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Dynamics and exchange fluxes of methane in the estuarine mangrove environment of the Sundarbans, NE coast of India

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HIGHLIGHTS

- Dynamics of methane in the world's largest mangrove forest, the Sundarbans.
- Distribution of methane and methanogenesis in the mangrove sediment.
- Dissolved methane in the estuarine water and its sources.
- Methane exchange fluxes at different environmental interfaces.
- Contribution of the Sundarbans in the atmospheric methane budget.

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ABSTRACT

The distribution and exchange fluxes of methane (CH_4) were measured in a mangrove vegetated island and its bordering estuarine system of the Sundarbans mangrove biosphere from June 2010 to December 2011 on monthly basis. The onset of methane production is evident in the forest sediment at about 25 cm deep sediment layer under strong redox condition having an average E_h value of −175.7 mV and showing a 2.8 folds increase in the pore water dissolved methane concentration at that depth in comparison to the surface layer. The average diffusive flux of methane from this methane producing layer to surface was calculated to be 591 ± 106 nmol m^{-2} d^{-1}. The depth profiles of NO_2^-–N, SO_4^{-2}–S, acid volatile sulphide, organic carbon and dissolved methane in the sediment cores from inter-tidal zones showed distinct trends representing signatures of denitrification, sulfate reduction and methanogenesis in the sediment layers. The methane emission from the sediment–atmosphere interface was observed to be maximum during monsoon and higher emission rates was recorded from upper littoral zone. The annual average atmospheric methane mixing ratio was 2.038 ± 0.07 ppmv. This mangrove biosphere was found to act as source for methane during monsoon while as sink during pre and postmonsoon seasons. Estuarine surface water showed a very high degree of super saturation about 2748 ± 730% for dissolved methane at an annual basis and act as a significant source of methane having an annual average exchange flux of 408 ± 110 nmol m^{-2} h^{-1}. A box model approach has been adopted at annual basis to understand the distribution and dynamics of methane in this mangrove environment.

1. Introduction

Methane is one of the most important greenhouse gases and an important component of atmospheric chemistry. CH_4 has the second-largest radiative force among the greenhouse gases (Ramaswamy et al., 2001). In the atmosphere methane influences the lifetimes of various chemically reactive trace gases owing to a photochemical feedback on the hydroxyl radical (Crutzen and Zimmerman, 1991; Krol et al., 1998). Wetlands, paddy fields, ruminants are most important source of atmospheric methane (Zhuang et al., 2009; Aluwong et al., 2011).

Wetlands (Tundra, Bog, Fen, Marsh, Swamp) covers only 5% of Earth's land surface (Matthews and Fung, 1987; Aselmann and Crutzen, 1989), it contributes about 20% of the total annual emission of CH_4 to the atmosphere and is the largest natural source of atmospheric CH_4 (Wuebbles and Hayhoe, 2002; Kapoor, 2005). The significant spatial and temporal variation in CH_4 emission from natural wetlands has been often studied (Christensen et al., 2003; Ding et al., 2004; Yang et al., 2006).

Tropical coastal mangrove dominated wetlands are one of the most productive ecosystems of the world (Odum and Heald, 1975) and there higher degree of leaf litters deposition together with faster

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tidal sedimentation offer inadequate period for aerobic decomposition of organic matter resulting organic carbon rich anoxic condition in the subsurface sediment layer. Moreover, the mangroves sediments are rich in clay content which reduces the porosity of the sediment and helps in the formation and retention of anoxic condition. This anoxic condition induces several microbial processes in the subsurface mangrove sediments like denitrification, sulphate reduction, methanogenesis, and other redox reactions, which may contribute significantly to nutrient turnover (Alongi and Christoffersen, 1992) and production of gases like methane, nitrous oxide, and hydrogen sulphide. In particular, gaseous methane is produced when the redox potential of the sediment reached to less than −150 mV (Wang et al., 1993). A major part of produced methane get dissolved in the pore water at *in situ* high pressure resulting significant super saturation, which finally released from the sediment to the atmosphere through diffusion of dissolved methane and the residual part undergoes ebullition as gas bubbles (Neue et al., 1997). The emitted methane at the soil atmosphere interface gets vertically mixed by the micrometeorological processes and atmospheric turbulence, enriching its atmospheric mixing ratio at regional level (Mukhopadhyay et al., 2002). The atmospheric methane also undergo photochemical transformation (Wagner et al., 2002) based on the prevailing regional climatic factors and finally constitute the mangrove biosphere–atmosphere fluxes at the boundary layer between the mangrove forest and the atmosphere.

The methane enriched pore water from mangrove forest infiltrate into adjacent estuarine system enriching the dissolved methane concentration in the water column. In the estuaries except the above source a significant amount of methane may be produced *in situ* in anoxic micro-environments such as organic-rich sediments or oxygen deficient waters (Iversen et al., 1987; Chanton et al., 1989; de Angelis and Lee, 1994). In estuarine water, a part of dissolved methane is oxidised by methane oxidising bacteria. The major part of dissolved methane escapes oxidation that enriches the surface mixed layer of the water column finally emitted at the air water interface (Upstill-Goddard, 2006).

So, the biogeochemistry of methane in the estuarine mangrove environment is complex and dependent on number of interacting factors, which prompted the present study aiming to investigate the nature of production of methane in the sediment, its transport in the mangrove environment and exchange fluxes at different interfaces from the Sundarban mangrove biosphere reserve.

2. Study location

Sundarbans is the largest mangrove forest in the world situated at the land ocean boundary of Ganges-Brahmaputra delta and the Bay of Bengal having an area of 10,200 sq. km of which, 4200 sq. km of reserved forest is spread over India and 6000 sq. km of reserved forest is in Bangladesh. The Indian part of Sundarbans is extended over an area of 9600 sq. km which is demarcated as Sundarbans Biosphere Reserve constituting of 4200 sq. km mangrove reserve forest, 1800 sq. km estuarine waterways and 3600 sq. km. reclaimed areas. The Indian part of the Sundarbans mangrove (Fig. 1) is crisscrossed by the estuarine phases of several rivers namely Mooriganga, Saptamukhi, Thakuran, Matla, Bidya, Gosaba and Haribhanga forming a large number of discrete islands. One of these Islands is the Lothian Island, covering an area of 38 km^2 has been notified as a sanctuary and is situated at the confluence of Saptamukhi river and Bay of Bengal. Mangrove like *Avicennia alba*, *Avicennia marina* and *Avicennia officinalis* are the dominant species, *Excoecaria agallocha* and *Heritiera fomes* are thinly distributed and *Ceriops decandra* is found scattered all over the island. The vertical tide range at the mouth varies from 5.2 m during spring to 1.8 m at the neap period. Mean current velocities range between 117 and 108 cm s^{-1} during low tide and high tide respectively (Mukhopadhyay et al., 2006). The sediment of study area is silty clay and composed of quartzo-feldspathic minerals like quartz, albite and microline (Ray et al., 2013). The present study has been carried out in the Lothian Island and Saptamukhi, which can ideally be taken as representative of Sundarbans mangroves to understand the dynamics and exchange fluxes of methane.

3. Materials and methods

Air, water and sediment samples were collected from inter-tidal zones of Lothian Island (21° 42.58′ N and 88° 18′ E) and Saptamukhi estuary (21° 42.396′ N and 88° 18.815′ E) from June 2010 to December 2011 on monthly basis for measuring following parameters.

3.1. Estimation of CH_4 concentration and physicochemical properties of sediment

Vertical column of sediment of 25 cm length were collected from upper littoral zone (ULZ), mid littoral zone (MLZ) and lower littoral zone (LLZ) from the inter-tidal area of the island. Collected sediment cores were sealed in polythene bags and stored in icebox for transportation to the laboratory. The sediment temperature was recorded *in situ* by using thermometer. In the laboratory, the sediment cores were sliced having 5 cm thickness representing 0–5, 5–10, 10–15, 15–20, 20–25 cm sediment column and immediately oxidation–reduction potential (E_h) were measured by platinum electrodes and Ag/AgCl reference electrode (Fiedler et al., 2003). Porosity of sediments was measured following Knab et al. (2009). For measurement of CH_4 concentration 3 cm^3 sediment samples collected with 5 ml cut-off syringes from each slices and sealed in serum vials containing 6 ml NaOH (2.5% w/v). The mixture was shaken and stored to achieve equilibrium between the slurry and the headspace. The methane concentration in the headspace was determined by gas chromatography (Varian CP3800 GC) fitted with chrompack capillary column (12.5 m × 0.53 mm) and a flame ionization detector (FID) with a mean relative uncertainty of ±2.9%. From the measured CH_4 concentration along sediment depth profile, the diffusive flux of CH_4 from deeper sediment to the surface was calculated using Fick's law of diffusion (Knab et al., 2009). The nitrite concentration of the sediment was measured taking 2 M KCl extract of sediment followed by standard spectrophotometric method (Grasshoff et al., 1983). The sulphate and acid volatile sulphide concentration in the sediment was measured using standard spectrophotometric methods (Mussa et al., 2009; Simpson, 2001). The organic carbon percentage in sediment was estimated by high temperature combustion technique (TOC analyser, Shimadzu SSM-5000A) with a mean relative uncertainty ± 2.8%.

3.2. Soil CH_4 emission rate measurement

During low tide condition CH_4 emission from the sediment surface was measured from ULZ and MLZ locations in every month during the study period using static Perspex chamber method as described by Purvaja et al. (2004) and enhancement in the CH_4 mixing ratio inside the chamber in comparison to the ambient air were measured using gas chromatography. Based on the methane enhancement in the chamber for a particular duration the methane emission from sediment surface was calculated.

3.3. Measurement of atmospheric CH_4 mixing ratio and meteorological parameters

Air samples for measurement of methane mixing ratio were collected from the top of a tower located in the middle of the island