



Isotope studies of a thick unsaturated zone in a semi-arid area of Southern Africa

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ABSTRACT

Unsaturated zone profiles ranging in depth from 8 m to 22 m were obtained by hand augering an aeolian sand cover in the southern reaches of the semi-arid Kalahari thirstland. Moisture contents were rather low (< 3 wt.%); in situ moisture chloride concentrations, measured by selective ion electrode following elutriation, are generally < 500 ppm. Deuterium in the moisture was measured mass spectrometrically by direct quantitative conversion to hydrogen on zinc metal of moist soil samples. A novel technique of direct equilibration was developed for oxygen-18 analysis. Neither a thermonuclear tritium peak nor a stable isotope evaporation inversion near the surface could be observed in any of the profiles. Remarkable differences both laterally and vertically are observed in most parameters measured between profiles taken a few tens of metres apart. At greater depths, these differences become less pronounced. Recharge estimates based on chloride differ markedly from those obtained from tritium. Although the stable isotope values of the underlying saturated zone are similar to moisture in the deeper sections of the unsaturated zone profiles, the markedly lower chloride concentrations point towards preferential or bypass flow as an important mechanism of ground water recharge in the area. This can be regarded as a benchmark site on account of the wealth of unsaturated zone data as well as the detailed and ongoing rainfall record.

1. INTRODUCTION

The aim of this project was to obtain profiles of moisture content, isotopic and chemical indicators of the movement of rain water in the unsaturated zone in appropriate semi-arid environments in southern Africa. The results obtained form part of a world-wide survey of unsaturated zone transport with particular aim of exploiting what may be the last opportunity of applying thermonuclear tritium as a tracer in the unsaturated zone. Since thermonuclear tritium in the southern Hemisphere rain water peaked in 1964, levels have declined to about the expected pre-bomb, or natural, values (IAEA 1992). It is however possible that in thick unconsolidated overburdens thermonuclear tritium might still be found, acting as a marker of water infiltrated during the latter 60's which has not yet completely traversed the unsaturated zone.

During the mid-seventies, the Environmental Isotope Group conducted research on unsaturated zone transport in what is now called Northwest Province of the Republic of South Africa (Verhagen et al. 1979a; 1979b). Sample profiles were obtained in effectively pure sand overburden down to depths of more than 20 metres. Tritium and stable isotope values were measured on the moisture extracted from these samples and conclusions reached on moisture transport.

In the framework of the present project the area was re-visited during late July 1997, and three depth profiles obtained. A further three profiles were obtained in a closely-adjacent location during August 1998. This report describes the sampling procedures and analytical techniques, discusses the results and presents the conclusions drawn from the available information.

2. DESCRIPTION OF STUDY AREA

The area of study (Fig. 1) is typified by flat, featureless terrain with low, semi-fixed dunes, at an altitude of about 1100 m.a.m.s.l. and can be described as semi-arid on the basis of Köppen and Thornthwaite classifications. The sandy surface cover and topography lead to little runoff. Average annual rainfall is 309 mm (6 stations), the average number of rain days per year is 26 and pan evaporation is 2400 mm a⁻¹. The vegetation is typical African bush savannah.

The underlying geology of the area is varied, ranging from the oldest rocks consisting of dolomites, banded ironstones, lavas and tillites, to the lower members of the Karoo sedimentary sequence with intrusives. The hard rocks are covered by the Kalahari deposits, a semi-consolidated sequence which in the study area may reach a thickness of more than 150 m and consists of basal gravels, clays, calcretes and sand. The thickness of the fine, windblown red to white sand cover may exceed 50 m.

3. FIELD WORK

3.1 Drilling programme for 1997

During July 1997, three unsaturated zone profiles were obtained by hand augering in the Hotazel/Black Rock area of South Africa (Fig. 1). The area was chosen in the light of the earlier study (Verhagen et al. 1979a,b) which showed a pure sand cover depth of more than 22m. The profiles sampled in the first year of study reached depths of 18 m, 15 m and 3 m respectively. The first profile (BH 10) was sampled every 1 m; the second (BH 11) every 0.5 m, and the shallow profile (BH 12) was sampled at 0.1 m intervals. For the deeper profiles, 3 kg samples of sand were taken in glass preserve jars and 1 kg parallel samples in tins supplied by the IAEA. The shallow profile samples were taken in tins only.

The augering site was within a few tens of metres of the profile drilled in 1977 on the farm Harefield (Fig. 1). As the area is well-vegetated, the site chosen was in the middle of a grassed clearing about 100 m across between moderately-sized, mainly acacia, trees. Although it was realised that this choice would introduce some bias in terms of vegetal representativeness, the aim was to avoid striking major roots, which previous experience had shown could frustrate hand, and even power, augering. Bh 11 was drilled some 20 m from BH 10. The shallow BH 12 was drilled halfway between them. Fine root material was noticed only down to about 50 cm. The profiles consisted of pure sand, with noticeable changes in colour and texture, somewhat coarser material being encountered at depths greater than 10 m.

The auger used was constructed from thin-walled steel tubing, 10 cm dia, 25 cm long with two inclined cutting blades. The auger stem was made up of 1 m length steel water pipes with threaded ends and standard pipe bushes. This provided a robust, but heavy system. A 4 m tripod with a hook at the top served to stabilise and vertically store the 5-6 m lengths into which the stem was separated during raising and lowering of the auger at greater depths.

A problem encountered was that the auger holes tended to curve away noticeably from the vertical at depths greater than about 10 m. This implied scraping of the auger head along the wall of the hole whilst being lowered and raised. For every augerful sampled, sand was therefore removed off the top and bottom of the contents of the auger, and the centre section transferred directly into the sampling vessel which was immediately closed. The depth of 18m (BH 10) was the maximum attainable with the equipment used, and took about 1.5 days to reach. A second hole (BH 11) was drilled to 15m, reached in about 1 day. BH 12 was drilled to about 3 m, but sampled every 10-15 cm, which took a few hours.

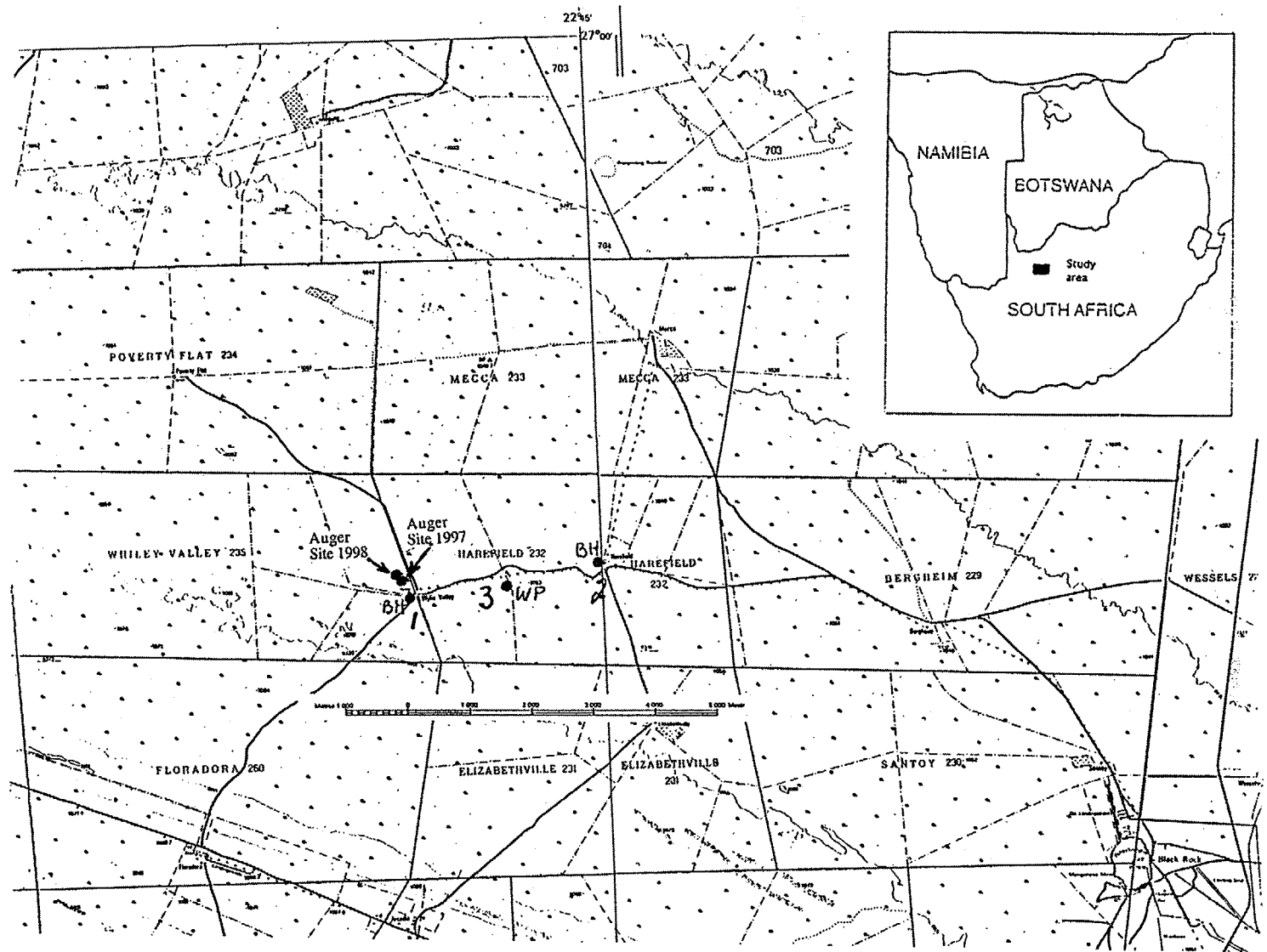


FIGURE 1.

3.2 Drilling programme for 1998

A further three hand auger boreholes were sunk in July 1998 in the same area as, and at a distance of some 70 metres from, the 1997 boreholes, in a different clearing between mainly acacia trees. BH 13 was sunk to 22 m over a period of two days. BH 14 and BH 15 were sunk at a distance of 25 metres to the west and north respectively to a depth of 8 metres. BH 15 struck a major root at a few metres depth. The position was moved about 1 metre and encountered no obstruction.

As with BH 10, 11 and 12 pure sand was encountered in these profiles, which changed in colour and texture with increasing depth. This allowed for assessing the extent of shallower material contaminating the deeper samples visually and removing it before the sample vessels were filled.

Narrower and longer auger heads were tried initially. They were found however to be less effective than the original auger head which was used for the profiles of 1997, and again for sinking BH 13.

Sand samples of 3 kg were taken in glass preserve jars. These were intended principally for tritium analysis. In addition to each of these, four 50 g samples for stable isotope analysis were taken in small glass bottles with double stopper. These were used following the experience gained from the first round of sampling, which showed that stable isotope values started shifting rapidly in tins, and in the larger glass vessels once they had been opened.

4. ANALYTICAL METHODOLOGY

4.1 Moisture contents

About 20g of sand is placed in a petri dish and weighed accurately on a precision balance. It is then placed in a drying oven at about 100°C overnight, allowed to cool and weighed again. The weight difference is taken to be loss of moisture, expressed as a weight percentage w.r.t. the original weight. When water was quantitatively distilled from the larger, 3 kg sand samples, the sample was weighed before and after distillation, as well as the water produced.

4.2 Chloride concentration

About 30 g of moist sand is placed in a small glass bottle, which is weighed accurately and 20 ml of de-ionised water is added. The bottle is tightly stoppered and shaken. It is then left overnight, again shaken, and left to settle. The electrical conductivity of the supernatant is determined with an E.C. meter and the Cl⁻ concentration is determined with an ion sensitive electrode. The Cl⁻ value in the soil moisture in both cases is determined by:

$$C_o = \frac{(C - C_i)M_w}{M_s p}$$

where C_o is the value in the soil moisture; C the measured value; C_i the blank value for de-ionised water; M_w the mass of water added; M_s the mass of soil and p the measured weight fraction of water (initial moisture contents).

4.3 Stable isotopes

The analytical procedures for stable isotopes developed during the project are described in some detail as they have some unique features which specifically suit them to unsaturated zone investigations.

4.3.1 Deuterium

Deuterium measurements are conducted by using a modification of a method developed by Darling (1989). Ampoules are made from Pyrex glass tubing (Fig. 2) with a constriction. 0.5 g specially prepared and dried Zn metal grains (20-30 mesh) are shaken through the lower constriction. A small plug of glass wool is then inserted and held in position by this constriction. The top of the ampoule is then drawn to a narrow neck, attached to a vacuum line and evacuated whilst heating to some 200°C.

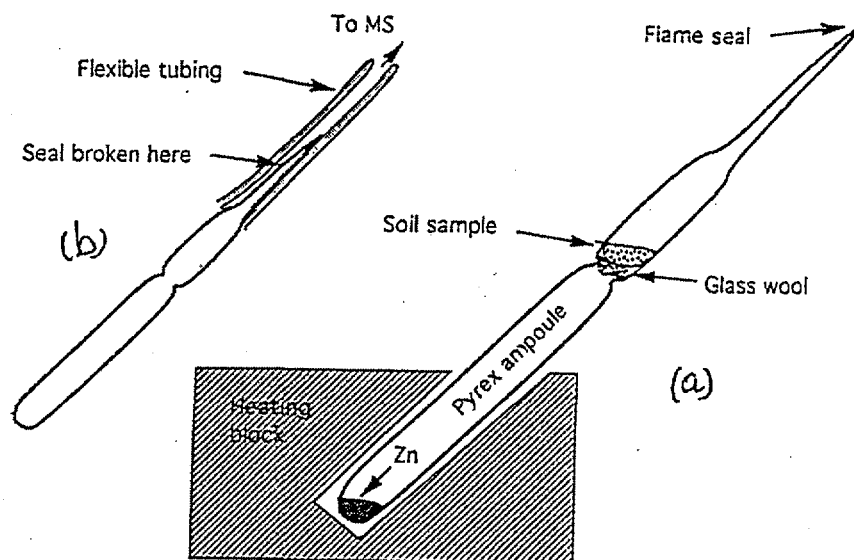


FIGURE 2.

After cooling down, the ampoule is removed from the vacuum line and about 0.5 g of moist sand (depending on the moisture content) shaken rapidly through the neck onto the glass wool. The ampoule is then immediately re-attached to the vacuum line, cooled to -80°C in dry-ice slurry and evacuated. The upper constriction is then flame-sealed. Ten such ampoules can be heated simultaneously at an angle in a heating block at ~ 500°C, with the sand just outside the block (Fig. 2a).

Complete reaction to H₂ is usually achieved within about 30 minutes, and checked by observing the absence of visible moisture condensation after touching the ampoule with a liquid nitrogen-soaked cotton wool plug.

Deuterium is then measured in an isotope VG 602C isotope ratio mass spectrometer. The ampoule is attached to the IRMS inlet with a length of flexible tubing (Fig. 2b). After evacuation,

the tip of the ampoule seal is broken by hand through the flexible tube and the hydrogen gas allowed to flow into the IRMS inlet reservoir.

To test the reproducibility of the method, deuterium measurements using the above-mentioned method have been conducted on

1. four aliquots from a moist sand sample and
2. four aliquots of sand, first dried in a drying oven and then moistened with water of known isotopic content.

The results are presented in Table 1.

Table 1. Deuterium test analyses

Sample	δD (‰)	Sample	δD (‰)
Soil Sample 1	-67.8	Wetted Sand 1	-2.2
Soil Sample 2	-66.1	Wetted Sand 2	-2.1
Soil Sample 3	-66.8	Wetted Sand 3	-3.9
Soil Sample 4	-67.3	Wetted Sand 4	-2.4
		Average Water	-2.6

The reproducibility of the δD measurements on the first set of moist samples is very good. The results for the measurements on the second batch of samples again shows good reproducibility as well as acceptable correspondence with the known isotopic value of the water used. The method therefore appears to be quantitative and fractionation corrections therefore need not be applied.

4.3.2 Oxygen-18

An approach similar to the direct conversion technique for deuterium was tried in two methods for oxygen-18 determination by equilibrating CO_2 directly with the moist soil/sand.

a) About 1g of sand is placed in a glass ampoule which is attached to a vacuum system via a capillary, pumped and CO_2 admitted. The ampoule is then flame sealed, removed from the vacuum line and left in a horizontal position to equilibrate. The CO_2 is transferred to the mass spectrometer inlet as described for deuterium measurements (above). It was found that oxygen isotope equilibration between the sand moisture and CO_2 proceeded rapidly, and was complete within a few hours. Reproducibility for duplicate samples was as good as for standard water equilibration.

b) The second method uses the water equilibration line for oxygen-18 determination. The advantage is that 17 aliquots can be handled simultaneously. Sand samples are weighed into individual bottles which are attached to the line. The bottles are evacuated through individual capillaries and CO_2 admitted. The bottles are left stationary, as the sand would not move under the shaking motion. It was found initially that equilibration proceeded only partially, the CO_2 not reaching the expected isotopic composition even after 24 hours. It was concluded that only the moisture in the uppermost layers of the sand sample was brought into mixing contact with the free CO_2 overlying the sand. The procedure was then changed by bringing the bottles into a near-horizontal position and tapping them gently, in order for the sand to spread out and develop cracks in its bulk. It was found that under these (again stationary) circumstances, equilibration was

complete in about 4 hours (Fig. 3). As with the first method, good reproducibility could be obtained in duplicate samples.

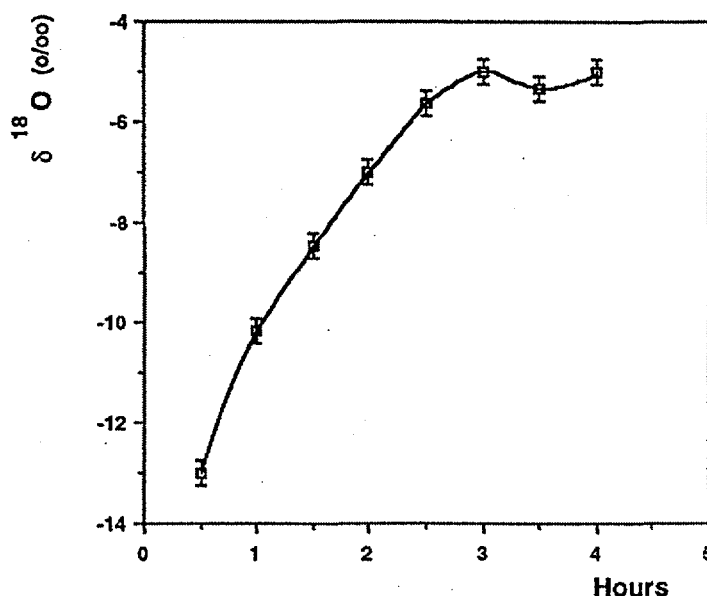


FIGURE 3.

As the moisture content of some of the augered sand samples was rather low (<2 wt.%), an amount correction may have to be applied to the measured $\delta^{18}\text{O}$ values. This correction (IAEA 1981) is given by:

$$\delta_G^0 = \left(1 + \alpha \frac{n}{N}\right) \delta_G - \alpha \frac{n}{N} \delta_G^i$$

where n and N are the gram-atoms of oxygen in the sample and CO_2 respectively; α is the fractionation factor; δ_G is the value for CO_2 after equilibration, δ_G^0 the value for an infinite amount of water ($n/N = 0$); δ_G^i is the initial value of CO_2 .

A test with standard water samples was run on the method, using the same gas both as the reference and for equilibration. An amount of sand typical for the study area was thoroughly dried and 20 g aliquots loaded into equilibrator bottles. Water standards were pipetted into the individual bottles to produce a 2% by weight moisture contents, the bottles were stoppered and shaken to homogenise the moisture. Duplicate bottles of each: SLAP, VSMOW, GISP and eight of a lab standard were then attached to the equilibration line and handled in the standard fashion for oxygen-18 analysis. The results are shown in Table 2.

The duplicate values of $\delta^{18}\text{O}$ for the standards and the multiple values for the lab standard show very good reproducibility (21.50 ± 0.13 ‰). The span between SMOW and SLAP is considerably lower than the theoretical value. In spite of this, the span corrected value for GISP is $\delta_{\text{SMOW}} = -24.86$ ‰ and for the lab standard the accepted value of -0.58 ‰ is obtained.

The corrections which would have to be applied to the results in terms of the moisture content of the sand needed to be assessed. Typical sand from the project area was thoroughly dried, moistened with a laboratory water standard to water contents ranging from 2% to 10%, and

subjected to typical equilibration with CO₂. The resulting δ¹⁸O values are shown in Fig. 4. Interestingly, the results follow an exponential dependence on moisture contents, which allows the results to be corrected using the best-fit exponential expression.

Table 2. Results of water standards

Laboratory Standard	δ ¹⁸ O (‰)
SMOW (a)	+21.89
SMOW (b)	+22.08
SLAP (a)	-24.32
SLAP (b)	-24.14
GISP (a)	+1.24
GISP (b)	+1.33
TAP Std (a)	+21.42
TAP Std (b)	+21.60
TAP Std (c)	+21.28
TAP Std (d)	+21.37
TAP Std (e)	+21.54
TAP Std (f)	+21.61
TAP Std (g)	+21.68
TAP Std (h)	+21.52

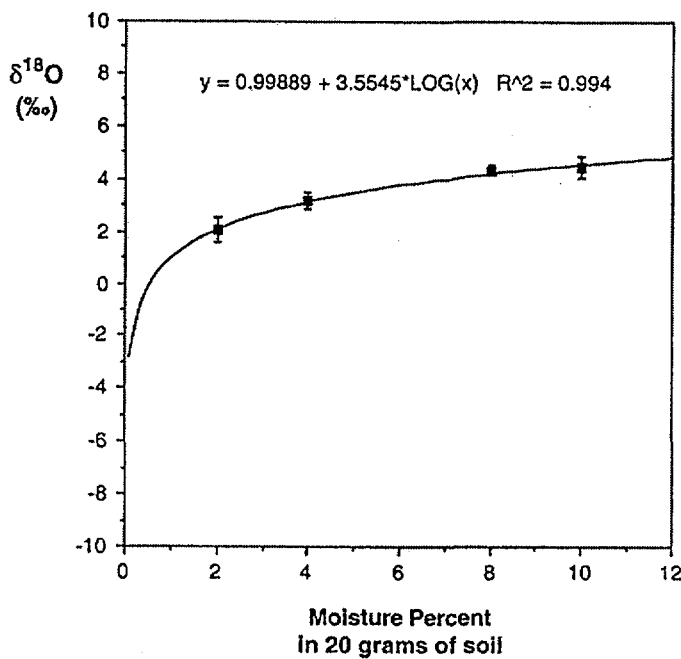


FIGURE 4.

4.3.3. Tritium

The entire contents of a 3 kg jar is weighed and then emptied into a metal vessel. The lid of the vessel has a vacuum tight seal and a conical joint on which is mounted a Liebig condenser with a 1 litre collection flask and a capillary connection to a vacuum pump. The metal vessel is first evacuated through the capillary (to limit the loss of water vapour) and then heated to about 100°C. The distillation proceeds until the sand is dry. Both the sand and the collected water are weighed. Typical moisture recovery is around 95%. The resulting fractionation is too small significantly to influence the tritium values.

Aliquots of 110 ml of water are made up, either from a single soil sample if it yielded enough water, or by adding some water from the previous, or following, sample. Sodium peroxide is added to the water to make a 2% NaOH solution and batches of samples are electrolysed to a final volume of 15 ml. The resulting enrichment in tritium is about 6x. A zero tritium blank sample and a tritium spike is run with each batch. The enriched samples are then vacuum distilled. A 10 ml sample is introduced into a standard counting vial, Ultima Gold scintillator is added, shaken and counted in a Packard 2770 TR/SL low-level liquid scintillation counter.

5. RAINFALL DATA

The owner of the farm Harefield had kept meticulous rainfall records since 1966 which are shown in Table 3 and Fig. 5. A few salient features are noted:

1. The rainfall is strongly seasonal. The wettest months are November - March. During the winter months May - August, rainfall is rare.
2. The rainfall for individual months is highly variable from year to year.
3. There is the suggestion of a cycle in annual totals of the order of 11 years (cf. long-term mean of 337 mm a⁻¹).
4. Unusually late rains fell shortly before the profiles were taken in July 1997.

6. THE SATURATED ZONE

The three boreholes sampled were all drilled into the basal gravels of the Kalahari sequence. In 1977 the rest levels in the area lay in the range of 70-80m (Verhagen et. al 1979(b)). Isotope and other observations on pumped water from the three boreholes are given in Table 4.

In all cases the radiocarbon values are rather low, the result for the Harefield pumped borehole suggesting that confined water is being extracted. If the deep aquifer is represented by the value of the pumped borehole on Harefield, then the Wylie Valley borehole can be interpreted as pumping a mixture of the old, confined and more recent ground water, overlying the clay. The Harefield wind pump (KSP 3) at 48 pMC produces much more recent water, with significantly more positive stable isotope values.

The model which emerges from these observations is the following. When infiltrating rain water reaches the saturated zone, it recharges the upper aquifer. The lower, below the aquitard/aquiclude, is said to be sub-artesian. The deeper aquifer will therefore react only very slowly, if at all, to such recharge events. Evapotranspirative losses will be more pronounced from the shallow, phreatic aquifer, which therefore becomes ephemeral. Following the high rainfall period 1973-75 (Fig. 5), there is a suggestion of significant rest level rises in the area. Based on the unsaturated zone profiles obtained in 1977, an infiltration rate of the order of 15% of mean annual rainfall was derived for the high rainfall period. Although the radiocarbon values are low, suggesting MRT values for the ground water in the range of 3000 years and greater, these should be interpreted in terms of the mixture of ground water likely to be produced by the deep boreholes

Table 3. Rainfall Data for Harefield Farm

	66	67	68	69	70	71	72	73	74	75	76	77	78	79	80	81
	65/66	66/67	67/68	68/69	69/70	70/71	71/72	72/73	73/74	74/75	75/76	76/77	77/78	78/79	79/80	80/81
Sep	0	0	0	0	0	0	0	0	0	8	3	9	40	15	12	16
Oct	0	7	0	30	13	0	24	0	41	10	0	23	23	6	58	0
Nov	17	0	2	0	9	30	5	25	50	28	42	3	39	2	7	81
Dec	3	25	7	36	19	16	67	0	106	18	211	7	76	23	36	18
Jan	47	81	38	18	16	29	127	5	191	85	218	104	38	47	18	34
Feb	46	93	0	66	20	102	12	106	121	19	55	51	80	171	74	103
Mar	8	25	43	6	0	56	114	85	251	54	156	280	77	11	52	39
Apr	11	42	40	76	31	11	80	68	31	43	68	39	19	22	17	0
May	0	49	68	17	19	46	0	0	16	7	13	0	0	0	0	0
Jun	11	0	13	0	9	0	0	0	0	0	0	0	0	0	0	0
Jul	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Aug	0	0	0	0	8	0	0	0	0	31	0	0	0	0	12	50
	143	322	211	249	144	290	429	289	807	303	766	516	392	297	286	341
	337	337	337	337	337	337	337	337	337	337	337	337	337	337	337	337

	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	Average
	81/82	82/83	83/84	84/85	85/86	86/87	87/88	88/89	89/90	90/91	91/92	92/93	93/94	94/95	95/96	#DIV/0!
Sep	0	0	3	0	13	41	36	15	0	0	0	0	0	0	0	4
Oct	31	77	6	18	10	0	5	11	0	0	12	8	88	2	14	0
Nov	108	37	75	23	7	32	33	37	22	0	8	25	22	0	26	74
Dec	92	17	73	9	83	33	37	118	25	50	28	17	12	15	74	75
Jan	0	40	17	41	9	55	21	169	48	128	0	16	29	34	49	109
Feb	70	0	0	59	47	212	206	59	54	81	16	115	40	0	71	33
Mar	17	29	94	23	17	62	49	17	28	169	44	23	56	84	0	21
Apr	33	31	13	9	14	60	77	112	73	0	11	0	0	0	13	42
May	0	12	22	0	8	0	7	30	0	0	0	0	0	26	6	69
Jun	0	7	0	0	7	0	0	0	0	43	0	0	0	0	0	70
Jul	17	0	0	0	0	0	0	0	0	0	0	0	0	0	0	79
Aug	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	37
	368	250	303	182	215	495	471	568	250	471	119	204	247	161	253	372
	337	337	337	337	337	337	337	337	337	337	337	337	337	337	337	#DIV/0!

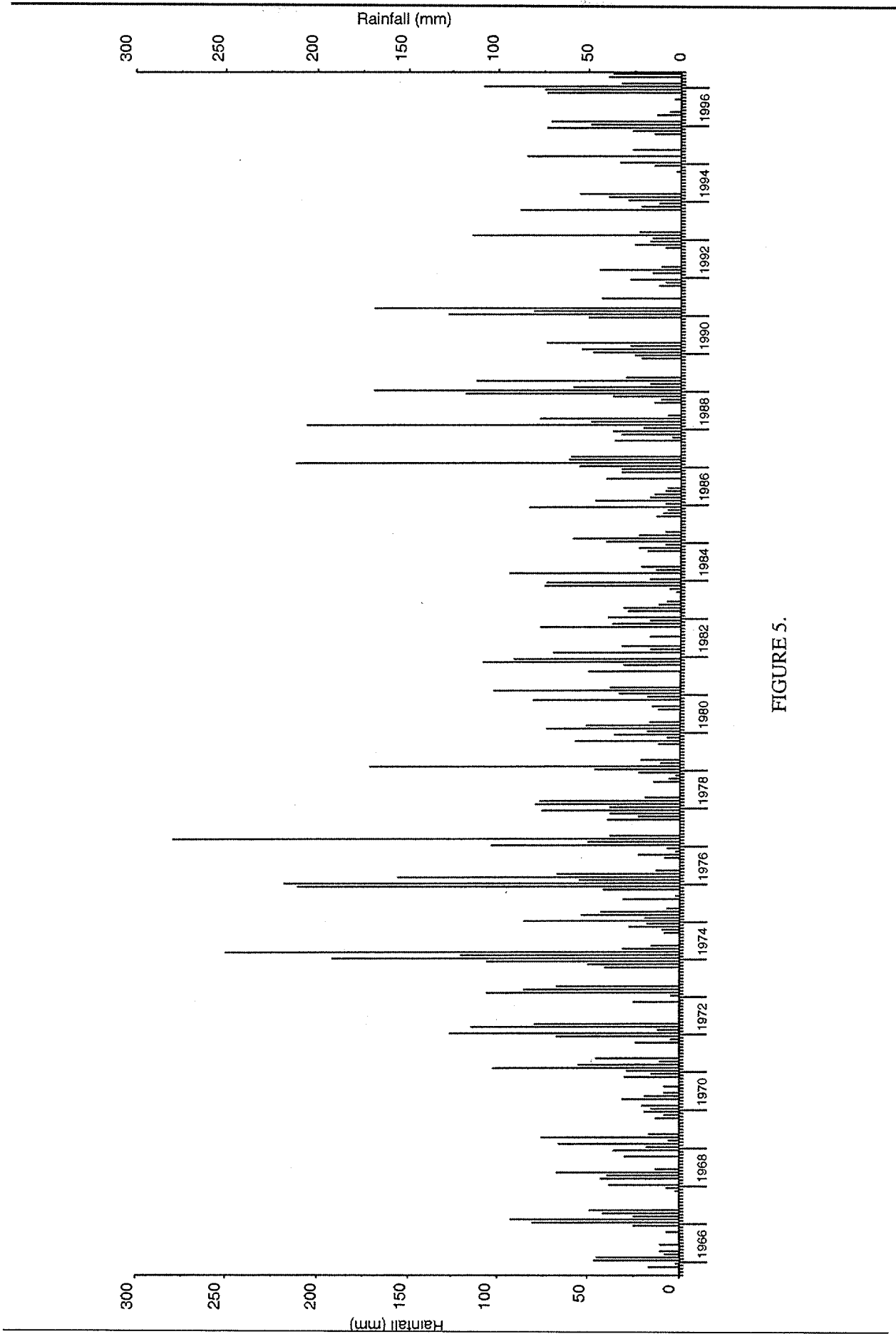


FIGURE 5.

Table 4. Borehole Data

Sample	Radiocarbon (pMC)	$\delta^{18}\text{O}$ (‰)	δD (‰)	[Cl] (mg/l)	E.C. ($\mu\text{S}/\text{cm}$)
KSP 1	33.6 \pm 1.6	-5.97	-41.5	34	300
KSP 2	9.2 \pm 1.4	-5.88	-39.6	90	670
KSP 3	48.2 \pm 1.7	-5.22	-35.7	88	660
Wylie V WP 1977	36.8	-5.80	-38.0		300

7. DATA FROM THE PROFILES

The results of the measurements on sand samples from the different auger boreholes taken during 1997 and 1998 are shown in Tables 5 to 10 and in Figs. 6 to 11.

These results are discussed in terms of each of the two sampling periods.

7.1 The 1997 Profiles: BH 10; BH 11; BH 12

7.1.1 *Moisture*

The moisture content of all three profiles shows a high near the surface. This suggests the influence of the relatively good 1996/97 (and possibly 1995/96) rainy seasons and the above-average late-season rain which had fallen in the area (see Table 3 and Fig. 5). The high is then followed in all three cases by a minimum around 1 metre; a maximum around three metres and then a gradual increase for Bh 10 and Bh 11 to a value of some 2.3 % near the bottom.

7.1.2 *Oxygen-18*

The inflection in $\delta^{18}\text{O}$ at the very top of the profile, typical of the evaporation front (Allison et al 1994; Barnes and Allison 1988), is not observed, even with the finer depth resolution in BH 12. The first ~ 0.10 metres of the sand column was effectively dry. There is a general decline in $\delta^{18}\text{O}$ values, the shapes of the observed curves placing them in the category of profiles characteristic of low recharge (Allison et al. 1994).

In BH 12 the top-most $\delta^{18}\text{O}$ value shows extreme enrichment which suggests that the upward trend in the $\delta^{18}\text{O}$ profiles for the other auger holes may continue to the surface. A strong transient drying condition following the rains which fell shortly before might have been recorded in this sampling episode. It might be speculated whether an inflection might have been recorded in the $\delta^{18}\text{O}$ profile for BH 12 had it been sampled at even smaller intervals. The rest of the profile gives values considerably more positive than for corresponding depths in the other two. It is as yet unclear whether this difference is an artifact of the measurements.

7.1.3 *Electrical conductivity and chloride concentration.*

The profiles of EC and Cl^- concentrations for BH 10 show strong similarities. This suggests that chloride makes up an important component of the overall mineralisation of the soil moisture. For the first 10 metres or so these parameters increase with increasing moisture content. It appears that near-surface mineralisation, increased by evaporation, is being washed down by successive infiltration events. The broad minimum between -11 m and -16 m coincides with a

Table 5. Borehole 10

Lab No.	Depth (m)	$\delta^{18}\text{O}$ (‰)	Tritium (TU)	Moisture (%)	Chloride (ppm)	EC. (μmhos)
KSP 4	0.3	+1.85		1.84	1316	2718
KSP 5	1.0	-1.90	11.2±0.6	1.08	630	1940
KSP 6	2.0	-2.97		1.92	337	1039
KSP 7	3.0	-2.82		1.85	964	3515
KSP 8	4.0	-3.86	7.2±0.5	1.57	746	1909
KSP 9	5.0	-5.59		1.55	355	812
KSP 10	6.0	-5.37		1.71	373	1168
KSP 11	7.0	-4.78	6.6±0.5	2.10	463	1380
KSP 12	8.0	-5.13		1.87	210	728
KSP 13	9.0	-6.55		2.35	286	959
KSP 14	10.0	-5.93	7.3±0.5	2.28	391	1537
KSP 15	11.0	-6.71		2.17	189	1013
KSP 16	12.0	-6.53	6.9±0.5	2.16	195	1293
KSP 17	13.0	-6.93		1.89	219	1191
KSP 18	14.0	-7.48	7.6±0.5	1.97	185	1457
KSP 19	15.0	-7.49		2.21	205	1403
KSP 20	16.0	-7.23	13.5±0.8	2.02	229	1484
KSP 21	17.0	-7.29		2.18	588	1837
KSP 22	18.0	-6.50	8.1±0.6	2.33	525	2465

Table 6. Borehole 11

Lab No.	Depth (m)	$\delta^{18}\text{O}$ (‰)	Moisture (%)	Chloride (ppm)	EC. (μmhos)
KSP 23	0.5	+0.09	3.11	152	270
KSP 24	1.0	-2.56	1.57	306	510
KSP 53	1.2	-1.34	1.64	265	468
KSP 25	1.6	-2.72	1.45	367	483
KSP 26	2.0	-1.98	1.56	234	481
KSP 27	2.4	-1.76	1.96	259	509
KSP 28	2.8	-2.10	2.24	178	335
KSP 54	3.0	-1.91	2.28	350	438
KSP 29	3.5	-2.89	2.34	206	320
KSP 30	4.0	-2.91	2.04	199	425
KSP 31	4.5	-3.52	2.14	257	552
KSP 32	5.0	-5.00	1.54	401	812
KSP 33	5.5	-4.79	1.69	830	1365
KSP 34	6.0	-4.83	1.70	268	708
KSP 35	6.5	-4.65	1.61	206	497
KSP 36	7.0	-4.74	1.67	255	660
KSP 37	7.5	-5.27	1.80	362	974
KSP 38	8.0	-5.77	2.05	289	978
KSP 39	8.5	-5.57	2.14	655	1770
KSP 40	9.0	-6.70	2.14	430	623
KSP 41	9.5	-5.35	2.21	171	544
KSP 42	10.0	-5.36	1.95	258	700
KSP 43	10.5	-5.68	1.87	394	1739
KSP 44	11.0	-6.34	2.28	221	745
KSP 45	11.5	-6.90	2.14	243	983
KSP 46	12.1	-6.38	2.18	288	893
KSP 47	12.6	-6.63	2.11	275	1406
KSP 48	13.1	-5.56	2.14	242	1356
KSP 49	13.6	-6.40	2.05	286	1446
KSP 50	14.1	-6.91	2.19	279	1200
KSP 51	14.6	-6.65	2.13	238	1249
KSP 52	15.0	-6.69	2.35	66	1468

Table 7. Borehole 12

Lab No.	Depth (m)	$\delta^{18}\text{O}$ (‰)	Moisture (%)
KSP 55	0.10	+8.82	2.98
KSP 56	0.25	+3.57	2.56
KSP 57	0.35	+2.44	2.79
KSP 58	0.45	+1.56	2.91
KSP 59	0.55	+2.24	1.48
KSP 60	0.60	+1.42	1.96
KSP 61	0.75	+2.75	1.00
KSP 62	0.85	+1.39	1.00
KSP 63	1.00	+1.21	1.08
KSP 64	1.10	+1.71	1.09
KSP 65	1.25	-1.56	1.17
KSP 66	1.35	-1.98	1.26
KSP 67	1.45	+0.75	1.30
KSP 68	1.55	-0.14	1.22
KSP 69	1.70	-0.13	1.33
KSP 70	1.80	-1.01	1.40
KSP 71	1.95	-1.11	1.58
KSP 72	2.05	-0.05	1.69
KSP 73	2.15	+0.54	1.65
KSP 74	2.30	-0.47	1.64
KSP 75	2.40	+0.83	1.65
KSP 76	2.55	-0.34	2.01
KSP 77	2.65	+0.68	2.22
KSP 78	2.80	-0.38	2.27

Table 8. Borehole 13

Lab No.	Depth (m)	$\delta^{18}\text{O}$ (‰)	δD (‰)	Deuterium Excess	Tritium (T.U.)	Moisture %	Chloride (ppm)	E.C. (μmhos)
KSP 79	0.5	-2.02	-16.5	-0.34		1.09	334	1614
KSP 80	1.0	-2.25			6.1±0.7	1.09	239	1615
KSP 81	1.5	-1.86	-24.8	-9.92		1.71	79	965
KSP 82	2.0	-0.87				1.71	118	755
KSP 83	2.5	-2.04	-36	-19.68		0.97	411	2349
KSP 84	3.0	-0.38			3.5±0.6	0.97	317	1471
KSP 85	3.5	-1.28	-18.2	-7.96		1.67	222	1051
KSP 86	4.0	-2.11				1.67	142	705
KSP 87	4.5	-2.3	-13.5	4.9		1.78	101	571
KSP 88	5.0	-2.51			3.2±0.6	1.78	133	694
KSP 89	5.5	-2.2	-20.5	-2.9		1.92	193	1077
KSP 90	6.0	-3.02				1.92	131	669
KSP 91	6.5	-3.68	-31.5	-2.06	5.7±0.7	2.05	132	538
KSP 92	7.0	-4.91				2.05	48	361
KSP 93	7.5	-4.74	-28	9.92	6.0±0.7	2.06	96	812
KSP 94	8.0	-4.61				2.06	176	763
KSP 95	8.5	-4.86	-33.4	5.48	3.3±0.6	2.33	116	508
KSP 96	9.0	-4.71				2.33	82	339
KSP 97	9.5	-4.98	-37.8	2.04	4.7±0.7	2.41	84	546
KSP 98	10.0	-5.12				2.41	78	685
KSP 99	10.5	-5.04	-39.7	0.62		2.25	84	741
KSP 100	11.0	-4.78				2.25	80	546
KSP 101	11.5	-4.86	-37.4	1.48	5.9±0.7	2.36	121	1053
KSP 102	12.0	-4.6				2.36	60	596
KSP 103	12.5	-4.66	-32.4	4.88	5.4±0.7	2.33	146	1382
KSP 104	13.0	-4.37				2.33	163	1242
KSP 105	13.5	-5.03	-30.8	9.44	6.5±0.7	2.40	166	1290
KSP 106	14.0	-5.84				2.40	146	1255
KSP 107	14.5	-5.56	-36.6	7.88	4.4±0.6	2.14	167	1502
KSP 108	15.0	-5.73				2.14	214	2373
KSP 109	15.5	-5.6	-37.9	6.9		2.56	130	1877
KSP 110	16.0	-5.39				2.56	110	1002
KSP 111	16.5	-5.73	-35.2	10.64	4.3±0.6	2.39	176	2116
KSP 112	17.0	-5.46				2.39	164	2635
KSP 113	17.5	-5.09	-31.8	8.92	4.3±0.6	2.62	153	1979
KSP 114	18.0	-3.66				2.62	129	1932
KSP 115	18.5	-4.72	-27.2	10.56		2.51	120	1309
KSP 116	19.0	-4.7				2.51	162	1783
KSP 117	19.5	-3.64	-25.9	3.22	5.5±0.7	2.49	135	1754
KSP 118	20.0	-3.12				2.49	151	1938
KSP 119	20.5	-1.96	-25.4	-9.72		3.00	115	1672
KSP 120	21.0	-2.32				3.00	146	1551
KSP 121	21.5	-2.86	-25.7	-2.82	5.2±0.7	2.93	132	1452
KSP 122	22.0	-2.85				2.93	127	1463

Table 9. Borehole 14

Lab No.	Depth (m)	$\delta^{18}\text{O}$ (‰)	Moisture %	Chloride (ppm)	E.C. (μmhos)
KSP 139	0.5	-1.86	0.66	629	2851
KSP 140	1.0	-3.09	1.36	170	457
KSP 141	1.5	-1.19	2.10	177	565
KSP 142	2.0	-1.02	1.51	167	405
KSP 143	2.5	-1.15	1.30	208	480
KSP 144	3.0	-0.27	1.51	196	447
KSP 145	3.5	-1.52	1.10	281	808
KSP 146	4.0	-1.89	1.44	221	440
KSP 147	4.5	-1.81	1.68	219	536
KSP 148	5.0	-2.18	1.35	303	1347
KSP 149	5.5	-2.34	1.03	440	2098
KSP 150	6.0	-2.69	1.63	231	509
KSP 151	6.5	-2.58	1.74	218	870
KSP 152	7.0	-3.17	1.26	228	564
KSP 153	7.5	-3.87	2.15	161	719
KSP 154	8.0	-3.09	1.92	214	891

Table 10. Borehole 15

Lab No.	Depth (m)	$\delta^{18}\text{O}$ (‰)	Moisture %	Chloride (ppm)	E.C. (μmhos)
KSP 123	0.5	-1.8	1.04	128	496
KSP 124	1.0	-2.24	0.97	167	504
KSP 125	1.5	-1.84	0.94	200	1043
KSP 126	2.0	-0.62	1.28	157	764
KSP 127	2.5	-1.44	1.08	182	812
KSP 128	3.0	-0.99	0.77	262	1234
KSP 129	3.5	-1.72	1.06	223	773
KSP 130	4.0	-2.22	1.41	174	973
KSP 131	4.5	-1.74	1.75	261	1107
KSP 132	5.0	-2.27	1.16	413	1885
KSP 133	5.5	-4.02	1.30	257	1368
KSP 134	6.0	-3.51	1.62	215	969
KSP 135	6.5	-4.73	1.64	389	1564
KSP 136	7.0	-4.9	1.67	184	708
KSP 137	7.5	-6.07	1.65	191	969
KSP 138	8.0	-5.98	1.99	176	491

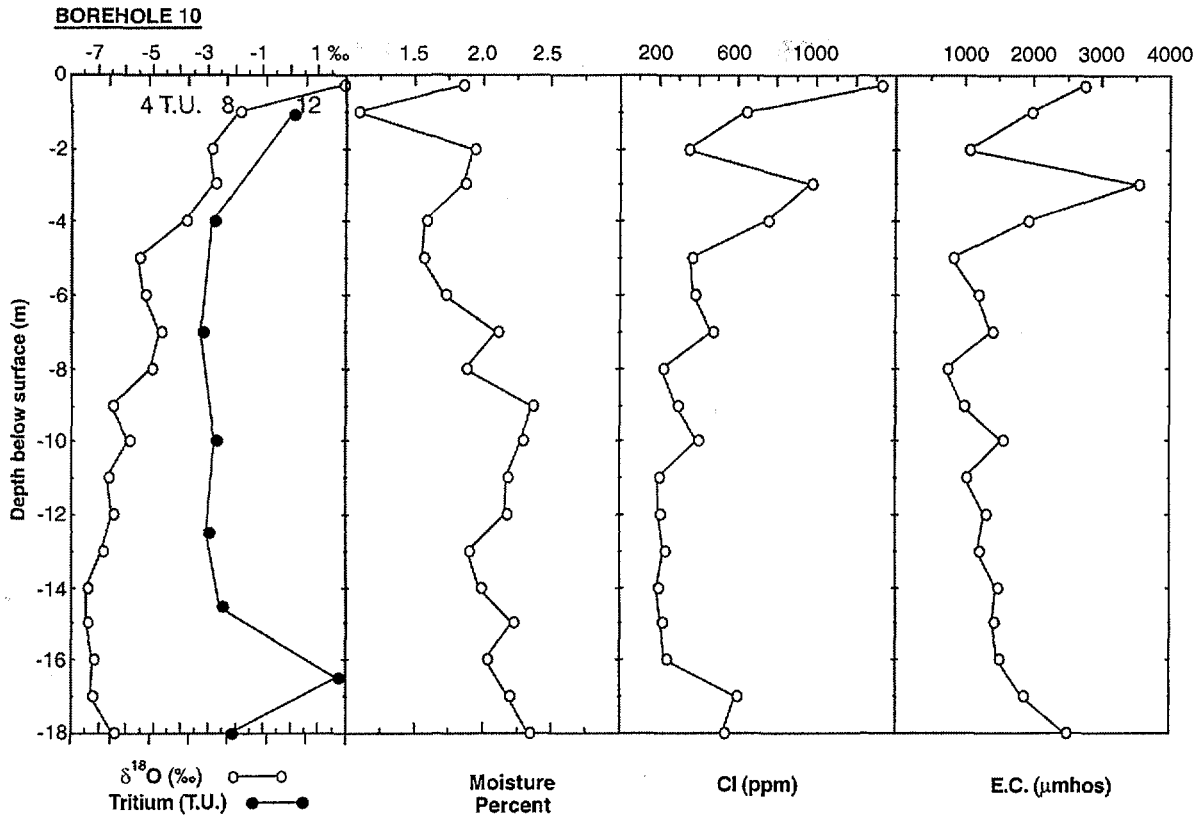


FIGURE 6.

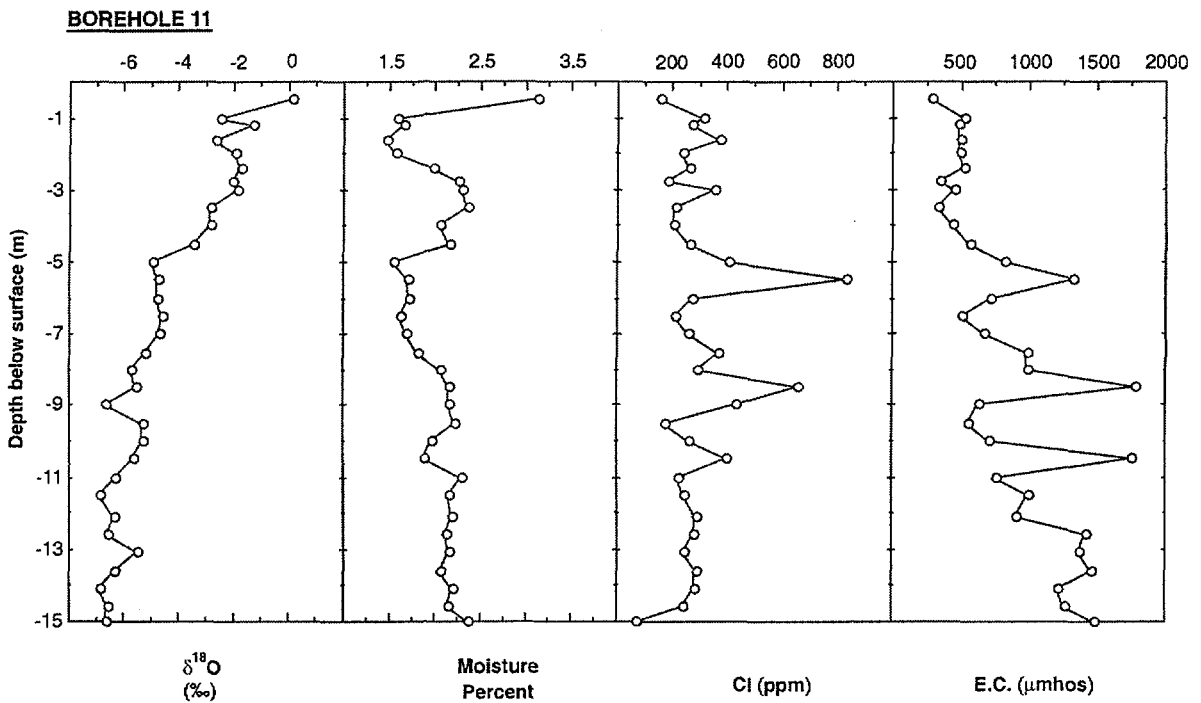


FIGURE 7.

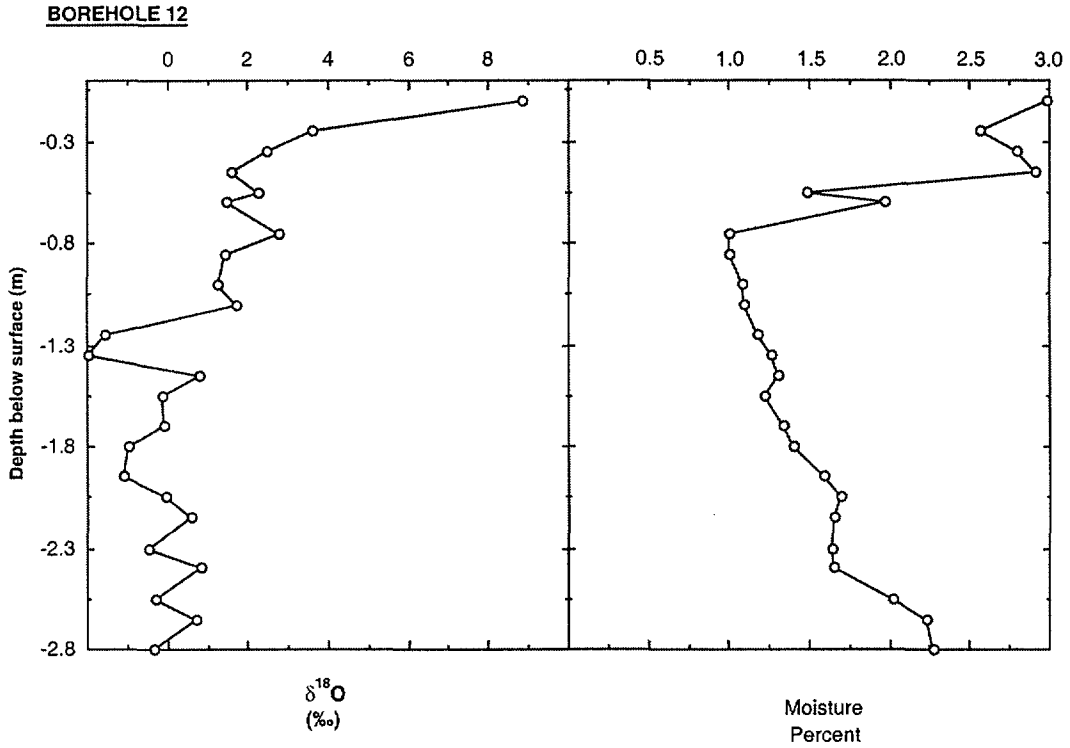


FIGURE 8

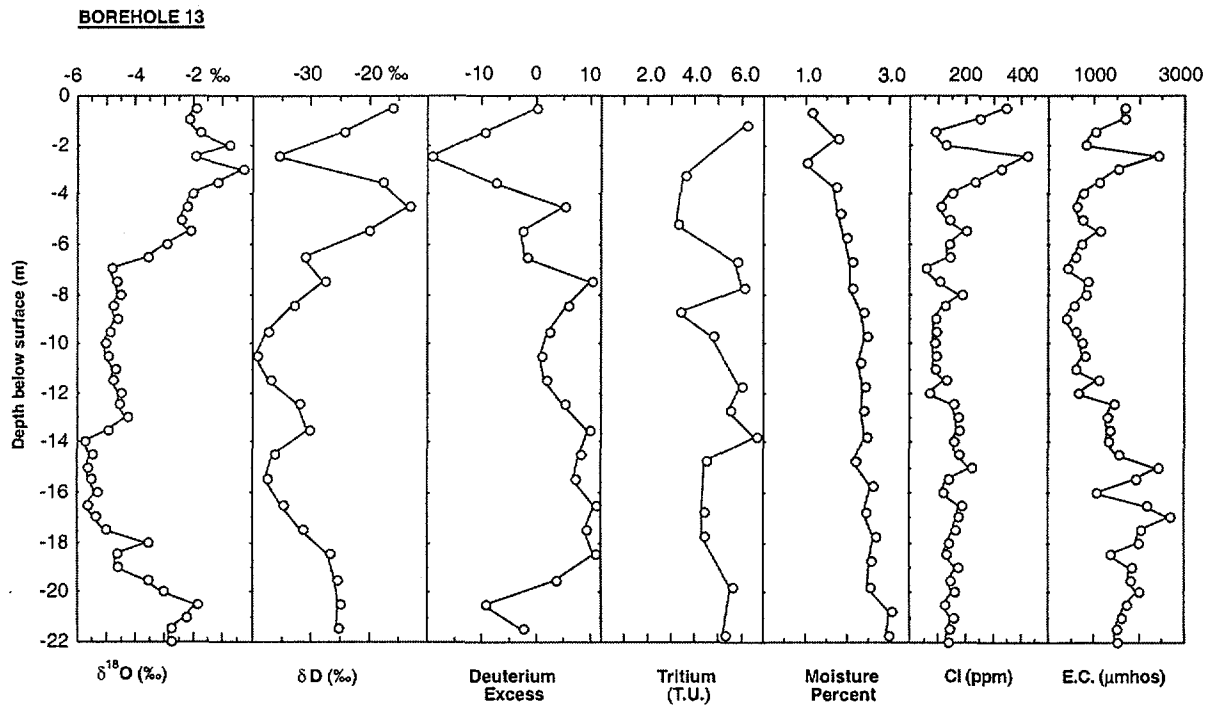


FIGURE 9.

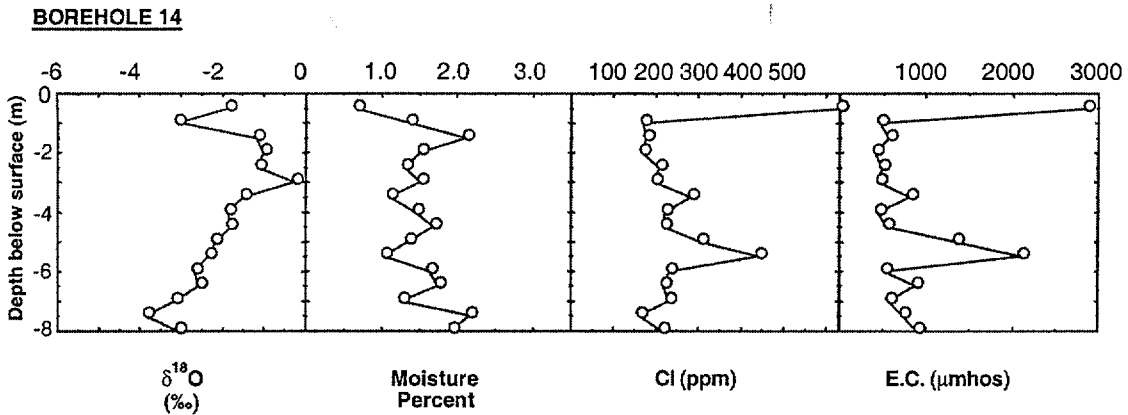


FIGURE 10

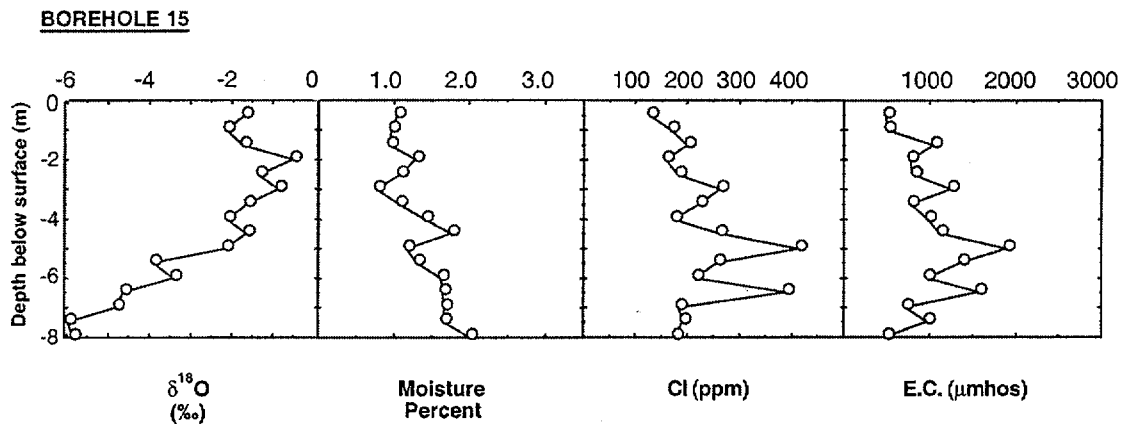


FIGURE 11

minimum in the $\delta^{18}\text{O}$ profile. All three parameters show a further rise near the bottom, which may represent the trailing edge of a displacement front. At this depth too, there is an erratic, but general increase in moisture content.

The relative shapes of the EC and Cl^- curves for BH 10 show $\text{EC}/[\text{Cl}^-]$ ratios ranging from about 3 to about 7. This is reversed near the surface, where the ratio drops to about 0.4, the EC probably representing almost pure chloride. Elsewhere, there is a substantial concentration of other ions, probably mainly HCO_3^- , which become quite dominant in the -11m to -16m zone, where the ratio rises to 7. This might identify the zone as being associated with a significant recharge period, on account of the increase in biogenic soil CO_2 .

The profiles of EC and Cl^- values for BH 11 differ in many ways from those of BH 10. The lower values near the top of the profile seem to reflect the relatively higher moisture contents there. In contrast to BH 10, Cl^- tends to be more prominent with an isolated peak at about -6.5 m. The tendency for the other ions, probably mainly HCO_3^- , to dominate, seen deeper in BH 10, is also evident here, at roughly the same values. Overall, there is much less correlation between EC and Cl^- values, and moisture and $\delta^{18}\text{O}$ profiles than seen in BH 10.

It is remarkable that the behaviour of these parameters can differ so greatly between profiles which are separated laterally by a distance roughly equalling their depths.

7.1.4 Tritium

In the 1997 sample set, tritium was measured only in 1998 and then only for BH 10. The results are presented in Table 5 and displayed in Fig. 6. Values higher than expected for average present-day rainfall are observed all the way down the profile. At 0 - 2 m and 16 - 17 m somewhat higher values are observed. These are not interpreted readily. Expected rainfall tritium values for 1997 in the area should lie around 5 TU. However, tritium values up to 15 TU were measured in rainfall samples taken at Johannesburg during the exceptional 1995/96 rainy season which produced monsoon-type conditions from southwards-moving tropical air masses.

When samples for tritium analysis were distilled during 1998, they included a zero sample which was made up by moistening a 3 kg thoroughly dried sand sample to 2% by weight with water at 0 TU. The water was retrieved by distillation and electrolysed as for the auger samples. The enrichment-corrected result of 0.0 ± 0.3 TU suggests that there is minimal tritium contamination during the entire laboratory sample handling procedure.

7.2 1998 Profiles: BH 13; BH 14; BH 15

7.2.1. Moisture contents.

Moisture was quantitatively distilled from the 3 kg samples for tritium analysis for BH 13. The moisture content was determined by weighing the entire sample before and after distillation. In the case of BH 14 and 15, sand aliquots were removed from the 3 kg samples and weighed before and after drying in the oven. The results are shown in Figs. 9, 10 and 11.

Compared with the 1997 profiles, the BH 13,14,15 showed much lower moisture content just below surface. This is ascribed to the absence of any rainfall for several weeks prior to sampling. As in the earlier profiles, the uppermost 8 metres or so show considerable variations with depth as well as differences in depth dependence over relatively small lateral distances. As in the 1997 results, the profiles become smoother at greater depths. The moisture profiles of the deeper boreholes BH 10, 11, 13 are compared in Fig. 12 and are quite similar, steadily increasing with depth in all three.

As yet, the controls on these moisture variations are not clear. A possible control could be found in the grain size distribution, as was observed in the profiles obtained in the earlier study of 1977.

7.2.2 Electrical conductivity and chloride concentrations

To depths of some 8 m the EC and Cl^- profiles for the deep boreholes BH 10 and BH 13 profiles are quite similar. As was observed for BH 10, the EC for the upper part of the BH 13 profile is dominated by Cl^- . Between 8 and 22 m, Cl^- values increase slightly from about 100 ppm to 150 ppm, whilst EC values increase from about 600 μS to well over 2000 μS . The ratio $\text{EC}/[\text{Cl}^-]$ ranges from 5 - 7 near the top of the profile, increasing to 10 - 15 near the bottom. This suggests the presence of at least one other ion, which, in the case of BH 10, was speculated to be HCO_3^- . This relative increase in HCO_3^- , and lower values of Cl^- , both suggest higher recharge rates with concomitant increase in vegetal activity and release of biogenic CO_2 .

7.2.3. Oxygen-18

Whereas the $\delta^{18}\text{O}$ profiles of BH 10,11,12 exhibited a sharp drop from positive values over the first metre or so from surface, no such drop is seen in the profiles for BH 13,14,15. The latter, taken shortly after late-season rain, show an increase in values down to some 4 m, and then

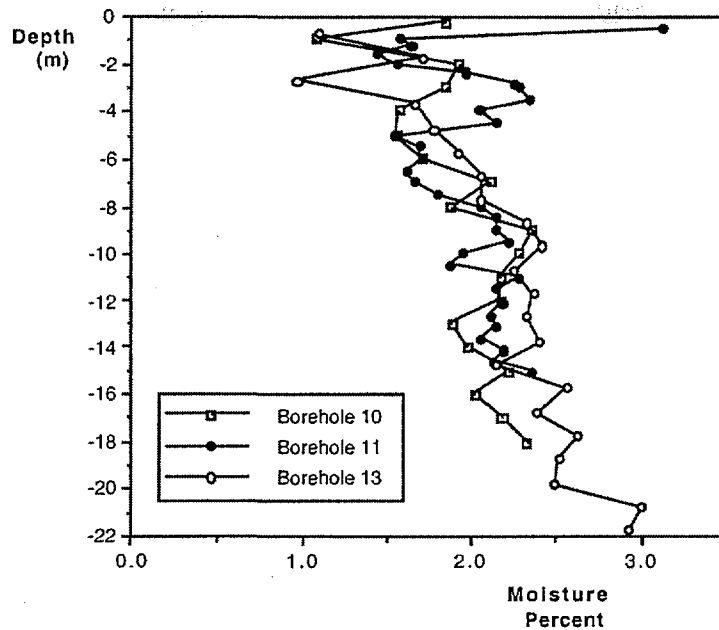


FIGURE 12

a decline to about 8 m. From there, BH 13 shows fairly constant values with a further minimum at 14 - 17 m. Then follows a marked increase down to 22 m. A minimum at about 14 m is also suggested in the $\delta^{18}\text{O}$ profiles of BH 10,11, but at values more negative than for BH 13.

All the $\delta^{18}\text{O}$ profiles analysed to date are plotted in Fig. 13 for comparison. Although there are considerable differences in the individual profiles, some general trends are observed:

- i. Only the 1997 profiles rise to values of 0 ‰ and more positive.
- ii. Around 2 m, all profiles lie in the range 0 ‰ to -2 ‰.
- iii. A decline of values is seen down to 14 m, albeit in the individual values for the profiles spanning a range of about 2 ‰
- iv. There then follows an increasing trend, which persists in BH 13 down to its final depth of 22 m.

The inflection at the very top in the $\delta^{18}\text{O}$ - depth curve, proposed as typical of the near-surface evaporation/drying front (Allison et al. 1994; Barnes and Allison 1988), is not observed in any. There is a general decline in $\delta^{18}\text{O}$ values, the shapes of the observed curves placing them in the category of profiles characteristic of low recharge (Allison et al 1994).

7.2.4 Deuterium

Deuterium analyses were performed on samples at depth intervals of one metre for BH 13. The direct soil moisture reduction system functioned very well, giving excellent reproducibility for duplicate samples (see Table 11). Although the shape of the δD profile resembles that of $\delta^{18}\text{O}$ for depths below 4 metres, it appears quite different for the shallower samples. Figure 14 shows a plot of δD vs $\delta^{18}\text{O}$ for BH 13 with the world meteoric water line (WMWL) for comparison. From this it is clear that evaporation has influenced a number of the points. The deuterium excess d for all the samples in the profile is plotted against depth, along with other parameters, in Figure 9. Low values for d are seen particularly for the shallow samples, confirming the conclusion from the $\delta^{18}\text{O}$ data that the high values above some 6 m depth are due to kinetic evaporation. It has

been remarked earlier that the classical evaporation front with its characteristic inflection is not visible in any of the profiles - albeit suggested in BH 12. Probably, the d values are produced by the process associated with such a front, but that repeated rapid infiltration events, which also appear to influence deeper sections of the profiles, distort the regular pattern such as observed by e.g. Allison et al. (1994).

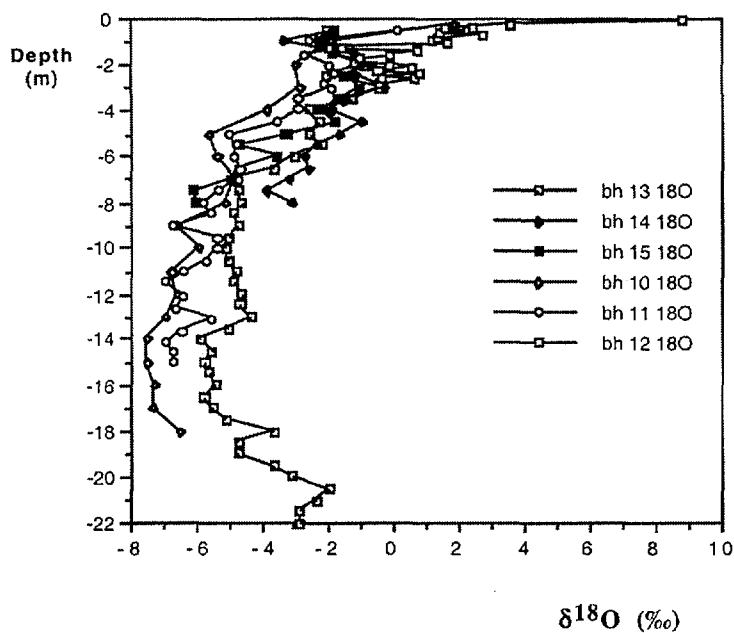


FIGURE 13

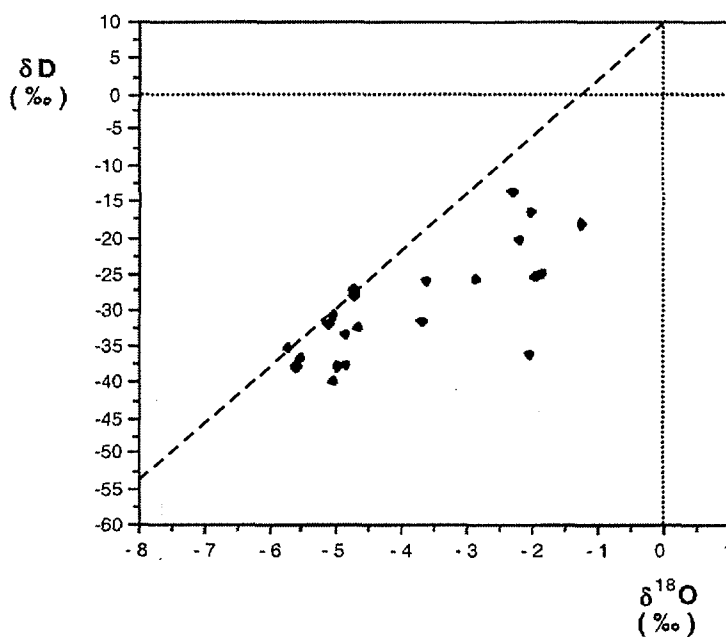


FIGURE 14

Table 11. Results for Deuterium Analyses

Sample Number	Normalised δD (‰)	Average δD (‰)
KSP 79	-16.8	-16.5
	-16.2	
KSP 81	-24.3	-24.8
	-25.3	
KSP 83	-36.2	-36.0
	-35.9	
KSP 85	-18.1	-18.2
	-18.3	
KSP 87	-14.0	-13.5
	-12.9	
KSP 89	-20.7	-20.5
	-20.2	
KSP 91	-32.0	-31.5
	-30.9	
KSP 93	-27.7	-28.0
	-28.3	
KSP 95	-33.9	-33.4
	-32.9	
KSP 97	-38.0	-37.8
	-37.6	
KSP 99	-39.9	-39.7
	-39.6	
KSP 101	-38.4	-37.4
	-36.4	
KSP 103	-31.9	-32.4
	-32.9	
KSP 105	-30.1	-30.8
	-31.5	
KSP 107	-35.8	-36.6
	-37.5	
KSP 109	-38.4	-37.9
	-37.4	
KSP 111	-34.9	-35.2
	-35.6	
KSP 113	-31.9	-31.8
	-31.8	
KSP 115	-26.9	-27.2
	-27.5	
KSP 117	-25.9	-25.9
	-25.9	
KSP 119	-25.4	-25.4
	-25.3	
KSP 121	-25.6	-25.7
	-25.9	

At greater depths, values for d then rise to around +10 ‰, confirming that these are associated with direct rainfall infiltration. Values of d drop somewhat near the very bottom of the profile. These are associated with an inflection in the $\delta^{18}\text{O}$ profile. This is tentatively interpreted as due to contamination of deeper auger samples by material from the near the top of the auger hole. Such contamination is not seen however in any of the other parameters measured for BH 13.

7.2.5 Tritium

The tritium results for BH 13 are given in Table 6 and plotted against depth in Fig. 9. On average, the values lie about 2 TU lower than those observed for BH 10 and the variations with depth are less pronounced. In neither case can the variations in tritium concentration be correlated with any of the other measured parameters.

8. RECHARGE ESTIMATES

8.1. Chloride concentrations

As there is no suitable time marker, such as a discernible tritium peak, no attempt was made to assess recharge by chloride balance for the entire moisture profiles. Recharge rate, or rather nett infiltration, was derived using the chloride mass balance equation:

$$C_P \cdot P = C_R \cdot R$$

where P is the precipitation, C_P the chloride concentration in rain, (about 1 ppm; Beekman et al. 1996), and C_R the chloride concentration in recharge. A mean annual rainfall of 336 mm is assumed (Table 3). The lowest chloride concentrations observed, and maximum recharge rate, are respectively:

$$\begin{aligned} \text{BH 10 (1997)} \quad C_R &= 200 \text{ ppm} ; R = 1.8 \text{ mm a}^{-1} \\ \text{BH 13 (1998)} \quad C_R &= 80 \text{ ppm} ; R = 5.0 \text{ mm a}^{-1} \end{aligned}$$

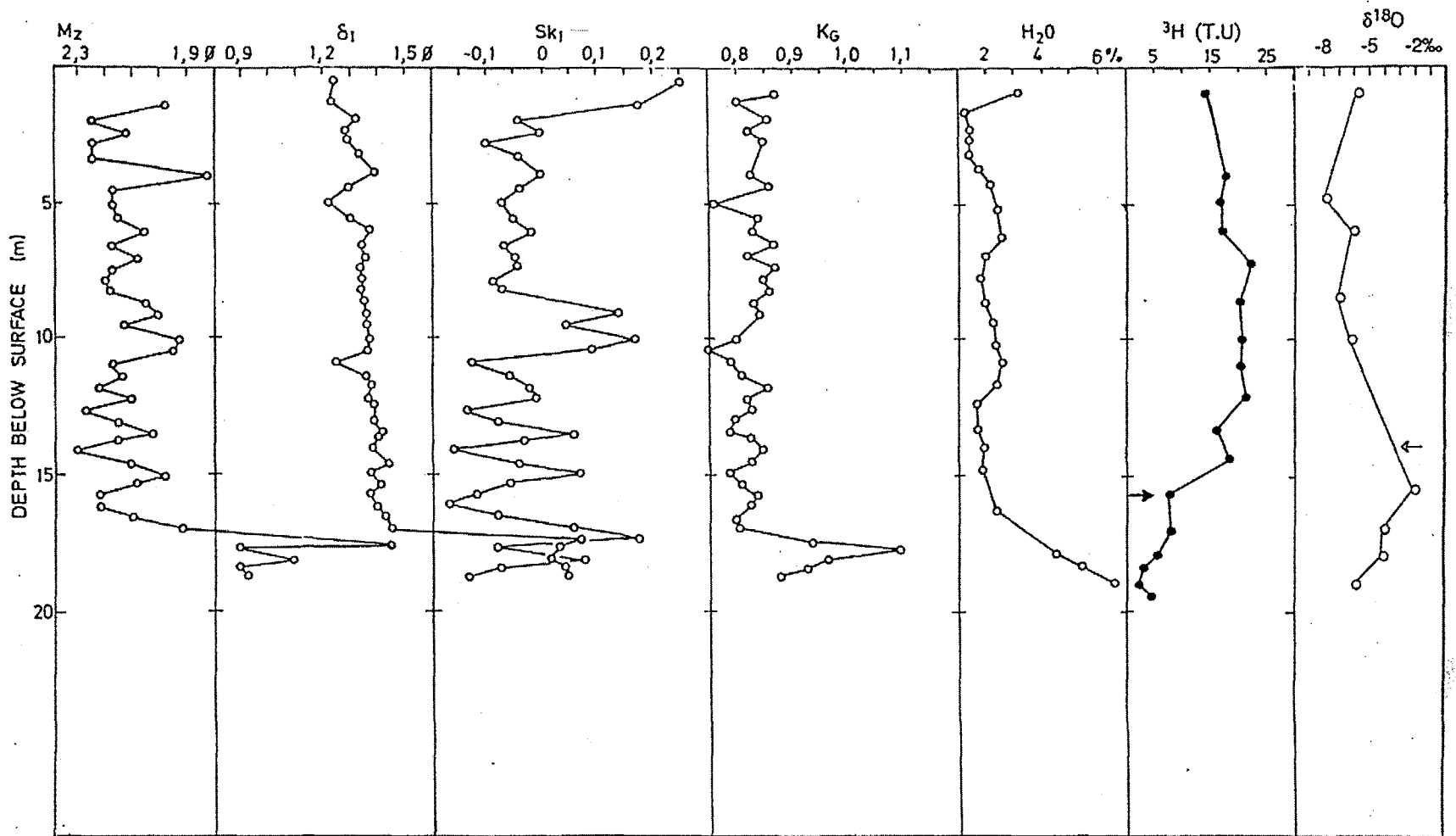
These figures reflect only the inferred downward movement of moisture as an equivalent water column in part of the profile, and set an upper limit to the inferred recharge rate to the underlying aquifer.

8.2 Tritium values

The result of zero TU for the control sample gives confidence that the tritium values measured in the deep profiles BH 10 and BH 13 are not influenced by any contamination in the laboratory. They both show that the moisture down to depths of 18 m to 22 m has a residence time of at most 3 tritium half-lives or 36 years. At a mean moisture content of 1.6 wt %, the equivalent water column to a depth of 22 m is 500 mm. Over a 36 year period this represents a recharge of some 4 % of annual rainfall, or some 13 mm a⁻¹ for an average annual rainfall of 337 mm. Estimates of maximum recharge on the basis of chloride balance and assuming 1 mg L⁻¹ of chloride in rainfall, lie in the range 1.8 - 5.0 mm a⁻¹. A more thorough analysis of the data may be required in order to understand this apparent discrepancy.

9. COMPARISONS WITH PREVIOUS STUDIES

As mentioned in the Introduction (Section 1.) several depth profiles were obtained at the Harefields site during an earlier project in 1977, down to depths of around 20 metres. The results for grain size analysis, moisture contents, tritium and $\delta^{18}\text{O}$ for BH 7, drilled during the 1977 study, are shown in Fig. 15.



Mean grain size, standard deviation, skewness, kurtosis, moisture content, 3H and ^{18}O concentrations for BH7. Closed arrow indicates bomb 3H cut off. Open arrow marks estimated level of penetration of the 1973-74 heavy rainfall period from ^{18}O increase.

FIGURE 15

Whereas the moisture contents down to some 16 m is in the range of values observed in the 1997/98 profiles, there is a sharp increase up to nearly 7 % at the bottom of BH 7. This coincides with a sharp decrease in grain size, standard deviation, and excursions in skewness and kurtosis. No such rises in moisture content were observed in the 1997/98 profiles, which probably do not show such dramatic changes in grain size distribution. However, grain size information for BH 10-15 may elucidate some of the observed changes with depth, as well as position, of the different boreholes.

In 1977 tritium concentrations reached a high of some 22 TU at a depth of 7 m in BH 7, declining to about 2 TU near the bottom at 18 m. At the time, this was interpreted as the extent of the bomb tritium "peak". The leading edge of the exceptional series of rainfall years (1974-1977; Fig 5) was interpreted as the point of inflection in the $\delta^{18}\text{O}$ profile at some 16m.

During 1997/98 values of 5 to 8 TU were found at the maximum depths reached in BH 10 and 13. This suggests that the remains of the bomb peak have moved to greater depths. Rain for the 1995/96 period in which tritium concentrations in excess of some 5 TU were measured, may have penetrated to various depths in the profiles.

The minimum in $\delta^{18}\text{O}$ seen in various profiles obtained in the present project at depths around 14 m to 16 m, may possibly be correlated with the minimum around 5 m to 7 m in the 1977 profiles.

The large discrepancy in the recharge assessments based on the depth of post-bomb tritium levels on the one hand and minimum values of Cl^- concentration on the other, could possibly be explained by repeated invasions of rainfall through preferential flow to depths of 20 m or more, followed by periods of evapotranspiration, which build up the chloride concentration but cause losses of tritium.

10. CONCLUSIONS

1. Given the home-made augering equipment used, 22 m is the maximum depth to which it is reasonably possible to drill and sample reliably.

2. The direct equilibration method for measuring oxygen-18 in the moisture in soil/sand samples continues to produce good reproducibility. The choice of CO_2 for the equilibration is important in that the $\delta^{18}\text{O}$ value of the gas should be not too different from that of the moisture. Correction of values for different moisture contents seems to be possible.

3. The direct soil water reduction method using zinc metal for deuterium measurements gives excellent reproducibility.

4. Both the 1997 and 1998 profiles show remarkable differences in both absolute values and depth dependence of the parameters measured (moisture content, electrical conductivity, chloride concentration, oxygen-18, and tritium) even over a few tens of metres separation at both sites selected.

5. Conclusions regarding depth dependence and evaporative enrichment near the surface based on oxygen-18 values alone, have been fully corroborated by deuterium measurements.

6. Measurable, post-bomb tritium values are observed to the maximum depth in both of the deepest profiles. A careful laboratory assessment of a blank subjected to the same procedure as the samples, gives confidence in the results obtained. Using tritium as an indicator of depth of penetration with time, only a lower limit to the inferred recharge can be set.

7. There is a wide discrepancy between minimum recharge rates estimated on the basis of the tritium profiles and the lowest values of $[Cl^-]$ observed. As yet, this discrepancy remains unresolved. A possible explanation would be repeated penetration of individual infiltration events to the depths attained by augering, and subsequent substantial water losses due to evapotranspiration.

8. The mechanism proposed under 7. might partially explain also the lateral and vertical variability of the other parameters measured. Grain size analysis is seen as important to remove the matrix factor from these variations.

9. The chloride concentrations for the three saturated zone water samples obtained in the area are nearly one order of magnitude below even the lowest values measured in the profiles. Therefore, it is concluded that either/or:

- a) there should be deeper sections of the unsaturated zone in which the chloride concentrations are much lower than observed down to some 20 m
- b) such infiltration as reaches the saturated zone, thereby constituting recharge, does so through bypass flow. This phenomenon was recognised early on by Smith et al. (1970) and has more recently been demonstrated by Beekman et al. (1996) in the unsaturated zone of a similar environment.

10. This Kalahari area can be regarded as a benchmark site on account of its relative accessibility, the wealth of information on the unsaturated zone obtained in this and earlier studies as well as the availability of detailed and ongoing local rainfall data.

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