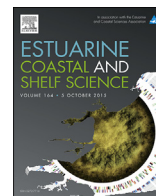




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Predicting the buffering of acid plumes within estuaries



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ABSTRACT

The acid buffering capacity of an estuary is directly proportional to the volume of buffering agents within the system. In areas with limited upstream inflows of buffering agents, the primary buffering agents are sourced from the diffusion of marine constituents. During dry periods the buffering capacity increases with tidal diffusion, whereas buffering decreases following estuarine flushing from high inflows. In acid sulphate soil environments, acidic discharges are generated immediately following rainfall events. Thereby, the diffusion of buffering agents throughout an estuary plays an important role in sustaining biogeochemical processes. Prediction of the transport and buffering dynamics of acidic plumes within estuaries allows severely impacted acid areas to be identified and catchment management strategies developed.

To simulate acid transport and tidal buffering processes within an estuary, a numerical model was modified and an acidic buffering module included within the hydrodynamic routine. An estuarine wide case study was then undertaken to demonstrate the effectiveness of the acid plume module in simulating onsite conditions and the role of buffering versus dilution. Acid discharged from the study domain, located on a tidal barrier estuary in south-eastern Australia, was monitored using fixed and boat mounted instrumentation to track far-field plume behaviour. The acidic plume monitoring results were converted to hydrogen proton concentrations and included within the water quality module to simulate acid buffering with bicarbonate ions diffusing from the marine boundary.

The validated model can be applied to single or multi-drain estuary networks to assess possible management options for reducing the impact of acid plume events. This enables the most effective management approach to be identified and assists in prioritising acid sulphate soil sites for remediation. This is particularly useful to natural resource managers in order to assess remediation sites that have the most environmental impact, or can be most effectively buffered, via model scenario tests.

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1. Introduction

Acid sulphate soil (ASS) is the name given to soils and sediments containing iron sulfides, the most common sulfide being pyrite (FeS_2) (DERM, 2009). Pyrite concentrations typically range from innocuous levels below 0.01% Chromium Reducible Sulfide (% S_{CR}), to extreme concentrations above 15.0% S_{CR} (Dent and Pons, 1995). Due to the low energy depositional environment in which they formed during the last major sea level rise period approximately 6500 years ago, subsurface concentrations in Australia are typically above the management action criteria of 0.05% S_{CR} set by Stone et al. (1998). ASS remain chemically inert under reducing conditions.

When atmospheric oxidation occurs as a result of climatic, hydrological or geological changes, the pyrite reacts to produce sulphuric acid and the oxidised soil is termed an actual acid sulphate soil (AASS).

The release of acidic plumes caused by oxidation of pyritic soils is well known to cause widespread environmental pollution in tidal estuaries resulting in large scale fish kills (Sammut et al., 1995, 1996; Pollard and Hannan, 1994; NSW Fisheries, 2001; Bush et al., 2004; Lin et al., 2004; Kroon and Ansell, 2006; Winberg and Heath, 2010) and impacting oyster health (Dove and Sammut, 2007a,b). To date, significant resources have been directed to understanding the generation and transport of acidic groundwater into adjacent drainage canals and adjoining creeks (Pease, 1994; Indraratna and Blunden, 1998; Blunden and Indraratna, 2000; Glamore, 2003; Johnston et al., 2003), but limited investigations (Glamore and Indraratna, 2001; Indraratna

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et al., 2002) have been undertaken to assess the transport and buffering dynamics of these acidic plumes after they have been discharged into an estuary.

The east coast of Australia has extensive acid sulphate soil deposits, exacerbated by floodplain drainage networks which lower the groundwater table (Naylor et al., 1995). In areas affected by ASS, one-way floodgates at the tidal junction of drainage networks increase pyrite oxidation (Indraratna and Blunden, 1998; Glamore, 2003), as floodgate invert levels are generally set to maintain floodplain water levels at low tide elevations. Since the pyritic layer is normally at the mid to high tide level, by maintaining drain water elevations lower than the pyritic layer, the one-way floodgates increase the hydraulic gradient between the drain water and the groundwater (Glamore and Indraratna, 2001). This gradient is particularly steep following significant rainfall events when groundwater levels are high and receiving water levels have returned to regular tidal levels (Johnston et al., 2003).

The risk of widespread acidic contamination to the estuary is offset in highly flushed estuaries with high acid buffering potential (Glamore and Indraratna, 2001). Acid buffering occurs when strong acids (hydrogen protons (H^+)) react with bicarbonate (HCO_3^-) or carbonate (CO_3^{2-}) inherent in marine waters. Similarly to salinity, bicarbonate diffuses from the ocean forming a buffering gradient throughout the estuary. Acidic plumes have the greatest environmental impact when the receiving water is predominantly freshwater and the acidic flux (concentration*discharge) is high. This has been shown to occur at the end of a freshwater hydrograph (Johnston et al., 2003). As such, during dry periods the buffering capacity of the entire estuary is high, while during wet events the buffering capacity is reduced.

The tail end of a freshwater hydrograph is a period of low dilution and high pollutant load, with limited buffering capacity in the receiving water. Johnston et al. (2003) identified that 90% of the total pollutant load is discharged over the last 10% of the flood hydrograph. During this period, the surface water has drained from the floodplain, forcing a strong gradient between high groundwater levels and low drain and estuary water levels (Indraratna et al., 1995). This drives the flow of acidic groundwater into the floodplain drainage network and the receiving estuary (Blunden and Indraratna, 2000). Acidic plumes have a long residence time within the estuary or can join with other acidic plumes to form a 'super-plume' (Glamore, 2003).

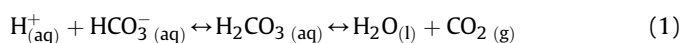
While the generation and export of acid from coastal floodplains is well understood, limited information about the fate and transport of acid plumes on an estuary wide scale is available (Indraratna et al., 2002; Nordmyr et al., 2008; Hipsey et al., 2014). Prediction of acidic plume transport, including buffering dynamics, would improve our understanding of acid plume dynamics allowing for the development of catchment wide management and remediation strategies (Rayner, 2010). The aim of this study was to develop an acid buffering module within an estuary wide water quality hydrodynamic numerical model to replicate acid plume dynamics within a dynamic estuary with varying acid/buffering potentials. Once verified and validated against an observed acid plume event, the acid buffering module can enable detailed understanding of multi-plume interactions and the role of onsite acid remediation within estuarine environments.

This paper outlines the development and validation of a hydrodynamic/water quality model to analyse acid plume transport and buffering dynamics. Modifications of a water quality model source code were undertaken to include the acid-bicarbonate buffering reaction. The RMA suite of models were utilised for this study, namely RMA-2 v7.5 (King, 2006a) for hydrodynamics and RMA-11 v4.4c (King, 2006b) for water quality modelling.

2. Model approach

The concentration of a single, conservative water quality constituent, such as salinity, is controlled by advection, dilution and diffusion with the modelling of conservative water quality constituents has been successfully undertaken for decades (Leendertse and Gritton, 1971; Hamilton, 1990; Lung, 1993; James, 1993; Simons et al., 1996; Chapra, 1997). However, as the concentration of acidity, or pH, is determined by advection, dilution, diffusion and buffering, the neutralisation reaction of acid by bicarbonate needs to be included in modelling exercises.

The approach was to develop an algorithm that simulates acid-buffering in estuaries. As the advection of acidic plumes is determined by hydrodynamic forces, the buffering reaction rate was added to a hydrodynamic-water quality modelling suite. Several stages of acid production occur during the buffering of acid sulphate soil leachate, however the majority of pH variation can be summarised using the buffering reaction as per Equation (1).



Although the above equation is a simplification of a complex process, modelling of the buffering reaction allows hydrogen protons (H^+) to represent both acidity and soluble metals. As this is an equilibrium process, the reaction was modelled using molar concentrations.

The acid neutralisation capacity (i.e. buffering capacity) of a solution is determined by alkalinity (Morel and Hering, 1993). Alkalinity is comprised of seven principal components with bicarbonate the dominant species at near neutral pH (Morel and Hering, 1993). As such, bicarbonate was chosen as the constituent representative of buffering capacity. Although carbonate has double the buffering capacity of bicarbonate, due to a double negative charge, it was not considered relevant in this study as carbonate is only present in significant quantities within marine conditions (Morel and Hering, 1993).

The RMA suite of finite element models was utilised. Hydrodynamic processes were simulated (Hofman et al., 2009) using RMA-2, and water quality was modelled using RMA-11. In its existing state, RMA-11 does not have the capability to model acid buffering reactions and subsequently the model source code was modified to incorporate two new constituents, [ACID] and [BICARB]. The modifications enabled the coupling of the two constituents, where the decay of each constituent is dependent on the presence of the other. As Equation (1) is a 1:1 reaction, the source code was modified to replicate this reaction ratio.

Code modifications were initially tested in a steady state box model. The advantage of a box model approach is it has no intrinsic variability, enabling the reaction to occur free from diffusion and advection effects, testing the effects of the model code modifications (Officer, 1980). The acid neutralisation reaction was tested over a range of constituent ratios, from 1:0.1 to 1:100,000. These constituent ratios were then tested over multiple time-steps from 0.1 h to 1 h. For each test case, the rate of reaction was the same, with a first order decay of constituents. Box model test results validated the numerical representation of Equation (1), with one part of bicarbonate buffering one part of acid. An example of the buffering reaction reproduced in a box model is shown in Fig. 1. The example box model depicts an initial condition of 500 parts of acid and 100 parts of bicarbonate. This reaction between the two constituents was found to reach equilibrium after approximately 15 h, with a final concentration of 400 parts of acid and zero parts of bicarbonate.

Due to the freshwater inflow within an estuary, coupled with the diffusion of saltwater from the ocean boundary, a dynamic

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