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# Sources and sinks of methane and nitrous oxide in the subtropical Brisbane River estuary, South East Queensland, Australia

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ABSTRACT

This study investigated sources and sinks of methane ($CH_4$) and nitrous oxide ($N_2O$) in the Brisbane River estuary, Australia. Field measurements and laboratory incubation experiments were performed to determine the contribution of the water column, sediments, influent creeks and discharge from a wastewater treatment plant, to greenhouse gas levels occurring in the estuary. Sampling was conducted at 16 sites along the estuary with more detailed studies at three of these sites, located in the lower, middle and upper estuarine reaches, respectively. The estuary is a source of $CH_4$ and $N_2O$ with their saturation in the surface water ranging between 2160 and 26,900% for $CH_4$, and between 140 and 230% for $N_2O$, relative to their respective atmospheric concentrations. Estuarine sediments were identified as sources for both $CH_4$ and $N_2O$, as concentrations for both dissolved gases were higher in the pore water at the sediment surfaces in comparison to the water column samples at all sites. Methane and $N_2O$ were produced in silty sediments as shown in sediment-water incubations. Creeks were detected to be a source of $CH_4$ and $N_2O$, as their concentrations were higher in the creek in comparison to those measured at the confluence of the creek and the estuary. The estuarine water column was identified as a $CH_4$ sink; however, the water column was neither a sink nor a source for $N_2O$. A wastewater treatment plant discharging effluent into the tributary did not contribute significantly to the creek $CH_4$ levels, but may have elevated $N_2O$ at the direct point of discharge. A budget estimate of the Brisbane River estuary showed that sediments are not the sole source of $CH_4$, as the sediment-water fluxes are far less (1%) than what is consumed, emitted or transported to the ocean. An unknown $CH_4$ source is therefore likely present, which is yet to be revealed. In contrast, most of $N_2O$ produced in the estuarine sediments was emitted to the atmosphere.

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## 1. Introduction

Coastal features such as estuaries have been identified as natural sources of the highly potent greenhouse gases (GHGs), methane ($CH_4$) and nitrous oxide ($N_2O$) (Bange, 2006; Ferron et al., 2007; Musenze et al., 2014). However, it is often difficult to quantify the gases and identify the estuarine sources and sinks where contributions from the water column and sediments (Law et al., 1992; Hovland et al., 1993; Bange, 2006; Zhang et al., 2008), freshwater streams (Ferron et al., 2007; Call et al., 2015), and sewage discharges need to be considered (De Angelis and Scranton, 1993; Toyoda et al., 2009; Beaulieu et al., 2010; Teixeira et al., 2010). Methane concentrations and emissions are found to be high in estuaries and streams surrounded by forests, agricultural land and peatlands (De Angelis and Lilley, 1987; Jones and Mulholland, 1998; Hope et al., 2001; Dawson et al., 2004; Kristensen et al., 2008). It is seen that high $N_2O$ concentrations and emissions occur in systems receiving freshwater from streams or groundwater, this is especially when fertilizer has been applied to the surrounding land producing nutrient rich runoff to the stream (Mosier et al., 1998; McMahon and Dennehy, 1999; Machefert et al., 2004).

There are many environmental and ecosystem factors that influence the levels of these GHGs in estuaries. Dissolved oxygen (DO) is a key factor governing whether $CH_4$ is produced or consumed within a system. In well-mixed estuarine systems, oxic processes occur in the water column and in oxygenated surface sediments. In contrast, anoxic processes primarily occur in the deeper, anoxic

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layers of the sediments. Methane is mainly formed by microbial methanogenesis under anoxic conditions (Canfield et al., 2005). However, $CH_4$ production can be strongly inhibited in sulfate-rich environments, e.g. estuaries or oceans (Poffenbarger et al., 2011; Marton et al., 2012), as methanogens and sulfate-reducing bacteria (SRB) compete for electron donors such as hydrogen or acetate (Oremland and Polcin, 1982; Kristjansson and Schönheit, 1983). Oxidation of $CH_4$, which can occur under aerobic as well as anaerobic conditions (Bange, 2006), conversely functions as a major $CH_4$ sink (De Angelis and Scranton, 1993). Similarly, the production or consumption of $N_2O$ is also influenced by DO. Nitrous oxide can be produced under oxic conditions as a byproduct of nitrification, or at oxic–anoxic interfaces, as an intermediate of denitrification (Ward, 1996; Codispoti et al., 2001). Denitrifying bacteria can also reduce $N_2O$ to nitrogen gas ($N_2$).

The contribution of coastal regions to the global $CH_4$ and $N_2O$ budgets is still uncertain (Ferron et al., 2007), and there are few studies that examine the production and consumption processes that ultimately determine $CH_4$ and $N_2O$ emissions from these aquatic systems. There is a severe lack of knowledge especially for estuaries located in subtropical regions, particularly in the Southern Hemisphere (Ortiz-Llorente and Alvarez-Cobelas, 2012). The few existing studies show that the estuarine sediments and water columns play important roles in producing $CH_4$ and $N_2O$ (Law et al., 1992; Hovland et al., 1993; De Bie et al., 2002; Bange, 2006; Zhang et al., 2008).

A recent study of the subtropical, Brisbane River estuary shows this to be a strong source of $CH_4$ and $N_2O$ all-year-round, with pronounced spatial and temporal variability in $CH_4$ and $N_2O$ surface water concentrations and emissions (Musenze et al., 2014). However, the sources and sinks of these GHGs in the surface waters of the Brisbane River estuary have not been studied.

The main objective of our study was to determine the $CH_4$ and $N_2O$ sources and sinks in the subtropical Brisbane River estuary. The driving research questions were if the 1) sediments, 2) water column, 3) creeks and 4) wastewater treatment plant (WWTP) effluent were a source or a sink of $CH_4$ and $N_2O$. To investigate the role of sediments, a measurement survey was conducted at 16 sites along the estuary during which the gradients of $CH_4$ and $N_2O$ between the sediment pore water and surface water concentrations were measured at all sites. The $CH_4$ and $N_2O$ sediment-water fluxes were further determined by laboratory incubations of sediment cores from three selected sites, located in the lower, middle and the upper estuarine reaches. Laboratory incubations of site water from these selected sites were also conducted to study the water column contributions. The $CH_4$ and $N_2O$ concentrations in surface water of a selected creek were measured, and compared to the $CH_4$ and $N_2O$ concentrations at the confluent. Finally, the $CH_4$ and $N_2O$ concentrations in the WWTP effluent discharged into the creek were measured to determine if wastewater effluent had a direct impact on the $CH_4$ and $N_2O$ levels in the creek.

## 2. Materials and methods

### 2.1. Study site

The Brisbane River estuary is located in the Southern Hemisphere in subtropical, South East Queensland, Australia. The estuary meanders through urban Brisbane and flows into Moreton Bay with an average annual discharge of $1400 \times 10^6$ $m^3$ (Eyre et al., 1998). The estuary has confluences with the Bremer River, Oxley Creek (Fig. 1), and smaller creeks (not illustrated in Fig. 1). The estuary has a surface area of 19 $km^2$ and a catchment area of 13,643 $km^2$. The natural tidal limit reaches 16 km along the length of the river from the estuary mouth (Holland et al., 2001), however,

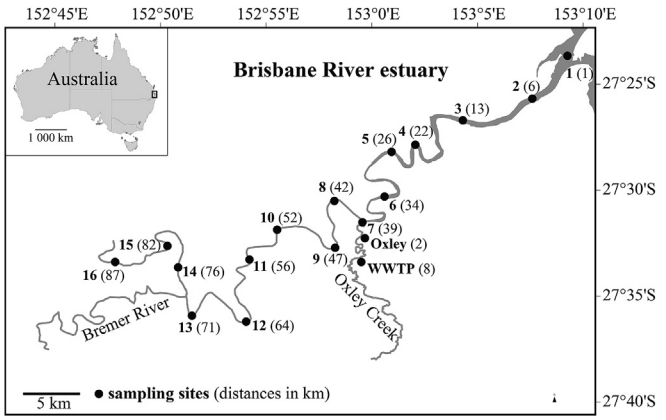


**Fig. 1.** Locations of the field sampling sites in the Brisbane River estuary, South East Queensland, Australia. Detailed studies were conducted at three selected sites, located in the lower- (site 2), middle- (site 7) and upper (site 15) reaches. Studies were also conducted upstream in Oxley Creek at the site Oxley and at the site WWTP. Site distances are given from the estuary mouth except for the Oxley and WWTP sites, which are given as distances from the creek/estuary confluence (site 7). The tidal limit and water exchange reaches 86 km.

it now extends to 86 km (Davie et al., 1990; Holland et al., 2001) due to major channel modifications (Eyre et al., 1998). Located in a subtropical region, the Brisbane River estuary has relatively high water temperatures (ranging between 16 and 29 °C among seasons (EHMP, 2014)), and can experience distinct, seasonal rainfall patterns in contrast to estuaries located in temperate regions. The sampling trip days were in the months of August and October, both belonging to the dry season (Yu et al., 2013) and had average water temperatures of 18 °C and 24 °C, respectively. Detailed characteristics of the 16 sampling sites chosen for the sampling survey such as the surface water temperature and salinity as well as the averaged water column DO concentrations can be found in Table 1.

### 2.2. Study design

In order to determine the contributions of the water column or sediments to act as $CH_4$ or $N_2O$ sources or sinks we measured the surface water and sediment surface pore water concentrations in an extensive sampling survey (sampling conducted in August 2013) at 16 sites along the Brisbane River estuary (Fig. 1, Table 1). Detailed data for each site such as the site distance from the estuary mouth, the GPS coordinates or the mid-channel water depth are presented in Table 1. Based on the survey results three sites which capture different estuarine sections were chosen for further detailed studies, 1 site in each of the lower- (site 2), the middle- (site 7), and the upper (site 15) estuary sections. This site choice covered one site (lower) which was highly impacted by the tides, one site (middle) which receives the majority of water from upstream reaches and creeks while still being impacted by the tides and one site (upper) which was located at the tidal limit and mainly received water from upstream freshwater reaches with no dilution of bay water. From these sites, water column and sediment-water samples were examined in laboratory incubation studies (sampling conducted in October 2013) to examine the role of the water column and sediments in $CH_4$ and $N_2O$ production or consumption. To examine the role of creeks to the river GHG levels, sampling was conducted at site 7 and upstream in Oxley Creek (sampling conducted in August 2013). Additionally, $CH_4$ and $N_2O$ concentrations in the Oxley WWTP effluent discharged to Oxley Creek (WWTP site sampling conducted in August 2013, Fig. 1) were examined to determine if the effluent was a source of these gases to the creek.

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