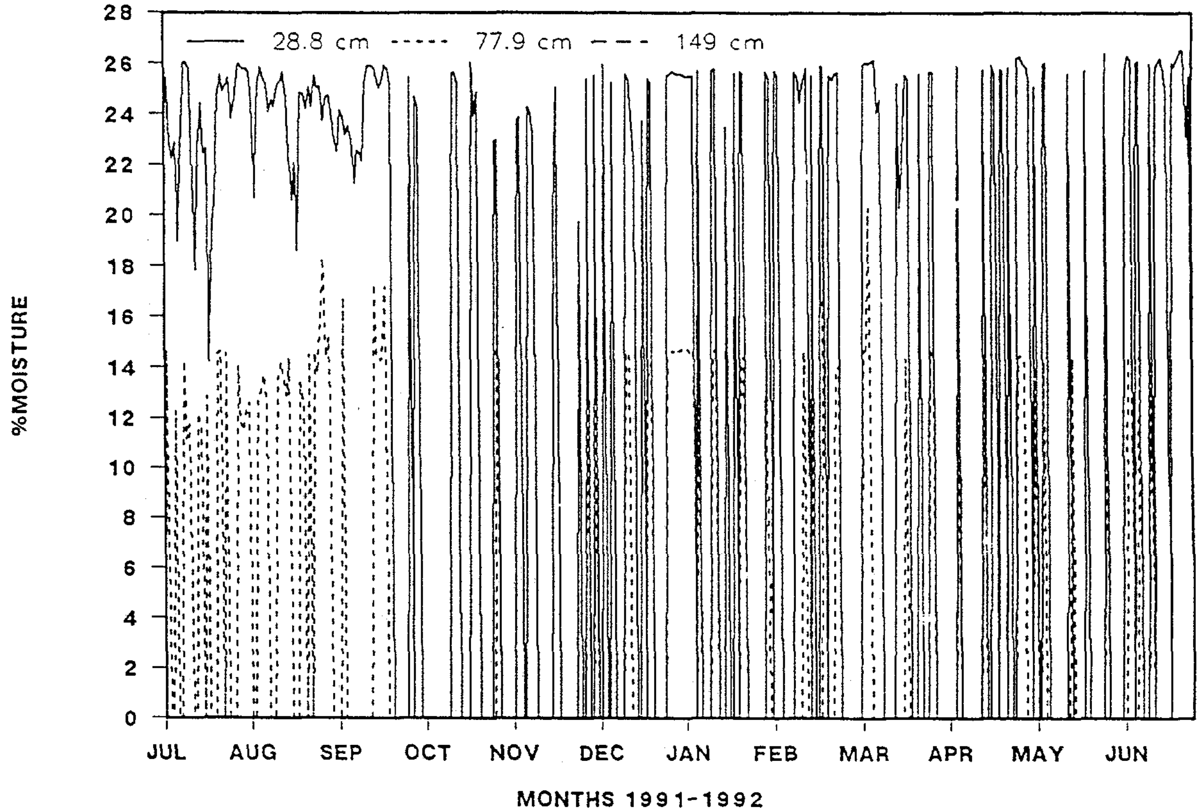

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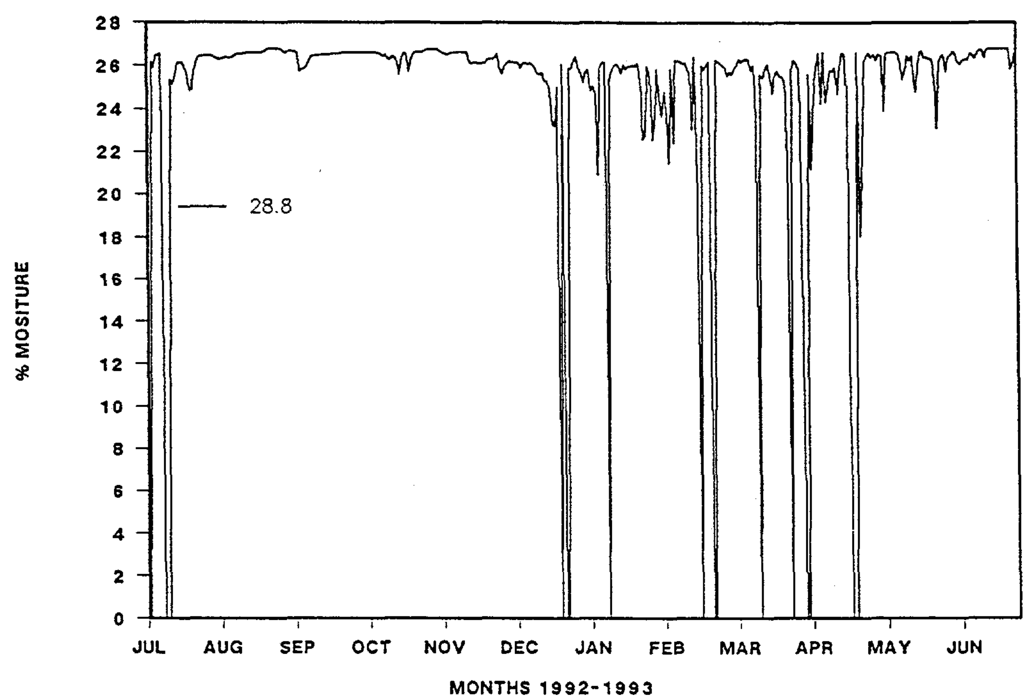

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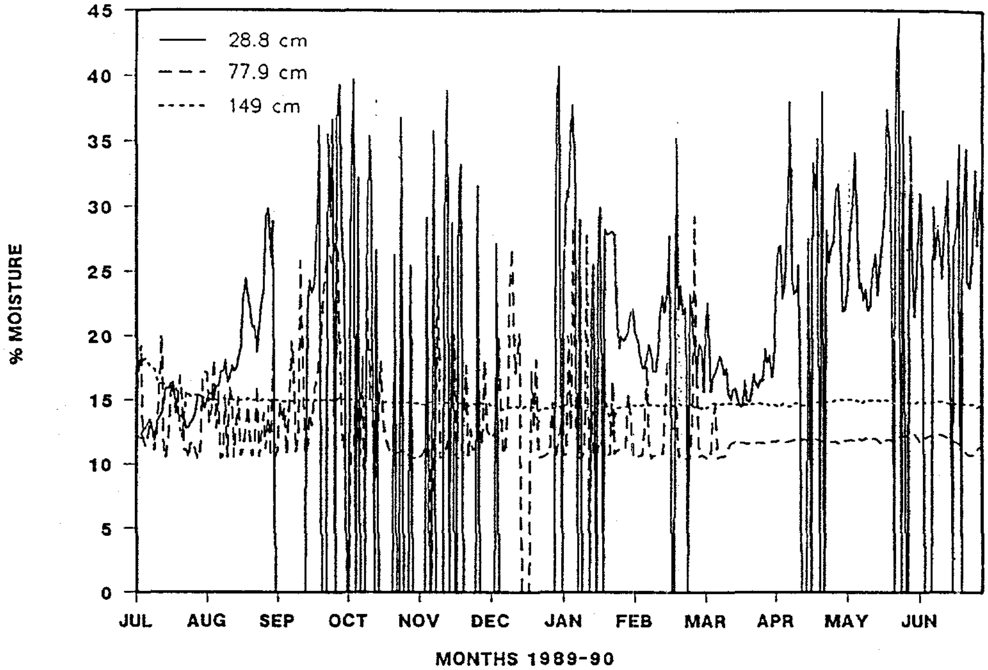

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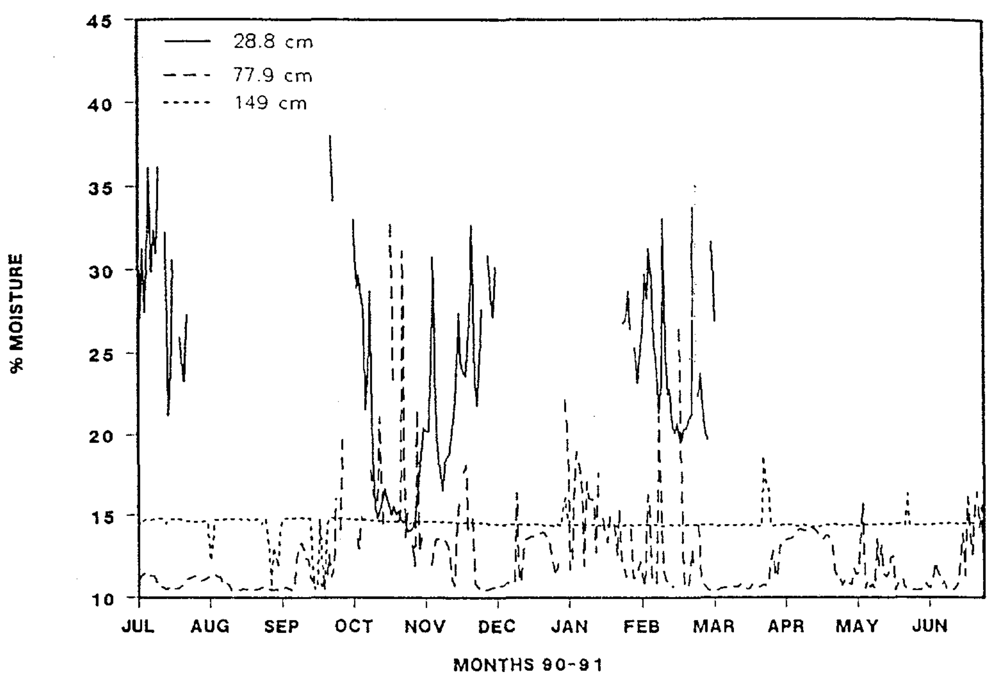

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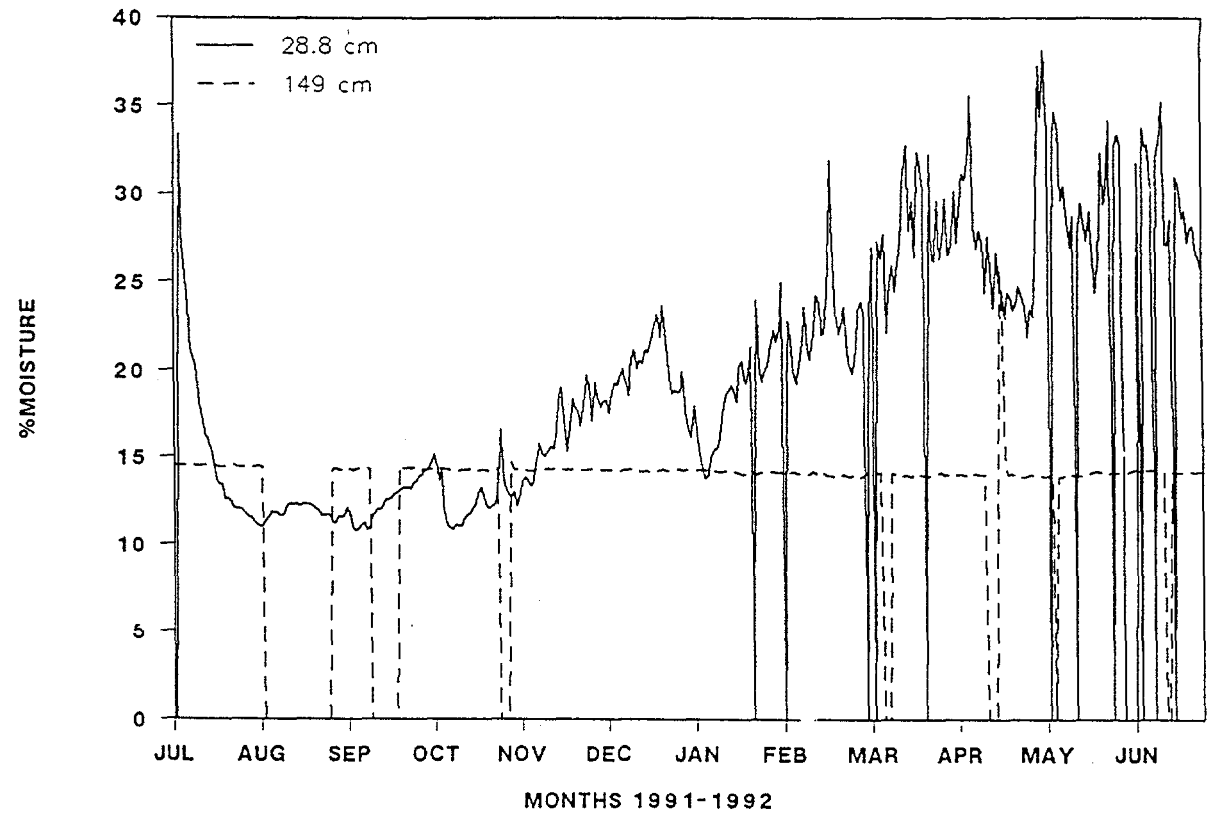

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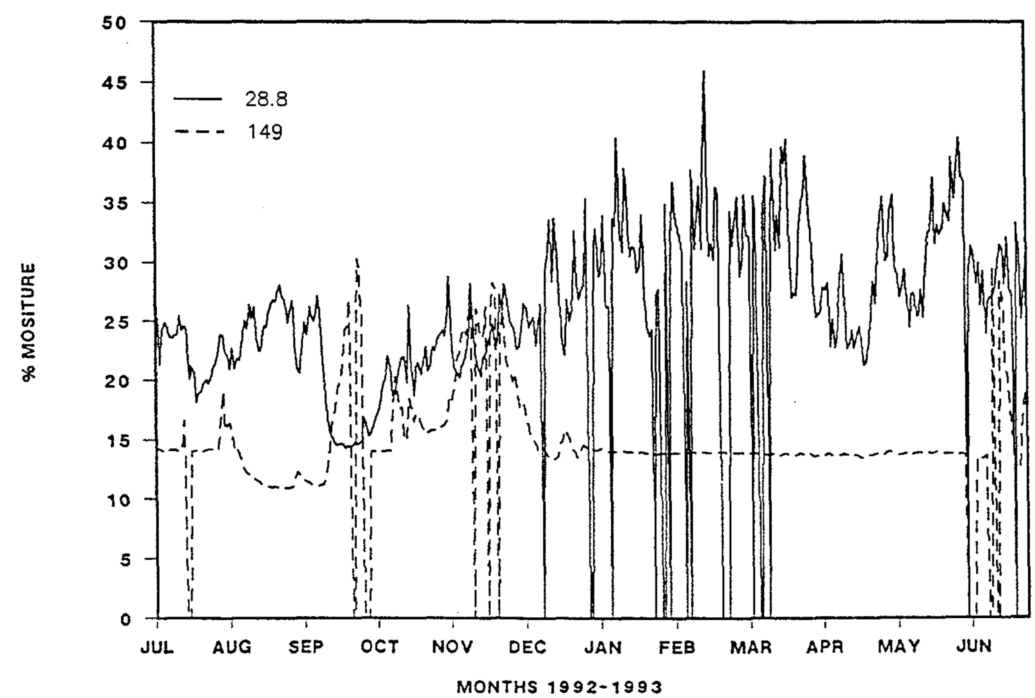

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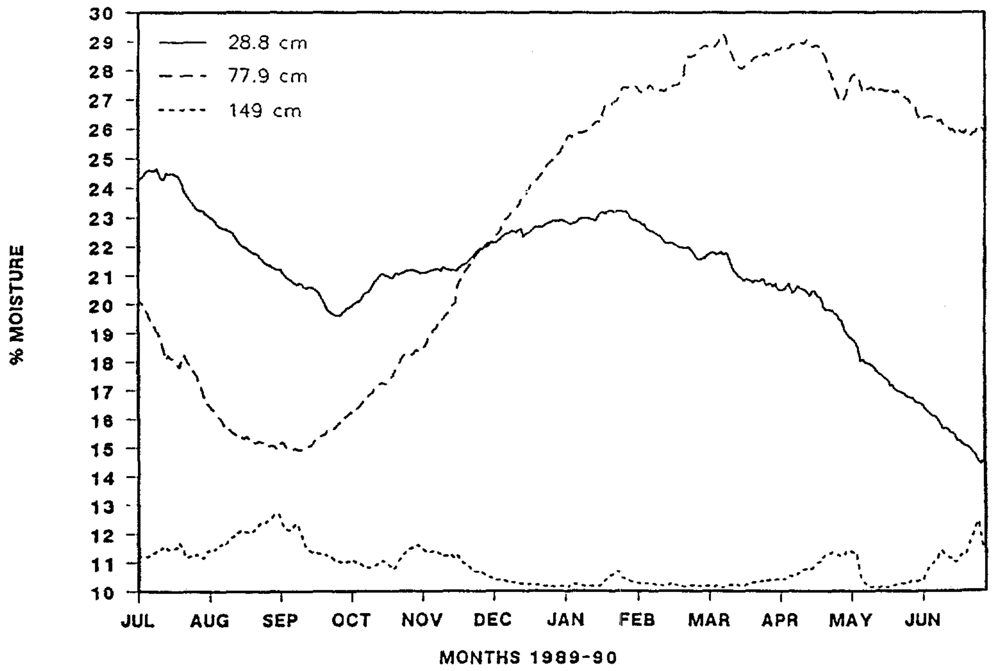

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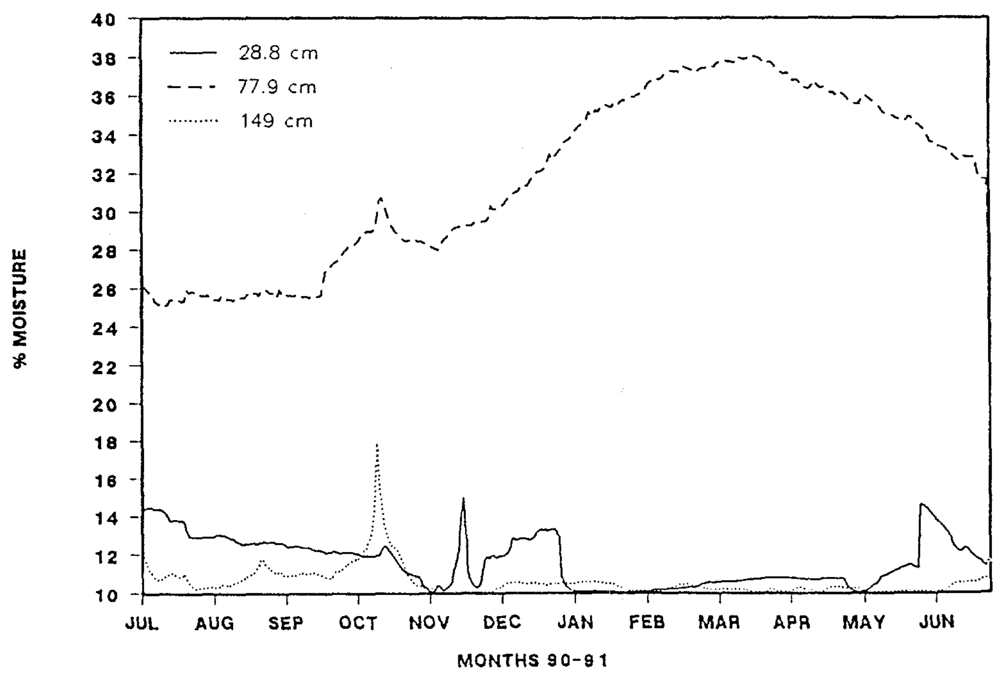

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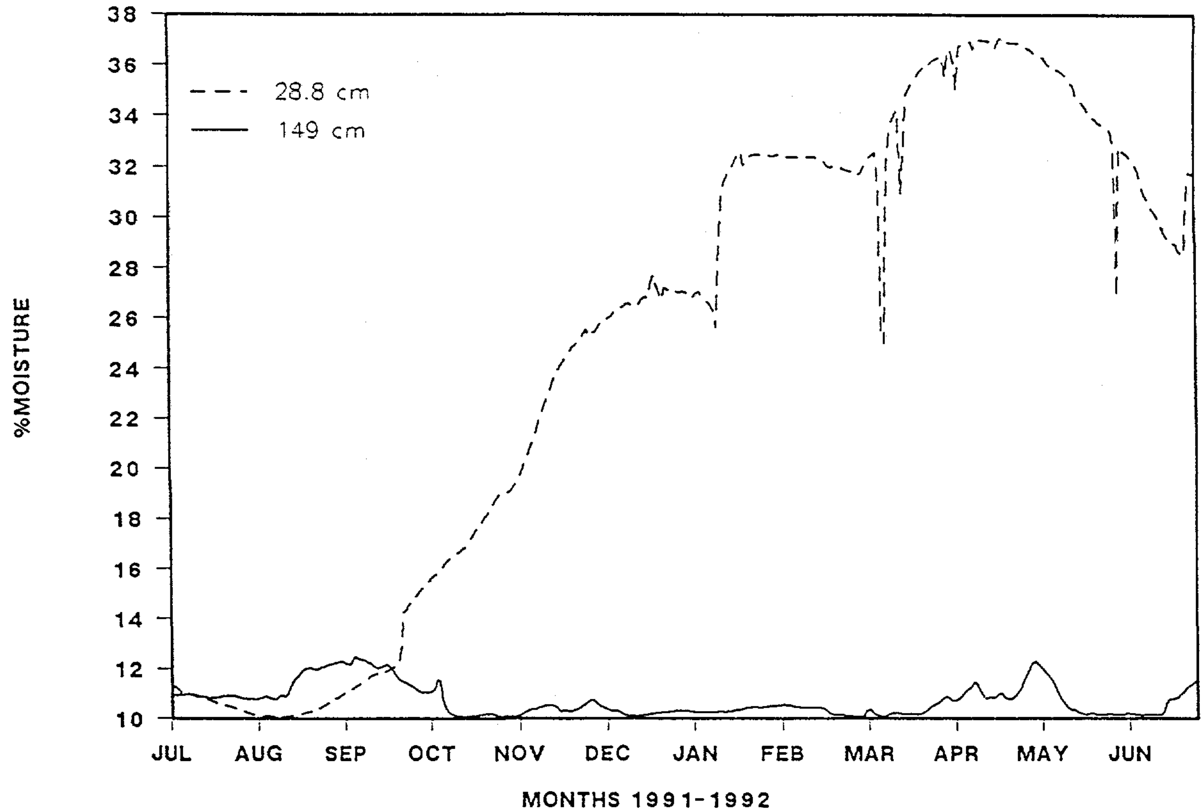

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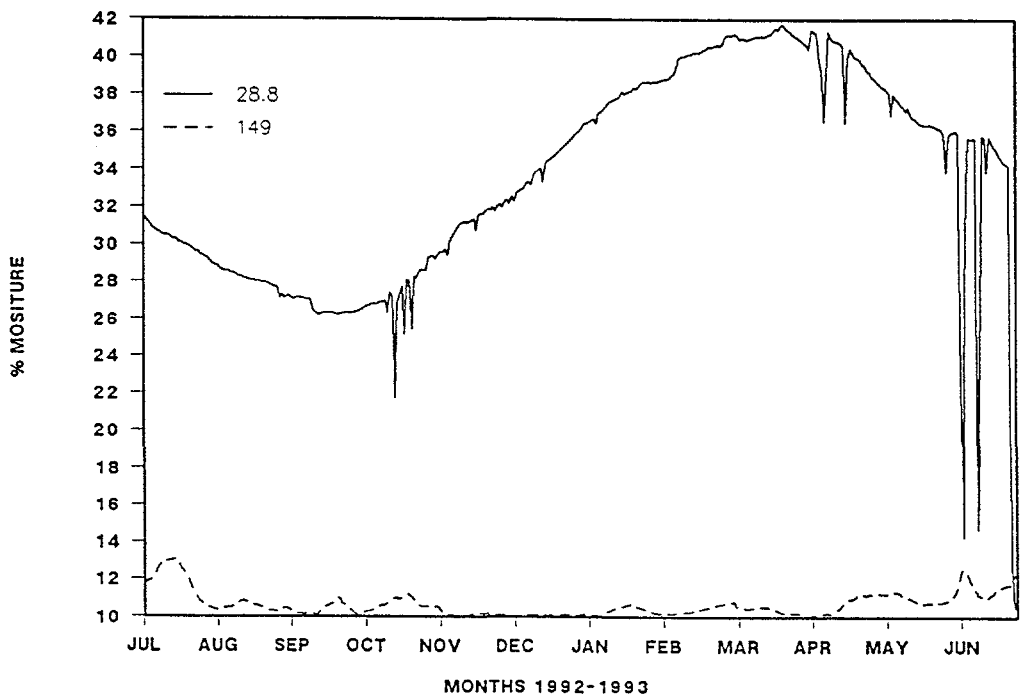

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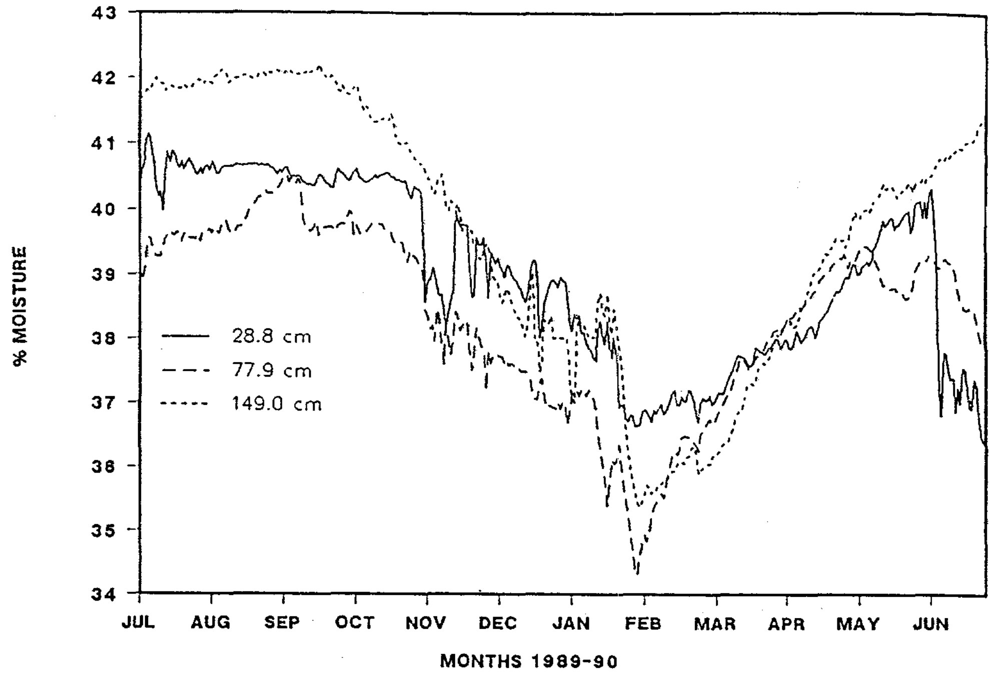

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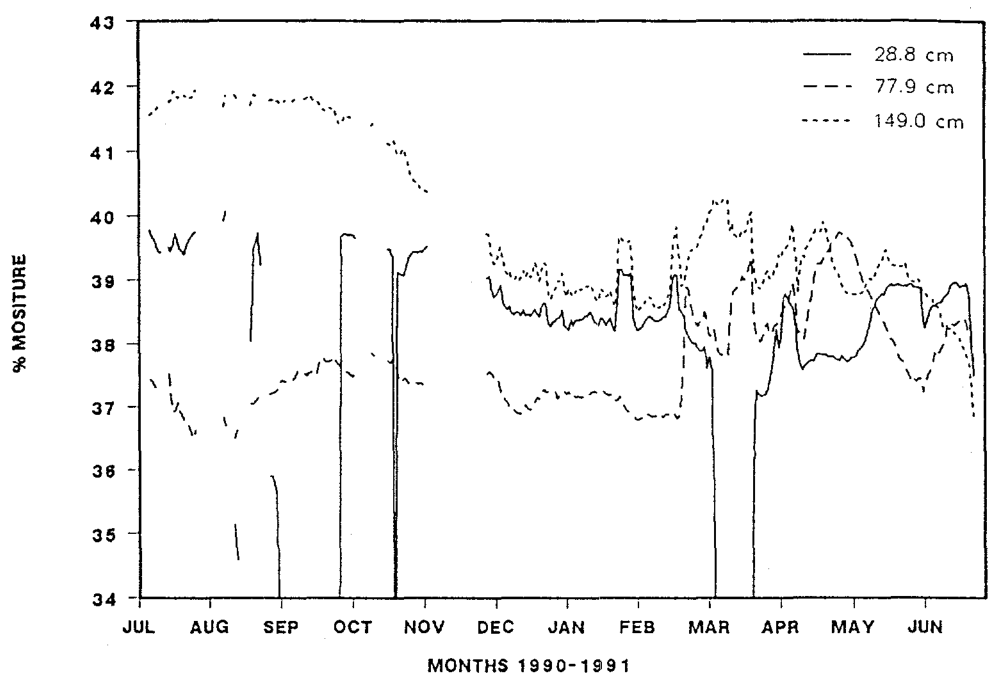

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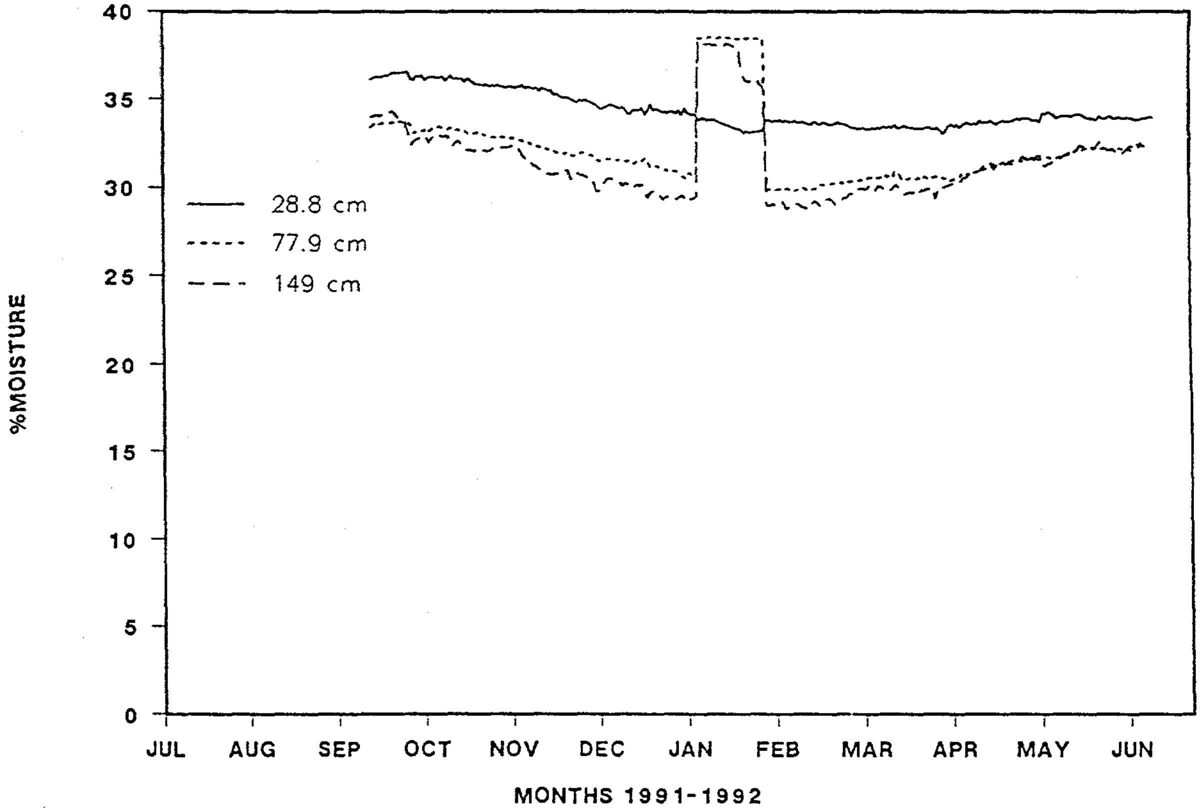

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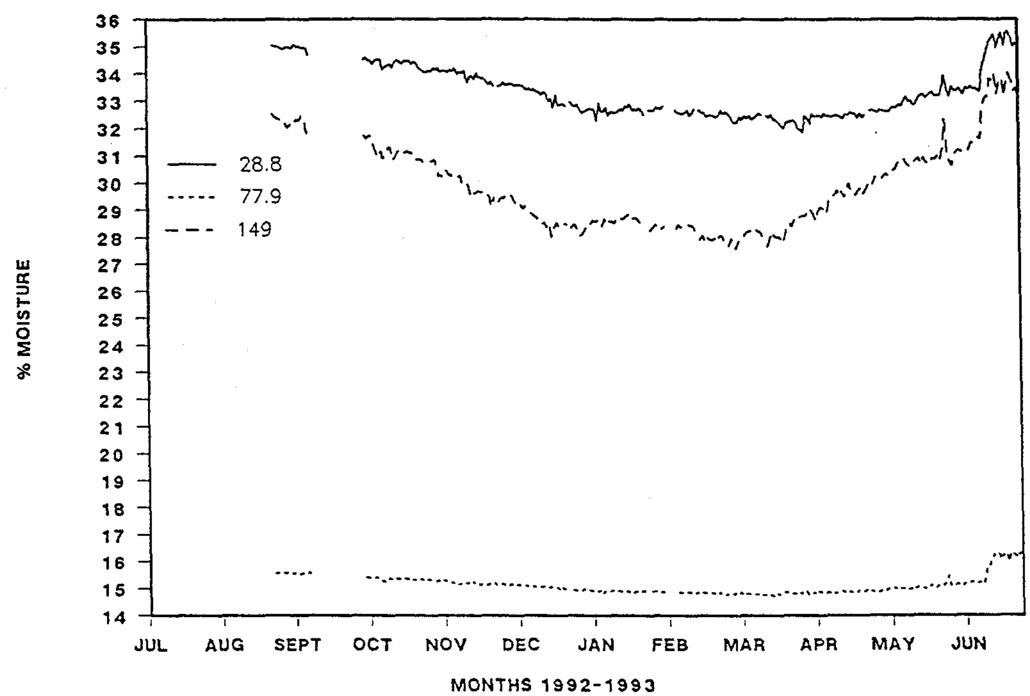

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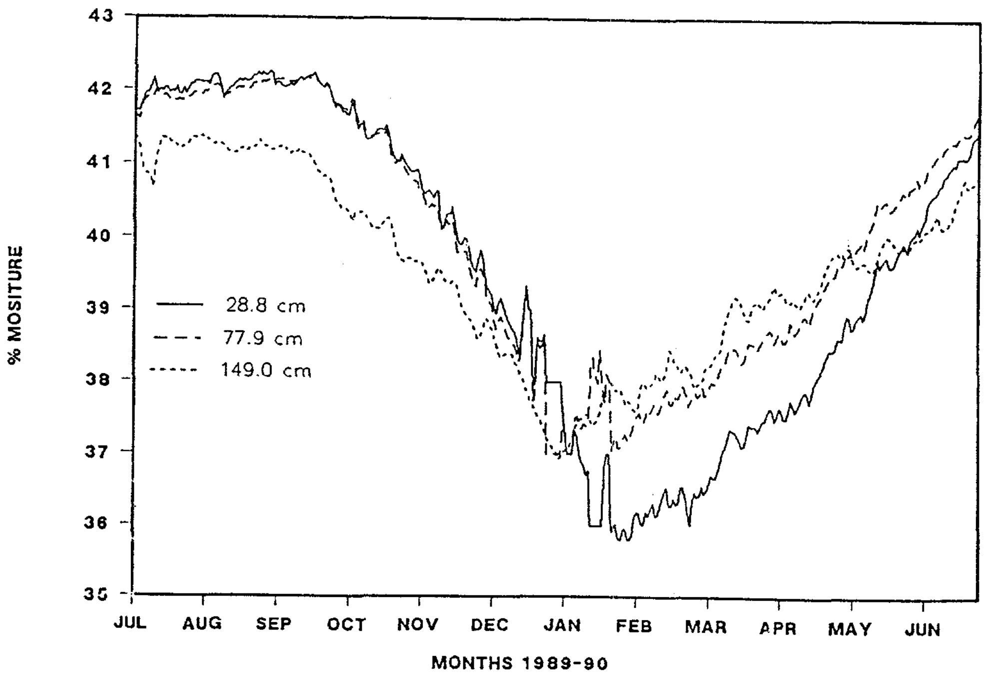

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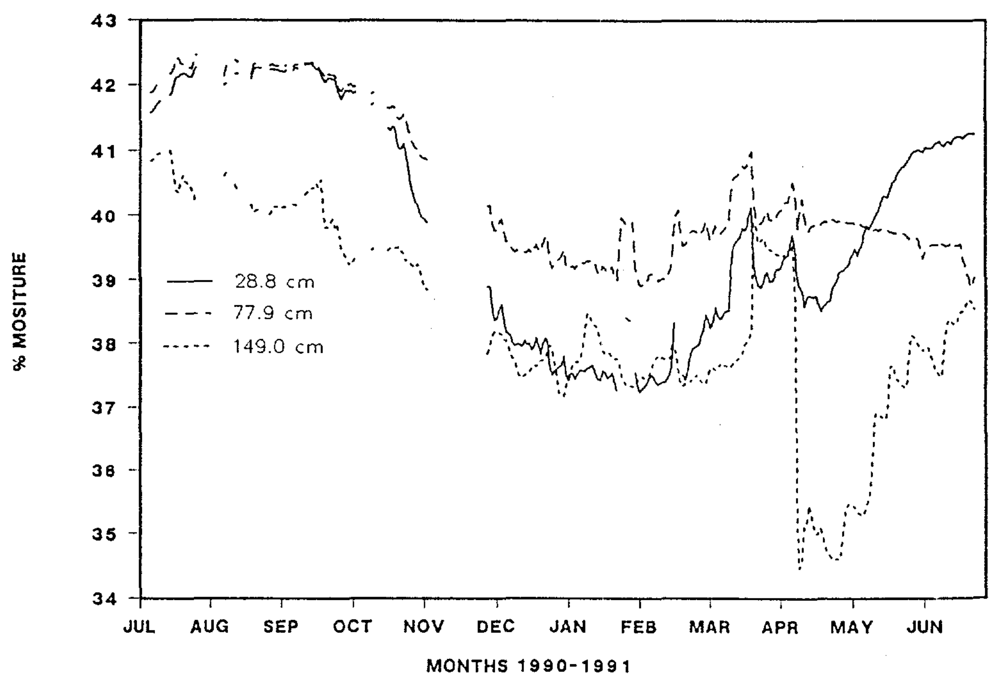

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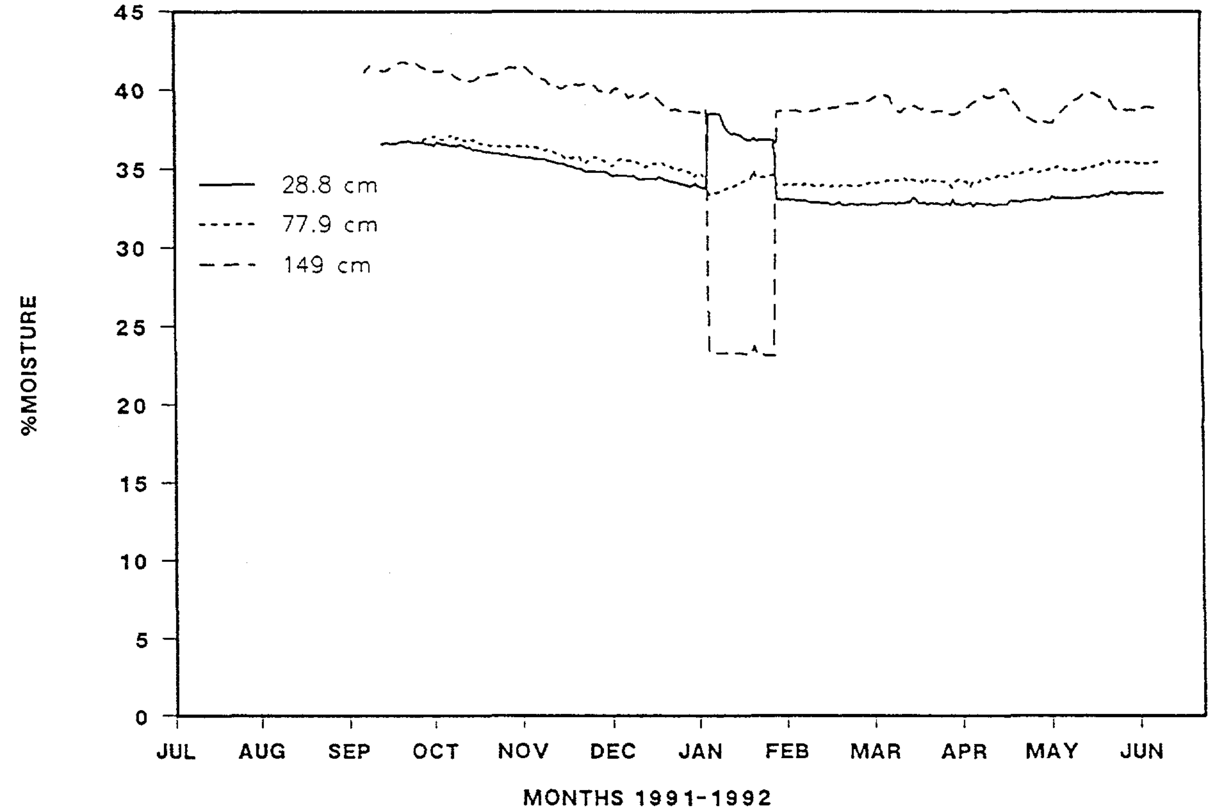

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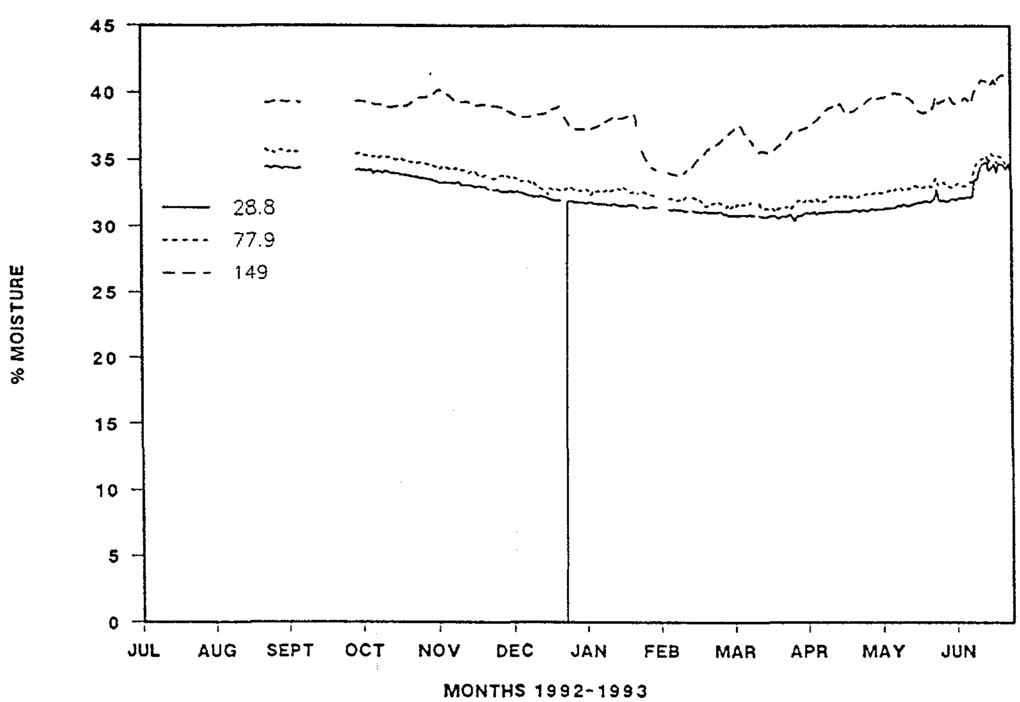

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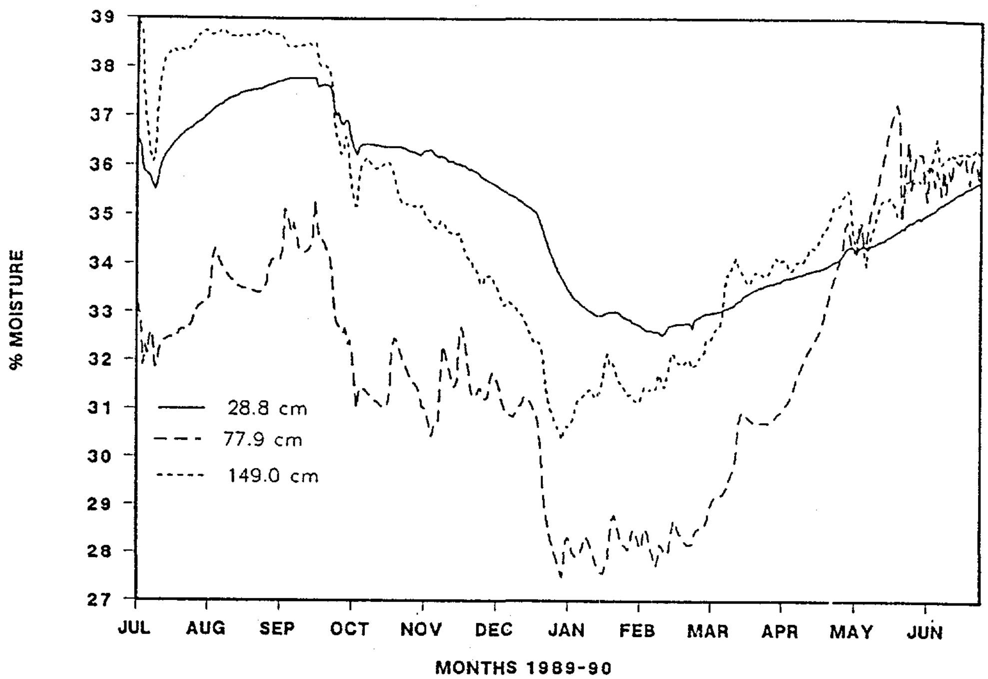

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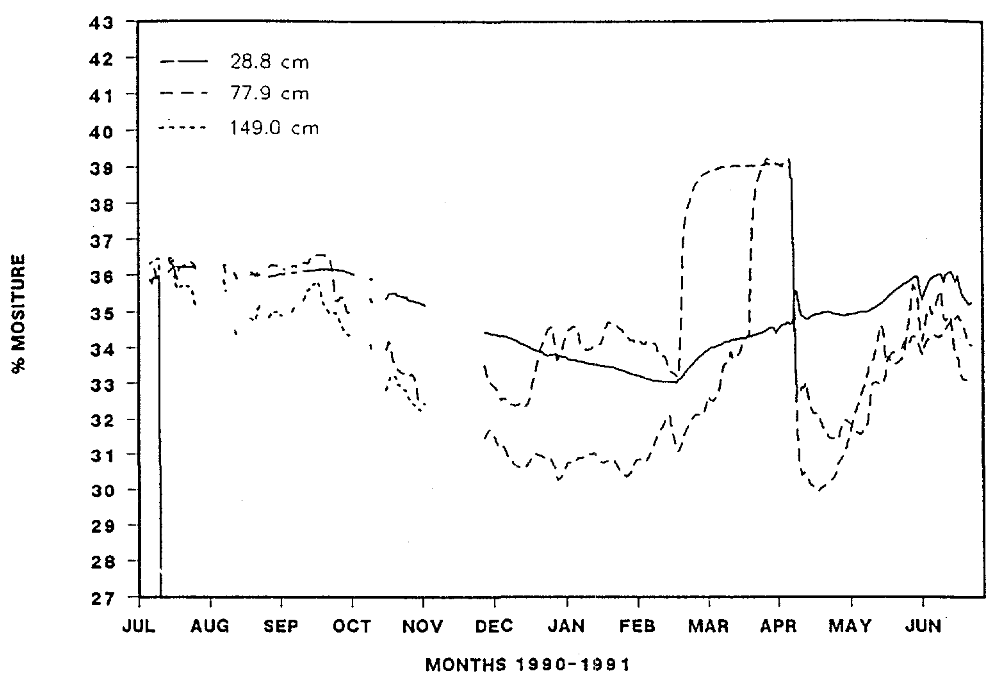

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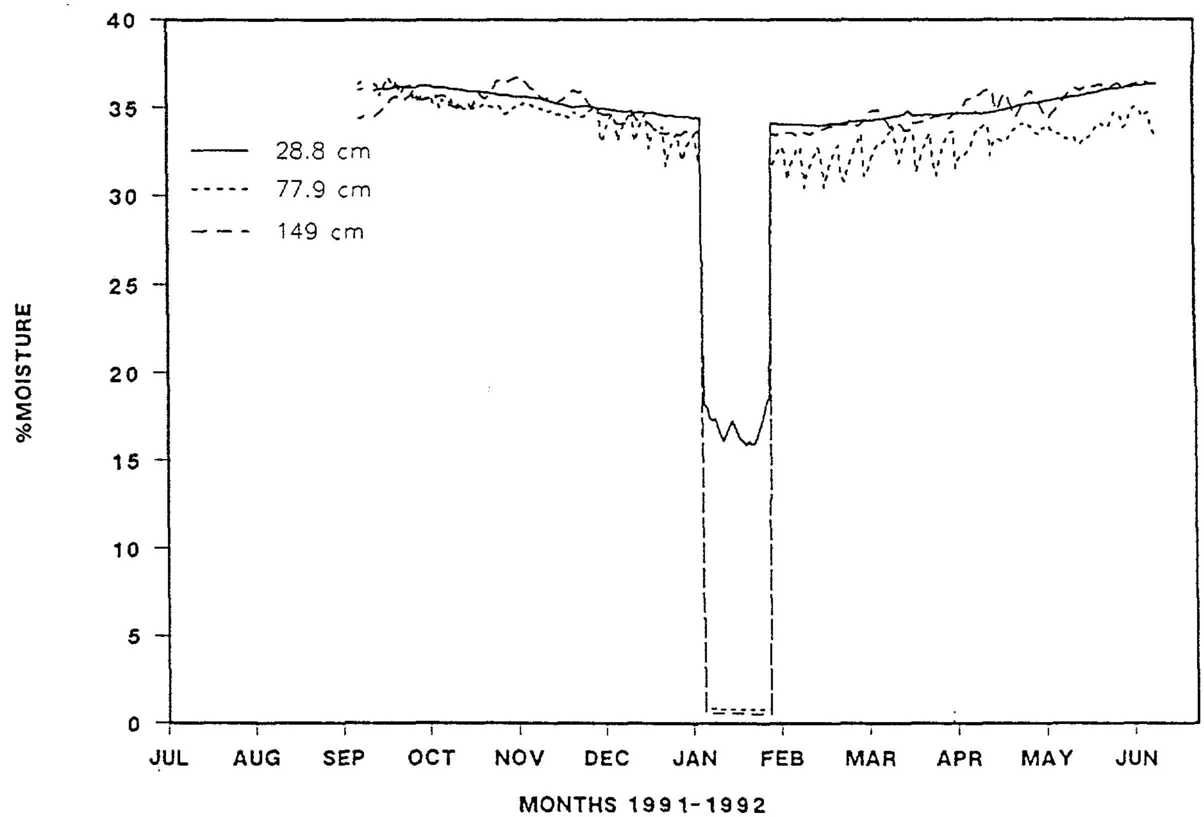

Figure C-31. ORNL lysimeter 3 soil moisture for 1991-92.

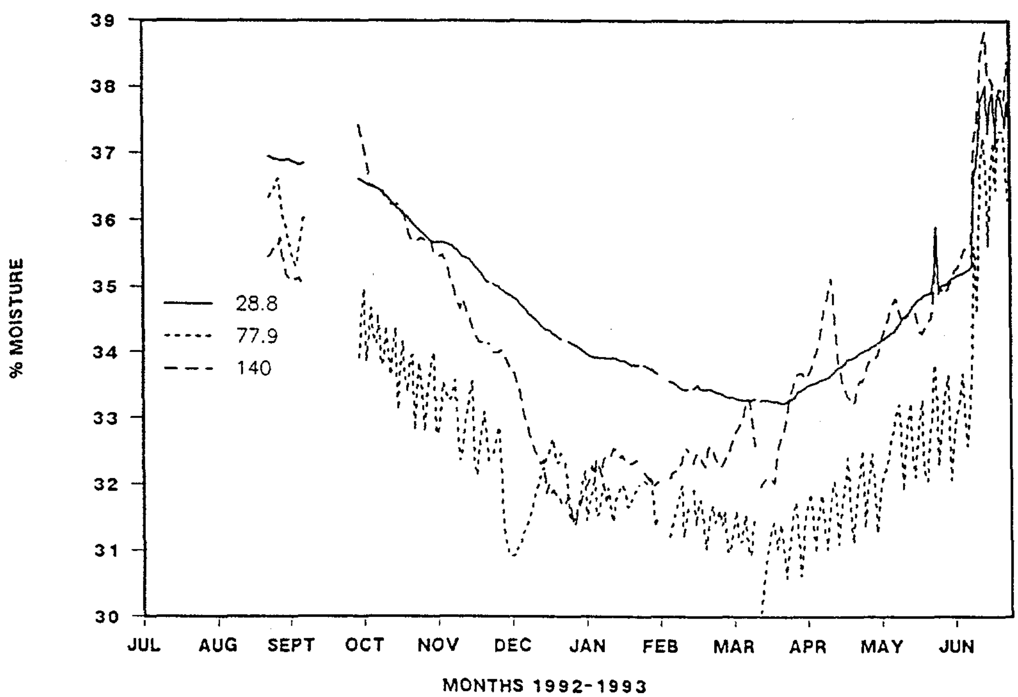

Figure C-32. ORNL lysimeter 3 soil moisture for 1992-93.

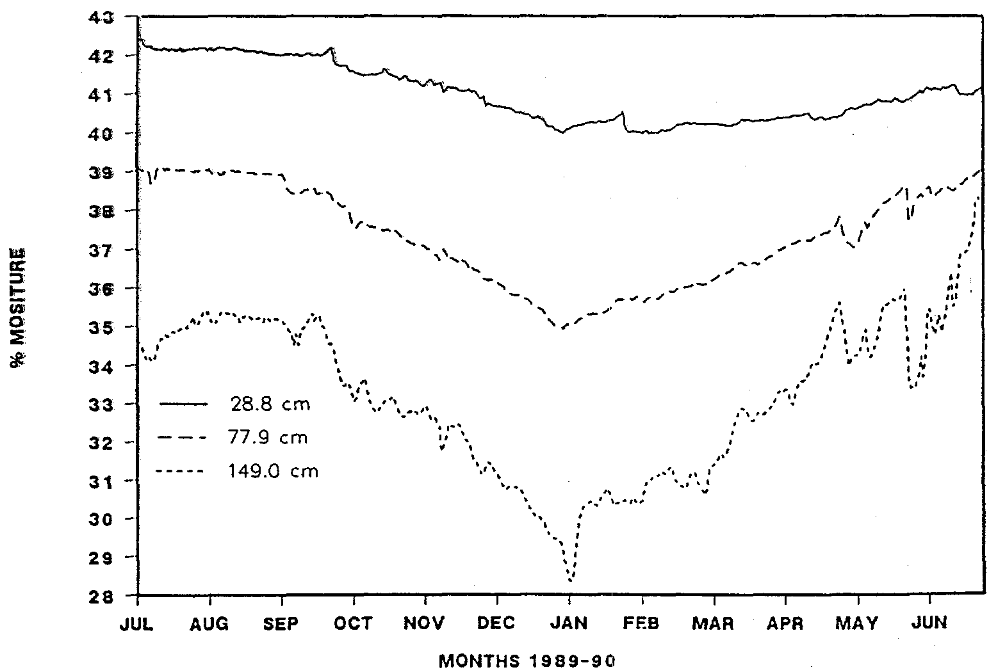

Figure C-33. ORNL lysimeter 4 soil moisture for 1989-90.

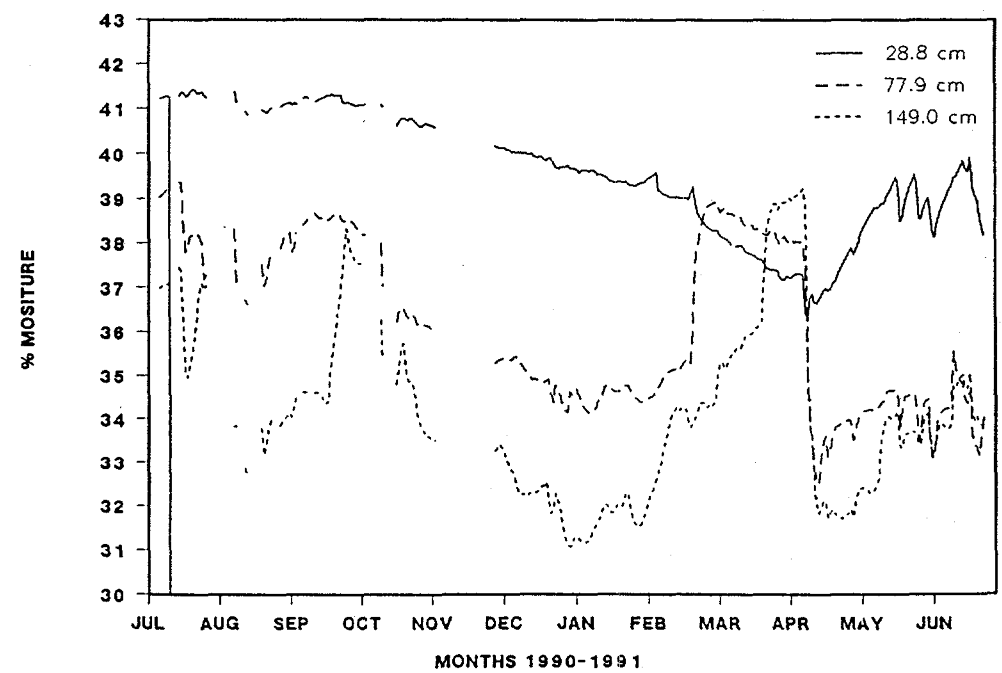

Figure C-34. ORNL lysimeter 4 soil moisture for 1990-91.

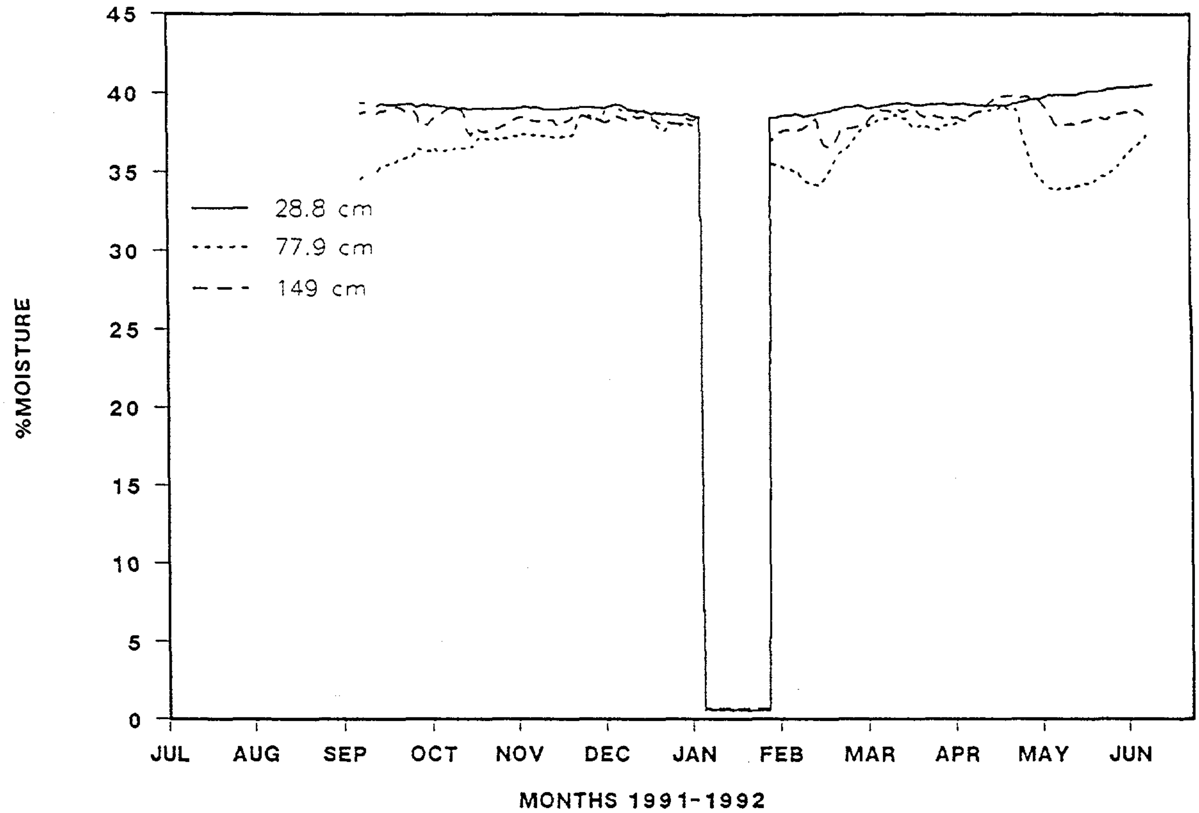

Figure C-35. ORNL lysimeter 4 soil moisture for 1991-92.

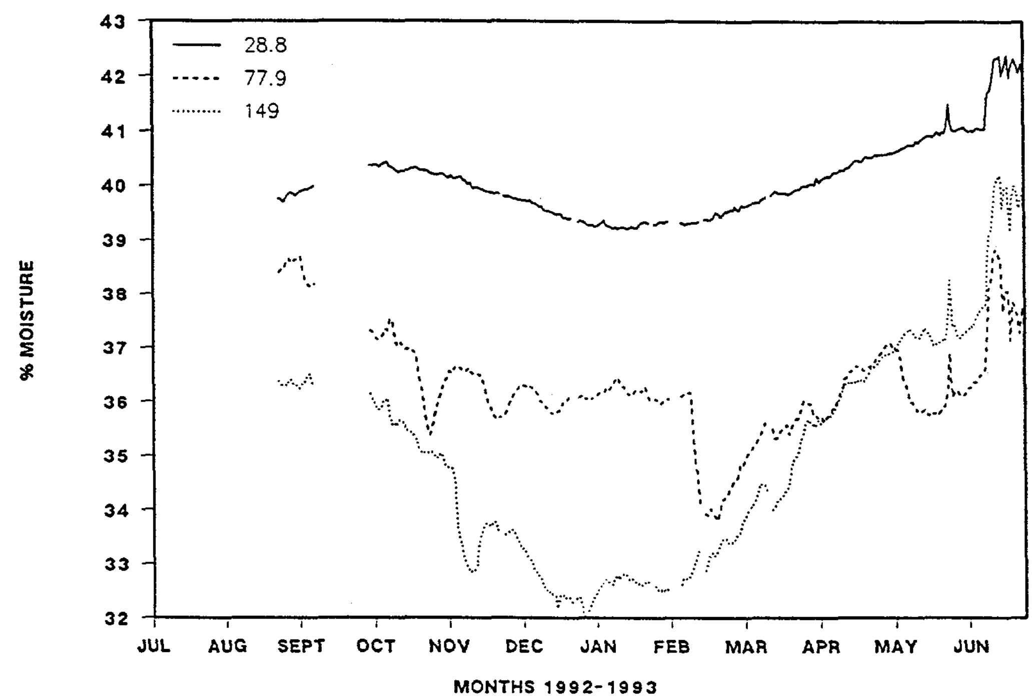

Figure C-36. ORNL lysimeter 4 soil moisture for 1992-93.

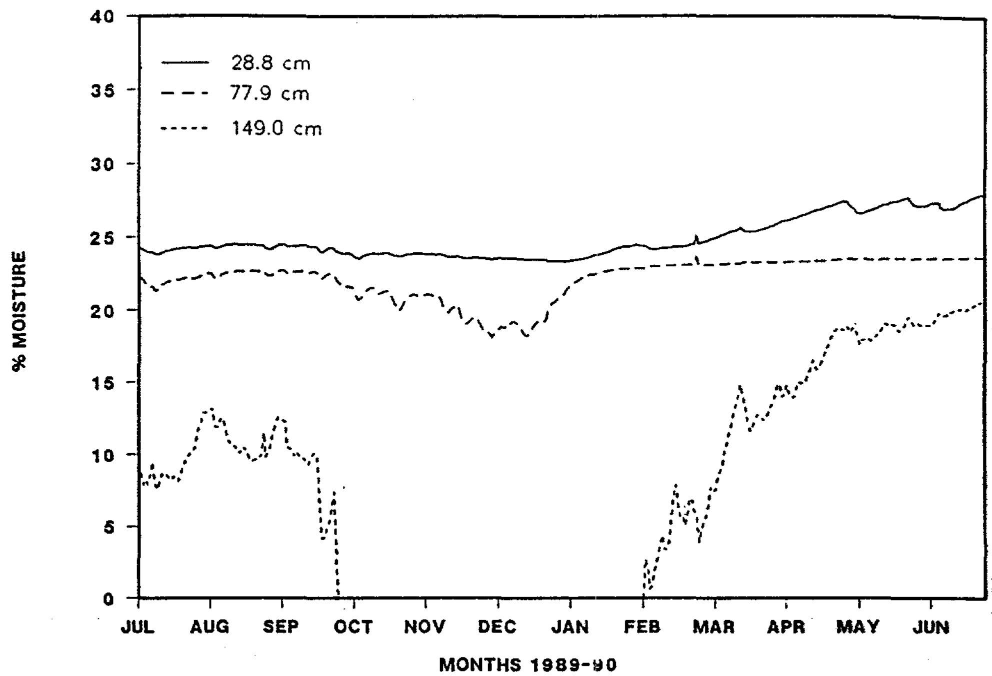

Figure C-37. ORNL lysimeter 5 soil moisture for 1989-90.

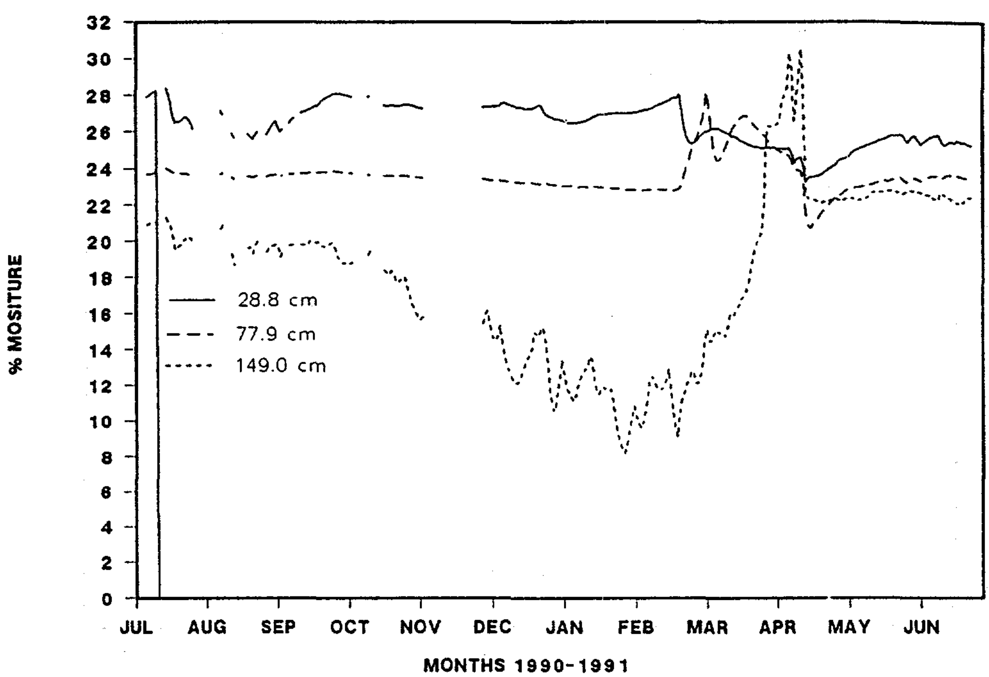

Figure C-38. ORNL lysimeter 5 soil moisture for 1990-91.

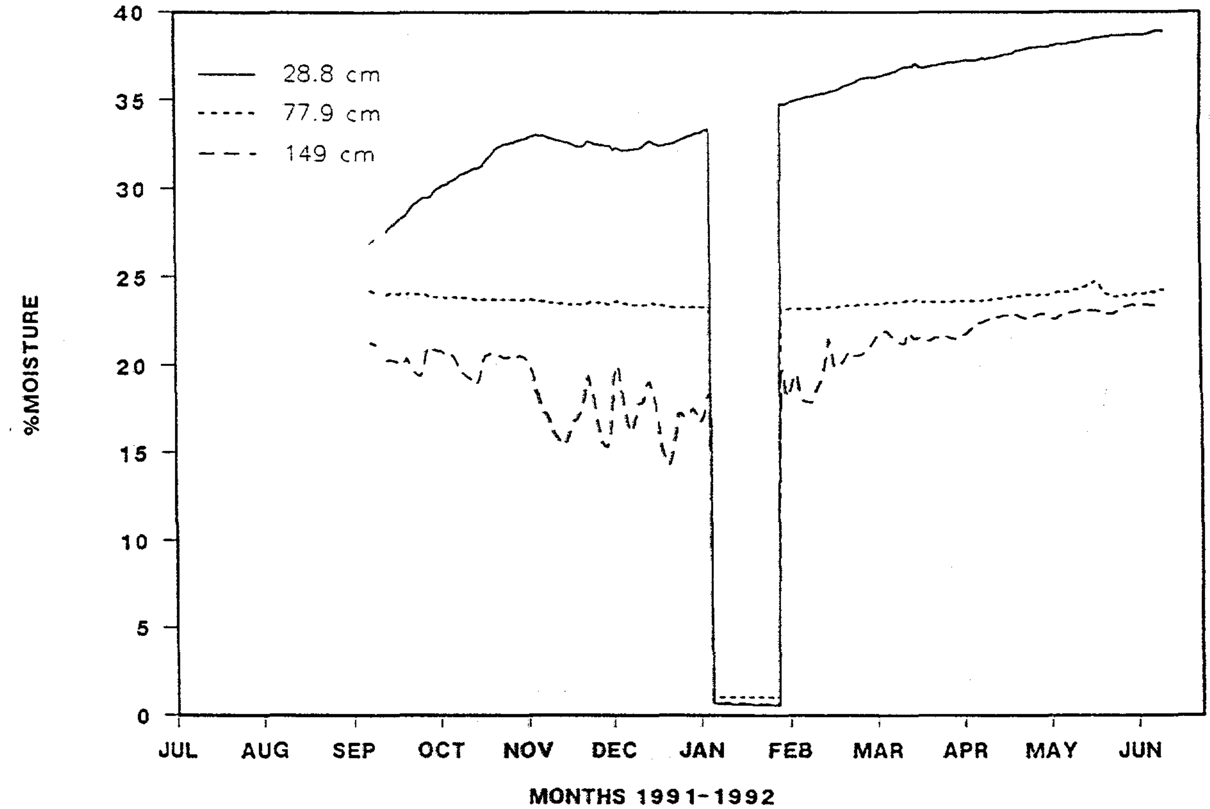

Figure C-39. ORNL lysimeter 5 soil moisture for 1991-92.

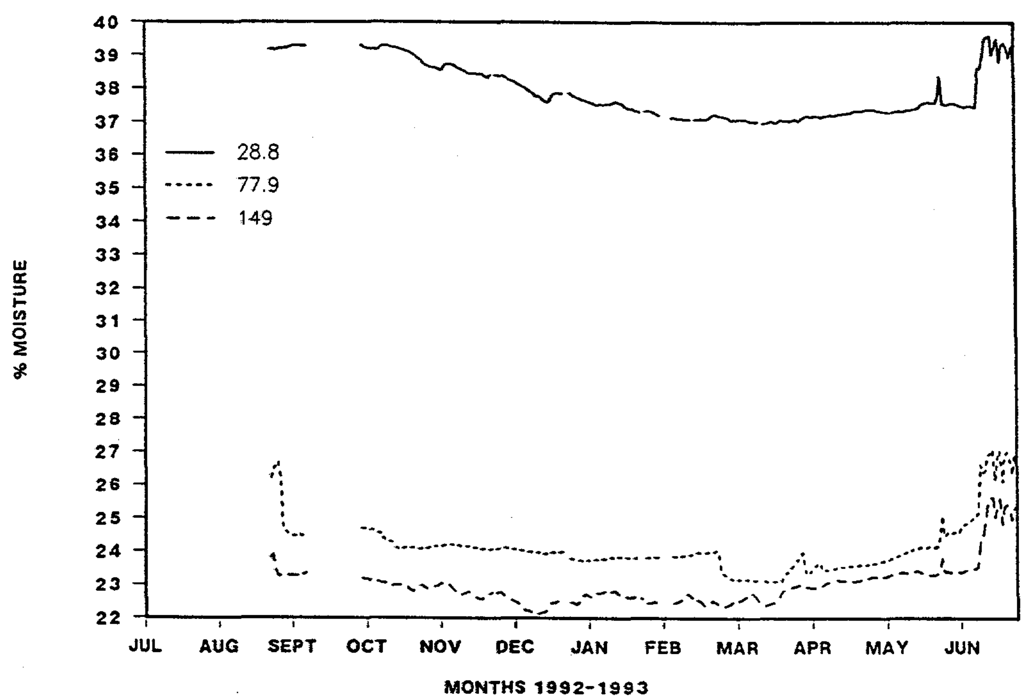

Figure C-40. ORNL lysimeter 5 soil moisture for 1992-93.

Appendix D

Soil Moisture Data—Gravimetric

Appendix D
Soil Moisture Data—Gravimetric

List of Tables

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

Lysimeter	Depth (cm)	% Moisture (dry weight)
1	0–41	17.4
1	41–62	20.8
1	62–82	21.2
1	82–107	21.4
1	107–133	21.9
1	133–153	22.6
1	153–182	23.0
1	182–202	23.3
2	0–41	18.5
2	41–62	22.3
2	62–82	21.1
2	82–107	22.6
2	107–133	22.6
2	133–153	23.3
2	153–182	23.8
2	182–202	23.8
3	0–41	22.2
3	41–62	22.7
3	62–82	24.7
3	82–107	24.9
3	107–133	24.6
3	133–153	24.6
3	153–182	24.9
3	182–202	24.3
4	0–41	22.3
4	41–62	22.6
4	62–82	22.7
4	82–107	22.7
4	107–133	24.2
4	133–153	24.0
4	153–182	23.8
4	182–202	23.9

a. Samples were collected on July 28, 1990.

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

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

a. Samples were collected on July 16, 1991.

Table D-3. Soil moisture percentage of ANL-E lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

		% Moisture (dry weight)	
Lysimeter	Depth (cm)	Gravimetric	Neutron probe
1	0–41	18.9	—
1	41–62	25.0	19.6
1	62–85	20.5	20.5
1	82–107	21.3	—
1	107–133	21.3	—
1	133–153	22.1	19.8
1	153–182	22.3	—
1	182–202	22.6	20.3
2	0–41	18.6	—
2	41–62	19.8	17.3
2	62–82	21.5	19.7
2	82–107	21.6	—
2	107–133	21.7	—
2	133–153	22.2	18.8
2	153–182	23.0	—
2	182–202	22.3	19.2
3	0–41	20.0	—
3	41–62	21.2	20.1
3	62–82	23.8	22.2
3	82–107	24.2	—
3	107–133	24.1	—
3	133–153	24.2	20.3
3	153–182	24.0	—
3	182–202	24.8	20.7
4	0–41	21.3	—
4	41–62	21.0	20.9
4	62–82	22.5	21.8
4	82–107	22.7	—
4	107–133	22.7	—
4	133–153	23.2	21.7
4	153–182	23.2	—
4	182–202	23.8	21.8

a. Samples were collected on July 29, 1992.

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

		% Moisture (dry weight)	
Lysimeter	Depth (cm)	Gravimetric	Neutron probe
1	0–41	15.5	—
1	41–62	22.6	19.9
1	62–85	26.5	20.8
1	82–107	24.0	—
1	107–133	24.2	—
1	133–153	23.6	22.0
1	153–182	22.9	—
1	182–202	22.9	23.3
2	0–41	16.2	—
2	41–62	19.9	19.0
2	62–82	20.1	20.1
2	82–107	21.6	—
2	107–133	22.0	—
2	133–153	22.1	21.8
2	153–182	22.2	—
2	182–202	23.0	24.2
3	0–41	18.2	—
3	41–62	21.9	18.9
3	62–82	24.2	22.3
3	82–107	24.2	—
3	107–133	23.2	—
3	133–153	23.9	22.6
3	153–182	24.6	—
3	182–202	23.6	23.7
4	0–41	20.3	—
4	41–62	25.6	21.2
4	62–82	28.6	22.5
4	82–107	25.0	—
4	107–133	27.0	—
4	133–153	22.9	23.4
4	153–182	24.3	—
4	182–202	24.7	23.8

a. Samples were collected on July 22, 1993.

Table D-5. Moisture profile of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	% Moisture (dry weight)
1	0–25	15.8
1	25–50	16.6
1	50–75	17.8
1	75–100	18.0
1	100–125	17.8
1	125–150	18.6
2	0–25	16.0
2	25–50	16.7
2	50–75	17.3
2	75–100	17.8
2	100–125	18.0
2	125–150	18.2
3	0–25	15.2
3	25–50	16.4
3	50–75	17.6
3	75–100	17.9
3	100–125	18.5
3	125–150	18.8
4	0–25	15.7
4	25–50	17.1
4	50–75	17.5
4	75–100	18.1
4	100–125	18.2
4	125–150	18.9

a. Samples were collected on July 10, 1990.

Table D-6. Moisture profile of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

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

a. Samples were collected on August 5, 1991.

Table D-7. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	% moisture (dry weight) Gravimetric	% moisture (dry weight) Neutron probe
1	0–41	12.0	12.7
1	41–62	14.5	15.9
1	62–85	15.6	16.3
1	82–107	15.8	16.5
1	107–133	15.9	15.8
1	133–153	17.4	16.8
1	153–182	18.7	17.2
1	182–202	19.4	17.7
2	0–41	10.6	12.7
2	41–62	14.2	15.8
2	62–82	13.2	16.5
2	82–107	15.9	16.5
2	107–133	16.7	14.9
2	133–153	18.6	17.7
2	153–182	18.1	17.7
2	182–202	18.3	18.0
3	0–41	17.2	17.2
3	41–62	17.9	18.3
3	62–82	17.5	18.7
3	82–107	18.9	18.7
3	107–133	19.4	17.9
3	133–153	18.8	18.7
3	153–182	19.2	19.0
3	182–202	19.9	19.5
4	0–41	14.2	14.5
4	41–62	16.2	19.2
4	62–82	18.7	19.6
4	82–107	19.5	20.0
4	107–133	19.7	17.8
4	133–153	20.1	19.8
4	153–182	20.1	20.5
4	182–202	21.5	21.2

a. Samples were collected July 15–27, 1992.

Table D-8. Soil moisture percentage of ORNL lysimeters 1 through 4 based on gravimetric measurement of water content.[a]

Lysimeter	Depth (cm)	% moisture (dry weight) Gravimetric	% moisture (dry weight) Neutron probe
1	0–41	12.3	13.0
1	41–62	14.8	15.7
1	62–85	16.3	15.8
1	82–107	16.6	16.3
1	107–133	17.7	15.2
1	133–153	18.0	16.4
1	153–182	18.7	16.8
1	182–202	19.5	17.5
2	0–41	10.0	13.4
2	41–62	13.8	16.2
2	62–82	14.1	16.2
2	82–107	13.5	15.8
2	107–133	14.6	14.2
2	133–153	17.0	17.0
2	153–182	17.4	17.0
2	182–202	16.7	17.6
3	0–41	12.3	17.5
3	41–62	15.0	18.5
3	62–82	16.0	18.7
3	82–107	15.9	18.7
3	107–133	16.9	17.8
3	133–153	17.6	18.6
3	153–182	19.0	18.9
3	182–202	18.0	19.4
4	0–41	12.2	15.5
4	41–62	14.3	19.1
4	62–82	15.4	20.2
4	82–107	15.9	19.8
4	107–133	16.6	17.9
4	133–153	17.8	19.4
4	153–182	18.9	19.9
4	182–202	19.0	20.9

a. Samples were collected on June 16, 1993.

Appendix E

Results of Beta and Gamma Analysis

Appendix E
Results of Beta and Gamma Analysis

List of Tables

Table E-1. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 5 (1989-1990).

Sample identification	Concentration (pCi/L)[a] Co-60 89 Oct	Co-60 90 Dec	Co-60 90 Mar	Co-60 90 Jul	Cs-137 89 Oct	Cs-137 90 Dec	Cs-137 90 Mar	Cs-137 90 Jul	Sr-90 89 Oct	Sr-90 90 Dec	Sr-90 90 Mar	Sr-90 90 Jul
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	7.9 ± 0.2	2.3 ± 0.1	11.4 ± 0.2	31 ± 1
Lys 4	<1	<1	<1	<1	<1	<1	<1	<1	<1	18.4 ± 0.3	<1	<1
Lys 5	<1	<1	<1	<1	<1	<1	2 ± 1	4.7 ± 1.6	384 ± 1	345 ± 1	452 ± 1	432 ± 1
Lys 1-3[c]	<5	<5	<5	<5	<5	<5	<5	<5	1.8E+4 ± 129	4,942 ± 50	2,762 ± 36	176 ± 12
Lys 2-3	<5	<5	<5	<5	965 ± 2	1,563 ± 55	2,556 ± 67	1,295 ± 53	1,946 ± 9	1,564 ± 55	2,480 ± 12	2,773 ± 11
Lys 3-3	<5	<5	<5	<5	<5	<5	<5	<5	7.9E+5 ± 1,452	6.7E+5 ± 1,343	1.0E+6 ± 1,603	1.01E+6 ± 1,531
Lys 4-3	<5	<5	<5	<5	<5	<5	<5	<5	2,353 ± 9	4,519 ± 13	3,321 ± 2	2,200 ± 10
Lys 5-3	<5	<5	<5	<5	1.9E+4 ± 134	1.5E+4 ± 50	2.1E+4 ± 141	2.7E+4 ± 234	9,377 ± 76	1.3E+4 ± 80	1.0E+4 ± 70	1.03E+4 ± 72

a. Concentration ± 2 sigma

b. 1-L subsample from leachate collector

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

Table E-2. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 6 (1990-1991).

Sample identification	Concentration (pCi/L)[a]								
	Co-60			Cs-137			Sr-90		
	Nov	Apr	July	Nov	Apr	July	Nov	Apr	July
Lys 1[b]	<20	<20	<20	<15	<15	<15	<1	<1	<1
Lys 2	<40	<20	<40	<35	<15	<35	<1	<1	<1
Lys 3	<40	<40	<20	<35	<35	<15	39 ± 1	43 ± 1	39 ± 2
Lys 4	<20	<40	<40	<15	<35	<35	<1	<1	59 ± 4
Lys 5	<20	<40	<20	<15	<35	<15	565 ± 4	661 ± 4	576 ± 8
Lys 1-3[c]	<30	<20	<20	<20	<15	272 ± 60	7,732 ± 34	7,892 ± 41	4,055 ± 31
Lys 2-3	<50	<20	<40	322 ± 41	184 ± 26	509 ± 60	3,420 ± 27	3,767 ± 37	4,169 ± 36
Lys 3-3	<50	<40	<40	<50	<35	232 ± 90	8.6E+5 ± 3,340	1.0E+6 ± 3,161	8.8E+5 ± 2,871
Lys 4-3	<50	<20	<20	<50	<15	<15	7,144 ± 47	9,046 ± 53	1.1E+4 ± 58
Lys 5-3	<50	<40	<40	2.3E+4 ± 1,318	3.9E+4 ± 2,498	5.6E+4 ± 2,480	1.1E+4 ± 216	1.1E+4 ± 177	1.7E+4 ± 197
Lys 1-1[c]	<50	<40	<20	<50	<35	<15	<10	<10	NA[d]
Lys 2-1	<30	<20	<20	<20	<15	<15	<10	<10	<10
Lys 3-1	<50	<40	<40	<50	<35	<35	570 ± 12	1,350 ± 23	876 ± 7
Lys 4-1	<30	<20	<20	<20	<15	<15	<10	<10	<10
Lys 5-1	<30	<40	<40	<20	<35	<35	808 ± 17	621 ± 15	961 ± 7

a. Concentration ± 2 sigma.

b. 1-L subsample from leachate collector.

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

d. Sample not available for analysis.

Table E-3. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 7 (1991-1992).

Sample identification	Concentration (pCi/L)[a] Co-60 Oct 91	Co-60 Dec 91	Co-60 Apr 92	Co-60 Jun 92	Cs-137 Oct 91	Cs-137 Dec 91	Cs-137 Apr 92	Cs-137 Jun 90	Sr-90 Oct 91	Sr-90 Dec 91	Sr-90 Apr 92	Sr-90 Jun 92
Lys 1[b]	<15	<5	<5	<5	<15	<5	<5	<5	<5	<5	<5	<5
Lys 2	<15	<5	<5	<5	<15	<5	<5	<5	<5	<5	<5	5.7 ± 4.3
Lys 3	<15	<5	<5	<5	<15	<5	<5	<5	74.2 ± 2.2	54.1 ± 8.1	48.7 ± 8.1	51.4 ± 8.1
Lys 4	<15	<5	<5	<5	<15	<5	<5	<5	<5	5.1 ± 4.3	4.6 ± 4.1	<5
Lys 5	<15	<5	<5	<5	<15	5.1 ± 2.2	<5	<5	1,302.7 ± 95.5	1,351 ± 54	1,162 ± 54.1	703 ± 27
Lys 1-3[c]	<15	<30	<30	<30	<15	<40	162 ± 105	<40	1.3E+4 ± 404	1.6E+4 ± 541	1.9E+4 ± 811	—[d]
Lys 2-3	<15	<30	<30	<30	320 ± 20	730 ± 54	270 ± 81	514 ± 81	3,365 ± 233	5,676 ± 270	7,838 ± 270	5,676 ± 270
Lys 3-3	<15	<30	<30	<30	<15	200 ± 70	<40	<40	91.8E+4 ± 9.5E+4	146E+4 ± 2.7E+4	2.2E+4 ± 541	108E+4 ± 2.7E+4
Lys 4-3	<15	<30	<30	<30	<15	<40	<40	<40	1.8E+4 ± 223	2.6E+4 ± 270	5,676 ± 270	2.7E+4 ± 7,203
Lys 5-3	<15	<30	<30	<30	14.0E+4 ± 250	6.2E+4 ± 2,703	4.3E+4 ± 2,703	9.2E+4 ± 2,703	1.6E+4 ± 239	2.1E+4 ± 1,351	5,135 ± 541	3.0 ± 2,703
Lys 1-1[c]	<15	35 ± 16	<30	<30	<15	<40	<40	<40	32.9 ± 5.7	32.4 ± 21.6	27.0 ± 21.6	<20
Lys 2-1	<15	<30	<30	<30	<15	<40	<40	<40	<5	91.9 ± 75.7	51.4 ± 27.0	<20
Lys 3-1	<15	<30	<30	<30	<15	<40	<40	<40	<5	2,324 ± 135	2,243 ± 102	1,865 ± 108
Lys 4-1	<15	<30	<30	<30	<15	<40	<40	<40	74.2 ± 2.2	1,703 ± 243	676 ± 189	20.0 ± 18.9
Lys 5-1	<15	<30	<30	<30	<15	<40	<40	<40	1,768 ± 40.8	1,270 ± 108	1,838 ± 162	2,324 ± 162

a. Concentration ±2 sigma.

b. One-L subsample from leachate collector.

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

d. None detected.

Table E-4. Results of beta and gamma analysis of ANL-E soil moisture and leachate samples, year 8 (1992-1993).

	Concentration (pCi/L)[a]											
	Co-60				Cs-137				Sr-90			
Sample identification	Oct 92	Dec 92	Apr 93	Jun 93	Oct 92	Dec 92	Apr 93	Jun 93	Oct 92	Dec 92	April 93	June 93
Lys 1[b]	0.3 ± 5.7	<5	<5	<5	6.5 ± 2.7	7.7 ± 1.2	<5	<5	3.5 ± 3.5	89 ± 2	1.1 ± 0.4	0.3 ± 0.4
Lys 2	-0.3 ± 3.2	<5	<5	<5	1.1 ± 3.2	<5	<5	<5	3.8 ± 3.5	1.5 ± 0.3	1.3 ± 0.4	0.3 ± 0.3
Lys 3	0.3 ± 6	<5	<5	<5	4.3 ± 5.7	<5	<5	<5	297 ± 27	228 ± 8	232 ± 10	224 ± 3
Lys 4	3 ± 4	<5	<5	<5	4.1 ± 2.4	<5	<5	<5	7 ± 4	5.7 ± 0.5	8 ± 0.5	3.4 ± 0.5
Lys 5	-2 ± 6	<5	<5	<5	3.8 ± 4.1	<5	<5	12 ± 2	1,162 ± 54	1,270 ± 20	910 ± 4	930 ± 18
Lys 1-3[c]	51 ± 114	<5	<5	<5	116 ± 92	<5	113 ± 17	<5	5,946 ± 270	1.7E+4 ± 166	1.4E+4 ± 127	3.0E+4 ± 232
Lys 2-3	8 ± 70	<5	<5	<5	378 ± 81	514 ± 77	338 ± 51	430 ± 20	8,378 ± 270	5,970 ± 85	184 ± 19	4,905 ± 91
Lys 3-3	-24 ± 15	<5	<5	<5	60 ± 146	<5	255 ± 38	32 ± 6	130E+4 ± 2.7E+4	140E+4 ± 2.3E+4	140E+4 ± 2E+4	140E+4 ± 2.3E+4
Lys 4-3	-16 ± 76	<5	<5	<5	-16 ± 62	<5	<5	<5	4.9E+4 ± 2,703	3.5E+4 ± 258	3.6E+4 ± 228	4.8E+4 ± 332
Lys 5-3	5 ± 127	<5	<5	6 ± 1	12.0E+4 ± 2,703	12.9E+4 ± 1.8E+4	10.1E+4 ± 1.5E+4	<5	3.2E+4 ± 2,703	2.4E+4 ± 260	2.5E+4 ± 239	2.8E+4 ± 370
Lys 1-1[c]	8 ± 32	<5	<6	<5	38 ± 24	<5	<5	15 ± 2	32 ± 16	27 ± 3	14 ± 2	16 ± 2.2
Lys 2-1	-11 ± 30	<5	<5	—[d]	7 ± 17	123 ± 18	<5	—[d]	15 ± 15	88 ± 12	2.6 ± 2.2	—[d]
Lys 3-1	-19 ± 62	<5	<5	<5	5 ± 65	<5	50 ± 7	18 ± 2	3,784 ± 270	3,292 ± 36	6,359 ± 46	4,739 ± 45
Lys 4-1	135 ± 18	<5	<5	—[d]	-35 ± 241	<5	<5	—[d]	186 ± 76	456 ± 29	176 ± 12	—[d]
Lys 5-1	8 ± 70	<5	<5	<5	-3 ± 68	11 ± 2	34 ± 5	43 ± 7	1,459 ± 81	1,619 ± 21	1,158 ± 21	1,045 ± 20
Lys 1-2	—	—	<5	—	—	—	37.3 ± 5.6	—	—	—	6.5 ± 3.3	—
Lys 2-2	—	—	<5	—	—	—	<5	—	—	—	2.6 ± 2.2	—
Lys 3-2	—	—	<5	—	—	—	<5	—	—	—	26.3 ± 3.7	—
Lys 4-2	—	—	<5	—	—	—	15.9 ± 2.4	—	—	—	5.2 ± 1.3	—
Lys 5-2	—	—	<5	—	—	—	223.7 ± 33.6	—	—	—	1,246 ± 19	—

a. Concentration ±2 sigma.

b. One-L subsample from leachate collector.

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

d. None detected.

Table E-5. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 5 (1989-1990).

Sample Identification	Concentration[a] (pCi/L) Co-60 Oct 89	Co-60 Jan 90	Co-60 Apr 90	Co-60 Jul 90	Cs-137 Oct 89	Cs-137 Jan 90	Cs-137 Apr 90	Cs-137 Jul 90
Lys 1[b]	-1.6 ± 4.0	0.5 ± 5.7	<108	3.2 ± 4.6	0.5 ± 3.5	1.3 ± 4.0	40.5 ± 37.8	0.8 ± 5.9
Lys 2	6.2 ± 3.8	2.7 ± 5.7	<108	0.8 ± 3.5	1.6 ± 4.6	5.1 ± 4.9	27.0 ± 59.5	0.5 ± 3.2
Lys 3	1.9 ± 7.0	0.8 ± 4.9	<81	0.75 ± 3.0	-3.2 ± 7.8	0.3 ± 3.8	2.76 ± 4.9	1.1 ± 4.9
Lys 4	-0.5 ± 3.2	-0.8 ± 3.5	<81	1.2 ± 7.0	0.3 ± 3.2	0.8 ± 3.0	<54	3.5 ± 6.2
Lys 5	2.4 ± 4.3	1.1 ± 5.1	<54	2.2 ± 4.3	64.9 ± 5A	37.8 ± 5.4	<81	22 ± 4.6
Lys 1-3[c]	13.5 ± 29.7	-13.5 ± 43.2	<54	2.7 ± 48.7	2.7 ± 29.7	-8.1 ± 35.1	<54	19 ± 35
Lys 2-3	5.4 ± 46.0	16.2 ± 59.2	<81	46 ± 38	2.7 ± 43.2	24.3 ± 54	<54	24 ± 57
Lys 3-3	46.0 ± 56.8	—[d]	—	—	27 ± 64.9	—	—	—
Lys 4-3	-8.1 ± 37.8	24.3 ± 35.1	<54	22 ± 103	2.7 ± 29.7	2.7 ± 043.7	<54	5 ± 76
Lys 5-3	8.6 ± 23.8	13.5 ± 32.4	<54	5.4 ± 62	595 ± 27	432 ± 27	514 ± 27	946 ± 81

Table E-5. (continued).

Sample Identification	Concentration[a] (pCi/L)							
	Sb-125				Sr-90			
	Oct 89	Jan 90	Apr 90	Jul 90	Oct 89	Jan 90	Apr 90	Jul 90
Lys 1[b]	<8.1	<54	<108	<8.1	2.7 ± 29.7	3.5 ± 3.2	48.7 ± 8.1	18.1 ± 5.1
Lys 2	<5.4	<54	<108	<8.1	3.8 ± 3.2	1.3 ± 2.54	0.3 ± 3.2	4.6 ± 3.2
Lys 3	<10.8	<108	<108	<8.1	0.4 ± 2.5	1.9 ± 3.0	0.8 ± 3.2	1.9 ± 2.7
Lys 4	<5.4	<54	<108	<8.1	2.7 ± 3.2	1.1 ± 3.0	2.7 ± 3.5	4.3 ± 3.2
Lys 5	<10.8	<54	<108	<8.1	194.6 ± 13.3	405 ± 27	297 ± 27	3.2 ± 27
Lys 1-3[c]	<54	<54	<81	<108	4.0E+4 ± 2,703	3.5E+4 ± 2,703	4.0E+4 ± 2,703	8.4E+4 ± 2,703
Lys 2-3	<54	<81	<81	<108	4,325 ± 270	4,325 ± 270	7,839 ± 270	8,920 ± 2,703
Lys 3-3	<108	—	—	—	2.2E+4 ± 270	—	—	—
Lys 4-3	<54	<54	<81	<108	62.2 ± 13.5	70.3 ± 13.5	183.8 ± 27	297 ± 27
Lys 5-3	<54	<54	<81	<108	175.7 ± 21.6	221.6 ± 24.3	460 ± 54	568 ± 27

a. Concentration ± 2 sigma.

b. 1-L subsample from leachate collection.

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

d. Sample not available.

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

Sample Identification	Concentration[a] (pCi/L)					
	Co-60			Cs-137		
	Nov 90	Apr 91	Jun 91	Nov 90	Apr 91	Jun 91
Lys 1[b]	0.5 ± 5.9	0.8 ± 5.9	29.7 ± 27.0	-0.8 ± 5.7	1.6 ± 5.7	8.1 ± 35.1
Lys 2	0.5 ± 4.1	0.5 ± 4.1	22.4 ± 23.8	-0.8 ± 3.5	0.8 ± 4.1	8.1 ± 35.1
Lys 3	6.5 ± 4.3	-1.6 ± 6.5	21.6 ± 29.7	2.2 ± 4.9	1.4 ± 5.7	8.1 ± 35.1
Lys 4	3.0 ± 5.9	0.8 ± 6.2	2.7 ± 37.8	-0.8 ± 7.0	1.1 ± 6.5	8.1 ± 37.8
Lys 5	-2.7 ± 5.9	0.3 ± 5.1	16.2 ± 37.8	24.1 ± 4.3	37.8 ± 5.4	156.6 ± 32.4
Lys 1-3[c]	6.2 ± 21.6	10.8 ± 37.8	19.7 ± 22.1	8.6 ± 22.7	5.4 ± 32.4	5.4 ± 29.7
Lys 2-3	21.6 ± 32.4	2.7 ± 37.8	8.1 ± 40.5	27.0 ± 27.0	-8.1 ± 37.8	10.8 ± 35.1
Lys 3-3	13.5 ± 32.4	15.1 ± 21.6	25.3 ± 24.3	8.1 ± 37.8	-5.4 ± 29.7	2.7 ± 32.4
Lys 4-3	-2.7 ± 29.7	1.9 ± 18.5	5.4 ± 40.5	7.6 ± 25.4	4.6 ± 23.0	2.7 ± 32.8
Lys 5-3	8.6 ± 23.8	21.6 ± 32.5	19.7 ± 23.8	892 ± 54	2,271 ± 2,970	2,970 ± 270
Lys 1-1[c]	11.1 ± 14.1	-7.8 ± 26.2	18.9 ± 40.5	2.7 ± 23.2	7.3 ± 22.7	8.1 ± 40.5
Lys 2-1	-11.1 ± 24.6	8.1 ± 35.1	7.6 ± 26.2	4.6 ± 29.7	-16.2 ± 30.4	0.5 ± 25.9
Lys 3-1	35.1 ± 32.4	-5.4 ± 40.5	1.1 ± 4.9	2.7 ± 29.7	-2.7 ± 37.8	0.5 ± 25.9
Lys 4-1	-2.7 ± 40.5	-8.1 ± 32.4	1.1 ± 5.4	18.9 ± 29.7	2.7 ± 27.0	0.8 ± 3.2
Lys 5-1	-2.7 ± 29.7	-2.7 ± 27.0	2.7 ± 29.7	-5.4 ± 25.1	54.1 ± 21.6	32.4 ± 21.6
Lys 1[b]	<8.1	<8.1	2.7 ± 83	15.7 ± 4.9	15.1 ± 4.6	70.2 ± 10.8
Lys 2	<8.1	<8.1	2.7 ± 92	0.8 ± 2.7	0.5 ± 4.9	13.5 ± 5.7
Lys 3	<8.1	<8.1	2.7 ± 83	0.5 ± 3.8	1.9 ± 3.0	4.8 ± 4.3
Lys 4	<8.1	<8.1	24 ± 95	0.0 ± 4.9	2.4 ± 4.9	51.3 ± 8.1
Lys 5	<8.1	<8.1	54 ± 81	541 ± 27	351 ± 27	486 ± 27

Table E-6. (continued).

Sample Identification	Concentration[a] (pCi/L)					
	Sb-125			Sr-90		
	Nov 90	Apr 91	Jun 91	Nov 90	Apr 91	Jun 91
Lys 1-3[c]	—[d]	—	—	9.2E+4 ± 2,703	7.0E+4 ± 2,703	8.1E+4 ± 2,700
Lys 2-3	—	—	—	1.0E+4 ± 270	8,920 ± 270	1.0E+4 ± 270
Lys 3-3	—	—	—	1.0E+5 ± 2,703	9.5E+4 ± 2,703	1.4E+5 ± 2,700
Lys 4-3	—	—	—	622 ± 54	351 ± 27	1,485 ± 54
Lys 5-3	—	—	—	730 ± 54	351 ± 27	1.1E+4 ± 270
Lys 1-1[c]	—	—	—	784 ± 27	1,135 ± 54	1,701 ± 81
Lys 2-1	—	—	—	12.4 ± 4.9	2.7 ± 9.5	164.7 ± 27
Lys 3-1	—	—	—	1.1 ± 7.6	3.8 ± 7.0	8.6 ± 84
Lys 4-1	—	—	—	10.0 ± 8.9	11.4 ± 10.0	5.7 ± 84
Lys 5-1	—	—	—	27.0 ± 24.3	40.5 ± 13.5	64.8 ± 16.2

a. Concentration ± 2 sigma.

b. 1-L subsample from leachate collection.

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

d. Sample not available.

Table E-7. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 7 (1991-1992).

Sample identification	Concentration (pCi/L)[a] Co-60 Sep 91	Co-60 Dec 91	Co-60 Mar 92	Co-60 Jun 92	Cs-137 Sep 91	Cs-137 Dec 91	Cs-137 Mar 92	Cs-137 Jun 92
Lys 1[b]	24.3 ± 59.5	2.7 ± 40.5	8.6 ± 20.0	-0.3 ± 4.9	18.9 ± 64.9	-2.7 ± 51.3	2.7 ± 32.4	-0.3 ± 5.4
Lys 2	32.4 ± 48.7	-18.9 ± 35.1	18.1 ± 26.5	-0.3 ± 4.9	43.2 ± 54.1	12.4 ± 26.2	-1.4 ± 25.7	1.4 ± 3.2
Lys 3	16.2 ± 48.7	2.7 ± 40.5	-8.1 ± 59.5	1.6 ± 3.0	54.1 ± 56.8	16.2 ± 37.8	5.4 ± 64.9	-0.5 ± 4.3
Lys 4	-35 ± 91.9	1.9 ± 26.2	2.7 ± 51.4	0.08 ± 4.1	-2.7 ± 75.7	13.5 ± 32.4	-32.4 ± 70.3	0.8 ± 4.1
Lys 5	10.8 ± 59.5	8.4 ± 22.7	-2.7 ± 59.5	1.6 ± 10.5	73.0 ± 40.5	56.8 ± 27.0	43.2 ± 59.5	35.1 ± 8.1
Lys 1-3[c]	-8.1 ± 37.8	24.3 ± 54.1	13.5 ± 56.8	-8.1 ± 35.1	8.1 ± 37.8	-16.2 ± 43.2	70.3 ± 32.4	56.8 ± 18.9
Lys 2-3	-8.1 ± 75.7	-16.2 ± 67.6	2.7 ± 43.2	-13.5 ± 70.3	-5.4 ± 48.7	21.6 ± 48.6	13.5 ± 32.4	21.6 ± 48.6
Lys 3-3	-43.2 ± 116.2	2.7 ± 45.8	-45.9 ± 102.7	-5.4 ± 70.3	32.4 ± 64.9	16.2 ± 35.1	10.8 ± 64.9	-40.5 ± 59.2
Lys 4-3	24.3 ± 32.4	-13.5 ± 86.5	-16.2 ± 97.3	-5.4 ± 70.3	5.4 ± 48.7	27.0 ± 51.4	35.1 ± 56.8	-5.4 ± 70.3
Lys 5-3	-13.5 ± 64.9	27.0 ± 62.2	21.6 ± 51.4	-10.8 ± 51.4	2,973 ± 270	1,324 ± 108	1,622 ± 108	3,243 ± 270
Lys 1-1[c]	-35.1 ± 83.8	24.3 ± 37.8	-10.8 ± 67.6	-54 ± 43.2	8.1 ± 43.2	27.0 ± 67.6	24.3 ± 64.9	24.3 ± 35.1
Lys 2-1	62.2 ± 62.2	-27.0 ± 89.2	-51.4 ± 94.6	13.5 ± 40.5	13.5 ± 48.7	10.8 ± 83.8	21.6 ± 43.2	5.4 ± 29.7
Lys 3-1	-2.7 ± 46.0	10.8 ± 8.1	18.9 ± 86.5	37.8 ± 54.1	40.5 ± 67.6	-8.1 ± 67.6	16.2 ± 56.8	48.6 ± 64.9
Lys 4-1	-10.8 ± 48.7	10.8 ± 12.6	8.1 ± 59.5	29.7 ± 54.1	-5.4 ± 32.4	-2.7 ± 59.5	24.3 ± 56.8	8.1 ± 73.0
Lys 5-1	16.2 ± 48.7	35.1 ± 105.4	18.9 ± 43.2	32.4 ± 48.6	27.0 ± 48.7	35.1 ± 64.9	43.2 ± 27.0	48.6 ± 48.6

Table E-7. (continued).

Sample identification	Concentration (pCi/L)[a]							
	Sb-125				Sr-90			
	Sep 91	Dec 91	Mar 92	Jun 92	Sep 91	Dec 91	Mar 92	Jun 92
Lys 1[b]	24 ± 184	-11 ± 86	-2.7 ± 56	-5.9 ± 14	75.7 ± 8.1	197 ± 189	170 ± 16.2	232 ± 16.2
Lys 2	103 ± 162	-11 ± 51	24 ± 51	-3.0 ± 8.6	3.5 ± 2.2	24 ± 162	10 ± 5.1	6.2 ± 3.8
Lys 3	-105 ± 211	-68 ± 119	-51 ± 167	0.3 ± 8.4	3.5 ± 1.9	43 ± 127	6.2 ± 4.1	1.4 ± 2.7
Lys 4	-81 ± 211	-24 ± 78	-51 ± 156	5.4 ± 12	5.4 ± 3.0	73 ± 138	2.2 ± 3.8	1.4 ± 3.0
Lys 5	38 ± 168	-5 ± 51	54 ± 132	3.2 ± 15	784 ± 27	702 ± 243	946 ± 27	1,189 ± 27
Lys 1-3[c]	—[d]	—[d]	—[d]	—[d]	9.7E+4 ± 2,703	10.0E+4 ± 2,703	8.4E+4 ± 2,703	9.5E+4 ± 2,703
Lys 2-3	—[d]	—[d]	—[d]	—[d]	1.2E+4 ± 207	1.3E+4 ± 811	1.0E+4 ± 541	1.2E+4 ± 270
Lys 3-3	—[d]	—[d]	—[d]	—[d]	16.2E+4 ± 2,703	12.4E+4 ± 2,703	15.4 ± 2,703	20.5E+4 ± 2,703
Lys 4-3	—[d]	—[d]	—[d]	—[d]	2,000 ± 135	2,243 ± 297	3,514 ± 270	4594 ± 270
Lys 5-3	—[d]	—[d]	—[d]	—[d]	3.2E+4 ± 2,703	2.7E+4 ± 2,703	2.2E+4 ± 811	3.2 ± 2,703
Lys 1-1[c]	—[d]	—[d]	—[d]	—[d]	3,244 ± 270	4,324 ± 541	4,865 ± 541	6,216 ± 270
Lys 2-1	—[d]	—[d]	—[d]	—[d]	70 ± 27	224 ± 165	249 ± 127	130 ± 18.9
Lys 3-1	—[d]	—[d]	—[d]	—[d]	460 ± 54	211 ± 162	254 ± 132	78 ± 21.6
Lys 4-1	—[d]	—[d]	—[d]	—[d]	73 ± 30	5 ± 122	43.2 ± 83.8	19.5 ± 15.9
Lys 5-1	—[d]	—[d]	—[d]	—[d]	176 ± 38	41 ± 119	227 ± 130	81.1 ± 21.6

a. Concentration ±2 sigma.

b. One-L subsample from leachate collector.

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

d. None detected.

Table E-8. Results of beta and gamma analysis of ORNL soil moisture and leachate samples, year 8 (1992-1993).

Sample identification	Concentration (pCi/L)[a]							
	Co-60				Cs-137			
	Oct 92	Dec 92	Mar 93	Jun 93	Oct 92	Dec 92	Mar 93	Jun 93
Lys 1[b]	0.8 ± 3.5	64.9 ± 5.4	1.6 ± 5.4	0.3 ± 6.8	1.6 ± 3.2	38 ± 5.4	1.4 ± 4.3	6.8 ± 4.6
Lys 2	0.3 ± 4.3	3.2 ± 3.0	3.2 ± 8.4	3.8 ± 7.3	4.1 ± 3.0	6.2 ± 2.7	1.1 ± 9.7	49 ± 11
Lys 3	4.3 ± 3.5	3.2 ± 2.4	4.6 ± 1.6	3.2 ± 13.2	2.7 ± 5.7	13 ± 2.7	-0.3 ± 5.7	16 ± 8
Lys 4	2.4 ± 3.8	5.7 ± 4.9	-7.3 ± 15.1	-5.4 ± 16	0.5 ± 4.3	3.2 ± 5.9	2.2 ± 9.5	22 ± 99
Lys 5	0.3 ± 3.8	4.3 ± 3.5	0.5 ± 11	0.5 ± 12	28 ± 2.7	30 ± 5.4	26 ± 9.5	12 ± 8.4
Lys 1-3[c]	-19 ± 81	-22 ± 70	35 ± 73	14 ± 54	135 ± 60	11 ± 54	43 ± 70	38 ± 62
Lys 2-3	8.1 ± 68	-13 ± 108	49 ± 65	-11 ± 68	-2.7 ± 76	49 ± 50	32 ± 76	2.7 ± 68
Lys 3-3	32 ± 54	19 ± 173	8.4 ± 30	30 ± 68	30 ± 65	-49 ± 208	6.8 ± 23	19 ± 76
Lys 4-3	32 ± 54	57 ± 38	5.4 ± 70	2.7 ± 43	-2.7 ± 70	8.1 ± 100	-30 ± 78	2.7 ± 38
Lys 5-3	22 ± 38	5.4 ± 108	62 ± 62	-19 ± 70	6,757 ± 270	108 ± 62	4,324 ± 270	8,649 ± 270
Lys 1-1[c]	32 ± 54	2.7 ± 70	-38 ± 97	8.1 ± 60	-2.7 ± 70	30 ± 65	22 ± 90	19 ± 60
Lys 2-1	-16 ± 76	19 ± 116	8.1 ± 84	24 ± 57	16 ± 70	2.7 ± 103	78 ± 43	-38 ± 76
Lys 3-1	11 ± 60	13 ± 103	30 ± 97	46 ± 54	35 ± 68	30 ± 78	-16 ± 89	-11 ± 154
Lys 4-1	-8.1 ± 73	38 ± 116	24 ± 87	49 ± 60	-14 ± 78	-2.7 ± 235	16 ± 70	11 ± 73
Lys 5-1	2.7 ± 60	11 ± 84	22 ± 70	8.1 ± 70	78 ± 68	95 ± 57	-30 ± 176	5.4 ± 73
Lys 1-2	—	—	—	21.6 ± 62.2	—	—	—	-5.4 ± 75.7
Lys 2-2	—	—	—	-16.2 ± 73.0	—	—	—	10.8 ± 70.3
Lys 3-2	—	—	—	-29.7 ± 75.7	—	—	—	27.0 ± 62.2
Lys 4-2	—	—	—	37.8 ± 29.7	—	—	—	8.1 ± 29.7
Lys 5-2	—	—	—	8.1 ± 62.2	—	—	—	324.3 ± 54.1

Table E-8. (continued).

Sample identification	Concentration (pCi/L)[a] Sb-125 Oct 92	Sb-125 Dec 92	Sb-125 Mar 93	Sb-125 Jun 93	Sr-90 Oct 92	Sr-90 Dec 92	Sr-90 Mar 93	Sr-90 Jun 93
Lys 1[b]	-2.7 ± 7.8	0.3 ± 12	-3.2 ± 11	-5.9 ± 14	324 ± 27	486 ± 27	432 ± 27	486 ± 27
Lys 2	0.3 ± 6.5	1.9 ± 8.4	0.3 ± 18	-3.0 ± 8.6	9.2 ± 3.8	20 ± 6.5	15 ± 3.0	22 ± 6.5
Lys 3	-1.6 ± 12	-0.8 ± 7.6	0.5 ± 17	0.3 ± 8.4	3.8 ± 3.2	10 ± 4.9	3.5 ± 1.9	7.8 ± 4.9
Lys 4	2.2 ± 9.5	-1.1 ± 14	-5.4 ± 26	5.4 ± 12	-0.3 ± 2.3	2.7 ± 3.8	-0.1 ± 1.4	1.6 ± 3.5
Lys 5	-2.2 ± 8.9	0.8 ± 9.2	1.6 ± 21	3.2 ± 15	1,838 ± 54	1,919 ± 81	811 ± 27	865 ± 54
Lys 1-3[c]	—[d]	—[d]	—[d]	—[d]	9.4E+4 ± 2,703	7.5E+4 ± 2,703	6.5E+4 ± 2,703	8.4E+4 ± 2,703
Lys 2-3	—[d]	—[d]	—[d]	—[d]	1.2E+4 ± 207	0.84E+4 ± 270	1.0E+4 ± 270	1.1E+4 ± 270
Lys 3-3	—[d]	—[d]	—[d]	—[d]	17.3E+4 ± 2,703	14.3E+4 ± 2,703	17.8E+4 ± 8,108	23.7E+4 ± 8,108
Lys 4-3	—[d]	—[d]	—[d]	—[d]	6,757 ± 270	5,946 ± 270	7,838 ± 270	7,838 ± 270
Lys 5-3	—[d]	—[d]	—[d]	—[d]	3.2E+4 ± 2,703	60 ± 22	1.0E+4 ± 541	1.4E+4 ± 541
Lys 1-1[c]	—[d]	—[d]	—[d]	—[d]	7,297 ± 270	6,757 ± 270	8,649 ± 270	1,027 ± 270
Lys 2-1	—[d]	—[d]	—[d]	—[d]	214 ± 24	222 ± 35	324 ± 27	351 ± 54
Lys 3-1	—[d]	—[d]	—[d]	—[d]	70 ± 11	103 ± 24	105 ± 16	138 ± 30
Lys 4-1	—[d]	—[d]	—[d]	—[d]	38 ± 11	76 ± 22	30 ± 19	76 ± 30
Lys 5-1	—[d]	—[d]	—[d]	—[d]	70 ± 14	62 ± 22	51 ± 24	108 ± 36
Lys 1-2	—	—	—	-54.1 ± 167.6	—	—	—	37.8 ± 10.8
Lys 2-2	—	—	—	32.4 ± 151.4	—	—	—	29.7 ± 18.9
Lys 3-2	—	—	—	-2.7 ± 162.2	—	—	—	7.0 ± 13.2
Lys 4-2	—	—	—	2.7 ± 73.0	—	—	—	13.0 ± 8.1
Lys 5-2	—	—	—	2.7 ± 170.3	—	—	—	23.8 ± 9.5

a. Concentration ±2 sigma.

b. One-L subsample from leachate collector.

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

d. None detected.

Appendix F

Results of Chemical Speciation

Appendix F
Results of Chemical Speciation

List of Tables

Table F-1. ANL-E results of chemical speciation, lysimeter moisture cups 1, 2, 3, and 5, July 1991.

		Cation					Anion			
Sample	Solidification agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
RAIN 1	—	2.3	0.22	0.038	<0.5	1.0	0.37	5.5	<3	3.7
RAIN 2		3.0	0.21	0.045	<0.5	1.2	0.38	5.5	<3	3.7
Lys 1-1	Cement	89	13	14	<0.5	52	3.7	<0.5	<3	45
Lys 1-3		68	12	9.7	<0.5	39	3.0	<0.5	<3	42
Lys 2-2	Cement	89	13	13	<0.5	52	4.9	<0.5	<3	41
Lys 2-3		82	19	15	<0.5	62	3.2	<0.5	<3	57
Lys 3-1	VES	85	6.6	11	<0.5	48	2.1	3.0	<3	28
Lys 3-3		79	8.2	16	<0.5	47	8.2	<0.5	<3	28
Lys 3-5		84	3.4	16	<0.5	48	1.8	<0.5	<3	28
Lys 4-2	VES	80	6.5	10	<0.5	45	4.6	<0.5	<3	40
Lys 4-3		81	4.4	10	<0.5	45	2.4	<0.5	<3	41
Lys 5-1	Cement	5.5	0.6	11	<0.5	2.8	0.7	6.2	<3	7.0
Lys 5-3		6.8	10	30	<0.5	3.6	1.1	5.5	<3	6.9
Lys 5-5		4.4	0.8	25	<0.5	2.1	0.9	12	<3	7.0

Table F-2. ANL-E chemical speciation results from lysimeter moisture cups 1, 2, 3, 4, and 5, April 1992.

Sample	Solidification agent	Cation					Anion			
		Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	90	13	12	0.60	57	3.8	3.4	<0.5	39
Lys 1-3		72	12	9.1	0.51	39	4.0	13	<0.5	50
Lys 1-4		114	8.7	12	0.65	61	2.8	<0.1	<0.5	44
Lys 2-2	Cement	81	10	11	<0.5	50	3.4	<0.1	<0.5	23
Lys 2-3		72	18	13	0.67	62	3.5	0.13	<0.5	33
Lys 2-4		116	6.8	13	0.62	61	4.9	0.15	<0.5	36
Lys 3-1	VES	39	5.4	12	<0.5	46	2.4	3.6	<0.5	17
Lys 3-3		80	6.9	14	<0.5	45	12	0.17	<0.5	22
Lys 3-5		88	3.1	15	<0.5	50	3.0	9.9	<0.5	32
Lys 4-2	VES	82	4.8	8.3	<0.5	42	5.0	0.49	<0.5	35
Lys 4-3		66	5.0	9.5	<0.5	38	5.7	0.17	<0.5	30
Lys 4-5		109	4.0	12	<0.5	53	4.1	<0.1	<0.5	39
Lys 5-1	Cement	9.0	1.5	9.2	0.82	4.6	2.2	3.8	<0.5	8.7
Lys 5-3		9.2	7.8	29	3.7	5.1	2.2	1.3	<0.5	8.3
Lys 5-5		7.2	1.0	16	1.7	3.2	3.0	5.3	<0.5	10

Table F-3. ANL-E chemical speciation results from lysimeter moisture cups 1, 2, 3, 4, and 5, June 1993.

Sample	Solidification agent	Cation					Anion			
		Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	88	11	12	<1	53	2.0	0.32	0.94	38
Lys 1-3		45	6.6	5.9	<1	23	3.6	1.2	2.4	48
Lys 1-5		32	0.89	2.8	<1	7.8	2.0	4.4	<0.5	20
Lys 2-2	Cement	89	7.9	11	<1	48	2.3	<0.1	<0.5	27
Lys 2-3		20	0.30	2.3	<1	4.1	0.98	1.7	1.5	6.1
Lys 2-4		90	5.0	10	<1	49	2.1	0.21	<0.5	36
Lys 3-1	VES	67	3.3	7.9	<1	41	1.5	4.4	1.6	20
Lys 3-3		83	6.0	14	<1	48	6.4	0.48	1.7	27
Lys 3-5		62	2.3	16	<1	46	1.5	1.8	1.2	26
Lys 4-1	VES	75	4.2	11	<1	47	4.9	0.28	<0.5	34
Lys 4-3		86	5.4	9.4	<1	45	1.6	0.23	<0.5	35
Lys 4-5		86	2.8	9.5	<1	40	1.5	<0.1	<0.5	30
Lys 5-1	Cement	6.6	<0.3	7.4	<1	3.0	0.57	3.7	<0.5	4.4
Lys 5-3		8.3	8.5	28	3.6	4.2	1.3	4.3	3.5	5.6
Lys 5-5		6.9	<0.3	168	<1	2.9	0.98	4.8	<0.5	5.2

Table F-4. ORNL results of chemical speciation for lysimeter moisture cup 1 and 3, July 1991.

		Cation					Anion			
Sample	Solidification agent	Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	41	5.2	20	0.09	1.5	0.91	0.57	0.2	26.0
Lys 1-3		53	4.5	26	2.59	1.7	0.95	37	0.2	21.3
Lys 2-1	Cement	40	3.7	20	0.05	1.2	0.90	13.0	0.2	9.6
Lys 2-3		36	4.7	34	2.47	1.1	0.28	6.9	0.2	7.6
Lys 3-1	VES	34	1.9	22	0.09	0.9	0.85	44	0.2	6.7
Lys 3-3		120	4.9	31	0.38	2.0	2.43	39	0.2	7.3
Lys 4-1	VES	5.4	4.8	16	0.15	0.8	1.94	4.64	0.2	15.0
Lys 4-3		4.9	6.9	16	0.15	1.0	1.33	1.14	0.2	17.3
Lys 5-1	Cement	9.2	0.3	10	1.24	3.4	4.03	1.96	0.2	4.2
Lys 5-3		11	2.3	29	2.47	4.2	0.79	7.77	0.2	1.0

Table F-5. ORNL chemical speciation results from lysimeter moisture cups 1 and 3, July 1992.

Sample	Solidification agent	Cation					Anion			
		Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	43	3.5	19	<0.4	1.6	1.8	1.1	<0.1	24
Lys 1-3		41	3.5	22	2.6	1.6	1.0	9.0	<0.1	22
Lys 1-5		40	0.28	15	<0.4	1.3	1.0	11	<0.1	13
Lys 2-1	Cement	39	2.7	18	<0.4	1.2	0.8	2.4	<0.1	8.5
Lys 2-3		34	2.9	29	2.0	1.0	0.8	3.7	<0.1	7.5
Lys 2-5		4.5	0.15	5.6	<0.4	0.4	0.9	4.1	<0.1	0.9
Lys 3-1	VES	34	1.9	19	<0.4	0.82	2.5	0.3	<0.3	6.2
Lys 3-3		34	2.8	25	<0.4	1.3	4.0	2.5	<1	7.6
Lys 3-5		5.1	0.44	8.3	<0.4	1.3	3.2	12.5	<0.3	0.6
Lys 4-1	VES	6.4	4.2	9.6	<0.4	0.94	1.9	3.3	<0.3	16.3
Lys 4-3		6.2	6.0	12	<0.4	1.1	2.1	5.9	<0.3	1.1
Lys 4-5		2.9	0.25	6.8	<0.4	0.56	0.9	6.6	<0.3	18
Lys 5-1	Cement	8.5	0.6	8.1	1.6	3.7	2.3	12.7	<0.3	6.9
Lys 5-3		13	2.3	19	3.0	3.1	3.2	30	<0.3	7.0
Lys 5-5		8.0	0.15	15	1.1	<0.02	3.7	14	<0.3	4.5

Table F-6. ORNL chemical speciation results from lysimeter moisture cups 1, 3, and 5, July 1993.

Sample	Solidification agent	Cation					Anion			
		Ca (mg/L)	Na (mg/L)	Si (mg/L)	K (mg/L)	Mg (mg/L)	Cl (mg/L)	NO_3 (mg/L)	PO_4 (mg/L)	SO_4 (mg/L)
Lys 1-1	Cement	34	3.1	18	<1	1.3	5.3	9.2	<3	2.4
Lys 1-3		33	2.8	23	2.3	1.4	0.6	3.8	<3	18
Lys 1-5		35	0.34	19	<1	1.8	2.3	5.4	<3	11
Lys 2-1	Cement	41	2.2	16	<1	1.4	<1.0	25	<3	7.7
Lys 2-3		36	2.4	28	2.3	1.1	1.3	33	<3	6.8
Lys 2-5		9.9	0.78	7	<1	1.2	3.9	25	<3	1.6
Lys 3-1	VES	30	1.6	17	<1	0.75	1.2	13	<3	4.9
Lys 3-3		37	4.3	33	<1	1.2	4.6	79	<3	4.6
Lys 3-5		3	0.25	9.9	<1	1.2	1.7	2.9	<3	2
Lys 4-1	VES	8.7	3.7	8.2	<1	1.9	4.2	1.4	<3	17
Lys 4-3		6.1	4.5	10	<1	0.98	1	2.4	<3	18
Lys 4-5		1.8	0.24	9	<1	0.39	2	12	<3	3.4
Lys 5-1	Cement	6.3	0.23	7.2	<1	3	1.7	9.2	<3	7.6
Lys 5-3		13	1.7	27	2.7	5.0	5.8	<1	<3	5.0
Lys 5-5		13	0.28	18	<1	3.3	26	11	<3	6.1

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J. W. McConnell, Jr., R. D. Rogers, J. D. Jastrow[1], D. S. Wickliff Hicks[2], W. E. Sanford[2], R. R. Brey[3], T. M. Sullivan[4], R. M. Neilson, Jr., L. D. Hilton

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Lockheed Idaho Technologies Company
Idaho Falls, ID 83415

[1]Argonne National Laboratory, Argonne, Il
[2]Oak Ridge National Laboratory, Oak Ridge, TN
[3]Formerly with Purdue University
[4]Brookhaven National Laboratory, Upton, NY

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11. ABSTRACT *(200 words or less)*

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