Agricultural and Forest Meteorology 213 (2015) 33–41

Contents lists available at ScienceDirect

Agricultural and Forest Meteorology

journal homepage: www.elsevier.com/locate/agrformet

Atmospheric fluxes and photo-oxidation of methane in the mangrove environment of the Sundarbans, NE coast of India; A case study from Lothian Island

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ARTICLE INFO

Article history:
Received 22 December 2014
Received in revised form 6 May 2015
Accepted 21 June 2015
Available online 4 July 2015

Keywords:
Methane
Atmospheric flux
Photo-oxidation
Formaldehyde
Ozone
Mangrove

ABSTRACT

Atmospheric fluxes of CH_4 across different boundaries and its photo-oxidation were measured in the Sundarbans mangrove forest during June 2010–December 2011. The annual mean CH_4 emission rate from intertidal sediment and adjacent estuarine surfaces were 7.06 and 0.14 mg m^{-2} d^{-1}, respectively. Together these sources contribute 10.89 Gg of CH_4 annually to the atmosphere, of which 99.17% was from sediment. The atmospheric CH_4 mixing ratio varied between 1.693 and 2.251 ppmv, having an average of 2.004 ppmv. CH_4 exchange flux from this mangrove biosphere to the atmosphere was 0.086 mg m^{-2} d^{-1}, contributing 0.30 Gg of CH_4 annually. The annual average rate of CH_4 photo-oxidation in the forest atmosphere was 3.25×10^{-9} mg cm^{-3} d^{-1} with maximum oxidation occurring during the monsoon period and minimum oxidation occurring during the post-monsoon period. Total CH_4 photo-oxidation within the atmospheric boundary layer of the Sundarbans was 9.26 Gg annually, which acts as sink for 85.03% of the emitted CH_4. The HCHO and O_3 mixing ratios in the forest atmosphere ranged between 2.14 ± 0.06–4.08 ± 1.61 ppbv and 14.66 ± 1.88–37.90 ± 0.91 ppbv, respectively, having maximal pre-monsoon and minimal monsoon periods. The CH_4 photo-oxidation mediated productions of HCHO and O_3 within the atmospheric boundary layer were 17.5 and 139 Gg yr^{-1}, respectively. A 'sources and sinks dependent' mathematical model for atmospheric CH_4 proposed in this study can explain the seasonal variability of CH_4 and showed significant correlation with the observed values of this regional atmosphere.

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1. Introduction

Methane (CH_4) is the most abundant organic trace gas in the atmosphere. Following water vapour and carbon dioxide, CH_4 is also the most abundant greenhouse gas in the troposphere, contributing to approximately 20% of global warming (Burton and Turner, 2003; http://www.eci.ox.ac.uk/research/energy/downloads/methaneuk/chapter01.pdf). Moreover, on a per molecule basis, it is a much more effective greenhouse gas than atmospheric CO_2. Until the early 1980s it was assumed that CH_4 was a stable, nonviable component in earth's atmosphere (Khalil, 2000), but ongoing measurement has reported that the atmospheric CH_4 mixing ratio has increased greatly over the 20th century from its pre-industrial value. The inventories of atmospheric CH_4 cover a wide variety of sources, both natural and anthropogenic. Wetlands are considered as largest natural source for atmospheric CH_4 contributing 177–284 Tg CH_4 annually to global atmospheric CH_4 budget (IPCC, 2013).

Mangrove inhabited tropical and subtropical coastal wetlands act as biogenic sources of CH_4, as a result of anaerobic microbial mineralization of organic material in the intertidal sediment (Alongi and Christoffersen, 1992; Mukhopadhyay et al., 2002; Purvaja et al., 2004). Methanogenesis in the mangrove sediment typically occurs below the sulphate reduction zone, depending upon the redox conditions, organic matter content and sediment texture (Dutta et al., 2013). CH_4 produced in the mangrove sediment enters into a complex biogeochemical cycle where it emits into the atmosphere through sediment and aquatic surfaces. The emitted CH_4 enriches the atmospheric mixing ratio at a regional level (Mukhopadhyay et al., 2002) and undergoes complex atmospheric photo-chemical transformation processes.

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http://dx.doi.org/10.1016/j.agrformet.2015.06.010
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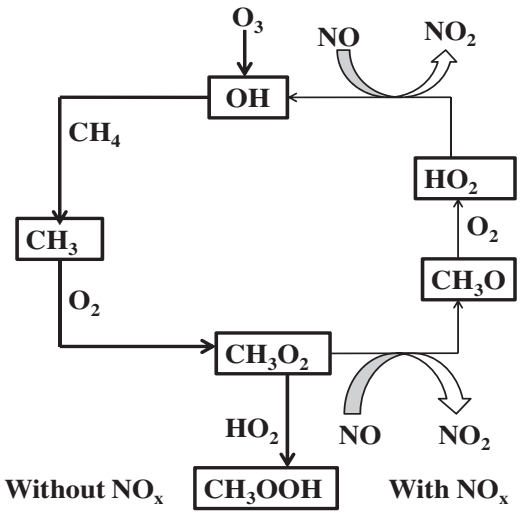

Fig. 1. Schematic of the CH_4 oxidation cycle. Bold arrows in the first half of the cycle indicate what happens without NO_x, while the thin arrows on the second half indicate processes that require NO_x.

In the atmosphere approximately 90% of CH_4 is removed by the process of photo-oxidation (Fletcher et al., 2004). Upon photo-oxidation, CH_4 is the dominant precursor for formaldehyde (HCHO) (Wagner et al., 2002). In the troposphere, the photo-oxidation process is initiated by a reaction of CH_4 with OH radicals and results in the formation of methylperoxy (CH_3O_2) radicals. Further transformation of CH_3O_2 is dependent on the ambient concentration of NO_x. In regions of high NO_x concentration, CH_3O_2 reacts with nitric oxide (NO) yielding a methoxy radical (CH_3O) which is rapidly converted to HCHO by a reaction with O_2. In this process NO is converted to nitrogen dioxide (NO_2), which in turn photo-dissociates, generating ozone (O_3) (Wuebbles and Tamaresis, 1993; Wayne, 1991).

At low NO_x concentrations, the CH_3O_2 radical may reacts with hydrogen peroxy radicals forming methylhydroperoxide (CH_3O_2H), or undergo self-reaction forming HCHO and methanol (CH_3OH) (Wubbles and Tamaresis, 1993). CH_3O_2H and CH_3OH have long lifetimes but may further break down to produce HCHO in the atmosphere (Wagner et al., 2002). The produced HCHO is either washed out by wet deposition or is oxidized into carbon monoxide (CO). So, atmospheric CH_4 chemistry (presented in Fig. 1 as adopted and modified from Wayne, 1991) is highly intricate, involving other gases like HCHO, O_3 and NO_x. Atmospheric CH_4 that escapes from photochemical oxidation gets vertically mixed in the atmosphere by micrometeorological processes and atmospheric turbulence, enriching the atmospheric mixing ratio at a regional level (Mukhopadhyay et al., 2002).

The Sundarbans mangrove ecosystem has been reported as a source for atmospheric CH_4 (Dutta et al., 2013; Mukhopadhyay et al., 2002). The impact of CH_4 emission at a regional level including biosphere–atmosphere exchange and subsequent enrichment of the atmospheric CH_4 mixing ratio have been studied by Mukhopadhyay et al. (2002). Moreover, Ganguly et al. (2009) reported that a significant amount of NO_x is emitted from the mangrove environment of the Sundarbans. The above background necessitates further insight to understand the fate of CH_4 emitted in the lower atmosphere of the Sundarbans mangroves. The experimental study described here aims to assess seasonal variability of CH_4 fluxes across different interfaces in the mangrove environment, variability in the CH_4 mixing ratio in the regional atmospheric boundary layer and the effect of micrometeorological factors to the former. In addition, a further objective is to understand the atmospheric fate of the emitted CH_4 at a regional level with a focus on its photo-oxidation involving other gases such as HCHO and O_3. Finally, a mathematical model has been proposed to predict contributory sources and sinks dependent atmospheric CH_4 mixing ratio, which helps to explain the seasonal variability of CH_4 in the regional mangrove atmosphere.

2. Study location

Sundarbans is the largest mangrove ecosystem in the world, situated in both India and Bangladesh at the land ocean boundary of Ganges–Brahmaputra delta and the Bay of Bengal. This extensive natural mangrove forest (10,200 km^2) was inscribed as a UNESCO world heritage site since 1997. The Indian Sundarbans Biosphere Reserve (SBR) extends over an area of 9600 km^2 is constituted of 44% mangrove reserve forest, 19% estuarine waterways and the rest as reclaimed areas. The Sundarbans mangrove forest accounts for 85% of the overall mangrove biodiversity as found in India (Kathiresan and Rajendran, 2005) and recent research suggests that the area may hold up to 69 species under 49 genera & 35 families (Mandal and Naskar, 2008). Among the mangroves, *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 Indian Sundarbans (Ganguly et al., 2009). The Indian region of the Sundarbans mangrove delta is crisscrossed by the estuarine phases of several rivers, namely the Mooriganga, Saptamukhi, Thakuran, Matla, Bidya, Gosaba and Haribhanga, which form a cluster of 102 low lying mostly uninhabited islands. One of these Islands is the Lothian Island (38 km^2), situated at the confluence of the Saptamukhi river and the Bay of Bengal. Lothian Island is one of three wildlife sanctuaries located in the buffer zone of the Sundarbans Biosphere Reserve, and was selected as an ideal representative for the Sundarbans region. This island is completely intertidal and occupied by thick, robust and resilient mangrove trees with a mean height of <10 m. On Lothian Island the annual mean litter fall rate was 97.8 gm dry wt. m^{-2} $month^{-1}$ with an above and below ground biomass of 3.85 and 1.34 dry wt. kg C m^{-2}, respectively (Ray et al., 2011). The Sundarbans has a typical tropical climate having three seasons of four months duration: premonsoon (February–May), monsoon (June–September) and post-monsoon (October–January). The tidal creeks and tidal water ways associated with the island are comprised of the estuarine part of the Saptamukhi River, which has no perennial source of freshwater and receives significant amounts of agricultural and anthropogenic runoff, especially during monsoon season. The estuary is polyhaline in nature being salinity maximal premonsoon and minimal monsoon (Dutta et al., 2015). The vertical tide range in the channels varies from 5.2 m during spring to 1.8 m at the neap period with mean current velocities ranging between 117 and 108 cm s^{-1} during low tide and high tide, respectively (Mukhopadhyay et al., 2006). A location map of Sundarbans showing Lothian Island is presented in Fig. 2.

3. Materials and methods

This study was carried out between June 2010 and December 2011 on a monthly basis covering diurnal variation in 3 h intervals for atmospheric samples and meteorological parameters from a watch tower located in the middle of the island (21°42′N and 88°18′E). CH_4 fluxes across sediment–atmosphere interface and water–atmosphere interface were measured from inter-tidal locations of the island during low tide conditions and mid-estuary locations of the Saptamukhi estuary, respectively.

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