Fluxes of methane and nitrous oxide from an Indian mangrove

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Fluxes of greenhouse gases, CH_4 and N_2O, were measured from a mangrove ecosystem of the Cauvery delta (Muthupet) in South India. CH_4 emissions were in the range between 18.99 and 37.53 mg/sq. m/d, with an average of 25.21 mg/sq. m/d, whereas N_2O emission ranged between 0.41 and 0.80 mg/sq. m/d (average of 0.62 mg/sq. m/d). The emission of CH_4 and N_2O correlated positively with the number of pneumatophores. In addition to the flux measurements, different parts of the roots of *Avicennia marina* were quantified for CH_4 concentration. Invariably in all the seasons, measured CH_4 concentrations were high in the cable roots, with gradual decrease through the pneumatophores below water level and the above water level. This clearly indicates the transport of CH_4 through the roots. We were able to establish that CH_4 was released passively through the mangrove pneumatophores and is also a source to the atmosphere. We present some additional information on transport mechanisms of CH_4 through the pneumatophores and bubble release from the mangrove ecosystems.

Keywords: Mangroves, methane, nitrous oxide, pneumatophores.

METHANE (CH_4) and nitrous oxide (N_2O) are atmospheric trace gases and contribute about 15 (CH_4) and 6% (N_2O) to the greenhouse effect[1]. Both N_2O and CH_4 have a long atmospheric residence time of about 114 and 12 years respectively[2] and since they are key compounds in the chemical reaction cycles of the troposphere and the stratosphere[3,4], their potential to directly or indirectly influence global climate is high[5,6]. Although atmospheric loading of N_2O is lower than CH_4 globally[7], the former is 310 times a more potent greenhouse gas (GHG) than CO_2 on a 100 yr timescale, while CH_4 is only 21 times more potent[2]. During 1990–99, CH_4 and N_2O concentrations increased by 0.007 and 0.008 ppb/yr respectively[2]. In recent years, although the slope of increase for CH_4 has slowed down, its concentration has doubled during the last 100 years[8]. The current tropospheric concentration[2] of N_2O is 317 ppb and that of CH_4 is 1.75 ppm.

Among the biogenic sources of CH_4 and N_2O, ecosystems in which anoxic conditions prevail are of particular significance, because under such conditions CH_4 and N_2O are produced as intermediate and end-products in microbial processes (methanogenesis, denitrification)[9,10]. Such emission has been observed in a number of natural wetlands that are considered to contribute between 22 and 40% to global source strength of CH_4 and N_2O respectively[7].

Mangroves occupy the intertidal zone of tropical and subtropical regions of the world, contributing to the biogenic sources of GHGs. They act as a buffer zone between land and sea as they prevent erosion, reduce currents, attenuate waves and encourage sediment deposition and accretion[11]. These ecosystems are characterized by a high rate of nutrient turnover and are therefore considered highly productive sources of organic matter[12]. Due to tidal flooding, anoxic conditions prevail at least periodically in the soil of mangrove ecosystems. Under these conditions denitrification and methanogenesis may significantly contribute to nutrient turnover[13,14]. From these features it may be concluded that anoxic wetlands are potential sources of CH_4 and N_2O since at a high rate of nutrient turnover, small leaks in nutrient cycles may result in high rates of emission of these GHGs[9]. However, despite the proposed possible significance of mangrove wetlands as sources of GHGs, detailed information on the contribution of mangrove forests to the overall source strength of natural wetlands for GHGs is scarce[15]. Since vegetation adapted to frequently flooded sites by the formation of aerenchyma may mediate the transport of trace gases between soil and atmosphere[16], it may be assumed that mangrove vegetation may also facilitate GHG exchange.

Thus there is need for quantification of the present and future potential of individual ecosystems as sources and sinks for atmospheric CH_4 and N_2O. The aim of the present study therefore was to characterize N_2O and CH_4 emissions from a mangrove ecosystem of the Cauvery Delta, South India and to elucidate the role of pneumatophores in mediating these emissions. As the study area is dominanted by *Avicennia* sp., all the flux measurements were carried out from this zone.

Materials and methods

Study site and vegetation

Muthupet mangrove is located at 10°20′N and 79°32′E of South India, covering an area of 68.03 sq. km, the majority of which is covered by mangrove creeks. Five rivers

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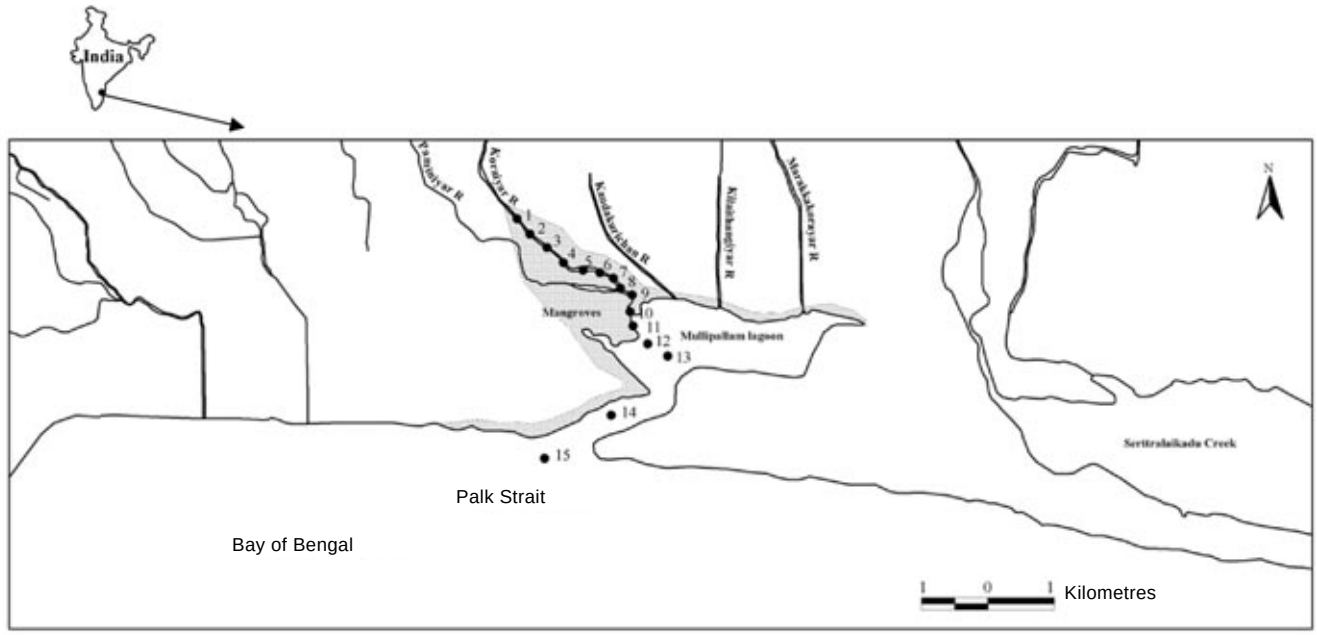

Figure 1. Map of study area – Muthupet, South India (with 15 sampling sites).

flow into these mangroves, among which the Koraiyar is quantitatively the most important (Figure 1). These rivers are highly seasonal and freshwater input into the system is directly dependent on rainfall. The rivers feed a network of mangrove creeks, which run into an extensive lagoon (Mullipallam lagoon), pass through Palk Strait and finally join the Bay of Bengal. These mangroves are subjected to a variety of anthropogenic inputs, including aquaculture (shrimp-farming effluent), and more diffuse and seasonal agricultural run-off. The system is typically characterized by a neap tidal range of ~0.4 m and a spring range of ~1 m. We sampled the mangrove system spatially at 15 locations that were selected based on inflow of rivers, run-off from agricultural sources and aquaculture outlets to the Bay of Bengal (Figure 1). The dominant species found is *Avicennia marina*, which is high salinity-tolerant. *Avicennia* has a highly differentiated root system with cable roots running parallel to the sediment surface and pneumatophores projecting from the cable roots upwards and connecting it to the atmosphere[17]. Our measurements therefore included the enclosure of these pneumatophores (numbers were enumerated during sampling) within the static chamber to determine the flux of CH_4 and N_2O through the pneumatophores, as described below.

Dissolved CH_4 in surface water

Temporal and spatial measurements of dissolved CH_4 were made in 15 locations from March 2002 to March 2003 (five seasons). Serum bottles of 100 ml capacity were filled with surface water and 0.1 ml of saturated mercuric chloride ($HgCl_2$) was added to inhibit phytoplankton activity[18]. The bottles were sealed and transported to the laboratory for analysis of dissolved CH_4. Exactly 50 ml of the surface water was subsequently transferred into 100 ml sealed bottles, which were preflushed with N_2. The bottles were hand-equilibrated for 10 min and the gas taken from the headspace was analysed under the gas chromatograph conditions, as described in detail below. For dissolved CH_4 measurements the headspace mixing ratios were converted to concentrations using a primary standard (14.2 ppmv CH_4 in N_2, National Physical Laboratory (NPL), New Delhi), which was run prior and between analyses.

Flux measurements (static chamber)

Seasonal measurements of CH_4 efflux at the sediment–water interface were made during the wet (September and December 2002) and dry (March 2002, June 2002 and March 2003) seasons at Forest Camp in the Muthupet mangrove covering the pneumatophores of *A. marina* as was done in previous studies[15]. Additionally, water-level fluctuations due to tides and air temperature were measured in parallel. Methane emission was calculated from the linear increase of mixing ratio with time, taking into account the actual volumes of the chamber[15].

Gas bubbles and aerenchyma

Gas bubbles entrapped in the sediments were collected using an inverted glass funnel (20 cm dia) fitted with a silicone rubber septum in its stem between 10 and 20 m

from the location where the static chamber was placed[15,19]. Gas samples were also collected from the aerenchyma of the cable roots (CR), and from the pneumatophores, which were submerged (below water level: BWL) and those that were exposed (above water level: AWL). Gas samples from at least five pneumatophores were drawn using a 1 ml syringe and were transferred to 5 ml Venoject-tubes. The cable root, located between 20 and 25 cm from the sediment surface, was sampled by excavating the sediment along the root (pneumatophore) zone until it was exposed. Care was taken at all times (during excavation) to retain about 3–5 cm of standing water to avoid artifacts.

Gas analysis

Methane measurements were carried out using standard procedure[15]. Pure methane (99.95%) and a secondary standard with a concentration of 1195 ± 2.6 ppm$_v$ CH_4 in nitrogen (Bhoruka Gases, Bangalore) were used for the analysis of CH_4 in gas bubbles and in the aerenchyma of cable roots and pneumatophores respectively.

Nitrous-oxide measurements were carried out in the HP-GC 5890 fitted with an Electron Capture Detector and a Poropak-Q column (mesh: 80/100). The column, injector and detector temperatures were 60, 100 and 250°C, respectively. The carrier was nitrogen at a flow rate of about 20 ml/min. For flux measurements, the gas chromatograph was calibrated using a secondary standard 5.78 ± 0.35 ppm$_v$ N_2O in nitrogen (NPL).

Gas samples were taken using pressure-lock syringes (Dynatech, Baton Rouge, USA) from the sampling tubes for gas chromatographic analysis. Linearity check of the gas chromatograph was made on a regular basis by injecting various volumes (0.1 to 1.0 ml) of the appropriate standards for CH_4 and N_2O.

Results

Fluxes of CH_4 and N_2O for five different seasons (Figure 2) indicate a distinct seasonal variability, with peak emissions during monsoon (December 2002) and the least in September for both trace gases. CH_4 flux ranged between 18.69 ± 0.92 and 37.47 ± 0.52 mg/sq. m/d, with a mean of 25.10 ± 0.80 mg/sq. m/d. For N_2O, the flux varied from 0.43 ± 0.023 to 0.81 ± 0.066 mg/sq. m/d, with an average of 0.63 ± 0.030 mg/sq. m/d. We observed a strong positive correlation ($r^2 = 0.82$) between the fluxes of CH_4 and N_2O in this study. In addition we also determined the effect of pneumatophores on the exchange of gases from the sediment to the atmosphere, by correlating emission with the number of pneumatophores (Figure 3). A strong positive correlation between the number of pneumatophores and trace gases (CH_4, $r^2 = 0.96$ and N_2O, $r^2 = 0.81$) was observed in the present study.

There was a strong CH_4 gradient from soil through roots to the atmosphere (Figure 4) during all the seasons. Concentration of CH_4 in gas bubbles ranged from 30.71% during December 2002 to 62.73% in September 2002. Invariably, the mean mixing ratio of CH_4 was highest in gas bubbles (42%) followed by cable roots, 373.3 ± 96.4 ppmv to submerged parts of pneumatophores, 50.5 ± 24.7 ppmv and pneumatophores above the water level, 8.2 ± 1.6 ppmv. The mixing ratio of CH_4 in the atmosphere was 1.8 ppmv. Spatial variability is the major limitation for accurate quantification of trace-gas fluxes over estuarine areas using chambers and therefore, the estuarine CH_4 flux from the water column was also quantified using a wind speed-based air–sea gas exchange model in order to account for the uncertainty associated with chamber flux estimates. The sea-to-air flux of CH_4 from the surface waters of the Muthupet mangrove is given by:

$$F = k\Delta C,$$

where F is the interfacial gas flux (mol unit/area/unit time), k the gas transfer velocity (cm/h) and ΔC the dif-

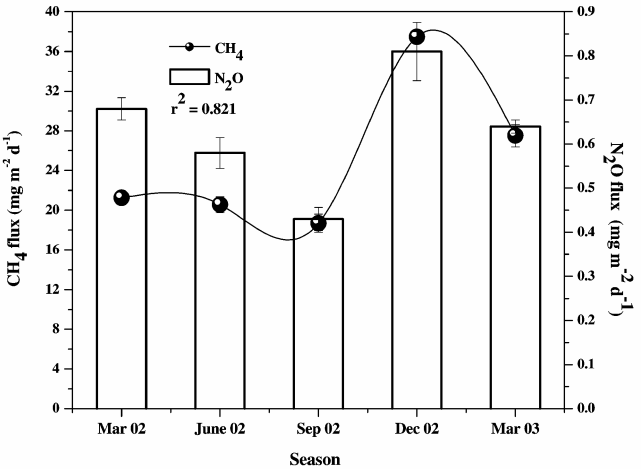

Figure 2. Correlation between fluxes of CH_4 and N_2O ($n = 3$) using static chamber technique from Muthupet mangrove.

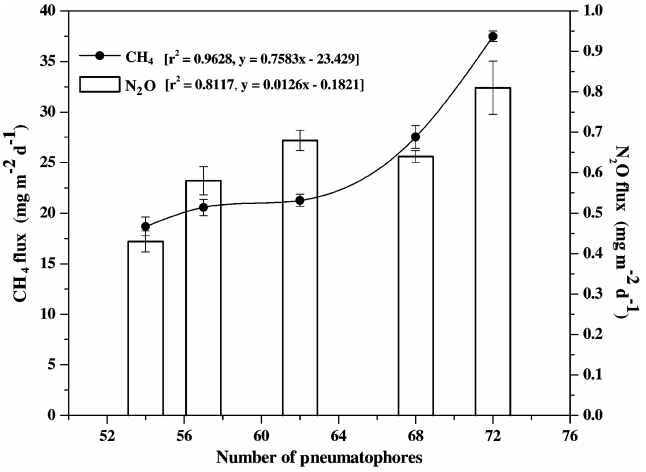

Figure 3. Correlation of CH_4 and N_2O flux rates ($n = 3$) with the number of pneumatophores.

ference in gas concentration (mol/l) between the air to water phases on either side of the interface. Various empirical relationships have been derived[20,21] for estimating *k*. Another relationship for estuaries (using data from the Hudson River estuary), takes into account non-wind speed-driven turbulence due to interaction with the seabed[22]. For this reason, all three methods were included here to aid comparison with similar studies, although the model developed by Clark is probably the most applicable to Muthupet mangrove. Muthupet mangrove functions as a significant CH_4 source and measurement of dissolved CH_4 in the surface water indicated distinct seasonal dynamics. The mean dissolved CH_4 concentration for all seasons and spatially was 225 ± 41.0 nmol/l, ranging from 182.75 ± 3.2 nmol/l in September 2002 to 267.69 ± 2.65 nmol/l in December 2002 (Figure 5). There was a decrease in estuarine CH_4 concentration with increasing salinity during all the seasons and a strong negative correlation ($r^2 = -0.8$) was observed (Figure 6). Dissolved O_2 concentration correlated weak negative correlation with dissolved CH_4 concentration ($r^2 = -0.55$), implying increased CH_4 fluxes at low O_2 concentrations, although not very obvious during our study period. We also did not observe any distinct changes in CH_4 concentration due to seasonal variations in dissolved nutrients – NO_3, NO_2 and PO_4 (with the exception of NH_4, which showed positive correlation; $r^2 = 0.76$); probably due to the fact that the mangrove ecosystems are in general a low-nutrient environment, especially with reference to their dissolved concentrations (Table 1).

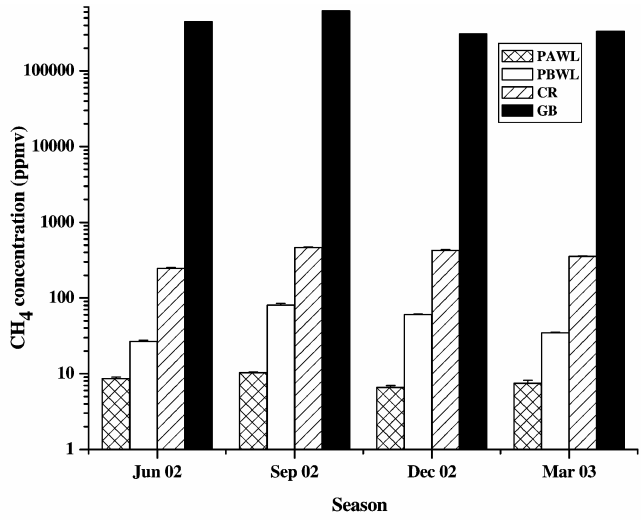

Figure 4. Transport of CH_4 ($n = 3$) through different parts of the pneumatophores (passive) and through gas bubbles (active). PAWL, Pneumatophores Above Water Level; PBWL, Pneumatophores Below Water Level; CR, Cable Roots; GB, Gas Bubbles.

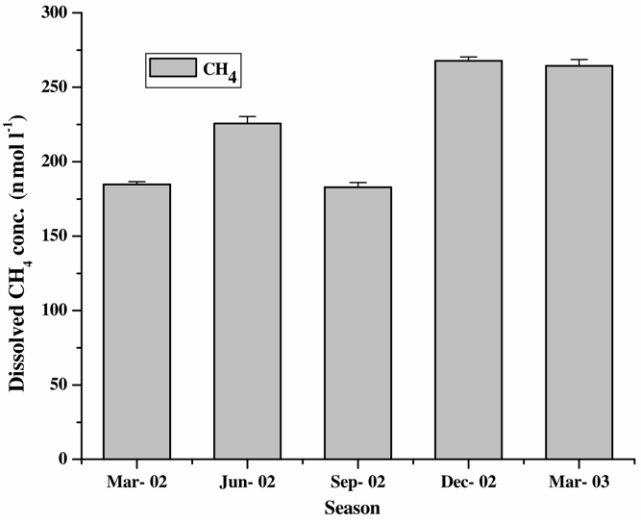

Figure 5. Mean value of dissolved CH_4 concentration in surface water of Muthupet mangrove during different seasons.

Discussion

The present work combined both CH_4 and N_2O emission measurements from the Indian mangrove ecosystems. The highest emissions were observed just after the monsoon season (December), which could have resulted from anthropogenic inputs from the seasonal rivers. The present results seem to be in accordance with earlier studies conducted by Purvaja *et al.*[15] on CH_4 emission from similar mangrove ecosystem in South India (Pichavaram). Table 2 compares the results of this study with similar CH_4 flux data for tropical coastal systems. We have observed an average flux of 9.2 g CH_4/sq. m/yr, as also reported by Purvaja *et al.*[15] for the Pichavaram mangrove (10.0 g CH_4/sq. m/yr). Kreuzwieser *et al.*[23] conducted similar measurements of trace gases from Australian mangroves and observed comparatively low fluxes, 0.18–3.07 g CH_4/sq. m/yr and 0.02–0.12 g N_2O/sq. m/yr. The higher flux of CH_4 (from the Australian mangroves) in the current study is due to anthropogenic additions from land-based sources. CH_4 emission in the Vembanad Lake (highly impacted by coconut husk decomposition)[24] was about 1690 g/sq. m/yr, more than two orders of magnitude higher than what was observed in the present study due to high anoxic stress. N_2O emission rates from Muthupet mangrove varied between 0.41 and 0.77 μmol/sq. m/h. Similar values for N_2O emissions were recently reported from Australian mangroves and other tropical ecosystems (Table 3). Corredor *et al.*[25] reported highest N_2O emission rates from mangroves of Puerto Rico and attributed this to the availability of nitrogenous substrates.

The increase in CH_4 and N_2O concentration with the number of pneumatophores substantiated the fact that vegetation also plays a major role in the transport of gases to the atmosphere. Schutz *et al.*[26] and Singh and Singh[27] studied the role of wetland plants as pathways for

Table 1. Environmental factors and nutrients from surface-water samples (n = 15) of the Muthupet mangrove

Season	pH	EC (μS/cm)	Total dissolved solids (mg/l)	Salinity	Dissolved oxygen (ml/l)	NH_4 (μmol/l)	NO_3 (μmol/l)	NO_2 (μmol/l)	PO_4 (μmol/l)	SO_4 (mmol/l)
Jun-02	7.4 ± 0.1	45.8 ± 12	22.9 ± 6	25.2 ± 6.1	9.4 ± 2.8	34.21 ± 14.3	3.31 ± 0.8	3.02 ± 0.8	0.87 ± 0.6	3.22 ± 2.5
Sep-02	7.4 ± 0.1	28.6 ± 27	14.1 ± 7	22.9 ± 6.6	8.59 ± 2.3	64.58 ± 33.3	7.28 ± 4.2	1.74 ± 1.1	0.81 ± 0.2	1.39 ± 1.1
Dec-02	7.2 ± 0.1	7.0 ± 2.6	3.5 ± 1.3	19.6 ± 6.9	12.8 ± 3.6	78.45 ± 11.7	5.06 ± 2.4	0.76 ± 0.4	0.45 ± 0.2	5.67 ± 2.8
Mar-03	7.0 ± 0.02	7.1 ± 0.8	3.6 ± 0.4	22.5 ± 6.8	10.6 ± 1.7	32.22 ± 18	4.23 ± 1.8	2.59 ± 1.0	2.90 ± 0.3	10.00 ± 3.6
Average	7.3 ± 0.2	22.2 ± 18.8	11.0 ± 9.4	22.6 ± 2.3	10.3 ± 1.8	52.37 ± 22.84	4.97 ± 1.7	2.02 ± 1.00	1.26 ± 1.11	5.07 ± 3.72

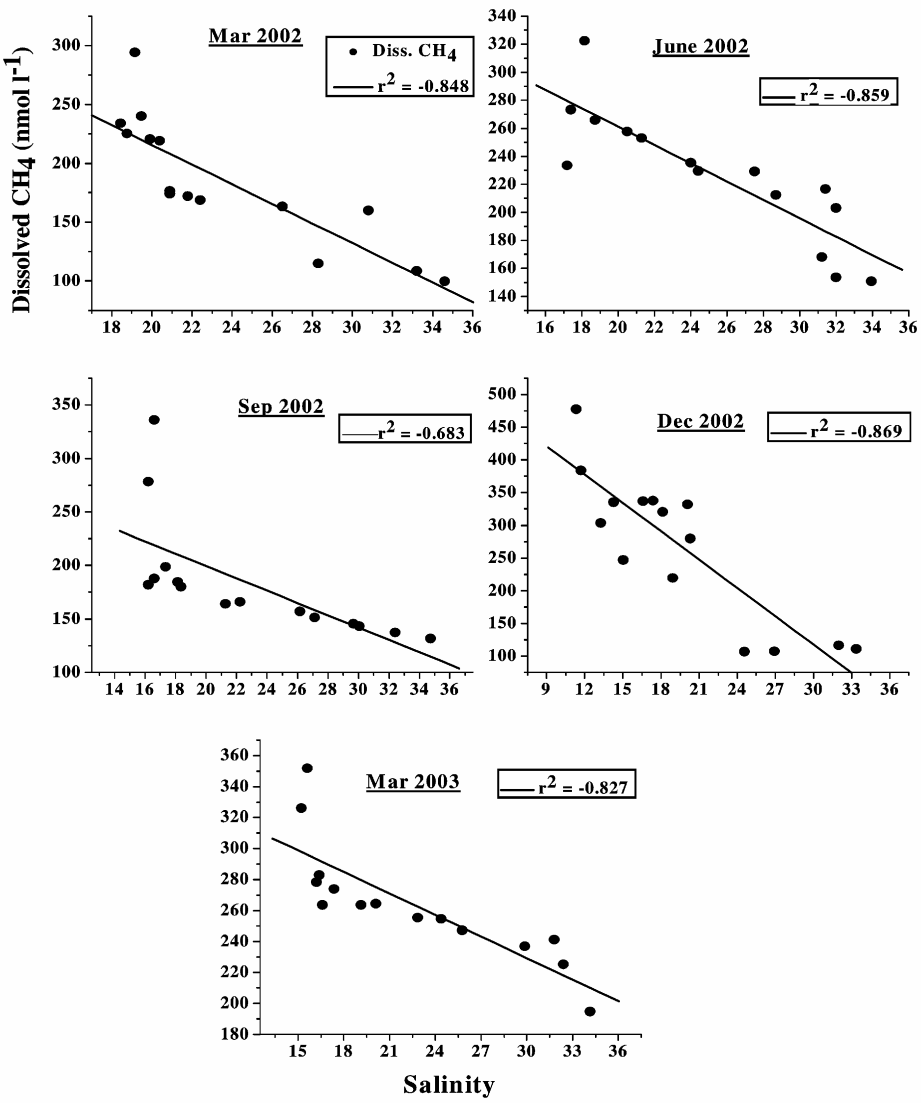

Figure 6. Correlation of dissolved CH_4 concentration with salinity.

gas transport from the root zone into the atmosphere, whereas Kreuzwieser *et al.*[23] demonstrated clearly the role of Australian mangrove vegetation as a transport path for CH_4 and N_2O between the sediment and atmosphere. In experiments under controlled conditions, Rusch and Rennenberg[16] showed that wetland tree species facilitate CH_4 and N_2O emission via their aerenchyma. It has been further strengthened by the recent findings of Purvaja *et al.*[15] from the Pichavaram mangrove.

Methane concentration in gas bubbles was high (Figure 4), reaching values comparable with those reported from freshwater wetlands and rice fields[28–31]. The concentra-

Table 2. Methane emission from mangroves and wetlands (updated from Purvaja *et al*.[15])

Ecosystem/site	Latitude	Flux (mg CH_4/ sq. m/d)	Flux (g CH_4/ sq. m/yr)	Notes and references
Mangrove (*Avicennia*), Muthupet, South India	10°N	18.9–37.5	9.2	Chamber method; flux through pneumatophores; the present study
Mangrove (*Avicennia*), Pichavaram, South India	11°N	7.4–63.7	10	Chamber method; flux through pneumatophores; 15
Pulicat Lake, South India (in press)	13°N	18.48–25.36	8	Chamber method; sediment–water fluxes; 36
Mangroves (*Rhizophora*), Queensland, Australia	27°S	0.02–0.35	–	Chamber method; flux through pneumatophores; 23
Coastal South Kalimantan, Indonesia	–	3.28–5.21	1.2–1.9	Chamber method; 37
Vembanad Lake, South India	9°N	223.2–4632	1690	Chamber method; sediment water fluxes; 24
Ranong Mangroves, Thailand	9.5°N	0.19–0.52	–	Chamber method; sediment water fluxes; 38
Mangroves (*Bruguiera*), Hainan	18°N		0.12–0.39	Chamber method; water surface and soil; 39
Mangrove, Sunderban	22°N	–16–32	43	Gradient, micrometeorological measurement; 40
Mangrove (*Avicennia*), Puerto Rico	18°N	4–42	n.a.	Chamber method; gradient from pristine to polluted; 41
Hudson Bay Lowlands, coastal marshes	50–58°N	84.3	8	Chamber method; coastal marshes; 42
Hudson Bay Lowlands, open fens	50–58°N	78.6	6.7	Chamber method; coastal marshes; 42

n.a.: not available.

Table 3. Comparison of nitrous oxide emissions from mangrove ecosystems world-wide using closed chamber

Ecosystem/site	Latitude	N_2O flux (μmol/sq. m/h)	Reference
Mangrove (*Avicennia*), Muthupet, South India	10°N	0.41– 0.77	This study
Mangroves, Queensland, Australia	27°S	–0.045–0.32	23
Red mangrove fringe, South West Coast of Puerto Rico	17°N	0.05	43
Red mangroves, Puerto Rico	17°N	0.54	44
Mangroves, Puerto Rico	17°N	0.12–7.8	25

tion of CH_4 from gas bubbles in the Pichavaram mangrove[15] was 30% compared with the present result. Pneumatophores primarily function as a conduit allowing atmospheric gases to be transported from the atmosphere to the roots, aiding diffusion of gases from the subsurface to the atmosphere. Gases, including O_2 are transported to the roots via the pneumatophores, both from the water column and the atmosphere. This promotes methane oxidation (data not provided here) and considerably reduces the overall flux of CH_4 to the atmosphere to just over ambient levels. The diffusion of gases, primarily CH_4, take place under low-O_2 conditions and has been observed to be tide-dependent. At high tide, flooding does not permit emission/diffusion of gases to the atmosphere, while at low tide, CH_4 emission via bubble ebullition (as discussed earlier) is obvious.

Thus, pneumatophores, facilitated by the aerenchyma[15], serve as one of the principal pathways through which CH_4 and other gases may be transported from the sediment to the atmosphere in this O_2-poor environment. It is hypothesized that when pneumatophores become flooded, gas transfer between the aerenchyma and the atmosphere becomes impeded, and vice versa[15]. Conceptually, surface-water CH_4 concentration can be described as a function of sources and losses[32]. Therefore, the Muthupet mangrove represents a consistent source of CH_4 to the atmosphere during all the seasons.

Conclusion

In summary, the present results support the view that mangroves act as sources of atmospheric CH_4 and N_2O. In addition, vegetation plays an important role as transport path for CH_4 and N_2O emissions, as previously observed for herbaceous species in other wetland ecosystems[33–35]. Therefore, in order to extrapolate CH_4 emissions from mangrove to global levels, composition, distribution of vegetation and quantity of different root types should also be taken into account.

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ACKNOWLEDGEMENT. The first author thanks the Council of Scientific and Industrial Research (CSIR), Government of India for providing the Senior Research Fellowship to carry out this work. PR is grateful to the Department of Science and Technology, Government of India for the financial support (SR/FTP/ES-59/2001) and to the Hanse Wissenschafts Kolleg, Delmenhorst, Germany for support.

Received 6 March 2007; revised accepted 7 December 2007