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Tidal and seasonal variation in carbonate chemistry, pH and salinity for a mineral-acidified tropical estuarine system

Sorya Proum [a,b], Jose H. Santos [c], Lee Hoon Lim [c], David J. Marshall [a,*]

[a] Environmental and Life Sciences, Faculty of Science, Universiti Brunei Darussalam, BE 1410 Jalan Tungku Link, Brunei Darussalam
[b] Chemistry Department, Royal University of Phnom Penh, Russian Federation Boulevard, Toul Kork, Phnom Penh, Cambodia
[c] Chemical Sciences, Universiti Brunei Darussalam, BE 1410 Jalan Tungku Link, Brunei Darussalam

HIGHLIGHTS

- Tidal fluxes and seasonal patterns in pH, salinity and carbonate chemistry are described for a tropical Southeast Asian estuary influenced by Acid Sulphate Soil (ASS) discharge.
- Heavy daily downpours had little effect on the water salinity and pH, whereas accumulative rainfall during the monsoon lowered these parameter's baselines.
- Remarkably low pH relative to salinity, extraordinary pCO_2 super-saturation, and carbonate under-saturation occurred extensively across the estuary.
- Mineral acidification is implicated in changing estuarine pH and elevating pCO_2 through the carbonate equilibrium system.

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ABSTRACT

Estuarine acidification and carbonate chemistry derive from multiple biogeochemical processes. Other than biogenic CO_2-acidification, estuaries can be acidified allochthonously through non-carbonate sources originating in freshwater and land ecosystems. The present study considered the carbonate chemistry of a nutrified, turbid, tropical mangrove estuary, influenced by acidic groundwater discharge from pyritic soils (Acid Sulphate Soils, ASS). We studied the spatial and temporal variation of the surface water pH, salinity, total alkalinity (TA), partial pressure carbon dioxide (pCO_2), dissolved inorganic carbon (DIC), and calcite (Ω_{cal}) and aragonite (Ω_{ara}) saturation, in the Brunei Estuarine System (BES), Borneo, Southeast Asia. pH and salinity for tidal to seasonal timeframes were determined from data collected half-hourly, logged at three stations (upper, middle and lower estuary); these data were correlated with rainfall incidence and intensity. Carbonate parameters were calculated from TA using discrete samples collected from six stations. pH (6.8–7.9) and salinity (4.2–28.2) increased expectedly seawards, due to tidal forcing and freshwater dilution at opposite ends of the estuary; amplitudes within a tidal cycle became expanded landwards and during spring tides. While, the overall effect of heavy daily downpours on estuarine salinity and pH was muted, cumulative rainfall during the monsoon season distinctly lowered parameter baselines; the response was again more pronounced in the upper estuary. In the mid-to-upper estuary, we observed a remarkably low pH relative to salinity, extraordinary pCO_2 super-saturation (13031 ± 4412 μatm) and carbonate undersaturation (Ω_{cal} and Ω_{ara} were 0.006–1.431 and 0.004–0.928, respectively). Although the relative contributions of heterotrophic metabolism and ASS-discharge to the estuarine pH and pCO_2 were not determined, both processes are implicated in increasing both acidity and CO_2 levels. This study contributes to the understanding of carbonate fluxes in mineral-acidified estuaries.
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1. Introduction

Estuarine carbonate systems are highly variable, both temporally (relative to tides and freshwater discharge) and spatially (within estuaries, locally and regionally). Understanding this variability is important to ecological frameworks (ecological structure and functioning), as well as marine, oceanic and atmospheric CO_2 flux contexts. Interest in marine carbonate systems has peaked against the backdrop of anthropogenic atmospheric CO_2 elevation (Noriega and Araujo, 2014); CO_2 in the atmosphere is rising at ~2 ppm per year (IPCC, 2013; NOAA, 2015), with increased CO_2 hydrolysis in oceanic surface water predicted to reduce the pH

* Corresponding author.
E-mail address: david.marshall@ubd.edu.bn (D.J. Marshall).

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by 0.3–0.4 by 2100 (Caldeira and Wickett, 2003; Kimmerer and Weaver, 2013). By lowering calcite and aragonite ($CO_3{}^{2-}$) saturation states, ocean acidification places calcifying organisms and ecological assemblages at greater risk (Orr et al., 2005; Kimmerer and Weaver, 2013). Contrasting with oceanic systems, estuarine CO_2 levels are mainly saturated to super-saturated, leading to net atmospheric CO_2 transfer (estuaries are generally sources of atmospheric CO_2; Borges and Gypens; Maher et al., 2015; Sadat-Noori et al., 2016). However, the carbonate chemistry of coastal systems where estuaries and oceans interact is spatially and temporally complex (Cai and Wang, 1998; Cai et al., 2011; Sunda and Cai, 2012; Call et al., 2015; Zhai et al., 2015).

Traditionally, estuarine studies focused on specific physical and chemical attributes of the water (salinity, temperature, dissolved oxygen and nutrients), with carbonate chemistry receiving limited consideration (but see Borges, 2005; Chen et al., 2012; Call et al., 2015). More recently, studies describing estuarine carbonate systems are geographically-skewed towards temperate and subtropical regions (see Laruelle et al., 2010; Noriega and Araujo, 2014; Maher et al., 2013; Maher et al., 2015) , with limited information available for tropical estuaries, especially in Southeast Asia (Koné and Borges, 2008). These estuaries are often associated with turbid river systems, productive terrestrial systems (back swamps and mangroves) and densely-populated urban areas, implying substantial nutrient (organic) loading. Excessive organic matter drives heterotrophy and aerobic respiration of resident benthic and pelagic organisms (Anderson et al., 2002; Wang, 2006; Bianchi and Allison, 2009; Cai et al., 2011; Howarth et al., 2011), which raises *in situ* water pCO_2. Additionally, pCO_2 in mangrove-dominated estuaries can be elevated by tidal pumping and within estuarine ground/porewater discharge (Borges et al., 2003; Bouillon et al., 2007; Maher et al., 2013). Furthermore, estuarine pCO_2 is affected by CO_2 importation (heterotrophy elsewhere in rivers and/or groundwater), CO_2 loss to the atmosphere (relating to wind speed and current flow; Borges et al., 2003; Zhai et al., 2005; Bouillon et al., 2007; Call et al., 2015) and exportation (relating to tidal flux and mixing; Rivkin and Legendre, 2001; Cai, 2011). Salinity robustly influences estuarine pCO_2, which is reduced at the higher salinities where an estuary enters the coastal ocean (close to equilibrium with atmospheric CO_2; Frankignoulle et al., 1998; Sarma et al., 2001; Sarma et al., 2011; Chen et al., 2012; Noriega and Araujo, 2014).

pCO_2 elevation lowers estuarine water pH (Eq. (1); Raymond et al., 2000). Nonetheless, estuarine acidification can arise through multiple other processes, including natural mineral acidic discharge (see Marshall et al., 2008). Such discharge derives from pyrite (FeS_2 in soils/sediments), which is formed under reducing conditions when marine sediments are inundated (see Acid Sulphate Soils, ASS; Dent, 1986; Powell and Martens, 2005; Grealish et al., 2008). When fossilized sediments that have been subject to sea-level-decline are disturbed by flooding or air-exposure, oxidation of the pyrite produces sulphuric acid (Fig. 1; Eq. (2); Stumm and Morgan, 1981; Dent, 1986; Schippers and Jorgensen, 2002).

$$CO_2 + H_2O \rightleftharpoons H_2CO_3 \rightleftharpoons HCO_3^- + H^+ \quad (1)$$

$$FeS_2 + 15/4O_2 + 7/2H_2O \rightarrow Fe(OH)_3 + 2H_2SO_4 \quad (2)$$

Van Breemen (1973)

Many coastal systems across the globe (floodplains, wetlands and estuarine embayments) are influenced by acidic groundwater infiltration (Walker, 1972; Sammut et al., 1996; Roach, 1997; Astrom and Bjorkland, 1995). Studies on the ecological consequences of ASS discharge have prominently been conducted in Australia

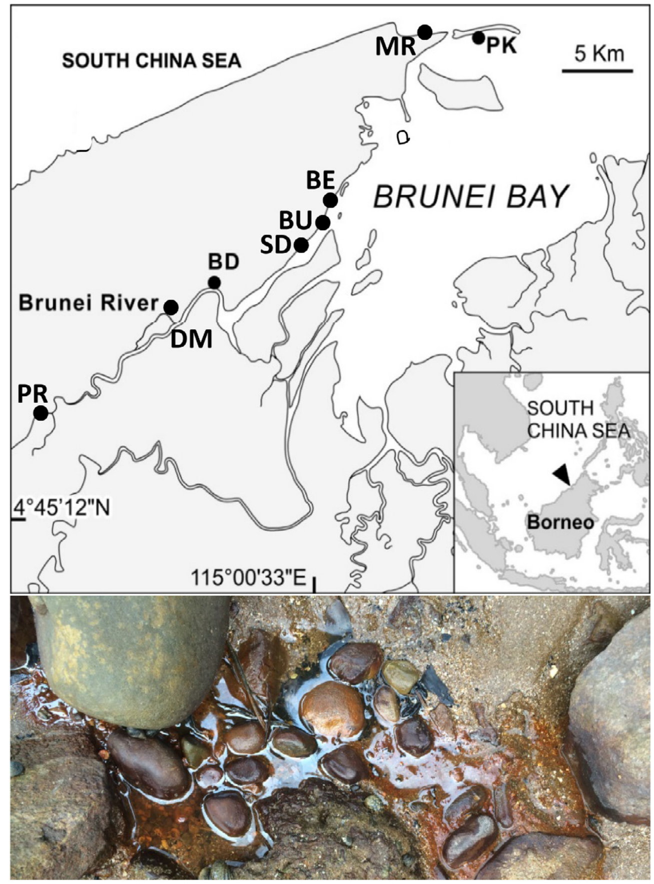

Fig. 1. Map of the Brunei Estuary System (BES) (upper panel). Sampling was carried out during Sep 2013–Mar 2014. Hydrological monitoring stations were Bandar (BD), Serdang (SD) and Pulau Pelumpong (PK). Discrete sampling for the carbonate system analysis was carried out at Kampong Parit (PR), Damuan (DM), Bandar (BD), Sungai Bunga (BU), Sungai Besar (BE), and Muara (MR). Lower panel shows a seep from pyrite-rich sediments at a nearby coastal locality, producing brown-staining precipitates of iron oxyhydroysulphate on pebbles and in the sediment (see Grealish et al., 2008). Oil-like bacterial films are associated with the seep.

(Brown et al., 1983; Callinan et al., 1993; Neal, 1993; Dove, 2003). More recent studies, also emanating from Australia, have explored the interaction of this discharge and the carbonate chemistry of estuaries (Maher et al., 2013; Ruiz-Halpern et al., 2015; Sadat-Noori et al., 2016; Jeffrey et al., 2016). Other cases might involve ASS, but this has not been explicitly indicated (see Chen and Borges, 2009; Laruelle et al., 2010; Chen et al., 2012; Noriega and Araujo, 2014; Call et al., 2015).

We investigated an ASS-influenced tropical mangrove estuary, the Brunei Estuarine System (BES) [Brunei Darussalam (Borneo), Southeast Asia; Grealish et al., 2008]. Other than representing a useful scientific model system, the BES is ecologically sensitive and socio-economically important to the country. It supports housing (an extensive water village, Kampong Ayer), artisanal fisheries, small-scale aquaculture, and several other ecosystem services, including acting as a significant nursery ground for many fish species. The circumstances of multiple source acidification (see Marshall et al., 2008) have stimulated studies on responses of species and ecological assemblages to low pH waters (Hossain et al., 2014; Bolhuis et al., 2014; Majewska et al., 2016; Proum et al., 2016, 2017). Like many other tropical systems, the BES is vulnerable to extreme weather patterns (flooding and drought). Flooding is

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