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### Spatial and temporal variation of nitrous oxide and methane flux between subtropical mangrove sediments and the atmosphere

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#### **Abstract**

We quantified spatial and temporal variations of the fluxes of nitrous oxide  $(N_2O)$  and methane  $(CH_4)$  and associated abjotic sediment parameters across a subtropical river estuary sediment dominated by grey mangrove (Avicennia marina). N<sub>2</sub>O and CH<sub>4</sub> fluxes from sediment were measured adjacent to the river ("fringe") and in the mangrove forest ("forest") at 3-h intervals throughout the day during autumn, winter and summer.  $N_2O$  fluxes from sediment ranged from an average of  $-4 \mu g$  to  $65 \mu g N_2O m^{-2} h^{-1}$  representing  $N_2O$  sink and emission. CH<sub>4</sub> emissions varied by several orders of magnitude from 3 µg to 17.4 mg CH<sub>4</sub> m<sup>-2</sup>h<sup>-1</sup>. Fluxes of N<sub>2</sub>O and CH<sub>4</sub> differed significantly between sampling seasons, as well as between fringe and forest positions. In addition, N2O flux differed significantly between time of day of sampling. Higher bulk density and total carbon content in sediment were significant contributors towards decreasing N<sub>2</sub>O emission; rates of  $N_2O$  emission increased with less negative sediment redox potential  $(E_h)$ . Porewater profiles of nitrate plus nitrite  $(NO_x^-)$  suggest that denitrification was the major process of nitrogen transformation in the sediment and possible contributor to  $N_2O$ production. A significant decrease in  $CH_4$  emission was observed with increasing  $E_h$ , but higher sediment temperature was the most significant variable contributing to CH<sub>4</sub> emission. From April 2004 to July 2005, sediment levels of dissolved ammonium, nitrate, and total carbon content declined, most likely from decreased input of diffuse nutrient and carbon sources upstream from the study site; concomitantly average CH<sub>4</sub> emissions decreased significantly. On the basis of their global warming potentials, N<sub>2</sub>O and CH<sub>4</sub> fluxes, expressed as CO<sub>2</sub>-equivalent (CO<sub>2</sub>-e) emissions, showed that CH<sub>4</sub> emissions dominated in summer and autumn seasons (82–98% CO<sub>2</sub>-e emissions), whereas N<sub>2</sub>O emissions dominated in winter (67–95% of CO<sub>2</sub>-e emissions) when overall CO<sub>2</sub>-e emissions were low. Our study highlights the importance of seasonal N<sub>2</sub>O contributions, particularly when conditions driving CH<sub>4</sub> emissions may be less favourable. For the accurate upscaling of N<sub>2</sub>O and CH<sub>4</sub> flux to annual rates, we need to assess relative contributions of individual trace gases to net CO<sub>2</sub>-e emissions, and the influence of elevated nutrient inputs and mitigation options across a number of mangrove sites or across regional scales. This requires a careful sampling design at site-level that captures the potentially considerable temporal and spatial variation of N<sub>2</sub>O and CH<sub>4</sub> emissions.

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#### 1. Introduction

Quantification of the trace gases, nitrous oxide ( $N_2O$ ) and methane (CH<sub>4</sub>) is a subject of great interest because accurate information is required to determine the contribution of these gases to global greenhouse gas fluxes (Khalil et al., 2002; Wuebbles and Hayhoe, 2002). The

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Intergovernmental Panel for Climate Change (IPCC, 2001) has reported increased concentrations in  $N_2O$  and  $CH_4$  since industrial times, a concern since both gases, although present in lower concentrations to that of  $CO_2$ , have 296 ( $N_2O$ ) and approximately 23 ( $CH_4$ ) times the global warming potential of  $CO_2$  over a 100-year time period (IPCC, 2001).

It is estimated that natural sources of  $N_2O$  and  $CH_4$  account for 44–54% of  $N_2O$  emissions (9.6–10.8 Tg  $N_2O$  year<sup>-1</sup>) and 30–40% of  $CH_4$  emissions (150–237 Tg  $CH_4$  year<sup>-1</sup>) (IPCC, 2001). Tropical soils and wetlands are considered significant natural sources, contributing approximately 22–27% ( $N_2O$ ) and 24% ( $CH_4$ ), respectively, towards this inventory (IPCC, 2001; Whalen, 2005). Most wetland studies, however, have been carried out in temperate areas. Empirical studies in tropical wetlands are rare, and estimates of  $N_2O$  and  $CH_4$  fluxes in tropical wetlands carry a high level of uncertainty (Purvaja and Ramesh, 2001; Purvaja et al., 2004). Tolhurst and Chapman (2005) noted that despite considerable effort, processes controlling variation in intertidal sediment properties over time remain poorly understood, as most studies have focused on only one time or spatial scale.

A major component of tropical and subtropical coastal wetlands are mangrove ecosystems, which occupy the intertidal zone of estuaries, bays, inlets and gulfs and part of the riparian zone (Alongi, 2002). Unlike temperate wetlands where temperature fluctuation is extreme, mangrove ecosystems are restricted to warm waters, with greatest abundance and species diversity found in subtropics and tropics (Alongi, 2002). Mangrove communities occur across a range of salinity levels and different types of sediments (Lovelock, 1993; Duke et al., 1998). Sediment properties of mangrove communities range from coarse sands to black fine estuary muds, and associated with sediment properties, different abiotic and biotic conditions prevail. Microbial processes affecting trace gas production are regulated by many parameters including oxygen availability, sediment temperature and water content, sediment redox potential  $(E_h)$ , salinity, pH, and microbially available reduced carbon and nitrogen sources (Bauza et al., 2002; Whalen, 2005). Mangrove sediment is often considered to be oligotrophic, but mangroves are not restricted to low nutrient environments (Feller et al., 2003). There is growing interest in using mangroves for treating domestic, agricultural and industrial wastewaters (Tam and Wong, 1999). Studies have reported some capacity for mangrove estuaries to tolerate intense shrimp pond effluent (Trott and Alongi, 2000), to remove ammonium from wastewater (Tam and Wong, 1999) and to depurate nitrate from treated-sewage effluent (Corredor and Morell, 1994). The extent of such nutrient storage and conversion capacity of mangrove sediments and the potentially associated trace gas emissions remains poorly understood (Gauiter et al., 2001; Alongi, 2002).

It has been suggested that trace gas emissions from coastal mangroves are negligible compared to trace gas emissions originating from wetlands (Sotomayor et al., 1994). However, Purvaja and Ramesh (2001) observed several human-induced factors that enhance CH<sub>4</sub> emissions from mangroves to the atmosphere, and there is evidence that additional nitrogen inputs in mangroves increased N<sub>2</sub>O emissions (Kreuzwieser et al., 2003). Increasingly, riverine mangrove sediments are considered to contribute to N<sub>2</sub>O and CH<sub>4</sub> emissions (Sotomayor et al., 1994; Corredor et al., 1999; Purvaja and Ramesh, 2001; Kreuzwieser et al., 2003). Another consideration is the presence of pneumatophores, the mangrove roots linking sediment to atmosphere and which have been implicated in increased emissions from mangrove sediments (Purvaia et al., 2004). The potential for mangrove sediments to have significant N<sub>2</sub>O and CH<sub>4</sub> emission is of concern since mangroves occupy 181 000 km<sup>2</sup> of coastline (Alongi, 2002). As human expansion continues along riverine and coastal shorelines, mangroves may be subject to anthropogenic inputs including sewerage, aquaculture and agriculture, which have potential for adding nutrients to mangrove ecosystems (Alongi, 2002).

Some research has been undertaken to measure mangrove gas fluxes using micrometeorological techniques (Mukhopadhyay et al., 2001), however most studies have used the "closed chamber technique", which is more readily accessible, inexpensive, easier to use, and which may capture very small variations in N<sub>2</sub>O not measurable by flux-gradient techniques (IAEA, 1992; Griffith et al., 2002). The cost-efficient set-up of closed chambers is offset by labour intensiveness if frequent measurements are performed (Dalal et al., 2003). In agricultural systems, attention has been given to sampling strategies for temporal (Buendia et al., 1998; Smith and Dobbie, 2001) and spatial variation (Ambus and Christensen, 1994; Ball et al., 1997; Weitz et al., 1999), however, no consensus exists for measurement protocols in wetland and mangrove ecosystems.

This study was undertaken in subtropical mangrove sediments along the Brisbane River, at a site located adjacent to a treated sewage outlet, which also receives diffuse nutrient inputs from upstream (Dennison and Abal, 1999). Using the closed chamber technique we measured gas flux from exposed estuary sediments at low tide. We aimed to determine whether distinct temporal and spatial variation occurs in N<sub>2</sub>O and CH<sub>4</sub> flux throughout the day and in different seasons, and whether spatial variation exists in flux. To address the second aim, we compared emissions from sediments close to the river edge ("fringe") and in the mangrove forest ("forest").

#### 2. Materials and methods

#### 2.1. Site description

Sampling was carried out approximately 46.5 km upstream from the mouth of the Brisbane River, in Chelmer, South East Queensland, Australia (27°33′S, 152°59′E), on the northern bank of a river estuarine fringe dominated by

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