GEOCHEMICAL HETEROGENEITY OF A COASTAL MANGROVE WETLAND.

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ABSTRACT

Preliminary geochemical studies of intertidal mangrove sediments from Polaris Point, Guam (13° 30′N 144° 45′E) showed a compositional transition from a terrestrial to a marine environment. Seven mangrove core samples were taken in a transect across the mangrove swamp during the October wet season. Mineralogy and major (Si, Al, Fe, Mn, Mg, Ca, P) and minor element (Ga, Zn, Ni, Co, Mn, Cr, Pb, Rb, Sr, V,Y, Zr) distribution in the mangrove swamp were determined using techniques of X-ray diffraction (XRD) and X-ray fluorescence (XRF). Element concentrations largely reflect the sediment mineralogy which includes terrestrially-derived volcanic material, marine-derived carbonate and clay minerals. High concentrations of the metals Ni, Co, Mn and Cr are indicative of the basaltic source, although some residual contamination may exist from a previous oil spill. Movement of a subsurface salt-wedge over a 12-hour tidal period is thought to be reflected by a negative correlation between the tidal height (m) and groundwater salinity (ppt), and a positive correlation between groundwater pH, Eh (mV) and Mn concentration (ppm). These results suggest further investigation of the temporal and spatial variations of transition elements in the mangrove swamp, together with the causes of these variations, is warranted.

INTRODUCTION

The macro and micro-scale interaction of physical, chemical and biological factors has made the study of geochemical processes in mangrove wetlands difficult to decipher. Geochemical studies have focused mainly on the capacity of mangrove swamps to immobilise pollutants (Harbison 1984, 1986, Larcerda *et al.* 1988, Saenger *et al.* 1990), yet the pollutant levels are often measured against adjacent marine sediments and do not take into account background concentrations and variations within the mangrove sediments themselves.

A study was carried out in the mangrove swamp off Polaris Point in Guam during the wet season months of October and November. In order to gain some insight into geochemical variability within mangroves, redox conditions across the mangrove wetland were compared with the mineralogy. These data were used to characterise and compare the geochemistry of the mangrove flats, as a transitional environment, with the marine and terrestrial extremes.

STUDY AREA

Guam (13° 30′N 144° 45′E) is the only island of the Marianas to have mangrove forests. They form part of the prograding shoreline on the lagoon edge of what is now Apra Harbour (Figure 1). This study was conducted at the southern end of Apra Harbour, where the transition from the terrestrial to the marine edge occurs over a distance of about one kilometre.

On Guam, the climate is tropical with relative humidity rarely less than 60 percent and mean annual temperature near sea level around 26°C (Tracey *et al.* 1964). There are two distinct seasons, a dry season from January through May and a wet season from July to November. Mean annual rainfall ranges from 2100 to 3000mm. Droughts and cyclones are moderately common in Guam. Storms may bring winds up to 250km/h and 650mm rain in a 24 hour period. In the southern volcanic half of the island, runoff to the sea averages about 11m³/s, with numerous seeps scattered along the streams (Ward *et al.* 1964).

Draining the Alifan limestone and the basaltic volcanics of the Alutom formation, the Laguas and Agueda Rivers are the main sources of freshwater and terrestrial sediment to the mangrove wetland of Polaris Point (Figure 1). A
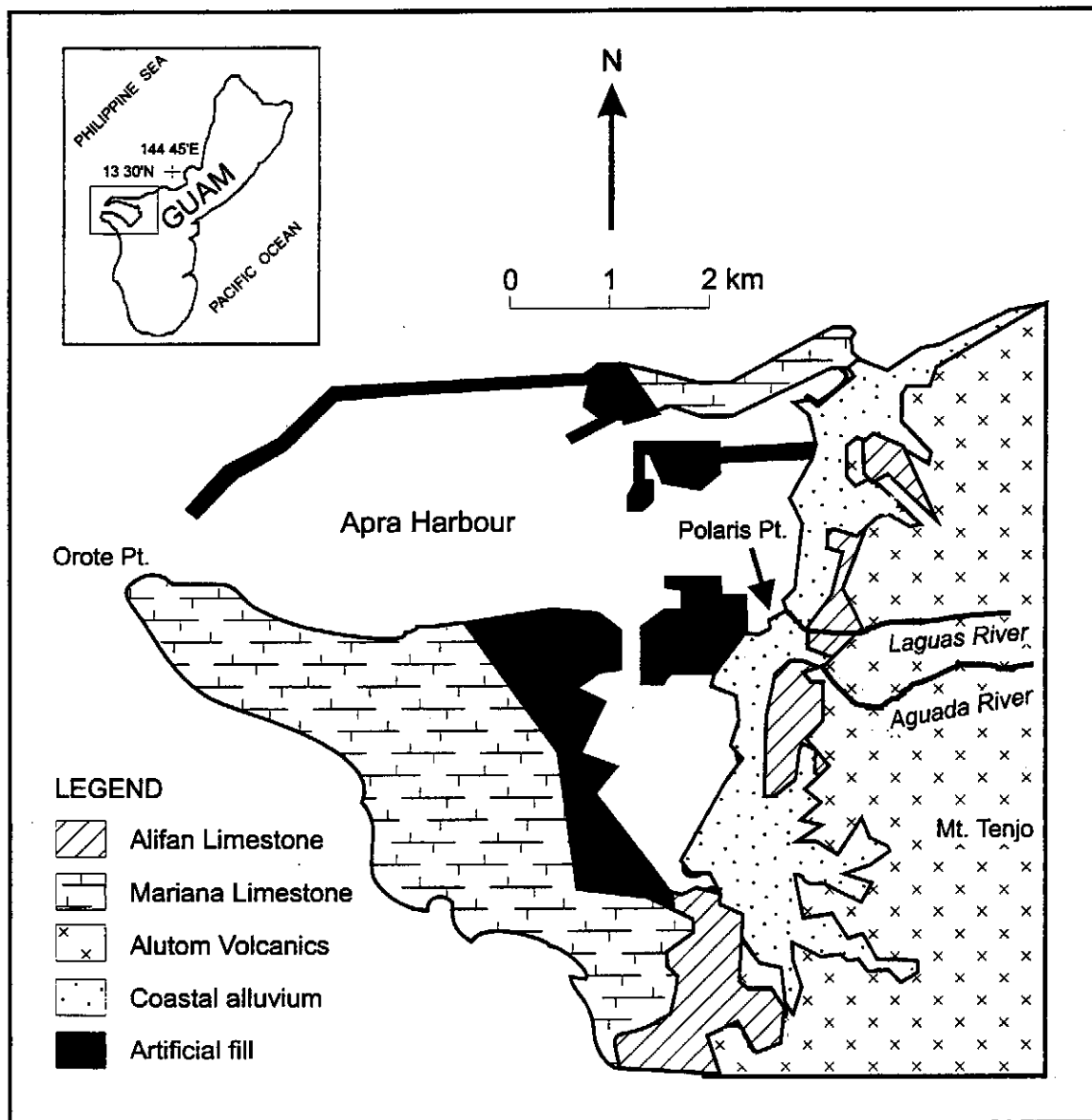
Figure 1 Location map and geology of study area.

tidal channel meanders across the wetland and a significant input of freshwater through the saturated zone maintains a saline front that extends to the seaward margin during the wet season. Average tidal range is estimated at 1.0m and complete inundation of the mangrove wetland only occurs at extreme high water (1.2m) or after heavy rain. Sediment within the mangrove swamp includes allochthonous material from both the seaward and terrestrial extremes, and autochthonous material formed under the prevailing conditions of the sedimentary environment.

Rhizophora sp. and *Avicennia* sp. generally grow in accordance with their increasing salinity tolerance. However, most of the mangal was artificially replanted after an oil spill in 1986 killed the previous mangrove forest. These mangroves exist in all except the emergent (+0.25m ASL) Holocene reef flat which is devoid of any vegetation. A diverse fauna, which includes crabs (*Uca* sp.), mudskippers (*Periopthalmus* sp.) and molluscs (*Gafrarium* sp., *Saccostrea* sp.), causes bioturbation of the sediment and affects sediment geochemistry.

METHODS

Polyethylene pipes (10 x 50cm) were used to take sediment core samples. In order to reduce contamination, the pipes were cleaned in HCl (conc.) and soaked in filtered (0.45μm) seawater for a week.

Seven cores were collected in an east-west (land-seaward) transect across the mangrove swamp at approximately 40m spacings to an average depth of 40cm. Core samples were stored at 4°C before being split and analysed. Petrographic descriptions were done at the Water and Energy Research Institute of the Western Pacific (WERI) at Guam University, according to standard soil methods (Berkman, 1989).

Sub-samples of sediment cores were analysed for mineralogy and geochemistry at the Geology Department of James Cook University, Australia. Mineralogy was determined from smear-mounted slides on a Rigaku Denki D2155 X-ray diffractometer (XRD) fitted with a post diffraction curved graphite crystal monachromator using CuKα. Major and minor element concentrations were determined from boron pellets and glass beads using a Siemen SRS 3000 X-ray fluorescence spectrometer (XPF). Major element concentrations are reported as oxides and minor elements as elements.

A Corning portable meter (battery-operated) was used to measure surface (0-2cm) sediment pH, Eh (mV) and temperature (°C), and an Aquafauna refractometer was used to determine surface water salinity. Measurements of groundwater pH, Eh (mV) and salinity (ppt) were measured hourly for 12 hours from a borehole situated approximately + 3.0m ASL at the eastern end of the mangrove swamp. Water samples were also collected over the 12 hour tidal cycle, using a 25ml plastic syringe fitted with a Swinex filter unit (0.2μm) and acidified with concentrated nitric acid (Analar). Filterable Mn was measured by Atomic Absorption Spectroscopy (AAS) on a Perkin Elmer model 560 within 24 hours of sampling.

RESULTS AND ANALYSIS

Qualitative sediment digestions with H_2O_2 and petrographic observation show that decaying organic matter from the present, and previous, modern mangrove forest comprises up to 80-90% of the sedimentary matter. In many of the cores, an horizon of oil-laden mangrove roots (10-15cm) overlies a layer of mottled grey silty-clay (15-20cm). In cores taken from the tidal creek and the seaward extreme, root matter is mixed with shelly-carbonate material. A surface horizon of silty-mud (< 63μm) is observed to decrease in thickness toward the seaward edge of the mangroves (Figure 2).
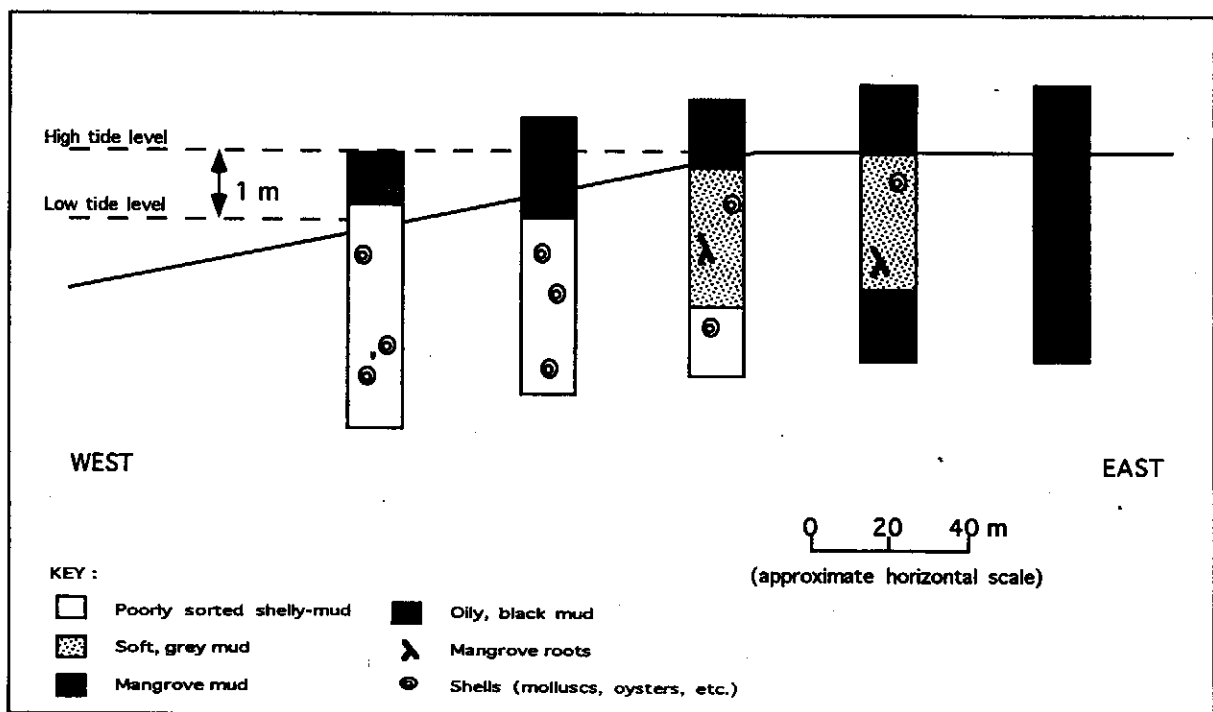

Figure 2 Schematic diagram of core stratigraphy in mangrove swamp at Polaris Point.

Combined results of petrographic observations and XRD analyses indicate that about 50% of the mineral fraction is quartz and 20% carbonate material. These carbonates (low magnesium calcite and aragonite) are most likely to be of marine/biogenic origin. The remaining fraction is made up of varying proportions of clay minerals and volcanogenic minerals (feldspar, biotite, pyroxene, olivine and heavy minerals). The clay mineral assemblage is dominated by smectite (d-spacing ~ 15 - 17.5Å) and mixed-layer clays (d-spacing ~ 29.10Å/15.25Å/7.60Å) with lesser amounts of hydrated kaolinite (halloysite) (d-spacing ~ 7.4Å). Pyrite (FeS_2) and jarosite ($KFe_3(OH)_6(SO_4)_2$) are also present in some of the clay rich sediments.

Eh-pH measurements show that these clay rich sediments (laden in some areas with residual oil from an oil spill in 1986) are quite reduced (pH 6.5, Eh -100 to -300mV) and contrast with the more oxidising conditions found in the shelly carbonate sediments within the tidal creek, Holocene reef and seaward margin (pH 7 -7.5, Eh 0 to + 100mV). Across the mangrove swamp, pH and Eh conditions change from pH 8 - 8.5 and + 400mV in the adjacent freshwater swamp, through pH 6.5 - 7 and -200 to -400 mV in the mangrove sediment, to pH 7.5 and -100 mV at the seaward edge. Salinity also increases across the mangrove swamp, ranging from zero at the landward (east) extreme to 33 - 35ppt at the seaward (west) extreme. Observations indicate that clay minerals accumulate at the edge of the mangroves in response to the increased salinity.

Measurements of groundwater over a 12-hour tidal cycle show a general trend of more oxidising conditions at high tide and less oxidising conditions at low tide (Figure 3). Manganese concentration also
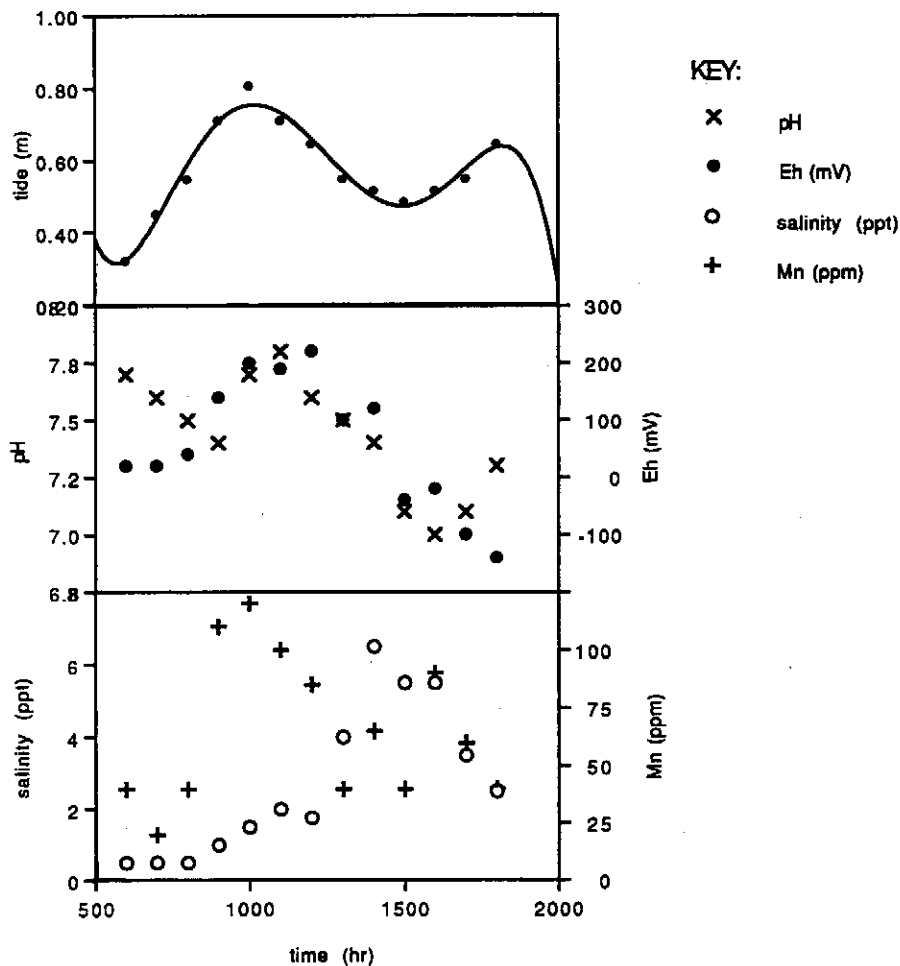

Figure 3 pH, Eh (mv), salinity (ppt), and Mn (ppm) measured over 12-hour tidal cycle.

shows a positive correlation with tide height ($r = 0.77$). The negative correlation between salinity and tide ($r = -0.17$) suggests there is a time-lag as saline water percolates the sediments at the landward end of the mangrove swamp.

Geochemical analyses reflect some chemical distinction of the adjacent terrestrial and marine sediments. Relative to volcanic material, mangrove sediments contain a high percentage of SiO_2 and lower percentage of metal oxides (Al_2O_3 Fe_2O_3, $TiO_2$and MgO) (Table 1). The concentration of CaO in the mangrove sediments is consistent with mixing of calcareous (marine) sediment in the nearshore environment.

Geochemical variations also reflect mineralogical variations within the mangrove swamp. For example, oxides of Al, Si, Na and K are greater in the clay mineral sediments accumulating between mangrove roots (core nos. 1 - 3, Table 1). A large percentage of CaO is found in sediments further seaward where a greater percentage of carbonate material is present

(core nos. 6 and 7, Table 1). From element correlations (Table 2), a negative association of 'terrestrial' (aluminosilicate) dominated elements (Al_2O_3, Fe_2O_3, MgO, Mn, Ti, Ga, Zr) with 'marine' (carbonate) elements (CaO, Sr) is evident. The strong negative correlation of CaO with MgO ($r = -0.93$) suggests MgO is not found in a carbonate phase (for example high magnesian calcite), but in a silicate phase such as olivine, pyroxene, biotite or their degraded products.

Elements with similar charge and ionic ratio should show geochemically coherent behaviour (Krauskopf 1967), and such behaviour is exemplified by CaO and Sr (correlation coefficient $r = 0.93$), K_2O and Rb ($r = 0.85$) and Fe_2O_3 and Mn ($r = 0.77$). However, there is a poor correlation of Al_2O_3 and the minor elements Ga ($r = 0.49$), Y ($r = 0.30$) and Zr ($r = 0.43$) which suggests that these elements are mainly associated with heavy mineral phases, such as zircon and apatite, rather than aluminosilicate (clay) or oxyhydroxide minerals. The transition

		CORE 1	CORE 2	CORE 3	CORE 4	CORE 5	CORE 6	CORE 7	BASALT*
SiO2		89.38	87.92	86.17	86.02	86.17	83.95	81.82	50.61
Al2O3		4.18	7.03	5.63	2.03	5.92	3.5	5.11	17.60
TiO2		0.19	0.18	0.14	0.09	0.14	0.03	0.12	0.55
Fe2O3		2.4	2.43	1.79	1.16	0.18	0.17	1.14	8.79
MnO	MAJOR	0.02	0.02	0.03	0.03	0.03	0.01	0.01	0.15
MgO		0.62	0.76	0.67	0.57	0.77	0.16	0.38	5.31
CaO		0.52	0.95	3.86	6.59	1.36	10.89	5.96	10.21
K2O		0.26	0.33	0.21	0.09	0.22	0.05	0.1	0.36
Na2O		0.32	0.53	0.62	0.03	0.61	0.13	— —	2.10
P2O5		0.02	0.07	0.09	0.02	0.09	0.08	0.08	0.08
Ga		58	52	42	— —	— —	20	36	— —
Zn		232	266	276	— —	— —	45	301	— —
Ni		132	136	108	— —	— —	25	107	24
Co		90	110	115	— —	— —	6	46	— —
Mn	MINOR	1933	1857	2669	— —	— —	528	876	— —
Cr		712	1096	1266	— —	— —	148	436	100
Pb		12	32	28	— —	— —	0	436	— —
Rb		107	96	90	— —	— —	17	73	8
Sr		960	768	1620	— —	— —	22264	5773	145
V		688	744	568	— —	— —	142	305	— —
Y		160	110	185	— —	— —	44	65	28
Zr		440	358	329	— —	— —	0	23	51

Table 1   Major element (wt%) and minor element (ppm) chemistry of mangrove swamp sediments. Cores are numbered according to an east (land) to west (sea) transect (Reagan and Meijer 1984).

	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	K_2O	Na_2O	P_2O_5	Ga	Zn	Ni	Co	Mn	Cr	V	Pb	Rb	Sr	Y	Zr
SiO_2	1																			
Al_2O_3	0.24	1																		
Fe_2O_3	0.8	0.63	1																	
MgO	0.72	0.77	0.95	1																
CaO	-0.8	-0.6	-1	-0.9	1															
K_2O	0.84	0.7	0.96	0.95	-0.9	1														
Na_2O	0.69	0.61	0.65	0.82	-0.6	0.76	1													
P_2O_5	-0.7	0.27	-0.5	-0.3	0.56	-0.4	0.03	1												
Ga	0.8	0.49	0.98	0.88	-1	0.9	0.53	-0.7	1											
Zn	0.13	0.69	0.69	0.72	-0.7	0.54	0.31	0	0.66	1										
Ni	0.56	0.67	0.94	0.89	-1	0.84	0.47	-0.4	0.94	0.87	1									
Co	0.69	0.73	0.91	0.98	-0.9	0.89	0.86	-0.2	0.83	0.72	0.85	1								
Mn	0.67	0.52	0.77	0.88	-0.7	0.75	0.9	-0.1	0.7	0.58	0.67	0.94	1							
Cr	0.57	0.77	0.78	0.93	-0.7	0.8	0.92	0.06	0.66	0.66	0.71	0.97	0.94	1						
V	0.85	0.65	0.99	0.97	-1	0.99	0.75	-0.5	0.95	0.61	0.89	0.93	0.81	0.82	1					
Pb	-0.7	0.07	-0.2	-0.3	0.15	-0.4	-0.6	0.26	-0.2	0.47	0.12	-0.3	-0.4	-0.3	-0.4	1				
Rb	0.65	0.58	0.96	0.91	-1	0.85	0.55	-0.5	0.96	0.83	0.98	0.89	0.78	0.76	0.91	0	1			
Sr	-0.5	-0.7	-0.9	-0.9	0.93	-0.8	-0.6	0.32	-0.9	-0.9	-1	-0.9	-0.8	-0.8	-0.9	-0.1	-1	1		
Y	0.7	0.3	0.74	0.79	-0.7	0.66	0.79	-0.3	0.71	0.51	0.63	0.86	0.96	0.82	0.76	-0.4	0.77	-0.8	1	
Zr	0.94	0.43	0.92	0.89	-0.9	0.91	0.78	-0.6	0.91	0.45	0.76	0.88	0.86	0.78	0.95	-0.5	0.85	-0.8	0.87	1

Table 2 Correlation matrix for seven sediment samples (r = 0.58 at 95% significance level).

metals (Zn, Ni, Co, Cr, V) show a high correlation with SiO_2, Al_2O_3, Fe_2O_3 and Mn (Table 2) which suggests that these minor elements are present within primary mineral phases as well as detrital clay and oxyhyroxide phases of the mangrove sediments. However, Pb does not show any significant correlation with any major element (Table 2).

A comparison of transition metals in the mangroves of Polaris Point with those reported from other sites (Table 3) shows concentrations up to ten times higher, although the results are much more variable than those reported from other mangroves (Larceda & Abrao 1984, Cook & Mayo 1980, Ward 1994). Basaltic volcanics typically have higher concentrations of transition metals than granitic or well-weathered sedimentary material (Krauskopf 1967). The mangrove flat is therefore predicted to have relatively higher levels of transition metals than the granitic areas where other studies have been based. The variability may also be a function of both the low numbers of core samples taken as well as the heterogeneity across the wetland.

DISCUSSION

Studies in Australia (Wolanski & Gardiner 1981, Wolanski & Ridd 1986) have shown how mangrove swamps control the flushing of freshwater material into the open sea, while simultaneously trapping marine material in their low energy areas. However, the coastal mangrove delta of Apra Harbour has not yet formed a complex drainage system and terrestrial material draining from the volcanic-limestone hills dominates over sediment brought in by the tide. Studies by Matson (1986) indicate that coastal sediments of Guam are impacted by different terrestrial runoff processes including discharge of aquifers into the intertidal zone and conservation of river load nearshore. The high energy and volume of flushing may account for the poor accumulation of clay size minerals and the high percentage of organic matter in comparison to other mangrove wetlands. Furthermore, the minerals present in the wetlands are dominated by fresh, and relatively unweathered, terrestrial minerals and this will clearly control the chemical composition determined by XRF.

In the alternating wet-dry climate, argillaceous-volcanic material would undergo rapid processes of reaction and removal involving dissolution, diffusion and mass transfer. Some unweathered material (olivine, pyroxene, amphibole) is likely to be transported directly into the mangroves by runoff, especially during the

LOCATION		Cr	Zn	Mn	Co	Pb	Ni
Polaris Point	x	632	209	1328	73	72	100
	sd	467.8	124.6	497.6	55.3	39.9	60.5
Sepetiba Bay[1]	x	13	58	256	5	14	8
	sd	1.1	7.0	15.5	0.3	0.9	0.4
Broad Sound[2]	x	65	41	449	12	13	23
	sd	17.8	14.3	448.8	12.2	13.1	23.4
Townsville[3]	x	47	36	408	8	18	12
	sd	13.5	8.8	163.4	2.7	5.1	4.1

Table 3   Comparison of heavy metals (ppm) in mangrove sediments.
(1. Larcerda & Abrao 1984, 2. Cook & Mayo 1980, 3. Ward 1994)

wet season. The high percentage of organic matter within the mangrove sediments may enhance or decrease the stability of certain minerals, such as quartz and calc-magnesium silicates, as a result of redox processes and an increase in H^+ activity (Cuff pers. comm. 1992).

Organics can also control the sedimentary cycling of trace metals directly, through complexation, or indirectly by providing a reducing capacity through sulphate reduction which assists redox reactions. In addition to the metals released from organic matter through sulphate reduction, the released anions HS^- and HCO_3^- can also precipitate authigenic mineral phases which are capable of absorbing or co-precipitating metals.

Studies have shown that the high content of organic matter and reducing conditions favour the accumulation of metals within (naturally occurring) sulphides in the mangrove sediments (Larceda & Abrao 1984, Harbison 1986, Yuudan et al. 1988). Heavy metals may also be derived from rusting war refuse dumped in Apra Harbour or from residual oil in the mangroves (P.B.E.C. 1982). The lack of correlation between Pb and SiO_2, Al_2O_3 or CaO suggests that Pb may be derived from a non-geological source, such as oil or batteries.

The neutral condition of the mangrove sediments is not unusual because most flooded soils are buffered by oxides as well as carbonate minerals, organic matter, clay minerals (Boto 1981), gypsum and jarosite (Mohr et al. 1972). However, unlike many coastal mangroves, there is a significant groundwater input via limestone rivers which would also provide a HCO_3 buffer to the mangrove sediment. Even small pH changes may increase the mobility of some elements resulting in complex reactions of absorption or isomorphous substitution with organic calcareous, aluminosilicate and oxyhydroxide phases.

During the dry season, more acidic conditions may eventuate from the oxidation of sulphides (e.g. pyrite) and elemental sulphur, forming H^+ and SO_4^{2-} ions. However, the pH may be buffered by the formation of gypsum or jarosite (Mohr et al. 1972). In addition, oxidation and reduction reactions of redox couples are not mutually exclusive, but may occur simultaneously within different micro environments (Lord & Church 1983), so that pyrite and jarosite may occur together in adjacent sedimentary horizons. Jarosite was also a characteristic feature of oxidised mangrove soils in Botany Bay, Australia, where the concentration of sulphate was considered to reflect the intensity of drainage (Lin & Melville 1992).

Manganese originates as Mn^{2+} in silicate minerals and as Mn^{4+} in relatively insoluble oxide minerals in volcanic bedrock and overlying limestones of south Guam. These minerals are normally liberated from bedrock by weathering and erosion, transported downstream and eventually deposited along the coast. The low redox, low pH and elevated bacterial activity in the mangrove swamp favours the transformation of manganese oxides to hydrated Mn^{2+} ions and organo-

complexes, and would account for the seaward decrease in both Fe and Mn.

The more oxidising conditions at high tide and less oxidising conditions at low tide are consistent with other studies (Harbison 1986, Larcerda *et al.* 1988), and should promote the oxidation of Mn^{2+} to Mn^{4+} (Harbison 1986, Larcerda *et al.* 1988). However, the dissolved concentration of manganese increases with rising tide. Furthermore, the increase in pH, Eh (mV) and manganese is not coincident with an increase in salinity. These results suggest that as the tide rises, a wedge of saline water diffuses through the mangrove swamp increasing the ratio of oxidising to reducing conditions within the sediment, and resulting in apparent increases in redox potential as well as the concentration of dissolved manganese.

Considerable changes in the concentration of dissolved Fe and Mn may also occur within millimetre distances of the mangrove roots (Clough pers comm. 1992). Combined with small changes in pH and salts, this small scale variation may be expected to compound over larger scales. Although many elements (some of which may be considered as nutrients e.g. Fe, Mn, Zn, Co) may be influenced by chemosynthetic processes, these are often masked by mineralogical factors and compounded by bioturbation within the mangrove sediments of Guam.

From these results, it would appear that the major elements (Si, Al, Ti, Fe, Mg, K, Na and Ca) are better indicators than minor elements of freshwater and marine influences. For example, the observed tendency for clay minerals to accumulate between the mangrove roots and at the freshwater-saline boundary (probably due to flocculation) is reflected by a greater concentration of Al, Na and K but a decrease in Ca within the sediments. High concentrations of silica and iron oxide together with low levels of aluminium favour the formation of sodium-rich smectite in marine mud (Garrels 1984) and is consistent with both the XRD and XRF geochemical results.

A dominance of smectite, hydrated kaolinite and smectite-kaolinite mixed-layer clays in the mangrove sediments is consistent with the findings of Carroll and Hathaway (1963), who reported halloysite, montmorillonite (smectite) and mixed-layer clays as the main clay mineral constituents on Guam. Halloysite may have formed either via hydration of kaolinite or from the transformation of 2:1 clay mineral phases, such as smectite.

The high concentration of SiO_2, especially as quartz, is unusual in a basaltic environment. In the mangrove swamps of Senegal, an enrichment of SiO_2 was due to dissolution of quartz and clay (smectite) together with the precipitation of amorphous silica involving the following mineral-solution equilibria (Marius 1985):

$$SiO_{2(s)} + 2H_2O = Si(OH)_{4(aq)}$$
$$K_{eq} = 1 \times 10^{-4} \ (25°C)$$
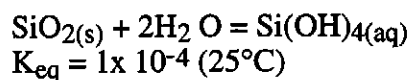
Although this process is possible, the concentration of quartz in the mangroves of Guam is more likely to be derived from undiscovered quartz veins, or from the marine environment.

In general, the fresh unweathered sedimentary material, and greater variability in concentration of minor elements compared to major elements within the mangrove sediments of Guam, suggests that element distribution is dominated by mineralogy and transport rather than transformation and other syngenetic geochemical processes. As in other mangrove swamps (Ward 1994), the influence of colloids is not great enough to be detectable over the more substantive influence of mineralogical factors.

The results of this study only provide an initial indication of the temporal and spatial variability present in the coastal mangrove wetland of Guam. Further studies, including grain size analysis and sequential extraction, are required to obtain a better understanding of the controls of element distribution in the mangrove sediments. In addition, hydrological and diagenetic processes should be determined in both wet and dry seasons, as seasonal shifts in the water table are likely to influence the spatial and the stratigraphic distribution of many metals.

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