



AUSTRALIAN ATOMIC ENERGY COMMISSION  
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LUCAS HEIGHTS

THE USE OF ENVIRONMENTAL URANIUM ISOTOPES IN THE  
STUDY OF THE HYDROLOGY OF THE BURDEKIN DELTA

by

B.L. CAMPBELL

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ABSTRACT

Analyses of bore water samples from the Burdekin Delta, Queensland, show considerable variation in both the uranium concentration and the  $^{234}\text{U}/^{238}\text{U}$  activity ratio. In many cases, the uranium concentration was closely correlated with the bicarbonate concentration, but not for waters with a very low uranium concentration. Mechanisms by which uranium can be removed from solution are discussed. They provide a basis for explaining the low uranium concentrations and, in some areas, the apparent lack of response of the aquifer to the input of uranium by recharge.

The  $^{234}\text{U}/^{238}\text{U}$  activity ratio is interpreted as a reflection of the extent of local interaction of infiltrating ground water with soil constituents. Measurement of the isotope activity ratio has confirmed the location of an area with significant recharge from the river into the aquifer system. The isotopic and supporting chemical data illustrate the complexity of the aquifer system.

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AQUIFERS; BOREHOLES; ENVIRONMENT; GROUND WATER; HYDROLOGY; ISOTOPE RATIO; QUEENSLAND; RADIONUCLIDE MIGRATION; URANIUM 234; URANIUM 238;

## CONTENTS

	<u>Page</u>
1. INTRODUCTION	1
2. HYDROLOGY OF THE INVESTIGATION AREA	2
3. RESULTS AND DISCUSSION	2
3.1 Uranium Concentration	2
3.1.1 $E_h$ -pH relationships	3
3.1.2 Complex ion formation effects with uranium	4
3.2 $^{234}\text{U}/^{238}\text{U}$ Activity Ratio	6
4. SUMMARY OF RESULTS	8
5. ACKNOWLEDGEMENT	8
6. REFERENCES	8

Table 1 Chemical Analyses of Burdekin Delta Water Samples

Table 2 Comparison of Correlation Coefficients of Uranium with  
Constituents of Bore Waters

Figure 1 Location of bores sampled in the Burdekin Delta. Upper values are the uranium concentrations ( $\mu\text{g } \ell^{-1}$ ) and lower values, where available, are the  $^{234}\text{U}/^{238}\text{U}$  activity ratios. Dashed lines represent surface recharge facilities.

Figure 2 Drill logs of selected sample bores (see Figure 1 for location). Depths are in metres from the surface.

Figure 3 Aqueous equilibrium diagram of the U-O<sub>2</sub>-H<sub>2</sub>O-CO<sub>2</sub> system at 25°C and 101 kPa with  $\Sigma\text{CO}_2 = 440 \text{ mg } \ell^{-1}$ . The total ionic activity of dissolved uranium species in equilibrium with solid phases is  $10^{-6}$ .

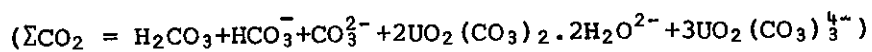


Figure 4 Relation between uranium concentration and bicarbonate ion concentration. The line of best fit is drawn with a slope of 1.88.

Figure 5 Burdekin Delta water level contours, June 1974.



## 1. INTRODUCTION

The intensive and expanding agricultural use of the Burdekin Delta plains, which are situated 80 km south of Townsville, Queensland, poses the continual problem of budgeting water input against consumption, development proposals and the maintenance of water quality. In this area, summer rainfall predominates but its variability puts agriculture at risk and irrigation must be used to ensure crop success [Archibald 1970]. For example, in the Lower Burdekin district, 99 per cent of the sugar cane produced is dependent on irrigation for the majority of the growing season [Ham 1972]. The excess water required is obtained by pumping from the aquifer system that underlies the Delta.

Storage in the aquifer is affected by high evaporation and a considerable run off of water, so the effective rainfall is much less than the actual rainfall. In the mid 1960s, the steadily increasing demands of an expanding agriculture led to a situation in which exploitation exceeded recharge; this had the effect of lowering the water table which resulted in salt water intrusion in some areas. Studies by O'Shea [1967], MacDonald [1970] and Weeks [1975] have indicated some of the measures already taken to replenish the underground water supplies artificially by pumping from the Burdekin River part of the unregulated flow into recharge pits and channels.

There are differing views on the relative importance to the total recharge of rainfall compared with inflow from the river. Early engineering evaluations by O'Shea [1967] indicated that rainfall was the major recharge source. On the other hand, Volker & Stark [1973] used a mathematical model to show that river recharge was very likely of much greater significance than had previously been envisaged. Support for their view came from studies of the aquifer system by the Australian Atomic Energy Commission using measurements of the environmental isotopes tritium and carbon-14 [Airey et al. 1974]. Some areas were identified in which there was a significant inflow to the aquifer system from the river.

Variations in uranium concentration and its naturally occurring isotopes uranium-234 and uranium-238 have also been used to identify natural waters and their sources [Campbell 1975]. Some preliminary data on water samples from the Burdekin Delta have shown significant variations of these isotopes according to locality. Additional data for the  $^{234}\text{U}/^{238}\text{U}$  activity ratio and chemical analyses of the water samples are presented in Section 3 to show the complexity of the recharge sources throughout the delta. A

satisfactory interpretation of these data also requires an understanding of such factors as  $E_h$ -pH and complex ion formation which significantly affect the mobility of uranium in an aqueous environment.

## 2. HYDROLOGY OF THE INVESTIGATION AREA

The Burdekin Delta plains comprise an area of about 615 km<sup>2</sup> bounded to the west by Stokes Range and Kelly's Mt. and to the south by Mt. Inkerman. The coastal margins consist essentially of littoral deposits up to 5 km wide. The plains are of quite low profile being generally less than 16 m above sea level. The extensive pediment slopes of Stokes Range and Kelly's Mt. and other outcrops in the area consist of undifferentiated granite, andesite and hornfels. The bedrock is mainly Permian granite and diorite and its depth increases in a north-easterly direction away from the pediment slopes of the west and south ranges.

An examination of borehole logs and seismic data has enabled some definition of the contours of the bedrock [Hopley 1970]. Contour variations could modify the subsurface drainage, but gross water movements are of course normal to the equipotential lines.

Watkins & Wolff [1960], after studying the available data, concluded that the Delta comprised a system of aquifers of unconsolidated sands and gravels variously separated by silts, silt clays and clays. On a broad scale, however, they thought that the sediments comprised a homogeneous aquifer unit. The sands containing the groundwater consisted essentially of quartz and felspar.

There are more than 300 bore holes and water sampling points throughout the Delta from which water level contours are established at regular periods or samples are taken for chemical analysis as in the present study. The bore holes are lined with polyvinyl chloride (PVC) pipe set to sample the desired water strata.

There are also extensive recharge facilities both north and south of the Burdekin River. Of the mean annual recharge ( $210 \times 10^6 \text{ m}^3$ ),  $100 \times 10^6 \text{ m}^3$  is pumped from the river. The balance is supplied by seasonal rains as groundwater infiltration. The location of these recharge lines relative to the bore holes sampled is shown in Figure 1.

## 3. RESULTS AND DISCUSSION

### 3.1 Uranium Concentration

The distribution of uranium concentration in groundwaters throughout the Delta is shown in Figure 1; there are significant variations in concentration, ranging from less than  $0.09 \mu\text{g l}^{-1}$  to  $14 \mu\text{g l}^{-1}$ . The Burdekin

River, which will be used as a basis for comparing levels, has a uranium concentration of  $1.4 \mu\text{g l}^{-1}$ .

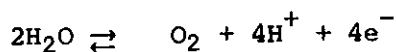
As a general conclusion, on the north side of the Burdekin River the uranium concentration falls off regularly as distance increases from either the river or recharge facilities. The average uranium concentration for all bores on the north side is  $0.14 \mu\text{g l}^{-1}$ , that is one tenth the uranium concentration in the Burdekin River. On the north side particularly, the lowest uranium concentrations were generally observed at bores fairly well removed from recharge lines, e.g. bores E9, C9, D7, L2B7, B7 and B5. Considering the level of the mean annual recharge, a uranium concentration closer to that of the Burdekin River would have been expected, but had not been observed. This lack of effect was also apparent until 1970 when according to MacDonald [1970] there was 'no evidence that either artificial or natural replenishment has improved water quality to any significant degree.....'.

A similar situation occurs on the south side of the Burdekin River, where the average for all bores with a uranium concentration less than the Burdekin River is still only  $0.38 \mu\text{g l}^{-1}$ . Even bores such as FG9 and H5, which are close to both the Burdekin River and a recharge line, have uranium concentrations less than this average. Only one bore, (A-B) S7, has a uranium concentration consistent with river water input and is further discussed in Section 3.2 in association with the isotope activity ratios.

Local variations in uranium concentration had not been expected to be so great. Factors most likely to influence the concentration of uranium in water are the redox potential, the presence of complexing agents and the uptake characteristics of the soil for various ions. It was generally expected that uranium would be retained in solution, not removed (see Section 3.1.2).

### 3.1.1 $E_h$ -pH relationships

The oxidation-reduction of water can be expressed by the reaction



for which the corresponding Nernst equation [Siegel 1974] linking  $E_h$  and pH is

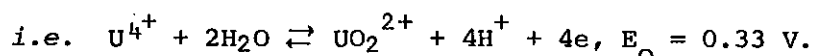
$$E_h = E_o + \frac{0.059}{4} \log \frac{[p\text{O}_2] [\text{H}^+]^4}{[\text{H}_2\text{O}]^2} .$$

The standard potential ( $E_o$ ) for this reaction is 1.23 V. Thus

$$E_h = 1.23 + 0.015 \log [pO_2] - 0.059 \text{ pH},$$

where  $pO_2$  is the partial pressure of oxygen and the activity of  $H_2O$  is 1. At a typical ground water pH of 7 and an oxygen partial pressure of 0.101 kPa ( $10^{-3}$  atmospheres), the  $E_h$  would be about 0.77 V. At a high oxidation potential such as this, the main ionic species of uranium in an essentially pure water system would be  $UO_2^{2+}$  and  $UO_2(OH)^+$  (Hostetler & Garrels [1962, Figure 3]).

At low oxidation potentials, reduction of  $UO_2^{2+}$  occurs



This oxidation potential is in the range for various natural aqueous environments studied by Baas Becking et al. [1960], and is applicable to the Burdekin Delta aquifer. Both ionic species would therefore be expected to occur in water with a pH ranging from slightly acidic to slightly alkaline, but the  $U^{4+}$  species has a high charge density and is more likely to undergo extensive hydrolysis with resultant precipitation. A well-defined reducing (or complexing) environment would be required to stabilise the tetravalent species as even small amounts of oxygen will cause oxidation of  $U^{4+}$  to  $UO_2^{2+}$ . Small amounts of oxygen, quite sufficient to maintain oxidising conditions, would be present in both infiltrating groundwater and recharge water flowing into channels and pits.

An explanation of the low uranium concentration at bore B7 may come from a study of its penetration of a mangrove mud layer (Figure 2). The redox potential in this area may be low enough to result in the precipitation of uranium from solution.

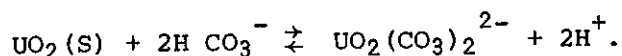
### 3.1.2 Complex ion formation effects with uranium

The solubility and mobility of uranium is markedly affected by some ionic species, particularly the carbonate ion. Groundwaters always contain both carbonates and dissolved  $CO_2$ , often in concentrations up to  $10,000 \text{ mg } \ell^{-1}$ . Figure 3 illustrates a closed system containing the equivalent of  $440 \text{ mg } \ell^{-1}$  of  $CO_2$  in solution and is adapted from the work of Hostetler & Garrels [1962, Figure 7]. In the usual pH range of groundwater [Baas Becking et al. 1960] and particularly those of the Burdekin Delta samples, the predominant ionic species of uranium is the dicarbonate ion  $(UO_2(CO_3)_2 \cdot 2H_2O)^{2-}$ . It is important to note that the uranium di- and tricarbonate species are both

stable, even under slightly reducing conditions.

The stability of the carbonate complex is dependent on pH; the transition from the dicarbonate form to  $\text{UO}_2^{2+}$  takes place at about pH 4.5. This is in substantial agreement with data based on titration studies [Miller 1958]. Under appropriate  $E_h$ -pH conditions, precipitation of uranium occurs as either  $\text{UO}_2$  (uraninite) or  $\text{UO}_2 \text{CO}_3$  (rutherfordine).

A typical reversible reaction between bicarbonate ion and an insoluble form such as  $\text{UO}_2$ , which will cause mobilisation of the uranium, is



This suggests that the solubility of uranium is both pH dependent and proportional to  $[\text{HCO}_3^-]^2$ , i.e.

$$\log [\text{uranium concentration}] = 2 \log [\text{HCO}_3^-] + \text{constant}.$$

Data for the Burdekin Delta are shown for convenience, in Figure 4 as a plot of  $\log [\text{HCO}_3^-]$  against  $\log [\text{uranium concentration} \times 10]$ . The least-squares line of best fit has a slope of 1.9 (s.d.  $\pm 0.3$ ), which is in excellent agreement with the theoretical slope of 2.0 based on the thermodynamic data obtained under laboratory conditions (Figure 3). The theoretical slope is found because the association constant for the  $\text{UO}_2(\text{CO}_3)_2^{2-}$  species ( $4 \times 10^{14}$ ) is much greater than that for corresponding complexes with chloride (0.8) or sulphate (50), the other major anionic components of groundwater.

This appears to be the first experimental demonstration of the applicability of the equilibrium diagram to uranium levels in groundwater samples collected over an extensive basin. A similar result has also been obtained for a series of samples collected in the Mereenie sandstone aquifer near Alice Springs, Northern Territory. In future, the  $\log [\text{HCO}_3^-]$  against  $\log [\text{uranium concentration}]$  plot will be used routinely to test whether the aqueous uranium chemistry in groundwater sampled from quartzose aquifers is normal. Substantial deviations from the theoretical slope of 2.0 will need to be explained.

The linear correlation coefficient  $r$  is 0.83

$$\text{i.e. } r = \frac{N\sum XY - (\sum X)(\sum Y)}{\sqrt{(N\sum X^2 - (\sum X)^2)(N\sum Y^2 - (\sum Y)^2)}}$$

where, for example,  $X = \log [\text{HCO}_3^-]$  and

$$Y = \log [\text{uranium concentration} \times 10] \quad .$$

This establishes that in the Burdekin Delta the bicarbonate ion is highly significant in solubilising the uranium. Consideration was given to the possibility that the uranium concentration in the Burdekin River, which is the basic artificial recharge input to the aquifer, may be related to volume flow. If this was the case, it was thought most unlikely that the uranium concentration would be proportional to the bicarbonate concentration with the theoretical slope of 2. There are often situations where this effect might well be important, and a systematic study of the variation of total uranium and  $^{234}\text{U}/^{238}\text{U}$  activity ratio with volume flow is planned for another area.

Correlation coefficients were determined for other ion constituents (Table 1) in the Burdekin Delta waters and have been compared with similar values obtained by Dall'Aglia [1972] in some Sicilian stream waters (Table 2). As was expected, the leaching capacity of water is related to its chemical composition and is reflected in the uranium to total dissolved solid (TDS) correlation.

Uranium concentrations significantly higher than Burdekin River water were measured at bores HJ5.2 ( $14 \mu\text{g l}^{-1}$ ), J6 ( $4.4 \mu\text{g l}^{-1}$ ), J7 ( $5.5 \mu\text{g l}^{-1}$ ) and HJ6A ( $3.8 \mu\text{g l}^{-1}$ ). Two of these (bores HJ5.2 and J6) also showed high carbonate contents. An explanation of the very high uranium and carbonate contents of bore HJ5.2 was found on examination of the drill log (Figure 2) which showed layers of limestone, granite and decomposed granite just below the screen level. Bore HJ6.1 is located about midway between bores HJ5.2 and HJ6A, and although this bore was not sampled, it also showed (Figure 2) a similar layering of limestone and granite below the screen. It seems reasonable to speculate that it may also show high carbonate and uranium levels which possibly might contribute to the chemical environment of a neighbouring bore such as HJ6A.

### 3.2 $^{234}\text{U}/^{238}\text{U}$ Activity Ratio

This ratio, in conjunction with the uranium concentration, has been found to reflect the origin and history of a particular water, providing in a sense, an isotopic fingerprint of groundwater masses [Campbell 1975]. Relative decreases in uranium concentration either by precipitation from solution or by simple dilution do not change the  $^{234}\text{U}/^{238}\text{U}$  activity ratio. Leaching from rock or soil can change both the  $^{234}\text{U}/^{238}\text{U}$  activity ratio and the uranium concentration. Time dependent processes, such as decay or enrichment, change only the isotope activity ratio. To establish criteria and conditions under which Osmond's mixing equations [Campbell 1975] may be

applied to the type of subsurface flow in the Burdekin Delta would require more data than are available. For these reasons, only general relationships will be used to illustrate the underlying usefulness of the environmental uranium isotope approach.

The action of meteoric waters in leaching the various earth materials of the area results in a runoff to the Burdekin River of water containing uranium with a  $^{234}\text{U}/^{238}\text{U}$  activity ratio of 1.38. This value would characterise any river water recharging the aquifer system either by inflow from the river banks or via artificial recharge lines and pits. Similarly, selective removal of either  $^{234}\text{Th}$  or authigenic  $^{234}\text{U}$  [Campbell 1975] during infiltration of ground water through different soil types will give rise to a  $^{234}\text{U}/^{238}\text{U}$  activity ratio in water which is characteristic of the source and enables discrimination against possible mixing patterns from otherwise equivalent sources.

Consider the two bores in which the  $^{234}\text{U}/^{238}\text{U}$  activity ratios are numerically the greatest. The higher of these values was measured at J8 (AR = 2.73) which is about 3 km north-west of Mt. Inkerman and located near a recharge line. The other bore FG4 (AR = 1.78) is located very close to the river. The tritium levels in these bores are respectively 7.1 TU and 7.6 TU (1 tritium unit (TU) = 1 tritium atom in every  $10^{18}$  hydrogen atoms), which indicates a high local recharge of meteoric water. The bores were sampled during the period December 1972 to May 1973, when the monthly rainfall measured at the Claredale Station in the Burdekin Delta varied from 3.9 TU to 13.8 TU. The annual weighted mean for 1972 was 7 TU. Although residence times cannot be calculated unequivocally, the tritium results are consistent with the assumption that the water percolated underground less than a year before sampling. The high  $^{234}\text{U}/^{238}\text{U}$  activity ratio may therefore reflect leaching from the top soil by a process of direct infiltration. In the case of bore J8, runoff from the granitic pediment of Mt. Inkerman may also be contributing to the high  $^{234}\text{U}/^{238}\text{U}$  activity ratio in the same way.

Bore (A-B) S7 is in an area believed to be a major source of recharge from the river. Both the uranium concentration and the  $^{234}\text{U}/^{238}\text{U}$  activity ratio are essentially the same as those in Burdekin River water. Thus, support is given to the view that river water is infiltrating the aquifer system in this region. Bore 1K5, and bore H6 which is close to a recharge line, also have activity ratios consistent with inputs of river water.

Bore G6.3 is located in a multibanded clay/sand structure. Its tritium level shows no input of modern water and consequently there must be restricted flow characteristics in its vicinity. The low  $^{234}\text{U}/^{238}\text{U}$  activity ratio of 1.17 is interpreted as indicating a limited interaction of groundwater with ground material having a low uranium content, e.g. clay.

As another example, water level contours (Figure 5) show the possibility of flow to bore HJ7 from either bore HJ6A or bore J7. The higher  $^{234}\text{U}/^{238}\text{U}$  activity ratio and lower uranium concentration at bore HJ7 contrast with lower  $^{234}\text{U}/^{238}\text{U}$  activity ratios and higher uranium concentrations in bores HJ6A and J7. It is unlikely, in this case, that there is any flow from either HJ6A to HJ7 or from J7 to HJ7.

#### 4. SUMMARY OF RESULTS

Measurements of uranium concentration and  $^{234}\text{U}/^{238}\text{U}$  activity ratio in bore water samples varied considerably, reflecting local hydrogeochemical areas. All data illustrate the complexity of the aquifer system and tend to support the view of Watkins & Wolff [1960] that it is a system of aquifers separated by aquicludes. Support is given to those studies which have indicated areas of inflow of water from the Burdekin River into aquifer storage.  $E_h$ -pH effects and complexing agents, particularly carbonates, play an important role in mobilising and transporting uranium.

#### 5. ACKNOWLEDGEMENT

The author wishes to record his appreciation of the constructive conversations and helpful suggestions of Dr P.L. Airey that have contributed to this paper. The chemical analyses of the water samples were carried out by the Department of Primary Industries, Queensland.

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TABLE 1  
CHEMICAL ANALYSES OF BURDEKIN DELTA WATER SAMPLES

Sample	TDS mg/ℓ	Ca mg/ℓ	HCO <sub>3</sub> mg/ℓ	Cl mg/ℓ	SO <sub>4</sub> mg/ℓ	pH	U μg/ℓ	<sup>234</sup> U/ <sup>238</sup> U
BURDEKIN R							1.4	1.38
H5	113	17	100	20		8.2	0.3	
H6	309	42	195	73	27	7.9	0.42	1.41
HJ5.2	1663	220	451	795	34	8.1	14	1.38
J6	839	75	523	260		8.2	4.4	
G6.3							0.88	1.17
HJ6A							3.8	1.24
HJ7	527	80	237	185	34	8.3	0.50	1.65
K7	351		85	180		7.4	0.29	
J7							5.5	1.39
JK8A	321	26	152	95	30	8.1	0.17	
J8							0.22	2.73
FG9	1944	120	231	1020	94	7.9	0.23	
H9	1101	70	272	425	144	8.3	0.54	
IK5							0.26	1.38
FG7							0.22	1.46
G5							0.30	
EF5.2	120	21	112	16		7.3	0.32	
FG4.7	74	14	73	7		7.7	0.14	1.78
PC16	95	9	84	17		7.9	0.12	
E2	219	32	134	70		7.7	0.14	
C1							0.23	
B3	848	70	121	230	270	7.5	0.17	
B5	329	30	277	30	36	8.0	<0.09	
L2B7	426	16	235	48	100	7.9	<0.09	
PR12	171	26	84	20	50	7.8	<0.09	
B7	1140	70	155	260	415	8.1	<0.09	
AN5	114	21	84	15		8.0	0.14	
E9						8.2	<0.09	
C9	1870	40	256	860	185	7.7	<0.09	
(A-B)S7	260	34	268	20	15	7.8	1.9	1.34
D6	179	6	146	16	23	7.6	0.15	
D7							<0.09	

TABLE 2  
COMPARISON OF CORRELATION COEFFICIENTS OF URANIUM  
WITH CONSTITUENTS OF BORE WATERS

	Total dissolved solids	Ca	HCO <sub>3</sub>	Cl	SO <sub>4</sub>	pH	Conductivity
BURDEKIN DELTA SAMPLES	0.55 (a)	0.82	0.83 (a)	0.51	-0.16	0.26	0.57
DALL'AGLIO (15)	-	0.64	0.24	0.45	0.64	-	-

(a) Determined for a log-log relationship.



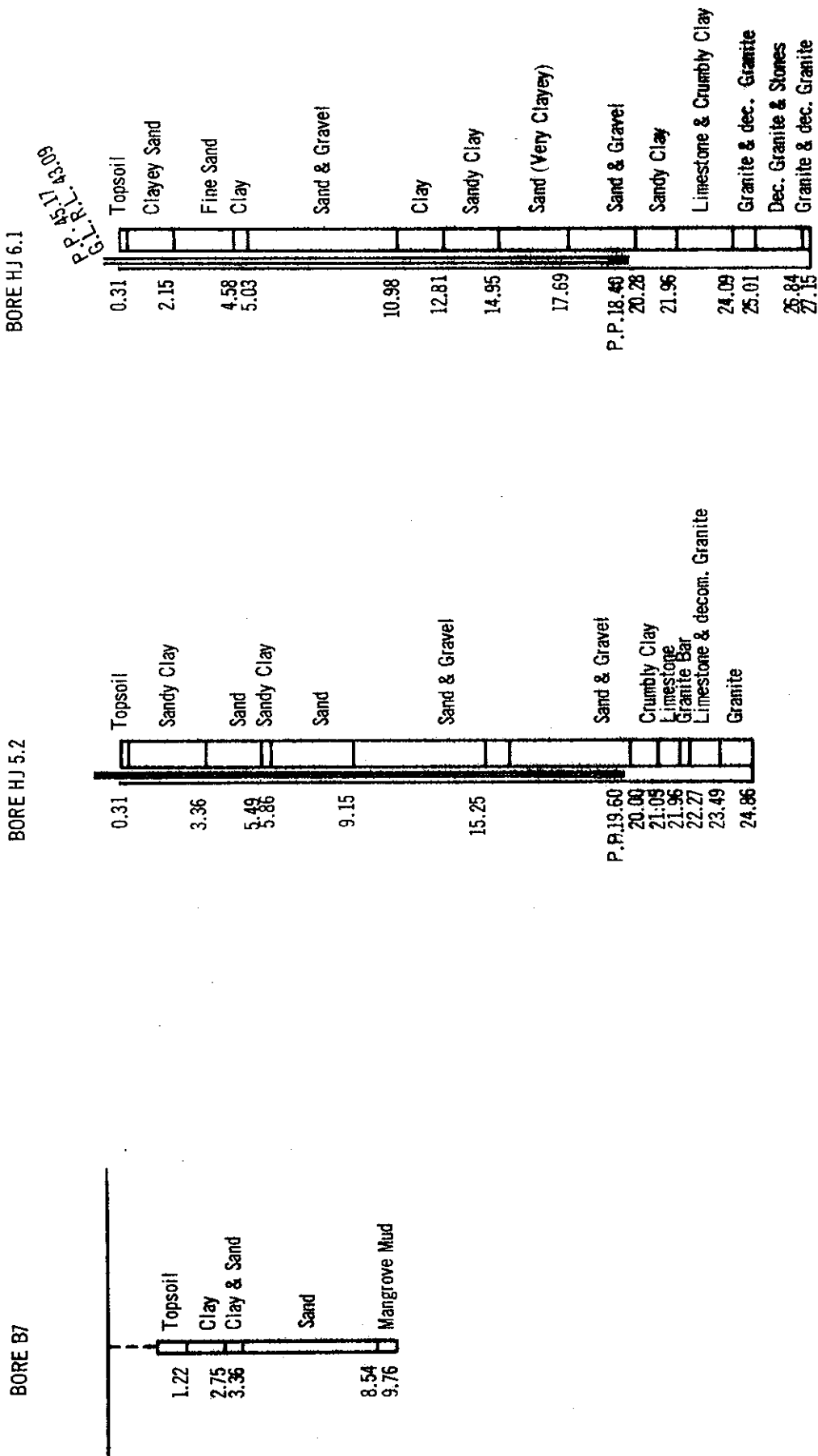


Figure 2 Drill logs of selected sample bores (see Figure 1 for location). Depths are in metres from the surface.

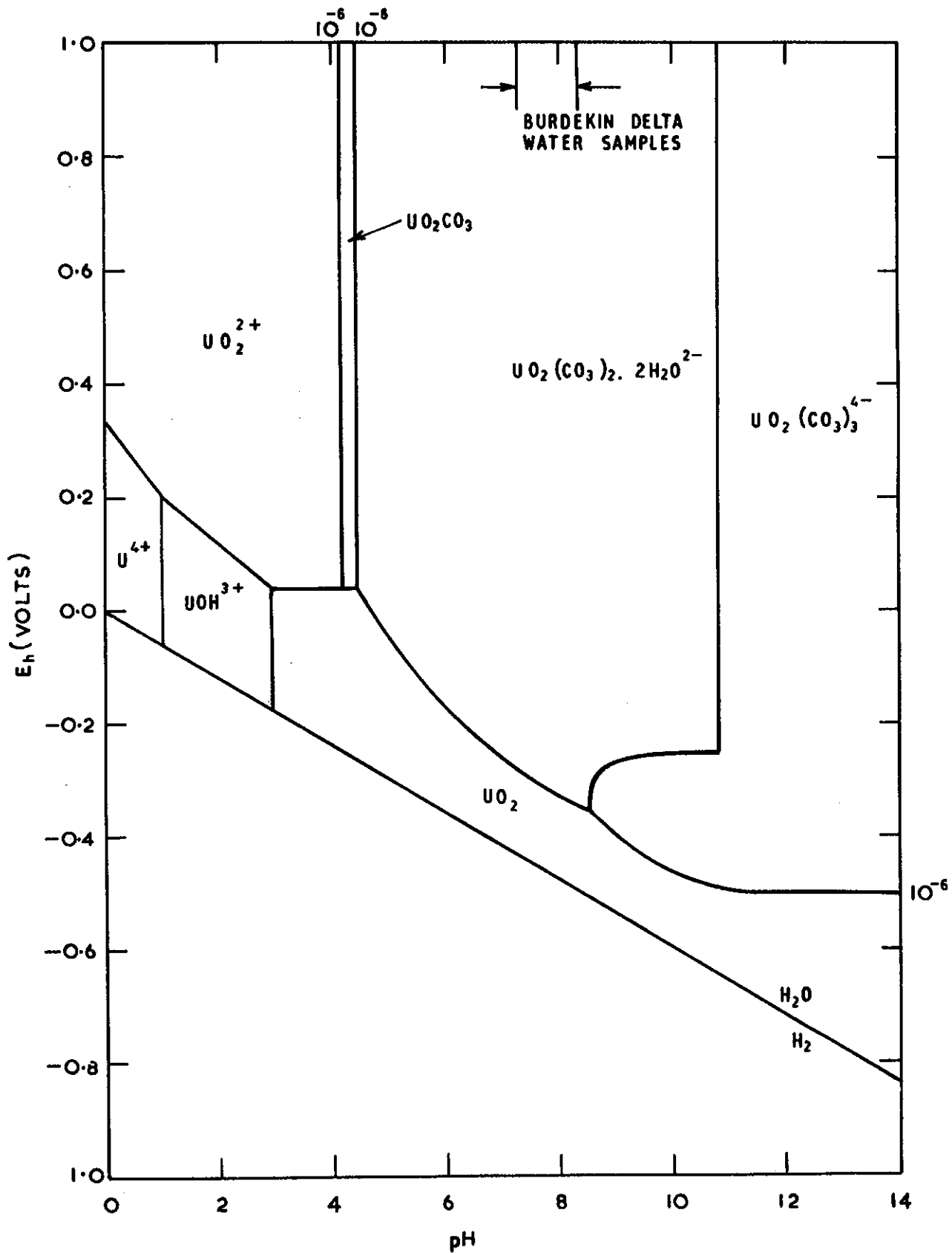
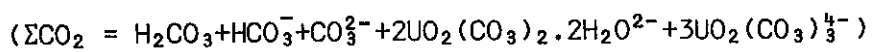


Figure 3 Aqueous equilibrium diagram of the U-O<sub>2</sub>-H<sub>2</sub>O-CO<sub>2</sub> system at 25°C and 101 kPa with  $\Sigma\text{CO}_2 = 440 \text{ mg } \ell^{-1}$ . The total ionic activity of dissolved uranium species in equilibrium with solid phases is  $10^{-6}$ .



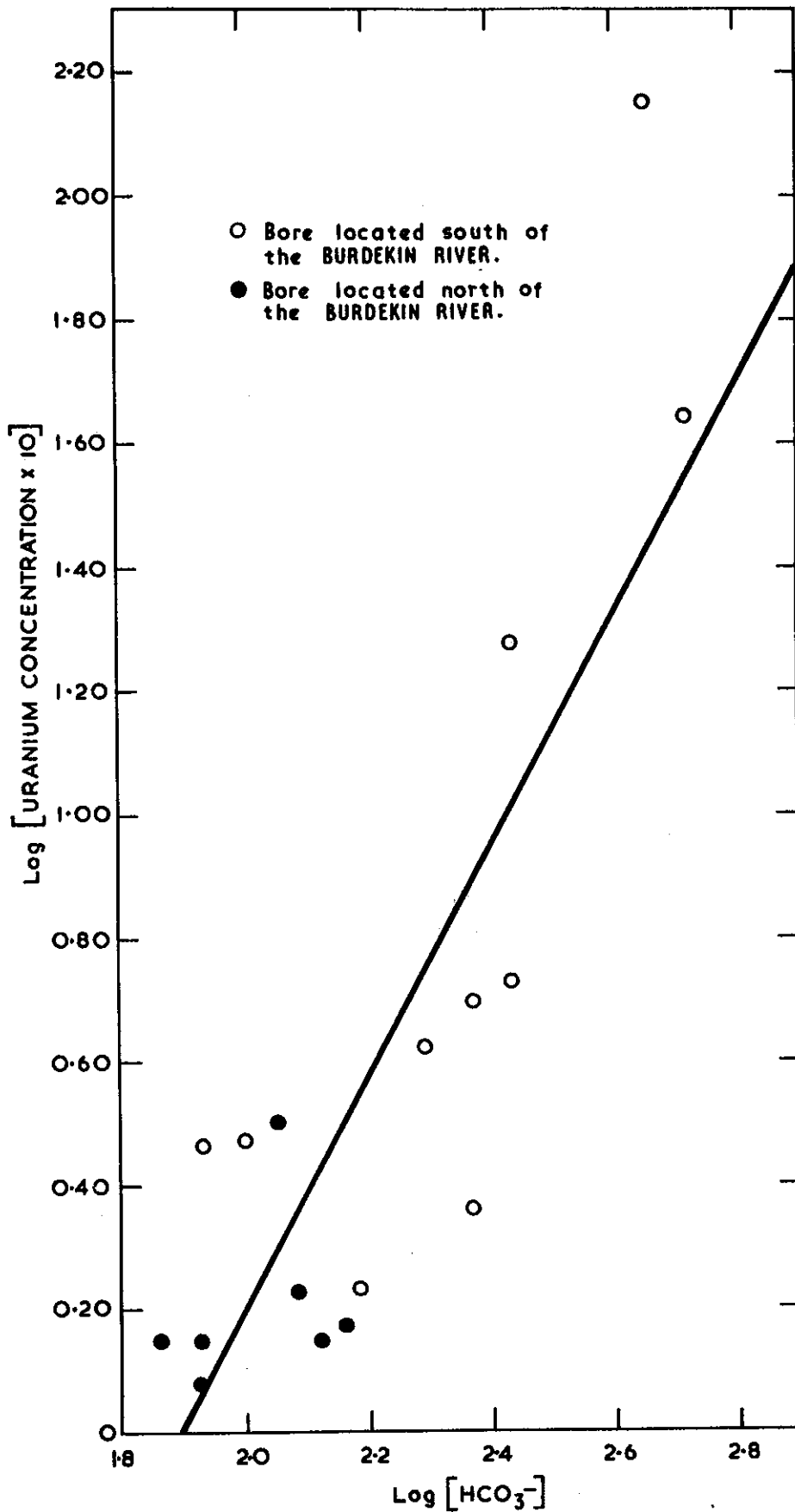


Figure 4. Relation between uranium concentration and bicarbonate ion concentration. The line of best fit is drawn with a slope of 1.88.

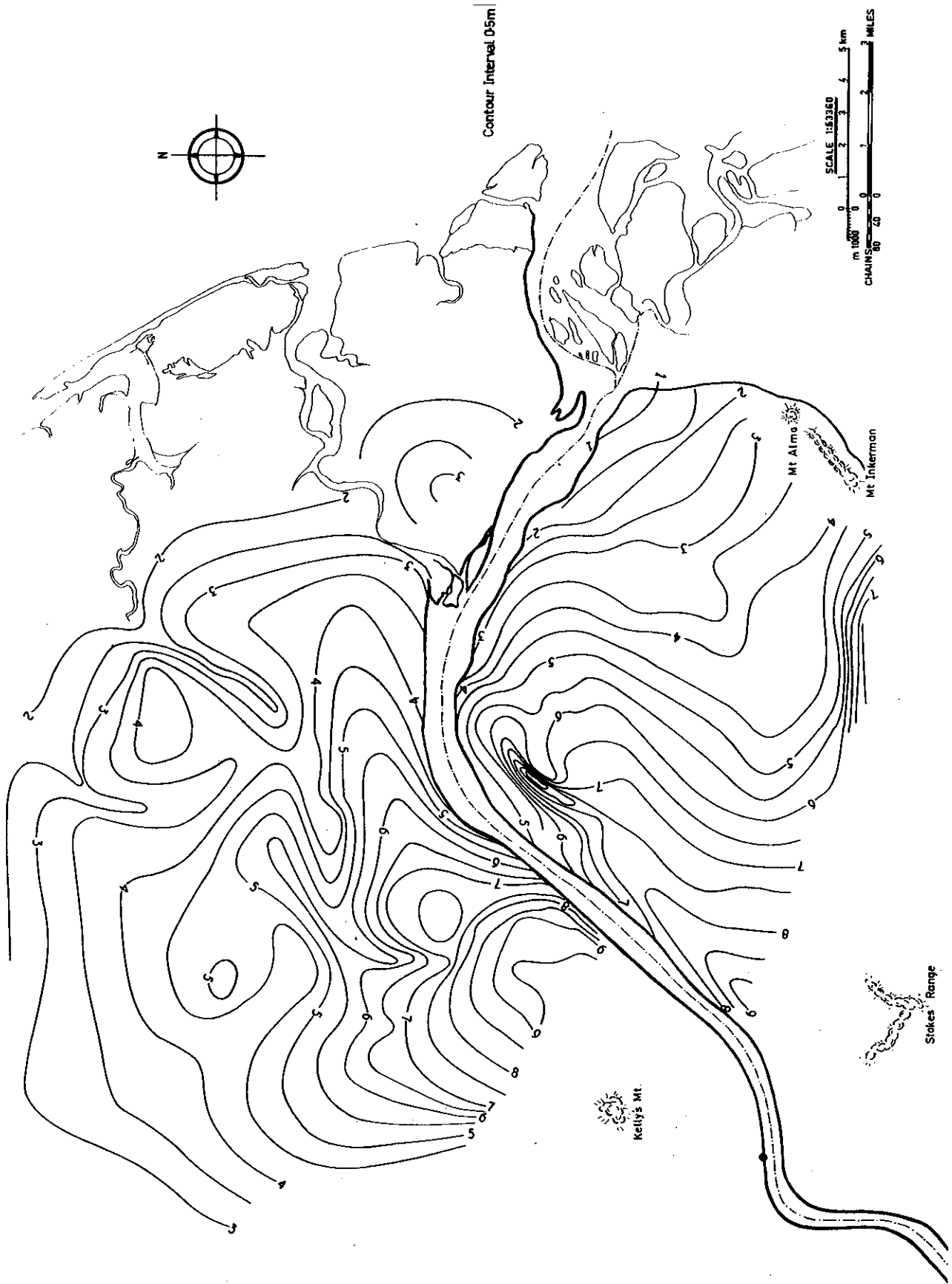


Figure 5 Burdekin Delta water level contours, June 1974.

