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AUSTRALIAN ATOMIC ENERGY COMMISSION
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ABSTRACT

Precipitation samples have been taken over a period of one calendar year to determine the variables in environmental tritium during single storm events. Where possible, comment is made on the hydrological implications of these variations.

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TRITIUM; GROUND WATER; RAIN; RAIN WATER; HYDROLOGY; AQUIFERS; ISOTOPE RATIO; NEW SOUTH WALES; RADIATION MONITORING

CONTENTS

1. INTRODUCTION

The study of groundwater dynamics using environmental tritium techniques relates the tritium levels in the sampled groundwater to an assumed input function. The input function is normally deduced from the variability in the tritium levels in the monthly rainfall samples from the 16 stations monitored, since 1970, by officers of the Australian Atomic Energy Commission (AAEC). It is assumed that the weighted mean of tritium levels in the rain falling over the recharge area is directly related to that entering the aquifer system. The validity of this assumption can be seriously challenged when applied to the arid and semi-arid areas where the potential evaporation rate can exceed mean annual rainfall by an order of magnitude. Under these conditions, the probability of an element of rainfall entering the saturated zone is small and may not be isotopically representative.

Because of the importance of the input function to tritium dating in general and to the study of recharge processes in particular, a very detailed investigation was made of the variation of tritium in rainfall over Northbridge, approximately 4 km north-east of Sydney, New South Wales. Samples were collected at two hourly intervals over a 12-month period from October 1976. A subsidiary aim of the work included an investigation of the significance of monthly average tritium levels reported by most laboratories including the International Atomic Energy Agency [IAEA 1975] and the AAEC [Calf *et al.* 1974,1977]. The results were interpreted in terms of a simplified storm model.

2. EXPERIMENTAL EQUIPMENT AND METHOD

The rainwater collection system is illustrated in Figure 1. Its main feature is a central dispensing table which rotates 30° at preset intervals once the circuitry has been activated by the pluviometer microswitch; the intervals can be varied from 1 to 10^6 s. Other features and operation are described in Appendix A. A two hourly sampling period was chosen; this was short compared with the duration of many rain events in the Sydney area, but usually long enough to allow collection of the minimum volume of 600 mℓ required for an assay [Calf 1976] from the 12 m^2 collection area. The collection area was a section of roof from which the drainage water had been diverted to the collection apparatus; it was sited approximately 10 m from the final point of collection. The apparatus was actuated manually at the start of each

24 h period, thus ensuring that the sampling time was accurately recorded.

To enable intercomparison with the other sampling stations located in the Sydney area and in other parts of Australia, monthly rainwater samples were also collected at Northbridge by the standard technique used by the AAEC Hydrology Group [Calf *et al.* 1974, 1977]. These samples were assayed by liquid scintillation counting after electrolytic enrichment. All results were recorded in tritium units (TU); the TU is defined as one atom of ^{3}H per 10^{18} atoms of ^{1}H and is equivalent to 7.2 disintegrations per minute (dpm) per litre or 3.24 pCi (118 mBq) per litre. All tritium results in this report have a standard deviation of either 8.5 per cent or 0.4 TU, whichever is the greater.

3. RESULTS

The results of the survey carried out between 20 October 1976 and 21 October 1977 are shown in Figures 2(a)-2(i). Where the rain events extended over more than one collection period, they have been labelled A...Z, AA...LL for easy reference. The mean rainfall precipitation during the 9 a.m. to 9 a.m. collection period, obtained from the Commonwealth Bureau of Meteorology, is distinguished by bars through the histograms.

The principal observation was a regular decrease in tritium levels with time during rain events lasting more than 12 h. These include the periods AB (single event), EE and JJ (Figure 2(a)). There was one exception to the pattern; in event Y, the levels of tritium show a general rise over the 24 h period. In events Q, R, W and BB, the tritium concentrations are steady within experimental error. Rain events over intervals shorter than 12 h cannot be usefully discussed since this is insufficient time in which to establish a trend.

4. DISCUSSION

4.1 Possible Influence of the AAEC Reactor HIFAR

It is necessary to consider the possible effect of small unavoidable releases of tritium from the heavy water-moderated materials testing reactor HIFAR. Since the levels of detection of tritium in rainfall are extremely low, even small releases might affect the results. This is, however, considered unlikely for the following reasons.

(a) The negligible effect of reactor releases can be found by relating the monthly average tritium results from Northbridge to those obtained at Brisbane in Queensland, Ryde and Campbelltown in New South Wales, Melbourne in Victoria and Hobart in Tasmania (Figures 5(a) & 5(b)). These results vary substantially because of effects of latitude, season, and proximity to the sea. From the histograms it can be seen that the inclusion of extra factors such as reactor proximity to explain the effects at Northbridge is unnecessary.

(b) It can be shown quite convincingly (Figure 6) that the effect of HIFAR on the tritium levels in rainfall at Northbridge is negligible by observing the excellent correlation between the tritium and D/H levels [Hartley 1978]. For example, in group AB the correlation coefficient is 93.2 per cent despite the fact that both the tritium and the D/H levels vary over a substantial range, i.e. 3.7 to 21.9 TU and -140.5/-2.9 per mille respectively. The isotopic ratios are reported in the conventional δ units defined with respect to the Standard Mean Ocean Water (SMOW)

$$\delta D = \left(\frac{R\ SAMPLE}{R\ SMOW} - 1 \right) \times 1000 \text{ per mille}$$

where R is the D/H atomic ratio [Payne 1968]. The lower the tritium level, the more enriched is the deuterium in the rainfall. If HIFAR were responsible for this, a positive correlation would be expected. Further, the impact of small releases from HIFAR on the D/H levels in rainfall would be insignificant because the normal environmental levels are of the order of 150 $\mu g\ g^{-1}$ (ppm) in the Sydney region, and the mass of water falling during an extended rain event is very high.

4.2 Explanation of Observations in Terms of a Simplified Storm Model

Precipitation is induced by a number of effects: instability; convergence; orographic uplift; and frontal uplift. Sydney, which is influenced principally by the south east trade wind system, probably experiences all these effects to some degree. A schematic representation of a cloud model developed by Chisholm [1970] is shown in Figure 5(a). It comprises an unmixed adiabatic core surrounded by an extensive boundary zone. The cloud top is frequently some kilometres above the ground. Tritium content and the tritium mixing ratio vary substantially over these elevations. Ehhalt [1974] studied sixty-two profiles of storms occurring in North America between 1966 and 1973. The example from

Florida is shown in Figure 5(b). Florida was chosen because it had a similar latitude and climatic zone to that of Sydney. Almost all the profiles showed that tritium content rose dramatically with increased altitude.

During the progress of a storm, turbulence frequently develops; this gives rise to a relatively high initial tritium level in rainwater owing to a contribution from vapour of high specific activity in the cloud layer. Over prolonged events, rainfall is formed increasingly from moisture transported into the storm area by the prevailing winds. As the vapour is derived principally from oceanic areas, the tritium level will only be of the order of a few TUs. The general decrease in tritium levels with time can be attributed to a dilution effect of this oceanic vapour. The discussion of the tritium results has been purposely made as general as possible. It has been kept independent of particular storm models since, in the Sydney region, orographic frontal and convection mechanisms occur from time to time. The discussion is valid for all cases in which the rain event is preceded by an uplift of cloud and the duration of the event is significantly extended by the input of oceanic vapour.

5. CONCLUSIONS

This study has been of use in developing an understanding of the nature of the variability of tritium content in precipitation in a temperate coastal region over a time scale that is much shorter than the normal monthly sampling period. The understanding of variation in the tritium levels during a discrete, extended rain event is, therefore, important when applying tritium measurements to studies of phenomena having characteristic times between days and a few months. These include:

(a) the turnover times of water in small lakes and reservoirs; and

(b) aspects of the transport of water through the unsaturated zone.

More importantly, this study has contributed to a clarification of those processes resulting in the isotopic biasing of water entering the saturated zone of an aquifer. The longer the rain event, the higher is the probability of recharge and the lower is the average tritium level. The understanding of these effects is important if tritium measurements are to be used for the absolute dating of groundwater. Further extension of this work should include:

(i) correlation of tritium data with D/H ratios; and

(ii) extension of the observations to other sampling stations, preferably located on a systematic grid.

6. ACKNOWLEDGEMENTS

The author is indebted to Mr G. Watt for his design of the electronic circuit used in the collection apparatus, and to Dr P.L. Airey for his assistance in the interpretation of some of the hydrological aspects.

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NOTES

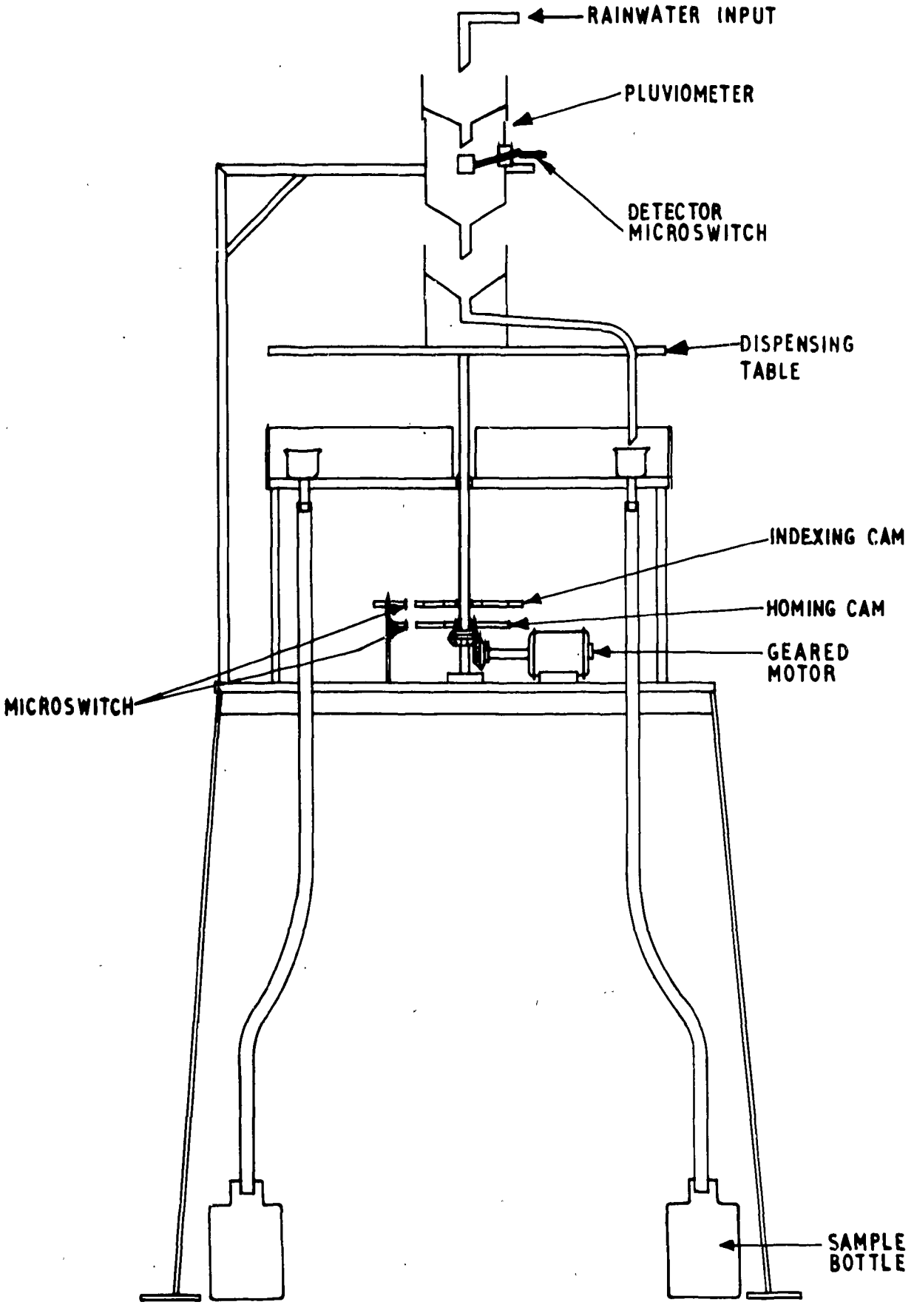

FIGURE 1 AUTOMATIC RAINWATER COLLECTOR

FIGURES 2(a) — 2(i)

VARIATION WITH TIME OF THE TRITIUM LEVELS (TU) IN RAINWATER SAMPLES TAKEN AT TWO HOURLY INTERVALS BETWEEN 16 OCTOBER 1976 AND 21 OCTOBER 1977. THE HORIZONTAL BARS ARE THE DAILY RAINFALL LEVELS (9 a.m. TO 9 a.m.) RECORDED AT THE SYDNEY REGIONAL OFFICE BY THE BUREAU OF METEOROLOGY. THE INTERVALS A, B, C, D-----NN FACILITATE REFERENCE FROM THE TEXT TO PARTICULAR RAIN EVENTS

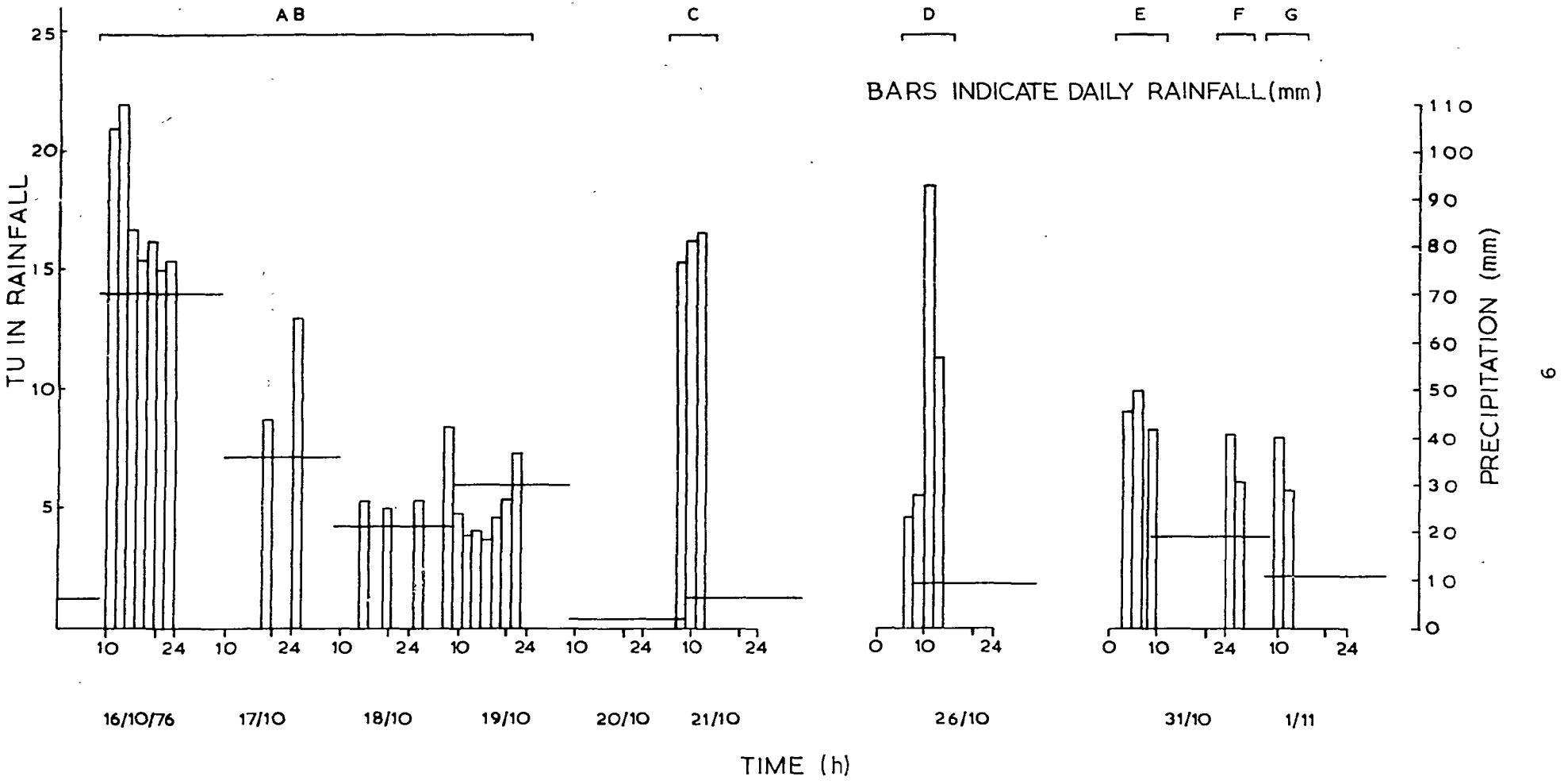
AB
C
D
E
F
G
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
0
110
100
90
80
70
60
50
40
30
20
10
0
10
24
16/10/76
17/10
18/10
19/10
20/10
21/10
26/10
31/10
1/11
TIME (h)

FIGURE 2(a)

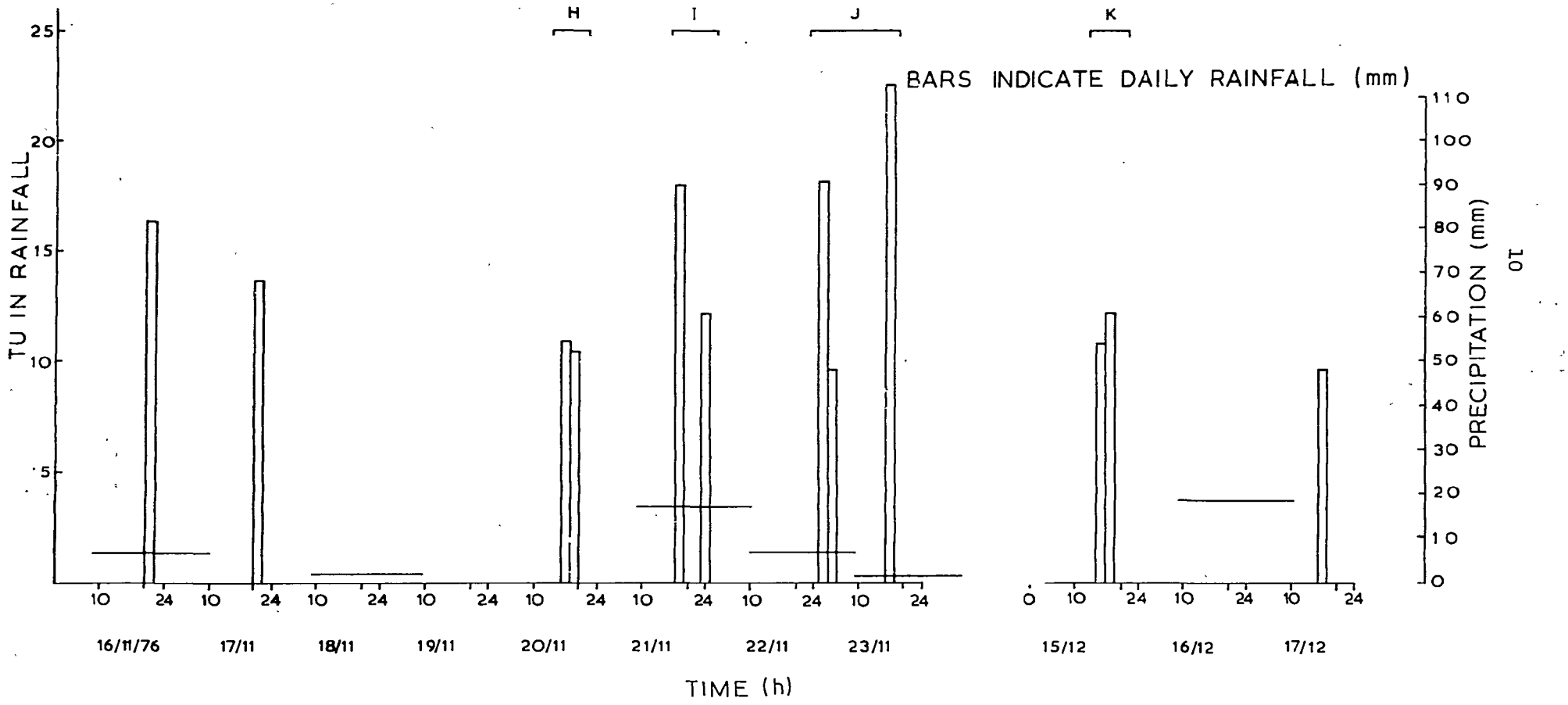
H
I
J
K
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
0
110
100
90
80
70
60
50
40
30
20
10
0
10
24
16/11/76
17/11
18/11
19/11
20/11
21/11
22/11
23/11
15/12
16/12
17/12
TIME (h)

FIGURE 2(b)

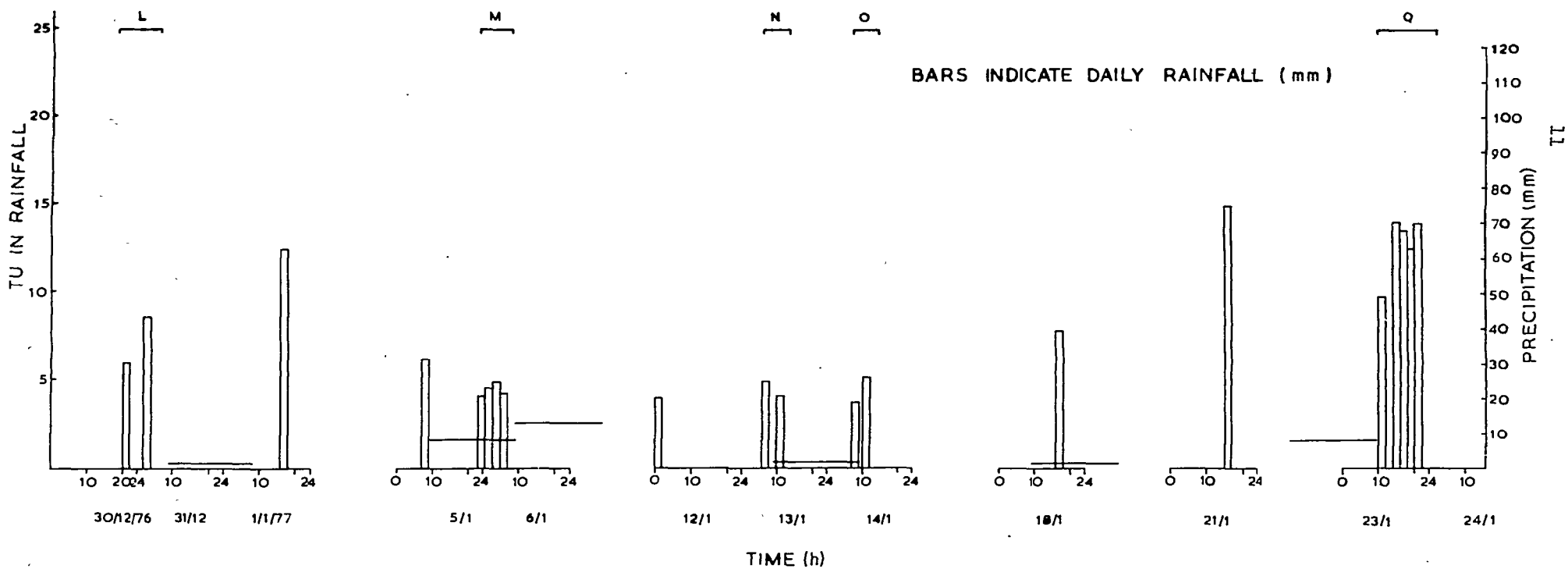
L
M
N
O
Q
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
120
110
100
90
80
70
60
50
40
30
20
10
30/12/76
31/12
1/1/77
5/1
6/1
12/1
13/1
14/1
18/1
21/1
23/1
24/1
TIME (h)

FIGURE 2(c)

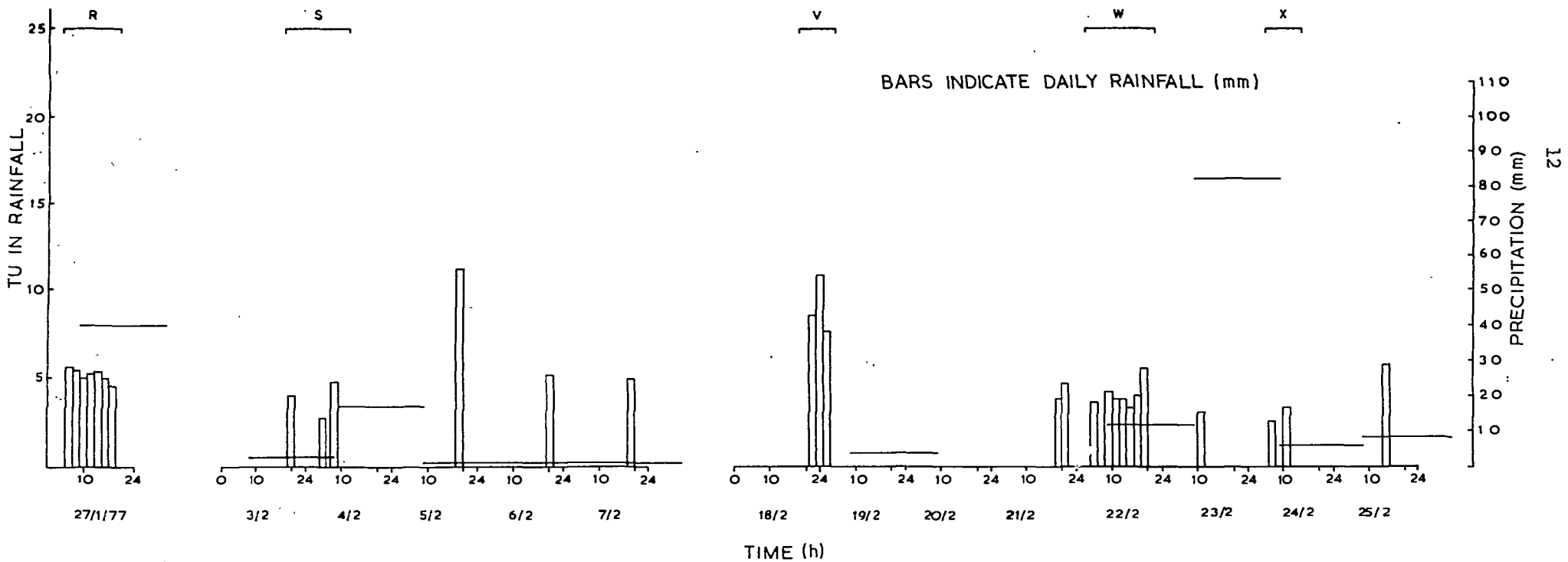
R
S
V
W
X
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
110
100
90
80
70
60
50
40
30
20
10
0 10 24 10 24 10 24 10 24 10 24 10 24
27/1/77
3/2
4/2
5/2
6/2
7/2
18/2
19/2
20/2
21/2
22/2
23/2
24/2
25/2
TIME (h)

FIGURE 2(d)

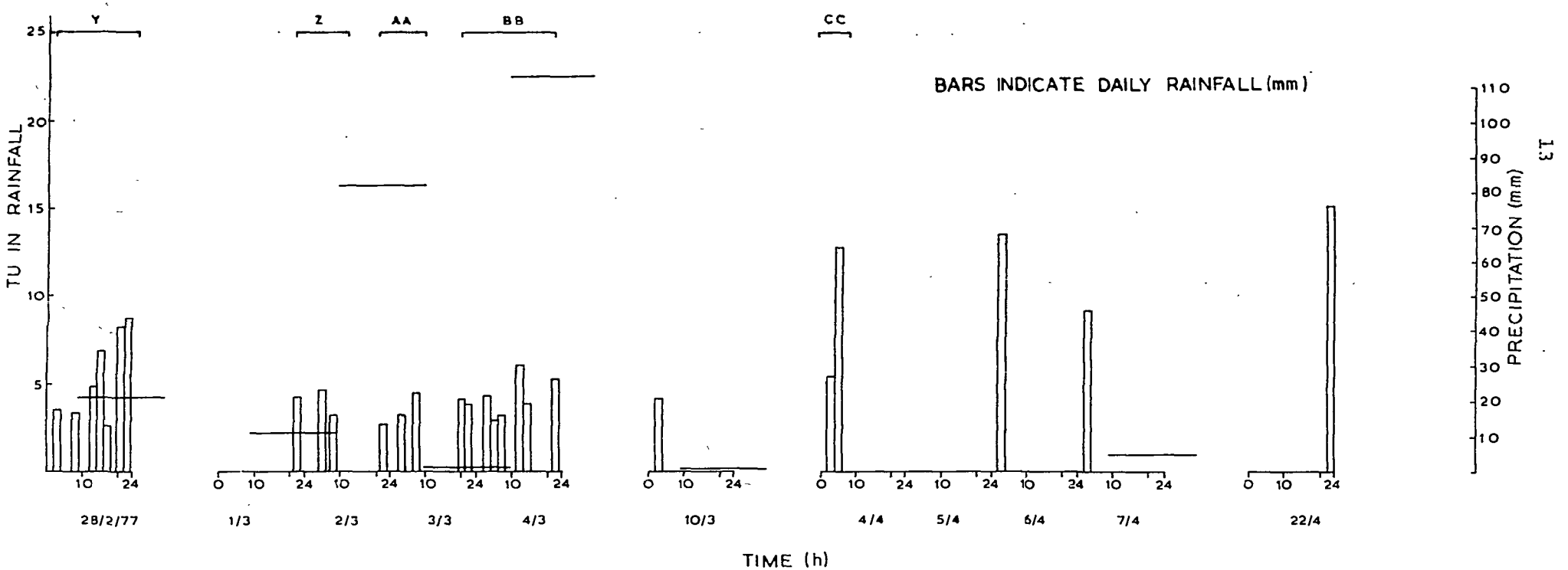
Y
Z
AA
BB
CC
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
110
100
90
80
70
60
50
40
30
20
10
0 10 24 10 24 10 24 10 24
28/2/77
1/3
2/3
3/3
4/3
10/3
4/4
5/4
6/4
7/4
22/4
TIME (h)

FIGURE 2(e)

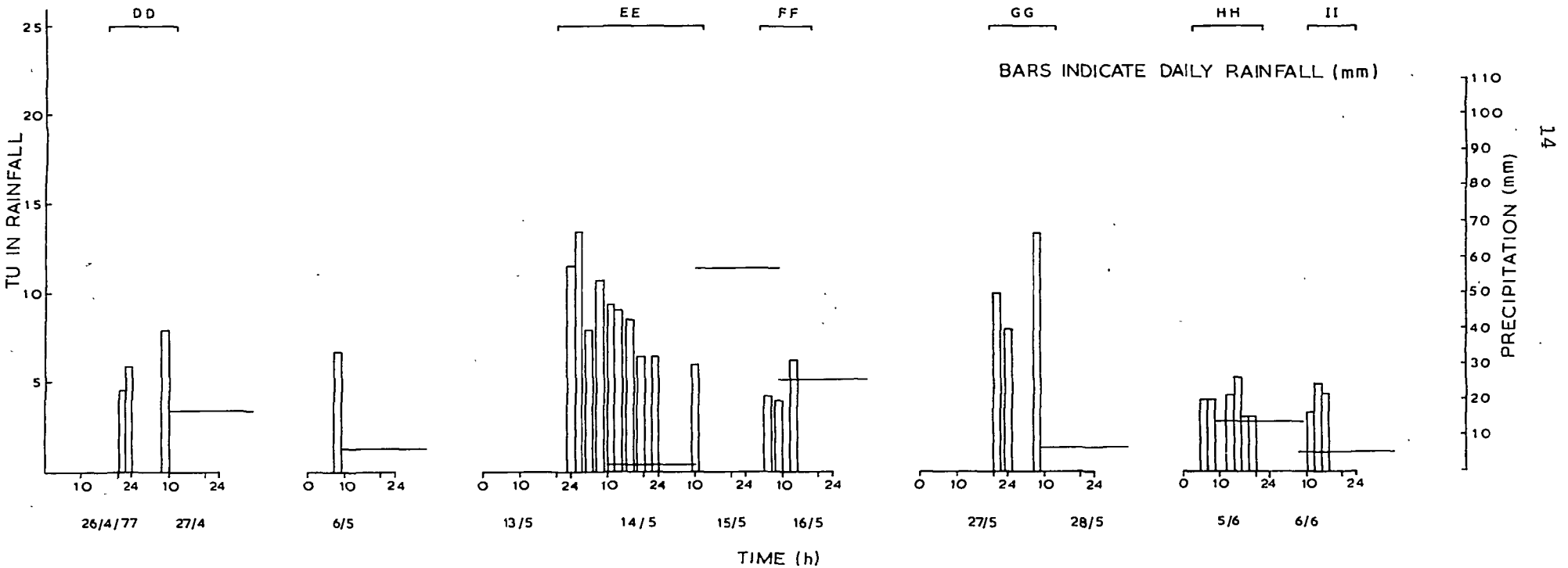
DD
EE
FF
GG
HH
II
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
110
100
90
80
70
60
50
40
30
20
10
0 10 24 10 24 10 24 10 24 10 24 10 24 10 24 10 24 10 24 10 24
26/4/77
27/4
6/5
13/5
14/5
15/5
16/5
27/5
28/5
5/6
6/6
TIME (h)

FIGURE 2(f)

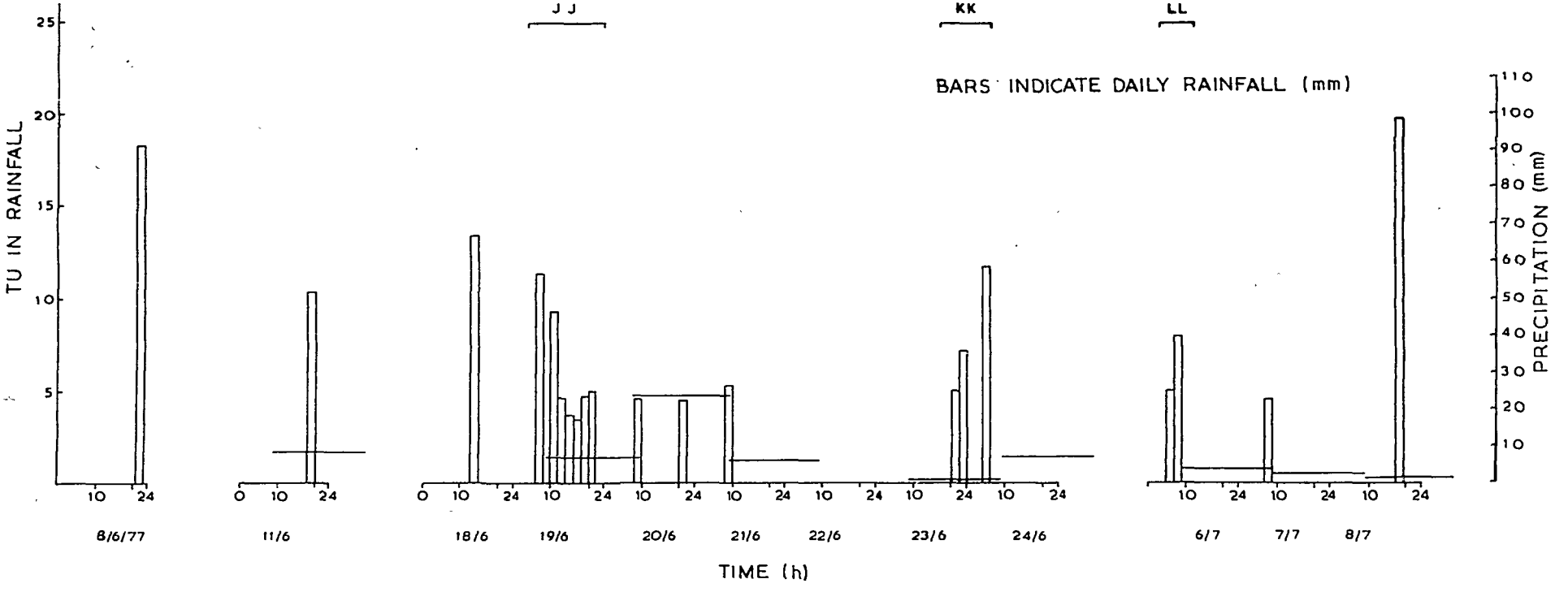
J J
KK
LL
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
110
100
90
80
70
60
50
40
30
20
10
10 24
0 10 24
8/6/77
11/6
18/6
19/6
20/6
21/6
22/6
23/6
24/6
6/7
7/7
8/7
TIME (h)

FIGURE 2(g)

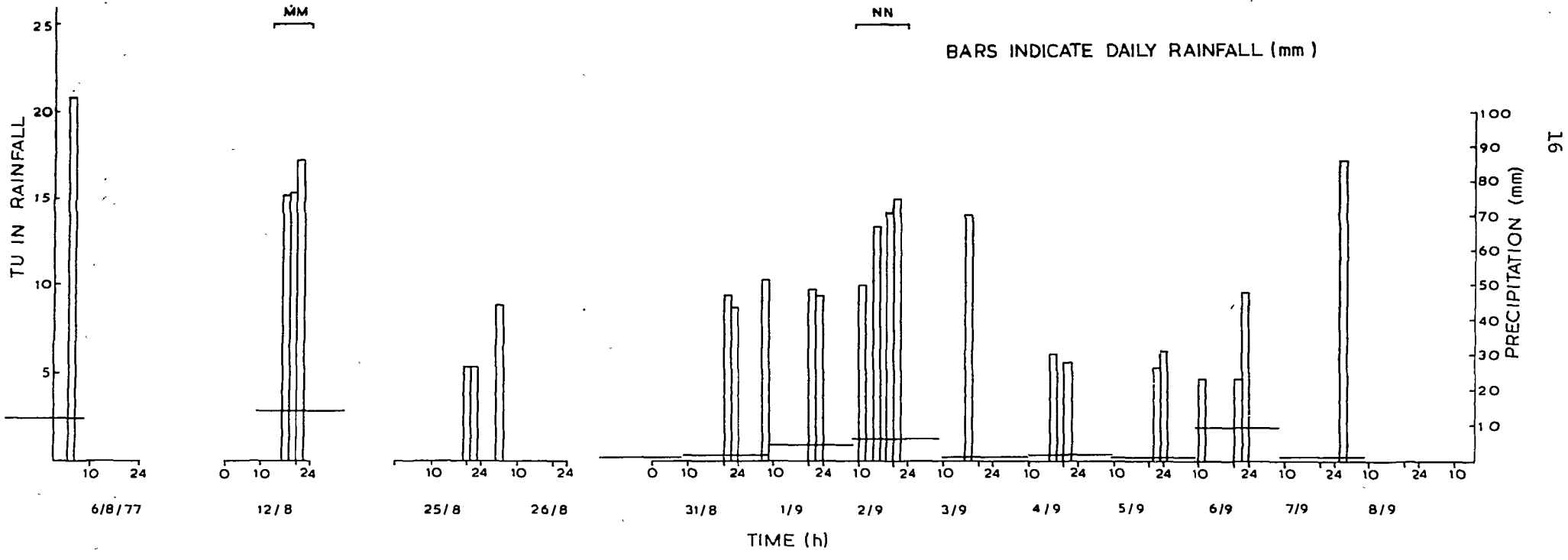
MM
NN
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
TIME (h)
6/8/77
12/8
25/8
26/8
31/8
1/9
2/9
3/9
4/9
5/9
6/9
7/9
8/9

FIGURE 2(h)

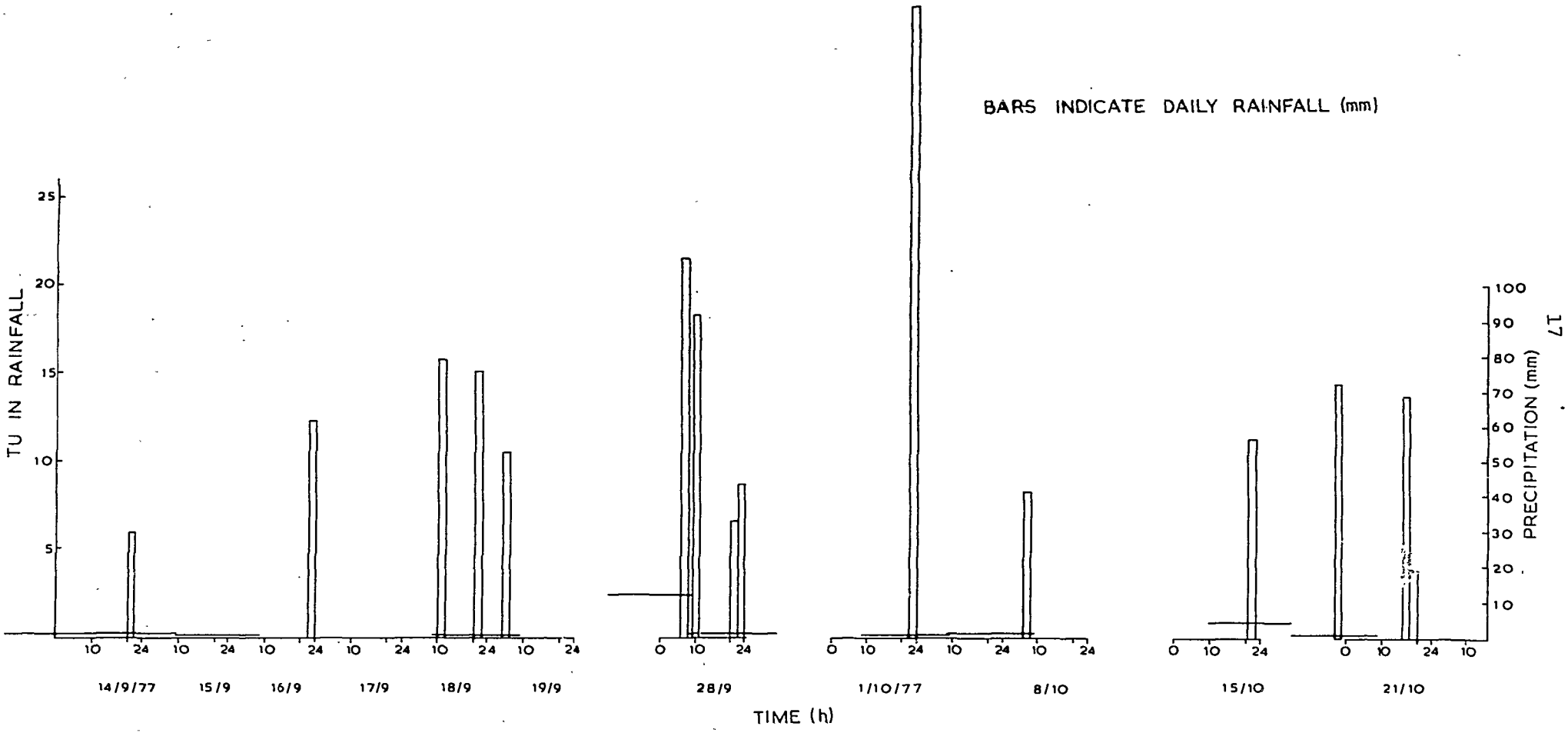
BARS INDICATE DAILY RAINFALL (mm)
TU IN RAINFALL
PRECIPITATION (mm)
25
20
15
10
5
100
90
80
70
60
50
40
30
20
10
0 10 24 10 24 10 24 10 24 10 24 10 24
14/9/77
15/9
16/9
17/9
18/9
19/9
28/9
1/10/77
8/10
15/10
21/10
TIME (h)

FIGURE 2(i)

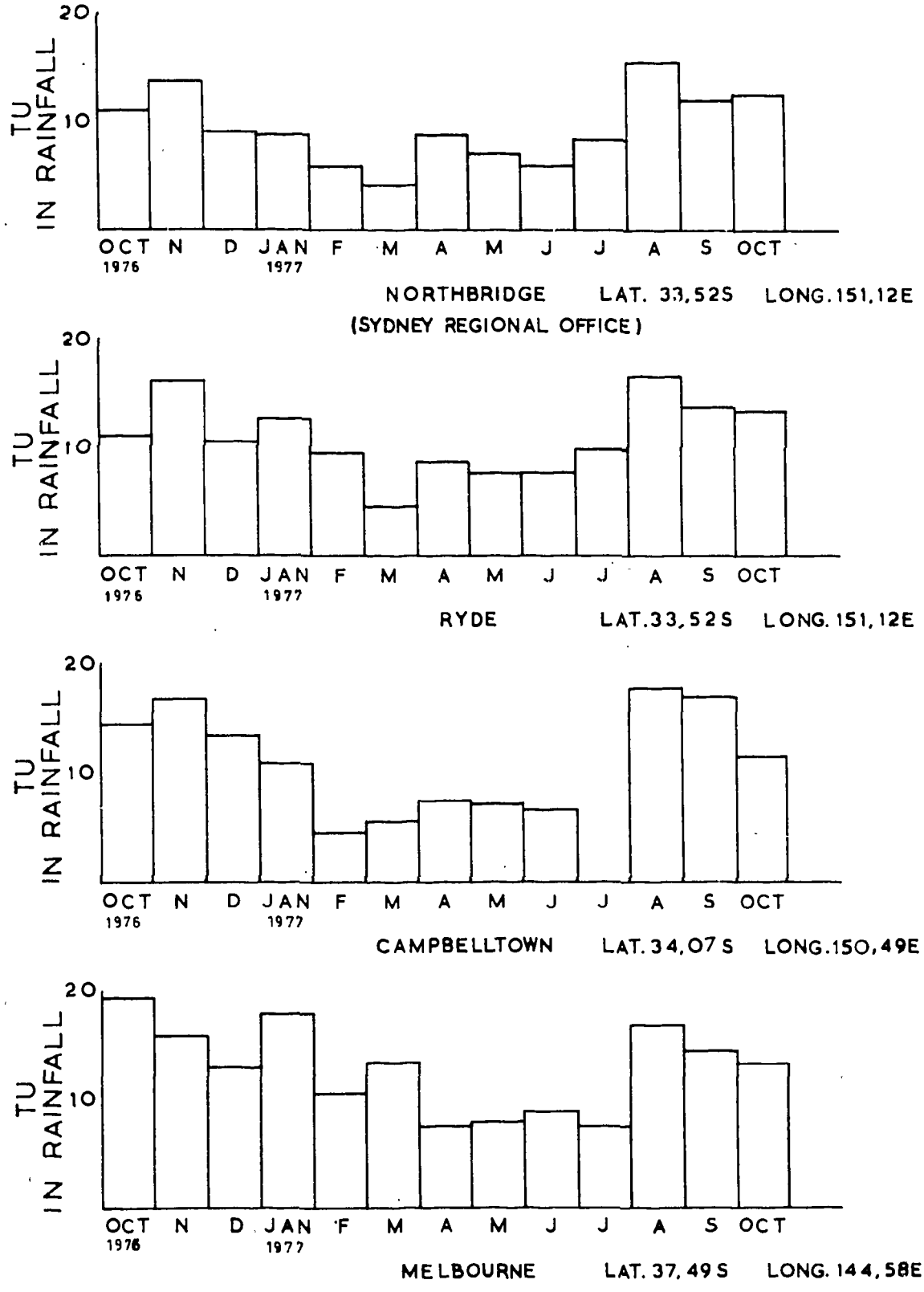

FIGURE 3 MONTHLY TRITIUM LEVELS IN RAINFALL

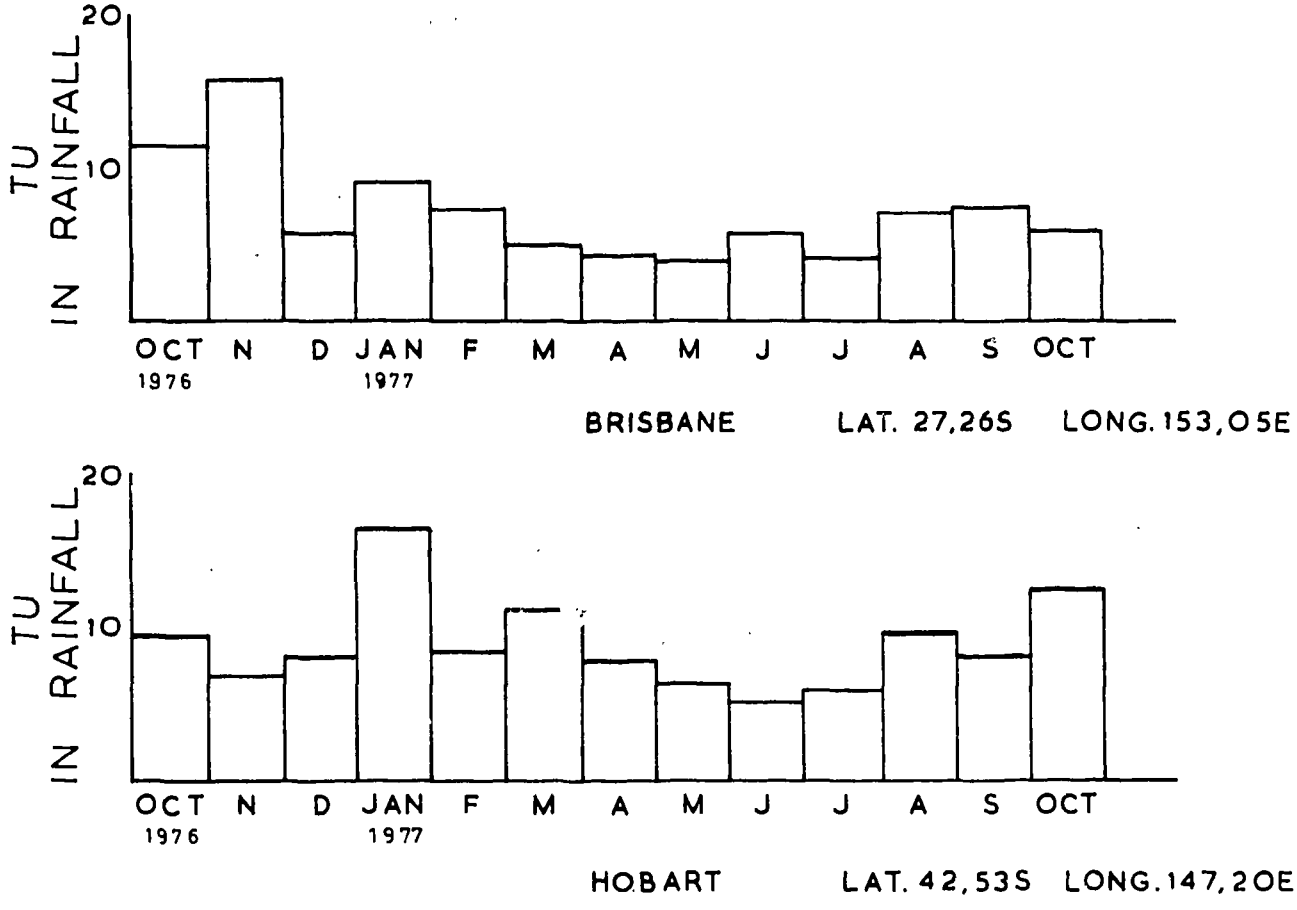

FIGURE 3 (Continued)

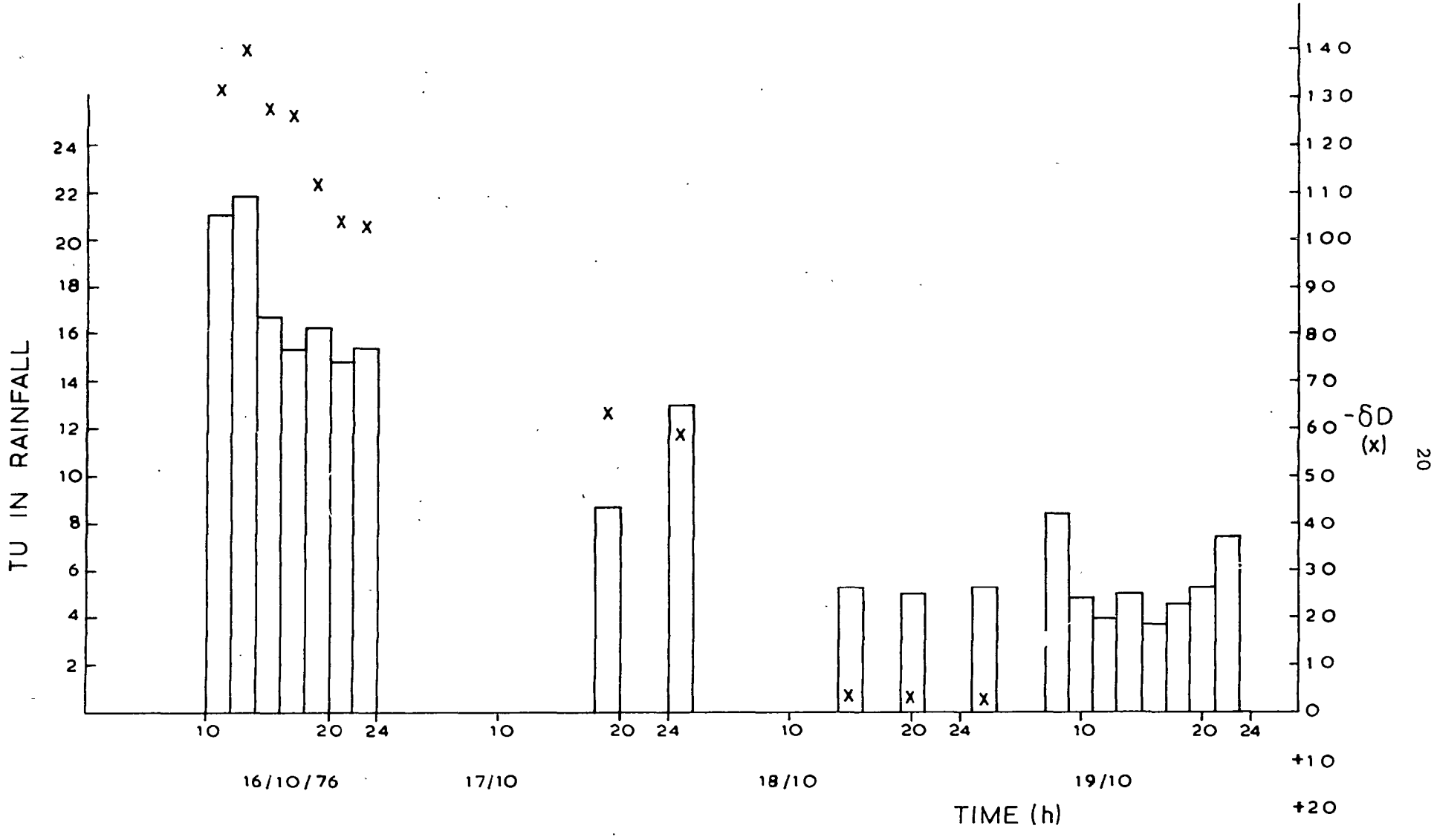

FIGURE 4 VARIATION WITH TIME OF THE TRITIUM LEVELS (TU) AND D/H RATIOS (X PER MILLE) FOR THE PERIOD 16 OCTOBER TO 19 OCTOBER 1976 (EVENT AB, FIGURE 2(a))

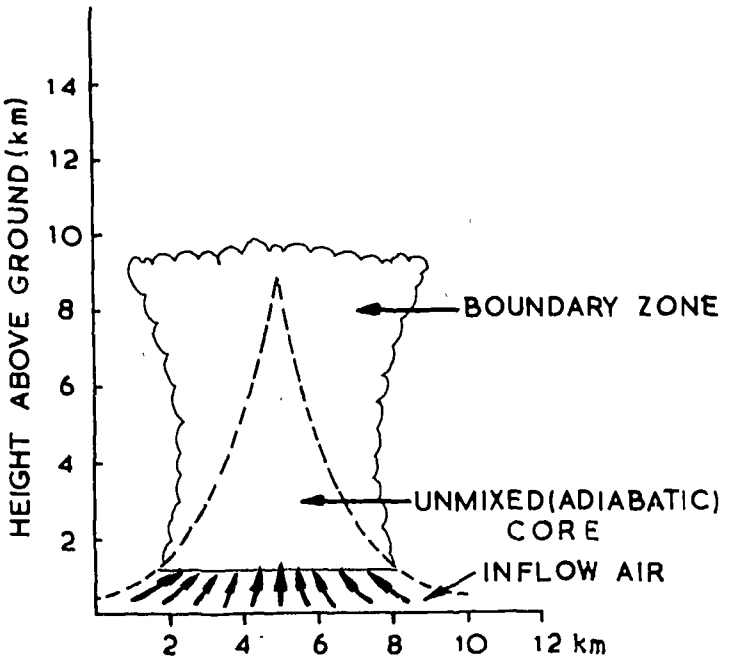

FIGURE 5(a) SCHEMATIC REPRESENTATION OF THE CLOUD MODEL [AFTER CHISHOLM 1970]

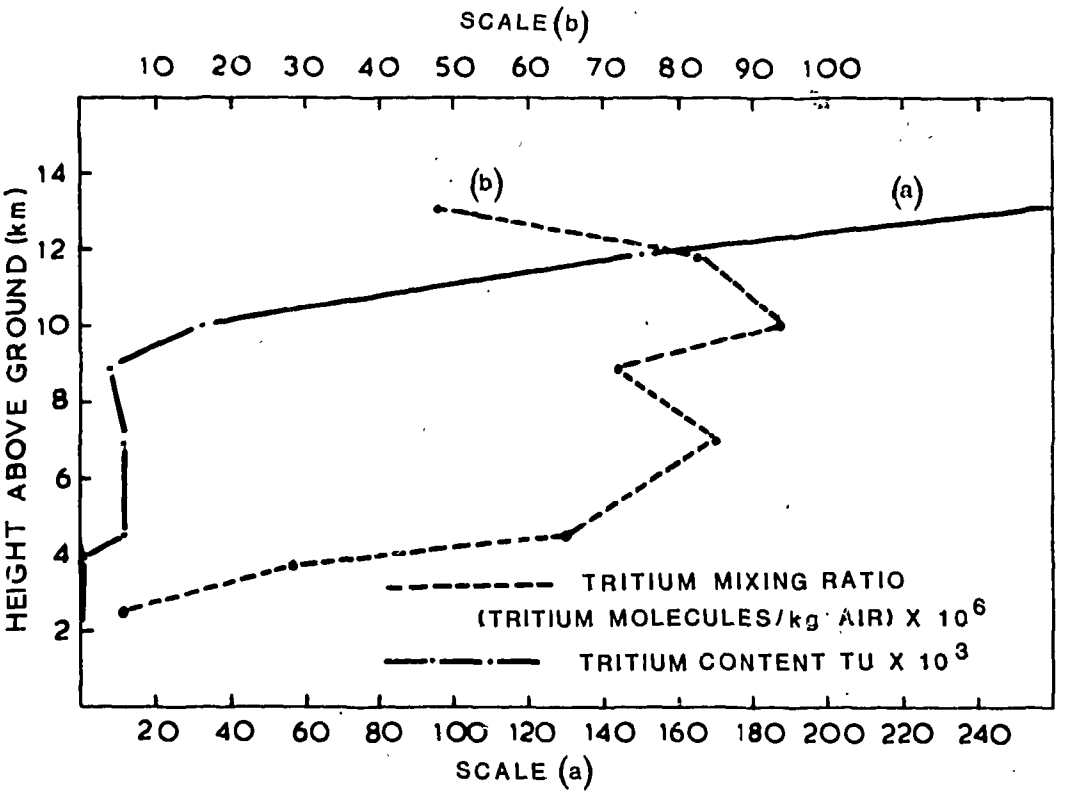

FIGURE 5(b) VARIATION WITH HEIGHT OF THE TRITIUM MIXING RATIO AND THE TRITIUM LEVEL OFFSHORE FROM THE EVERGLADES, FLORIDA, 29 FEBRUARY 1972 [AFTER EHHALT 1974]

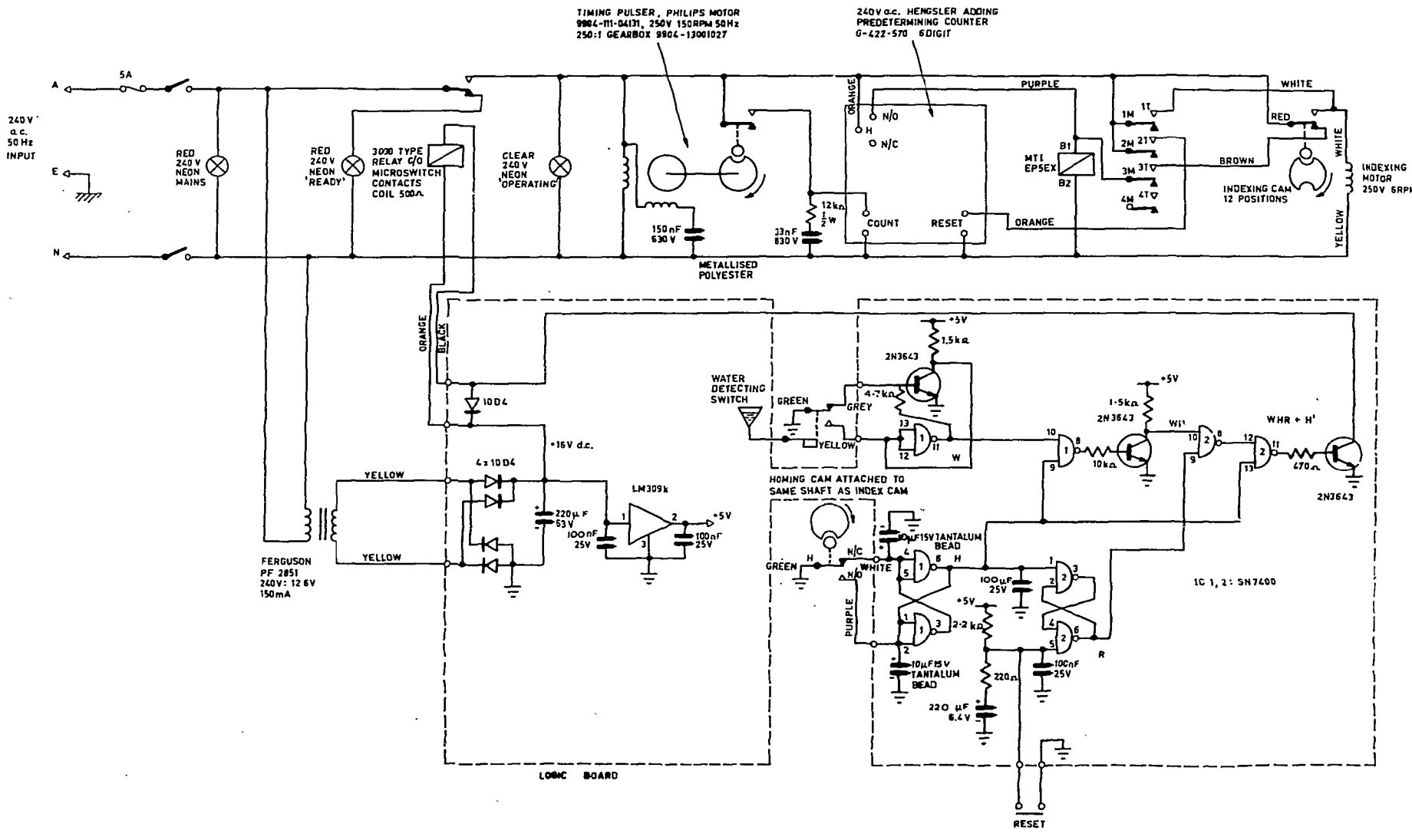

FIGURE 6 AAEC TYPE 533 CYCLE TIMER

APPENDIX A

AUTOMATIC RAINWATER COLLECTOR

A.1 RAINWATER COLLECTOR MECHANICS AND ELECTRONICS

The rainwater collector drive circuitry (Figure 1) consists of a motor and reduction gearbox to rotate the sample holders, an electro-mechanical timer to determine the collection time of each sample, cams which delineate the rest position and the 12 collection positions of the sample holder, a water detection switch, and logic circuitry to initiate, sequence and complete the collection of 12 samples.

A.2 OPERATION

Two cams are attached to the driveshaft of the collector: the *homing cam* has only one lobe which stops the drive motor by operating a microswitch at the rest position; the *indexing cam* has 12 lobes which stop the collector drive at 12 sampling positions.

Operation is commenced when (a) the pluviometer detector cup fills and trips the detector microswitch thus producing a signal W , (b) the sampler is in the homed position (as detected by the homing cam micro-switch) and a signal H is present, or (c) when the previous samples have been collected and the collector has been reactivated by pressing the reset push button producing a signal R . The drive motor responds to a signal which is the logical product of W, H & R, (D = WHR), and drives the collector to the first collection position.

As soon as the homing cam moves away from the rest position (which is half way between two sampling positions), the motor drive Command D is derived solely from a signal from the homing cam indicating that it is not a rest position, *i.e.* D = H'. (This ensures that if power fails midway through a sampling sequence, the correct signals are re-established on power resumption for the sequence to be continued through to the rest position. Thus in logic notation the motor drive command signal is D = W.H.R + H'. The relay which provides 240 V d.c. power to the drive motor, the 1 s impulsing motor and the 6-digit electro-mechanical preset accumulator is driven by D. During collection of samples, D remains at 1 , the relay is continuously energised and the sequencing through the 12 positions is entirely electro-mechanical.

When the transport reaches the first collection position, the lobe actuates the indexing microswitch which removes the a.c. supply from the

indexing motor. During this time, the accumulator counts up to the preset figure in 1 s increments until the preset figure is reached (1 s to 10^6(-1) s). At that point, the indexing motor is re-energised and the counter is reset to zero. The process repeats when the indexing microswitch connects with the reset indexing lobe on the cam. After the twelfth sample has been completed, the indexing motor is re-energised until it drives the collector to the homed position, at which point the drive signal D becomes zero (H' = 0, R = 0, W may be 0 or 1, D = H W R + H' = 0) and the relay driven by D drops away to prevent further operation until the reset push button is operated and the rainwater pluviometer cup is sufficiently full to activate the water detection switch. This prevents a second set of water samples being taken while the first set remains uncollected.