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Impacts of acid leachate on water quality and fisheries resources of a coastal creek in northern Australia

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Abstract. Reclamation of ~700 ha of mostly tidal wetlands in north Queensland in the early 1970s disturbed potential acid sulfate soils resulting in acid leaching into coastal waterways. The wetlands had been isolated from tidal inundation through the construction of a levee with tidal gates on the major creeks. Acid leachate caused the pH values in impounded, freshwater reaches of Firewood Creek to drop to <4 for much of the year and increased total dissolved aluminium and iron concentrations. Flood rains caused large volumes of acid water from the creek to discharge into its tidal reaches, resulting in environmental problems including episodic fish kills. Factors contributing to the fish mortalities included a temporary reduction of pH, elevated iron and aluminium concentrations and occasional low dissolved-oxygen concentrations. Fish kills were observed on three occasions in three years but did not occur every time acid water was discharged. Fish species diversity in the tidal reaches also declined during the wet season and this correlated with high concentrations of dissolved aluminium. Although episodic flood events resulted in the acidification of the tidal reaches of Firewood Creek, no major deleterious effects on the main estuary were found. Rehabilitation options for this site include restoration of full or partial tidal flooding.

Introduction

Acid sulfate soils develop as a result of the drainage of parent materials that are rich in pyrite (Dent 1986). When exposed to air, pyrite is oxidized to produce sulfuric acid (Dent 1986). In Australia, major soil acidification occurs in Holocene estuarine sediments of coastal floodplains, and the drainage from acid sulfate soils contained in these sediments is the major cause of stream acidification (Sammut *et al.* 1995). Although acidification can occur naturally (Brown *et al.* 1983), Lin *et al.* (1995) maintain that most increases in the discharge of acidic oxidation products into rivers and estuaries are the direct result of land reclamation and drainage. In areas where potential acid sulfate soils exist, soil excavation and land drainage can often promote the oxidation and hydrolysis of pyrite and subsequent transportation of acid drainage into waterways.

In lakes and streams, some fish species begin to show stress from elevated hydrogen ion concentrations when pH levels decline below 6.5; however, for most species the critical pH level is between 5.0 and 6.0 (Brocksen *et al.* 1992). The sulfuric acid produced can further react with a variety of minerals in the sediments to increase soluble concentrations of pH-dependent elements, for example iron and aluminium, to levels that are toxic to biota (Lin *et al.*

1995). Dissolved aluminium can have deleterious effects on aquatic biota, particularly fish under acid stress (e.g. Driscoll et al. 1980; Ramamoorthy 1988; Brocksen et al. 1992; Sammut et al. 1995); Driscoll et al. (1980) suggested that inorganic aluminium forms were the major species of concern. Callinan et al. (1993) and Sammut et al. (1996) have attributed injury and death of fishes to high concentrations of inorganic, cationic aluminium species in naturally acidified waters. Dent (1986) notes that direct toxicity by dissolved aluminium is greatest in fresh and brackish waters that are weakly buffered. Callinan et al. (1993) recorded pulses of acidic, deoxygenated water that originate from acid sulfate areas during the early stages of most major rainfall events, and they speculated that the temporal association between reductions in pH and dissolved oxygen concentrations might be a result of oxidation of ferrous iron to ferrihydrite.

Fish exposed to waters with elevated hydrogen ion concentrations and toxic levels of pH-dependent elements can exhibit a variety of pathologies. Laboratory studies suggest that fish that inhabit waters with elevated acid and aluminium levels and low calcium concentrations can exhibit severe disruption of internal body-ion balances and/or severe respiratory stress (Reader and Dempsey 1988); either response can be lethal. High calcium concentrations generally reduce mortality and sublethal physiological disturbances associated with low pH or with various trace elements (Reader and Dempsey 1988). Sammut et al. (1995) suggest that acidification of downstream reaches can result in displacement of fish into other river locations, which interferes with recruitment and dispersion processes, and increases pressure on remaining food resources and shelter. They also contend that reproduction could be adversely affected through loss of spawning sites and that interference with downstream and return migration could lead to reduction in populations of estuarine-spawning freshwater fish upstream of acid sulfate soil areas. Hyne and Wilson (1997) concluded that exposure of the early life stages of Australian bass to acid sulfate leachate could result in significant mortalities. Recruitment failure may be one of the first and most apparent responses of fish to acidification (Brocksen et al. 1992), and some laboratory studies (e.g. Mount et al. 1988a, 1988b) suggest that young fish have higher sensitivities to acid and aluminium toxicity.

In coastal aquaculture operations, the consequences of acid water runoff from pond dykes after rainfall range from chronically low yields to extensive kills of culture organisms (Simpson and Pedini 1985). Commercial offshore fisheries may be seriously affected by disturbance of potential acid sulfate soils (Dent 1986), and Callinan *et al.* (1993) suggested that epizootic ulcerative syndrome, a fish disease linked to acid runoff, could threaten aquaculture and fisheries in the Asia-Pacific region. In northern New South Wales, the construction of extensive drainage canals for flood mitigation has resulted in the episodic discharges of acid water into tidal areas. These discharges are thought to be responsible for extensive fish kills and outbreaks of red spot disease (Virgona 1992).

Other water-quality attributes, such as high concentrations of calcium and silicon, can lessen the toxicity of inorganic aluminium to fish (Brocksen et al. 1992; Brown et al. 1983, Callinan et al. 1993). The release of pyrite oxidation products into estuarine waterways in the Pearl River Delta region of southern China does not appear to cause fish kills probably because of dilution from large fresh water discharges and control of acid output through water management practices (Lin et al. 1995). In response to toxic acid/aluminium exposure, some fish species are able to acclimate physiologically or undergo longer-term adaptation by genetic selection (Mount et al. 1990). Communities of acidophilic biota can become established in streams where acidification has persisted (Brocksen et al. 1992).

This paper describes an investigation into seasonal changes in pH, dissolved oxygen, total monomeric aluminium and iron in coastal watercourses that are affected by acid drainage around Cairns, Australia. We also present evidence linking fish kills and changes in abundance and diversity of fish and mud crabs (*Scylla serrata*) in a tidal



Fig. 1. Study sites in Trinity Inlet. Water quality stations are labelled 1–12. Bottom inset: biological sampling sites in Firewood Creek. S prefix, seine netting sites; G prefix, gill-net sites; crab pots were set in the cross-hatched area.

creek to episodic discharges of acid water and discuss reinstating tidal flooding as a rehabilitation option for the area.

Methods

The study was undertaken in two consecutive phases. During the first phase (September 1994 to June 1996), detailed surveys were undertaken of the water quality at 12 sites in the Firewood Creek and Magazine Creek catchments and in Trinity Inlet adjacent to the mouth of Firewood Creek. In addition, observations of fish kills occurring in these catchments were recorded. In the second phase, from October 1997 to October 1998, measurements were made of temporal changes in the abundance and diversity of fish and the abundance of mud crab populations in the tidal section of Firewood Creek and from control sites elsewhere in Trinity Inlet. Water quality measurements were also made in association with this biological sampling.

Study sites

The study sites (Fig. 1) were on Trinity Inlet in north Queensland, a relatively deep estuary opening into a shallow bay that forms the port for the city of Cairns ($16^{\circ}56'S$, $145^{\circ}46'E$). It has maximum tidal amplitude of ~3.2 m, and rainfall in the area is seasonal with most falling during the hot summer months (Fig. 2). Daily rainfall data were obtained from the Australian Bureau of Meteorology for the Cairns



Fig. 2. Surface water pH, salinity and daily rainfall (mm) at sites inside Firewood Creek (Sites 4, 7), Magazine Creek (Site 1) and in Trinity Inlet (Sites 10–12), September 1994 to June 1996; \triangle , surface pH; \blacklozenge , surface salinity; solid bars represent daily rainfall recorded at the Cairns airport.

Airport meteorological station, ~8 km north-west of the mouth of Firewood Creek; data for seven days prior to each biological survey were pooled for analyses.

Trinity Inlet has no significant permanent freshwater inflow but does have extensive tidal wetlands that are drained by numerous tidal creeks. In the early 1970s, >700 ha (~14% of the total wetlands of Trinity Inlet) were isolated from tidal inundation by levees and tidal gates. Tidal gates were installed across the two major tidal creeks (Hills and Firewood Creeks) draining the site to prevent infiltration of salt water, and a third creek (Magazine Creek) was completely blocked. Although most of the area remains undeveloped, the levees and tidal gates remain. The site was purchased in 2000 by the Queensland State Government which is developing a management plan for the area. Hicks et al. (1999) estimated that 110 ha of the site was severely acidified and that it had produced 1.5×10^9 moles H⁺ since drainage in the 1970s. Using the Australian soil taxonomy system (Isbell 1996), they classified the soils at their drained study sites as either sulfuric extratidal hydrosol or sulfuric redoxic hydrosol. The soil textures at those sites were silty, clay loams in the top 30-60 cm of the profile and medium clays below them to 400 cm deep. Height differences between adjacent sites inside and outside the levee recorded in November 1995 suggested some shrinkage of the soils in the impounded area (Russell, unpublished).

Water quality

Water quality samples including determinations *in situ* of pH, dissolved oxygen, conductivity and salinity were taken from sites inside the levee,

the tidal section of Firewood Creek and Trinity Inlet ~100 m upstream and downstream of its confluence with Firewood Creek. Water samples were also taken inside the mouth of Magazine Creek and in Falls Creek. One litre surface (top 20 cm) water samples and surface physiochemical (pH, dissolved oxygen, temperature, salinity, conductivity and turbidity) measurements were taken at all twelve sites. Additional bottom water samples (~30 cm off the bottom) and physiochemical measurements were taken at Sites 8 and 9 in the tidal reaches of Firewood Creek. To determine the full effect of water draining from the impounded area, samples were taken as near to low tide as practical. From 28 September 1994 to 11 February 1995, sampling frequency was approximately once every two weeks. The first discharge from the impounded section of Firewood Creek in the 1994/95 wet season resulted from heavy rainfall during 11-18 February 1995. To assess its effect on the water quality of the tidal section of Firewood Creek, the sampling frequency was increased to every low tide for this period. This frequency then decreased to one sample per day until 4 March 1995, reverted to once every two weeks until 29 September 1995 and reduced to monthly sampling until 26 June 1996 (although a small number of extra samples were taken in February and March 1996 to correspond with seasonal monsoonal rains). Physicochemical measurements were taken for Firewood and Seelee/Falls Creeks in conjunction with the biological sampling between October 1997 and October 1998.

Water samples were initially chilled on ice before being frozen and stored prior to laboratory analyses. A Horiba model U10 meter measured pH, temperature, dissolved oxygen, salinity, conductivity and turbidity. It was auto-calibrated with a standard phthalate pH solution before each sampling event. The modified Winkler method (American Public Health Association 1992) was used to confirm the accuracy of dissolved oxygen determinations made with the Horiba probe and was also used when the meter was malfunctioning. A Yeocal Model 606-SDL data logger was used to obtain hourly measurements of pH, conductivity, depth and dissolved oxygen during critical peak flow periods in March 1995. Before use, the pH probe was calibrated with buffer solutions (pH 4 and pH 10) and the dissolved oxygen sensor was calibrated against saturated distilled water. Total monomeric aluminium concentrations were determined by the catechol-violet method and with a Lachat QuickChem flow-injection auto-analyser equipped with a Teflon sampler probe on samples which were filtered to 0.1 μ m. The calibration range was between 0.003 and 0.3 mg L⁻¹, and a 1000 mg L⁻¹ stock standard aluminium solution (Australian Chemical Reagents) was used to make up a set of six working standards from 10 to 300 μ g L⁻¹. As a quality control, standard additions (0.1 mg L⁻¹ spike) and working standards (25 µg L⁻¹) were analysed approximately every 20 samples. For the iron analyses, samples were first filtered through combusted, acid-washed GFF filters (0.7 µm pore size) and then analysed with a Varian Spectra AA600 atomic absorption spectrophotometer. The limit of detection for dissolved iron was 0.2 mg L⁻¹. Spot samples were taken at selected sites (both impounded and tidal) for analysis of calcium and sulfate concentrations under wet-season and dry-season conditions. Calcium and sulfate were determined by inductively coupled plasma atomic-emission spectrometry with detection limits of 2.0 and 0.2 mg L⁻¹ respectively. On 17 February 1995 (wet season) samples were taken from Sites 4-6, 8, 10 and 11, and on 26 October 1995 (dry season) samples were taken at Sites 6-8. Spot samples for nitrate analysis were taken at Site 4 (n = 13), Site 7 (n = 8) Site 8 (n = 16), Site 9 (n = 16), Site 10 (n = 11), Site 11 (n = 11) and Site 12 (n = 9) between September 1994 and March 1996. Nitrate (NO₃-T) concentrations were determined by a continuous-flow analyser using the Griess-Ilosvay reaction. Nitrate was first converted to nitrite-N with a cadmium column. The nitrite-N was coupled with N-(1-naphthyl) ethylenediamine dihydrochloride to form a red-purple dye which was proportional to the concentration of nitrite-N. Limit of detection for nitrate was 0.001 mg L^{-1} .

Biological sampling

Monthly fish surveys were conducted in Firewood Creek and Seelee/Falls Creeks between October 1997 and October 1998, and crab surveys were undertaken from October 1997 to September 1998. Selee Creek (Fig. 1) was initially selected as a control site in October 1997, but to avoid any effects from a nearby aquaculture facility it was replaced by Falls Creek in subsequent surveys. To standardize biological sampling procedures, surveys were conducted around the same lunar (first quarter) and tidal phase.

Seine netting was undertaken at low tide at three sites in Firewood Creek (S1, S2 and S3, Fig. 1) and at the mouth of Falls Creek and 500 m upstream. Single sweep samples using a 25 m by 25 mm stretched mesh (SM) monofilament net were taken between October 1997 and October 1998. Initially, there were also two control sites in Falls Creek, but sampling at this site was discontinued in January 1998 after a large estuarine crocodile was sighted adjacent to the sampling sites. Common species in the seine net hauls were measured *in situ* and released, and the others were returned to the laboratory for identification.

Larger fish were sampled with monofilament gill-nets set in the tidal sections of Firewood (Sites G1, G2, Fig. 1) and Falls Creek. Gill-net surveys were conducted in Firewood Creek from October 1997 to October 1998 and in Falls Creek from November 1997 to October 1998. Three monofilament gill-nets were used, of 50 mm SM (33 m length, 33 mesh drop and 14 ply line), 100 mm SM (33 m length, 33

mesh drop and 24 ply line) and 150 mm SM (33 m length, 33 mesh drop and 50 ply line). Each net was set up to 1.5 h before dusk and checked hourly until after the tidal change or when there was no longer sufficient water depth for the net to fish effectively or for the nets to be checked and retrieved. Soak time (period that the net was fishing) varied from 2.17 h to 6.5 h between months and sites depending on the tidal regime and site topography. In addition, on two occasions, small mesh (75 mm and 100 mm SM) nets were set overnight in Firewood Creek upstream of the levee. All fish were identified and measured before being released. Where identification was uncertain, specimens were kept for more detailed taxonomic studies. As netting effort varied between months and sites because of tidal regime and topography, fish catches were standardized to the number of hours fished. Monthly catch per unit effort (total number of fish caught per hour fished) was calculated for the gill-net surveys for each month by pooling the effort for all nets. Ten circular collapsible Munyana crab pots (0.75 m diameter, 60 mm SM) were set in both Firewood and Falls Creeks (see inset, Fig. 1) for ~24 h. All pots were checked twice during each 24 h cycle. All crabs were identified, measured (carapace width), sexed and tagged before being released. Catch per unit effort was calculated as the number of crabs caught per pot per lift.

Cast-net samples were taken in a monofilament cast net (2.4 m diameter, 25 mm stretched-mesh) at low tide. Up to ten casts were made in both Firewood and Falls Creeks and samples from each creek were pooled and returned to the laboratory for identification and measurement. Effort for these cast-net surveys could not be standardized, and data from this sampling method were used only to further describe the fish communities in each creek system.

Samples of recently dead or moribund fish from a fish kill on 19 January 1996 were fixed in 4% buffered formaldehyde and sent to the Oonoonba Pathology Laboratory, Townsville, for histological examination. Fixed tissues were dissected and processed routinely as 4 μ m sections then stained with haematoxylin and eosin prior to light microscopy.

Analyses

The Genstat® statistical package was used to develop multiple stepwise regression models to relate catch per unit effort and species diversity to the environmental parameters site, season, rainfall (daily and total for seven days prior to sampling), pH, salinity, temperature, turbidity, dissolved oxygen, conductivity and dissolved aluminium concentration. Seine-net catch data were normalized (log_e(catch+1)) before being used in the model. Since species diversity is a useful indicator of adverse water quality conditions in higher trophic levels of an estuary (Bechtel and Copeland 1970), species diversity was calculated from the Shannon–Weaver formula, $H' = -\sum N_i / N \log_e(N_i / N)$, where N_i is the number of the *i*th species in the collection, N is the total number of individuals in the sample and H' is the diversity of the sample. Separate species diversities were calculated for the seine-net samples and the gill-net samples. A paired t-test was used to test for significant differences in the gill-net catch rates between the impact and control sites.

Results

Water quality

Impounded waters of Firewood and Magazine Creeks

Time-series data on total dissolved aluminium, iron and dissolved oxygen were recorded at six sites, but in this paper these data are presented for only three representative sites: Site 1 (Magazine Creek), Site 7 (upper Firewood Creek) and Site 4 (lower Firewood Creek).



Fig. 3. Total monomeric aluminium concentrations (mg L^{-1}) in surface waters from Magazine (Site 1) and Firewood (Sites 4 and 7) creeks and in Trinity Inlet (Sites 10–12), December 1994 to March 1996.

The wet season of 1994/95 was delayed and significant rains did not fall until February 1995. The rainfall lowered salinities at all sites and caused the release of acid leachate resulting in a pronounced decline in pH at Site 4 (Fig. 2). After heavy rains, water overflowed into the Firewood Creek catchment from the nearby Hills Creek. The rugged, relatively pristine upper catchment of Hills Creek was not affected by acid leachate and its discharge into Firewood Creek resulted in an increase in pH at Site 7. An increase in the height of the water table could also have resulted in a localised temporary reduction in the amount of acid being produced. Although there were some fluctuations in salinity, the pH generally remained low at the two impounded Firewood Creek sites for the rest of the study (Fig. 2).

In the impounded waters of Magazine Creek (Site 1), the salinity increased during the dry season to a maximum of about 20 with the pH generally between 2.5 and 4 and similar to those recorded at sites in the adjacent Firewood Creek (Fig. 2). As the levee prevents any tidal intrusion into the impounded part of Magazine Creek, salinity changes were most likely the result of salt water infiltration through the levee. In the wet season, salinity fell as a result of rainfall or freshwater inflow but there was a corresponding rise in the pH to about 7 (Fig. 2). This is largely the reverse of the relationship at Site 4; perhaps the pH of the runoff from the

catchment of Magazine Creek was relatively high and acted to dilute receiving waters sufficiently to cause a rise in the pH.

Aluminium concentrations also appeared to be related to rainfall events within the catchment (Fig. 3). At Site 4, values dropped during February 1995 from >31 mg L⁻¹ to 0.05 mg L⁻¹ in less than a week. During this same period >460 mm of rain fell and pH increased suggesting dilution of the impounded surface waters. Some extremely high values, occasionally >70 mg L⁻¹ were recorded at Site 7.

Iron concentrations (Fig. 4) showed a similar pattern to the dissolved aluminium concentrations. At Site 1, there was a general increase during late 1994 and early 1995 but concentrations were generally relatively low throughout the remainder of the year. Concentrations at Sites 4 and 7 increased during February 1995 as a result of seasonal rains to 62 mg L⁻¹ and 186 mg L⁻¹ respectively (Fig. 4) then fluctuated considerably throughout the remainder of 1995.

Dissolved oxygen concentrations at Sites 1, 4 and 7 are shown in Fig. 5. At Site 1, most observations were between 60% and 100% saturation with the exception of one sample taken in January 1995 that was 17%. At Site 4, most dissolved oxygen observations were >60% except during February and March 1995 (Fig. 5). Several samples were supersaturated (Fig. 5), possibly as a result of photosynthetic activity by aquatic macrophytes and algae. At Site 7 samples



Fig. 4. Iron concentrations (mg L^{-1}) in Magazine (Site 1) and Firewood (Sites 4 and 7) creeks and in Trinity Inlet (Sites 10–12), September 1994 to December 1995.

with low dissolved oxygen concentrations were recorded in January 1995 and in the period July to September 1995.

In February 1995, spot water samples from Sites 4, 5 and 6 had calcium concentrations of 209, 290 and 350 mg L⁻¹ respectively and sulfate concentrations of 1660, 2782 and 3825 mg L⁻¹ respectively. In October 1995, the concentrations of calcium at Sites 6 and 7 were 93 and 203 mg L⁻¹ respectively and sulfate levels were 1969 and 1013 mg L⁻¹. Median nitrate concentrations at Sites 4 and 7 were 0.08 and 0.07 mg L⁻¹ respectively.

Tidal waters of Firewood Creek

The water quality in the tidal section (Sites 8 and 9) of Firewood Creek was largely determined by the inflow of waters from the upper catchment and the tidal cycle. During the drier months there was little or no freshwater flow in Firewood Creek but seasonal summer rains caused flow events. The tide gates on the levee interrupt freshwater flows, and discharges were controlled largely by tidal height with impounded water being released only during the lower part of the tidal cycle. The incoming tide stopped the discharge by shutting the gates. The height of fresh water backed up behind the levee acted to prolong this discharge.

The influx of seasonal floodwaters was responsible for variations in the pH in the tidal waters of Firewood Creek. For the drier parts of the year (May–December), pH was generally 7–8 (Fig. 6) and within the limits recommended by Chapman (1992) for fish and other aquatic life in natural waters. At Site 8, immediately downstream of the tidal gates, the pH of both top and bottom waters dropped to around 3 during flood events caused by monsoonal rains in February and March 1995 (Fig. 6). During this event, the pH at Site 9 at the mouth of Firewood Creek also dropped. There was a similar heavy rainfall event in early 1996 that also resulted in acidification of the downstream tidal reaches of Firewood Creek.

During small flow events, such as the one that occurred in the first week of August 1995 (76 mm, Fig. 6) or at the tail end of major events, the flow may be insufficient to lower the pH in bottom waters. In such circumstances, at low tide, a layer of fresh, low-pH water flows over a wedge of relative high-salinity estuarine water with a normal pH. During high



Fig. 5. Dissolved oxygen concentration (%) in Magazine (Site 1) and Firewood (Sites 4 and 7) creeks and in Trinity Inlet (Sites 10–12), September 1994 to June 1996.



Fig. 6. pH at Sites 8 and 9 and daily rainfall (mm, black columns) recorded at Cairns airport, September 1994 to June 1996: \triangle , surface pH samples; \blacksquare , bottom pH samples; \diamondsuit surface salinity.

flow events, the volume and associated turbulence was sufficient to mix the waters to give relatively uniform lower pH values through the water column. Only during major rainfall events was the discharge of acid waters sufficient to depress pH in the lower water column at the mouth of Firewood Creek (Site 9). The relationship between salinity, pH and tidal cycle was evident in the bottom waters at Site 8 during a heavy rainfall event in March 1995 (Fig. 7), with the operation of the tidal gates at low tide resulting in a discharge of acidic fresh water which, in turn, resulted in a fall in both salinity and pH. During neap tides, for example from 7 to 9 March, the height of the low tide may not be sufficient for the tidal gates to operate on every cycle. The 434 mm of rain which fell from 12 to 14 March caused a large increase in flow which, in turn, caused the gates to remain open for longer, thereby increasing the time that tidal waters of Firewood Creek remained acidified (Fig. 7). Even in such a substantial flood, there was a wedge of relatively high-salinity estuarine water at Site 8 during high tides.

Total monomeric aluminium concentrations at both sites varied within and among years but peaked during the flood event in February 1995 (Fig. 8). Those at Site 8 (closest to



Fig. 7. Dissolved oxygen (% saturation), pH, specific conductance $(dS m^{-1})$ and depth (m) at Firewood Creek at hourly intervals, 3–14 March 1995. Vertical axis plotted on log scale. Depths are indicative, not true tidal height.



Fig. 8. Total aluminium concentrations (mg L⁻¹) in surface waters at Sites 8 and 9: \blacktriangle , from December 1994 to March 1996 (primary Y axis); \Box , November 1997 to March 1999 (secondary Y axis).

the tidal gates) were generally higher than at Site 9 (creek mouth). Concentrations in the surface waters were generally higher than in bottom waters.

Iron concentrations peaked during the major flow events in February 1995 (58 mg L^{-1} at Site 8 and 37 mg L^{-1} at Site 9; Fig. 9) and, with a few exceptions, generally remained low for the remainder of the sampling period. Once waters are transported to pH-neutral localities, dilution or precipitation (Sammut *et al.* 1995) generally results in lower iron and aluminium concentrations. In Firewood Creek, as distance downstream from the tidal gates increased there was a corresponding decrease in the iron and aluminium concentrations.

Oxygen concentrations in surface waters fell to as little as 1% saturation during the February 1995 flood event (Fig. 10). During the same period, the oxygen levels in the bottom waters were considerably higher. For the remainder of the sampling period, both surface and bottom waters followed a similar pattern. Oxygen concentrations recorded by the data logger (Fig. 7) suggest that there was a diurnal pattern with concentrations falling during the night and

increasing during the day presumably as a result of photosynthetic activity. A discharge of cooler, fresh water from inside the impounded area acted to increase the dissolved oxygen concentrations in the tidal sections of the creek. Turbulence created as this water was discharged through the tidal gates also assisted in increasing oxygen saturation levels.

In February 1995, a spot water sample taken at Site 8 had a calcium concentration of 175 mg L^{-1} and a sulfate concentration of 1747 mg L^{-1} . Later, in October 1995, the concentrations of calcium in two water samples from Site 8 were 265 and 295 mg L^{-1} respectively and sulfate concentrations were 1854 and 1935 mg L^{-1} respectively. At this time, median nitrate concentrations at Sites 8 and 9 were 0.06 and 0.05 mg L^{-1} respectively.

Tidal waters of Trinity Inlet

Water quality was measured in Trinity Inlet both upstream (Site 10) and downstream (Site 11) of the mouth of Firewood Creek and at the mouth of Magazine Creek (Site 12). For most of the year, pH values at these sites were between 7 and



Fig. 9. Iron concentrations (mg L^{-1}) at Sites 8 and 9, September 1994 and December 1995: \diamond , surface samples; \blacksquare , bottom samples.



Fig. 10. Dissolved oxygen concentrations (%) at Sites 8 and 9, September 1994 to June 1996: \blacklozenge , surface samples; \Box , bottom samples.

8, but they were slightly depressed during February 1995 and January 1996 (Fig. 2). Although the lowest value recorded (4.7 at Site 11) was not as low as values within Firewood Creek in the same period, it was probably a direct result of acid leachate discharge. The tidal section of Magazine Creek was not connected to the upper reaches behind the levee, and the slightly depressed pH during those periods of heavy rainfall may have resulted from lower surface-water salinity.

Total monomeric aluminium concentrations at Sites 10, 11 and 12 (Fig. 3) were much lower than those recorded during the same period from waters in both the tidal and impounded reaches of Firewood Creek.

Iron concentrations in surface waters at Sites 10, 11 and 12 (Fig. 4) were generally much lower than at other sites and, with the exception of the major flow event during February 1995, did not exceed 1 mg L^{-1} . The spike during mid–late February 1995 corresponded with a major flood event that resulted in the discharge of acidic water from Firewood Creek.

Dissolved oxygen concentrations at Sites 10, 11 and 12 remained relatively high (mostly >70%) (Fig. 5) although the intensive sampling during the flood in February 1995 revealed some decreases. Concentrations during the wet season months (January–April) were generally lower than during other times of the year.

In February 1995, spot water samples from Sites 10 and 11 had calcium concentrations of 285 and 212 mg L^{-1}

respectively and sulfate concentrations of 1390 and 2064 mg L^{-1} respectively. Median nitrate concentrations at Sites 10, 11 and 12 were 0.03, 0.04 and 0.04 mg L^{-1} respectively.

Fish kills

Dead fish were observed in the lower tidal section of Firewood Creek on 11-15 and 24 February 1995 and on 19 January 1996. None were observed during the wet season of 1997/98. During 8-15 February 1995, 233 mm of rainfall caused considerable runoff through the tidal gates resulting in depressed pH levels and high aluminium and iron concentrations in the tidal section of Firewood Creek. During this period, the pH dropped to as low as 2.8 and dissolved aluminium increased to up to 41 mg L^{-1} . A fish kill of several hundred fish, primarily gobies (Mugilogobius sp. and Acentrogobius janthinopterus) but including mullet (Mugilidae) and pikey bream (Acanothopagrus berda) was observed in this area. Seine netting at the mouth of Firewood Creek (Site 9) on 14 February 1995 yielded a small number of fish species including herring (Herklotsichthys castelnaui) and diamond-scale mullet (Liza vaigiensis). It was likely that initially the discharge severely affected resident fish species, particularly benthic species such as gobies, but that more mobile species such as herring and mullet vacated and later recolonized the creek after tidal exchange had improved the water quality. The next fish kill, on 24 February 1995, coincided with the start of the next period of heavy rain when 121 mm fell in the catchment;



Fig. 11. Monthly catch rate (CPUE, number of fish caught per hour) from gill-net surveys, October 1997 to October 1998:), \bigcirc , Firewood sites; \bigcirc , Falls and Seelee creeks; solid columns represent cumulative rainfall for seven days prior to sampling.

mortalities included glass perchlets (Ambassis gymnocephalus), herring (Escualosa thoracata), shad (Anodontostoma chacunda), anchovies (Stolephorus sp.) and black-tipped pony fish (Leiognathus splendens). Although heavy rains continued into March 1995 no more mortalities were observed. The single fish kill observed during the 1995/96 wet season occurred on 19 January after nearly 100 mm of rain had fallen in the previous two days. Moribund fish and prawns included H. castelnaui, A. gymnocephalus, Stolephorus sp., toothed pony fish (Gazza minuta), the estuarine gudgeon (Butis butis), L. splendens, garfish (Hemiramphus regularis ardelio) and pug-nose pony fish (Secutor sp.).

Histological examination of the gills of herring and anchovy sampled on 19 January 1996 revealed acute lesions consisting of degeneration and necrosis of the lamellae epithelium (Ian Anderson, Queensland Department of Primary Industries, personal communication).

Fish assemblages

By means of seine-net, gill-net and cast-netting techniques, 5452 fishes from 128 species and 45 families were sampled in Firewood, Falls and Seelee creeks; 114 species (42 families) were caught in Firewood Creek but only 70 species (32 families) in Falls and Seelee creeks. The number of species found in Firewood Creek was only slightly lower than the 134 taxa found in the area by Coles *et al.* (1993) in 1988, and higher than 91 species recorded by Blaber (1980).

Although the number of fish families and species caught by gill-net in Firewood Creek (21 families, 37 species) and Falls/Seelee creeks (20 families, 31 species) were comparable, the catch rates in Firewood Creek were significantly greater (paired *t*-test, df 12, t = -2.25, P = 0.044).

Catch rates at Firewood Creek were variable, with relatively high catches during October and November 1997 and March, August and September 1998 (Fig. 11). Except in October 1997, catch rates at the Falls/Seelee creeks control sites were lower and less variable. Rainfall immediately



Fig. 12. Fish species diversity (Shannon Weaver Diversity Index) from gill-net surveys, October 1997 to October 1998: \bigcirc , Firewood sites; \bullet , Falls and Seelee creeks; solid columns represent cumulative rainfall for seven days prior to sampling.



Fig. 13. Monthly catch rate (CPUE, number of fish caught per shot) for Firewood Creek seine-net shots, October 1997 and October 1998: \Box , bund wall; \triangle , anabranch; X, mouth; solid columns represent cumulative rainfall (mm) for seven days prior to sampling.

prior to sampling appeared to have little influence on catch rates.

Firewood Creek had a higher species diversity than Fall/Seelee Creeks during most months (Fig. 12). Exceptions to this were in August and September, when a decline in species diversity was related to a seasonal influx of a large number of migratory baitfish, particularly mullet and herring. Helmke (unpublished) also observed an increase in baitfish catches during this period at other localities in Trinity Inlet.

The effect of a range of environmental parameters including rainfall and pH on gill-net catches was tested with a multiple stepwise-regression model. Inclusion of any of these parameters in the model failed to explain any of the variation in the catches.

Catches from monthly seine-net shots at three sites in Firewood Creek (Fig. 13) were generally low during October–February. Catches at the wall site (Site S1), which was closest to the discharge point, were lowest when there were periods of rain immediately prior to the sampling (Fig. 13). In relatively dry periods, e.g. May–August, catches increased. The high numbers of fish caught at the mouth and anabranch sites in mid year were due to schools of small mobile fish, predominantly *H. castelnaui*,



Fig. 14. Monthly fish species diversity (Shannon Weaver Diversity Index) for seine net shots in Firewood Creek, October 1997 and October 1998: \Box , bund wall; \triangle , anabranch; \bigcirc , mouth; solid columns represent cumulative rainfall (mm) for seven days prior to sampling.

A. chacunda and L. splendens. Although this reflected seasonality in the occurrence of these groups, perhaps the increasing salinity and reduced acid discharge also allowed these fishes to colonize these waters. In a multiple stepwise-regression model, there was a significant correlation (P = 0.003) between $\log_e(\text{number of fish +1})$ caught at the wall site and rainfall for the previous week. This model explained 61.1% of the variation and was not significantly improved through the addition of extra terms.

Changes in species diversity for seine-net samples at the three seine-netting sites in Firewood Creek are shown in Fig. 14. Site S1, which was immediately downstream of the wall and which would be expected to be most influenced by discharges, showed the most fluctuation in species diversity. At this site, species diversity in January 1998 was zero and less than one in February 1998. A multiple stepwise-regression model suggested that dissolved aluminium explained >63% of the variation. Inclusion of other terms did not significantly improve the model. Increases in dissolved aluminium concentrations as a result of discharges from the upstream reaches of Firewood Creek appeared to be a significant factor in the reduction of local species diversity in estuarine waters. The anabranch of Firewood Creek showed less variability than the sites at the mouth and at the wall. The anabranch site, which was out of the direct flow-path of the acidic discharges, appeared to be less affected by releases of leachate than were the other two sites.

Mud crabs

The total numbers of mud crabs caught were 269 at the Firewood Creek site and 115 in Falls and Seelee creeks. The CPUE at Firewood Creek, downstream from the levee wall where acid water was discharged, was always greater than at the two control sites (Fig. 15). In Firewood Creek, the CPUE fell during December–April (the wetter, warmer months). A significant proportion of the variation in catch rate (Fig. 15) could be explained by site, season and minimum pH; once



Fig. 15. Mud crab catch per unit effort (CPUE, number caught per pot lift), October 1997 to September 1998: ○, Firewood Creek;
♠, Falls/Seelee Creeks; solid columns represent cumulative rainfall (mm) for seven days prior to sampling.

site and season had been included in the multiple regression model, minimum pH (lowest pH recorded in the previous 24 h) was also significant in explaining catches.

Approximately 14% of the 257 crabs tagged in Firewood Creek and 8% of the 103 crabs tagged in Falls Creek were recaptured. Only one tagged crab was recaptured outside of Firewood Creek suggesting that there was little movement in response to the discharge of acid water.

Discussion

Land reclamation, drainage and installation of structures for flood mitigation can lead to the oxidation of pyritic soils and the production of sulfuric acid (Virgona 1992; Lin et al.1995; Preda and Cox 2000). The drainage and reclamation works at East Trinity lowered the water table and exposed potential acid sulfate soils to oxidation. This process was responsible for the low pH values and high total monomeric aluminium, iron, sulfate and calcium concentrations observed in the impounded waters of Firewood and Magazine Creeks. In addition to finding low pH values and elevated concentrations of aluminium, iron, sulfate and calcium, Hicks et al. (1999) also found the impounded waters of Firewood Creek to have silicon levels between 8 and 64 mg L^{-1} . These elevated concentrations are probably due to acid-induced dissolution of aluminosilicates within the soil, and the high calcium values may have originated partially from shell grit layers found throughout the site (D. Smith, Queensland Department of Natural Resources, personal communication). There was no evidence that any other processes (e.g. oxidization of nitrogenous fertilizers) were responsible for the acid production.

Although a few fish species appeared tolerant of these conditions, fish stocks in the impounded waters of Firewood Creek were considerably lower than in the tidal waters of the inlet. Sammut *et al.* (1996) maintained that such acid reservoirs act as a chemical barrier to fish migration not present under natural conditions. It appears that some fish

species are very tolerant of, or become acclimated to, acid waters and high aluminium concentrations (Mount et al 1990, Lin et al. 1995). In Kalimantan, Klepper et al. (1992) noted that certain fish species appeared well adapted to acid conditions and contrasted this to temperate regions where fish were almost completely absent from waters where the pH was < 4.2–4.9. A large natural fish kill in the Northern Territory, Australia, was attributed to a combination of natural acid water runoff and resultant elevated aquatic aluminium levels, but tarpon biotoxic (Megalops *cvprinoides*) were seen feeding on the surface during the fish kill and hence were apparently more tolerant to acid water than many other native Australian fishes (Brown et al. 1983). Dent (1986) noted interspecific and intraspecific variation in aluminium tolerance, and Howells et al. (1994) suggested that there was an acclimatory response in some fish exposed to sublethal conditions of pH, aluminium and calcium which allowed them greater resistance to short-term peaks of aluminium in association with episodes of acidity. At the East Trinity site, M. cyprinoides, barramundi (Lates calcarifer) and rainbow fish (Melanotaenia splendida) were all found behind the levee (Site G3) in waters where the pH regularly dropped to about 3 and the total aluminium reached $>35 \text{ mg L}^{-1}$.

There were some unexpected variations in the water quality pattern observed at impounded sites at East Trinity after heavy rainfall. For example, at Sites 1 and 7, the onset of wet-season rains resulted in an increase in the pH of surface waters presumably due to dilution whereas, at the same time, the pH at other impounded sites (Sites 2–5) was depressed. Factors such as sub-catchment soil types and volume of floodwaters received from other areas affected the pH of the creeks.

Although some discharges triggered by flood rains resulted in the acidification of the tidal section of Firewood Creek, there was little evidence of effects on the water quality of Trinity Inlet; in this large, well flushed estuarine system, acid discharges from Firewood Creek are diluted, dispersed and buffered, thereby minimizing effects except in the tidal reaches of the creek and its immediate surrounds. This contrasts with some confined riverine environments in northern New South Wales where Sammut et al. (1996) reported acid drainage to be responsible for the acidification of >90 km of a tributary stream of the Richmond River, New South Wales. Although they may produce more insidious effects such as periodic outbreaks of epizootic ulcerative syndrome (Callinan et al. 1993), most of the discharges of acidic water into the tidal reaches of Firewood Creek did not result in observable fish kills.

Natural controls and management interventions can minimize the degree of estuarine degradation due to acid sulfate soils (Lin *et al.* 1995). In Firewood Creek, the discharge would need to comprise a large volume of acid water with high concentrations of biotoxic metals before a fish kill would occur. Because of the restricted nature of the tidal reaches of Firewood Creek, tidal flushing is apparently sufficient to allow fish to recolonize soon after the main flow event has abated. Although episodic acidification associated with freshwater discharges on low tide may continue for some time after a major flood event or during small flow events, mixing appears limited and estuarine fish use the saltwater wedge as a refuge or to penetrate upstream and inhabit the lower parts of the system. Under these circumstances the saltwater wedge does not have elevated levels of biotoxic dissolved metals and the pH is close to normal. Sammut et al. (1995) reported that acid pulses in tidal sections of the Richmond River can persist for months during low tides and that they can move backwards and forwards as a slug when mixing and seawater neutralization are limited. In all but very large flood events, tidal flushing appears sufficient to disperse acid water quickly from the lower reaches of Firewood Creek.

The environmental effects of acid discharges are varied. Under some circumstances, the hydrogen ions and dissolved species of aluminium and iron can result in fish kills (Sammut *et al.* 1995) or reduced fish diversity and abundances. Ramamoorthy (1988) found that rainbow trout (*Salmo gairdneri*) mortalities from alum sludge at pH <4.5 were probably due to colloidal particles and acid- and aluminium-induced stress. It has also been suggested that acid leachate may trigger ulcerative diseases in fish (Callinan *et al.* 1993) and reduce growth rates in some species (Klepper *et al.* 1992). Although there were a few occurrences of epizootic ulcerative syndrome in the fish sampled during the present study, there are no data from un-impacted north Queensland estuaries for comparison.

At East Trinity, fish kills and moribund fish and prawns swimming erratically on the surface were observed on a number of occasions during the 1994/95 and 1995/96 wet seasons but not in the 1997/98 season. Many of these fish were smaller, less mobile species including gobies and eleotrids. Although the magnitude of these fish kills was difficult to gauge, these events appeared to be localized, restricted to a small section of Firewood Creek downstream of the tidal gates. Histological examination of some specimens suggests the pathology of gill damage to be similar to that described by Sammut et al. (1995) in fish taken from an acidified section of the Richmond River in northern New South Wales. Although there were no observed fish kills in the 1997/98 wet season, acidic discharges affected the fish species diversity in seine-net samples in the tidal reaches of Firewood Creek with the diversity at one site reduced to zero. The reduction of diversity was correlated species with increased concentrations of dissolved aluminium, and low fish abundances were related to heavy rainfall during the previous seven days. Other authors, including Chairuddin et al. (1990) as cited by Klepper et al. (1992), have also found a strong decrease in species diversity and total biomass of fish in acid sulfate soil areas. Changes in aluminium speciation are likely to be a major factor governing the occurrence of fish kills in Firewood Creek. In acidified lakes and streams in the north-eastern USA, Driscoll et al. (1980) noted that aluminium speciation was highly variable and its effect on fish was also variable. Complexation of aluminium with organic ligands seems to eliminate its toxicity and organically chelated aluminium was significantly correlated with total organic carbon (TOC) (Driscoll et al. 1980). Those authors suggest that, despite moderately low pH and high total aluminium concentrations, waters with high organic carbon content may be suitable for successful fish production because of the ameliorating effect of organic complexation on aluminium toxicity. TOC was not measured in the present study, but between 7 January and 9 March 1998 the TOCs of five samples collected in the tidal section of Firewood Creek by Hicks et al. (1999) were high at 10–14 mg L^{-1} . Spot sampling during the present study (in 1995) and samples collected by Hicks et al. (1999) in 1997 and 1998 indicated high calcium and silicon concentrations in the impounded waters. High concentrations of calcium and silicon can lessen the toxicity of inorganic Al to fish (Reader and Dempsey 1988; Brocksen et al. 1992). Since conditions in natural waters are rarely in equilibrium, a potential for toxicity will remain if Al speciation changes in response to fluctuations in water quality (Howells et al. 1994). Further testing may clarify the effects that factors such as organic carbon, silicon and calcium have on the toxicity of the aluminium in the discharges from Firewood Creek.

The tidal reaches of Firewood Creek, despite the periodic inflows of acid water, supported a relatively productive recreational fishery for mud crabs. The reasons for this are unclear, although the anabranch section of the creek contained a relatively shallow, protected area. Habitat preferences of this species can include relatively shallow bays (Hyland et al. 1984). Since the present study recorded crab catches in only one year, it is not possible to draw inferences from the lower catch rate at Firewood Creek during the wetter, warmer months. However, a similar trend was observed in other parts of Trinity Inlet in 1997 and 1998 (Helmke et al. in press). Although the influx of water with low pH and high Al concentrations does appear to reduce significantly the numbers of mud crabs caught in pots, those resident during the wet season still appeared to be feeding. No evidence was found of mortalities nor was there any evidence from the tagging studies to suggest any significant exodus of crabs from Firewood Creek during periods when acid water was being discharged. They may have avoided exposure by temporarily retreating into burrows. Generally, high-flow events do not last more than about a week and the evidence suggests that even under these conditions a wedge of relatively clean estuarine water is able to penetrate into the creek during some parts of the tidal cycle. Whether through the adoption of avoidance strategies or by other means, these bottom-dwelling mud crabs appear less severely affected by the acid discharges than the mid-water biota.

Iron may also have a deleterous effect on aquatic fauna and flora. Sammut *et al.* (1996) noted that red-brown flocs of iron hydroxide or iron oxyhydroxide formed in waters at pH >2.7 and were transported downstream in suspension or coated stream beds and banks. Iron deposits can coat the gills of fish and crustaceans thereby impairing gas exchange (Simpson and Pedini 1985), but no evidence was found of this during the present study. Simpson and Pedini (1985) also noted that iron hydroxide can coat benthic algae thus rendering them inedible for the meiofauna. In the impounded area of Firewood Creek, emergent vegetation, particularly reeds, has been encrusted with iron precipitates but any long-term effects are unknown.

Occasionally, other factors including depleted oxygen concentrations may also affect fishes. Waters that have low pH and high Al concentrations tend to be depleted in dissolved oxygen (Hyne and Wilson 1997). During this study, the dissolved oxygen concentrations at most sites generally remained high throughout the year, although at many sites the values did decrease slightly during the wetter, summer months. There were periodic pulses of deoxygenated water discharged into the tidal section of Firewood Creek during some heavy rainfall events. During one such rainfall event in early February 1995, dissolved oxygen levels during an early morning low tide were as low as 2% at Site 8 and 1% at Site 9. These levels quickly recovered with the incoming tide but, together with low pH, high monomeric aluminium and iron concentrations, undoubtedly contributed towards a fish kill that was observed during this event. Low oxygen concentrations were not consistently associated with the discharge of high volumes of floodwaters from the impounded section of Firewood Creek (e.g. March 1995). During high flow periods, the slightly elevated oxygen concentrations were possibly a result of the large amount of turbulence associated with the discharge. There are a number of possible reasons why the oxygen levels inside the impounded area were occasionally depressed. Graczyk and Sonzogni (1991) noted that storm-water input could cause an influx or re-suspension of oxygen-demanding materials. Bishop (1980) recorded natural fish kills in the Northern Territory that resulted from oxygen deprivation due to exposure of anoxic bottom waters disturbed by flood rains. The decrease in oxygen levels in February 1995 may also be related to chemical processes, particularly pyrite oxidation, occurring in the impounded reaches of Firewood Creek. Sammut et al. (1995) noted that the oxidation of ferrous iron (an initial product of pyrite oxidation) to iron hydroxide consumes oxygen and releases hydroium ions (H₃O⁺), thereby decreasing dissolved oxygen concentrations and pH.

The results highlight the complexities of predicting the impact that discharges of acid water can have on fisheries resources. At East Trinity fish kills did not result from every flood event and appeared largely dependent on changes in, and interaction between, various chemical components of the water. Wilson et al. (1999) also contend that large rainfall events did not always produce significant changes to water chemistry and that small rain events could produce large changes if the prevailing conditions were suitable. Lin et al. (1995) noted that dilution from large freshwater discharges helped to minimize the impacts of pyrite oxidation products on the estuarine waterways of the Pearl River delta in China. Preda and Cox (2000) suggest that towards the end of the wet season, prolonged flushing of pyrite oxidation products led to short-term recovery of the aquatic system including a neutral pH and lower concentrations of dissolved metals. Perhaps the inter-annual variability at East Trinity is related to variations in the level of the watertable; this fell considerably in the dry period of 1994, but remained high throughout most of 1995 and 1997 which were relatively wet years. This suggests that raising the impounded water level may cause a rise in the watertable thus covering or partially covering some of the acid producing soil, preventing its oxidation. In northern New South Wales, Wilson et al. (1999) found that the magnitude of changes to the estuarine waters was dependent on the position of the watertable - and hence the available pore space - and on the store of acidic water in the floodgated drains at the time of rainfall.

Watertable management may be an appropriate rehabilitation method for East Trinity, even though Hicks *et al.* (1999) suggest that tidal re-flooding of the East Trinity site would not establish the reducing conditions necessary for neutralization of acid or acid byproducts. In New South Wales, White *et al.* (1997) suggested that a practical solution to an acid sulfate problem was to raise the water table thus sealing the acid sulfate soil layer, but they acknowledged the difficulties of maintaining watertable depth in areas with a pronounced dry season.

There is little evidence that the episodic acidic discharges are having any significant overall impact on either the water quality or the attendant fisheries of Trinity Inlet. The discharges are, however, a chronic localized problem that will be addressed through rehabilitation of the East Trinity site.

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