

A GEOCHEMICAL COMPARISON OF MODERN AND HOLOCENE MANGROVE SEDIMENTS, TOWNSVILLE REGION, NORTH QUEENSLAND

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ABSTRACT

The geochemistry of coastal sediment is important in terms of potential soil acidification and the resultant mobilisation of metals. This paper reports geochemical analyses for mangrove sediments from the Townsville region, north Queensland. Vibrocores were taken from modern fringing and riverine mangrove swamps, from a recently buried mangrove swamp (ca. 200 B.P.), and from buried Holocene mangroves of the inner shelf of Cleveland Bay. Although the average pH (ca. 5) and Eh (ca. -250 mV) is similar for both the modern and early Holocene mangrove sediments, the pH and Eh are more variable in the younger mangrove sediments. Strongly acidic conditions (ca. pH 2.5) exist in the recently buried mangrove sediments, mainly caused by the degradation of organic matter. The porewater chemistry of early Holocene mangrove sediments suggests that acid-sulphate material may be preserved under marine and anoxic conditions because of limited exchange with seawater. The clay-rich nature of the mangrove sediments in the central Great Barrier Reef region enhances their chemical buffering capacity, the main buffers being clay (silicate) minerals, carbonates, iron-oxyhydroxides and organic matter. This leads to a reduced potential for acid-sulphate soil development and metal mobilisation.

INTRODUCTION

Acidification of mangrove swamps is a major environmental problem in the coastal floodplains of eastern Australia (Lin & Melville, 1992; Sammut *et al.*, 1994) and in agricultural areas elsewhere in the world (Moormann & Pons, 1974; Gong & Zhang, 1993). In tropical regions, increased evaporation and salinity during the dry season may increase soil acidity and

mobilisation of metals. Mangrove swamps and their associated fine-grained sediments occur along much of the modern north Queensland coast, and have done so through much of the mid and late Holocene, as revealed by seismic surveys and core transects (Thom *et al.*, 1975; Cook & Mayo, 1980; Way, 1987; Harris *et al.*, 1990; Carter *et al.*, 1993). In the Townsville region of northeastern Queensland, mangroves occur adjacent to the modern tidal inlets, and preserved mangrove sediments occur within the Holocene coastal wedge (Carter *et al.*, 1993).

The last post-glacial sea-level rise flooded the Great Barrier shelf at ca. 9.5 ky B.P. and peaked at 5.5 ky B.P. at 1 - 2 m above modern levels (Beaman *et al.*, 1994; Larcombe *et al.*, 1995). The ancient mangrove sediments studied here are derived from mangrove swamps which were drowned during the late stages of sea-level rise (ca. 7.8 and 6.9 ky B.P.) before sea-level fell to present day levels (Carter *et al.*, 1993; Larcombe *et al.*, 1995). Throughout the Holocene, terrestrially-derived sediments have diluted marine-derived components on the inner shelf (Belperio, 1978; Carter *et al.*, 1993), and the dominance of fine-grained terrigenous sediments in the mangrove swamps is maintained by a combination of onshore-directed sediment transport on the inner-shelf and probably the hydrodynamics of the mangrove systems themselves (Wolanski *et al.*, 1990, 1992, Larcombe & Ridd, 1996).

Recent evidence for episodic sea-level rise in the north-east Australian region suggests that the buried mangrove swamps may have been deposited during stillstands or slight regressions in sea level (Larcombe *et al.*, 1995) which would have produced sub-aerial exposure of some mangrove sediments. The long-term wetting and

drying of the early Holocene mangrove sediments caused by sea-level changes contrasts with the continuous short-term tidal and seasonal wetting and drying of the modern sediments. By comparing the geochemistry of mangrove sediments from a range of ages and environmental conditions, we examine the effects of burial and exposure on sediment chemistry, including the potential for development of acid-sulphate soils.

LOCATION

The study area is situated in the Townsville region, north Queensland, approximately 19°20'S, 146°35'E (Fig. 1). We present data from mangroves of three different age groups, modern, recently buried, and early Holocene.

Town Common. Cores were taken approximately at mean sea level. One core is from a recently-buried (*ca.* 200 B.P.) dead mangrove forest (TC 1), and the other is from an adjacent living mangrove forest (TC2). A further vibrocore at *ca.*+ 1.5 m A.H.D. was taken from riverine mangroves of Gordon Creek (GC1), a site described by Larcombe & Ridd (1995, 1996).

(ii) Early Holocene mangrove sediments.

Three vibrocores (KG931-VC1, VC4 and VC5) were collected in a north-south transect across Cleveland Bay (Fig. 1), located from previously logged and dated Holocene mangrove deposits (Carter *et al.*, 1993; Larcombe & Carter, in prep.). The early Holocene mangrove sediments are buried beneath modern marine sediments

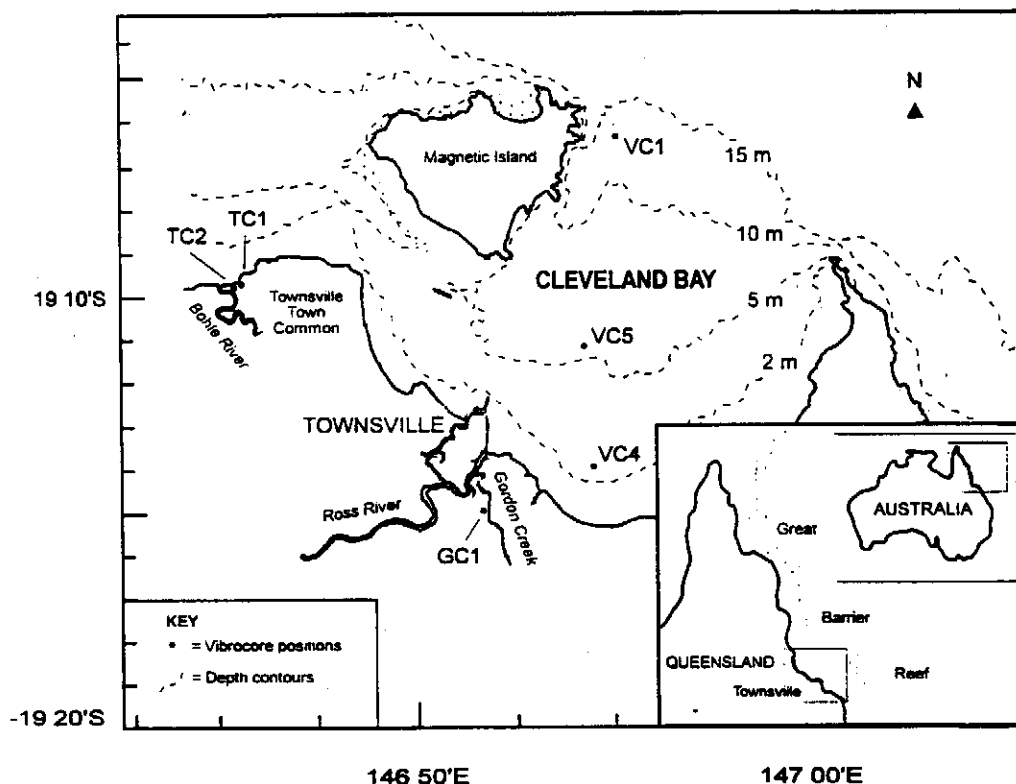


Figure 1: Location map, showing sample sites of modern mangrove sediments (GC1 & TC2); recently buried mangrove sediments (TC1) and Early Holocene mangrove sediments (sampled from vibrocores VC1, VC4 & VC5 taken in Cleveland Bay

(i) Modern mangrove sediments.

Two vibrocores were taken from the fringing mangroves off the Townsville

on the inner shelf (Fig. 2) and overlie transgressive and pre-transgressive Pleistocene sediments (Carter *et al.*, 1993; Ward *et al.*, 1996). The modern and

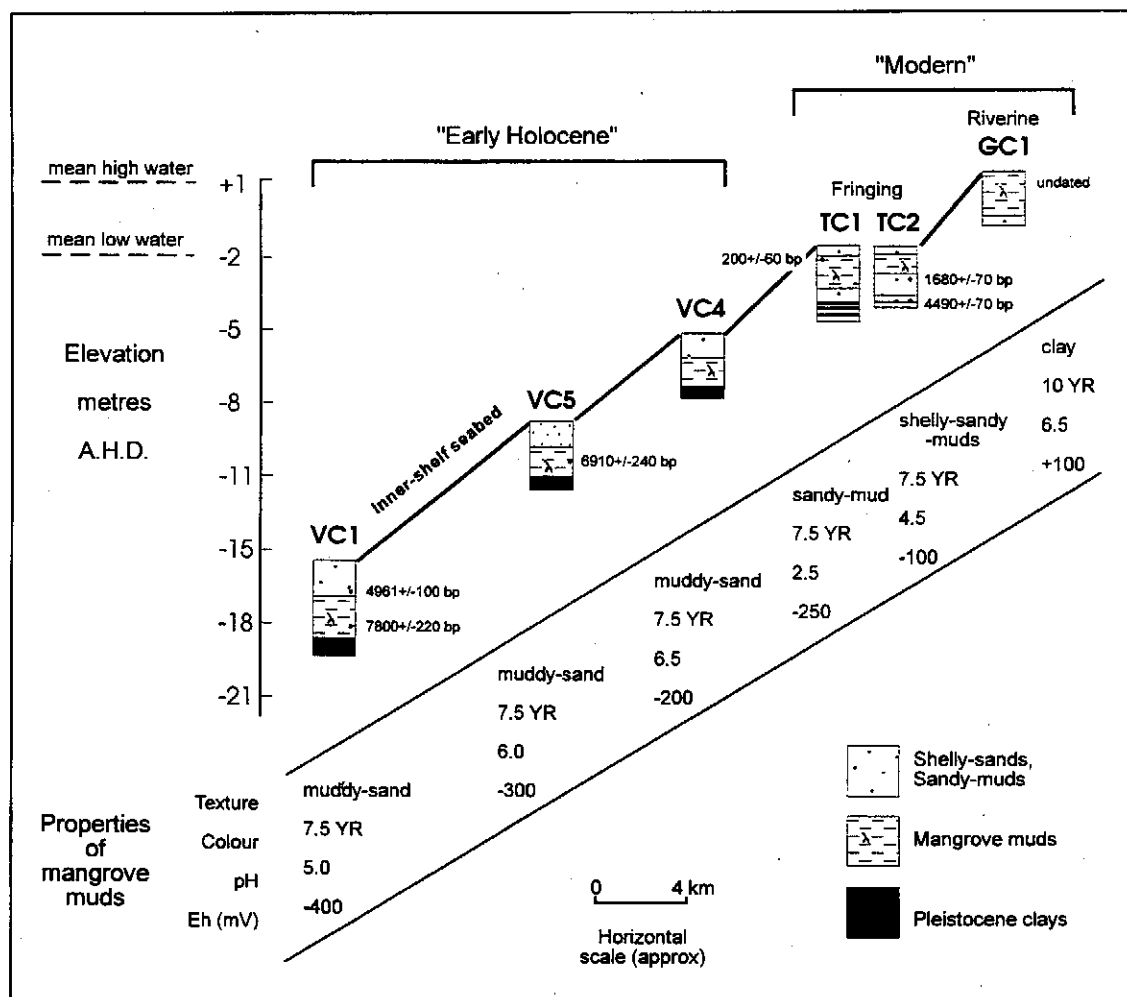


Figure 2: Composite cross-section through Cleveland Bay showing summary core logs and the stratigraphic relationship of the studied mangrove sediments. Radiocarbon age data given here are tabulated fully in Larcombe *et al.* (1995) and Larcombe & Carter (in prep).

ancient mangrove sediments we studied were of similar texture, and included dark muddy-sands and silty-clays which would have been deposited in swamps, and coarse sands similar to those found in the modern mangrove channels. The sediments have a relatively low organic content, generally less than 5%, and most have less than 5% carbonate (Carter *et al.*, 1993).

METHODS

(i) Sediments

Land vibrocores were taken using 3 m long core barrels, whilst marine vibrocores were collected from the R.V. James Kirby using

4.5 m long core barrels. Following collection, cores were cut into 25 cm lengths, sealed in nitrogen-filled polyethylene plastic and stored at 4 °C prior to further sampling and analysis. Using a nitrogen-filled glove-box, a sediment sample was taken from the centre of every alternate 25 cm length. Measurements of sediment pH and redox were taken with a TPS portable meter. Sediment mineralogy was determined from smear-mounted slides on a Rigaku Denki D2155 X-ray diffractometer (XRD) fitted with a post-diffraction curved graphite crystal monochromator using CuK α radiation. Clay minerals were identified according to the scheme of MacKinnon and Cuff (1992). Samples were divided into three

grain size fractions (sand, 200 - 63 μm , silt, 63 - 20 μm , and clay, <20 μm) by washing through nylon mesh with filtered seawater and then oven-drying at 60°C. The concentrations of major and minor elements in these fractions were determined from pressed boric acid pellets and glass beads, using a Siemens SRS 3000 X-ray fluorescence spectrometer (XRF). Major element concentrations are reported here as oxides (wt%), and minor elements are reported as elements (ppm). The organic carbon and sulphur contents were determined using a Leco SC-444DR analyser, after removal of carbonate using dilute HCl (10%).

(ii) Porewater

A total of 30 porewater samples were obtained by centrifugation of sediments. Under nitrogen, ca. 100 g of sediment was transferred into 250 ml polyethylene centrifuge tubes, which had been washed with Ultrix HNO₃ (5%) and spun for 10 min at 3000 rpm in a MSE centrifuge. Using acid-washed 50 ml syringes, porewater was then passed through 0.33 μm Millex in-line filters into washed 10 ml polyethylene tubes, acidified with 0.1 mL of conc. HNO₃, and stored at 4°C.

Dissolved Fe²⁺ and Mn²⁺ concentrations were measured on a Varian Spectra AA-400 Zeeman Corrected Graphite Furnace (GF-AAA). Concentrations of Ca²⁺, Sr²⁺, K⁺ and SO₄²⁻ were measured using a Varian AA-300 Atomic Absorption Spectrophotometer (AAS).

RESULTS

Mineralogy

(i) Modern mangrove sediments.

The surface shelly sandy-muds of the fringing mangroves are comprised of quartz (ca. 40%), carbonate (aragonite & calcite, 10%), alkali feldspars (mainly albite, 20%), clay minerals (mixed-layer clays, smectite, illite and kaolinite, 10%), accessory minerals (5%), evaporite minerals (e.g. gypsum, halite, 5%) and organic matter (5%). A similar mineral assemblage is found in the underlying

mangrove muds but with more clay minerals (up to 60%) and organic matter (10%), and less quartz (10%), alkali feldspars (10%), and carbonate (5%). The riverine clay sediments are similar to those of the fringing mangroves, while underlying coarse sandy sediments (probably representing the basal sediments of an infilled mangrove channel), are almost wholly made up of quartz (ca. 70%) and feldspar (ca. 20%).

(ii) Early Holocene mangrove sediments.

Early Holocene mangrove muds consist of clay minerals (mixed-layer clays, smectite, illite and kaolinite, 60%), quartz (10%), carbonate (aragonite & calcite, 5%), alkali feldspars (mainly albite, 10%), accessory minerals (5%), evaporite minerals (e.g. gypsum, halite, 5%), and organic matter (5%). The overlying modern bay sediments comprise more quartz (up to ca. 30%), feldspar (20%) and carbonate (10%), and less clay minerals (30%). The underlying Pleistocene sediments are almost wholly comprised of clay minerals (80%), with some quartz and organic matter (each 10%).

pH and Redox

(i) Modern mangrove sediments.

Most samples are acidic (ca. pH 5) and anoxic (Eh -250 mV) (Fig. 3). The riverine mangrove sediments of Gordon Creek are less acid (ca. pH 6) and are oxidising (+100 mV). Within the fringing mangrove sediments of the Town Common, the surface sandy sediments are porous, less acid (ca. pH 5.5) and are more oxidising (+10 mV) than the underlying muddy sediments (pH 4.5, Eh -250 mV). The recently buried mangrove muds are even more acid (pH 2.5 - 4) and more strongly reducing (Eh -350 mV).

(ii) Early Holocene mangrove sediments.

The mangrove sediments are more acid (pH 4 - 6) and reducing (ca. Eh -250 mV) than both the overlying shoreline sands (pH 6, Eh -150 mV) and surficial bay sediments (pH 6, Eh -100 mV). Redox is more oxidising in the Pleistocene sediments beneath the mangrove muds (Fig. 3). Acidity is lower in VC1 (ca. pH 4.6), the

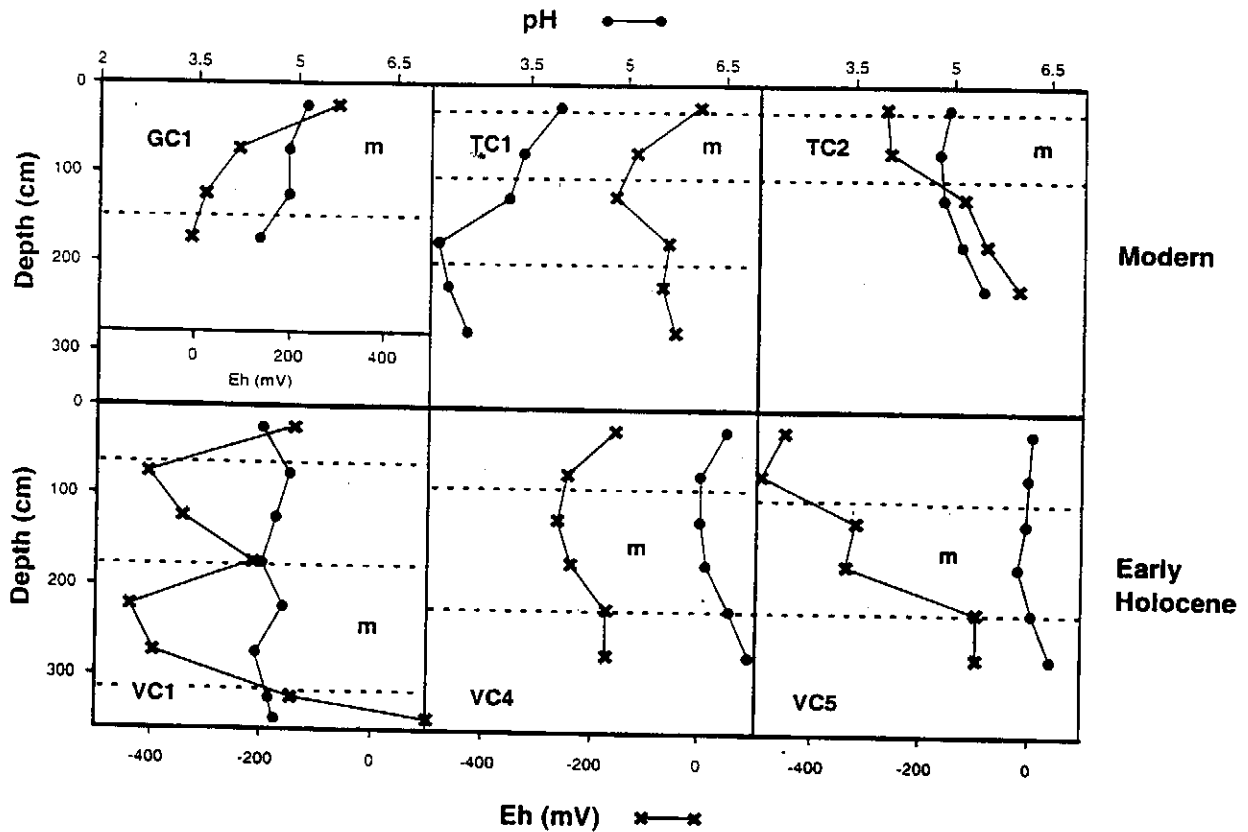


Figure 3: pH (dots) and Eh (crosses) profiles for each core. Horizontal dashed lines represent position of lithological boundaries (see Fig 2), and "m" marks the mangrove mud unit in each core. (Note the different scale for Eh for core GC1).

oldest sampled mangrove, than in cores VC4 and VC5 (pH 6.3). There is considerably less variation in the pH profiles in the early Holocene stratigraphy than in the modern mangrove sediments (Fig. 3).

Sediment geochemistry

Overall, the sediment geochemistry of the modern mangrove sediments is very similar to those of the early Holocene, and reflects the mineralogy and, to a lesser extent, the redox conditions of the sediments. High concentrations of CaO, K₂O, Rb and Sr in the sand fraction reflect the presence of carbonate and feldspar (Table 1) (see also Ward *et al.*, 1995). In contrast, the silt and clay fractions have negligible amounts of carbonate and feldspar, shown by their virtual absence on XRD traces. This probably indicates that they have been dissolved, an interpretation supported by

slightly lower concentrations of CaO, K₂O, Rb and Sr in these fine fractions (Table 1). These dissolved elements are found enriched in the porewater (e.g. Ca²⁺, Sr²⁺) or in the clay fraction probably associated with the dominant smectitic and mixed-layer clays.

The sand and silt fractions of these sediments contain high concentrations of SiO₂, Al₂O₃, Ti & Zr reflecting the presence of quartz and other resistant minerals such as ilmenite, rutile, monazite and zircon. Al₂O₃, Ti and Zr are also associated with aluminosilicates in the clay fraction.

High concentrations of Fe₂O₃, Mn, Ni, Co, Pb, Zn, Cr and V occur in the silt and clay fractions of the modern and early Holocene mangrove sediments (Table 1), especially in the riverine mangroves, most

	HOLOCENE		MODERN		HOLOCENE		MODERN		HOLOCENE		MODERN	
	mean	SD	mean	SD	mean	SD	mean	SD	mean	SD	mean	SD
	<200 μm fraction											
n = 10												
SiO ₂	78	13.5	71	6.0	53	11.3	63	5.3	37	11.2	42	12.0
Al ₂ O ₃	8	4.1	11	1.3	17	6.7	12	1.6	16	6.2	13	2.5
Fe ₂ O ₃	2	1.3	2	1.0	5	0.8	4	0.8	4	1.3	5	0.6
MgO	1	0.7	1	0.3	2	0.4	1.4	0.2	2	0.8	3	0.8
CaO	3	2.2	3	1.6	3	2.3	3	1.5	1	1.2	2	0.7
Na ₂ O	2	2.1	3	1.2	6	2.5	5	2.0	9	6.7	12	4.6
K ₂ O	1	0.5	2	0.2	1	0.3	2	0.2	1	0.4	1	0.4
P ₂ O ₅	0.05	0.02	0.06	0.02	0.08	0.01	0.08	0.02	0.07	0.03	0.07	0.01
n = 17												
Pb	12	6	14	4	21	4	19	6	21	4	19	4
Rb	67	27	90	6	99	14	93	14	104	15	106	8
Zr	169	32	295	102	374	158	504	254	212	103	263	112
Zn	25	13	30	9	46	10	40	9	40	13	37	7
Ni	10	7	8	4	18	4	13	3	17	5	15	2
Co	8	6	6	3	12	3	8	3	11	4	9	2
Mn	383	217	461	152	454	186	410	170	383	199	365	165
Cr	38	12	37	9	54	17	52	15	44	19	49	13
Ti	2574	819	3242	793	4360	1572	4443	1631	3665	1886	3775	1099
V	36	16	43	10	63	17	54	16	48	30	52	11

All data in ppm

Table 1. Summary of element concentration (ppm) for Modern and Early Holocene mangrove sediments, showing division of elements in <20 μm, 20-63 μm, and 63-200 μm size fractions.

likely reflecting the presence of aluminosilicates, or Al, Fe or Mn-oxyhydroxides. Under the prevailing redox conditions, these elements could also occur within sulphide phases (Mohr, 1972; Curtis, 1987), although these were not detected by XRD and thus, if present, have concentrations below 4 %.

On average, the early Holocene mangrove muds contain slightly more organic carbon (0.89%) and sulphur (0.77%) than the modern mangrove sediments (0.77% organic C, 0.51% S). The dead mangrove sediments may contain more organic carbon (average 1.0%) and sulphur (0.61 %) than the living mangroves (0.40% organic C and 0.50% S). However, with both these comparisons, variability is high and the differences may not be significant.

Porewater chemistry

Porewaters of the modern and early Holocene mangroves are depleted in Fe^{2+} and Mn^{2+} (Fig. 4). However, concentrations are significantly higher in the underlying Pleistocene sediments, where concentrations are up to five hundred times local seawater concentrations (ca. Fe^{2+} 0.8 ppm, Mn^{2+} 1.0ppm). Ca^{2+} is slightly enriched in both the modern and early Holocene mangrove porewaters (ca. 500 ppm). Both K^{+} and Mg^{2+} are depleted (360 ppm and 920 ppm respectively) in the early Holocene mangroves, and enriched (490 ppm and 1850 ppm respectively) in the modern mangroves. Sr^{2+} is eighty times depleted (ca. 0.1 ppm) in the modern (living) mangroves, and slightly enriched (ca. 10ppm) in the early Holocene mangroves. SO_4^{2-} is depleted (ca. 1600 ppm) in the modern (dead and living) mangroves.

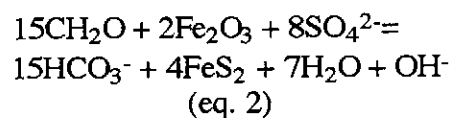
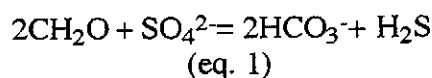
DISCUSSION

Sediment redox and acidity

In areas subject to continuous wetting and drying, consequent variations in the redox potential of the sediments can result in intense diagenetic reactions (Mohr *et al.*, 1972; Lord & Thomas, 1983; Arakel *et al.*, 1990; Fletcher *et al.*, 1993). Such variation

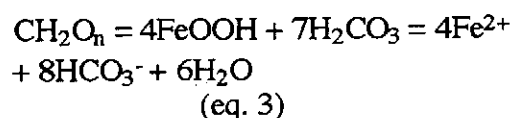
will occur in the mangroves in response to tidal and seasonal processes and longer-term changes in sea level. Sulphate reduction is generally thought to predominate in mangrove sediments due to the continuous supply of seawater sulphate and organic matter (Fry *et al.*, 1982; Aller, 1986). The smell of hydrogen sulphide is evident in the studied mangrove sediments.

The reduction reactions can be written as:



The average ratio of organic carbon to sulphur is approximately unity, indicating that there is a fairly constant rate of organic matter decomposition, sulphate reduction and pyrite formation (Berner, 1982). These results are consistent with other data in the region where some of the carbon is refractory carbon, indicating sedimentary reworking (Woolfe *et al.*, 1995).

The depletion of sulphate in the modern mangrove sediments, including the recently buried mangroves, suggests that the depletion is not due to metabolic processes. It is probably a result of sulphide formation, and precipitation of authigenic sulphate minerals such as gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) or jarosite ($\text{KFe}_3(\text{OH})_6(\text{SO}_4)_2 \cdot 2\text{H}_2\text{O}$). However, as a result of the low sulphate concentration, mineralisation may also proceed via Mn^{4+} or Fe^{3+} reduction (eq. 3). Fe-reduction is promoted by the low reactivity and/or concentration of organic matter (ca. <2% C_{org}), high concentration of reactive iron and manganese in the mangrove sediments, and physical reworking of sediments by tides and burrowing organisms (Kristensen *et al.*, 1992; Aller *et al.*, 1986).



In general, the modern and early Holocene mangrove sediments are more oxidising at the surface than at depth (Fig. 3). The

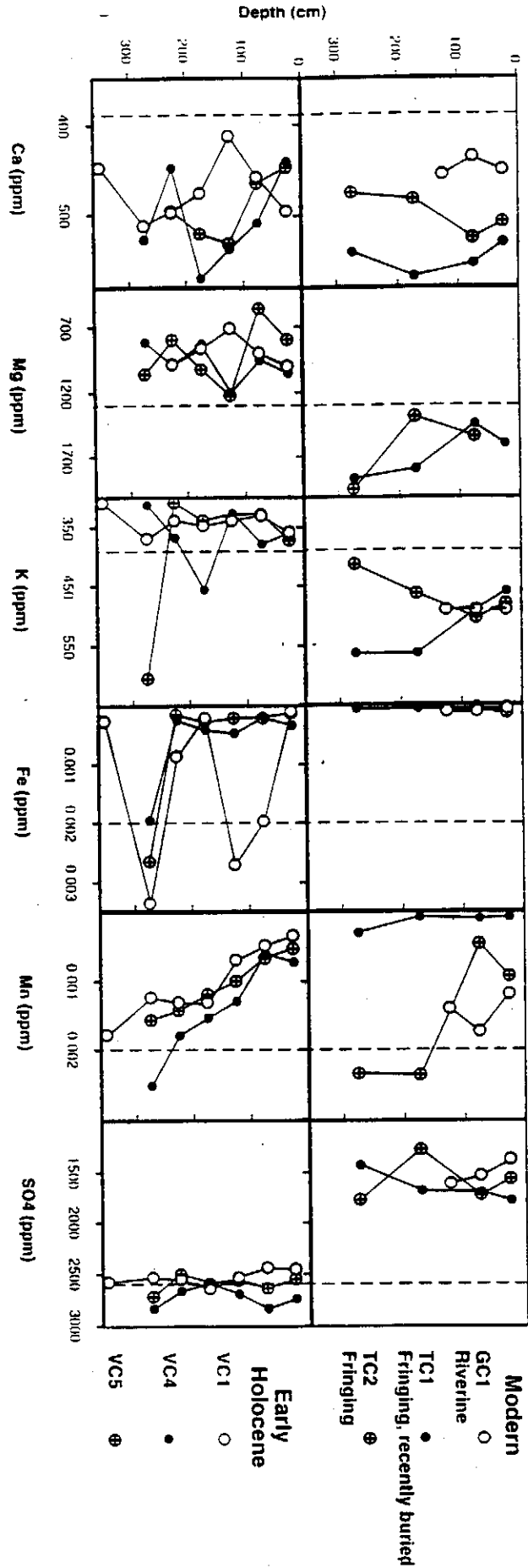
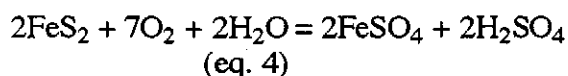


Figure 4: Concentrations of the major elements in sediment porewaters (ppm). The composition of the local seawater (Cuff, 1975) is shown by a vertical line.

increase in redox potential and concentrations of the redox sensitive elements Fe^{2+} and Mn^{2+} in the porewaters of the Pleistocene sediments suggests that fresh or brackish water conditions exist, which may influence redox reactions in the overlying Holocene mangrove sediments (see also Ward *et al.*, 1995). Redox potential is lowest in those sediments with little or no oxygen input from growing plants (Fry *et al.*, 1982; Carlson *et al.*, 1983) or through bioturbation (Smith *et al.*, 1991). In addition, the absence of vegetation results in greater evaporation, increasing salinity, soil acidity and the potential for mobilisation of metals. Similar observations have been made in other parts of the central Queensland coast (Cook and Mayo, 1980) and elsewhere (Moorman and Pons, 1975; Khoa, 1993; Sammut *et al.*, 1994). Reducing conditions in the mangrove sediments allow iron sulphides to remain inert and harmless, as "potential acid sulphate soils" (Lin & Melville, 1992). Upon aeration, the sediments may become "actual acid sulphate soils" due to the oxidation of iron and other metal sulphides to form sulphuric acid (eq. 4).



Acid-sulphate soil hazards

Evidence is increasing for an episodic rise in post-glacial sea-level in NE Australia (summarised in Larcombe *et al.*, 1995). Along the central GBR coastline, periods of sea-level stasis or fall would have increased rates of coastal progradation and sediment accumulation, including a significant proportion of mangrove sediments. At 8.2 ky B.P., some early Holocene mangrove sediments may have been exposed in a supratidal position, and would have had the potential for development into actual acid-sulphate soils. Acid conditions would have dissolved carbonate, producing porewaters initially enriched in Ca^{2+} and Sr^{2+} , and these elements, along with Mg^{2+} , may have subsequently been fixed in clay minerals. Precipitation of jarosite ($\text{KFe}_3(\text{OH})_6(\text{SO}_4)_2 \cdot 2\text{H}_2\text{O}$) via oxidation of iron sulphides (Mohr *et al.*, 1972) would account for the depletion of K^+ in the porewater, and is consistent with the mottled colouring of many of these

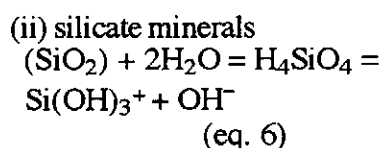
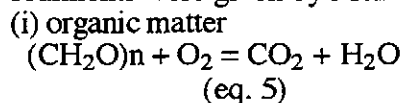
sediments.

Subsequent inundation by seawater in the next episode of sea-level rise (8 ky B.P.) might be expected to partly neutralise the sediments and restore porewater concentrations to normal seawater values. However, our results suggest this was not the case. Porewaters of the early Holocene mangrove sediments are enriched in Ca^{2+} and Sr^{2+} , and depleted in Mg^{2+} and K^+ , suggesting there has been little exchange with seawater since their inundation and burial. Porewater exchange may have been limited by rapid burial of these coastal sediments beneath transgressive inner-shelf sediments during a vault in sea level. Further, the mangrove sediments are relatively impermeable, so whilst remaining buried and permanently saturated, these early Holocene sediments have had little potential for oxidation, except locally through their exposure at the edge of the dredged main shipping channel through Cleveland Bay.

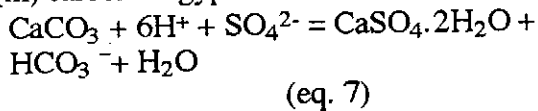
It is not unusual for mangrove porewaters to be enriched in sulphate (Thornton & Giglioli, 1965; Carlson *et al.*, 1983; Lin & Melville, 1992). However, this was not observed in our study and we consider that sulphate is probably being precipitated as sulphate minerals such as gypsum and jarosite. Acidic conditions may exist in such sediments through the presence of soluble chlorides and decaying organic matter.

Sediment buffering

Acid sulphate soils develop when the inherent buffering capacity of the sediments is insufficient to neutralise the increased acidity formed by the oxidation of iron pyrite (FeS_2). The main buffers and typical reactions which operate in mangrove sediments were given by Mohr *et al.* (1972):

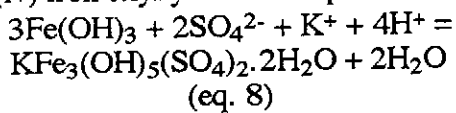


(iii) carbonate/gypsum



and,

(iv) iron oxyhydroxides/sulphates



Carbonate buffering operates in our modern mangrove sediments, where there is an excess of CaCO_3 over oxidisable sulphur. The sediments are buffered between pH 5 and pH 7 by the formation of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$. In the absence of carbonate, the sediments may be buffered down to pH 3.5 by the formation of jarosite. The pH can only be lowered further by complete dissolution of $\text{Fe}(\text{OH})_3$ and/or by oxidation of stable polysulphides (Mohr *et al.*, 1972; Thornton & Giglioli, 1965). In the sediments of the recently buried mangroves, the pH is only 2.5 which suggests that the latter reactions may be occurring. The pH may also be decreased by organic acids formed from the breakdown of plant material.

The relatively high concentrations of trace metals within the mangrove sediments reflects their fine-grained nature and low Eh. The high clay-mineral and oxyhydroxide contents of these mangrove muds not only reduces potential metal mobilisation and toxicity through adsorption and complexation but also significantly buffers the sediments (to pH 3), in contrast to mangrove peats. The overall concentration of metals in the modern and early Holocene mangrove sediments are similar, reflecting the relatively unpolluted nature of the modern mangroves.

In general, clay-rich organic sediments are more oxidising and less acidic than organic-rich peats (Senaratne *et al.*, 1990). The higher acidity of the organic-rich peats reduces plant decomposition and retards the action of sulphate-reducing bacteria (Berner, 1984), leading to a reduced total sulphur content (Bustin & Lowe, 1987; Senaratne *et al.*, 1990). However, it is not the abundance but rather the form of

sulphur that is the important factor in the formation of H_2SO_4 , and of these, non-stoichiometric Fe-sulphide (eg. greigite) and Mn-sulphide compounds and organic-bound sulphides are the most significant. It is likely that the mangrove sediments studied contain all forms of sulphur found in most peats, including pyritic, sulphate (mainly gypsum) and organic sulphur.

CONCLUSION

The clay-rich nature of the early Holocene and modern mangrove sediments of the central GBR region restricts the potential development of acid-sulphate soils. Clay minerals and oxyhydroxides effectively buffer the acidity developed through sulphate-reduction and organic matter degradation, and act to immobilise potential toxic metals. The more anoxic and acidic sediments of the recently buried mangroves (those buried within the last 200 yrs) highlight the active role mangroves themselves have on controlling sediment salinity and acidity.

Early Holocene mangrove sediments buried on the inner shelf of the central Great Barrier Reef are anoxic and acidic. Their porewaters are relatively enriched in Ca^{2+} and Sr^{2+} , and depleted in Mg^{2+} and K^+ , consistent with preservation by rapid burial (possibly following a vault in sea level) and with little subsequent exchange with seawater.

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