



## Research papers

# High porewater exchange in a mangrove-dominated estuary revealed from short-lived radium isotopes



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## ABSTRACT

We hypothesise that mangroves play an important role in groundwater exchange processes in subtropical and tropical estuarine waters. To investigate this, multiple high resolution time series measurements of radium across a tidal estuary (Coffs Creek, NSW, Australia) were performed as well as a spatial survey in both bottom and surface layers. Results from the spatial survey revealed increasing radium concentrations in parts of the estuary surrounded by mangroves. The average radium concentration in estuary areas lined with mangroves was 2.5 times higher than the average concentration at the mouth of the estuary and 6.5-fold higher than upstream freshwater areas. Additionally, the area enriched in radium coincided with low dissolved oxygen concentrations, implying that porewater exchange may drive anoxia. A radium mass balance model based on  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  isotopes at different sections of the estuary confirmed higher porewater exchange rates from areas fringed with mangrove vegetation. Estimated porewater exchange rates were  $27.8 \pm 5.3$  and  $13.6 \pm 2.1 \text{ cm d}^{-1}$  ( $0.8 \pm 0.1$  and  $0.4 \pm 0.1 \text{ m}^3 \text{ s}^{-1}$ ) based on  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  isotopes, respectively. The average saline porewater exchange was  $\sim 10$ -fold larger than the upstream surface freshwater inputs to the estuary. We suggest that mangrove environments within subtropical estuaries are hotspots for porewater exchange due to the complex belowground structure of crab burrows and the effect of tidal pumping. Because porewater exchange releases carbon and nitrogen from coastal sediments, development and modification of mangrove areas in subtropical estuaries have a significant effect on coastal biogeochemical cycles.

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## 1. Introduction

Submarine groundwater discharge (SGD) can include either or both the flow of fresh terrestrial groundwater and/or porewater exchange (saline recirculated seawater) into coastal waters. Where sediments are saturated, both the terms groundwater and porewater can be used to describe the water below the sediment surface (Burnett et al., 2003). The origin of SGD can be from shallow and/or deep aquifers and the mechanisms driving SGD can include hydraulic gradient, tidal pumping, wave setup and density driven water convection occurring over various time and spatial scales (Santos et al., 2012a). The role of SGD is now recognized as a major component of water balances (Kwon et al., 2014; Sadat-Noori et al., 2016c), as a significant pathway for solute delivery to aquatic systems (Rodellas et al., 2015; Hong et al., 2017; Tait et al., 2017) and as a driver of biogeochemical processes in coastal environments

(Moore, 2006a,b). However, due to SGD's heterogeneous nature both in space and time, quantifying SGD remains a challenge (Burnett et al., 2006; Porubsky et al., 2014).

Natural geochemical tracers are now widely used to quantify SGD (Burnett et al., 2006; Trezzi et al., 2017). Radioactive nuclides such as radium isotopes ( $^{223}\text{Ra}$ ,  $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ) have been used to quantify SGD rates in numerous aquatic systems including estuaries (Charette et al., 2013), lakes (Wang et al., 2016), rivers (Peterson et al., 2008a,b), embayments (Stewart et al., 2015; Wang et al., 2016) and the continental shelf (Moore, 2000). Radium is a rare-earth metal produced from the decay of uranium/thorium and its concentration is often much higher in coastal groundwater relative to surface waters. The short-lived radium isotopes  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  have half-lives of 11.4 and 3.6 days which are on the same time scales of physical processes usually related to tidally-driven SGD and mixing in coastal waters (Burnett et al., 2008). Radium is attached to sediment particles and is released to solution when exposed to brackish or saline water due to ion exchange processes. This makes radium an ideal tool for quantifying recirculated seawater and saline groundwater (Moore, 2006a,b). However,

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the labour-intensive nature of radium sampling often hinders the collection of large datasets.

Estuaries act as a pathway for solutes to enter the coastal ocean especially at times of high flow (Maher et al., 2015). Previous studies have shown that estuaries are significant sources of greenhouse gases, nutrients and dissolved carbon to the coastal ocean and have highlighted the important role SGD plays in this delivery process (Sadat-Noori et al., 2016a,b; Reading et al., 2017). SGD can also deliver low oxygen waters to coastal environments and cause hypoxia (McCoy et al., 2011; Peterson et al., 2016). Low dissolved oxygen (DO) concentrations in the water column can disrupt key biogeochemical processes such as respiration and nitrification (Roberts et al., 2012). However, the role of SGD and porewater exchange processes in influencing hypoxia and anoxia conditions in coastal estuarine waters is still poorly understood.

Mangroves can play a significant role in biogeochemical processes in tropical coastal waters (Bouillon et al., 2008; Stieglitz et al., 2013; Tait et al., 2016). Mangroves can sequester carbon from the atmosphere, store large amounts of carbon below-ground, provide a major pathway for carbon to travel from land to ocean and can facilitate nutrient cycling (Sanders et al., 2016; Sippo et al., 2017). This makes mangroves within estuarine ecosystems an important component of the global carbon cycle (Bouillon et al., 2008). Additionally, mangroves support many burrowing animal species (Nagelkerken et al., 2008). Burrows create large pore spaces in sediments that enhance the interaction between surface waters and porewater. Crab burrows can be extensive with previous studies suggesting that the subsurface area can be up to six times greater than the aboveground area (Stieglitz et al., 2000). Therefore, any alteration to mangrove habitat, including aquaculture expansion, coastal development and urbanization could alter SGD or porewater exchange rates and the release of dissolved carbon and nutrients to marine coastal waters.

SGD hotspots along rivers and estuaries can occur in areas with different vegetation, modifications and drainage settings. Sadat-Noori et al. (2015) used multiple radon ( $^{222}\text{Rn}$ ) time series stations along the length of a subtropical tidal estuary to detect groundwater discharge rates at the mouth, mid-section and upstream parts of an estuary. The study showed a well-defined area of high groundwater discharge near sand dunes. Additionally, the study showed that multiple time series stations decreased the overall uncertainty of groundwater discharge estimates from 41% with one station to 23% with four by breaking down the estuary into smaller sections which reduces errors related to upscaling. Another way to further reduce uncertainties in mass balance models is through the use of radium. A radium mass balance model does not require estimating evasion rates which often have large uncertainties due to the use of empirical models used in evasion calculations. However, to date there has been no multiple radium isotope time series measurements in estuaries to quantify SGD rates from various sections within the estuary.

In this study, we hypothesize that estuarine areas surrounded by mangroves play an important role in groundwater exchange processes. Previous studies have used time series radium measurements to quantify SGD rates with a measuring station at the mouth of the estuary which cannot reveal the spatial distribution of SGD (Peterson et al., 2008a; Sadat-Noori et al., 2015; Tait et al., 2017). This study builds on the literature by combining multiple radium time series measurement stations along an estuarine gradient accompanied with a detailed spatial survey to quantify SGD rates and identify SGD hotspots. With our detailed sampling strategy, uncertainties can be reduced when estimating SGD rates.

## 2. Material and methods

### 2.1. Study Site

The study was conducted at Coffs Creek estuary located at Coffs Harbour, NSW, Australia ( $33^{\circ}17'48''\text{S}$ ,  $153^{\circ}08'14''\text{E}$ ) (Fig. 1). Coffs Creek estuary is a tidal estuary, 6 km long, an average of 40 m wide and has an average depth of 0.6 m. The estuary's lower and central parts are surrounded by mangroves and saltmarsh vegetation with the highest densities in the mid estuary. Mangroves in Coffs Creek estuary cover an area of about 187,000  $\text{m}^2$ . The estuary has a catchment area of about 25  $\text{km}^2$  of which 80% is dominated by urban development and agriculture, while just ~16% is considered undisturbed (Roper et al., 2011). The estuary accumulates sands in the first 4.5 km from the mouth of the estuary as a result of waves and storms. In the upper part of the estuary, silt and clay fractions increase. The region receives an average annual rainfall of 1600 mm, has a sub-tropical climate with hot and wet summers and cold and dry winters. The wettest and driest months are February and September, respectively. The water demand of Coffs Harbour is around 18 ML per day. Potable water for the city is pumped from the local rivers outside the catchment (Orara River, Nymboida River and Shannon Creek) and the wastewater from the city is treated before the effluent is disposed offshore (Coffs Harbor City Council, 2013).

A field campaign was conducted from the 1<sup>st</sup> to 7<sup>th</sup> March 2016. Although sampling was conducted in what is normally the wettest time of the year, El Niño conditions led to drier than normal conditions and thus lower than expected surface water flow in the estuary. The area did not receive any rainfall two weeks prior to the time series data collection. However, it received a total of 11.7 mm of rainfall during the sampling week with 7.2 mm on 4<sup>th</sup> March. Overall, no surface water runoff was observed during sampling from stormwater drains flowing into the estuary.

### 2.2. Radium time series measurements

For the time series experiment, radium samples were collected at five sites (A, B, C, D, E) along the length of the estuary over six consecutive days with the first site (Site A;  $30^{\circ}17'47.38''\text{S}$ ,  $153^{\circ}8'11.85''\text{E}$ ) being located at the mouth of the estuary and the last site (Site E;  $30^{\circ}17'35.99''\text{S}$ ,  $153^{\circ}6'38.31''\text{E}$ ) at the freshwater endpoint of the estuary (Fig. 1). Sites B ( $30^{\circ}17'30.97''\text{S}$ ,  $153^{\circ}7'25.97''\text{E}$ ), C ( $30^{\circ}17'41.17''\text{S}$ ,  $153^{\circ}7'5.40''\text{E}$ ) and D ( $30^{\circ}17'40.35''\text{S}$ ,  $153^{\circ}7'13.13''\text{E}$ ) were located in between. For all sites (except E), radium samples were collected every 1–2 h for at least 24 h to capture two full tidal cycles and diurnal patterns. Site E samples were collected every 6 h for 24 h as it was not influenced by tides. To account for stratification in the water column between Sites C and D, radium samples for these sites were collected from the top and bottom layer of water column.

For each sample, estuary water was pumped from mid estuary into 100 L barrels which were drained through manganese-impregnated fibre at about  $1 \text{ L min}^{-1}$  with the fibres absorbing the dissolved radium from the water (Moore, 2010). The fibres were then transported to the laboratory within a day, rinsed with radium-free water to remove residual salt and particles, partially dried and placed in a Radium Delayed Coincidence Counter (RaDeCC) for measuring  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  activities (Moore and Arnold, 1996) with analytical uncertainties calculated following Garcia-Solsona et al. (2008). Thereafter, samples were stored for a period of more than one month ( $>5$  half-lives of  $^{224}\text{Ra}$ ) before being reanalysed through the RaDeCC to measure  $^{228}\text{Th}$  activity to estimate the  $^{224}\text{Ra}$  excess.

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