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Methane and nitrous oxide emissions from a subtropical estuary (the Brisbane River estuary, Australia)

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HIGHLIGHTS

- The estuary is a strong source of atmospheric methane and nitrous oxide.
- Emissions had strong spatial-temporal variability with unclear seasonal patterns.
- Dissolved gas saturation comparable to that in tropical rivers and polluted estuaries.
- Emissions are dominated by N2O, which positively correlated with NOx concentrations.
- Currently existing models contribute to uncertainty in emission estimates.

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ABSTRACT

Methane (CH_4) and nitrous oxide (N_2O) are two key greenhouse gases. Their global atmospheric budgeting is, however, flout with challenges partly due to lack of adequate field studies determining the source strengths. Knowledge and data limitations exist for subtropical and tropical regions especially in the southern latitudes. Surface water methane and nitrous oxide concentrations were measured in a subtropical estuarine system in the southern latitudes in an extensive field study from 2010 to 2012 and water–air fluxes estimated using models considering the effects of both wind and flow induced turbulence. The estuary was found to be a strong net source of both CH_4 and N_2O all-year-round. Dissolved N_2O concentrations ranged between 9.1 ± 0.4 to 45.3 ± 1.3 nM or 135 to 435% of atmospheric saturation level, while CH_4 concentrations varied between 31.1 ± 3.7 to 578.4 ± 58.8 nM or 1210 to 26,430% of atmospheric saturation level. These results compare well with measurements from tropical estuarine systems. There was strong spatial variability with both CH_4 and N_2O concentrations increasing upstream the estuary. Strong temporal variability was also observed but there were no clear seasonal patterns. The degree of N_2O saturation significantly increased with NO_x concentrations ($r^2 = 0.55$). The estimated water–air fluxes varied between 0.1 and 3.4 mg N_2O m^{-2} d^{-1} and 0.3 to 27.9 mg CH_4 m^{-2} d^{-1}. Total emissions (CO_2-e) were N_2O (64%) dominated, highlighting the need for reduced nitrogen inputs into the estuary. Choice of the model(s) for estimation of the gas transfer velocity had a big bearing on the estimated total emissions.

1. Introduction

Methane (CH_4) and nitrous oxide (N_2O) are two atmospheric trace gases that have attracted great scientific attention. They are potent greenhouse gases (GHG) with respective global warming potentials of around 25 and 300 times that of carbon dioxide (CO_2), on a 100-year horizon (Ehhalt et al., 2001; IPCC, 1995, 2007). Nitrous oxide is also a strong Ozone depleting substance (Ravishankara et al., 2009). Together, these long-lived greenhouse gases contribute nearly 30% of the total warming due to greenhouse gases resulting from anthropogenic influences (Cicerone and Oremland, 1988; IPCC, 2007). The current atmospheric CH_4 concentration is nearly triple its pre-industrial level and N_2O is also around 20% higher than its pre-industrial level. Moreover, their concentrations are still on the rise (IPCC, 1995; Rigby et al., 2008).

Aquatic systems are likely significant sources for both atmospheric CH_4 and N_2O (IPCC, 2007; Seitzinger et al., 2000). Coastal systems (and specifically estuaries) are presumed to be a strong aquatic source of emissions. They are estimated to contribute up to 60% and 75% of the respective global oceanic N_2O and CH_4 emissions (Bange et al., 1994, 1996; Seitzinger and Kroeze, 1998). The reported concentrations and fluxes for both CH_4 and N_2O are, however, widely ranging (Bange et al., 1996; Upstill-Goddard et al., 2000) especially along climatic

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regional divides (Seitzinger et al., 2000). Both dissolved gas concentrations and fluxes (especially for CH_4) have also been found to have high spatial and temporal variability in coastal systems (Bange, 2006). For estuaries, spatial variability has been specially linked to the activities within the surroundings drained by these systems. For instance, elevated dissolved GHG concentrations and fluxes have been reported in rivers and estuarine segments draining farmlands, within vicinities of industrial and domestic wastewater effluent discharges, and generally high organic matter input terrestrial systems (Beaulieu et al., 2010; Richey et al., 1988; Law et al., 1992; Machefert et al., 2004; Toyoda et al., 2009). On a temporal basis, both concentrations and fluxes of CH_4 and N_2O are generally higher in the warmer summer season and low in the cold winter season (Beaulieu et al., 2010; Clough et al., 2007; de Angelis and Scranton, 1993). However, the absence of clear seasonal patterns has also been reported (Beaulieu et al., 2008; Stow et al., 2005). This highlights a high degree of systems diversity and the importance of region specific measurements.

Attempts have been made at establishing regional and global aquatic GHG emission budgets (Bange, 2006; Cicerone and Oremland, 1988). However, uncertainties and immense challenges still surround these estimates (Cicerone and Oremland, 1988; Nevison et al., 1995). One such major challenge is that there are limited emission measurements (Bange, 2006; Beaulieu et al., 2010; US EPA, 2010) from tropical and subtropical aquatic systems, especially in the southern latitudes, despite the fact that aquatic systems in tropical and subtropical regions have been reported to be strong sources of GHGs (Bastviken, 2009; Richey et al., 1988; Koné et al., 2010). Additionally, many emission estimates are often biased by limited spatial and temporal coverage (Nevison et al., 1995). Given the big range in source strengths with the apparent spatial and temporal variability, it is now evident that the knowledge gap due to lack of a good understanding of regional emissions hampers effective global GHG budgeting. Emission studies from the currently understudied systems will help bridge this gap and improve GHG accountability.

This study is aimed at quantifying CH_4 and N_2O emissions from a subtropical estuarine system — the Brisbane river estuary. Special attention was paid to assessment of both temporal and spatial variability. Measurements were undertaken at 18 monitoring stations along the estuary over a period of two years (October 2010 (spring) to August 2012 (winter)). CH_4 and N_2O fluxes were estimated using the thin boundary layer approach with the gas transfer velocity estimated on the basis of both wind speed and bottom flow turbulence.

2. Materials and methods

2.1. Physical setting and monitoring stations

The Brisbane River (344 km) is the longest river in Southeast Queensland, Australia. It is dammed in the upper reaches forming lake Wivenhoe and meanders through the city of Brisbane before discharging into Moreton Bay. Heavy dredging for bottom sand extraction has led to increased riverbed and bank erosion, high turbidity, increased sedimentation in Moreton Bay, and changes in tidal hydraulics. Dredging extended the river's tidal influence limit from 16 km to 85 km upstream (O'Brien et al., 2001). The river has a catchment area of 13,100 km^2 (Allen et al., 2011). The studied estuarine section (86 km) (Fig. 1) has a stream network length of 2475 km formed by 19 tributaries and creeks (HWPL, 2010). The estuary also receives effluents from seven wastewater treatment plants (wwtp), all of which perform biological chemical oxygen demand (COD) and nitrogen removal with the discharges of 40–100 mg COD L^{-1} (Law, unpublished data) and 2.2–50 mg N L^{-1} (Farre et al., 2010), respectively. The estuary has also been at the centre of major flooding events with the January 2011 flood being the most recent devastating one.

For the purpose of this work, we have divided the estuary into three sections: lower section (0–33 km), middle section (33–60 km) and upper section (60–86 km). Measurements were done from 18 stations within the estuary (Fig. 1). Sixteen of these stations are part of the bigger monitoring network established under the environmental health monitoring programme (EHMP) – Department of Environment and Resources Management (DERM) – Queensland Government, being used for monthly estuarine water quality monitoring. The 2 non-EHMP stations were established to monitor the impact of the Bremer River at the confluence with the Brisbane River estuary. All stations are described by their location (as distance) from the sea.

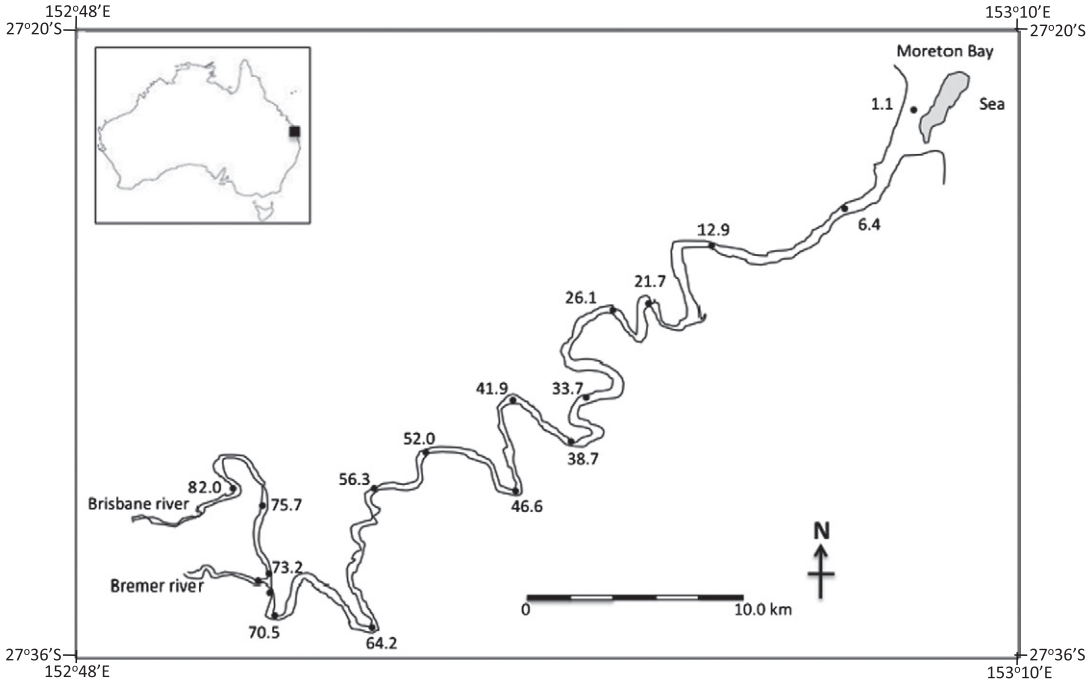

Fig. 1. Map of the Brisbane River estuary. Solid dots are stations that were used for field sampling. Numbers are relative distances (km) from the estuary's mouth/sea. Inset is the map of Australia showing location of the study area.