SPATIAL AND TEMPORAL VARIATIONS OF SOIL GAS ^{220}RN AND ^{222}RN AT TWO SITES IN NEW JERSEY*

Adam R. Hutter
Environmental Measurements Laboratory, U.S. Department of Energy
New York, NY

ABSTRACT

Soil gas ^{220}Rn and ^{222}Rn concentrations have been measured at sites in Chester and Aberdeen, New Jersey. Two years of ^{222}Rn and ^{220}Rn data were obtained from a depth of 0.85 m, followed by two subsequent years from depths of 0.28, 0.56, 0.85 and 1.28 m. ^{220}Rn and ^{222}Rn variations before the first winter that sample tubes were installed were larger than later, indicating that the soil structure, disturbed during installation of the tubes, may significantly redistribute after the first winter, thus ensuring that the sample is drawn from near the bottom of the tube. At the Chester site, autumn ^{222}Rn concentrations were found to be up to 10 times higher than winter values, variations larger than predicted assuming diffusion-only transport. Spatial variations up to an order of magnitude are observed over distances of a few meters. ^{220}Rn concentrations are typically ~ 2 to 3 times higher during summer than during winter. At the Aberdeen site, ^{220}Rn and ^{222}Rn concentrations were about an order of magnitude less than the lowest Chester site values, with no statistically significant temporal or spatial variations observed. Permeability measurements, thought to be an indicator of parameters controlling soil gas ^{222}Rn variations, show no correlation with ^{220}Rn or ^{222}Rn at either site.

INTRODUCTION

The element Rn, discovered in 1900 by Dorn who named it radium emanation, is a gaseous product of the U and Th decay series. There are 20 known isotopes of Rn, with ^{222}Rn, ^{220}Rn, and ^{219}Rn being alpha emitters. ^{222}Rn, colloquially referred to as radon, has a half-life of 3.825 days. ^{220}Rn, commonly termed thoron, has a half-life of 54.5 seconds. ^{219}Rn, known as actinon, has a half-life of 3.92 seconds, too short to be observable in most environmental situations.

It has been estimated that ~ 10% (or ~ 13,000 per year) of all lung cancer deaths in the U.S. are directly attributable to exposure to Rn and its decay products (Lubin and Boice 1989). Both ^{220}Rn and ^{222}Rn are significant contributors to the indoor radiation environment; it has been estimated that 10-20% of the effective dose equivalent from all indoor Rn progeny is due to ^{220}Rn progeny (UNSCEAR 1982). The source of indoor Rn in the majority of U.S. homes is the soil gas (Nero and Nazaroff 1984; Sachs et al. 1982; Li et a;. 1992). In addition to studying the source characteristics and dynamics of Rn, the data from the current study will be used in future studies to address the need for a greater understanding of soil gas transport mechanisms, in light of the critical need to understand the migration of pollutants in the vadose zone (Narasimhan et al. 1990; Dorr and Munnich 1990).

This paper details a project aimed to enhance our understanding of the factors, and their interactions, that influence the temporal and spatial variations in soil gas ^{220}Rn and ^{222}Rn, with an emphasis on applying the results to problems of soil gas transport in general. Therefore, the specific goals of this study are to: (1) develop the first data set of long-term variations of soil gas ^{220}Rn concentrations; (2) add to the understanding of long-term variations in soil gas ^{222}Rn concentrations; and (3) study certain aspects of geological setting and soil dynamics that influence soil gas ^{222}Rn and ^{220}Rn concentrations.

The first study to establish that ^{222}Rn and ^{220}Rn are contained in soil gas was performed by Elster and Geitel (Elster and Geitel 1902). Recent investigations of soil gas concentrations of ^{222}Rn have shown temporal and spatial variations to be large and often not well understood showing: (1) high summer and low winter ^{222}Rn (Rose et al. 1990); (2) high winter/spring and low summer/fall (Klusman and Jaacks 1987; Schumann et al. 1989); and (3) no annual pattern (Fleischer et al. 1980).

The dominant process of ^{222}Rn migration under most conditions has generally been accepted to be diffusion (Tanner 1980; Schroeder 1965). The diffusion length (defined as the distance in which an e-fold decrease in ^{222}Rn concentration occurs) depends upon the diffusion coefficient and the ^{222}Rn decay constant. The diffusion coefficient depends on soil parameters and atmospheric variables such as humidity, porosity, barometric pressure, wind, precipitation, etc. (Kvasnicka 1980), and can be theoretically determined from the soil water saturation fraction (Rogers and Nielson 1991). For dry soil of normal porosity, a typical diffusion length is ~ 1 m, which translates into an upper limit of ^{222}Rn transport on the order of a few meters (Tanner 1964). However, it has been shown that in soils with connected air-filled pores, bulk flow processes may be locally significant (Fleischer and Mogro-Campero 1979). If ^{222}Rn migration is caused by a flow of soil air, i.e., advection, ^{222}Rn transport distances can exceed 100 m (Malmqvist and Kristiansson 1984). It is thought that ^{222}Rn entry into houses is largely controlled by advection resulting from a pressure stack effect induced by a temperature difference between the building and soil (Nazaroff 1992).

The soil gas ^{222}Rn concentration described by simple diffusion can be modified by many factors, including temperature (Okabe 1956; Kraner et al. 1964; Wilkening et al. 1972; Megumi and Mamuro 1973; Colle et al. 1981; Washington and Rose 1992), barometric pressure (Rudakov 1985; Clements and Wilkening 1974), precipitation (Fleischer and Mogro-Campero 1979), wind direction and speed (Kraner et al. 1964), soil moisture (Washington and Rose 1990), soil porosity and permeability (Nazaroff and Sextro 1989). Additionally, bedrock type and structure, as well as uranium and uranium progeny concentrations and distributions in the rock and soil, can also affect ^{222}Rn concentrations (Washington and Rose 1992).

A model incorporating temperature effects of soil gas ^{222}Rn concentration variations is given by Washington and Rose (1990) for depths below which diffusional transport to the surface is negligible, i.e., ~ 1 m:

$$C_{Rn} = 10^3 C_{Ra} E\rho_b / P[F(K_T - 1)+1] \quad (1)$$

where: C_{Rn} = ^{222}Rn concentration, Bq m^{-3}; C_{Ra} = parent concentration, Bq kg^{-1}; E = emanation coefficient; ρ_b = dry bulk density; F = water saturation fraction; K_T = partition coefficient of ^{222}Rn between unit volumes of air and water, C_{water}/C_{air} (values vary from 0.525 at 0 °C to 0.226 at 25 °C (Battino 1979)); P = volume fraction of total pore space (porosity). Variations in the ^{222}Rn concentration of up to a factor of about four can be accounted for due to effects of F and K_T. For a warm (25 °C) and moist (F = 0.95) soil with C_{Ra} = 30 Bq/kg, E = 0.2, and ρ = 1.5 g cm^{-3}, the ^{222}Rn concentration is calculated to be 78.3 kBq m^{-3} (2116 pCi L^{-1}), but when cold (0°C) and dry (F = 0.05), the ^{222}Rn concentration is calculated to be 21.2 kBq m^{-3} (573 pCi L^{-1}).

MATERIALS AND METHODS

Site descriptions

Fig. 1 shows the locations of the two New Jersey field sites. The Chester site is located on the edge of the Reading Prong in central northern New Jersey. The site is situated in an open grassy field on the side of a hill, and is underlain by granitic gneiss. Solid-walled , stainless steel tubes of 0.95 cm I.D. and 0.85 m length were permanently installed at six sampling locations at this site in September, 1989. These tubes were emplaced so that they are flush with the surface, and flexible tubing is attached for sample connections. The sampling tubes (referred to as positions 1, 2, 3a, 4a, 5 and 6) are each spaced linearly ~ 3 m apart, with an additional sampling tube (3b and 4b) placed at two of the locations. Positions 3a and 3b are ~ 6 cm apart, as are positions 4a and 4b. Additionally, in August, 1992, tubes were emplaced at positions 2 and 5 for sampling depths of 0.28, 0.56 and 1.28

m. These positions were chosen because of differences observed in the first 2 years of data, specifically, position 2 showed strong seasonal variations in the soil gas ^{220}Rn and ^{222}Rn concentrations, whereas position 5 did not.

The Aberdeen site is located on the border of the Coastal Plain and Piedmont Provinces, with one layer of coastal plain sediments, mainly sand and gravel, to a depth of ~ 20 m. Four sampling tubes (0.95 cm I.D., 0.85 m length), arranged linearly and spaced ~ 3 m apart (referred to as positions 1, 2, 3, and 4a) with one additional sampling tube (referred to as position 4b), were emplaced in May, 1990. The sampling tubes at positions 4a and 4b are ~ 6 cm apart. Tubes for 0.28, 0.56 and 1.28 m depth sampling were emplaced at positions 4a and 4b in November, 1993. There is virtually no vegetation in the area surrounding the positions at the Aberdeen site.

Sampling

The field procedures used in this study are reported in the EML Procedures Manual (in press). Briefly, a soil gas sample is collected into a 165 mL scintillation cell using a battery-operated pump at a flow rate of ~ 2 L min^{-1} for 1 min. Sampling was performed on a bi-weekly frequency and the samples were analyzed for ^{220}Rn and ^{222}Rn. Additionally, a flow meter and pressure gauge were used to estimate soil permeability in order to indirectly determine the effects of changes in soil moisture and porosity on the ^{220}Rn and ^{222}Rn concentrations (Nazaroff and Sextro 1989). Permeabilities of typical soils range over six orders of magnitude, with diffusion being the dominant process at the low end and advection being dominant at the high end. The soil permeability (k) for each position was estimated from flow and pressure differential measurements at the time of sampling using the following equation (Rogers and Nielson 1991):

$$k = (5.2 \times 10^{-4} \bullet Q) / \Delta P \qquad (2)$$

where k = soil permeability (m^2), Q = flow rate ($m^3\ s^{-1}$), and ΔP = suction pressure (Pa)

^{220}Rn and ^{222}Rn Analysis

The methodology for the ^{220}Rn and ^{222}Rn analyses used in this project is detailed and discussed in Hutter (1995). In summary, the method requires a 1-min count as soon as the sample has been drawn into a scintillation cell, and a 5- or 10-min count at least 5 minutes after the soil gas sample has been obtained. The ^{222}Rn concentration is determined from the second count. Once the ^{222}Rn concentration is determined, the counts due to ^{222}Rn and progeny during the 1-min count can be calculated and subtracted from the total in the 1-min count. The remaining counts in the 1-min count are due to ^{220}Rn and progeny. The overall uncertainty when using this method to measure typical soil gas ^{220}Rn and ^{222}Rn concentrations, i.e., > ~ 3 kBq m^{-3} (80 pCi L^{-1}) was calculated to be 18.9% and 10.6% (90% confidence levels), respectively, determined from analyses of duplicate field measurements (180 for ^{222}Rn and 138 for ^{220}Rn). The lowest ^{220}Rn concentrations that can be measured using this technique while maintaining an overall error no greater than about 30% are ~ 500 Bq m^{-3} (13 pCi L^{-1}). The ^{220}Rn measurement uncertainty can be decreased if multiple measurements are obtained and are the results arithmetically averaged.

In order to better characterize each site, the soil depth and/or depth to bedrock was determined using an electrical resistivity method. At the Chester site, the soil thickness was found to be ~ 2 m, below which is granitic gneiss bedrock. At the Aberdeen site, there is a > 20 m thick single-layer of Cretaceous lignitic sands, below which is assumed to be basement rock.

Quality Assurance

Quality assurance of the soil gas ^{220}Rn and ^{222}Rn data was assessed by the following tasks:

(1) Calibrations performed on a semi-annual basis in the EML Radon, Thoron, and Progeny Exposure Facility following standard procedures (HASL-300 1992);
(2) background measurements performed on all cells for every measurement;
(3) duplicate measurements performed at a rate of 1 in 10;
(4) replicate counting of samples at a rate of 1 in 10; and
(5) error analysis of soil gas ^{222}Rn and ^{220}Rn measurements.

RESULTS

In order to measure the reproducibility of any observed seasonal variations, at least 2 years of data for any sampling location is needed. Soil gas ^{222}Rn measurements were obtained from the Chester site beginning in September, 1989 and continue to the present. Soil gas ^{220}Rn measurements began in March, 1990 at this site. The ^{222}Rn and ^{220}Rn data, as well as the soil permeability data, collected over this period at a depth of 0.85 m from the Chester site, are presented in Fig. 2. At the Aberdeen site, soil gas ^{222}Rn and ^{220}Rn concentrations and permeability data, obtained at a 0.85 m depth from May, 1990 until May, 1992, are presented in Fig. 3.

The soil gas ^{220}Rn and ^{222}Rn data from the profiled samples, i.e., depths of 0.28, 0.56, 0.85 and 1.28 m (at positions 2 and 5 at the Chester site, positions 4a and 4b at the Aberdeen site) are shown in Figs. 4 through 7, while Fig. 8 shows the soil permeability data obtained at these positions during these times.

Table 1 shows a statistical summary of ^{220}Rn and ^{222}Rn concentrations for the Chester, NJ site. Fourier analyses were performed on the Chester data in order to examine any seasonal cycles. The ratio of high to low values calculated following these analyses are shown in Table 1, as well as the day of the year when the maximum concentration was found following a fit of the transformed data.

Table 1 Analysis of ^{220}Rn and ^{222}Rn data obtained from Chester, NJ site calculated from data obtained from September 1989 to May, 1995.

Position	Isotope	Depth (m)	Min (kBq m^{-3})	Max (kBq m^{-3})	Mean (kBq m^{-3})	StDev (kBq m^{-3})	High-Low Fourier Fit	Max D.O.Y.
1	^{222}Rn	0.85	119	483	268	98	2.2	355
	^{220}Rn	0.85	133	302	210	40	1.4	220
2	^{222}Rn	0.28	7	419	108	100	17.5	285
	^{220}Rn	0.28	38	280	160	57	2.1	215
	^{222}Rn	0.56	34	557	188	132	7.9	295
	^{220}Rn	0.56	68	505	186	68	2.5	215
	^{222}Rn	0.85	31	832	276	196	8.4	295
	^{220}Rn	0.85	62	824	180	59	1.8	240
	^{222}Rn	1.28	419	1344	775	201	1.5	280
	^{220}Rn	1.28	224	1112	480	148	1.5	240
3a	^{222}Rn	0.85	107	1672	937	292	1.7	345
	^{220}Rn	0.85	170	944	487	146	1.8	255
3b	^{222}Rn	0.85	82	1125	607	234	1.6	325
	^{220}Rn	0.85	158	709	351	122	1.8	350
4a	^{222}Rn	0.85	41	723	298	163	2.2	275
	^{220}Rn	0.85	103	658	280	106	1.5	235
4b	^{222}Rn	0.85	25	621	267	162	1.9	280
	^{220}Rn	0.85	118	447	253	75	1.1	230
5	^{222}Rn	0.28	18	425	107	84	-	-
	^{220}Rn	0.28	31	245	142	49	1.5	210
	^{222}Rn	0.56	11	477	175	100	-	-
	^{220}Rn	0.56	85	600	197	76	1.7	218
	^{222}Rn	0.85	42	678	354	128	1.3	270
	^{220}Rn	0.85	86	433	242	80	1.6	225
	^{222}Rn	1.28	267	994	636	142	1.1	270
	^{220}Rn	1.28	111	806	296	111	1.2	275
6	^{222}Rn	0.85	131	699	339	123	1.4	330
	^{220}Rn	0.85	152	494	297	76	1.5	220

DISCUSSION

Chester, NJ site

A first inspection of the data shows some obvious trends (Fig. 2a). ^{222}Rn concentrations vary among all the positions by nearly two orders of magnitude. The values observed ranged from a low of 30 kBq m^{-3} to a high of 1672 kBq m^{-3} (810 to 45,200 pCi L^{-1}). Position 3a had the highest ^{222}Rn concentration for each measurement day, followed closely by 3b, two positions that were only ~ 6 cm apart. Positions 4a and 4b, on the other hand, typically had the lowest ^{222}Rn values. Positions 5 and 6 were normally higher than positions 1 and 2. These consistent spatial variations are probably due to inhomogeneous parent nuclide concentration in the soil, rather than due to variables such as soil moisture changes. The parent nuclide concentrations have not been measured at each position because doing so would require permanent destruction of the soil, and studies are ongoing. However, surface gamma-spectra show inhomogeneity at this site, which is assumed to be present at depth as well (Miller pers comm.).

The highest correlations in the ^{222}Rn concentrations at the Chester site are between duplicate pair positions, specifically, positions 4a and 4b, and positions 3a and 3b, with correlation coefficients (r) of 0.96 and 0.85, respectively. A trend of decreasing ^{222}Rn correlation coefficients with increasing distance between the positions is also observed. For example, the highest ^{222}Rn correlation coefficient for position 1 is with position 2 ($r \sim 0.8$), with the next nearest positions, 3 and 4, having smaller correlation coefficients ($r \sim 0.6$), and positions 5 and 6 even less ($r \sim 0.3$). This trend is observed in nearly all the ^{222}Rn data for each position.

Seasonal cycles in the ^{222}Rn concentrations can be seen with the lowest values occurring approximately from December until March, with increasing values from early spring until late autumn when the highest values are observed. ^{222}Rn concentrations from position 2 show the strongest seasonal trend with a greater than 10-fold increase from low to high seasons. Positions 4a and 4b show about a five-fold variation, whereas positions 1, 3a, and 3b show more modest seasonal variations on the order of two to three fold-variations from high to low values. Positions 5 and 6 show suspect, at best, seasonal cycles.

Another observation with important consequences occurs in the data when the tubes were first emplaced in the fall of 1989. As can be seen, until the winter period ended, the variations observed in the soil gas ^{222}Rn concentrations are very large compared to after this period. The widely variable concentrations are thought to be due to the disruption of the soil structure when the tubes were emplaced. During these first few months, there may have been cavities along the outside tube walls, that allowed soil gas from shallower depths, with lower concentrations, to reach the end of the tube and be sampled. After the freezing and thawing of the soil occurred during the winter months, the soil may have settled sufficiently to seal these cavities and ensure that the soil gas being sampled was from near the 0.85 m depth of the tube end. Soil permeability variations (see Figs. 2 and 8) corroborate this hypothesis. These data indicate that a one-time sampling of the soil gas by emplacing a tube into the soil and taking a sample of the soil gas may not give an accurate assessment of the soil gas ^{222}Rn concentration from the depth of sampling.

The ^{220}Rn data for the Chester site look very different than the ^{222}Rn data (Fig. 2b). The spatial variations among the positions are much less, with a total range of 62 to 944 kBq m^{-3} (1675 to 25500 pCi L^{-1}), and also with less temporal variations. Seasonal cycles can be observed at some of the positions, i.e., positions 2, 3a and 4b, with two to three-fold variations, but not at others. The seasonal variations in ^{220}Rn are not similar to the observed ^{222}Rn variations. The ^{220}Rn concentrations are again lowest during the winter months, but are highest in the summer months, gradually decreasing during the fall, in contrast to the ^{222}Rn concentration variations that increased virtually all summer and into the fall. The magnitude and timing of these observed seasonal cycles in the ^{220}Rn concentration closely match trends expected assuming diffusion-only transport. However, even if advective processes were occurring in the soil gas at the Chester site, the transport distance of ^{220}Rn is so short, due to its short half-life, that the seasonal cycles would still be expected to follow diffusional equations.

The ^{220}Rn correlation coefficients between positions for the Chester site do not exhibit the same trends as observed for ^{222}Rn. The ^{220}Rn correlation coefficients are smaller than for ^{222}Rn, with the highest being ~ 0.6, and they average 0.47 for all the positions with a standard deviation of 0.13. Additionally, there are no discernible trends with spatial distribution, which is to be expected since there were fewer temporal and spatial variations in the cumulative data for ^{220}Rn than for ^{222}Rn.

In an attempt to understand the relative importance of the measured factors affecting the observed ^{222}Rn and ^{220}Rn variability in the soil gas, correlations were performed for all of the positions at the Chester site for each measured parameter (^{222}Rn, ^{220}Rn and permeability) with available on-site meteorological data, i.e., temperature, barometric pressure, wind speed, and relative humidity. Unfortunately, sufficient meteorological data is only available for the 1990 - 1992 period, coinciding with the period when data was obtained at the 0.85 m depth. These analyses do not show strong correlations between Rn concentrations and the meteorological parameters. A few of the correlations of temperature with ^{222}Rn and/or ^{220}Rn have an r value of about 0.5, expected due to the seasonal variations in the ^{222}Rn and ^{220}Rn concentrations observed at some of the positions. The barometric pressure shows virtually no correlation with any of the other measured parameters. Relative humidity effects on the soil gas ^{222}Rn and ^{220}Rn concentrations are minimal, at best. Poor correlations among the meteorological parameters and soil gas ^{222}Rn and ^{220}Rn concentrations were expected, since atmospheric and meteorological effects on the soil gas ^{222}Rn and ^{220}Rn concentrations are small at the sampling depth of 0.85 m. If sufficient meteorological data were available for subsequent years, the variations observed in ^{222}Rn concentrations at shallow depths would be expected to correlate better with temperature and pressure than was observed for the 0.85 m data.

In 1992 tubes of 0.28, 0.56 and 1.28 m depth were added to the existing 0.85 m depth tubes at positions 2 and 5 at the Chester site. These profiled ^{220}Rn and ^{222}Rn data show consistent trends with the earlier data from the 0.85 m depth (see Figs. 4 and 5). For instance, variations in the ^{220}Rn and ^{222}Rn soil gas concentrations were much larger during the first few months that the tubes were installed compared to after the first winter, as were soil permeability variations. Seasonal patterns are found at position 2 for depths 0.28, 0.56 and 0.85 m, but not at the 1.28 m depth, where over a 2-year period the arithmetic mean is 775 kBq m^{-3} with a standard deviation of 200 kBq m^{-3}, which is only about twice the uncertainty in a single ^{222}Rn measurement. The magnitude of the seasonal ^{222}Rn concentration variations decrease with increasing depth at both positions. For position 2 at the 0.28 m depth, the seasonal variation is > 10 fold, whereas at the 0.56 and 0.85 m depths the seasonal variations are ~ 7 to 8 fold. The pattern of increasing ^{222}Rn concentration throughout the summer and fall, with a drastic decrease during early winter, observed in the earlier 0.85 m data, is followed at the 0.28, 0.56, and 0.85 m depths. The seasonal variations in the ^{220}Rn concentrations are more modest, ~ 2 fold, with the highest concentrations occurring during the summer. In both the ^{220}Rn and ^{222}Rn profiled data, there is a marked difference in concentration magnitude at the 1.28 m depth compared to the shallower depths. This pattern is attributed to a soil horizon change, which typically occurs at ~ 1 m for this soil type and region.

As is consistent with the previous data obtained at the 0.85 m depth at position 5, no seasonal patterns are discernible in the profiled data at this position (see Fig. 5a). The ^{222}Rn concentration increases with increasing depth, with arithmetic means and standard deviations of 109 ± 84, 175 ± 100, 354 ± 128 and 636 ± 142 kBq m^{-3} at depths of 0.28, 0.56, 0.85 and 1.28 m, respectively. The ^{220}Rn concentrations, shown in Fig. 5b, also show increases with depth, with arithmetic means and standard deviations of 142 ± 49, 197 ± 76, 242 ± 80 and 296 ± 111 kBq m^{-3} at depths of 0.28, 0.56, 0.85 and 1.28 m, respectively. Due to its short half-life and limited transport distance, the ^{220}Rn sampled was generated very close to the end of the sampling tube, in comparison to ^{222}Rn that may have been transported significant distances. From the ^{220}Rn and ^{222}Rn data, there seems to be a soil horizon change between the 0.85 m and 1.28 m depth.

The mechanism of transport mode has been hypothesized to be largely dependent upon the permeability of the soil, i.e., soil permeability may be a good indicator of the dominant transport mode. At permeabilities > 10^{-11} m^2, advective transport is thought to occur, whereas diffusive transport is thought to be dominant at permeabilities < 10^{-11} m^2 (Nazaroff and Sextro, 1989). Since ^{222}Rn variations are observed that are larger than can be explained by diffusion alone, a correlation of ^{222}Rn with permeability was expected to show that advection may be important as a

control of soil gas ^{222}Rn variations. These correlations are, however, for the most part, very poor, e.g., $r < 0.25$ (soil permeability data are presented in Figs. 2 and 8). These poor correlations suggest that either soil permeability, as measured using the technique described, is not a good indicator of parameters thought to have strong effects on the soil gas ^{222}Rn and ^{220}Rn concentrations (e.g., soil moisture, porosity), or that these factors themselves are not as important in controlling the soil gas ^{222}Rn and ^{220}Rn concentrations as has been thought. With the data obtained it is not possible to test these scenarios.

Aberdeen, NJ site

The magnitude of the ^{222}Rn and ^{220}Rn concentrations obtained at the Aberdeen site are about one order of magnitude less than those observed at the Chester site (see Figs. 3, 6 and 7). In addition, both spatial and temporal variations are much smaller with essentially constant ^{222}Rn concentration values over the entire 4 year period. There is a broader band of ^{220}Rn concentration variation at the Aberdeen site, shown in Figs. 6b and 7b, but this is due to the higher error associated with the ^{220}Rn measurements compared to the ^{222}Rn measurements. The lack of seasonal variations in the ^{222}Rn and ^{220}Rn concentrations is thought to be due to the soil being very well-drained, and thus, having little seasonal variation in soil moisture, thought to be a major factor affecting variations in soil gas ^{222}Rn and ^{220}Rn. At this site, the 1σ of the ^{222}Rn and ^{220}Rn measurements taken as a whole are 19% and 36%, respectively, i.e., 12.2 ± 2.3 kBq m^{-3} (330 ± 63 pCi L^{-1}) and 9.4 ± 3.4 kBq m^{-3} (250 ± 92 pCi L^{-1}). These values are about twice the 1σ error of the duplicate measurements used for the total soil gas measurement uncertainty.

Both the ^{222}Rn and the ^{220}Rn correlation coefficients for the Aberdeen site are small compared to the Chester site (average ~ 0.3), and in general show no strong relationships, except for the ^{222}Rn between positions 2 and 3 ($r \sim 0.7$). No trends among the ^{222}Rn and ^{220}Rn concentrations at the Aberdeen site were expected since the site is fairly homogeneous, i.e., all measurements have essentially been sampled from a normally distributed population.

The profiled ^{220}Rn and ^{222}Rn data, shown in Figs. 6 and 7 (tubes of 0.28, 0.56, and 1.28 m depth were installed in 1993), also show no seasonal cycles. Taking the data obtained from positions 4a and 4b collectively (the tubes are only ~ 6 cm apart) the ^{222}Rn concentration increases with increasing depth, with arithmetic means of 6.6, 8.8, 11.0, and 13.4 kBq m^{-3}, respectively at the 0.28, 0.56, 0.85 and 1.28 m depths. The ^{220}Rn concentrations seem to be similar for all depths, with arithmetic means of 6.9, 4.5, 8.1, and 8.8 at the depths of 0.28, 0.56, 0.85 and 1.28 m, respectively.

Once again, variations in soil gas ^{220}Rn and ^{222}Rn concentrations do not show good correlations with measured soil permeability variations (see Figs. 3 and 8).

CONCLUSIONS

A one-time sample of soil gas for the purpose of measuring ^{222}Rn and ^{220}Rn concentrations as a source term for indoor concentrations may be accurate only to an order of magnitude due to seasonal effects and the disruption of the soil structure.

^{222}Rn concentrations at the Chester site are highest during September/October, corroborating previous research and supporting the speculation that wintertime testing of indoor ^{222}Rn concentrations may not give the most conservative estimate of exposure, as is desired. Some ^{222}Rn variations are too large to be adequately explained by diffusion-only models. Spatial variations in the soil gas ^{222}Rn and ^{220}Rn concentrations can be large (up to an order of magnitude) over distances of ~ 3 m.

At the Aberdeen site, no seasonal ^{222}Rn variations are observed, also corroborating previous research.

Long-term cycles in the soil gas ^{220}Rn concentration have been established at two geologically different sites. The highest soil gas ^{220}Rn concentrations occur during mid-summer, coinciding with predictions from diffusion-only models. The controlling parameters of these variations are thought to be local soil conditions, such as soil moisture, water saturation fraction and temperature, among others.

Permeability measurements, thought to be a major indicator of parameters controlling soil gas ^{222}Rn variations, show no correlation with ^{222}Rn or ^{220}Rn at any of the sites. This perhaps indicates that soil permeability, as measured, does not accurately reflect parameters thought to control soil gas ^{220}Rn and ^{222}Rn, i.e., soil moisture and porosity.

ACKNOWLEDGMENTS

The author would like to thank EML reviewers A. Cavallo, H. Feely, and H. Beck and anonymous reviewers for constructive suggestions which markedly improved the paper. The EML editor, N. Chieco, is thanked for once again paying timely attention to detail. The author is especially grateful to H. Feely for guidance he provided.

* Permission granted by Elsevier Science to re-print this paper from The Science of the Total Environment, Special Issue on the Natural Radiation Environment, presented at the NRE VI International Symposium, June 5-9, 1995, Montreal, Canada, to be published in 1996.

REFERENCES

Battino, R. Radon-222. Clever, H. Lawrence, ed. Krypton, xenon, and radon (IUPAC solubility data series: vol. 2), Pergamon Press. 1979:227 - 241.

Clements, W. E.; Wilkening; M. H. Atmospheric pressure effects on ^{222}Rn transport across the earth-air interface. J. Geophys. Res. 79: 5025-5029; 1974.

Colle, R.; Rubin, R. J.; Knab, L. I.; Hutchinson, J. M. R. Radon transport through and exhalation from building materials: A review. U. S. Dept. of Commerce: Nat. Bur. Stds. Publ. C13.46:1139 (NBS TN 1139), 97 p.; 1981.

Dorr, H.; Munnich, K. O. ^{222}Rn flux and soil air concentration profiles in West Germany. Soil ^{222}Rn as tracer for gas transport in the unsaturated zone. Tellus, 42B:20-28; 1990.

Elster, J.; Geitel, H. Beschreibung des Verfahrens zur Gewinnung vorübergehend radioaktiver Stoffe aus der atomosphärischen Luft. Physik Zs., 3:305 - 310; 1902.

EML Procedures Manual. USDOE Report HASL-300, 27th Edition, Vol. 1. Available from NTIS, U.S. Department of Commerce; 1992.

EML Procedures Manual. USDOE Report HASL-300, 28th Edition, Vol. 1 (in press).

Fleischer, R. L.; Hart, H. R.; Mogro-Campero, A. Radon emanation over an ore body - search for long distance transport of radon. Memoir No. 38, New Mexico Bureau of Mines and Mineral Resources, pp. 380-390; 1980.

Fleischer, R. L.; Mogro-Campero, A. Radon enhancements in the earth: Evidence for intermittent upflows. Geophys. Res. Lett., 6:361-364; 1979.

Hutter, A. R. A Method for Determining Soil Gas ^{220}Rn (Thoron) Concentrations. Health Phys. 68:835-839; 1995.

Klusman, R. W.; Jaacks, J. A. Environmental influences upon mercury, radon, and helium concentrations in soil gases at a site near Denver, Colorado. J. Geochem. Explor. 27:259-280; 1987.

Kraner, H. W.; Schroeder, G. L.; Evans, R. D. Measurements of the effects of atmospheric variables on Rn^{222} flux and soil gas concentrations. Adams, J. A. S.; Lowder, W. M., eds. The Natural Radiation Environment. The University of Chicago Press, Chicago, 1964:191-215.

Kvasnicka, K. Radon concentration in the soil air measured by track detectors. Nucl. Instrum. Methods. 147:599-604; 1980.

Li, Y.; Schery, S. D.; Turk, B. Soil as a source of indoor ^{220}Rn. Health Phys. 62:453-457; 1992.

Lubin, J. H.; Boice, J. D. Estimating Rn-induced lung cancer in the United States. Health Phys. 57:417-427; 1989.

Malmqvist, L.; Kristiansson, K. Experimental evidence for an ascending microflow of geogas in the ground. Earth Planetary Sci. Lett. 70:407-416; 1984.

Megumi, K.; Mamuro, T. Radon and thoron exhalation from the ground. J. Geophys. Res. 78:1804-1808; 1973.

Miller, K. Personal communication. USDOE, Environmental Measurements Laboratory, New York; 1995.

Narasimhan, T. N.; Tsang, Y. W.; Holman, H. Y. On the potential importance of transient air flow in advective radon entry into buildings. Geophys. Res. Lett. 17:821-824; 1990.

Nazaroff, W. W. Radon transport from soil to air. Rev. Geophys. 30:137-160; 1992.

Nazaroff, W. W.; Sextro, R. G. Technique for measuring the indoor ^{222}Rn source potential of soil. Environ. Sci. Tech. 23:451-458; 1989.

Nero, A. V.; Nazaroff, W. W. Characterizing the source of radon indoors. Rad. Protec. Dos. 7:23-39; 1984.

Okabe, S. Time variation of the atmospheric radon-content near the ground surface with relation to some geophysical phenomena. Memoirs of the College of Science. Univ. of Kyoto, 28A:99-115; 1956.

Rogers, V. C.; Nielson, K. K. Correlations for predicting air permeabilities and ^{222}Rn diffusion coefficients of soils. Health Phys. 61:225-230; 1991.

Rose, A. W.; Hutter, A. R.; Washington, J. W. Sampling variability of radon in soil gases. J. Geochem. Explor. 38:171-191; 1990.

Rudakov, V. P. Baric variations in subsoil radon. Geochem. Inter. 22:14-18; 1985.

Sachs, H. M.; Hernandez, T. L.; Ring, J. W. Regional geology and radon variability in buildings. Env. Int. 8:97-103; 1982.

Schroeder, G. L.; Kraner, H. W.; Evans, R. D. Diffusion of radon in several naturally occurring soil types. J. Geophys. Res. 70:471-474; 1965.

Schumann, R. R.; Owen, D. E.; Asher-Bolinder, S. Weather factors affecting soil-gas radon concentrations at a single site in the semiarid western U.S. Proc. of the 1988 E.P.A. Symposium on Radon and Radon Reduction Technology 2. Publication EPA/600/9-89/006B; 1989:3.1-3.13.

Tanner, A. Radon migration in the ground: A review. Adams, J. A. S.; Lowder, W. M., eds. The Natural Radiation Environment. The University of Chicago Press, Chicago, 1964:161-276.

Tanner, A. Radon migration in the ground: A supplementary review. Gesell, T. F.; Lowder, W. M., eds. The Natural Radiation Environment III. USDOE Report CONF-780422, Vol. 1, 1980:5-56.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). Ionizing Radiation: Sources and Biological Effects. New York: Report to the General Assembly, United Nations; 1982.

Washington, J. W.; Rose, A. W. Regional and temporal relations of radon in soil gas to soil temperature and moisture. Geophys. Res. Lett. 17:829-832; 1990.

Washington, J. W.; Rose, A. W. Temporal variability of radon concentration in the interstitial gas of soils in Pennsylvania. J. Geophys. Res. 97:9145-9159; 1992.

Wilkening, M. H.; Clements, W. E.; Stanley, D. Radon-222 flux measurements in widely separated regions. Adams, J. A. S.; Lowder, W. M.; Gesell, T. F., eds. The Natural Radiation Environment II. ERDA Rept. CONF-720805-P2, Vol. 2. 1972:717-730.

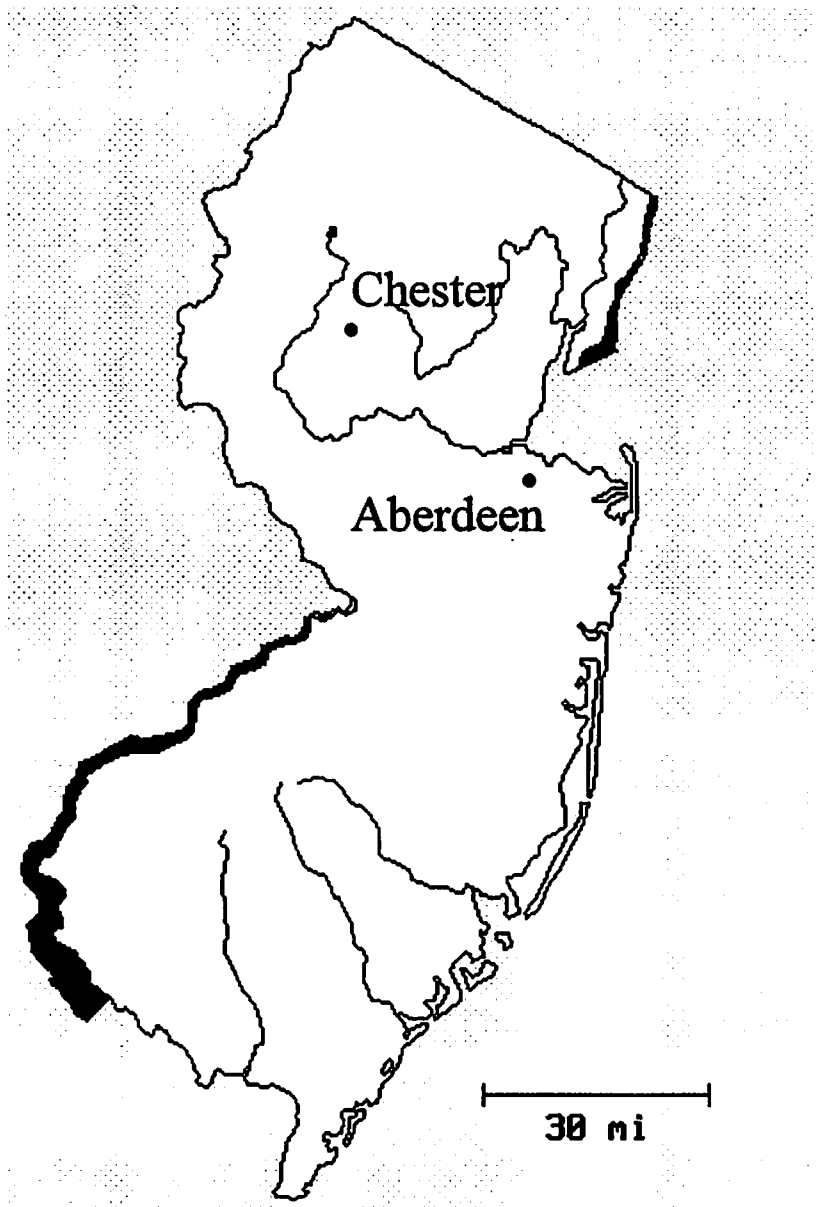


Fig. 1. Map of New Jersey showing location of sampling sites.

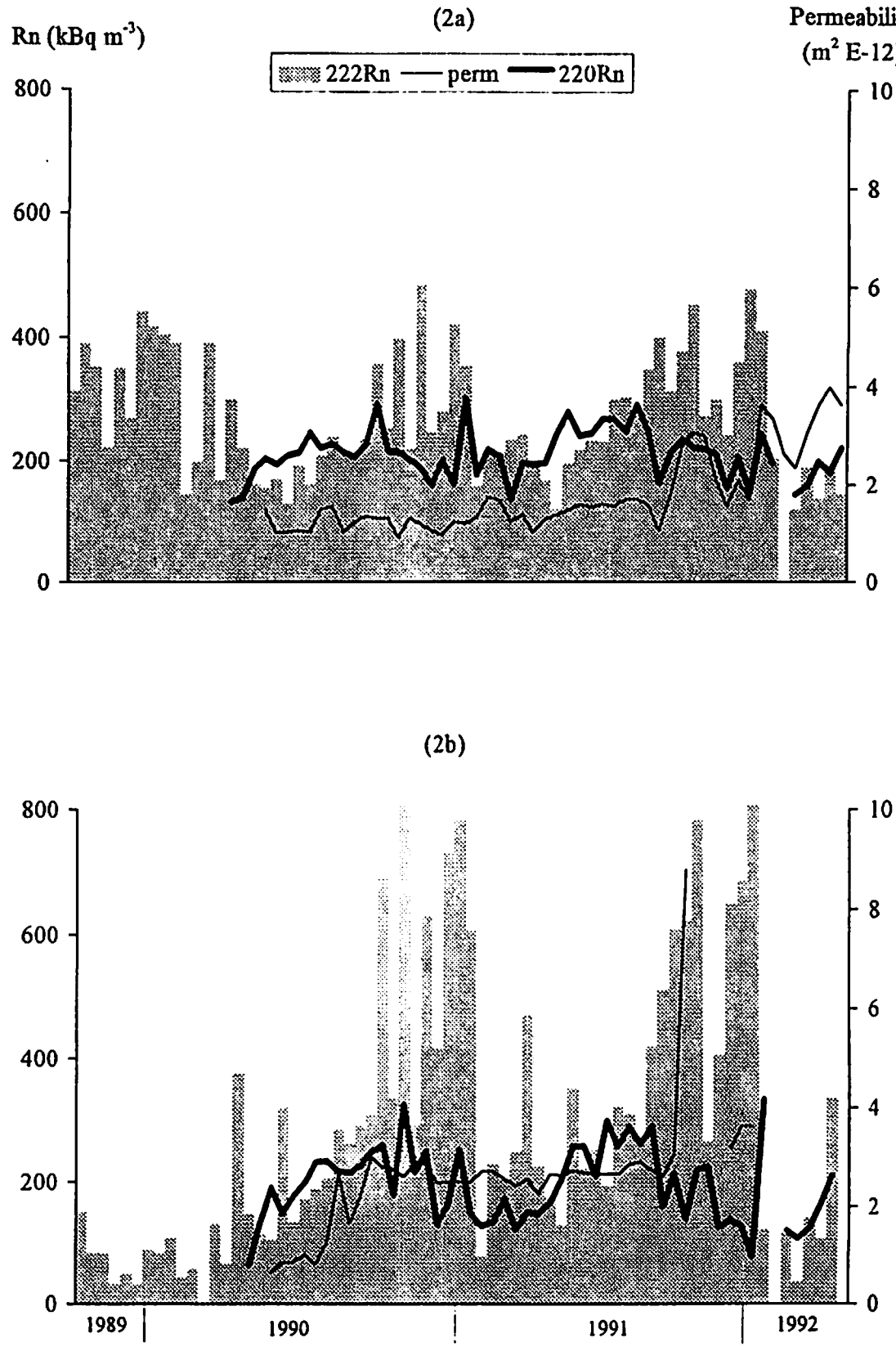
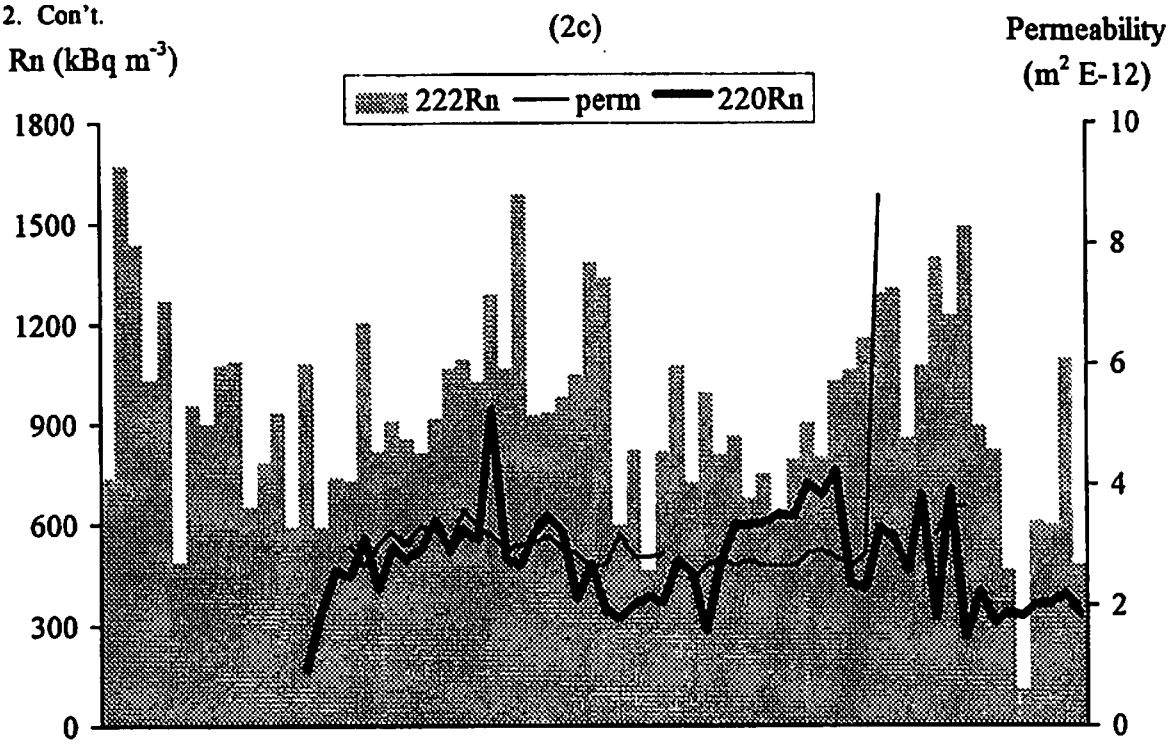


Fig. 2. ^{220}Rn, ^{222}Rn and permeability data from the Chester site obtained from 0.85 m depth tubes from September, 1989 to May, 1992. Position 1 data are shown in (a), (b) shows position 2 data, (c) shows position 3a data, (d) shows position 3b data, (e) shows position 4a data, (f) shows position 4b data, (g) shows position 5 data, and (h) shows position 6 data.

Fig. 2. Con't.


(2c)
Rn (kBq m-3)
Permeability
(m2 E-12)
222Rn
perm
220Rn
1800
1500
1200
900
600
300
0
10
8
6
4
2
0

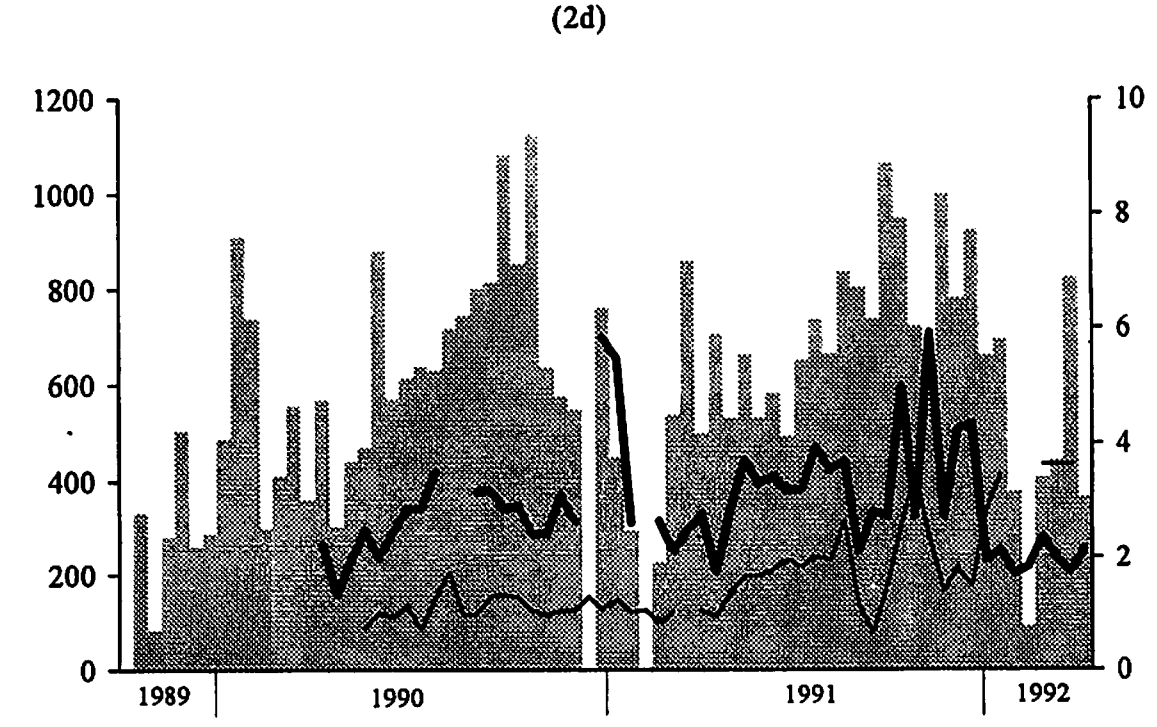
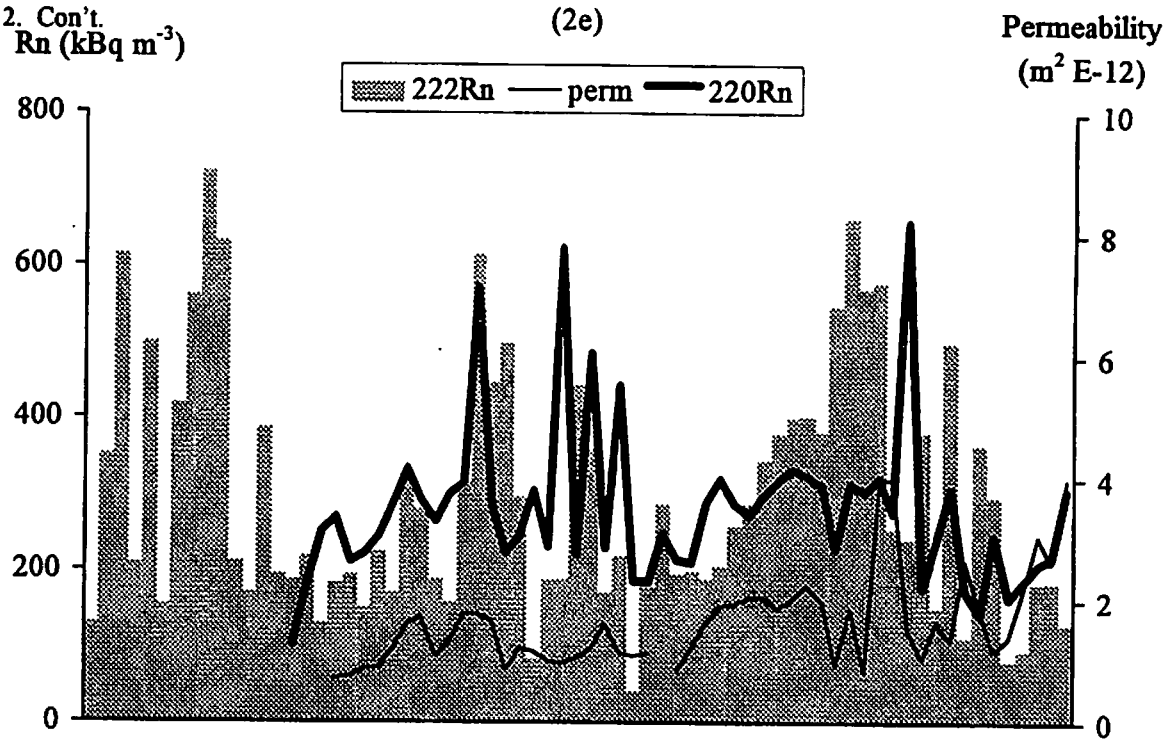

(2d)
1200
1000
800
600
400
200
0
10
8
6
4
2
0
1989
1990
1991
1992

Fig. 2. Con't.

(2e)



(2f)

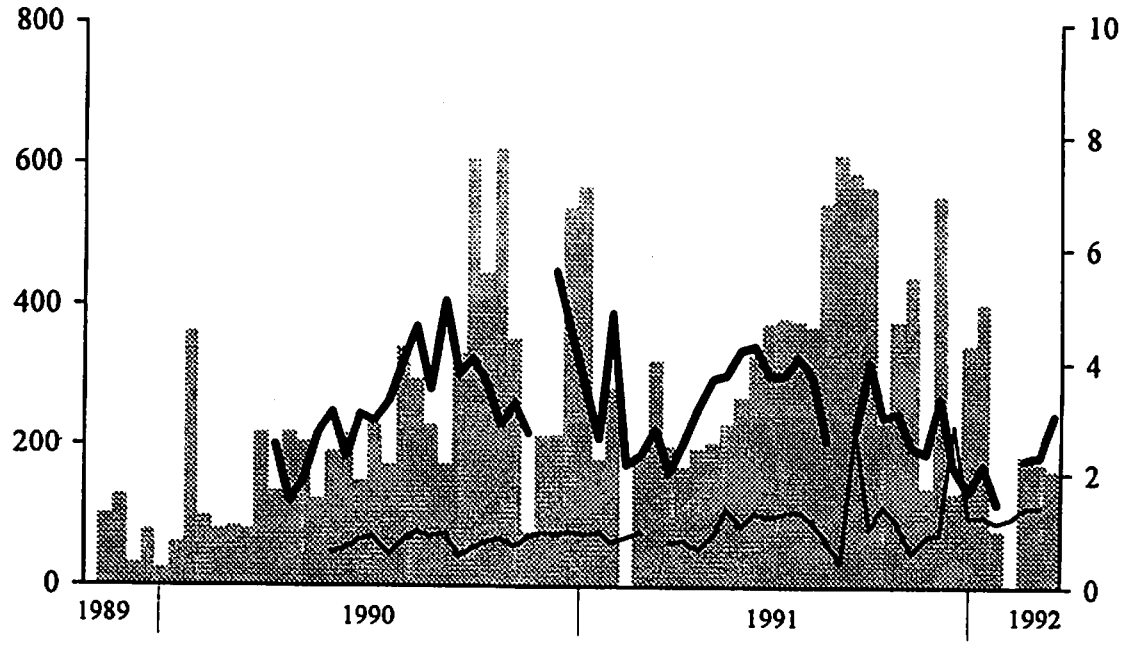
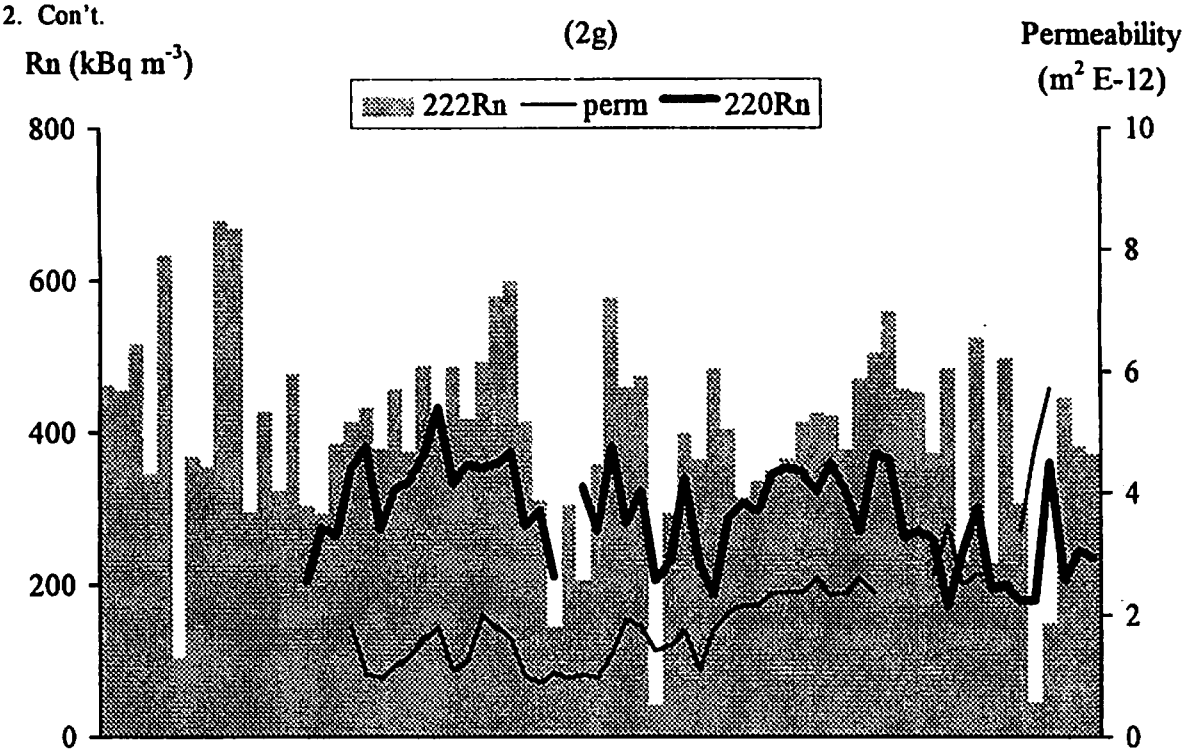
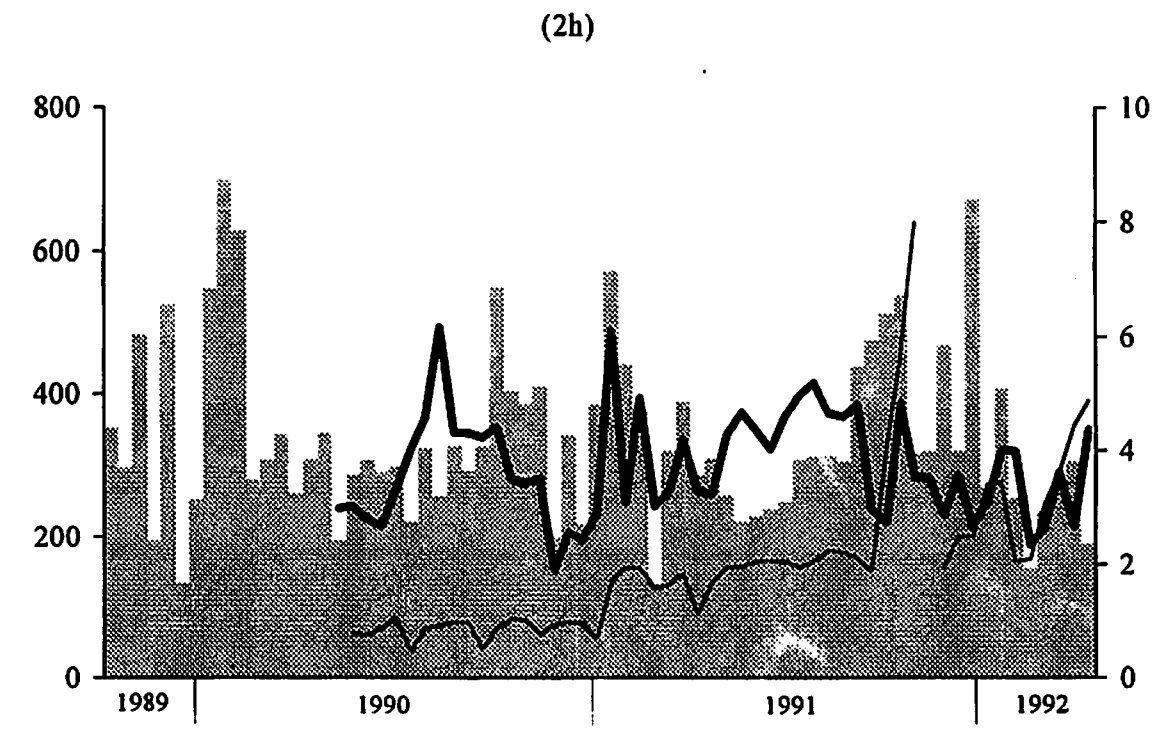


Fig. 2. Con't.


(2g)
Rn (kBq m-3)
Permeability
(m2 E-12)
222Rn
perm
220Rn
800
600
400
200
0
10
8
6
4
2
0


(2h)
800
600
400
200
0
10
8
6
4
2
0
1989
1990
1991
1992

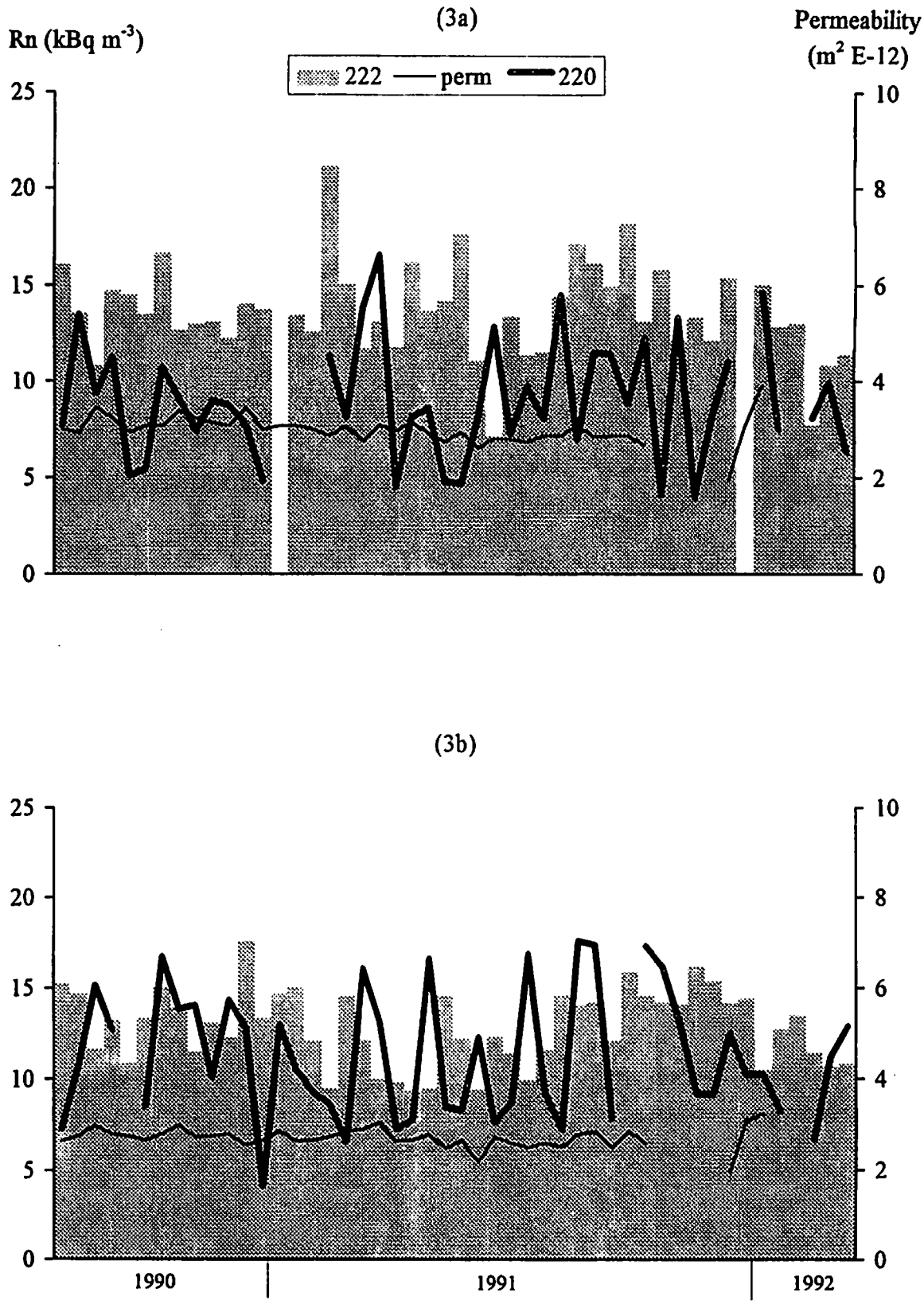
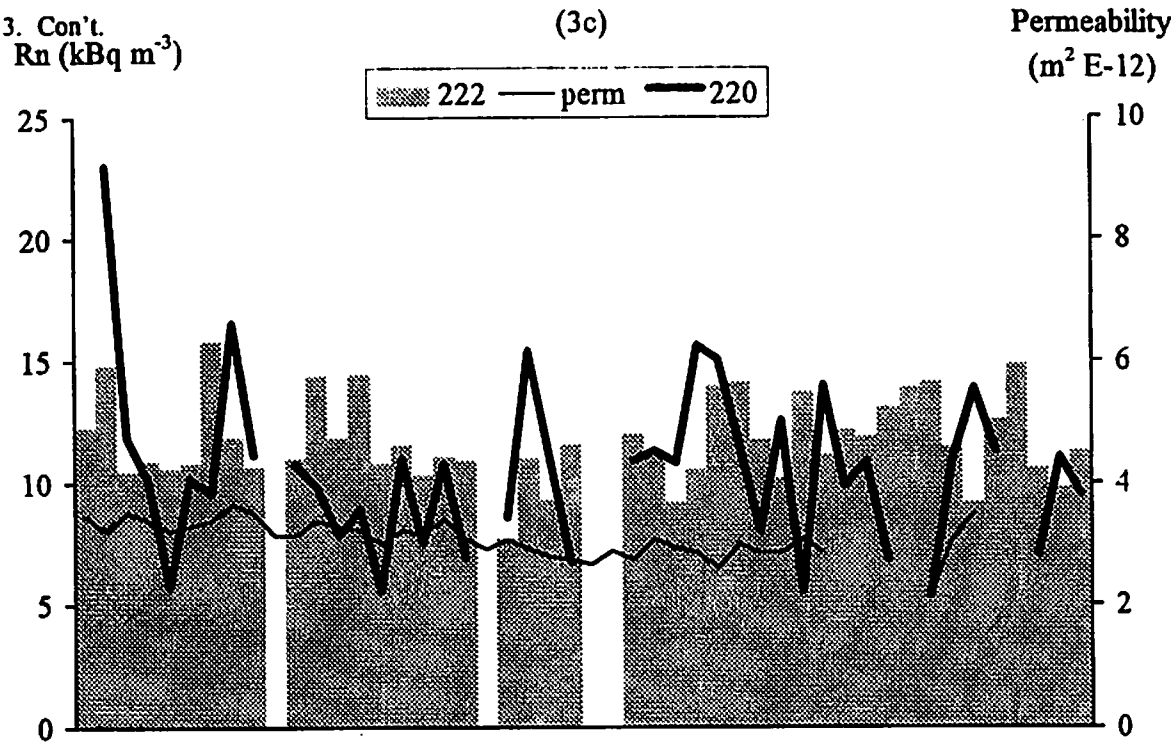


Fig. 3. ^{220}Rn, ^{222}Rn and permeability data from the Aberdeen site obtained from 0.85 m depth tubes from May, 1990 to May, 1992. Position 1 data are shown in (a), (b) shows position 2 data, (c) shows position 3 data, (d) shows position 4a data, and (e) shows position 4b data.

Fig. 3. Con't.

(3c)



(3d)

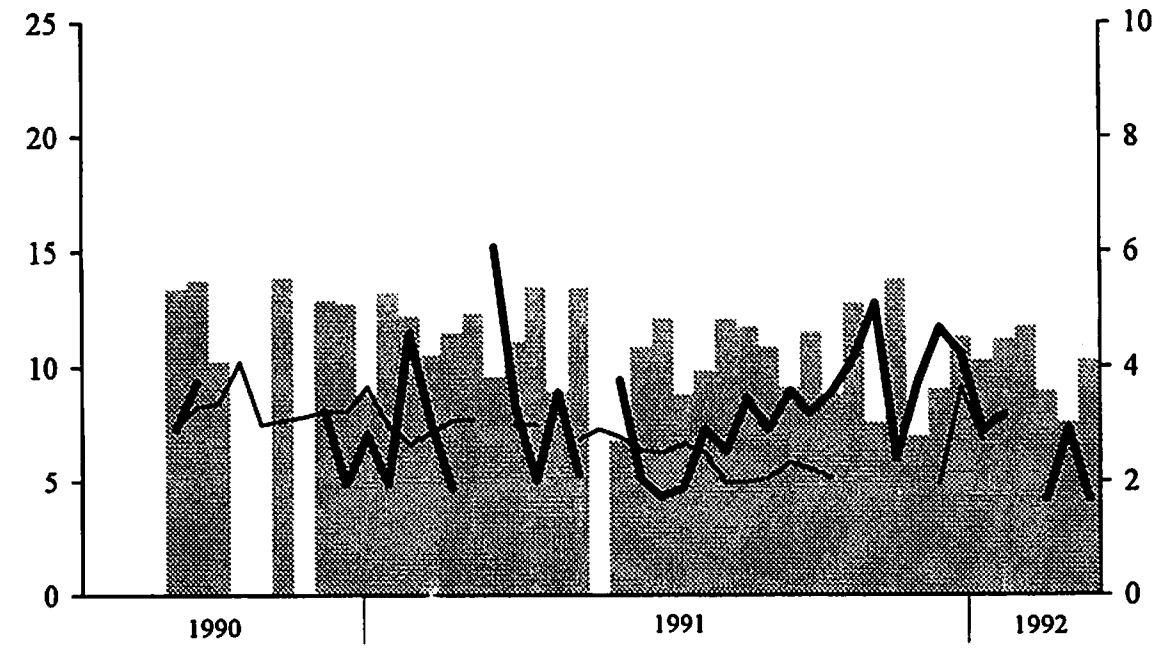
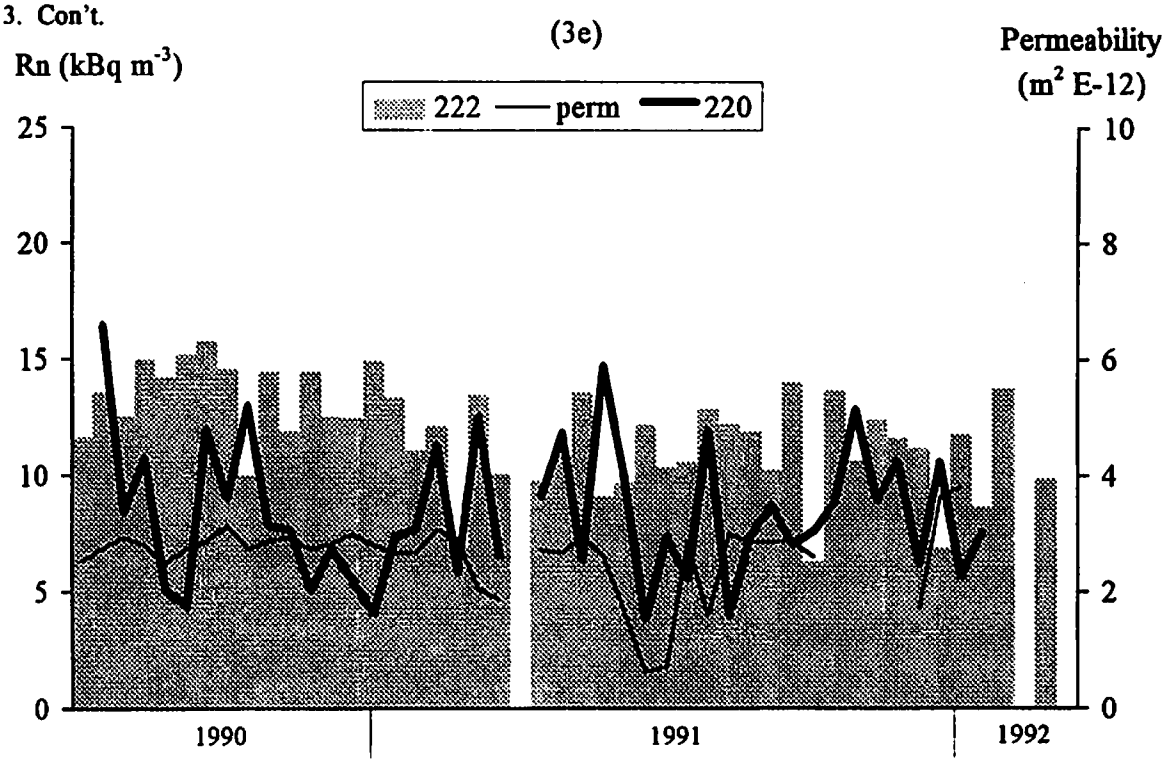
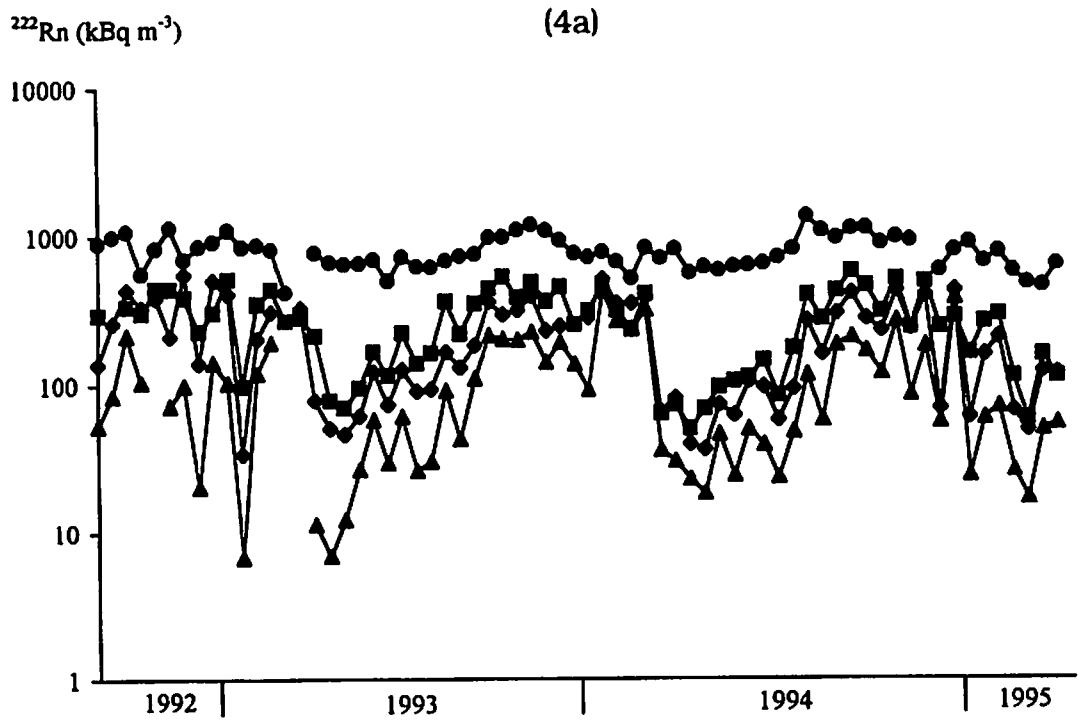


Fig. 3. Con't.


(3e)
Rn (kBq m-3)
Permeability
(m2 E-12)
222
perm
220
25
20
15
10
5
0
10
8
6
4
2
0
1990
1991
1992



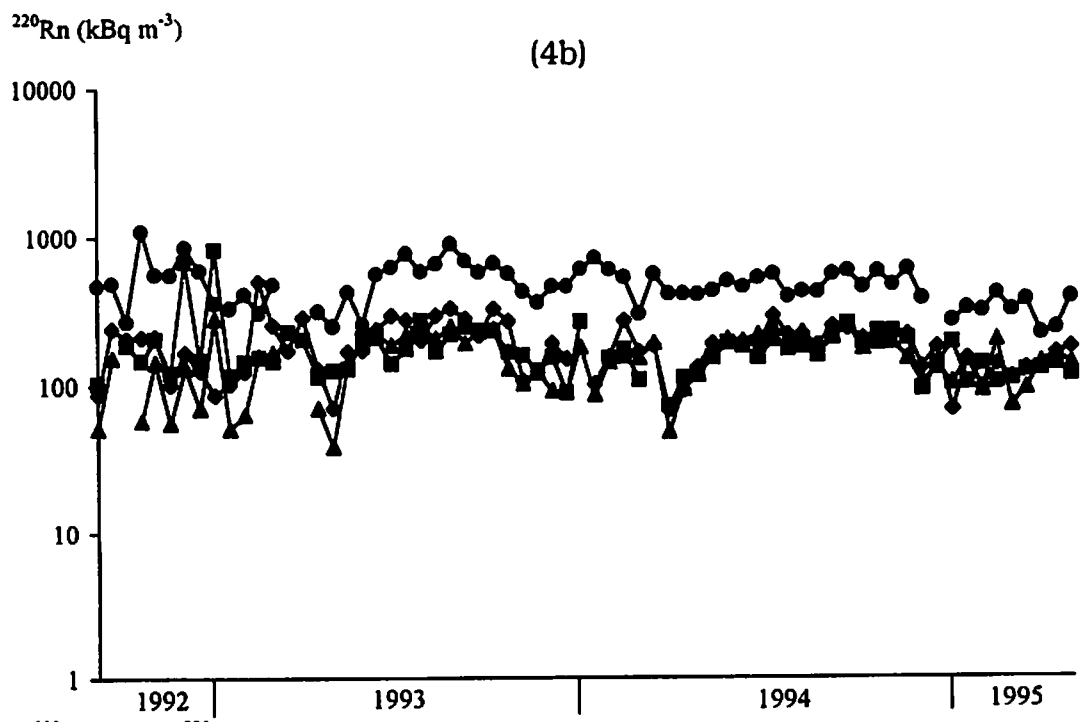
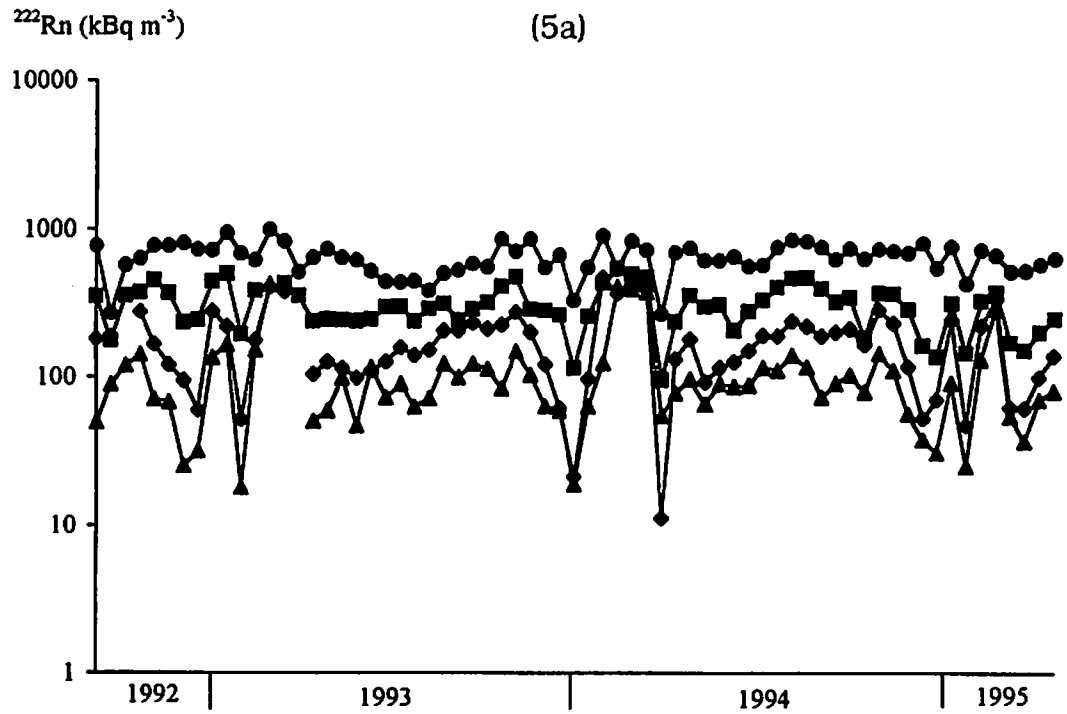


Fig. 4. ^{222}Rn (a) and ^{220}Rn (b) from the Chester site position 2 obtained from 0.28, 0.56, 0.85 and 1.28 m depths from August, 1992 to March, 1995.



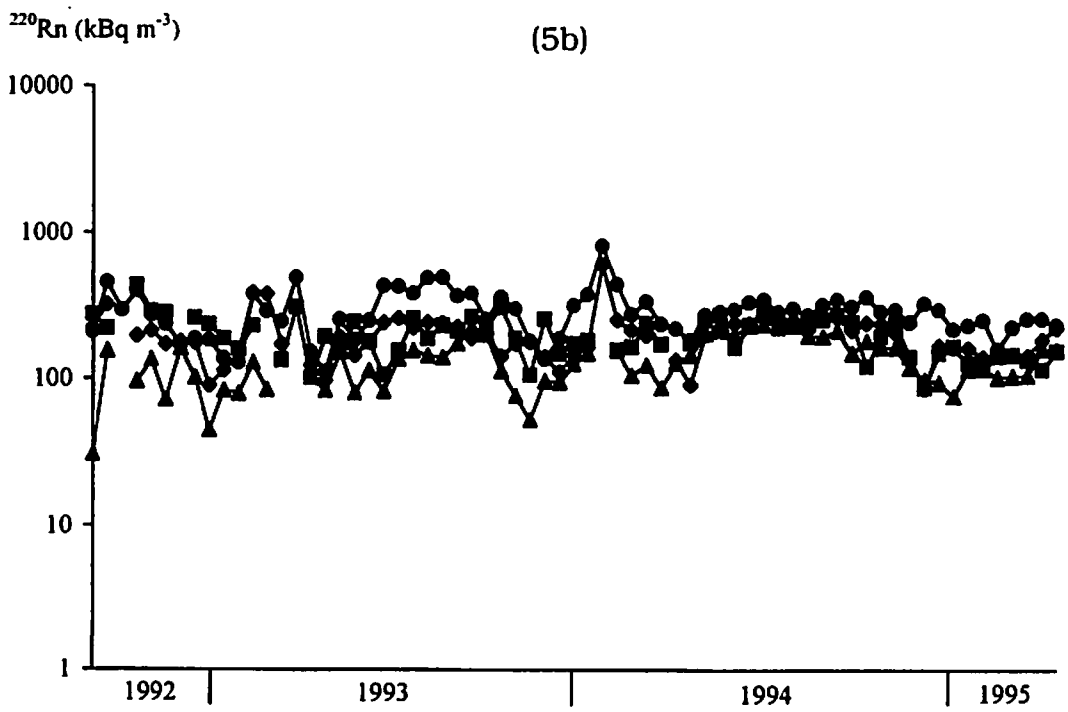
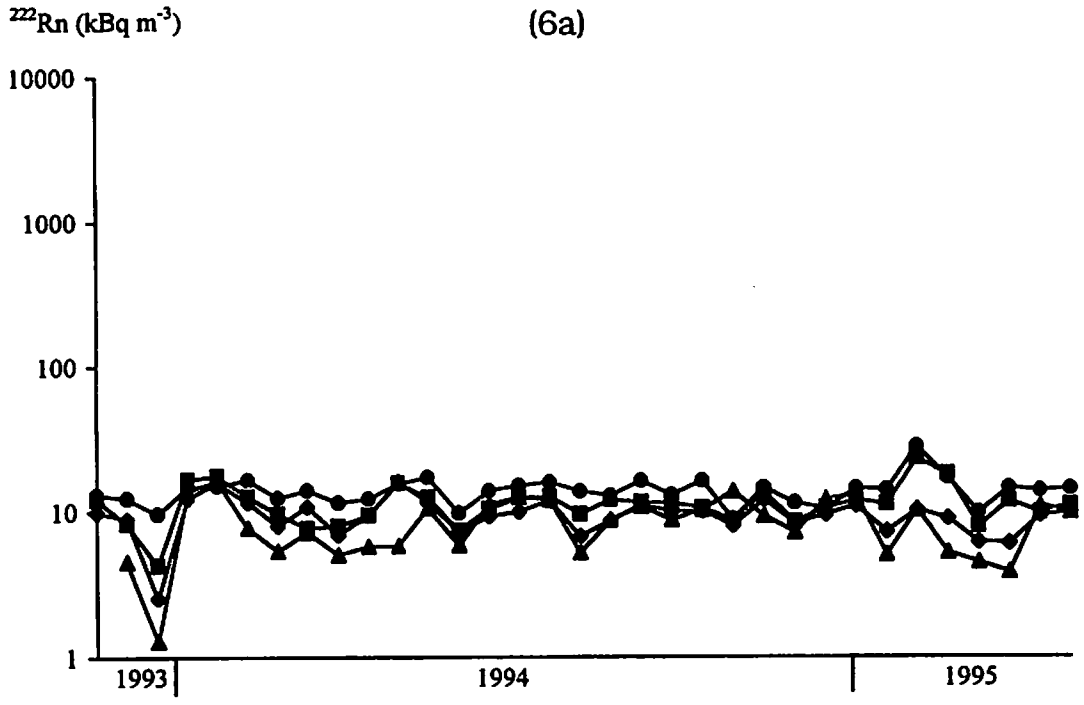


Fig. 5. ^{222}Rn (a) and ^{220}Rn (b) from the Chester site position 5 obtained from 0.28, 0.56, 0.85 and 1.28 m depths from August, 1992 to March, 1995.



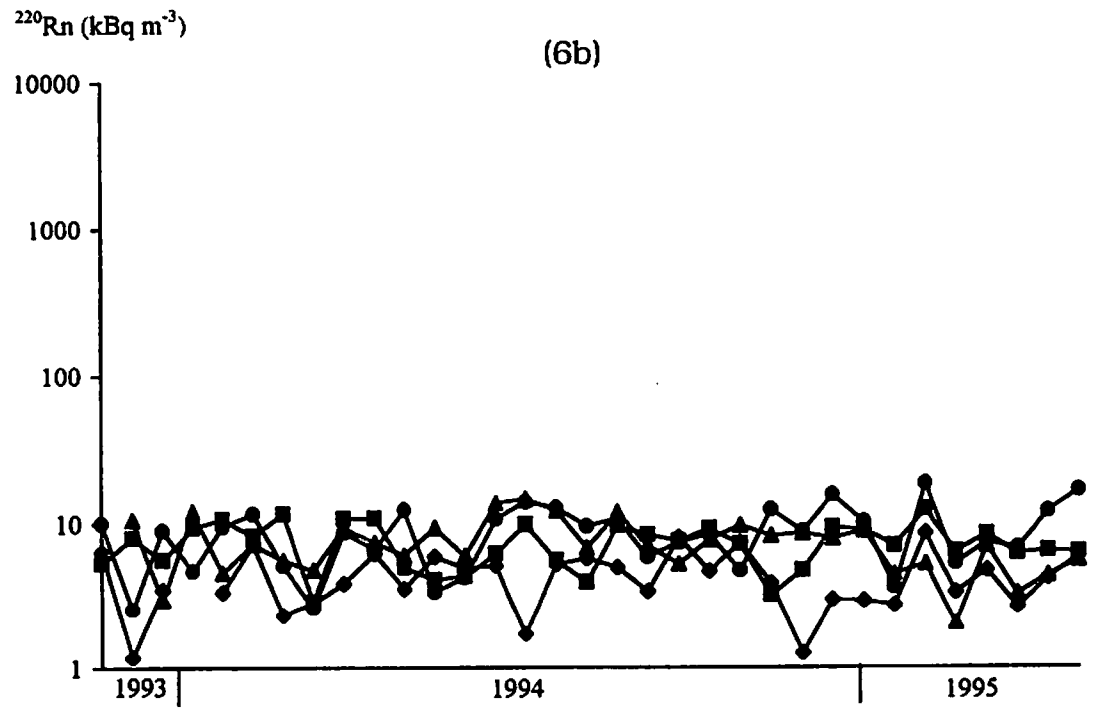
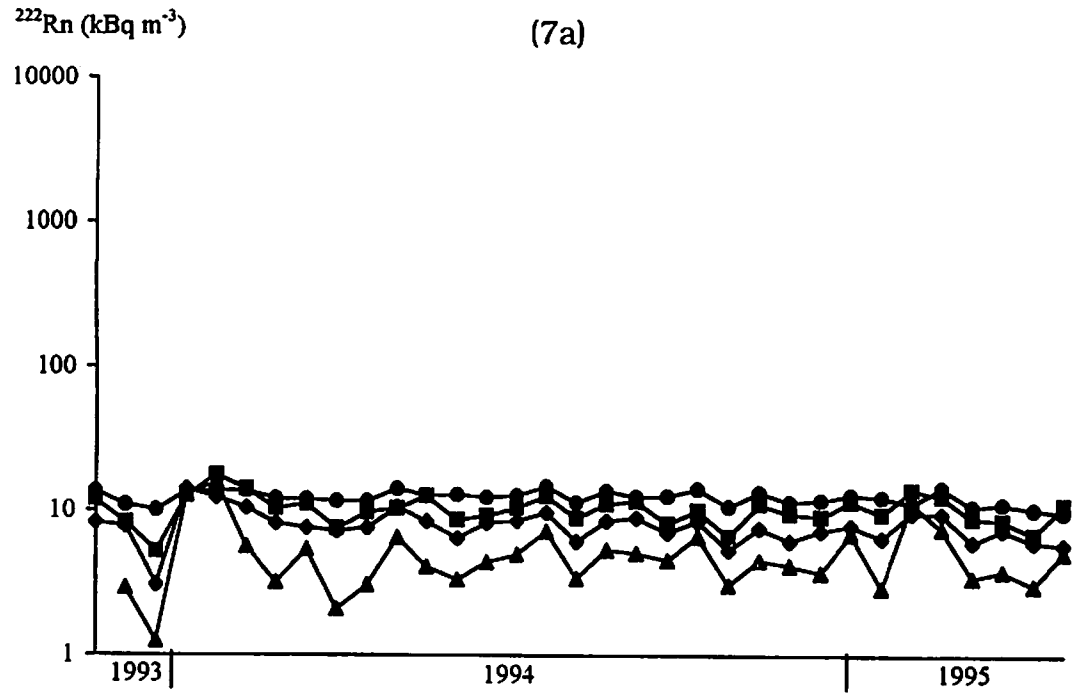


Fig. 6. ^{222}Rn (a) and ^{220}Rn (b) from the Aberdeen site position 4a obtained from 0.28, 0.56, 0.85 and 1.28 m depths from November, 1993 to March, 1995.



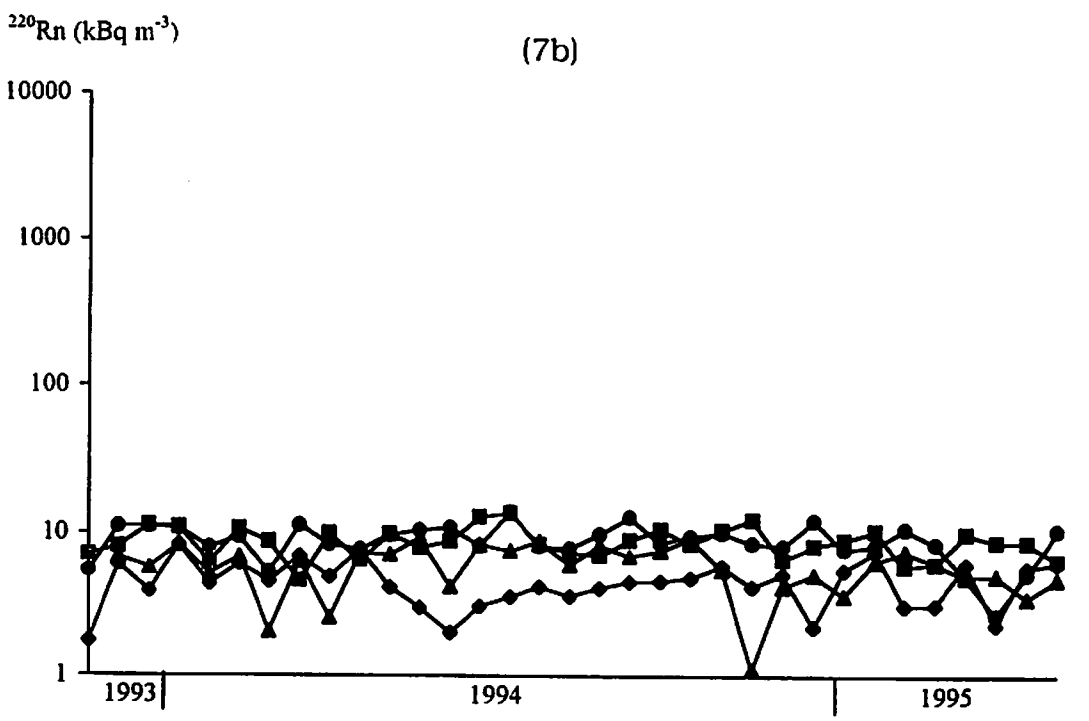


Fig. 7. ^{222}Rn (a) and ^{220}Rn (b) from the Aberdeen site position 4b obtained from 0.28, 0.56, 0.85 and 1.28 m depths from November, 1993 to March, 1995.

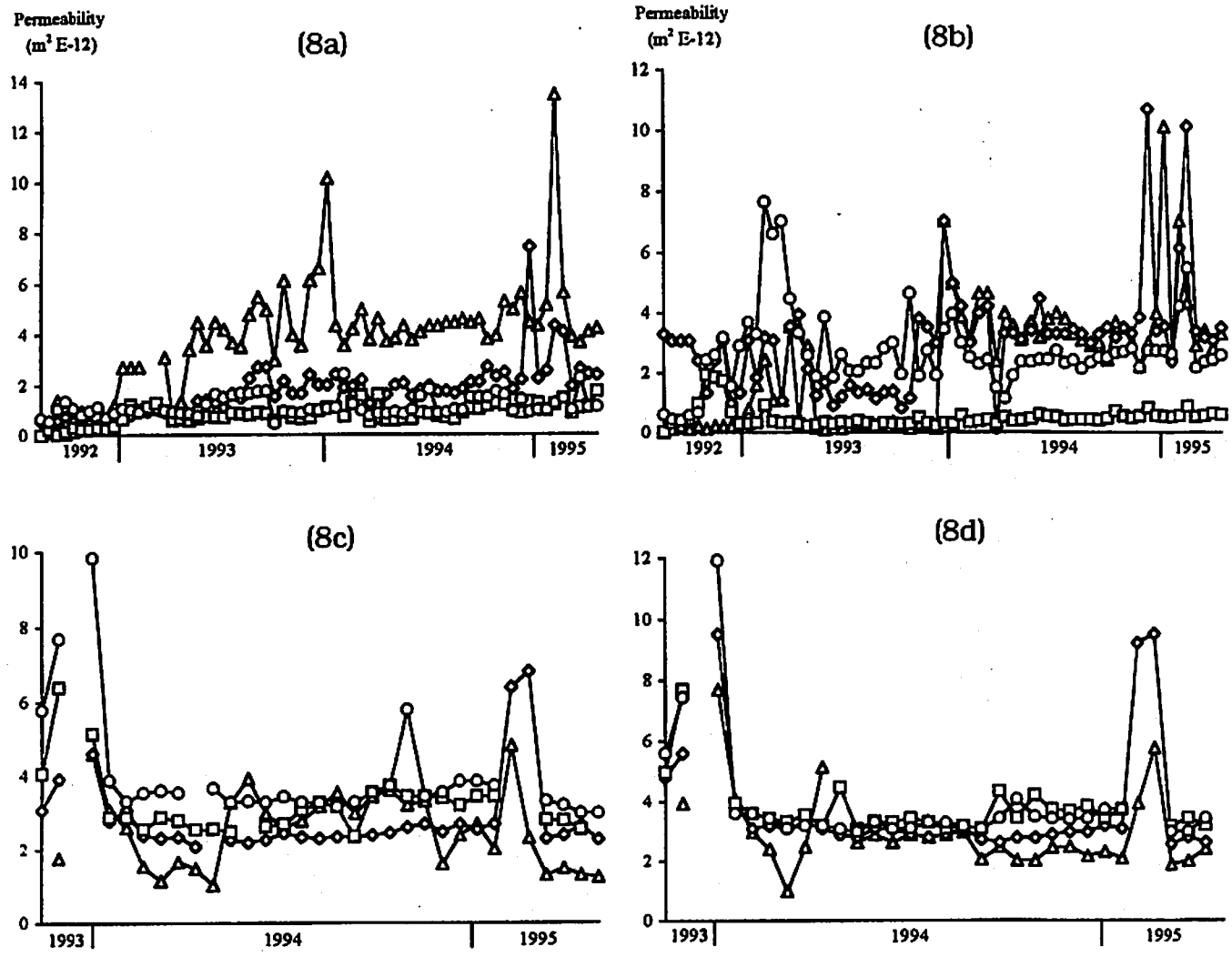


Fig. 8. Permeability data from the Chester site position 2 (a) and position 5 (b) obtained from August, 1992 to March, 1995 and from Aberdeen site position 4a (c) and position 4b (d) obtained from November, 1993 to March, 1995.