DISTRIBUTION OF GASES IN THE UNSATURATED ZONE AT A LOW-LEVEL RADIOACTIVE-WASTE DISPOSAL SITE NEAR SHEFFIELD, ILLINOIS

By Robert G. Striegl

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CONVERSION FACTORS

Readers who prefer inch-pound or centimeter-gram-second (cgs) units of measurement rather than the International System (SI) units used in this report may use the following conversion factors:

Multiply SI unit	By	To obtain inch-pound or cgs unit
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
gram (g)	0.03527	ounce, avoirdupois (oz)
liter (L)	0.2642	gallon (gal)
kilopascal (kPa)	0.009869	atmosphere (atmos)
	0.1450	pound per square inch (psi)
pascal (Pa)	0.007501	Torr
becquerel (Bq)	27.02	picocurie (pCi)
	2.702×10^{-11}	curie (Ci)

- -

Temperature in degrees Celsius (°C) can be converted to degrees Fahrenheit (°F) as follows:

$$°F = 1.8 \times °C + 32$$

- -

Sea level: In this report "sea level" refers to the National Geodetic Vertical Datum of 1929 (NGVD of 1929)--a geodetic datum derived from a general adjustment of the first-order level nets of both the United States and Canada, formerly called "Mean Sea Level."

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ABSTRACT

The unsaturated zone is a medium that provides pneumatic communication for the movement of gases from wastes buried in landfills to the atmosphere, biota, and ground water. Gases in unsaturated glacial and eolian deposits near a waste-disposal trench at the low-level radioactive-waste disposal site near Sheffield, Bureau County, Illinois, were identified and the spatial and temporal distribution of the partial pressures of those gases were determined for the period January 1984 to January 1986. Methods for the collection and analyses of the gases are described, as are geologic and hydrologic characteristics of the unsaturated zone that affect gas transport. The identified gases, which are of natural and of waste origin, include nitrogen, oxygen, argon, carbon dioxide, methane, propane, butane, tritiated water vapor, ^{14}carbon dioxide, and ^{222}radon. Concentrations of methane and ^{14}carbon dioxide originated at the waste, as shown by partial-pressure gradients of the gases in the unsaturated zone; ^{14}carbon dioxide partial pressures exceeded natural background partial pressures by factors greater than 10^6 at some locations. Variations in partial pressures of oxygen and carbon dioxide were seasonal among piezometers because of increased root and soil-microbe respiration during summer. Variations in methane and ^{14}carbon dioxide partial pressures were apparently related to discrete releases from waste sources at unpredictable intervals of time. No greater-than-background partial pressures for tritiated water vapor or ^{222}radon were measured.

INTRODUCTION

Background

Disposal of low-level radioactive wastes in the unsaturated zone creates a potential for the production of radioactive gases in waste-disposal trenches, and for the subsequent movement of radionuclides to the atmosphere, biota, and ground water. To date, there have been few measurements of gas partial pressures in the unsaturated zone near buried low-level radioactive wastes, and estimates of the transport of gaseous radionuclides from waste-disposal sites have necessarily been based on few data (Lu and Matuszek, 1979; Kunz, 1982). In order to improve the understanding of the behavior of gases near buried wastes and to obtain more extensive data from which gas transport could be estimated, a field investigation was conducted at the low-level radioactive-waste disposal site near Sheffield, Illinois (fig. 1).

Purpose and Scope

This report identifies the gases present in the unsaturated zone near an existing waste-disposal trench and determines the spatial and temporal distributions of the partial pressures of those gases. The report also discusses methods used for the collection and chemical analyses of gases from the unsaturated zone, presents partial pressures of gases collected at the Sheffield site from January 1984 to January 1986, and describes observed changes in gas partial pressures. Physical and chemical properties of the unconsolidated sediments that comprise the unsaturated zone at the study section also are described.

Acknowledgments

Appreciation is extended to Herbert Haas and James Devine of the Institute for the Study of Earth and Man at Southern Methodist University for performing ^{14}carbon analyses; to Frank Markun of Argonne National Laboratories for performing ^{222}radon analyses; and to the Oceanography and Limnology Program at the University of Wisconsin at Madison for providing laboratory and office facilities. Technical input and review from David Armstrong, Department of Civil and Environmental Engineering, University of Wisconsin at Madison is acknowledged.

DESCRIPTION OF THE SHEFFIELD SITE

Physical Setting

The low-level radioactive-waste disposal site is located on 8.1 ha (hectares) of rolling terrain, 5 km (kilometers) southwest of Sheffield in Bureau County, Illinois (fig. 1). The Pennsylvanian bedrock that underlies the site is covered by 3 to 30 m (meters) of unconsolidated glacial and eolian deposits, into which waste-disposal trenches were cut and filled. Eight rock-stratigraphic units that range in composition from silty clay to pebbly sand have been described in the unconsolidated deposits (Foster and Erickson, 1980; Foster, Erickson, and Healy, 1984; Foster, Garklavs, and Mackey, 1984). The average thickness of the unsaturated zone at the site is 12.2 m (Healy and others, 1986); ground-water flow beneath the waste is primarily to the northeast in a pebbly-sand unit (Foster, Erickson, and Healy, 1984).

The climate in northwestern Illinois is continental, having warm summers and cold winters. The mean annual temperature at the site is about 10.3 °C (degrees Celsius), and the mean barometric pressure is about 98,600 Pa (pascals). The mean annual precipitation is 900 mm (millimeters), based on 43 years of record (U.S. Department of Commerce, 1939-82) from nearby weather stations at Kewanee and Hennepin, Illinois. The maximum annual precipitation for that period was 1,330 mm, and the minimum was 650 mm. Severe convective rainstorms are typical during spring and summer (Huff, 1979), and precipitation averages 80 mm per month during April through July. Late fall and winter are driest; precipitation averages 50 mm per month during November through

March (Healy and others, 1986). Of a mean annual precipitation of 940 mm during 1982-84, it was estimated that 660 mm evapotranspired, 160 mm ran off, and 120 mm recharged the aquifer or was stored in the unsaturated zone (Healy and others, 1987).

History of Site Operation

Low-level radioactive wastes were buried at the Sheffield site from August 1967 to April 1978, during which time 21 trenches (fig. 2) were filled with about 90,000 m^3 (cubic meters) of material that contained about $2.22x10^{15}$ becquerels (60,000 curies) of activity (Illinois Department of Nuclear Safety, written commun., 1979). The exact contents of the burial trenches are unknown, and inventories of disposed waste are descriptively poor and quantitatively inaccurate (Kahle and Rowlands, 1981). Buried wastes include resins, carcasses of laboratory animals, solidified liquid wastes, glassware, building materials, clothing, containerized gases, paper, building and construction materials, and cleanup materials (Foster, Erickson, and Healy, 1984). Waste-containment vessels include steel drums, wooden crates, plastic containers, concrete casks, and cardboard cartons.

Nineteen of the trenches were excavated in undisturbed material, and two trenches were constructed above the existing land surface. All trenches, with the possible exception of trench 18 (fig. 2), were completed in the unsaturated zone. In early years of site operation, waste was randomly placed in trenches; in later years, waste was stacked in the trenches in an orderly fashion (Mark Bower, US Ecology, Inc., oral commun., 1987). Waste was covered with about 0.3 m of compacted clayey silt, on which about 0.5 m of fill material was mounded. Trench caps were smoothly contoured and seeded with grasses and clover (Healy and others, 1986).

Of the 21 trenches, trench 2 is located closest to the area studied. The excavated dimensions of trench 2 are 140 m long by 18.3 m wide by 7.6 m deep; the trench was filled from the west to the east during August 1968 to March 1971 (fig. 3; Kahle and Rowlands, 1981). When completed, the volume of trench 2 was 19,500 m^3, and the trench contained about 12,950 m^3 of fill and 6,550 m^3 of waste. Radioactive waste in trench 2 had a total activity of $3.87x10^{14}$ becquerels (10,450 curies), a mass of 17,110 kg (kilograms), and included 7,725 g (grams) of ^{235}uranium and 4,970 g of ^{239}plutonium (Illinois Department of Nuclear Safety, written commun., 1979).

DESCRIPTION OF THE STUDY SECTION

Location and Installation of Boreholes

The geologic section studied is located along a 46 m line that runs east to west from the approximate center of, and perpendicular to, the east wall of trench 2 (fig. 2). Three boreholes (A, B, and C) were augered to bedrock using a 153-mm-diameter hollow-stemmed auger. No drilling fluids were used.

Continuous core samples were collected through the auger stem using a 51-mm-diameter steel push tube in fine-grained deposits, and a 25-mm-diameter split-spoon sampler in coarse-grained deposits. Depending on the texture of the unconsolidated sediments, the coring devices were driven between 250 and 900 mm beyond the auger stem, then withdrawn. The cored hole was then augered to the depth of the collected sample and the process was repeated. Cores were sealed from the atmosphere and reserved for physical and chemical analyses.

Boreholes A, B, and C were located at respective distances of 12, 29, and 46 m from the east endwall monument of trench 2. Borehole A was located immediately east of the disposal-site boundary, as close to the boundary fence as possible (augering was not allowed within the site boundary). Boreholes B and C were augered at respective locations of 17 m and 34 m to the east of borehole A. The land surface slopes downward to a road cut approximately 6 m east of borehole C.

Stratigraphy and Physical Properties of Geologic Deposits

At the study section, weathered deposits of Pennsylvanian-age shale and coal of the Carbondale Formation are overlain by four Pleistocene glacial and eolian units having nearly parallel contacts, and a total thickness of about 17 m (fig. 4). The oldest unit is the Toulon Member of the Glasford Formation (Willman and Frye, 1970), which is followed in ascending sequence by the Radnor Till Member of the Glasford Formation, Roxana Silt, and Peoria Loess. Peoria Loess is developed into a modern soil near the land surface. Characteristics of the bedrock and the unconsolidated sediments at the site are discussed at length in Foster and Erickson (1980); Foster, Garklavs, and Mackey (1984); and Foster, Erickson, and Healy (1984).

Movements of gases in the unsaturated zone are affected by physical and chemical properties of the unsaturated-zone sediments. Table 1 summarizes the particle sizes, bulk densities, air- and water-filled porosities, surface areas, and carbonate contents of core samples collected at boreholes A, B, and C. Montmorillonites comprised the largest fraction of clay minerals in the Peoria Loess, Roxana Silt, and Radnor Till Member, followed in abundance by illites, and kaolinites plus chlorites. Illites comprised the largest fraction of clay minerals in the Toulon Member, followed in abundance by kaolinites plus chlorites, and montmorillonites.

FIELD PROCEDURES FOR GAS COLLECTION

Installation of Gas Piezometers

Five gas piezometers were nested in each borehole at depths of 1.8 to 17 m below land surface (fig. 5). The piezometers, which were constructed of 13-mm-diameter polyvinylchloride pipe, were slotted with 0.8-mm-thick slots that were spaced about 5 mm apart along the deepest 0.5 m of pipe. Within several hours after augering, each borehole was backfilled with washed silica sand to the desired altitude of the lowest piezometer screen. The piezometer

Table 1.--Physical and chemical properties of glacial and eolian deposits in the unsaturated zone

[g/cm^3, grams per cubic centimeter; m^2/g, square meters per gram; µm, micrometers. Dashes indicate no analysis performed]

	Particle size[1] (percent by weight)			Bulk density[2] (g/cm^3)	Total porosity[3] (dimensionless)	Air-filled porosity[3] (dimensionless)	Water-filled porosity[3] (dimensionless)	Surface area[4] (m^2/g)	Carbonate fraction[5] (by weight)	Calcite/dolomite ratio[6] (of carbonate fraction)
	Sand >62 µm	Silt 4-62 µm	Clay <4 µm							
Peoria Loess	4	81	15	1.50	0.43	0.30	0.13	9.82	0.28	0.14
Roxana Silt	2	81	17	1.50	.43	.30	.13	--	.04	.11
Radnor Till Member (Glasford Formation)	18	53	29	2.15	.32	.02	.30	26.4	.05	.12
Toulon Member (Glasford Formation)	84	10	6	1.60	.35	.29	.06	2.14	.15	.13

[1] Pipette analysis (Day, 1965).

[2] Core method (Blake, 1965).

[3] Mercury porisimeter (Vomocil, 1965).

[4] Dynamic nitrogen adsorption (E. Busenberg, U.S. Geological Survey, Water Resources Division, written commun., 1986).

[5] Gravimetric loss by selective dissolution in hydrochloric acid.

[6] Chittick analysis (Dreimanis, 1962).

was then installed, and the annulus was backfilled with silica sand around the screen to an altitude about 0.5 m above the screen. About 0.3 m of bentonite, 0.5 m of portland cement grout, and another 0.3 m of bentonite were backfilled over the sand. The bentonite absorbed excess water from the cement grout, and the cement sealed possible pathways for pneumatic communication between piezometers. To avoid bridging of backfill materials within the boreholes and contamination of borehole sidewalls with grout and bentonite, all backfill materials were funneled through 76-mm-diameter polyvinylchloride pipes to the altitudes at which they were deposited. Each borehole was backfilled with parent material, which was screened through a 2-mm mesh, to about 0.5 m below the desired altitude of the next piezometer. The sequence was repeated for each piezometer installation. The uppermost 1 m of each borehole was filled with portland cement grout.

With the exception of the Toulon Member and the Peoria Loess, gas piezometers were screened near the upper boundary of each lithologic unit (fig. 4). Two piezometers were screened in the Toulon Member at each borehole--one near the top of the unit and the second at the contact between the Toulon Member and the Carbondale Formation. Piezometer screens were placed 1.8 m below the land surface in the Peoria Loess. Altitudes of the land surface, the midpoints of piezometer screens, lithologic contacts, and the termini of augering at boreholes A, B, and C are listed in table 2. The screens of piezometers B1 and C1 apparently became clogged soon after installation, possibly because of a rise in the water table. Because of the clogging, it was possible to collect only one sample from piezometer B1; no samples were collected from piezometer C1.

The boreholes were augered on December 6, 1983, and piezometer installation was completed on December 8, 1983. The piezometers were sealed from the atmosphere and allowed to equilibrate with soil gases until January 19, 1984, when the first samples were collected. The piezometers were kept sealed from the atmosphere throughout the study.

Collection of Gas Samples

Gas samples were grouped into four categories, each requiring a unique sampling procedure: (1) nonradioactive gases, including nitrogen (N_2), oxygen plus argon (O_2+Ar), carbon dioxide (CO_2), methane (CH_4), ethane (C_2H_6), propane (C_3H_8), and n- and para-butane (C_4H_{10}); (2) tritiated water vapor ($^3HOH_{(v)}$); (3) ^{14}carbon dioxide ($^{14}CO_2$); and (4) ^{222}radon (^{222}Rn).

All samples were collected using peristaltic pumps connected to the gas piezometers by silicone-rubber tubing. Flow rates were set at 600 mL (milliliters) per minute. The relatively slow flow rate was maintained so that a minimum of pneumatic disturbance would be imparted to the unsaturated zone around the piezometer screens. The primary mechanism for gas movement in the unsaturated zone is considered to be ordinary diffusion (Keen, 1931; Evans, 1965; Hillel, 1980; Weeks and others, 1982; Thorstenson and others, 1983), so it is important to not create convection currents by use of excessive pumping rates or by sampling too frequently. Prior to collecting samples, at least 20 piezometer-volumes of gas were pumped from each piezometer to make certain that the samples collected represented the gas in the formation.

Table 2.--Altitudes of the land surface, the midpoints of piezometer screens, lithologic contacts, and termini of boreholes A, B, and C, in meters above sea level

	Borehole A	Borehole B	Borehole C
Land surface	238.50	238.30	237.60
Piezometer screen, depth 5	236.70	236.50	235.75
Peoria Loess - Roxana Silt contact	235.45	236.00	235.30
Piezometer screen, depth 4	234.80	236.60	233.90
Roxana Silt-Radnor Till Member contact	232.10	231.60	231.50
Piezometer screen, depth 3	231.20	231.00	230.30
Radnor Till Member-Toulon Member contact	227.85	227.95	227.85
Piezometer screen, depth 2	226.90	226.75	226.05
Piezometer screen, depth 1[1]	224.90	221.45	220.85
Terminus of borehole	224.70	221.25	220.65

[1] The midpoints of the piezometer screens at depth 1 are at the contact of the Toulon Member of the Glasford Formation and the underlying Carbondale Formation.

Nonradioactive Gases

Samples of nonradioactive gases were collected downstream of the peristaltic pump into 300-mL glass gas-sampling bulbs. The bulbs had a flow-through design, and each was fitted with a rubber septum for subsampling with a syringe. Upon connecting a sampling bulb to the peristaltic pump, the bulb was left in a flow-through position for at least 20 minutes. The downstream seal was then closed, followed by closure of the seal nearest the pump. The collected sample was therefore slightly overpressured within the bulb. Overpressuring the sample allowed the laboratory analyst opportunity to ascertain that the seals and septa did not leak prior to analysis. Samples were discarded if the pressure was lost, and when possible, replacement samples were collected.

Tritiated Water Vapor

Samples for $^{3}HOH_{(v)}$ and $^{14}CO_2$ analyses were collected simultaneously using the sample-collection system shown in figure 6. In that system, unsaturated-zone gases were pulled from the piezometer through a freeze trap that was immersed in a slurry of solid CO_2 (dry ice) and methanol. Gases were cooled in the freeze trap, and water vapor was frozen as ice that accumulated on the inside of the trap. A column containing color indicating silica gel was placed downstream of the freeze trap to monitor its efficiency for trapping the water vapor. Upon completion of sample collection, the accumulated ice was thawed, the volume of collected water was measured, and the sample was transferred to a scintillation vial for tritium analysis.

^{14}Carbon Dioxide

Following removal of the water vapor by the freeze trap, the remaining gases were pulled through a 500-mL gas-wash bottle that contained 300 mL of 5 molar potassium hydroxide (KOH) solution. When the gases bubble through the KOH solution, CO_2 reacts with the KOH to form aqueous potassium bicarbonate complex ($KHCO_{3(aq)}$) and remains in solution. Haas and others (1983) found this sampling procedure was at least 99.9 percent efficient for removing CO_2 from a gas mixture, and that it caused no isotopic fractionation of the sample. At least 0.25 mole (6 standard liters) of CO_2 was collected for each sample. Sampling time, which depended on the partial pressure of CO_2 in the unsaturated zone, ranged from 5 to 9 hours. After adequate sampling time had elapsed, sample-collection systems were closed from the atmosphere and from the piezometer, and the samples were transported to the laboratory in the gas-wash bottles in which they were collected.

^{222}Radon

Samples for ^{222}Rn analyses were collected into evacuated steel cannisters that were provided by Argonne National Laboratory. Like the nonradioactive gas samples, the ^{222}Rn samples were overpressured upon collection. Since the cannisters were single ended, flow-through sampling was not possible. Sample integrity was, therefore, dependent on a vacuum being maintained in the evacuated cannisters prior to sampling. Partial loss of vacuum could result in dilution of samples by atmospheric air. If partial loss of vacuum was suspected, suspect cannisters were not used.

LABORATORY ANALYSES OF GAS SAMPLES

Nonradioactive Gases

Nonradioactive gases (N_2, O_2+AR, CO_2, CH_4, C_2H_6, C_3H_8, and C_4H_{10}) were analyzed by the U.S. Geological Survey, Water Resources Division, Branch of Analytical Services, Arvada, Colorado. The gases were subsampled by syringe through the septa in the gas bulbs in which they were collected and injected into a gas chromatograph having a thermal conductivity detector. The gases were separated on a 2.1-mm inside diameter by 4-m long stainless-steel column packed with chromosorb 102. The chromatograph was programmed to hold the sample at -80 °C for 3 minutes, increase the sample temperature at the rate of 30 °C per minute until reaching 30 °C, increase the sample temperature at the rate of 10 °C per minute from 30 °C to 150 °C, and hold at 150 °C for 25 minutes. Argon was not separable from oxygen under the analytical conditions. Laboratory precision was 0.1 percent for N_2 and O_2+Ar, 0.01 percent for CO_2, and 0.1 ppm (parts per million) for CH_4, and C_3H_8 at concentrations greater than the detection of 0.5 ppm.

Tritiated Water Vapor

Because the volume of condensed water-vapor samples was small (generally less than 2 mL), samples were diluted to 10 mL with distilled water before adding the appropriate amount of scintillation cocktail. Samples were counted according to liquid-scintillation counting methods described by Thatcher and others (1977) at the U.S. Geological Survey, Water Resources Division, Illinois District laboratory at Urbana, Illinois. Detection limits were around 7.40 becquerels per liter of water. No samples exceeded the detection limits.

^{14}Carbon Dioxide

Samples for $^{14}CO_2$ analyses were stored as $KHCO_{3(aq)}$ in the gas-wash bottles in which they were collected until they could be hydrolyzed back to CO_2. Because all samples were counted at the radiocarbon-dating laboratory at the Institute for the Study of Earth and Man at Southern Methodist University, it was necessary to dilute the samples to below-modern ^{14}C activities to avoid contamination of the ^{14}C counting systems.

Samples were hydrolyzed and diluted using the vacuum manifold shown in figure 7. A gas-wash bottle holding the sample was connected to the manifold, and cold, 10 molar hydrochloric acid was dripped into the $KOH\text{-}KHCO_{3(aq)}$ solution. The resulting hydrolytic reaction yielded water vapor and CO_2, which were cryogenically separated in a series of three freeze traps. The first two traps were immersed in a solid CO_2-methanol slurry that froze the water vapor from the sample; the third trap was immersed in liquid nitrogen, which froze the CO_2 sample. Upon completion of the hydrolysis of a sample, a stopcock between the second and third freeze trap was closed and the solid CO_2 was allowed to warm and expand into the upper portion of the manifold. The amount of the CO_2 gas was determined by expanding the gas into a known volume and measuring the pressure. An aliquot of the sample was then diluted in the manifold using pure CO_2 that had negligible ^{14}C activity as a dilutant. Complete mixing of the sample and the dilution gas was insured by freezing and reexpanding the mixture several times. The diluted and mixed sample was cryogenically pumped into a stainless-steel cylinder and shipped to the radiocarbon laboratory at the Institute for the Study of Earth and Man, Southern Methodist University. At the laboratory, the sample was reexpanded, converted to benzene, and analyzed by liquid-scintillation counting techniques (Herbert Haas, Institute for the Study of Earth and Man, Southern Methodist University, oral commun., 1984; Thatcher and others, 1977).

^{222}Radon

Radon samples were express shipped to Argonne National Laboratory on the day they were collected. Samples were counted in silver-activated zinc-sulfide coated counting chambers using alpha-scintillation counting techniques (Lucas, 1977). Samples were usually counted within one ^{222}Rn half-life (3.82 days) after collection; measured activities were corrected to account for radioactive decay that had occurred since the time of collection.

SOURCES OF ERROR IN GAS PARTIAL PRESSURES

Reported gas partial pressures include uncertainties that result from sampling error, analytical error, and other possible errors including possible contamination of backfill or boreholes. Sampling error results from the failure of a field procedure to accurately collect a representative sample of gases from the unsaturated zone or from the failure of a sample-holding procedure to maintain the collected sample in an unchanged state until laboratory analyses are performed. Analytical errors include the inaccuracy that results from the laboratory preparation of samples for measurement by an analytical instrument and the inaccuracy of the laboratory measurement itself. Accidental contamination of backfill or boreholes could alter partial pressures of specific gases found at affected locations.

Sampling errors had the potential for being largest, and, because of that potential, were commonly the easiest to recognize when screening data for accuracy. The potential for large sampling errors was greatest for samples that were collected instantaneously and that were held as gases until laboratory analyses were performed. These included samples collected for determination of nonradioactive gases and for ^{222}Rn. Three potential causes for sampling error were recognized for those samples: (1) Samples were collected at a time when they did not actually represent the gases in the unsaturated zone near the location of the piezometer screen; (2) leakage of the sample container occurred after the sample was collected and resulted in loss or contamination of the sample; and (3) leakage of the sample container occurred prior to collection of the sample and resulted in contamination of the sample.

For the first case, the sample analyzed may represent some admixture of gases from within the piezometer pipe volume, air that may have been introduced to the system when the sampling pump was being connected, and gases from the unsaturated zone near the piezometer screen. In order to evaluate the volume of gases that needed to be pumped from a piezometer before a representative sample could be collected, two time series of ^{222}Rn samples were collected at piezometers B4 and C4 on December 1, 1984 (fig. 8). Graphs of the ^{222}Rn activities of those samples versus the number of piezometer volumes of gas pumped show that activities stabilized after about 20 piezometer volumes were pumped.

The second cause of sample error occurred when samples were collected when outside temperatures were as low as -25 °C. At those temperatures, o'rings and stopcocks on the sampling bulbs commonly leaked, causing loss of sample. Because of this, not all February 1985 samples were successfully collected. On occasion, leakage or breakage was caused by routine sample handling. So that leaks could easily be detected by the laboratory analyst, samples of nonradioactive gases and ^{222}Rn were collected under a small overpressure. In instances where samples leaked to ambient pressures, those samples were not analyzed. Replicates of leaked samples were collected when possible.

Because ^{222}Rn samples were collected in single-ended evacuated cannisters, it was possible that some air could leak into the sample cannisters between the time of evacuation and the time of sample collection causing some dilution of the final sample. When leakage of air into evacuated cannisters was suspected, the suspect cannisters were not used for sample collection. However, since the only criterion that could be used in the field to check for leaks was the audible presence or absence of a vacuum when the cannister was opened to the pump line, it is assumed that some atmospheric dilution of samples occasionally occurred. Activities of samples that indicated obvious leaks were excluded from the final data.

Samples for $^{14}CO_2$ and $^{3}HOH_{(v)}$ analyses required long pumping times that integrated over any instananeous aberrations that may have caused concern for nonradioactive gas and ^{222}Rn samples. However, since the intended use of $^{14}CO_2$ and $^{3}HOH_{(v)}$ samples was for isotopic analyses, it was desired that those samples be collected quantitatively so that no isotopic fractionation would occur. Fractionation could cause a bias toward the heavier isotopes in the samples. Haas and others (1983) demonstrated that the collection procedure for the $^{14}CO_2$ samples was quantitatively accurate. In order to insure that the $^{3}HOH_{(v)}$ trap was quantitatively efficient, color-indicating silica gel was placed in-line downstream of the $^{3}HOH_{(v)}$ trap. No changes in the color-indicating gel were observed in the field.

Analytical error was reported by contract laboratories for ^{222}Rn and ^{14}C activities. Error associated with ^{222}Rn counting ranged from 1.07 to 6.16 percent, had a median of 2.08 percent, a mean of 2.58 percent, and a standard deviation of 0.96 percent (Frank Markun, Argonne National Laboratory, written commun., 1984-86). Errors were greatest for samples with low activities.

Error associated with benzene conversion and liquid-scintillation counting of $^{14}CO_2$ ranged from 0.32 to 26.4 percent, had a median of 1.99 percent, a mean of 3.34 percent, and a standard deviation of 4.78 percent (Herbert Haas, Institute for the Study of Earth and Man, Southern Methodist University, written communs., 1984-86). Excluding six samples that had errors that exceeded 8.9 percent, the analytical error for $^{14}CO_2$ had a median of 1.35 percent, a mean of 1.89 percent, and a standard deviation of 1.37 percent. As with ^{222}Rn, the largest errors were associated with low activities.

The net sampling and analytical error for nonradioactive gases and for ^{222}Rn was evaluated by determination of the mean of absolute differences between replicate samples, the standard deviation of absolute differences between replicate samples, and the relative error. The relative error was calculated as twice the sum of the absolute differences divided by the sum of all paired measurements. Table 3 summarizes those statistics for 18 pairs of nonradioactive gas samples and for 13 pairs of ^{222}Rn samples.

Table 3.--Means of absolute differences, standard deviations of absolute differences, and mean relative errors determined from analyses of replicate pairs of nonradioactive gases and ^{222}radon

[Pa, pascals]

	N_2	O_2+Ar	CO_2	CH_4	^{222}Rn
Mean of absolute differences	89 Pa	89 Pa	37 Pa	0.030 Pa	1.49 becquerels
Standard deviation of absolute differences	81 Pa	87 Pa	53 Pa	0.031 Pa	1.13 becquerels
Mean relative error, in percent	0.1	0.5	1.3	5.2	2.8
Number of replicate pairs	18	18	18	18	13

Because the activities of $^{14}CO_2$ samples were unexpectedly high, all samples required dilution with CO_2 having negligible ^{14}C activity before they could be submitted to the radiocarbon laboratory at Southern Methodist University for counting. Dilution ratios ranged from 1:10 to about 1:10,000, with the more radioactive samples from boreholes A and B requiring a two-step dilution. Because of limitations in sampling equipment and funds, it was not possible to collect replicate $^{14}CO_2$ samples to evaluate the relative error associated with sample collection and analyses. However, because piezometers A1 and A2 were both screened in the Toulon Member of the Glasford Formation, it was possible to infer the relative error of $^{14}CO_2$ measurements by evaluating the relative differences between partial pressures at those piezometers. Table 4 lists the mean of absolute differences between nine pairs of nonradioactive gas samples and six pairs of $^{14}CO_2$ samples that were collected on the same days at piezometers A1 and A2. The relative differences for N_2, O_2+Ar, CO_2, and CH_4 ranged from 0.2 to 6.1 percent; these were similar to the relative errors listed for those gases in table 3. The relative difference for $^{14}CO_2$ partial pressures between A1 and A2 was 21.2 percent. It is thought that 21.1 percent approximates the upper limit for the sum of $^{14}CO_2$ sampling error plus analytical error because it is calculated for samples collected at two close but different locations, and because all of the samples that were compared required two-step dilutions in the laboratory.

Table 4.--Means of absolute differences, standard deviations of absolute differences, and mean relative differences between gas partial pressures at piezometers A1 and A2 for nonradioactive gases and $^{14}CO_2$

	N_2	O_2+Ar	CO_2	CH_4	$^{14}CO_2$
Mean of absolute differences, in pascals	67	100	70	0.087	3.73×10^{-6}
Standard deviation of absolute differences, in pascals	67	118	90	0.179	6.13×10^{-6}
Mean relative difference, in percent	0.2	0.7	1.9	6.1	21.1
Number of pairs	9	9	9	9	6

Calculated partial pressures of $^{14}CO_2$ are a product of the ^{14}C activity of the CO_2 that was measured by the radiocarbon-dating laboratory and the partial pressure of the CO_2 determined from analysis of the nonradioactive gas sample. Therefore, any errors in CO_2 partial-pressure measurements are inherent in the calculated $^{14}CO_2$ partial pressures. Where CO_2 samples leaked, it was necessary to calculate $^{14}CO_2$ partial pressures using the time-averaged mean for CO_2 at that location. Those calculations are in error by a factor that is equal to the absolute difference between the quotient of the true CO_2 partial pressure divided by the time-averaged mean CO_2 partial pressure and unity.

It is suspected that the silica sand that was used for backfill at piezometer A3 was premixed with lime. Coincidentally, ^{222}radon activities were unusually high, and methane partial pressures were unusually low at A3 relative to patterns in concentrations that were observed at surrounding piezometers. Radium sulfates commonly coprecipitate with lime during sediment deposition and could be a source for the high ^{222}Rn activities (J. Otton, U.S. Geological Survey, Geologic Division, oral commun., 1987). Consequently, ^{222}Rn activities for A3 were eliminated from the data set. It is not known if methane is preferentially adsorbed or in any other way affected by lime. Methane partial pressures for A3 are, therefore, presented with the caution that they may not accurately represent the location from which they were collected. Contamination of boreholes or backfill was suspected only at piezometer A3.

SPATIAL AND TEMPORAL DISTRIBUTIONS OF GAS PARTIAL PRESSURES

With the exception of ^{222}Rn, which is presented in units of radioactivity per standard liter of gas sample, gas concentrations are presented in units of pressure. On days when samples were collected, barometric pressure at the study section ranged from 97,600 to 99,200 Pa (pascals) and averaged 98,300 Pa. The average barometric pressure during the period of study was 98,600 Pa. Measurements of the differential between barometric pressure at the land surface and barometric pressure at the depths of the piezometer screens showed no measurable differences in pressure over the long term. For the sake of convenience and clarity, all partial pressures are presented as products of their measured volume fractions and of the average barometric pressure during the study period (98,600 Pa). Where listed, means of all partial pressures and of ^{222}Rn activity are presented as time-averaged means that are weighted to account for unequal periods of time between collection of samples.

Nonradioactive Gases

The gases that are collectively referred to here as nonradioactive gases comprise 0.999972 of the lower atmosphere where they are present in volume fractions of 0.78084 for N_2, 0.20946 for O_2, 0.00934 for Ar, 0.00033 for CO_2, and less than 0.000002 for CH_4 (Moran and Morgan, 1986). For comparison, average volume fractions of seven air samples collected at 0.3 m above the ground at borehole A from March 23, 1984, to October 30, 1985, were 0.7909 for N_2, 0.2089 for O_2+Ar, less than 0.0004 for CO_2, and 0.0000019 for CH_4.

Nitrogen

The lower atmosphere is the source for N_2 in the unsaturated zone, where it behaves as a stagnant gas, its partial pressures changing conservatively in response to seasonal changes in other gases, primarily O_2 and CO_2. Partial pressures of N_2 measured in the study section are listed in table 5, and their time-averaged means are listed in table 6. The partial pressures of N_2 versus time are shown in figures 9-13, and time-averaged mean N_2 partial pressures versus depth are shown in figure 14. Because the sum of the mean O_2 plus CO_2 partial pressures generally decrease with depth below the land surface, N_2 partial pressures generally increase with depth (fig. 14). Partial pressures of N_2 at 1.8 m below the land surface were similar at all boreholes and were seasonally cyclic, having early spring peaks, late summer troughs, and a range of about 4,000 Pa (fig. 9). Similar seasonal cycles having less amplitude were observed at 3.7 m below the land surface (fig. 10), and partial pressures were similar between boreholes at that depth. In the Radnor Till Member of the Glasford Formation, at 7.3 m below the land surface, seasonal cycles were also observed but were more pronounced at borehole A (figs. 11 and 14) than at boreholes B and C (fig. 11). As distance from a source of seasonality increased (either vertically from the root zone in the upper piezometers or horizontally from the trench in the Toulon Member of the Glasford Formation), cycles were damped in amplitude and were phase-shifted in time.

The observed patterns in partial pressures of N_2 infer three characteristics about the unsaturated media. First, connected pore spaces in the Peoria Loess and the Roxana Silt are sufficient to allow uniform horizontal communication of gases within those units, and vertical communication of gases between the units and the atmosphere. Second, the low air-filled porosity and poor interconnection between pore spaces in the Radnor Till Member allows for heterogeneity in the distribution of gases within the unit and impedes the movement of gases from overlying units. Finally, removal of the natural geologic materials during trench construction, and replacement of those materials with waste having high air-filled porosity, has allowed for vertical mixing of gases in the trenches. Because of that mixing, seasonal cycles of N_2 partial pressures that occurred within the trenches were observed at piezometers in the Toulon Member, which has a high air-filled porosity and good interconnection between pores.

Oxygen plus Argon

The atmosphere is also the primary source for O_2+Ar in the unsaturated zone. The partial pressures of O_2+Ar generally decrease with depth below the land surface as oxygen is consumed by root and soil-microbe respiration and by mineral oxidation (tables 5 and 6, figs. 15-20). Although the gas chromatography column and detector used to analyze gas samples did not separate O_2 from Ar, it was assumed that the partial pressure of the inert gas Ar was essentially constant at about 920 Pa. Observed changes in the sum of O_2+Ar partial pressures were, therefore, attributed to change in O_2 partial pressure. Partial pressures of O_2 at depths of 1.8 and 3.6 m were seasonally cyclic, having peaks in fall and troughs in summer (figs. 15 and 16). Seasonality was better defined at the 3.6 m depth than at the 1.8 m depth, probably because the shallower depth was more affected by local disturbances in barometric pressure and water content and by local differences in respiration rates. As with N_2 partial pressures, the O_2+Ar partial pressures were least similar between boreholes in the Radnor Till Member (fig. 17). Seasonal influences of the trench on O_2+Ar partial pressures in the Toulon Member were similar to those observed for N_2 partial pressures (figs. 18 and 19).

Table 5.--Partial pressures of N_2, O_2+Ar, CO_2, and CH_4 at boreholes A, B, and C, in pascals

[m, meters]

Date	N_2	O_2+Ar	CO_2	CH_4
	(1) Piezometer A5, 1.8 m below the land surface			
03-20-84	79,800	16,300	2,510	0.14
06-08-84	78,900	16,100	3,570	.16
08-16-84	76,300	17,100	5,200	.14
10-26-84	77,700	17,700	3,180	<.05
02-21-85	78,000	19,500	1,050	.09
06-12-85	76,900	18,900	2,760	.06
09-20-85	77,100	19,700	1,770	<.05
10-30-85	77,600	19,000	1,980	<.05
10-30-85	77,700	18,900	1,970	<.05
01-15-86	78,600	17,900	2,050	<.05
	(2) Piezometer A4, 3.7 m below the land surface			
01-20-84	79,100	17,500	2,070	0.12
03-20-84	79,200	16,800	2,630	.18
06-08-84	78,500	18,800	1,310	.29
08-16-84	77,700	19,500	1,410	.16
10-26-84	77,500	19,800	1,280	.10
06-12-85	78,100	17,600	2,930	.23
08-29-85	77,000	18,200	3,410	<.05
09-20-85	76,700	18,500	3,350	.05
10-30-85	77,300	18,100	3,170	<.05
01-15-86	78,600	17,400	2,600	.36
	(3) Piezometer A3, 7.3 m below the land surface			
03-20-84	79,300	16,800	2,540	0.71
06-05-84	79,000	15,900	3,680	.61
06-08-84	79,200	15,900	3,560	.63
08-16-84	78,700	15,700	4,170	.33
10-26-84	78,800	16,200	3,620	.19
02-21-85	78,200	18,800	1,540	.41
02-21-85	78,400	18,700	1,470	.48
06-12-85	78,300	16,500	3,790	.74
06-12-85	78,300	16,500	3,790	.74
08-29-85	77,700	17,100	3,820	.35
10-30-85	77,600	17,000	4,030	.32
01-15-86	79,100	15,700	3,770	.72
01-15-86	79,100	15,600	3,880	.73

Table 5.--Partial pressures of N_2, O_2+Ar, CO_2, and CH_4 at boreholes A, B, and C, in pascals--Continued

Date	N_2	O_2+Ar	CO_2	CH_4
(4) Piezometer A2, 11.6 m below the land surface				
01-19-84	80,400	14,100	4,140	1.54
03-20-84	79,400	15,500	3,690	.97
06-05-84	78,900	15,700	3,960	1.08
06-08-84	78,900	15,700	4,000	.99
08-16-84	79,300	14,900	4,390	1.38
08-16-84	79,400	14,800	4,400	1.38
10-26-84	90,500	14,300	3,820	1.38
10-26-84	80,400	14,400	3,840	1.28
02-21-85	79,100	15,600	3,870	1.77
06-12-85	79,500	15,300	3,800	2.76
08-29-85	80,100	14,700	3,830	1.68
10-30-85	80,300	14,400	3,910	1.18
01-15-86	79,700	15,200	3,720	.84
(5) Piezometer A1, 13.6 m below the land surface				
01-19-84	80,400	14,100	4,120	1.55
03-20-84	79,500	15,100	3,970	1.08
06-08-84	79,100	15,700	3,800	.99
08-16-84	79,300	14,900	4,390	1.38
10-26-84	80,400	14,600	3,660	1.18
06-12-85	79,500	15,400	3,760	2.76
08-29-85	80,000	14,800	3,830	1.77
10-30-85	80,200	14,500	3,950	1.18
01-15-86	79,800	15,200	3,690	.84
(6) Piezometer B5, 1.8 m below the land surface				
03-21-84	79,600	18,000	1,030	<0.05
06-08-84	79,300	18,100	1,140	.09
08-16-84	76,700	18,800	3,090	.15
10-26-26	77,700	19,600	1,320	.10
02-21-85	78,000	18,900	1,720	<.05
06-11-85	77,100	19,000	2,470	<.05
08-28-85	77,200	19,800	1,620	.06
09-20-85	76,900	20,000	1,710	<.05
10-30-85	77,300	19,300	2,010	<.05
01-15-86	78,200	18,300	2,040	.06

Table 5.--Partial pressures of N_2, O_2+Ar, CO_2, and CH_4 at boreholes A, B, and C, in pascals--Continued

Date	N_2	O_2+Ar	CO_2	CH_4
	(7) Piezometer B4, 3.7 m below the land surface			
01-20-84	78,400	18,400	1,810	0.07
03-21-84	79,100	17,600	1,940	<.05
06-08-84	78,600	17,300	2,760	.06
08-16-84	77,400	17,400	3,770	<.05
10-26-84	77,000	18,200	3,430	.18
02-21-85	78,000	18,300	2,260	<.05
06-11-85	77,900	18,000	2,700	<.05
08-28-85	76,800	18,700	3,100	.06
09-20-85	76,500	19,100	2,990	<.05
10-30-85	77,300	18,700	2,630	<.05
01-15-86	78,400	18,000	2,220	.14
	(8) Piezometer B3, 7.3 m below the land surface			
03-21-84	80,800	15,100	2,700	0.63
06-08-84	79,400	16,600	2,660	.49
08-16-84	79,300	17,200	2,150	.43
10-26-84	79,500	15,700	3,460	.58
02-21-85	79,800	15,300	3,560	.56
06-11-85	79,500	15,500	3,660	.97
08-28-85	79,400	15,700	3,520	.89
10-30-85	79,400	15,600	3,640	.80
01-15-86	79,600	15,400	3,660	.68
	(9) Piezometer B2, 11.6 m below the land surface			
01-19-84	79,800	15,300	3,540	0.71
03-21-84	79,900	14,900	3,800	.62
03-21-84	80,000	14,900	3,760	.72
06-08-84	79,000	16,500	3,090	.52
08-16-84	79,400	15,600	3,580	.57
10-26-84	79,300	15,500	3,790	.67
06-11-85	79,400	15,500	3,740	.98
08-28-85	79,500	15,500	3,640	.91
10-30-85	79,500	15,400	3,710	.82
01-15-86	79,600	15,300	3,730	.65

Table 5.--Partial pressures of N_2, O_2+Ar, CO_2, and CH_4 at boreholes A, B, and C, in pascals--Continued

Date	N_2	O_2+Ar	CO_2	CH_4
(10) Piezometer B1, 16.8 m below the land surface				
03-21-84	80,100	16,100	2,460	0.99
(11) Piezometer C5, 1.8 m below the land surface				
06-08-84	78,500	17,500	2,610	0.08
08-14-84	74,800	19,400	4,410	.12
10-25-84	77,600	19,200	1,770	.08
02-20-85	77,600	19,700	1,290	<.05
06-11-85	77,600	19,800	1,230	.06
08-28-85	77,300	20,200	1,070	.10
09-20-85	77,100	20,300	1,200	<.05
10-30-85	77,300	20,000	1,300	.06
(12) Piezometer C4, 3.7 m below the land surface				
01-20-84	77,400	19,600	1,620	0.07
03-22-84	78,000	18,900	1,640	<.05
06-08-84	78,600	17,700	2,260	.14
08-14-84	77,300	19,700	1,550	.18
10-25-84	77,400	20,000	1,190	.06
06-11-85	78,000	18,300	2,310	.15
08-28-85	76,800	19,100	2,720	<.05
09-20-85	76,700	19,400	2,470	<.05
10-30-85	77,300	19,200	2,100	<.05
01-15-86	77,900	19,500	1,210	<.05
(13) Piezometer C3, 7.3 m below the land surface				
03-22-84	79,500	15,600	3,490	0.25
06-08-84	80,200	15,100	3,360	.28
08-14-84	78,500	16,200	3,870	.31
10-25-84	79,500	15,500	3,660	.34
02-20-85	79,300	16,100	3,250	.28
06-11-85	79,300	15,500	3,800	.36
08-28-85	79,100	15,700	3,850	.47
10-30-85	79,000	15,900	3,700	.43
01-15-86	79,000	16,000	3,600	.38

Table 5.--Partial pressures of N_2, O_2+Ar, CO_2, and CH_4 at boreholes A, B, and C, in pascals--Continued

Date	N_2	O_2+Ar	CO_2	CH_4
	(14) Piezometer C2, 11.6 m below the land surface			
01-19-84	79,300	15,700	3,630	0.34
03-22-84	79,400	15,500	3,690	.26
06-08-84	79,500	15,200	3,900	.29
08-14-84	78,500	16,300	3,850	.31
10-25-84	79,200	15,700	3,710	.46
06-11-85	79,400	15,400	3,840	.35
08-28-85	79,000	15,700	3,890	.47
10-30-85	79,100	15,800	3,750	.44
01-15-86	79,100	15,700	3,800	.39

Table 6.--Time-averaged mean partial pressures of N_2, O_2+Ar, CO_2, and CH_4 at boreholes A, B, and C, in pascals

Meters below land surface	Borehole		
	A	B	C
	(1) N_2		
0	77,000	77,000	77,000
1.8	77,800	77,800	77,200
3.6	77,900	77,800	79,000
7.3	78,400	79,500	79,300
11.6	79,700	79,500	79,300
13.6	79,800		
	(2) O_2+Ar		
0	21,600	21,600	21,600
1.8	18,200	19,000	19,500
3.6	18,400	18,100	17,700
7.3	16,800	15,800	15,700
11.6	15,000	15,500	15,600
13.6	15,000		
	(3) CO_2		
0	33.0	33.0	33.0
1.8	2,610	1,830	1,870
3.6	2,300	2,740	1,880
7.3	3,380	3,280	3,610
11.6	3,900	3,660	3,680
13.6	3,860		
	(4) CH_4		
0	0.18	0.18	0.18
1.8	0.08	0.07	0.07
3.6	0.17	0.08	0.09
7.3	0.47	0.68	0.35
11.6	1.54	0.76	0.39
13.6	1.56		

Carbon Dioxide

Carbon dioxide is naturally produced in the unsaturated zone as an end product of the microbial decomposition of organic materials, by root respiration, and by the dissolution of carbonate minerals. Most CO_2 is produced in the root zone and diffuses from that zone to the atmosphere and to underlying deposits (Reardon and others, 1979; Thorstenson and others, 1983). Production of CO_2 is seasonally cyclic, and CO_2 partial pressures are greatest during the peak growing period for surface vegetation and when warm, moist soil conditions enhance microbial production (table 5, figs. 21-25).

At the Sheffield site, an additional source of CO_2 is produced in the waste-disposal trenches by the microbial decomposition of buried wastes. It was hypothesized that the source of CO_2 from the trenches would be identifiable from CO_2 partial pressures within the piezometer network. However, quantitative isolation of the portion of CO_2 that was contributed from the waste material at each sampling location was not possible, and a pattern showing higher mean CO_2 partial pressures at borehole A than at boreholes B and C was not observed (table 6, fig. 26). The inability to quantitatively separate the waste-produced CO_2 from naturally produced CO_2 was a function of two factors: (1) The large amount of spatial variability in CO_2 partial pressures that occurred within a single, presumably homogeneous, lithologic unit at the depths of CO_2 production (table 5, figs. 21-23) was not expected. Previous studies (Reardon and others, 1979; Thorstenson and others, 1983) observed variability in CO_2 partial pressures between distantly separated locations at different lithologic sections but did not address spatial variablity within a single lithologic section. Partial pressures from the Sheffield site indicate that both within-section variability and between-section variability need to be evaluated before regional models of the CO_2 cycle can be applied. (2) Because of the amount of temporal and spatial variability of CO_2 partial pressures, a much more frequent sampling scheme would be necessary to construct quantitatively accurate CO_2 mass balances.

Qualitatively, the presence of a CO_2 source at the waste-disposal trench is suggested by the occurrence of annual partial-pressure peaks of CO_2 that are usually higher at borehole A than at boreholes B and C (table 5, figs. 21-25).

Methane

The atmosphere is a source for CH_4 in the unsaturated zone. Other sources can include transport from natural gas deposits and from sites of microbial decomposition of modern- and paleo-hydrocarbons in the soil and underlying geologic deposits. At the Sheffield site, microbial decomposition of buried wastes is a source for methane that results in a downward gradient of CH_4 partial pressures from the waste source. The gradient is most evident in the Toulon Member (tables 5 and 6, figs. 27-32). At depths of 1.8 and 3.6 m below the land surface, the CH_4 gradient is influenced by methanotrophic bacteria in the soil that consume CH_4 from the overlying atmosphere and from underlying geologic environments. At those depths, the bacteria maintain mean CH_4 partial pressures at about one-third of the atmospheric CH_4 partial pressure (table 6, fig. 32). The bacteria responsible for the consumption of CH_4 in soils at less-than-atmospheric partial pressures have yet to be isolated (M. Lidstrom, California Institute of Technology, oral commun., 1987), but the pattern observed at Sheffield does not appear to be unique. Similar methane partial-pressure profiles have been measured in the arid southwest at the Nevada Test Site (D. Thorstenson, U.S. Geological Survey, Water Resources Division, written commun., 1987).

At 7.3 m below the land surface, methane partial pressures appear to be mostly influenced by the impedance effect of the low air-filled porosity in the Radnor Till Member. However, there is an anamoly in the partial pressure pattern at that depth that warrants discussion. Since the physical movements of gases in the unsaturated zone are generally considered to be dominated by diffusion (Weeks and others, 1982; Thorstenson and others, 1983), it would be expected that in the absence of CH_4 consumers, like those present in the root zone, there would be lateral gradient in CH_4 partial pressures coincident to increased distance from the source. Such a gradient was present in the Toulon Member (figs. 30-32) but not in the Radnor Till Member where only the first two of nine measured CH_4 partial pressures were greater at borehole A than at borehole B (fig. 29) and where time-weighted mean CH_4 partial pressures were higher at borehole B than at borehole A (fig. 32). The reason for this anomaly is not known, but because CH_4 partial pressures at piezometers A1 and A2 in the Toulon Member mimic each other (figs. 29 and 30), and because CH_4 partial pressures at B2 and C2 in the Toulon Member mimic CH_4 partial pressures at B3 and C3 in the Radnor Till Member (figs. 30 and 31), the questionable values can be isolated to those from piezometer A3. Possible causes of the lower partial pressures at A3 include natural variation within the Radnor Till Member and contamination of the backfill used at A3 when the piezometer was installed.

Although it was not possible to isolate methane and count its radioactivity for this study, it was previously shown that CH_4 collected from a tunnel beneath trench 2 (fig. 2) was enriched in ^{14}C and ^{3}H (C. Kunz, New York State Department of Health, written commun., 1983). It was assumed that CH_4 at the study section had specific ^{14}C and ^{3}H activities that were similar to those of CO_2 and water vapor at the same locations. Partial pressures of CH_4 appear to cycle annually, with peaks occurring during summer months (figs. 27-31). However, production of CH_4 also appears to be dominated more by source-related factors than by season (figs. 29-31).

Ethane, Propane, and Butane

Because C_2H_6, C_3H_8, and C_4H_{10} were found at the tunnel and were shown to have ^{3}H activities that increased with molecular weight (C. Kunz, New York State Department of Health, written commun., 1983), it was hypothesized that partial pressure gradients for those gases could be found at the study section. However, C_2H_6, C_3H_8, and C_4H_{10} were not routinely detected (detection limit 0.5 ppm) in gas samples. Propane was detected at concentrations of 1.2 ppm at piezometer A1 and at 1.3 ppm at A2 in January 1984. In March 1984, 0.7 ppm of C_3H_8 was measured at A1, and trace amounts were detected but not quantified at A2, A3, B2, and B3; trace amounts of iso-butane were detected but not quantified at A1, A2, B2, and B3. In June 1984, trace amounts of C_3H_8 were detected but not quantified at A2 and trace amounts of iso-butane were detected but not quantified at A2 and A3. Ethane and n-butane were not detected in any samples. When detected, C_3H_8 and iso-butane were believed to be of waste origin.

Radioactive Gases

Tritiated Water Vapor

It was hypothesized that the microbial decomposition of wastes containing tritium, in combination with the release of tritium from other waste sources, would produce an identifiable gradient of $^3HOH_{(v)}$ in the unsaturated zone. However, no samples had tritium activities that exceeded background conditions (approximately 7.40 becquerels per liter of water), therefore, no gradient was observed at the locations of the gas piezometers. It is believed that the low $^3HOH_{(v)}$ partial pressures resulted from the large molar gradients that exist for the isotopic exchange of tritium from water vapor to liquid water in the unsaturated zone. For the water and air-filled porosities found in the Toulon Member (table 1), the molar ratio of liquid water to water vapor per unit volume is greater than 20,000:1. Consequently, where specific tritium activities of the water vapor exceed specific tritium activities of the infiltrating pore water, there is a strong tendency for $^3HOH_{(v)}$ to dilute into the pore water rather than be transported as a vapor. Measurable gradients of $^3HOH_{(v)}$ would, therefore, be located very close to the waste source.

^{14}Carbon Dioxide

Aerobic decomposition of organic wastes that contain ^{14}C produces $^{14}CO_2$ that diffuses from the waste source through surrounding deposits. The waste source was sufficient to create very high $^{14}CO_2$ partial pressures that sometimes exceeded atmospheric $^{14}CO_2$ partial pressures by factors greater than 10^6 at piezometers A1 and A2, 12 m horizontal from the east wall of trench 2 (tables 7 and 8, figs. 33-37). Consequently, $^{14}CO_2$ partial-pressure gradients were large, having time-averaged partial pressures that decreased by a factor of 34 from A1 and A2 to C2, and by a factor of 640,000 from A1 to the land surface. Production of $^{14}CO_2$ appears to follow an annual cycle with peaks in summer. However, production is strongly source related and is apparently dominated by pulse-type releases from discrete sources. This is evidenced by a drop in $^{14}CO_2$ partial pressures by a factor of 10 or more at borehole A between August 1984 and February 1985.

^{222}Radon

Radioactive decay of ^{226}Ra, which may occur naturally or be of waste origin, emanates ^{222}Rn. Much of the ^{226}Ra that naturally occurs in glacial sediments is present as radium-sulfate precipitates on particle surfaces. The ^{222}Rn that is emanated from a source may diffuse through the unsaturated zone, be transported by convective air masses, or radioactively decay near its location of emanation. Because of its relatively short half-life (3.82 days), and

Table 7.--Partial pressures of CO_2 and $^{14}CO_2$ at boreholes A, B, and C, in pascals

[m, meters]

Date	CO_2	$^{14}CO_2$	Date	CO_2	$^{14}CO_2$
(1)	Piezometer A4, 3.7 m below land surface		(2)	Piezometer A3, 7.3 m below land surface	
03-20-84	2,630	2.01×10^{-5}	03-20-84	2,540	1.00×10^{-5}
06-05-84	1,310	0.81×10^{-5}	06-05-84	3,620	1.53×10^{-5}
08-16-84	1,410	1.44×10^{-5}	08-16-84	4,170	1.23×10^{-5}
10-26-84	1,280	0.05×10^{-5}	10-26-84	3,620	3.65×10^{-5}
02-21-85	2,300	0.15×10^{-5}	02-21-85	1,470	0.12×10^{-5}
06-12-85	2,930	0.36×10^{-5}	06-12-85	3,790	0.38×10^{-5}
08-29-85	3,410	0.46×10^{-5}	08-29-85	3,820	0.80×10^{-5}
10-31-85	3,170	0.34×10^{-5}	10-31-85	4,030	0.39×10^{-5}
(3)	Piezometer A2, 11.6 m below land surface		(4)	Piezometer A1, 13.6 m below land surface	
03-20-84	3,690	3.77×10^{-5}	06-05-84	3,800	2.22×10^{-5}
03-23-84	3,690	3.86×10^{-5}	10-26-84	3,660	5.76×10^{-5}
08-16-84	4,400	6.08×10^{-5}	02-21-85	*3,860	0.40×10^{-5}
10-26-84	3,830	4.03×10^{-5}	06-12-85	3,760	0.38×10^{-5}
02-21-85	3,870	0.68×10^{-5}	08-29-85	3,830	0.59×10^{-5}
06-12-85	3,800	0.41×10^{-5}	10-31-85	3,950	0.43×10^{-5}
08-29-85	3,830	0.48×10^{-5}			
10-31-85	3,910	0.43×10^{-5}			
(5)	Piezometer B4, 3.7 m below land surface		(6)	Piezometer B3, 7.3 m below land surface	
03-21-84	1,940	1.80×10^{-7}	03-21-84	2,700	7.15×10^{-6}
06-08-84	2,760	6.51×10^{-7}	08-15-84	2,150	6.99×10^{-6}
08-15-84	3,770	7.31×10^{-7}	02-21-85	3,560	1.74×10^{-6}
06-11-85	2,700	0.59×10^{-7}	06-11-85	3,660	1.68×10^{-6}
08-28-85	3,100	0.73×10^{-7}	08-28-85	3,520	1.58×10^{-6}
10-30-85	2,630	0.59×10^{-7}	10-30-85	3,640	1.71×10^{-6}
(7)	Piezometer B2, 11.6 m below land surface		(8)	Piezometer C4, 3.7 m below land surface	
03-21-84	3,810	1.63×10^{-5}	06-07-84	2,260	0.70×10^{-8}
06-07-84	3,090	1.97×10^{-5}	08-14-84	1,550	1.10×10^{-8}
08-14-84	3,580	1.52×10^{-5}	10-25-84	1,190	0.21×10^{-8}
10-26-84	3,790	1.08×10^{-5}	02-20-85	*1,870	0.23×10^{-8}
02-21-85	*3,660	0.07×10^{-5}	06-10-85	2,310	0.48×10^{-8}
06-11-85	3,740	0.19×10^{-5}	08-28-85	2,720	1.40×10^{-8}
08-28-85	3,640	0.18×10^{-5}	10-30-85	2,100	0.25×10^{-8}
10-30-85	3,710	0.21×10^{-5}			
(9)	Piezometer C3, 7.3 m below land surface		(10)	Piezometer C_2, 11.6 m below land surface	
03-22-84	3,490	8.57×10^{-7}	06-06-84	3,900	5.05×10^{-7}
08-14-84	3,870	8.04×10^{-7}	08-14-84	3,850	6.72×10^{-7}
10-25-84	3,660	3.83×10^{-7}	10-25-84	3,710	4.94×10^{-7}
02-20-85	3,250	6.21×10^{-7}	06-10-85	3,840	9.25×10^{-7}
06-10-85	3,800	8.70×10^{-7}	08-28-85	3,890	10.00×10^{-7}
08-28-85	3,850	9.11×10^{-7}	10-30-85	3,750	9.67×10^{-7}
10-30-85	3,700	9.37×10^{-7}			

*CO_2 estimated as time-averaged mean.

Table 8.--Time-averaged mean partial pressures of $^{14}CO_2$ at boreholes A, B, and C, in pascals

Meters below land surface	Borehole		
	A	B	C
0	3.96×10^{-11}	3.96×10^{-11}	3.96×10^{-11}
3.7	5.80×10^{-6}	3.54×10^{-7}	5.90×10^{-9}
7.3	1.19×10^{-5}	3.89×10^{-6}	7.45×10^{-7}
11.6	2.54×10^{-5}	7.88×10^{-6}	7.48×10^{-7}
13.6	2.03×10^{-5}		

the tortuous path through which gases must travel, much of the ^{222}Rn that is emanated in the unsaturated zone decays near its location of origin.

Maximum and time-averaged mean activities of ^{222}Rn (tables 9 and 10, fig. 38) were similar within geologic units and between boreholes. This indicates that the source for the measured activities was natural; not of waste origin. The time-averaged mean activities can be regarded as background activities for their respective geologic units. Because of the large variability (table 9) and the measurement error associated with ^{222}Rn activities, it warrants mention that determination of background ^{222}Rn activities for risk assessment purposes should require multiple measurements over time.

Other Gases

Although not a subject of this field investigation, it is important to note that several volatile organic compounds have been measured in samples of ground water collected at the Sheffield site. These include trichloromethane; di-, tri- and tetrachloroethane; dichloropropane; dioxane; tri-, tetra- and perchloroethene; dichloropropene; cyclohexene; benzene; chlorobenzene; toluene; carbon tetrachloride; chloroform; and methylene chloride (Goode, 1986). All have the potential to be gaseous carriers of radioisotopes in the unsaturated zone. ^{85}Krypton was measured in samples collected from a dry well at the site and from the tunnel (C. Kunz, New York State Department of Health, written commun., 1983).

Table 9.--Radioactivity of ^{222}Rn in gases collected at boreholes A, B, and C, in becquerels per liter

[m, meters]

Date	^{222}Rn	Date	^{222}Rn	Date	^{222}Rn
(1) Piezometer A5, 1.8 m below land surface		(2) Piezometer A4, 3.7 m below land surface		(3) Piezometer A2, 11.6 m below land surface	
08-15-84	76.46	08-15-84	10.84	08-15-84	9.09
08-28-84	38.04	08-15-84	12.09	08-28-84	8.31
11-05-84	72.02	08-28-84	24.10	11-05-84	7.84
11-05-84	73.08	11-05-84	55.30	11-05-84	7.89
02-21-85	44.11	11-05-84	56.36	02-21-85	7.84
06-12-85	60.36	12-01-84	43.55	06-12-85	8.44
08-29-85	53.19	06-12-85	59.70	08-29-85	8.91
03-29-85	55.55	08-29-85	47.19		
(4) Piezometer A1, 13.6 m below land surface		(5) Piezometer B5, 1.8 m below land surface		(6) Piezometer B4, 3.7 m below land surface	
08-15-84	8.24	08-15-84	44.96	08-15-84	51.10
08-28-84	4.73	11-05-84	71.11	11-05-84	51.17
11-05-84	5.10	11-05-84	67.04	12-01-84	49.84
02-21-85	6.28	02-21-85	78.36	02-21-84	46.51
06-12-85	8.30	06-11-85	41.23	06-11-85	53.18
08-29-85	7.98	08-28-85	32.52	08-28-85	53.55
(7) Piezometer B3, 7.3 m below land surface		(8) Piezometer B2, 11.6 m below land surface		(9) Piezometer C5, 1.8 m below land surface	
08-15-84	7.49	08-15-84	7.22	08-15-84	26.05
11-05-84	13.75	08-28-84	4.63	08-20-84	17.41
02-21-85	14.57	11-05-84	5.68	11-05-84	47.52
06-11-85	10.91	02-21-85	7.47	02-20-85	59.46
08-28-85	13.35	06-11-85	7.30	06-11-85	13.73
				08-28-85	15.22
(10) Piezometer C4, 3.7 m below land surface		(11) Piezometer C3, 7.3 m below land surface		(12) Piezometer C2, 11.6 m below land surface	
08-15-84	45.40	08-15-84	13.98	08-15-84	6.88
11-05-84	50.89	08-28-84	8.15	08-28-84	6.87
12-01-84	49.88	11-05-84	13.37	11-05-84	8.13
06-11-85	56.63	02-20-85	10.26	02-20-85	7.52
08-28-85	52.30	06-11-85	12.89	06-11-85	7.55
		08-28-85	14.68	08-28-85	7.67

Table 10.--Time-averaged mean radioactivities of ^{222}Rn at boreholes A, B, and C, in becquerels per liter

[Dashes indicate no value]

Meters below land surface	Borehole		
	A	B	C
0	0.04	0.04	0.04
1.8	56.17	56.57	35.04
3.7	48.68	50.89	52.21
7.3	---	12.51	12.11
11.6	8.22	6.42	7.61
13.6	7.34		

CONCLUSIONS

Field investigation of the spatial and temporal distributions of gases in the unsaturated zone defined partial-pressure gradients for CH_4 and $^{14}CO_2$ that originated at the waste source. Partial pressures of $^{14}CO_2$ at piezometers A1 and A2 exceeded atmospheric $^{14}CO_2$ partial pressures by factors greater than 10^6 for some measurements. Releases of the waste-produced gases were apparently from discrete, confined sources and were not necessarily related to season.

Partial-pressure gradients that were expected to occur for $^3HOH_{(v)}$, ethane, propane, and butane were not found. It is likely that gradients for those gases occur nearer to the waste source.

Activities for ^{222}Rn did not exceed what were probably background activities for each geologic unit. Those background activities were fairly constant within units and between boreholes.

Although not a subject of this investigation, distributions of volatile organic compounds near low-level radioactive waste are regarded as an important topic for continued field research.

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FIGURES 1 to 38

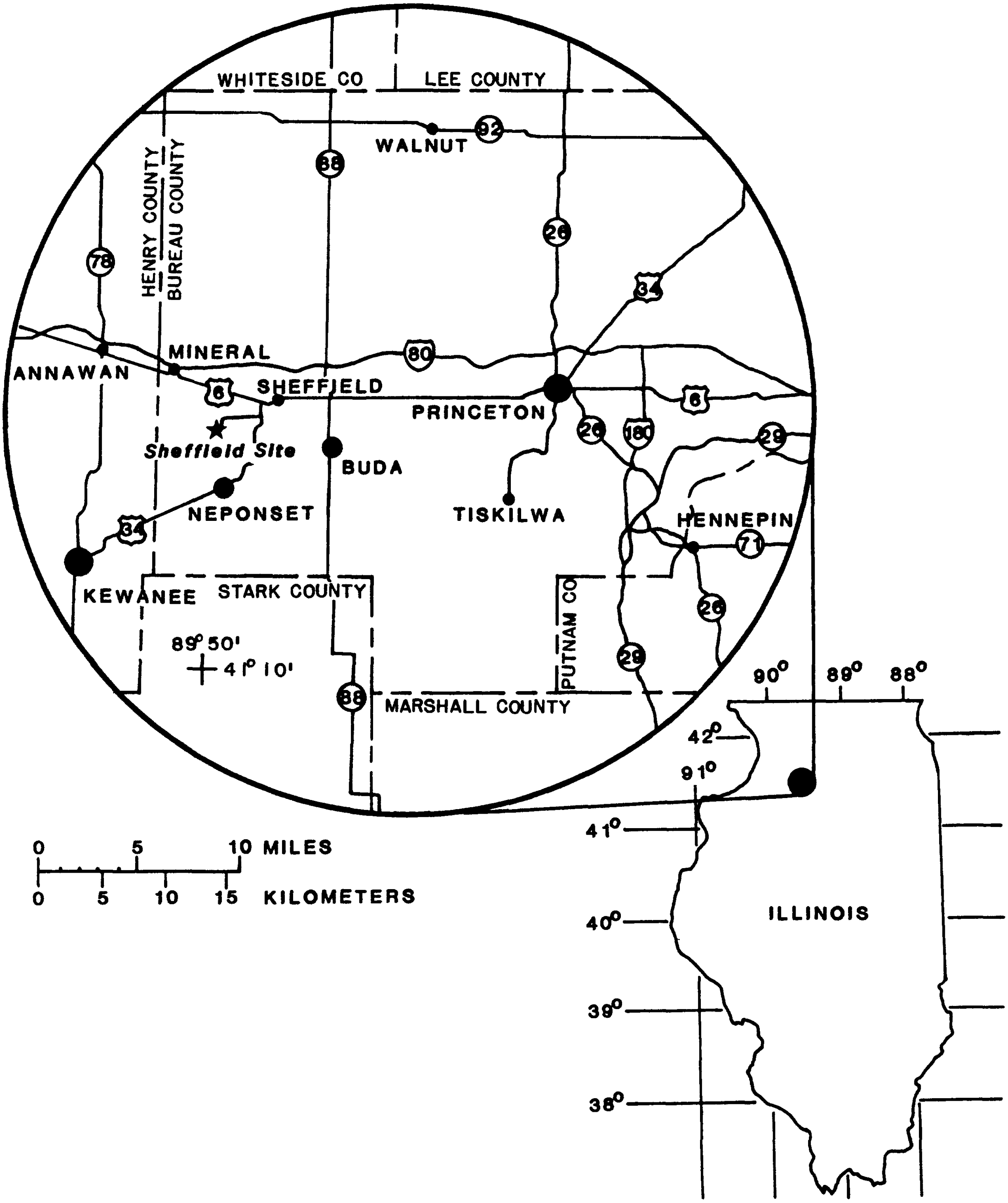


Figure 1.--Location of Sheffield low-level radioactive-waste disposal site.

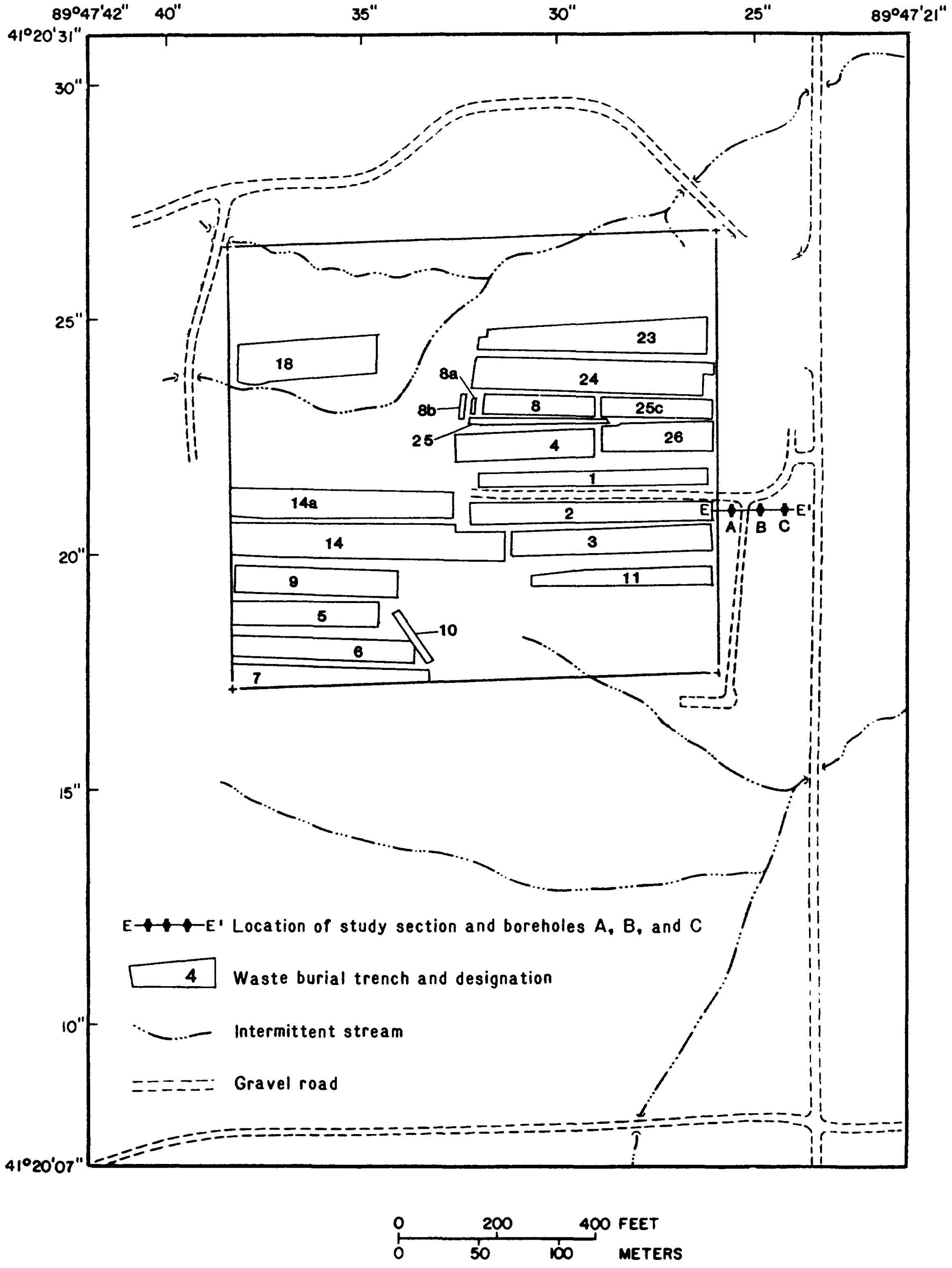
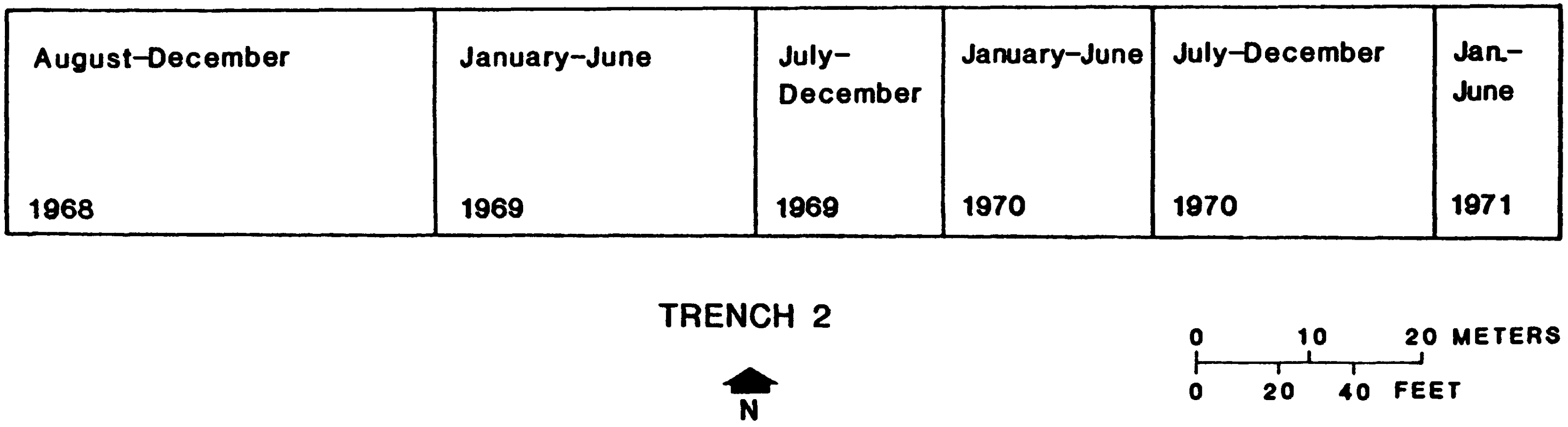


Figure 2.--Locations of the trenches, the study section, and boreholes A, B, and C at the Sheffield site.



Date opened: August 16, 1968

Date closed: March 31, 1971

Length: 140 meters

Width: 18.3 meters

Depth: 7.6 meters

Volume: 19,500 cubic meters

Radioactivity of contents:

3.87×10^{14} becquerels (10,450 curies)

Figure 3.--History of waste disposal in trench 2.

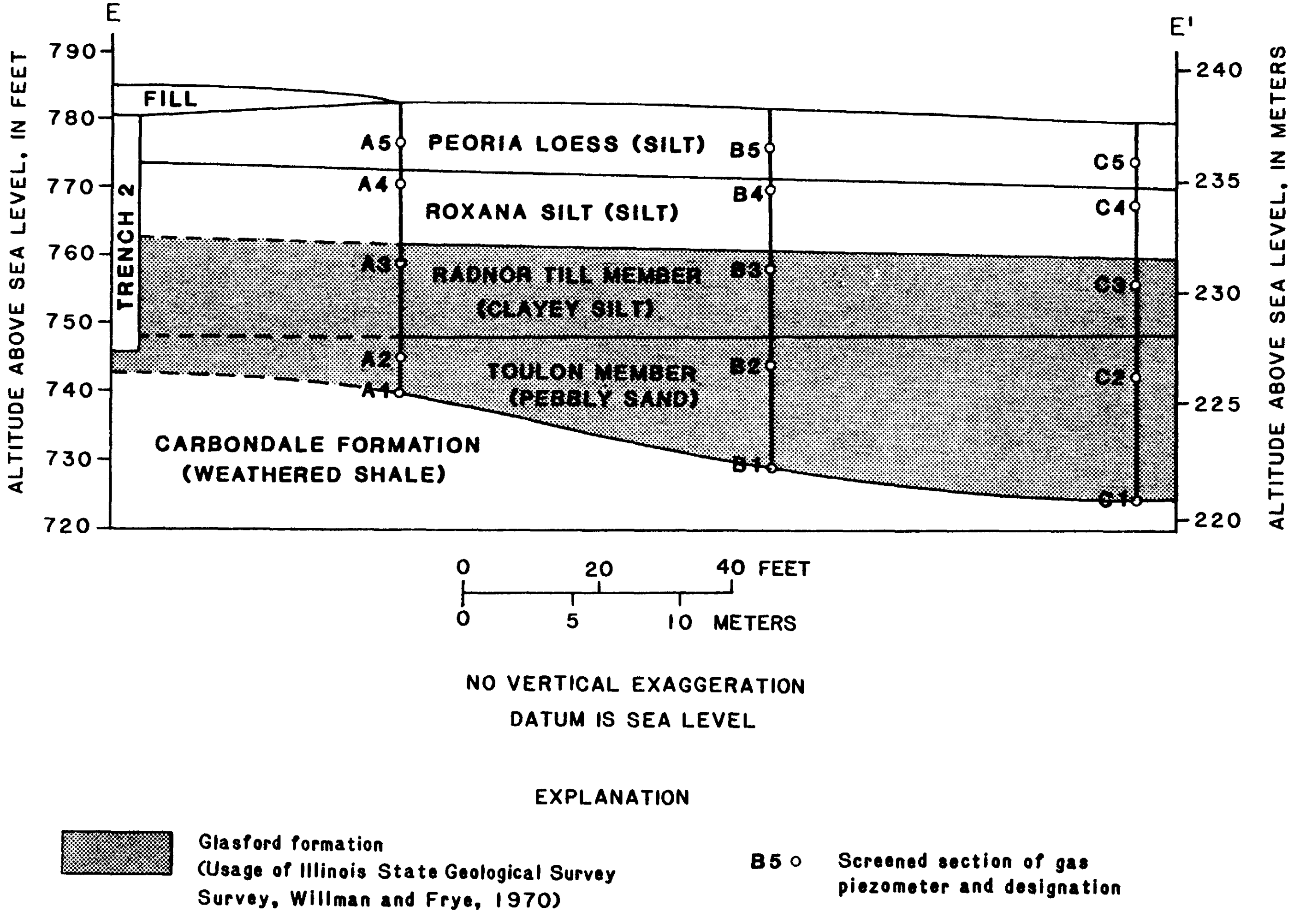


Figure 4.--Lithology of the study section and locations of gas piezometers.

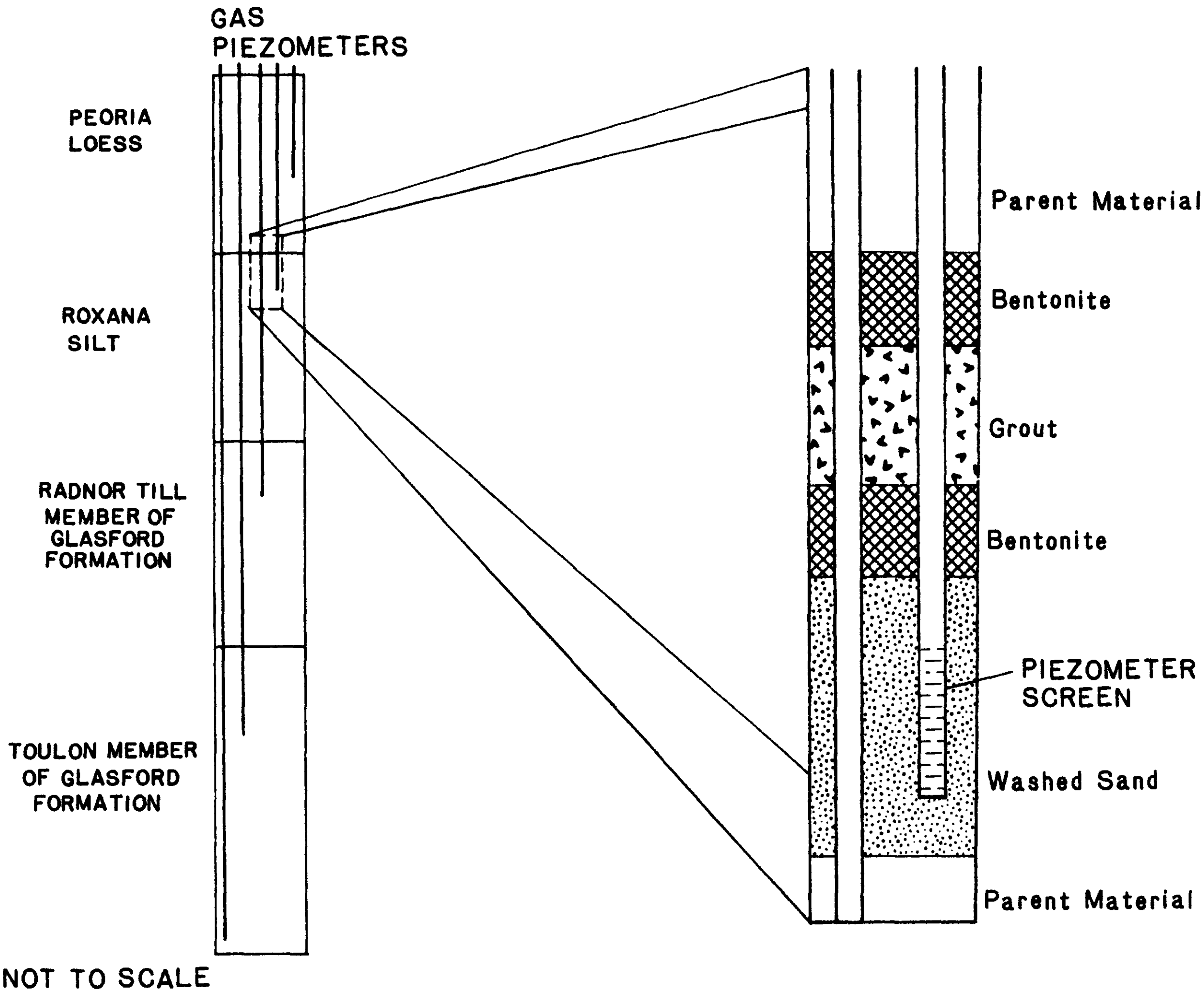


Figure 5.--Typical gas piezometer installation.

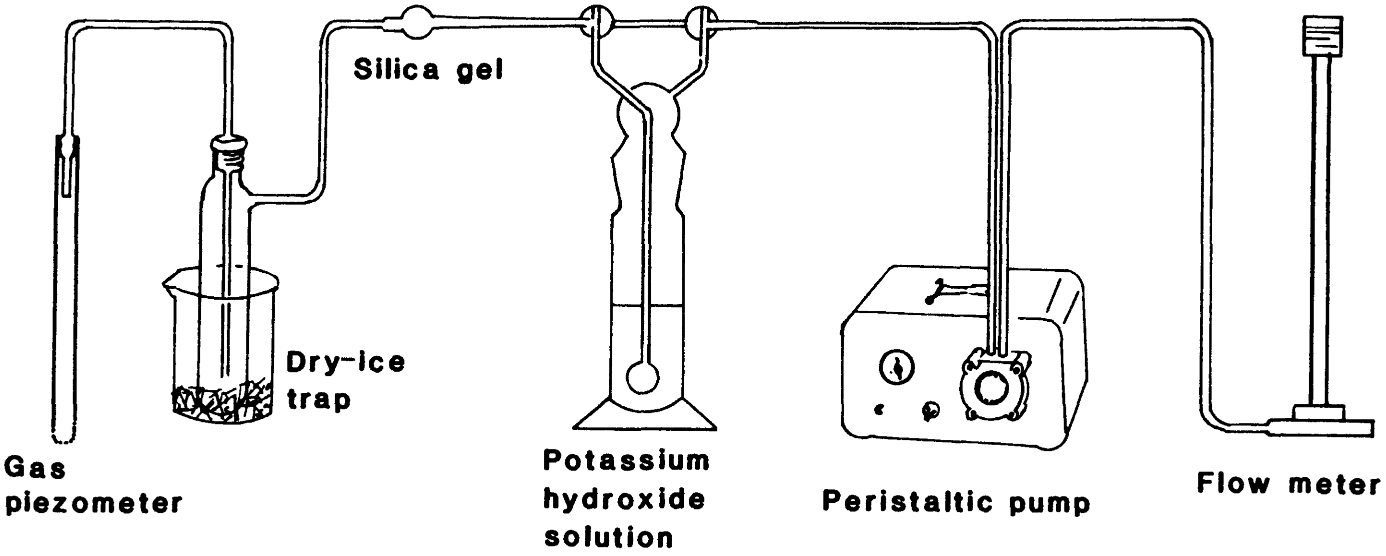
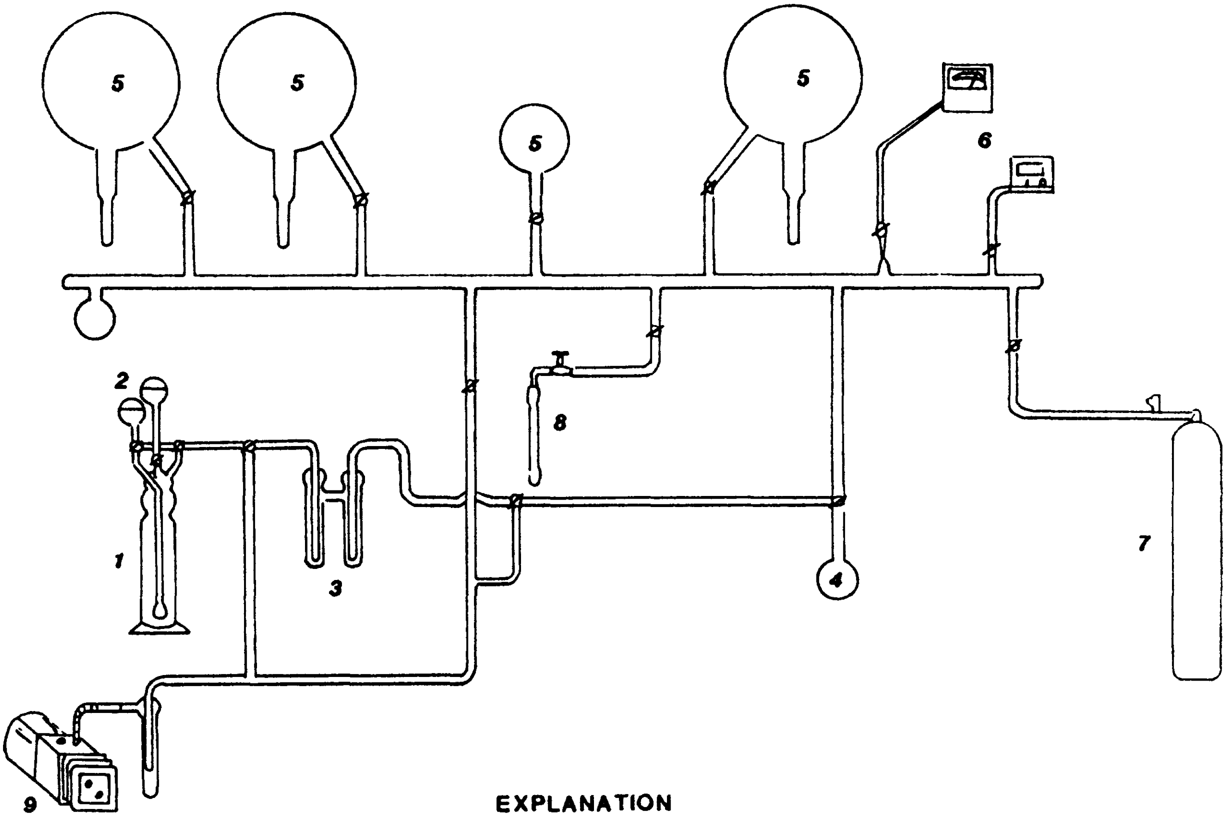


Figure 6.--Flow-through system used for collecting water vapor and carbon dioxide.



EXPLANATION

1. **Gas-wash bottle/sampler**
2. **Hydrochloric acid**
3. **CO_2 solid-methanol freeze traps**
4. **Liquid nitrogen freeze trap**
5. **Gas expansion and dilution bulbs of known volume**
6. **Transducers for measuring gas pressures**
7. **CO_2 for sample dilution**
8. **Stainless steel sample holding cylinder**
9. **Vacuum pump**

Figure 7.--Gas manifold used for the dilution of carbon dioxide.

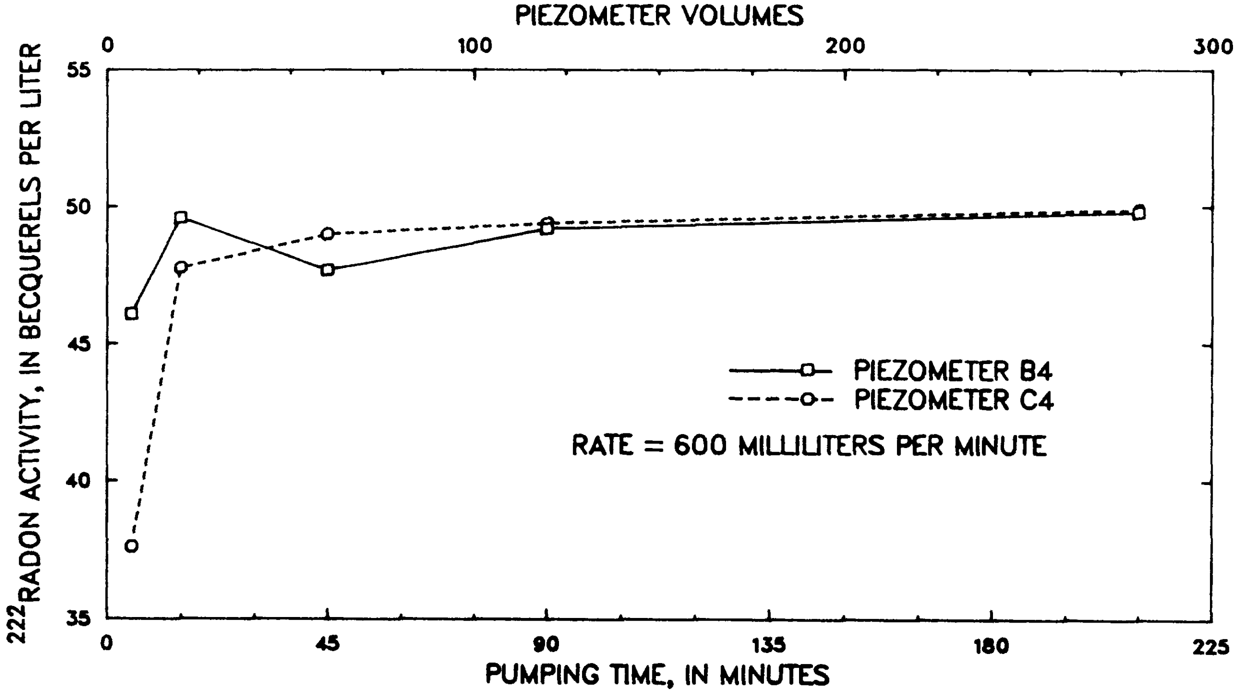


Figure 8.--Relation of change in ^{222}radon activity to piezometer volumes pumped and pumping times at piezometers B4 and C4 on December 1, 1984.

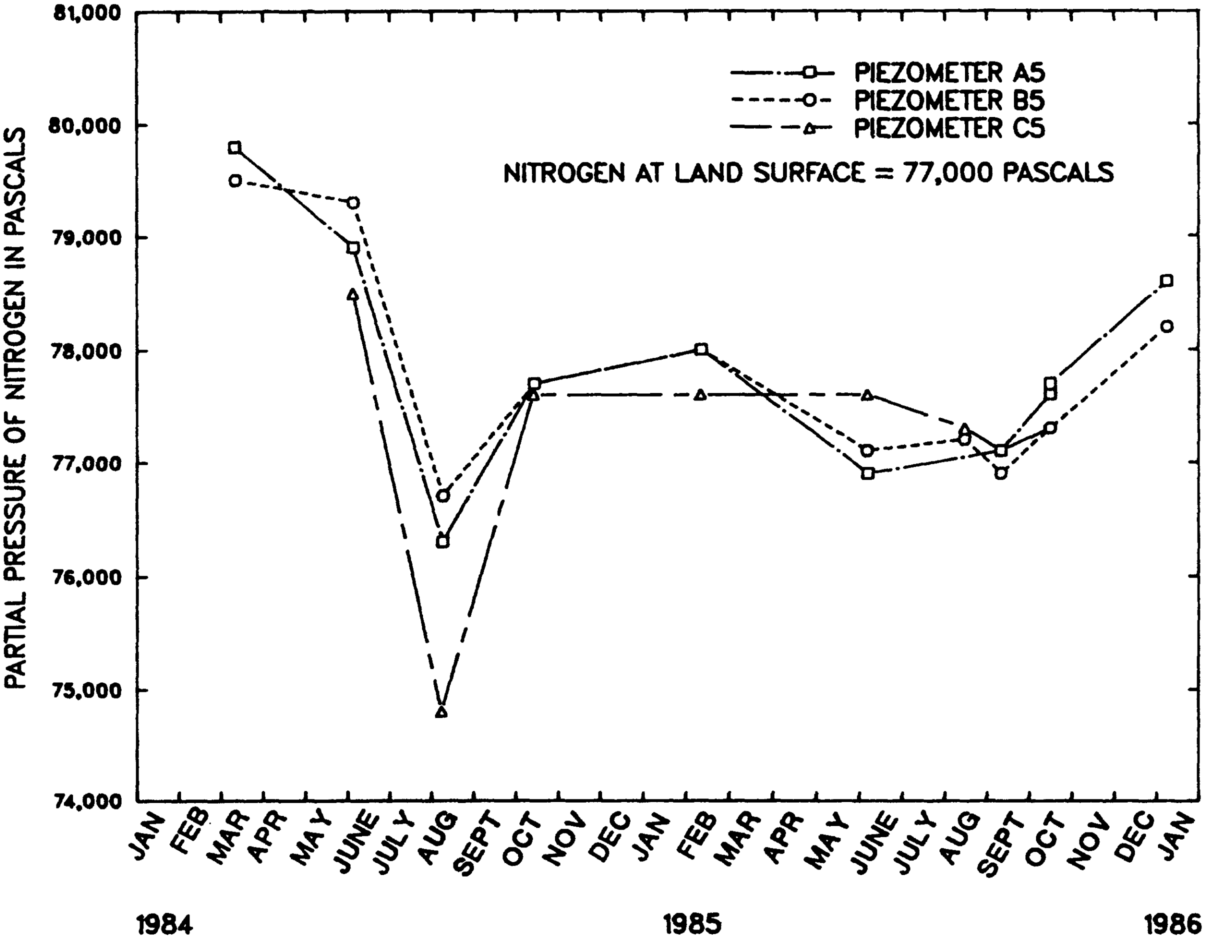


Figure 9.--Temporal changes in partial pressure of nitrogen 1.8 meters below land surface at boreholes A, B, and C.

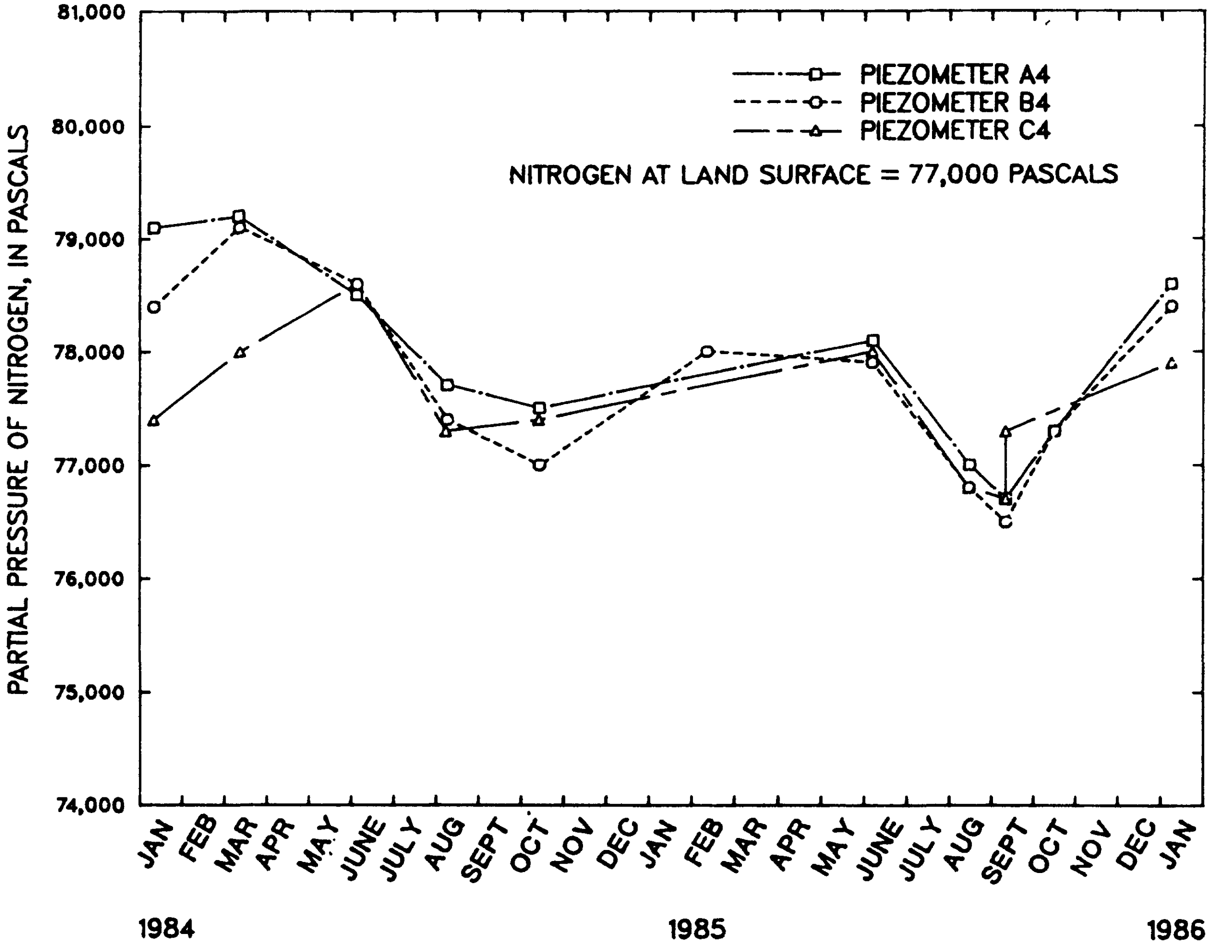


Figure 10.--Temporal changes in partial pressure of nitrogen 3.7 meters below land surface at boreholes A, B, and C.

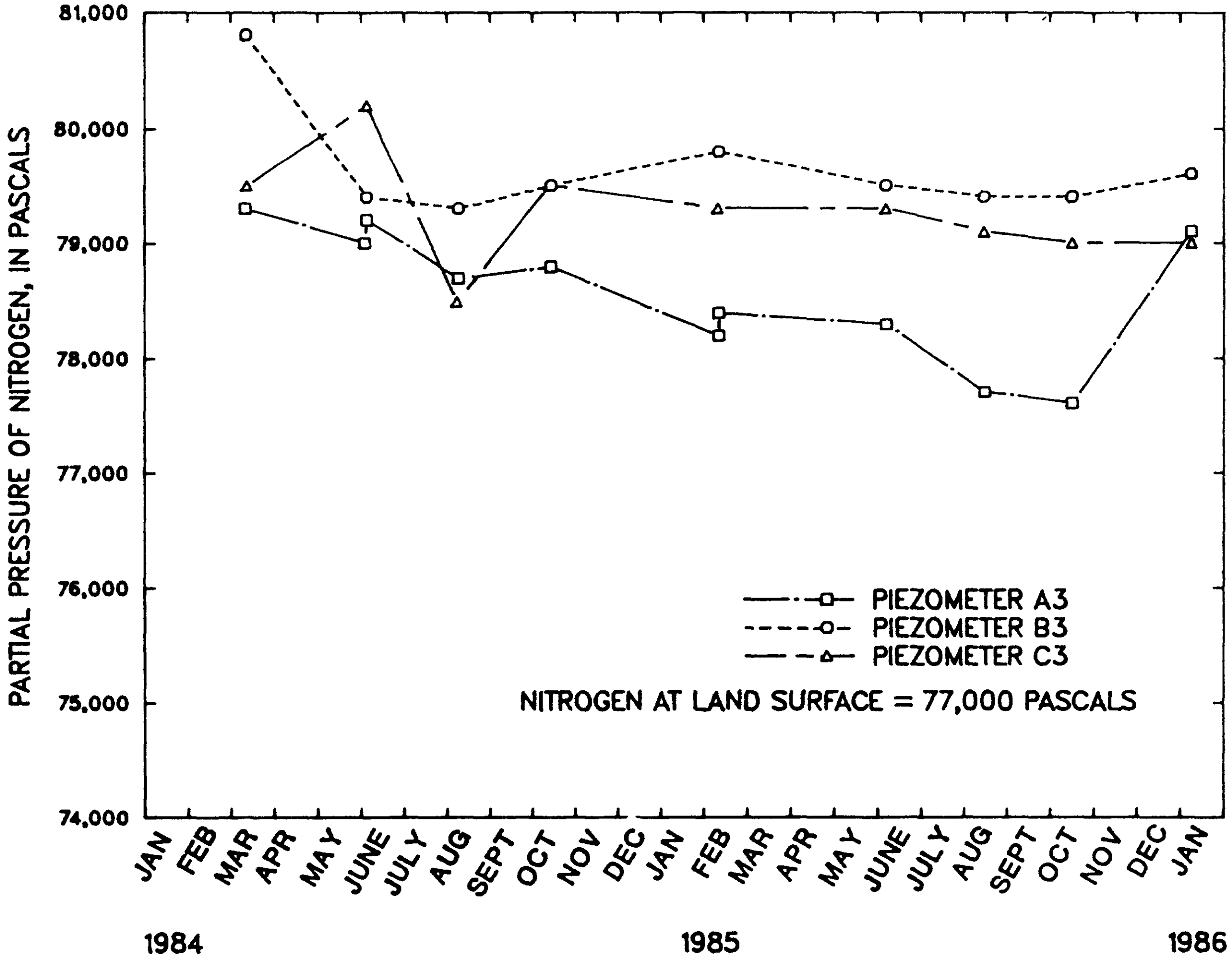


Figure 11.--Temporal changes in partial pressure of nitrogen 7.3 meters below land surface at boreholes A, B, and C.

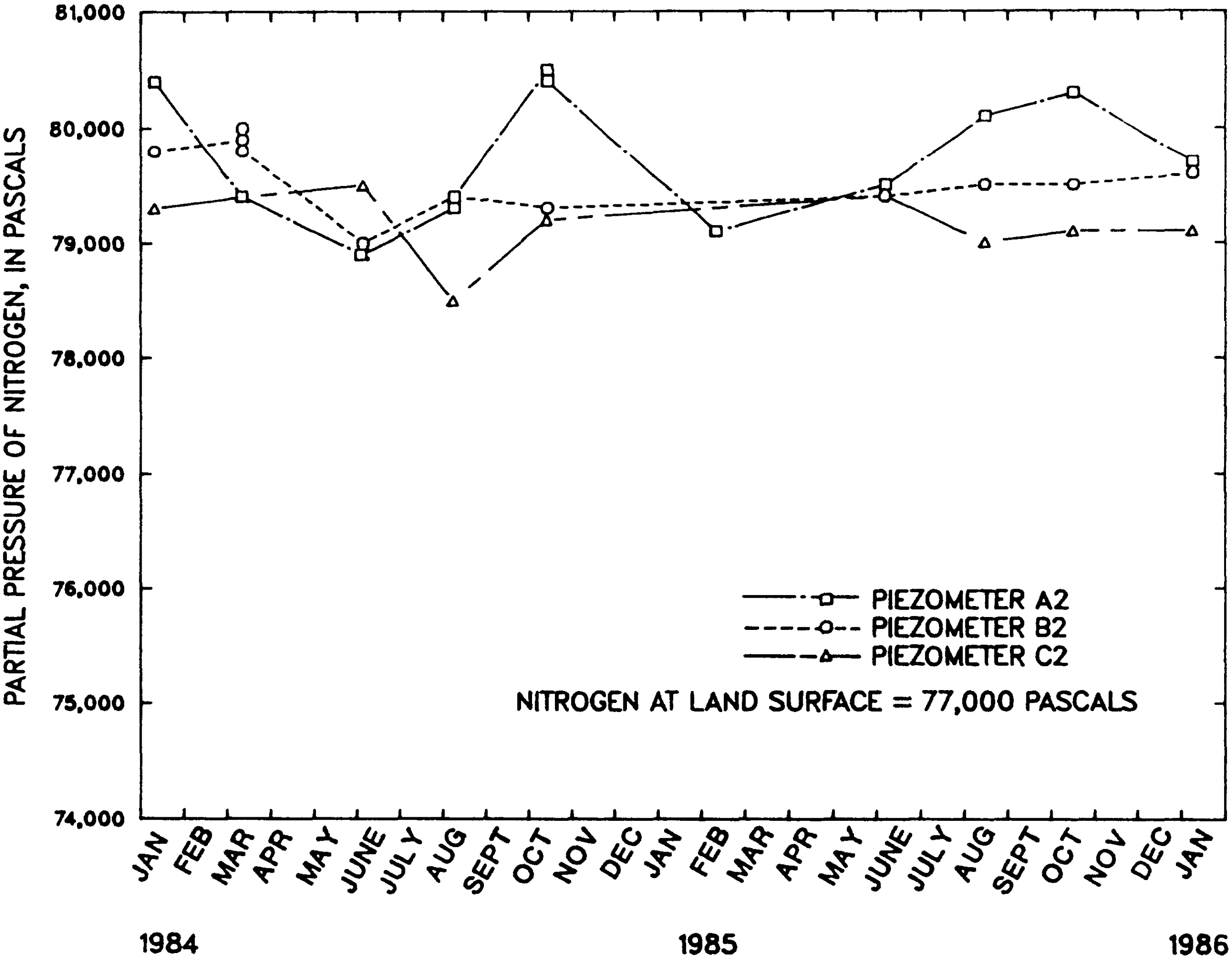


Figure 12.--Temporal changes in partial pressure of nitrogen 11.6 meters below land surface at boreholes A, B, and C.

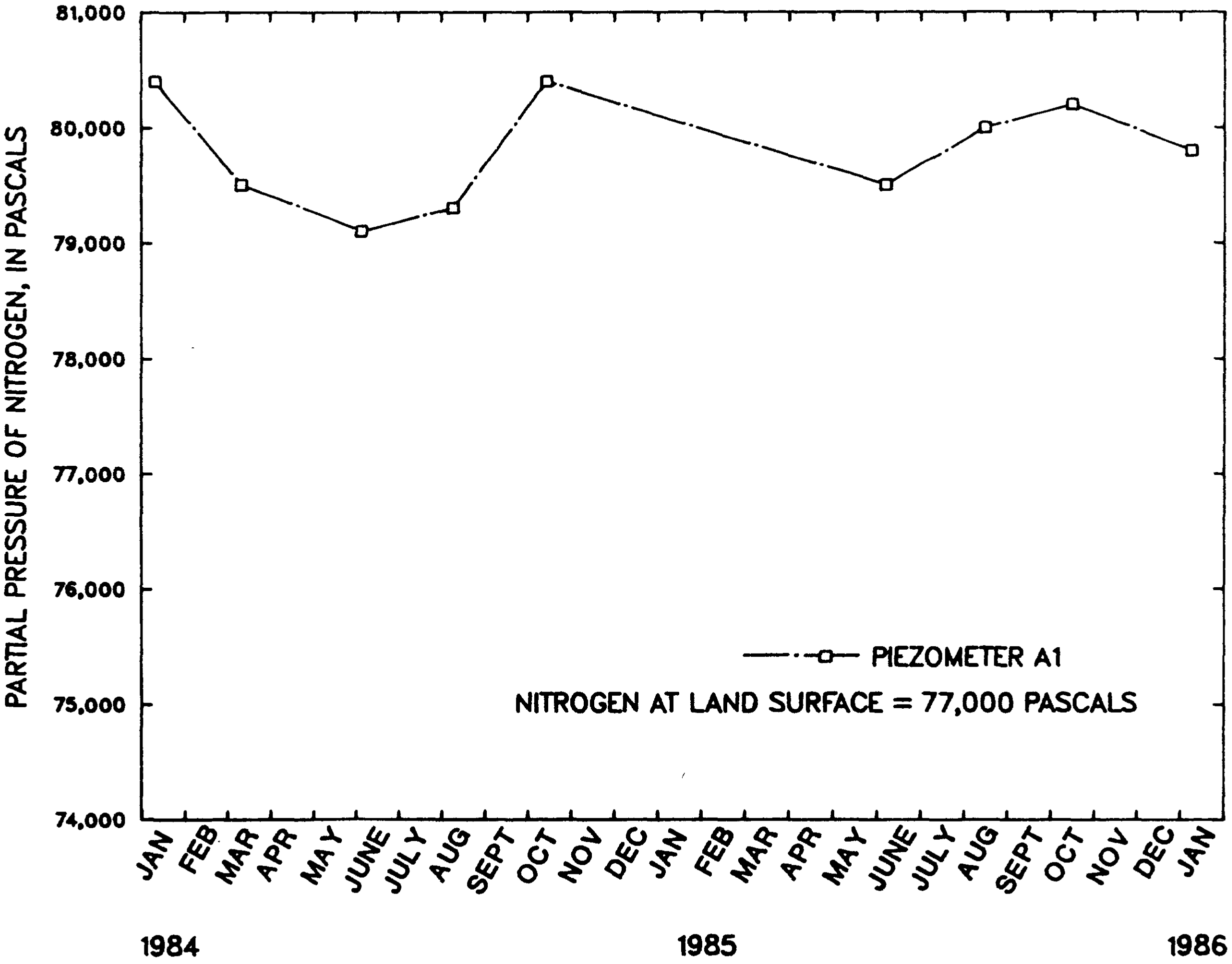


Figure 13.--Temporal changes in partial pressure of nitrogen 13.6 meters below land surface at borehole A.

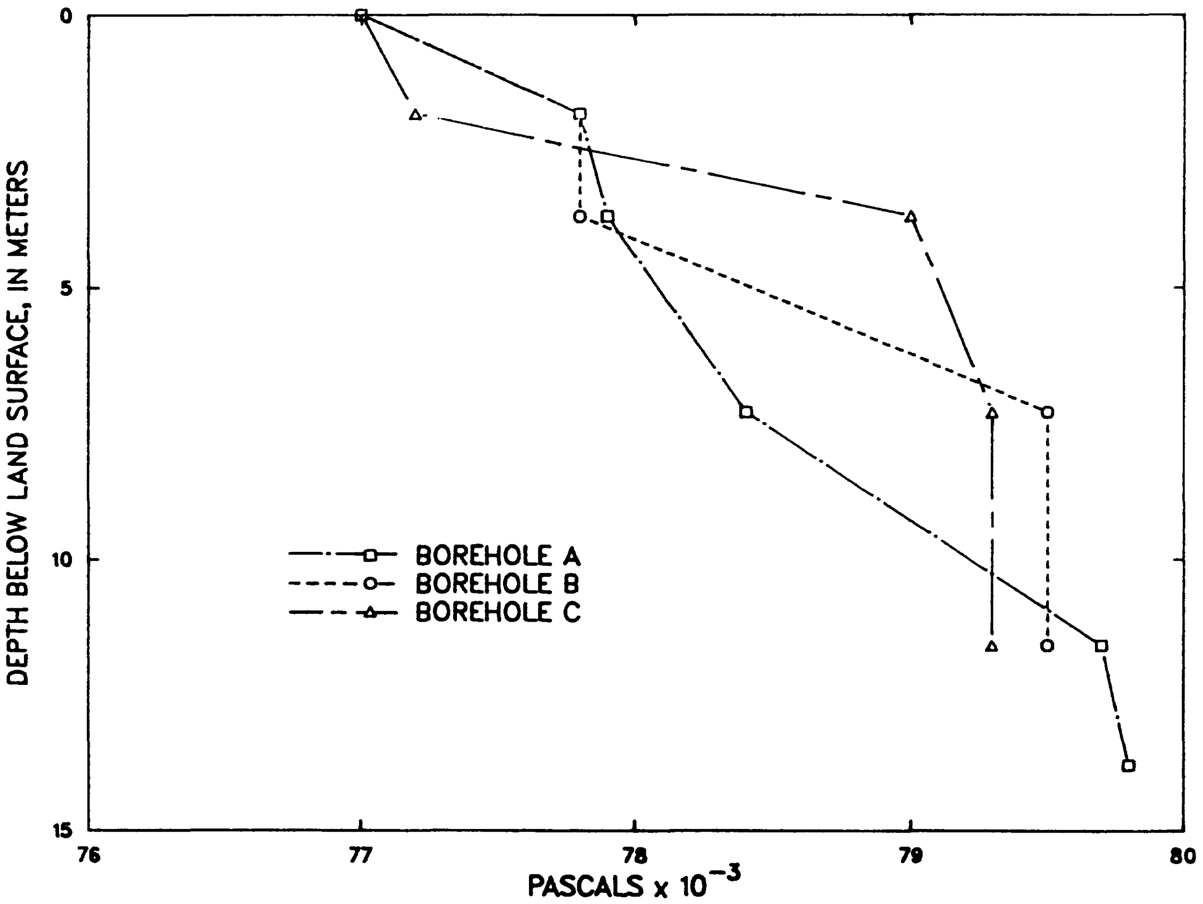


Figure 14.--Changes in the time-averaged mean partial pressure of nitrogen at boreholes A, B, and C as a function of depth.

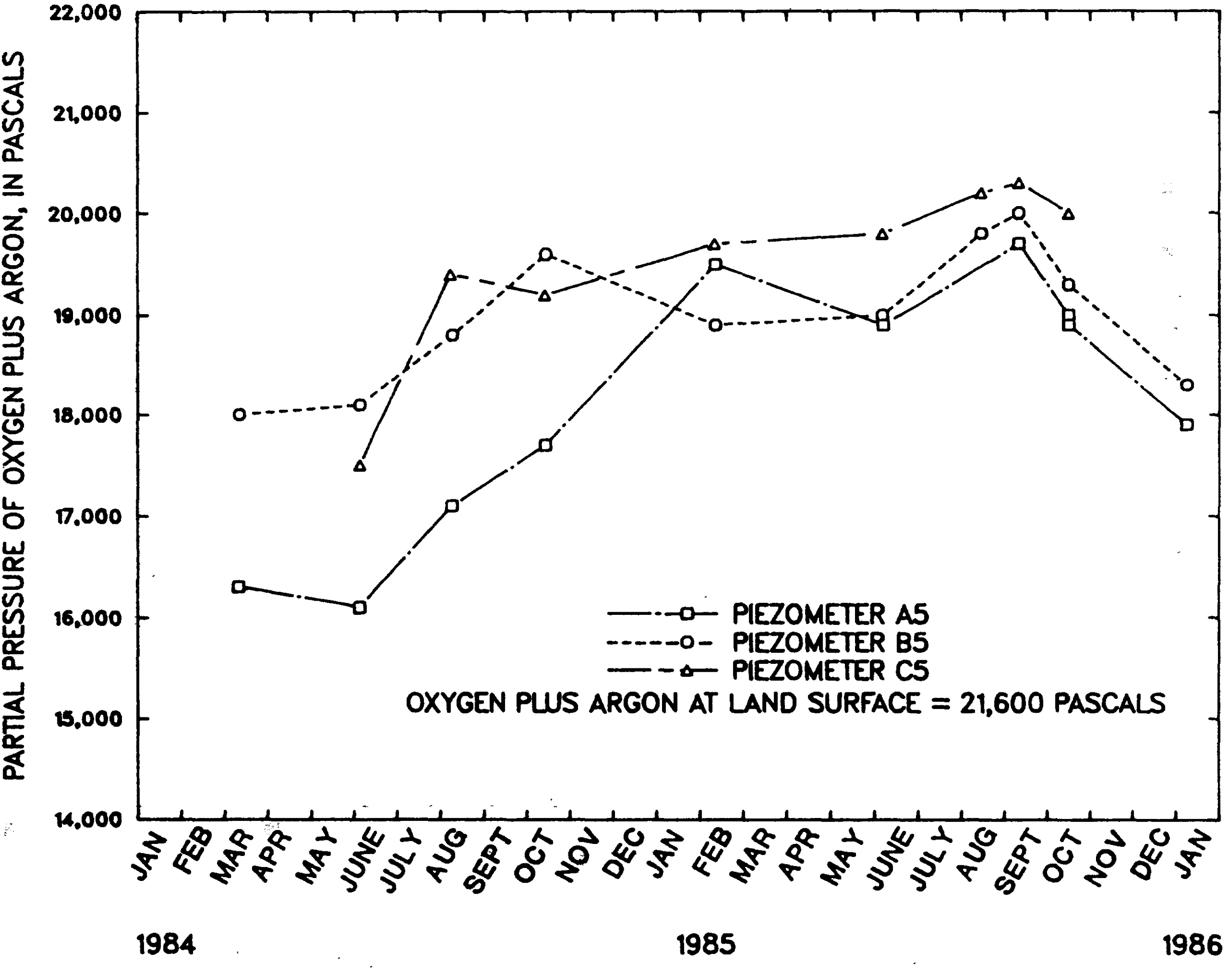


Figure 15.--Temporal changes in partial pressure of oxygen plus argon 1.8 meters below land surface at boreholes A, B, and C.

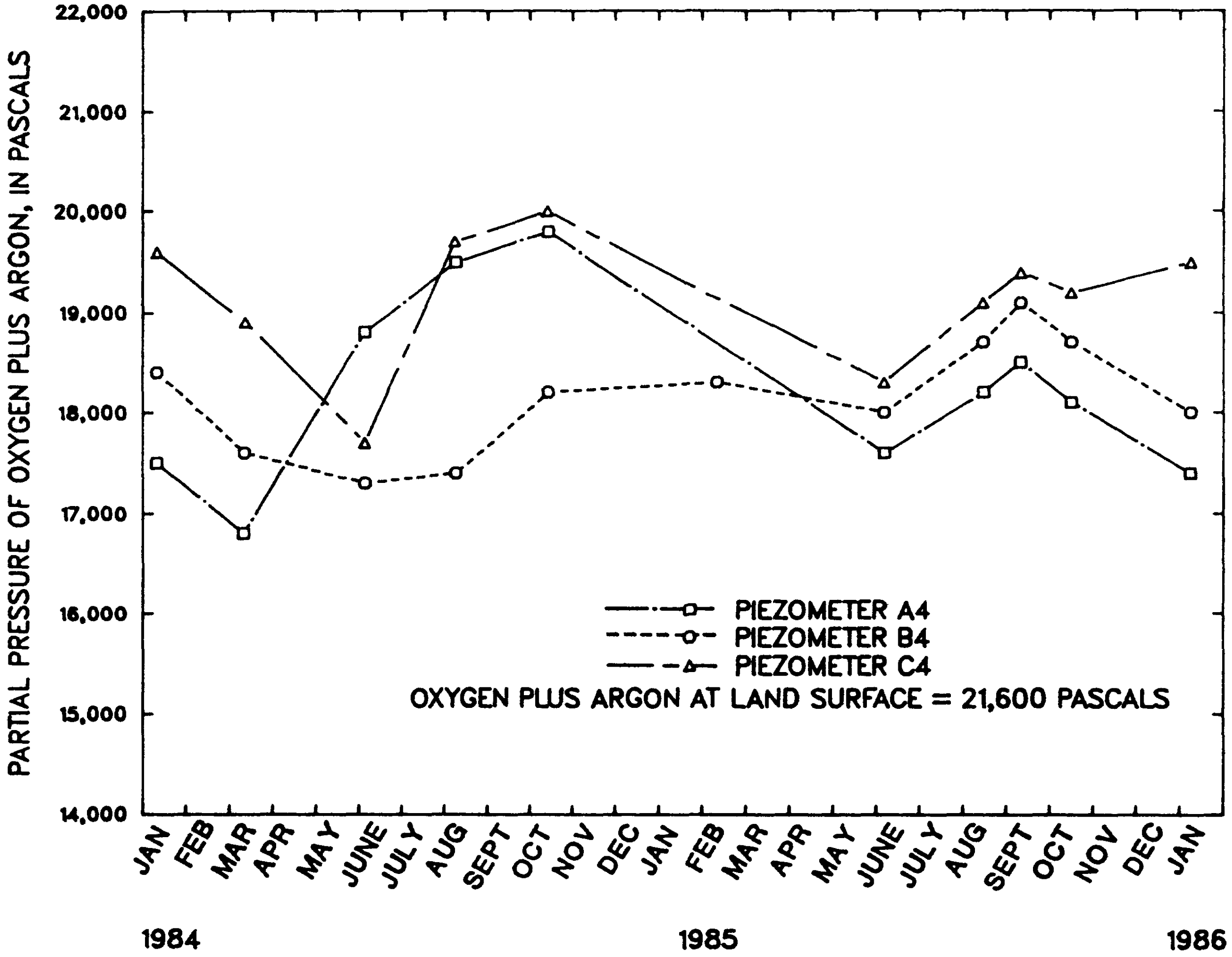


Figure 16.--Temporal changes in partial pressure of oxygen plus argon 3.7 meters below land surface at boreholes A, B, and C.

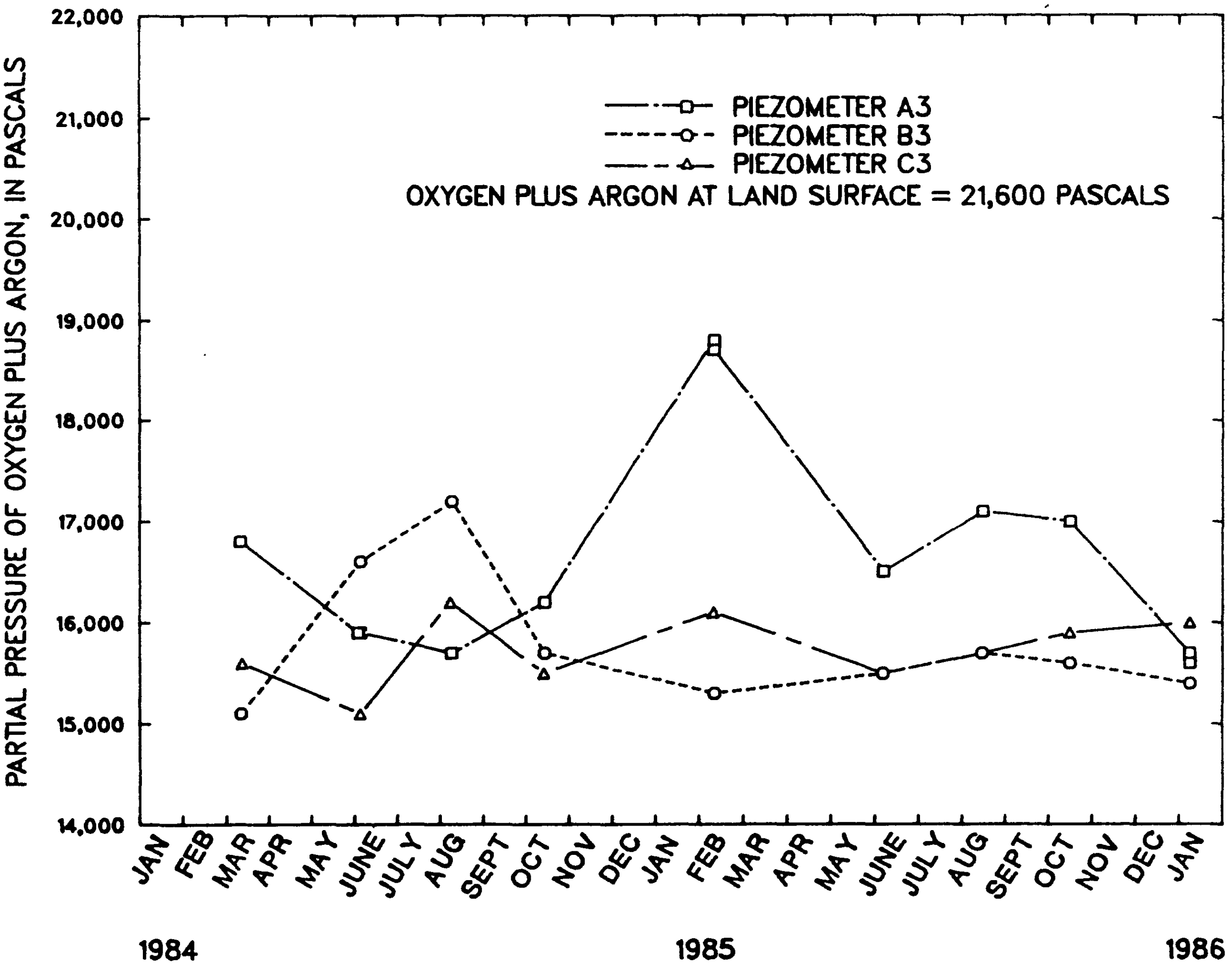


Figure 17.--Temporal changes in partial pressure of oxygen plus argon 7.3 meters below land surface at boreholes A, B, and C.

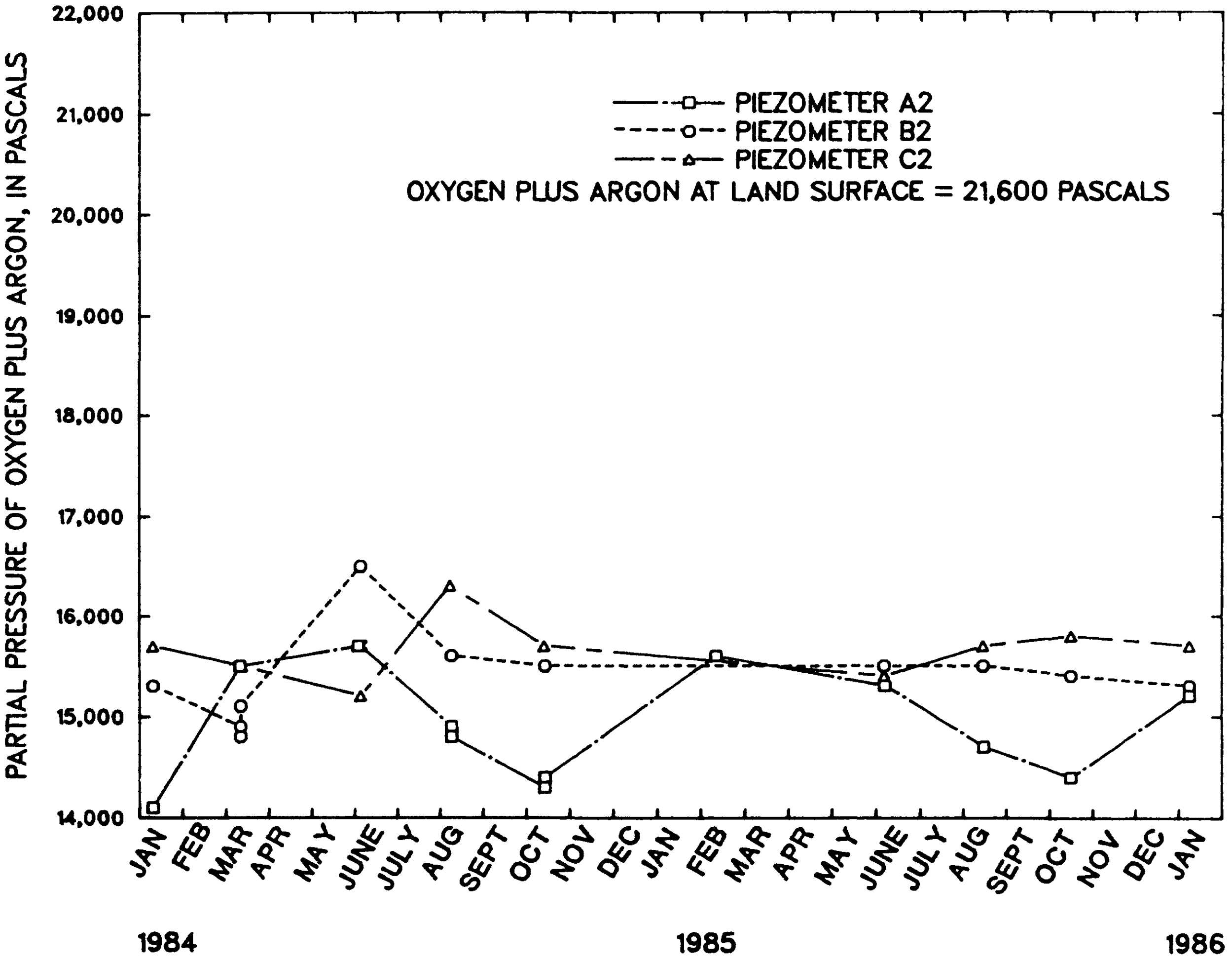


Figure 18.--Temporal changes in partial pressure of oxygen plus argon 11.6 meters below land surface at boreholes A, B, and C.

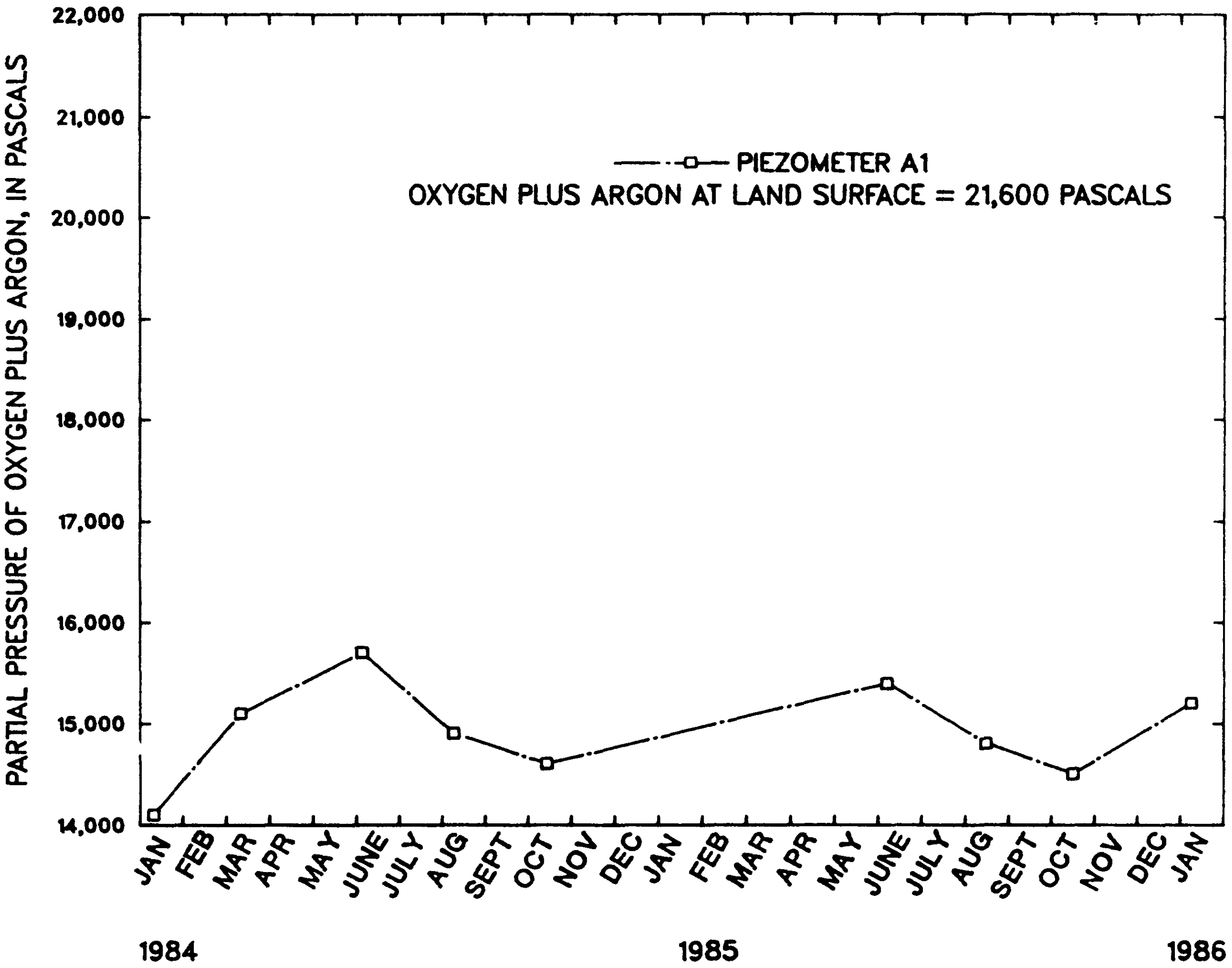


Figure 19.--Temporal changes in partial pressure of oxygen plus argon 13.6 meters below land surface at borehole A.

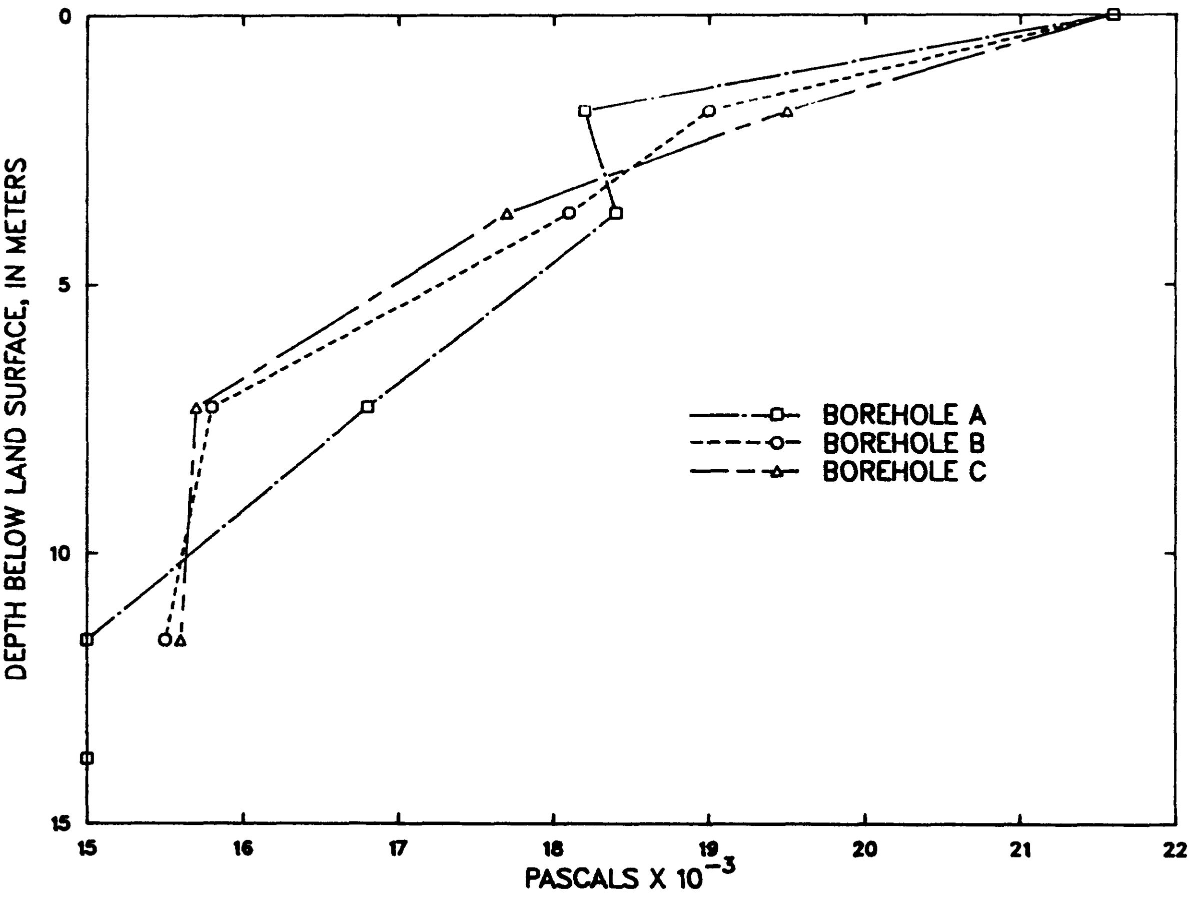


Figure 20.--Changes in the time-averaged mean partial pressure of oxygen plus argon at boreholes A, B, and C as a function of depth.

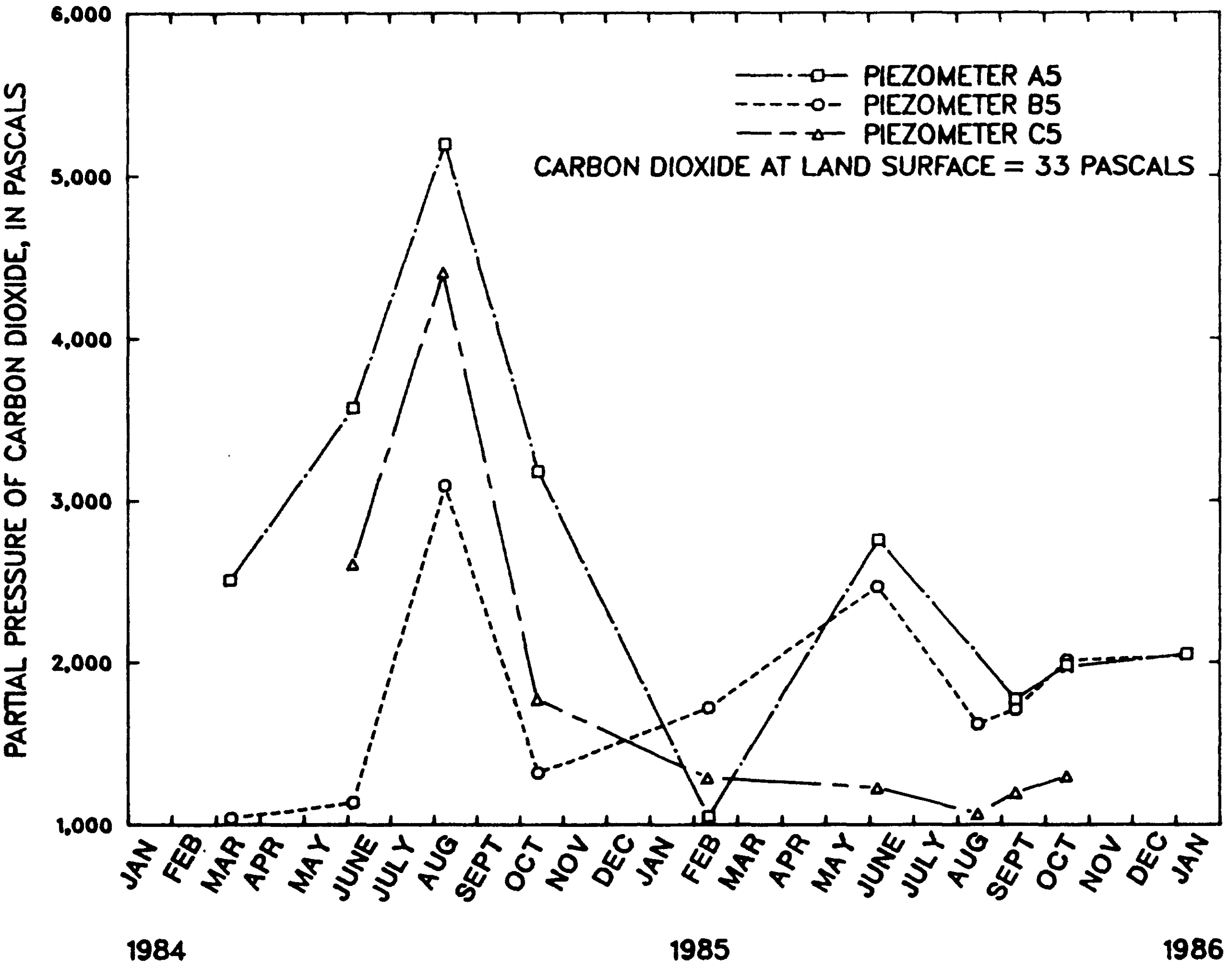


Figure 21.--Temporal changes in partial pressure of carbon dioxide 1.8 meters below land surface at boreholes A, B, and C.

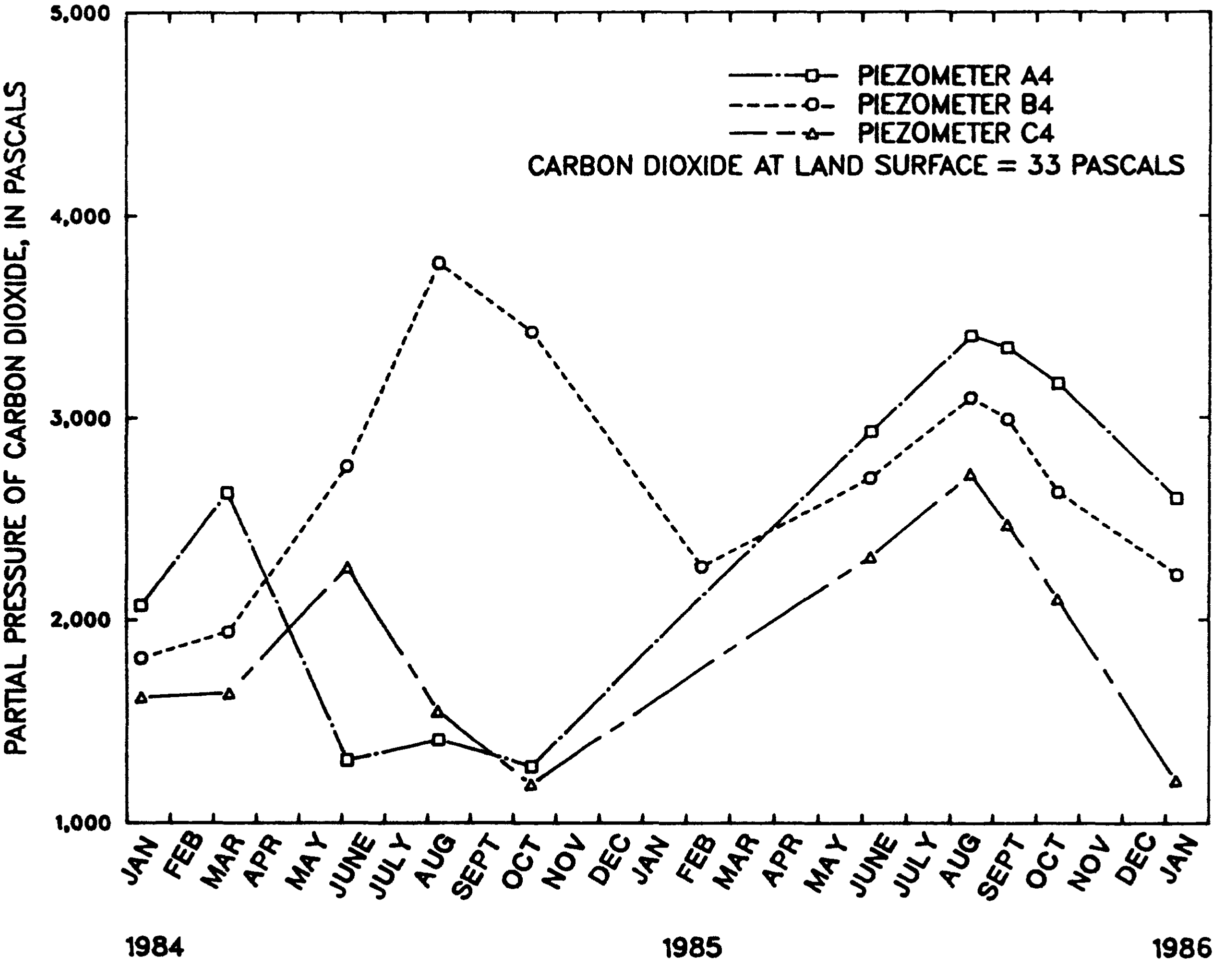


Figure 22.--Temporal changes in partial pressure of carbon dioxide 3.7 meters below land surface at boreholes A, B, and C.

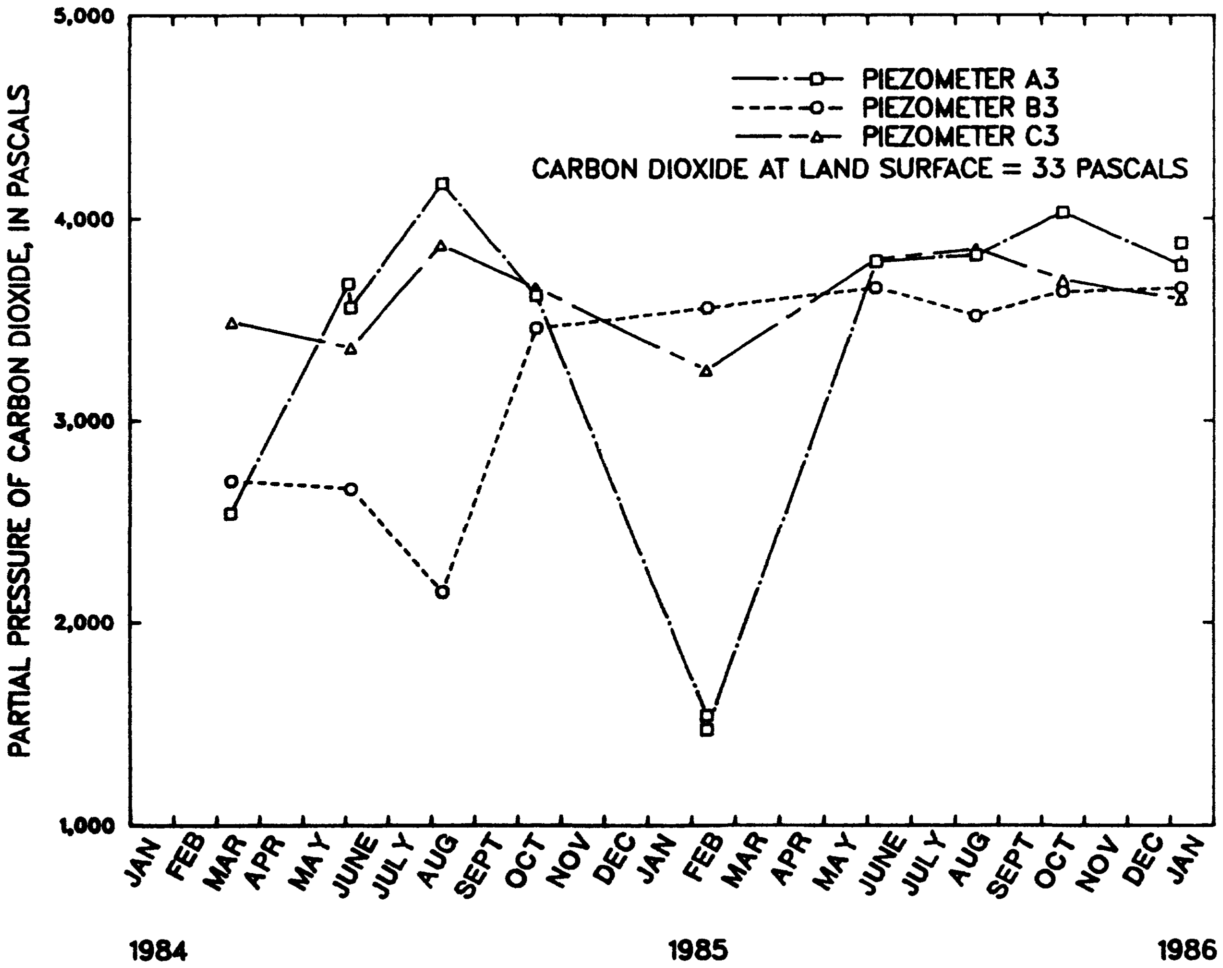


Figure 23.--Temporal changes in partial pressure of carbon dioxide 7.3 meters below land surface at boreholes A, B, and C.

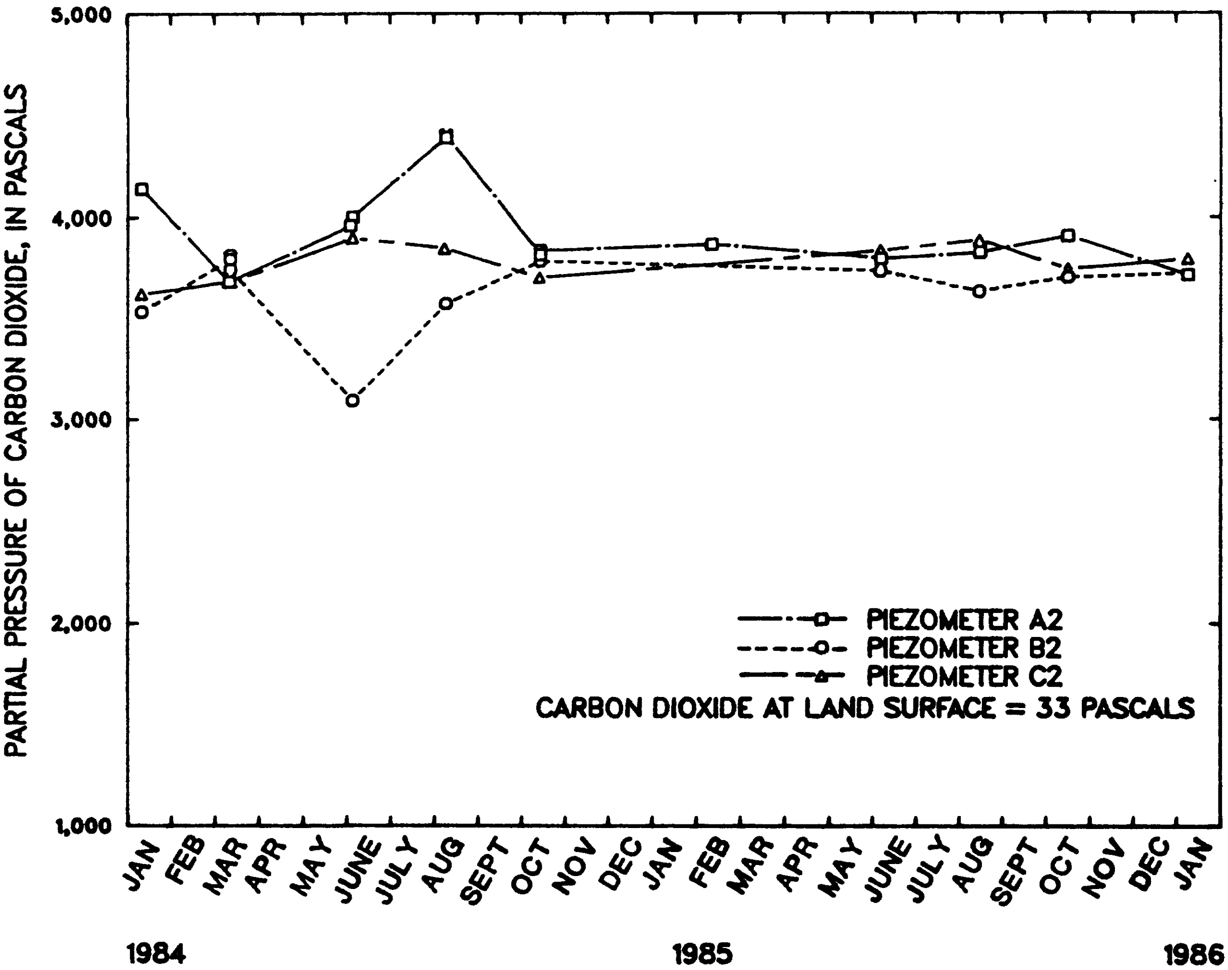


Figure 24.--Temporal changes in partial pressure of carbon dioxide 11.6 meters below land surface at boreholes A, B, and C.

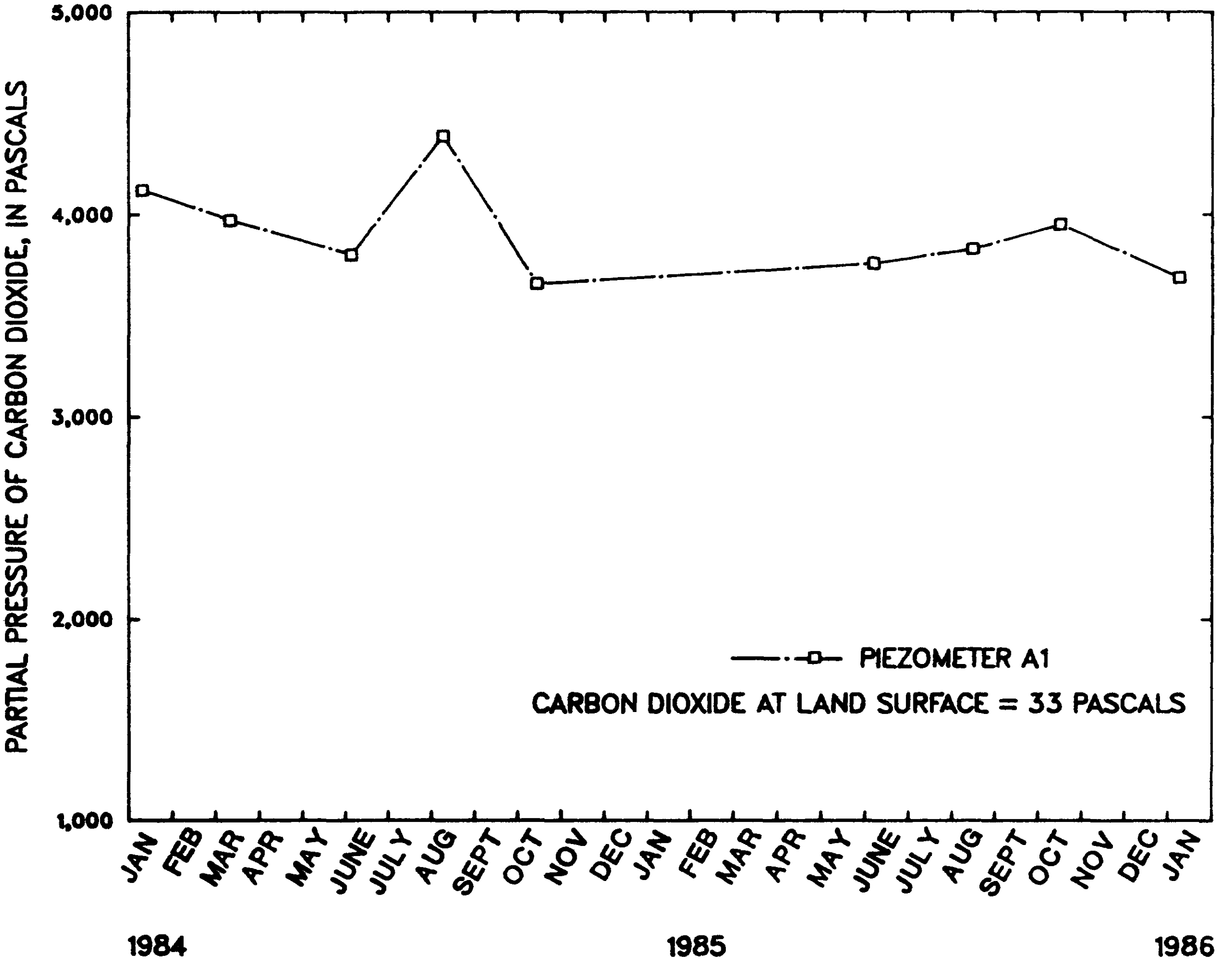


Figure 25.--Temporal changes in partial pressure of carbon dioxide 13.6 meters below land surface at borehole A.

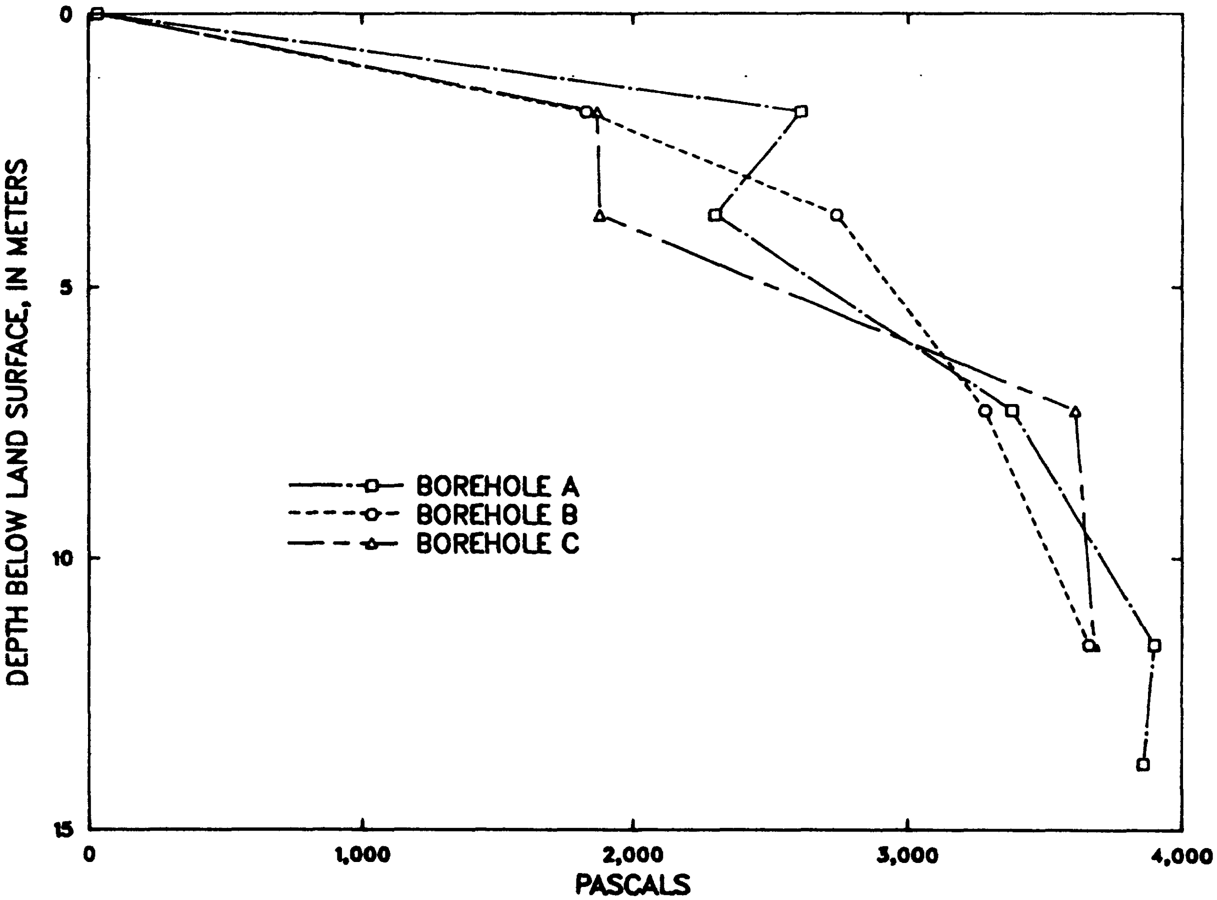


Figure 26.--Changes in the time-averaged mean partial pressure of carbon dioxide at boreholes A, B, and C as a function of depth.

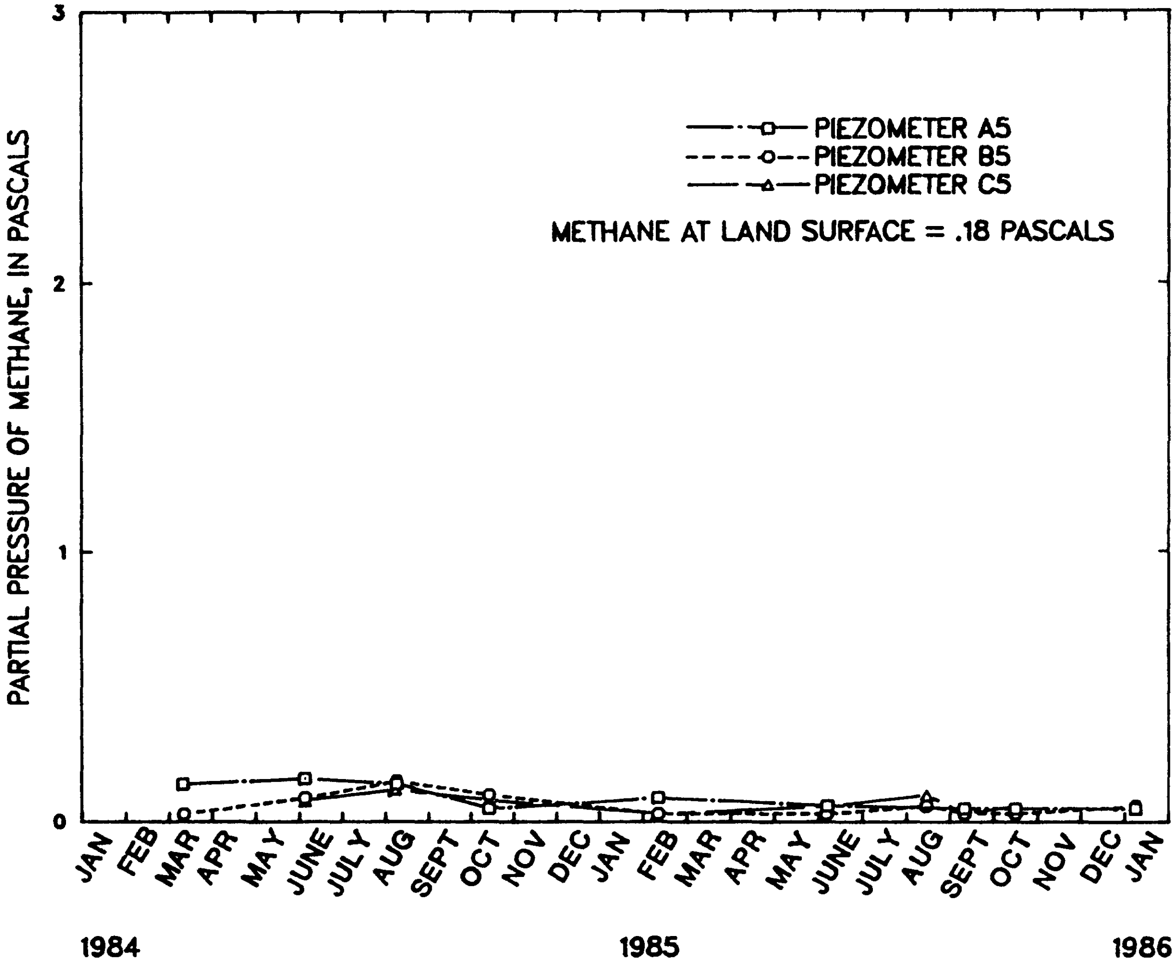


Figure 27.--Temporal changes in partial pressure of methane 1.8 meters below land surface at boreholes A, B, and C.

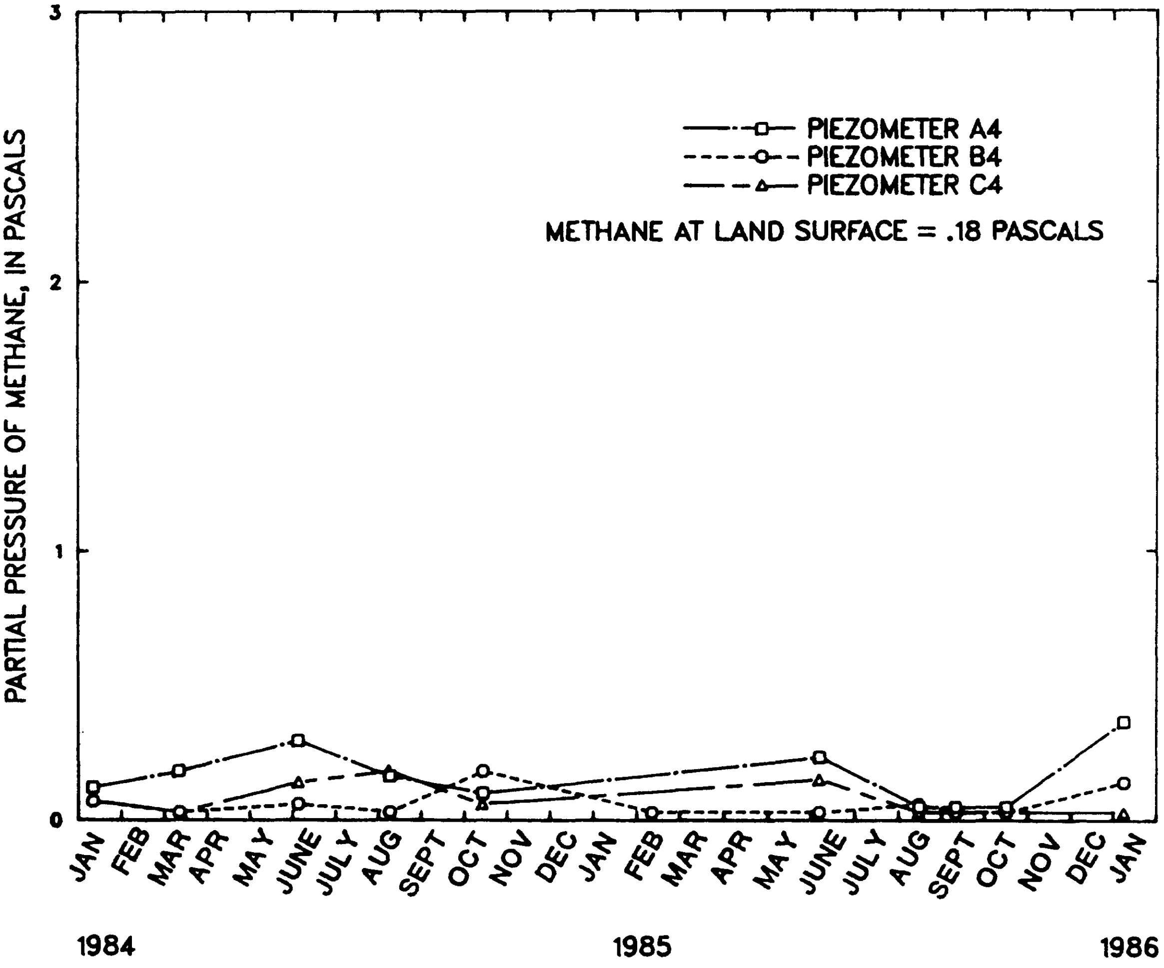


Figure 28.--Temporal changes in partial pressure of methane 3.7 meters below land surface at boreholes A, B, and C.

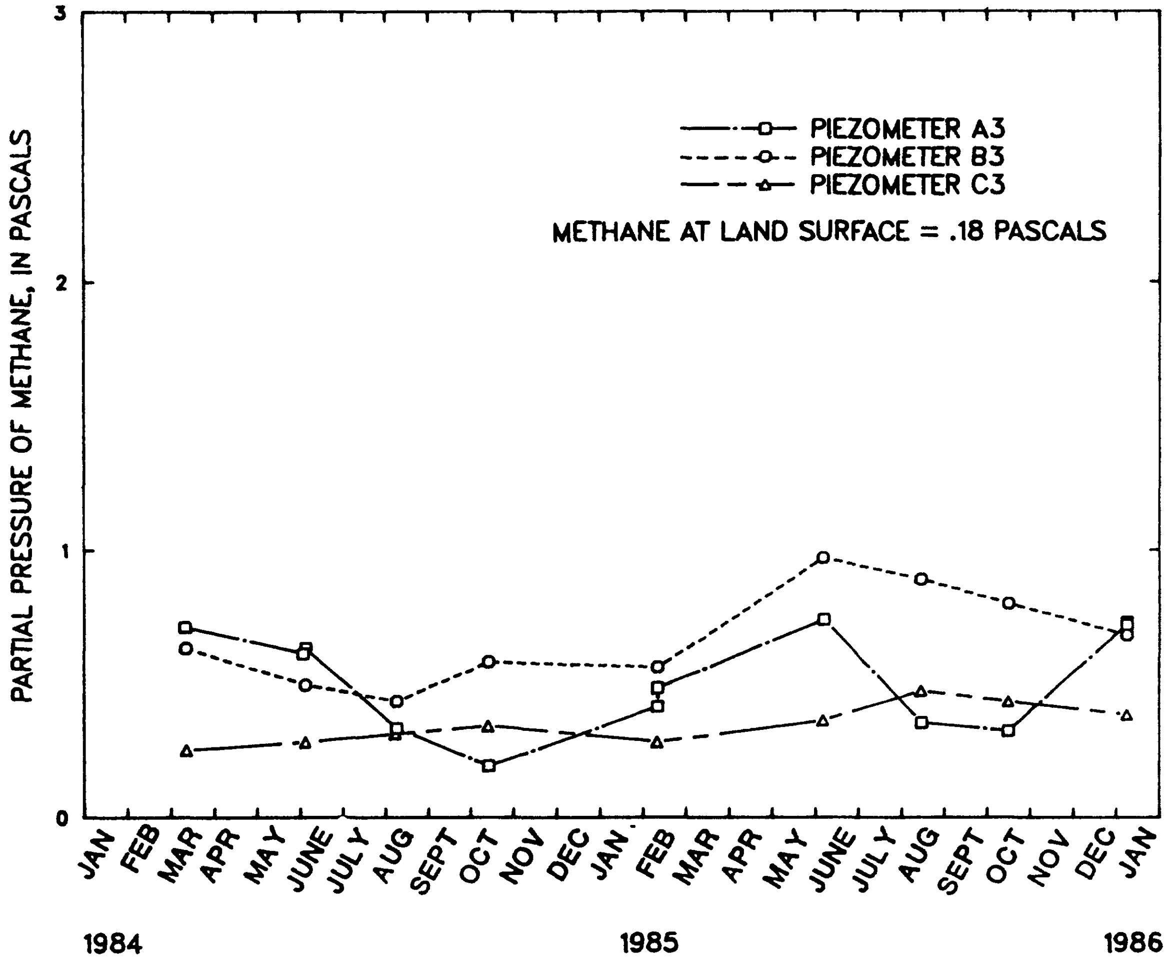


Figure 29.--Temporal changes in partial pressure of methane 7.3 meters below land surface at boreholes A, B, and C.

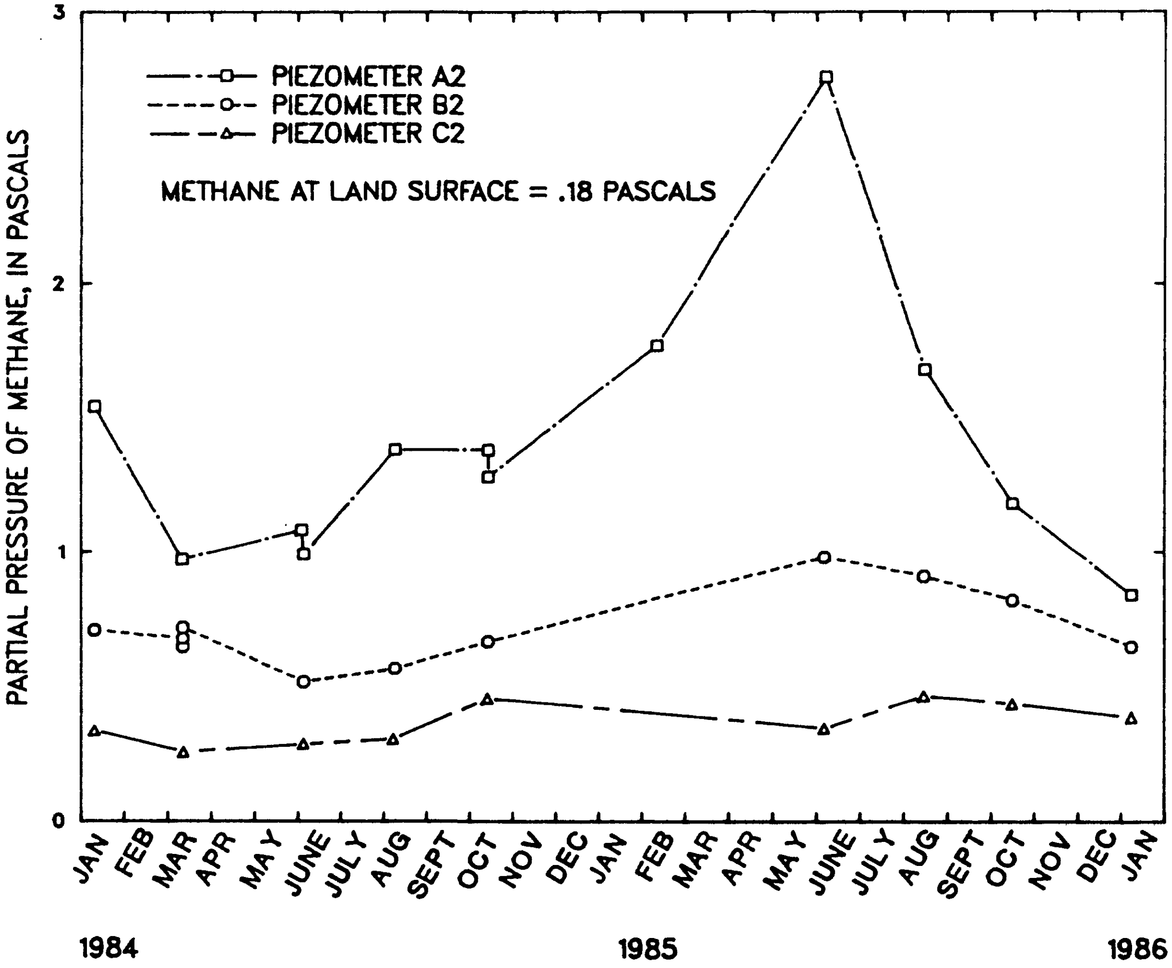


Figure 30.--Temporal changes in partial pressure of methane 11.6 meters below land surface at boreholes A, B, and C.

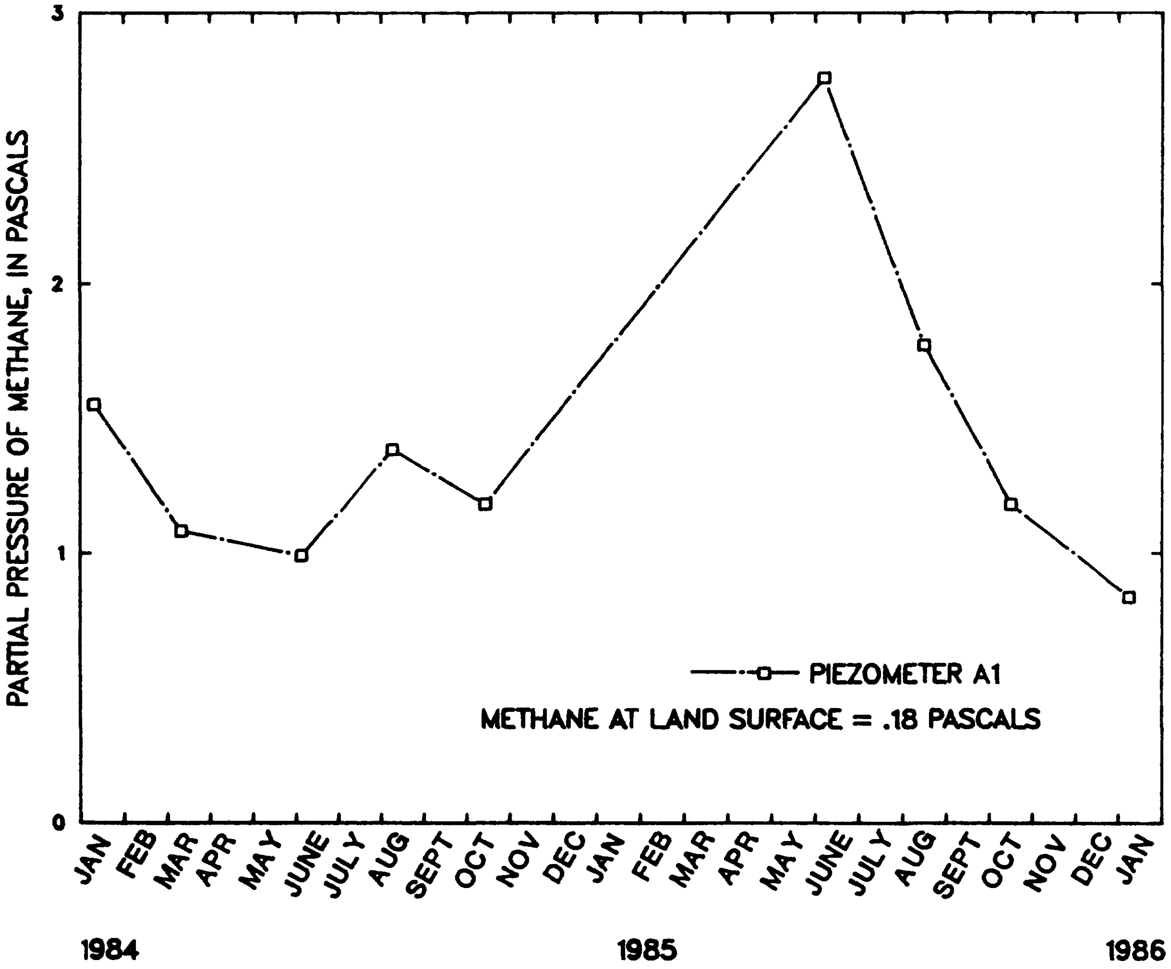


Figure 31.--Temporal changes in partial pressures of methane 13.6 meters below land surface at borehole A.

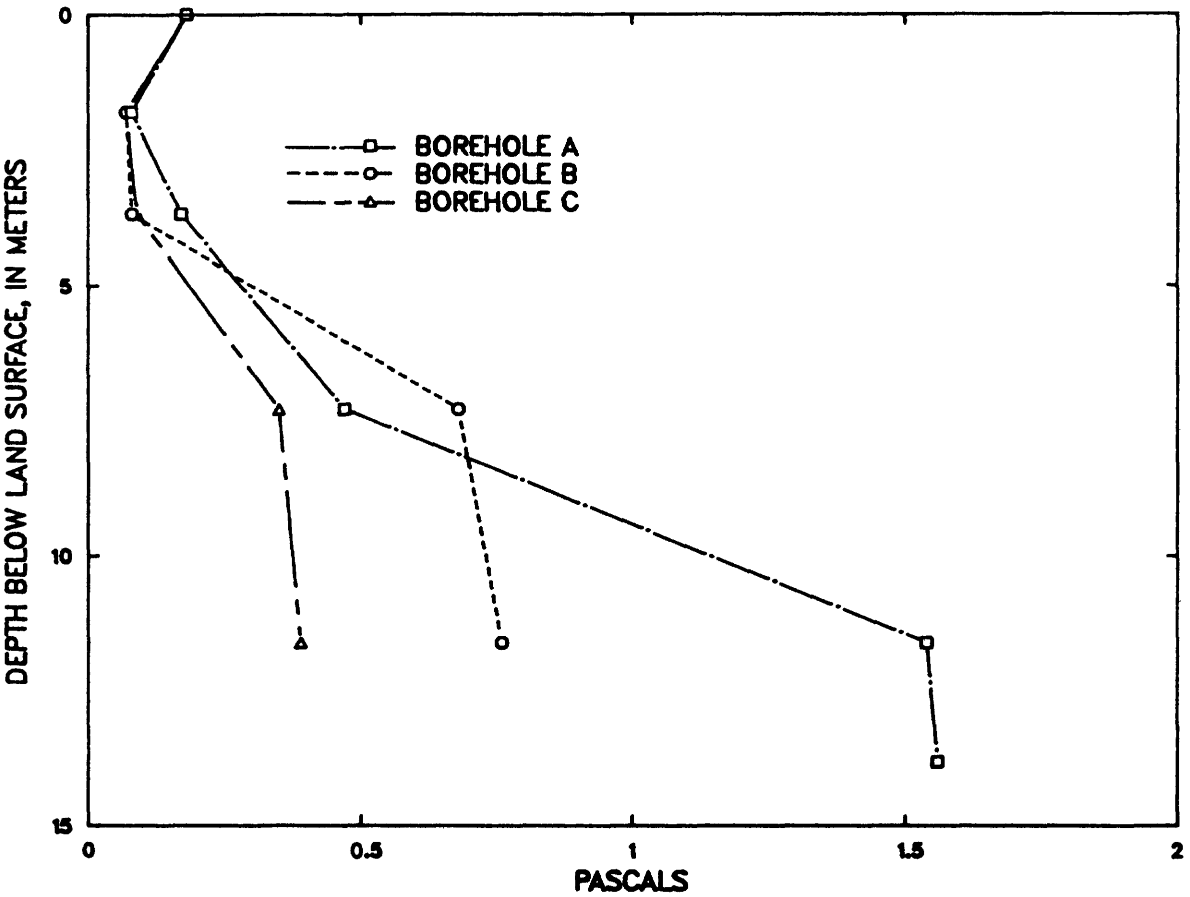


Figure 32.--Changes in the time-averaged mean partial pressure of methane at boreholes A, B, and C as a function of depth.

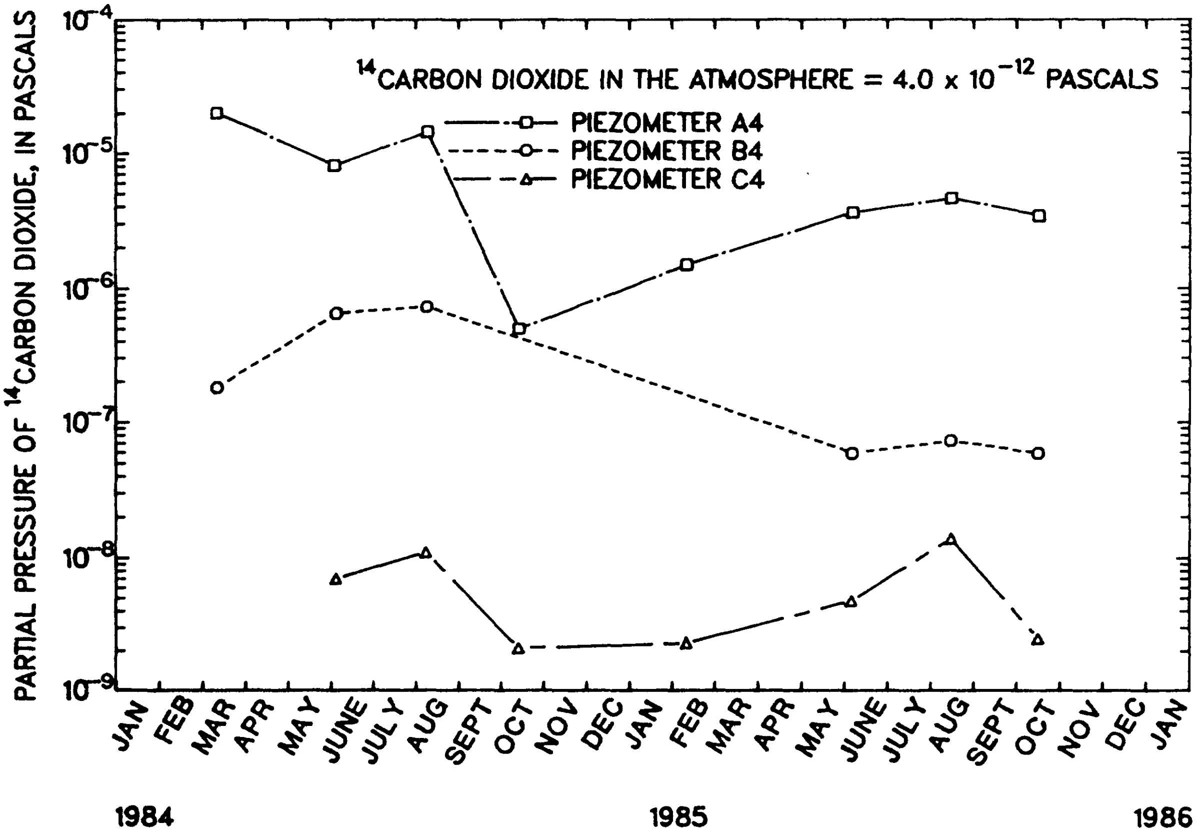


Figure 33.--Temporal changes in partial pressure of ^{14}carbon dioxide 3.7 meters below land surface at boreholes A, B, and C.

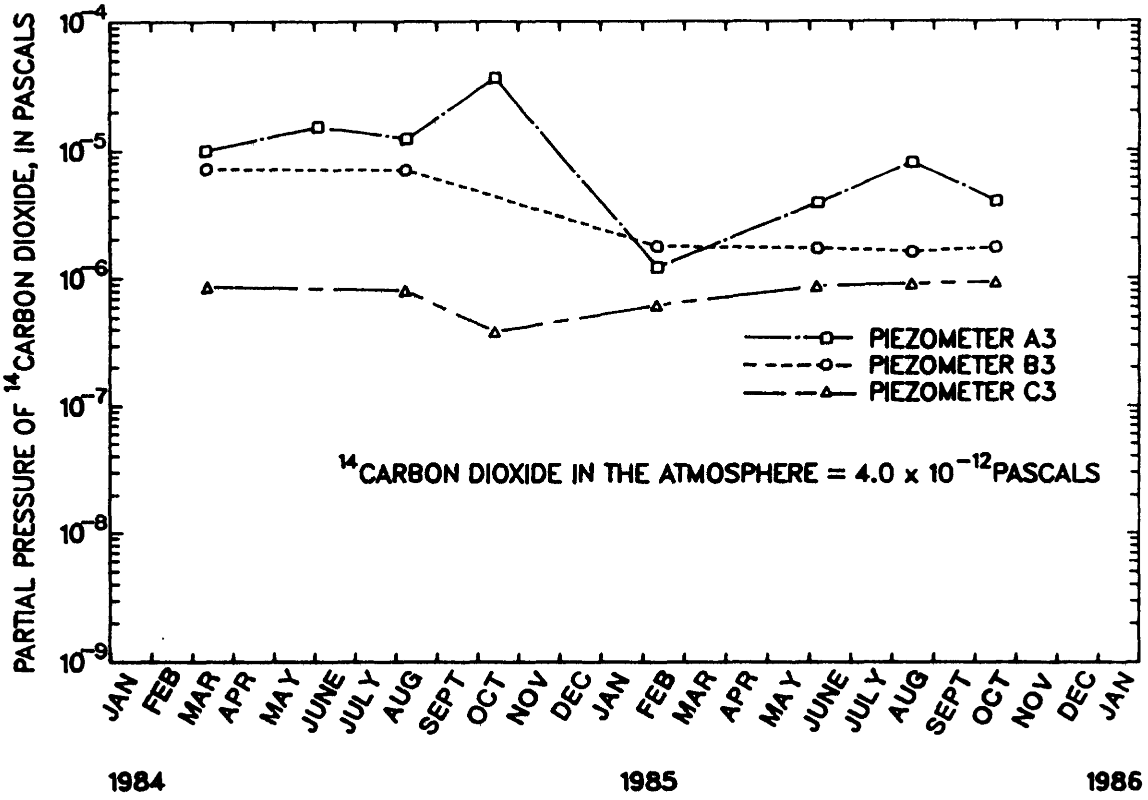


Figure 34.--Temporal changes in partial pressure of ^{14}carbon dioxide 7.3 meters below land surface at boreholes A, B, and C.

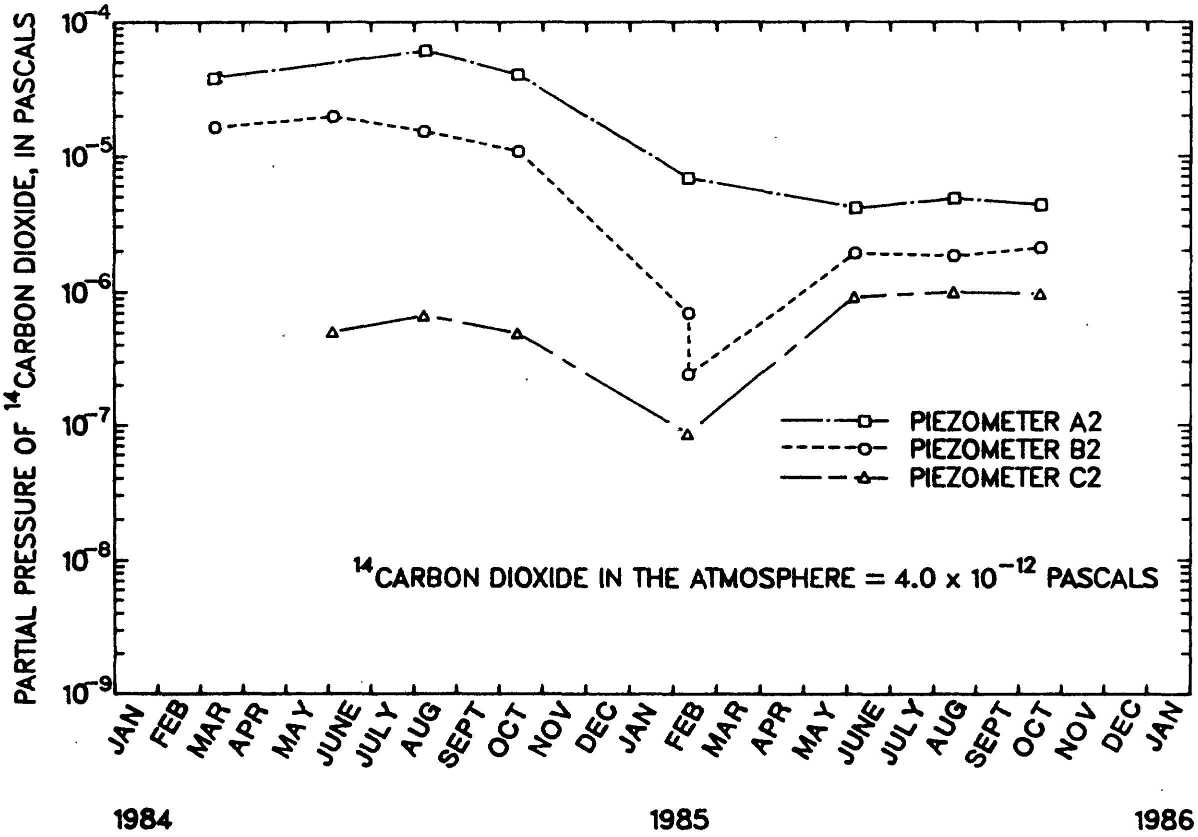


Figure 35.--Temporal changes in partial pressure of ^{14}carbon dioxide 11.6 meters below land surface at boreholes A, B, and C.

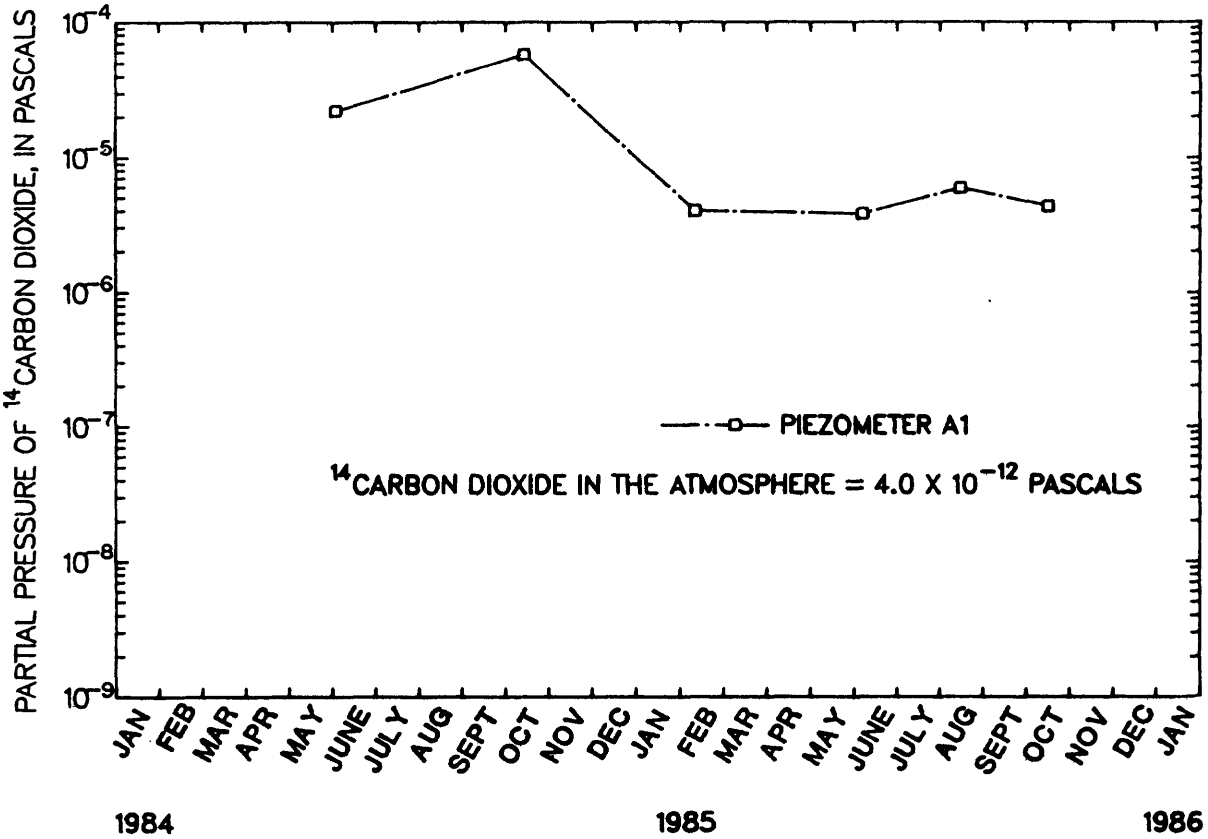


Figure 36.--Temporal changes in partial pressure of ^{14}carbon dioxide 13.6 meters below land surface at borehole A.

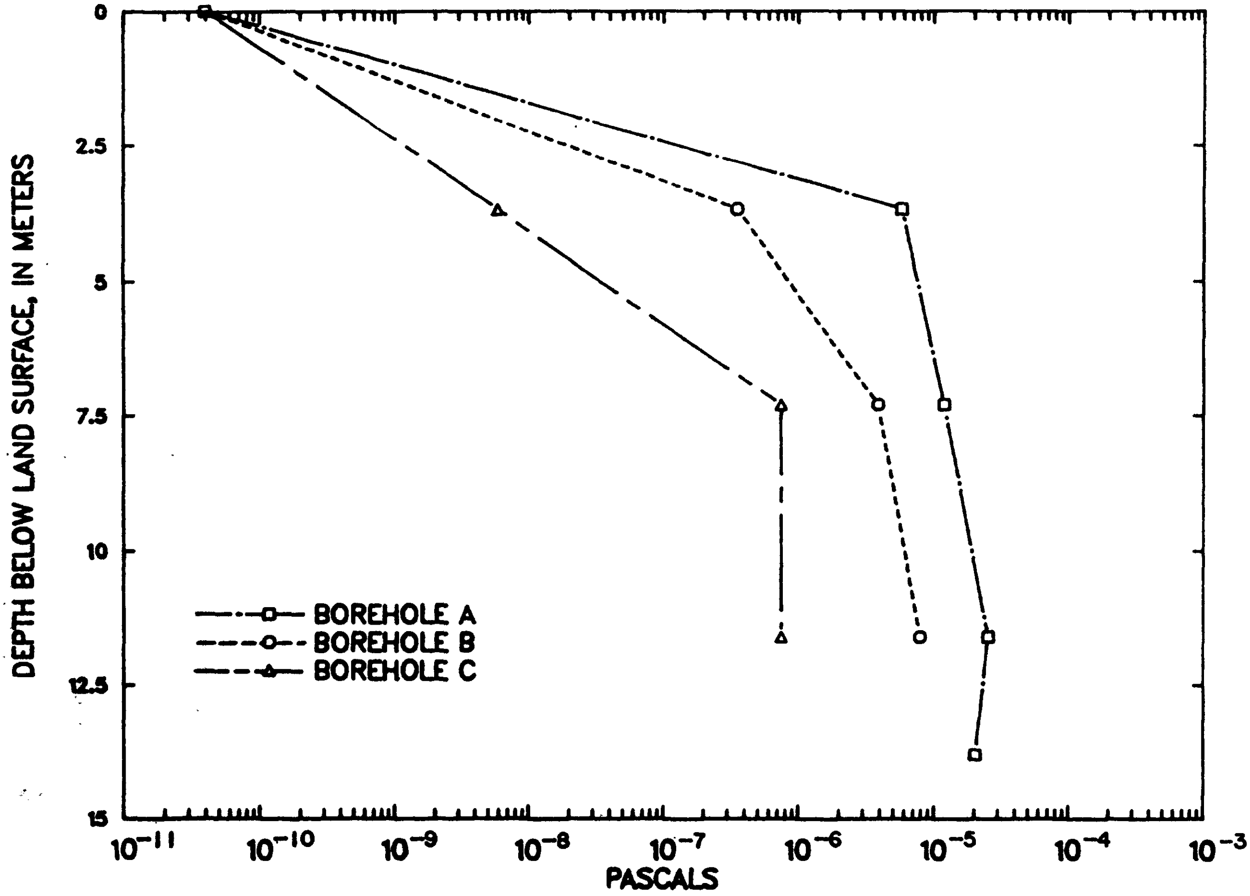


Figure 37.--Changes in the time-averaged mean partial pressure of ^{14}carbon dioxide at boreholes A, B, and C as a function of depth.

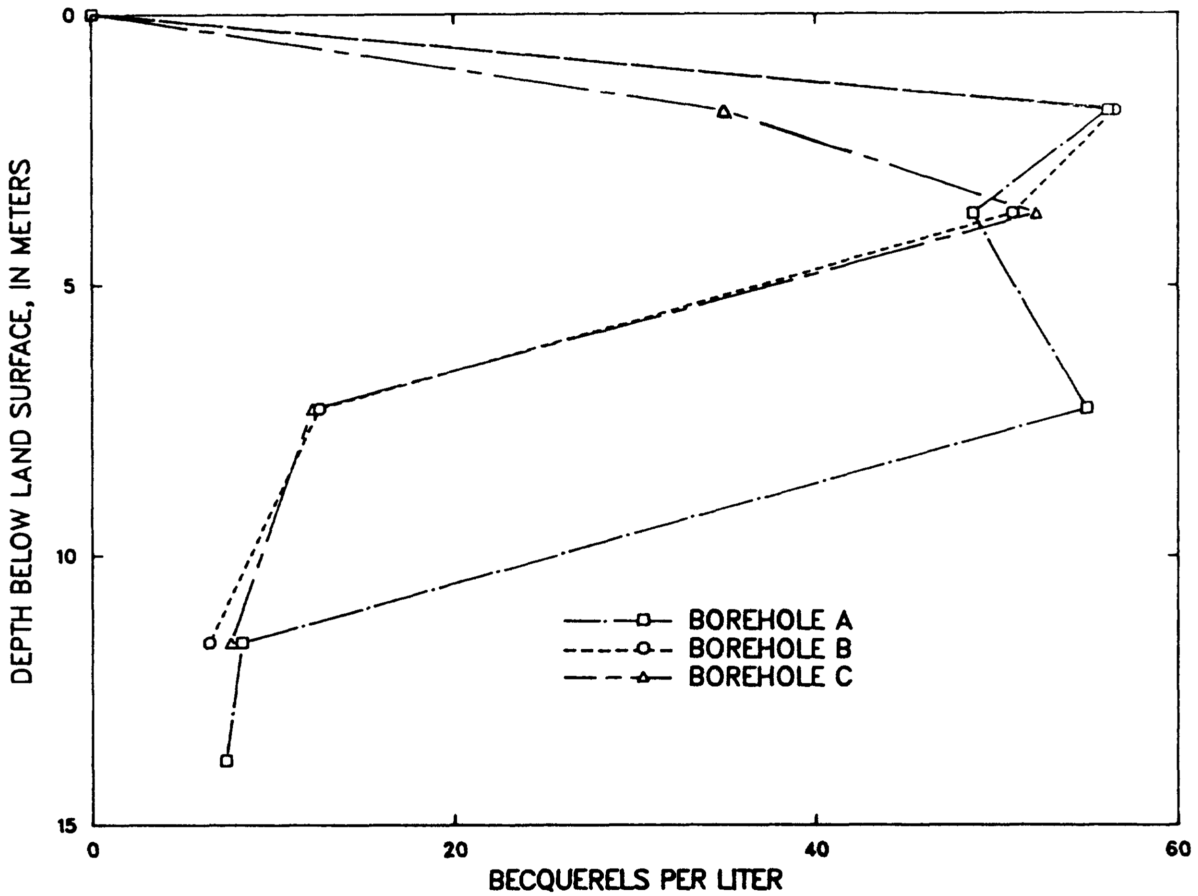


Figure 38.--Changes in the time-averaged mean activities of ^{222}radon at boreholes A, B, and C as a function of depth.

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