

Influence of river discharge on the distribution and flux of Methane in the coastal Bay of Bengal

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Abstract

Rivers are known to be strong sources of methane (CH_4) to the atmosphere and their discharges may also enhance its flux from the coastal ocean. In order to examine this, CH_4 fluxes to the atmosphere were examined along the coastal Bay of Bengal during peak discharge period. The saturation of CH_4 in the Ganges river was relatively lower ($208 \pm 7\%$) than that of Indian peninsular rivers ($907 \pm 136\%$). The Ganges discharges into the northwestern (NW) while the peninsular rivers discharge into the central and southwestern (SW) regions of the coastal Bay of Bengal and influence the concentration and saturations of CH_4 in the western coastal Bay of Bengal. Methane was higher (4.4 ± 0.9 nM) and more supersaturated ($208 \pm 37\%$) in the SW coastal Bay of Bengal than in the NW region (2.5 ± 0.1 nM and $116 \pm 2.6\%$ respectively). The higher concentrations of CH_4 in the SW region were also influenced by mild coastal upwelling. The efflux of CH_4 is estimated as 57.4 ± 19.3 and $14.5 \pm 2.3 \mu\text{mol m}^{-2} \text{d}^{-1}$ from SW and NW regions, respectively, and an average of $38.9 \pm 11.3 \mu\text{mol m}^{-2} \text{d}^{-1}$. Our study suggests that the western coastal waters of the Bay of Bengal act as strong source of CH_4 to atmosphere. The total flux from Indian coastal waters (estuaries, mangroves and coastal waters) is estimated as $21.9 \times 10^9 \text{ g CH}_4 \text{y}^{-1}$ out of which $\sim 70\%$ is from the coastal Bay of Bengal and 20% from estuaries.

1. Introduction

Methane (CH_4) is an important green house gas with an atmospheric mixing ratio of ~ 1.8 ppmv and is increasing at a rate of 0.7% per year (Forester et al., 2007; Ciais et al., 2013). The global warming potential of CH_4 is about 25 to 40 times higher than that of carbon dioxide (Forester et al., 2007; Shindell et al., 2009; Khalil et al., 2007). Hence, atmospheric CH_4 plays an important role in the Earth's radiation balance and atmospheric chemistry (Cicerone and Oremland, 1988; Lashof and Ahuja, 1990). Higher concentrations of CH_4 in the atmosphere reduce the oxidizing capacity of the troposphere by depleting OH radicals (Hein et al., 1997). Methane is produced both naturally (wetlands) and anthropogenically (livestock and other agricultural practices, decay of organic waste in municipal solid waste landfills, and leakages from natural gas systems) (Forester et al., 2007; Ciais et al., 2013). Methane undergoes, as part of natural processes, anaerobic or aerobic oxidation in sediments (Boetius and Wenzhöfer, 2013), aerobic oxidation mediated by aerobic methane-oxidizing bacteria in the water column (Reeburgh, 2007; Knoe et al., 2010; Borges and Abril, 2011), and photochemical reactions in the atmosphere by which it is removed (Wuebbles and Hayhoe, 2000, Allan et al., 2007). Kirschke et al. (2013) studied decadal variability in atmospheric CH_4 using different models and observed stabilization of CH_4 levels during the 1999 -2006 period which they attributed to decreasing-to-stable fossil fuel emissions and stable-to-increasing microbial emissions. In the years that followed, they observed a rise in global CH_4 levels attributed to enhanced (natural) wetland and fossil fuel emissions.

CH_4 in marine environment originates from either external sources such as methane-enriched fresh water, submarine ground water discharge, or internally through bacterial degradation of organic matter and associated microbial processes in particles and in guts of zooplankton (Scranton and McShane, 1991; Houvland et al., 1993; Martens and Klump, 1980; De Angelis and Lee, 1994; Karl and Tilbook, 1994; Holmes et al., 2000; Boetius et al., 2000). Methane is also produced through aerobic process under nutrient limiting conditions (Karl et al., 2008; Damm et al., 2010; Grossart et al., 2011). The efflux of methane from coastal areas and estuaries is an order of magnitude higher than that from the open ocean (Bange et al., 1996). Though coastal regions cover only about 15% of the total oceanic surface area, they account for more than 75% of global marine CH_4 emissions (Bakker et al., 2014). The estimate of CH_4 emission from marine waters (i.e. open ocean, continental shelves, estuaries and continental margin seeps etc) varies from 8.3 to 45.9 Tg $\text{CH}_4 \text{ yr}^{-1}$, out of which the share of continental shelves alone ranges from 0.38 to 7.3 Tg $\text{CH}_4 \text{ yr}^{-1}$ (Bakker et al., 2014).

It has been reported that CH₄ is mainly produced in estuarine systems and subsequently transported to coastal regions (Bange, 2006; Jayakumar et al., 2001; Zhang et al., 2010; Upstill-Goddard et al., 2000). Therefore river discharge enhances CH₄ fluxes from the coastal ocean. Significantly higher concentrations of CH₄ were observed in Jiaozhou Bay (Zhang et al., 2007), Dalian Bay (Wang et al., 2011) and Sanggou Bay (Hou et al., 2016) and they were attributed to discharge of CH₄-rich water through the adjacent rivers.

The northeastern Indian Ocean (Bay of Bengal) receives a huge amount of freshwater discharge from the major rivers such as Ganges ($0.44 \times 10^{12} \text{ m}^3 \text{ y}^{-1}$), Godavari ($0.11 \times 10^{12} \text{ m}^3 \text{ y}^{-1}$), Krishna ($0.07 \times 10^{12} \text{ m}^3 \text{ y}^{-1}$), and Mahanadi ($0.06 \times 10^{12} \text{ m}^3 \text{ y}^{-1}$) (Kumar et al., 2005). The river discharge peaks during southwest monsoon (June to September) when the Indian subcontinent receives significant rainfall (Mukhopadhyay et al., 2003; Sarma et al., 2011; Krishna et al., 2015). The discharge of freshwater stratifies coastal waters and inhibits vertical mixing (Sarma et al., 2013). Peninsular river discharge (Godavari, Krishna, Mahanadi) is found to be a significant source of CO₂ and N₂O to coastal waters and the atmosphere while discharge from the glacial river Ganges, a sink (Sarma et al., 2012a; Rao et al., 2013). The impact of discharge on coastal water concentrations of CH₄ is unknown as no measurements were carried out during the discharge period, when nearly 85% of the river water flux takes place. Berner et al. (2003) measured concentrations of CH₄ in the Northern Bay of Bengal during winter monsoon and reported that the mouth regions of Ganges and Brahmaputra rivers are strong sources of CH₄ to atmosphere. Several recent studies in Indian estuaries revealed that 3 to 20 times higher organic carbon transport to the adjacent coastal waters takes place during the discharge period when compared to the dry period (Sarma et al., 2014; Krishna et al., 2015; Gawade, 2015). Rao and Sarma (2016) found super saturation of CH₄ in the Indian estuaries (210 to 22,340%) during the discharge period. CH₄ showed a linear relation with organic load in the estuary, and was quickly diluted in the coastal water as evident from an inverse relation with salinity. The aim of this study was to understand the influence of discharge from Indian estuaries on concentrations and fluxes of CH₄ in the coastal Bay of Bengal during southwest monsoon, when river discharges are at their peak.

2. Material and methods

2.1. Study area

This study was carried out on board ORV SagarNidhi (#SN 42) during 23 July to 10 August 2010, representing the peak river discharge period of the southwest monsoon, along the east coast of India. Coastal waters studied were in southern (S) and northern (N) transects off each major estuary,

Krishna (KS, KN transects), Godavari (GS, GN) and Mahanadi (MS, MN)) and off the minor estuaries, Vamsadara (VD) and Hyadri (HD) (Fig. 1). A total of 73 stations were occupied. Transect V (off Visakhapatnam city) was chosen to represent locations without discharge. Samples were collected from the coast to locations that were 70 to 100 kms offshore. Simultaneous sampling was also done in the major estuaries namely Krishna, Godavari, Mahanadi and Ganges in order to examine the characteristics of the discharged water vis a vis the coastal Bay of Bengal. Three to 5 surface water samples were taken over a 10 km distance from the mouth to upstream in each estuary. In order to assess the spatial variation along the coastal Bay of Bengal, the study region was tentatively divided into northwestern (NW) (VD, HD, MN, and MS transects) and southwestern (SW) (KS, KN, GS, GN and V transects) regions.

2.2. Sampling and analyses

2.2.1. Hydrographic parameters

Surface water samples were collected using a Seabird Conductivity-Temperature-Depth (CTD)-rosette system fitted with 5 L Niskin bottles. Analysis for nutrients (nitrate, nitrite, ammonium, phosphate and silicate) and dissolved oxygen (DO) were completed shortly after collection of the samples. DO was estimated by potentiometric method following Carritt and Carpenter (1966) using Tritrinado 835 Metrohm autotitrator. Nutrients were analyzed following standard procedures (Grashoff et al., 1992) using an auto analyzer. The analytical precision, expressed as standard deviation, was $\pm 0.07\%$ RSD for DO whereas for nitrate+nitrite, ammonium, phosphate and silicate corresponding values were ± 0.02 , 0.02 , 0.01 and $0.02 \mu\text{M}$ respectively. For Chlorophyll-*a* (Chl-*a*), 2 to 5 L water of each sample was filtered through a GF/F filter (Whatman) and phytoplankton biomass retained on the filter was extracted in N, N Dimethyl formamide (DMF) at 4°C in the dark for 12 h, followed by spectrofluorometric (Varian Eclipse Fluorescence spectrophotometer, USA) analysis according to Suzuki and Ishimaru (1990). The analytical precision for Chl-*a* analysis was $\pm 4\%$. The suspended particulate matter (SPM) measurement was based on (dry) weight gain of the filter ($0.22 \mu\text{m}$ pore size, polycarbonate) when 1 L sample water was filtered. Winds were measured continuously along the cruise track using an automated weather station (AWS) installed on the ship at 10 m above sea level. River discharge data were obtained from dam authorities of respective rivers.

2.2.2. Measurements of CH_4

For CH₄, surface water samples were collected in 60 ml amber colored bottle immediately after the DO sampling. Samples were poisoned with 0.5 ml of saturated HgCl₂ to prevent microbial activity, and their analysis was completed on board within 2 hours after collection. A multiphase head space equilibration technique (McAuliffe, 1971) coupled with Gas Chromatographic (GC) analysis was used for the purpose. Briefly, a 25 ml aliquot was equilibrated with an equal volume of ultra pure helium in an airtight syringe by vigorously shaking at room temperature for 5 minutes using a wrist action shaker. After equilibrium, helium in the head space was dried over drierite and injected into a gas chromatograph (Agilent – 6820, USA) through a 5-ml sampling loop and separated over a Molecular Sieve 5A(80/100 mesh) at 35⁰C. The peak detection was by a Flame Ionization Detector (FID). The detector was calibrated at regular intervals using a standard gas mixture of CH₄(9.8 ppm; Spectra Gases, USA) diluted to different concentrations so that the dilutions closely represent the ambient concentration of CH₄. Using the NIST standard reference material as calibration standard, the concentration was computed from the calibration curve. The linearity of the detector with time was checked three times daily. Ambient concentrations of CH₄ in air samples were measured by collecting air samples in 60 mL gas tight BD syringes with stop cocks and injecting the samples directly into the GC in the same manner as for the sample. The uncertainty (1σ) in CH₄ measurements was less than ±2%, which was estimated from the analysis of duplicate water samples stored in a refrigerator. The precision of CH₄ measurement was always better than 1% (at 1σ level).

2.2.3. Estimation of CH₄ saturation and fluxes

The surface saturation of CH₄ was calculated as

$$\text{CH}_4\text{saturation} = \{[\text{CH}_4]_{\text{surface}} / [\text{CH}_4]_{\text{equ}}\} \times 100$$

Where [CH₄]_{surface} is the concentration of CH₄ in surface water and [CH₄]_{equ} is the solubility of CH₄ *in situ* sea surface temperature (SST) and salinity. The [CH₄]_{equ} was computed using solubility coefficients of Wiesenburg and Guinasso (1979). During the cruise, atmospheric CH₄ was measured and its mean atmospheric mole fraction was found to be 2.03 ± 0.06 ppm (n= 162). This value was used for calculating [CH₄]_{equ} and fluxes.

The sea to air fluxes of CH₄ were estimated using daily mean wind speed data, difference in its concentration between water and air, transfer velocity coefficient given by Ho et al., 2006 and its solubility coefficient calculated as per Wiesenburg and Guinasso (1979). The hourly wind speeds data obtained from automated weather station (AWS) installed on board (Astra Microwave Products Limited, Hyderabad, India). Hourly measured wind speed data was averaged to obtain daily mean and

was used for transfer velocity calculations. The Schmidt number (Sc) for CH_4 was calculated from temperature (t), according to the polynomial fit given by Wanninkhof (2014), and u_{10} , which is the wind speed normalized to a height of 10 m above the sea surface (Thomas et al., 2005).

$$F = k \cdot S_{gas} [\Delta_{gas}]$$

where F is flux, k is gas transfer velocity ($m\ d^{-1}$), S_{gas} is the solubility coefficient of CH_4 ($mol\ m^{-3}\ atm^{-1}$) (Wiesenburg and Guinasso, 1979) and Δ_{gas} is the difference in CH_4 concentration between water and air.

One-way analysis of variance (ANOVA) was used to calculate significance of the spatial variation in different physicochemical parameters using STATISTICA software. Three dimensional figures were drawn using Ocean Data View and two dimensional plots using Grapher software.

3. Results

Sea surface temperature and salinity varied from 26.79 to 29.98⁰C and from 20.58 to 34.10 respectively in the study region (Table 1). Hydrographic properties displayed significant spatial variability along the coastal Bay of Bengal. The surface waters of the SW region were cooler, more saline, and contained less dissolved oxygen (DO) and suspended matter (TSM) than those in the NW region. In contrast, higher concentrations of nutrients e.g., nitrate, ammonium and phosphate were observed in the NW than SW region (Fig. 2, Table 1). The concentrations of nitrate and ammonium in the surface waters ranged from 0 to 13.2 and 0 to 3.8 μM , respectively with higher concentrations occurring in the SW (mainly off Godavari river estuary, GN) region that decreased towards the NW (Fig. 2e,f). Similarly phosphate showed lower concentrations in the NW (0.2-0.5 μM ; mean of $0.4 \pm 0.1\ \mu M$) than in the SW region (0.2-0.7 μM ; mean of $0.6 \pm 0.3\ \mu M$; $t=14.8$ and $p < 0.001$ Fig. 2g; Table 1). However, Chl-a was more enriched in the NW (4.8 to 7.0 $mg\ m^{-3}$) than in the SW waters ($0.3-5\ mg\ m^{-3}$; $t=9.4$ and $p < 0.001$; Fig. 2h).

The concentrations of CH_4 in the surface waters ranged between 1.9 and 23.6 nM in the entire study area, and the concentrations were slightly higher in the SW (2.4-11.5 nM, mean $4.5 \pm 0.9\ nM$) than in the NW region (2.4-2.6 nM, $2.5 \pm 0.1\ nM$) and the classification was highly significant ($t=7.6$, $p < 0.001$; Table 1; Fig. 3a). The CH_4 saturation varied from 93 to 1088% and the mean saturation was higher in the SW (111.4-528.5%, mean: 208%) than in NW (109.2-119.6%, 116%) ($t=7.7$ and $p < 0.001$) (Fig. 3b). The estuaries of the Ganges, Mahanadi, Godavari and Krishna rivers were also characterized by higher concentrations of CH_4 and a higher CH_4 saturation in surface waters (4.7 to 36.1 nM and (196-1407% respectively; Table 2) than the coastal Bay of Bengal (2.4-11.5 nM and

109.2-528.5% respectively; Table 1). High wind speeds (13.1-25.5 m s⁻¹; Table 1) caused by local atmospheric depression prevailed during the study period and the speeds were higher in the NW (17.2-25.5 m/s; mean of 21.4±2.3 m/s; Table 1) than the SW region (13.1-20.2 m/s; mean of 16.9±1.2 m/s).

The concentrations of CH₄ and their saturation correlated inversely with salinity, temperature, dissolved oxygen, and positively with dissolved inorganic nitrogen, suspended matter, and pCO₂ in the coastal Bay of Bengal (Table 3). No significant relation was observed with phytoplankton biomass (Chl-*a*).

4. Discussion

4.1 Spatial variations in hydrographic properties

Cooler surface temperature (28.1±0.2 °C) and saline waters (33.3±0.2) in the SW coastal Bay of Bengal suggested the occurrence of coastal upwelling (Sarma et al., 2012a). Weaker stratification due to the discharge being less from the peninsular rivers (<4000 m³s⁻¹; Sarma et al., 2012b) allowed upwelling of subsurface waters to surface in the SW region. The occurrence of mild upwelling along the east coast of India was noticed earlier but confined close to the coast (Murty and Varadachari, 1968; Shetye et al, 1991; Gopalakrishna and Sastry, 1985; Sarma et al., 2013). Intensified upwelling due to decreased river discharge caused by a positive Indian Ocean Dipole (IOD) was reported off Godavari estuary (Sarma et al., 2015). In contrast in the NW coastal Bay of Bengal, low salinity (24.9±0.6) and warmer sea surface (29.7±0.1 °C), the upper ocean was stratified and the southwest winds could not rupture stratification (Sarma et al., 2013).

A sharp gradient in salinity and temperature was noticed around 18° N where the northward flowing high salinity western boundary current and southward flowing low salinity East India Coastal Current (EICC) meet (Shetye et al., 1993; Sarma et al., 2012a) resulting in the development of a salinity front. Although the peninsular rivers such as Mahanadi (2121 m³ s⁻¹), Baitarani (903 m³ s⁻¹), Subarnalekha (392 m³ s⁻¹) (Sarma et al., 2012b), discharge to the NW region, the discharge from the Ganges (15000 m³s⁻¹; Sarma et al., 2012a) dominated. The salinity gradient between surface and subsurface (50 m) was low (~0.5psu) in the SW than in the NW region (~13 psu) (Sarma et al., 2013). River discharge brings huge amounts of TSM to the coastal waters of Bay of Bengal. Though the influence of discharge i.e., terrestrial influence was high in the NW region, the TSM was higher in the SW region. The contribution of SPM by the Ganges river was small since its major flow is towards the west (Rao et al. 1988). Similarly, nitrate concentrations decreased from the SW (5.5 ± 3

μM) towards the NW region ($0.6 \pm 0.3 \mu\text{M}$), whereas phosphate ($0.3 \pm 0.2 \mu\text{M}$) did not show a significant difference (Fig. 3d, f). Though high nutrient concentrations were found in the SW region, relatively high Chl-*a* concentrations occurred in the NW ($3.1 \pm 0.4 \text{ mg m}^{-3}$) compared to that in the former ($1.5 \pm 0.3 \text{ mg m}^{-3}$) (Fig. 3b). The contrasting patterns in nutrients, TSM and Chl-*a* in the two regions meant that phytoplankton (chl-*a*) flourished better in the more intense light of the clearer waters of the NW region, consuming the nutrients (Fig. 3b). Hence, significant differences in CH_4 concentrations are expected between NW and SW coastal Bay of Bengal.

4.2 Spatial variations in concentrations and saturation of CH_4 in the coastal Bay of Bengal

The estuaries were highly saturated with respect to CH_4 and are major source of CH_4 to the coastal regions (Bange, 2006, Jayakumar et al 2001). This was addressed by the simultaneous samplings done. At three to five stations from the mouth of the estuary to about 10 km towards the upper estuary on the same day of sampling along the coastal transects (Table 2). The CH_4 concentrations were significantly higher in the monsoonal estuaries of Mahanadi (23.7-29.4 nM; mean of 25.7 ± 1.8 nM), Godavari (10.4-36.1 nM; mean of 23.2 ± 4.2 nM) and Krishna (11.8-27.5 nM; mean of 19.3 ± 4.5 nM) while the lowest CH_4 concentration was found in the Ganges estuary (4.7-5.2 nM; mean of 5 ± 0.1 nM). This study suggests that monsoonal river estuaries contain higher concentrations of methane than the glacial river estuary of the Ganges, which is consistent with prior work (Rao and Sarma, 2015). Lower concentrations of CH_4 in the NW (2.4 to 2.6 nM) than in the SW region (2.4 to 11.5 nM) (Table 1) suggest that the spatial variations are also governed by the CH_4 in the discharged water. Within the SW region, the CH_4 concentration and saturations off the Godavari estuary (GS, GN; 3.1 to 11.5 nM and 144.7-528% respectively) and the Krishna estuary (KS, KN; 2.4 to 3.6 nM; 111.4 to 138.2% respectively) were higher than those off Visakhapatnam (V; 2.5 ± 0.2 nM; $119.5 \pm 6.3\%$) where no major river discharge occurs suggesting that the source of CH_4 was the estuary where CH_4 was produced and that the CH_4 was transported to the adjacent coastal waters (Table 1). High concentrations of CH_4 in the Mahanadi estuary (25.7 ± 1.9 nM; $1095 \pm 68\%$) are non-typical for the NW region; however, are subjected to quick dilution by the CH_4 -poor coastal waters in which the chemical signals of Ganges river are strongly preserved due to its enormous size. This signal is lost as the water flows further south by dilution. The inverse relationship of CH_4 saturation with salinity ($r^2 = -0.57$; $p < 0.001$; Fig 4a; Table 3) does indicate that runoff is a major source of CH_4 even in the NW region, but the saturations are much less than in the case of estuaries of the SW region. (Fig. 2c). Methane saturation shows an inverse relation with DO saturation (Fig. 4b) and no significant relation with either zooplankton biomass or Chl-*a* concentration. The linear correlation of

CH₄saturation with pCO₂and TSM are highly significant ($r^2=0.49$, $p<0.001$ and 0.31 , $p<0.01$ respectively)andsupportthat input of fresh water with its greater heterotrophic activity mayexplain the highCH₄concentrations in the western coastal Bay of Bengal. Similar results were also reported in the European estuaries (Borges and Abril, 2012).

Concentrations and saturation of CH₄ in the coastal Bay of Bengal are less than those reported for Chinese coastal waters of the Jiaozhou Bay (Zhang et al., 2007), Dalian Bay (Wang et al., 2011) and Sanggou Bay (Hou et al., 2016) (Table 4). Such higher concentrations in the Chinese Bays were attributed to riverine CH₄inputs andby ground water sources. The concentrations of CH₄ in the rivers dischargingtothe Chinese Bays were 3 to 5 times higher than thosedischargingto the Bay of Bengal (Table 4). Relatively lower CH₄concentrations were observed in the Aegean (Bange et al., 1996) and Yellow (Zhang et al., 2004) seas where there is not significant input of riverine CH₄. This study suggests that river discharge is a major source of CH₄ to coastal waters in the western coastal Bay of Bengal.

Occurrence of mild upwelling further added CH₄ from subsurface to the surface waters ofthe SW region.The occurrence of mild upwelling and its impact on nutrients (Sarma et al., 2013) and phytoplankton pigments (Bandyopadhyay et al., 2017) along the southwestern coast of Bay of Bengal were reported. The increasing concentration of CH₄with depth at most of the stations sampled indicated that the subsurface waters can be a source, but only in the case of SW coastal water. The vertical distribution further suggested thatCH₄ concentrations in the upper 100 m were relatively lower in the NW compared to SW region. The isoline of 4nM CH₄ contour was situated below 40 m in the NW region, which was uplifted by20 m in the SW region (Fig. 5). On the other hand, the difference in CH₄ concentrations between surface and 50 m depth were large in the SW region when compared to NW region. The upsloping of isothermals and isohalines were reported (Sarma et al., 2013)as in the present case for CH₄in line with the suggestion that coastal upwelling enhanced CH₄ concentrations in the upper water column in the SW region. Thus,the CH₄ concentrations in the coastal Bay of Bengal were influenced by both physical processes namely upwelling in the coastal region and discharge of over saturated CH₄ waters from estuaries. The mean CH₄ saturation in the Bay of Bengal ($168.8\pm 21.9\%$) was found to be lower than that of the Arabian Sea (710 ± 582 ;Jayakumar et al., 2001) during southwest monsoon season.

4.3. Sea-to-air flux of CH₄ in the coastal Bay of Bengal

The sea-to-air flux of CH₄ inthecoastalBay of Bengalranged from $13.6-248.5 \mu\text{mol m}^{-2}\text{d}^{-1}$ with a mean of $47.6\pm 4.5 \mu\text{mol m}^{-2}\text{d}^{-1}$ in SW region while it was $24.4-68.4 \mu\text{mol m}^{-2}\text{d}^{-1}$ with a mean of 44.1 ± 1.1

$\mu\text{mol m}^{-2}\text{d}^{-1}$ in the NW region (Table 1). The wind speeds over the coastal Bay of Bengal were high (mean 18.8 m s^{-1} ; range, $13.1\text{-}25.5 \text{ m s}^{-1}$) during southwest monsoon period. The methane fluxes from the Bay of Bengal are higher compared to many oceanic regions in the world with the exception of the Black Sea ($53 \mu\text{mol m}^{-2} \text{ d}^{-1}$; Amouroux et al., 2002) and Dalian Bay (113.46 ± 146.82 ; Wang et al., 2011) (Table 4). The higher fluxes obtained ($46.3 \pm 3.5 \mu\text{mol m}^{-2} \text{ day}^{-1}$) are a result of vertical CH_4 gradients and high wind speeds during the study period. Our study suggests that the strength of the coastal Bay of Bengal as a source of CH_4 depends on the characteristics of the discharging water and its feedback on vertical mixing.

Rao and Sarma (2016) estimated a CH_4 flux rate of $13.4 \pm 5 \mu\text{mol m}^{-2} \text{ d}^{-1}$ from Indian estuaries which is ~ 30 times lower than that from European estuaries ($414 \pm 493 \mu\text{mol m}^{-2} \text{ d}^{-1}$; Upstill-Goddard and Barnes, 2016). This was attributed to a lower residence time of water in the Indian estuaries (2-10 days) than European estuaries (2-70 days). The total flux of CH_4 from the Indian estuaries amounted to $3.9 \times 10^9 \text{ g CH}_4 \text{ y}^{-1}$ (Table 5). The Indian mangroves were reported as a strong source for the atmospheric CH_4 (Biswas et al., 2007; Chauhan et al., 2008; Krithika et al., 2008; Dutta et al., 2013). Chauhan et al. (2008) estimated a mean CH_4 flux to the atmosphere from this source as $5.9 \mu\text{mol m}^{-2} \text{ d}^{-1}$ which amounts to $0.1 \times 10^9 \text{ g CH}_4 \text{ y}^{-1}$. The CH_4 fluxes from the west coast of India (Arabian Sea coast) during wet and dry periods were calculated as 11.25 and $1.37 \mu\text{mol m}^{-2} \text{ d}^{-1}$ respectively (Jayakumar, 1999). In comparison, the CH_4 flux from the east coast of India (Bay of Bengal coast) amounted to only $1.1 \mu\text{mol m}^{-2} \text{ d}^{-1}$ during dry period (Rao, 2013), which is about ~ 40 times higher than fluxes estimated from the Bay of Bengal during wet period ($46.3 \mu\text{mol m}^{-2} \text{ d}^{-1}$; this study). In the Arabian Sea, the concentration of CH_4 was only somewhat higher during the wet ($3.6 \pm 0.3 \text{ nM}$) period than the dry period ($2.2 \pm 0.02 \text{ nM}$) but wind speeds were significantly higher during former ($18.8 \pm 0.5 \text{ m s}^{-1}$) than latter ($2.7 \pm 0.3 \text{ m s}^{-1}$), leading to much higher fluxes during the former than the latter season (Jayakumar, 1999). Compared to the mean flux rate of the eastern coastal Arabian Sea ($0.23 \times 10^9 \text{ g y}^{-1}$), the rate of the western coastal Bay of Bengal was $15.7 \times 10^9 \text{ g y}^{-1}$ (Table 5). Overall, the fluxes of CH_4 from Indian coastal bodies are in the order, western coastal Bay of Bengal (70% of the total $\sim 22 \times 10^9 \text{ g y}^{-1}$ (Table 5) > Indian estuaries (20%) > the eastern coastal Arabian Sea (10%). The CH_4 flux from the Indian coastal waters ($1.7\text{-}46.3 \mu\text{mol m}^{-2} \text{ d}^{-1}$) was lower than Chinese ($0.8\text{-}113.4 \mu\text{mol m}^{-2} \text{ d}^{-1}$), European ($1.6\text{-}53 \mu\text{mol m}^{-2} \text{ d}^{-1}$) and American (5 to $86.5 \mu\text{mol m}^{-2} \text{ d}^{-1}$) coastal waters whereas it was more than from the African coastal waters ($0.1\text{-}1.7 \mu\text{mol m}^{-2} \text{ d}^{-1}$). The higher fluxes calculated in the case of the Bay of Bengal were strongly influenced by the very high

wind speeds during the sampling period, which aspect needs to be addressed by a high temporal resolution study for a more realistic estimation.

5. Summary and conclusions

The influence of river discharge on concentrations and fluxes of CH₄ was studied along the west coast of the Bay of Bengal during peak discharge period. Significant differences in hydrographic properties were observed between the SW and NW regions of the study area e.g., upwelling in the former and a strong surface stratification in the latter. The rivers that join the SW coastal Bay of Bengal (Godavari and Krishna Rivers) had higher concentrations of CH₄ whereas the R. Ganges that joins at the NW region with its large discharge had lower concentrations of CH₄. This study suggests that freshwater is the major source of CH₄ to the coastal Bay of Bengal as evidenced from a decrease in CH₄ concentrations from estuaries to the open ocean in association with an increase in salinity. Coastal upwelling acted as a secondary source of CH₄ in the surface water of the SW region. The mean CH₄ saturation (168.8±21.9%) in the Bay of Bengal was found to be less than that reported for the Arabian Sea (710±582%) during southwest monsoon season. The mean CH₄ flux was 46.3±3.5 μmol m⁻² day⁻¹ from the Bay of Bengal which is lower than reported for other coastal regions in the world, except in the African coastal waters. The fluxes of CH₄ from the Indian coast (especially Bay of Bengal) are likely overestimated due to prevailing high wind speeds. This study further suggests that decrease in river discharge due to natural processes, such as weakening of monsoon, El Nino or Indian Ocean Dipole (IOD), or anthropogenic processes, such as interconnecting of rivers, construction of more dams, may reduce fluxes of CH₄ to the atmosphere. The interconnection of Indian rivers is already underway and more dams are proposed on Indian rivers. Reduced river discharge may promote coastal upwelling during southwest monsoon (Sarma et al., 2015) that may enhance CH₄ concentrations and fluxes to the atmosphere. Nevertheless, it would be interesting to monitor time-series variations of concentrations and fluxes of CH₄ along the Indian coast to examine possible variations in future.

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Legend to figures

Figure 1: Sampling locations in the western coastal Bay of Bengal. Isobaths of 250, 500 and 1000 m were also shown. Acronyms K, G, VD, HD, and M stand for rivers Krishna, Godavari, Vamsadara, Hyadri and Mahanadi whereas N and S attached to the river names represent north and south of the mouth of the estuary respectively.

Figure 2: Distribution of a) Salinity b) Temperature ($^{\circ}\text{C}$) c) DO (μM) d), TSM(mg l^{-1}), e) nitrate (μM), f) phosphate (μM), and g) Chl-*a* (mg m^{-3}) in the west coast of the Bay of Bengal

Figure 3: Distribution of c) CH_4 (nM), d) CH_4 Saturation (%) in the west coast of the Bay of Bengal.

Figure 4: Relationship of CH_4 (nM) with (a) salinity and (b) DO saturation (%) in coastal Bay of Bengal.

Figure 5: Vertical distribution of CH_4 along different transects. X- axis shows distance from the coast (km).

Figure 1:

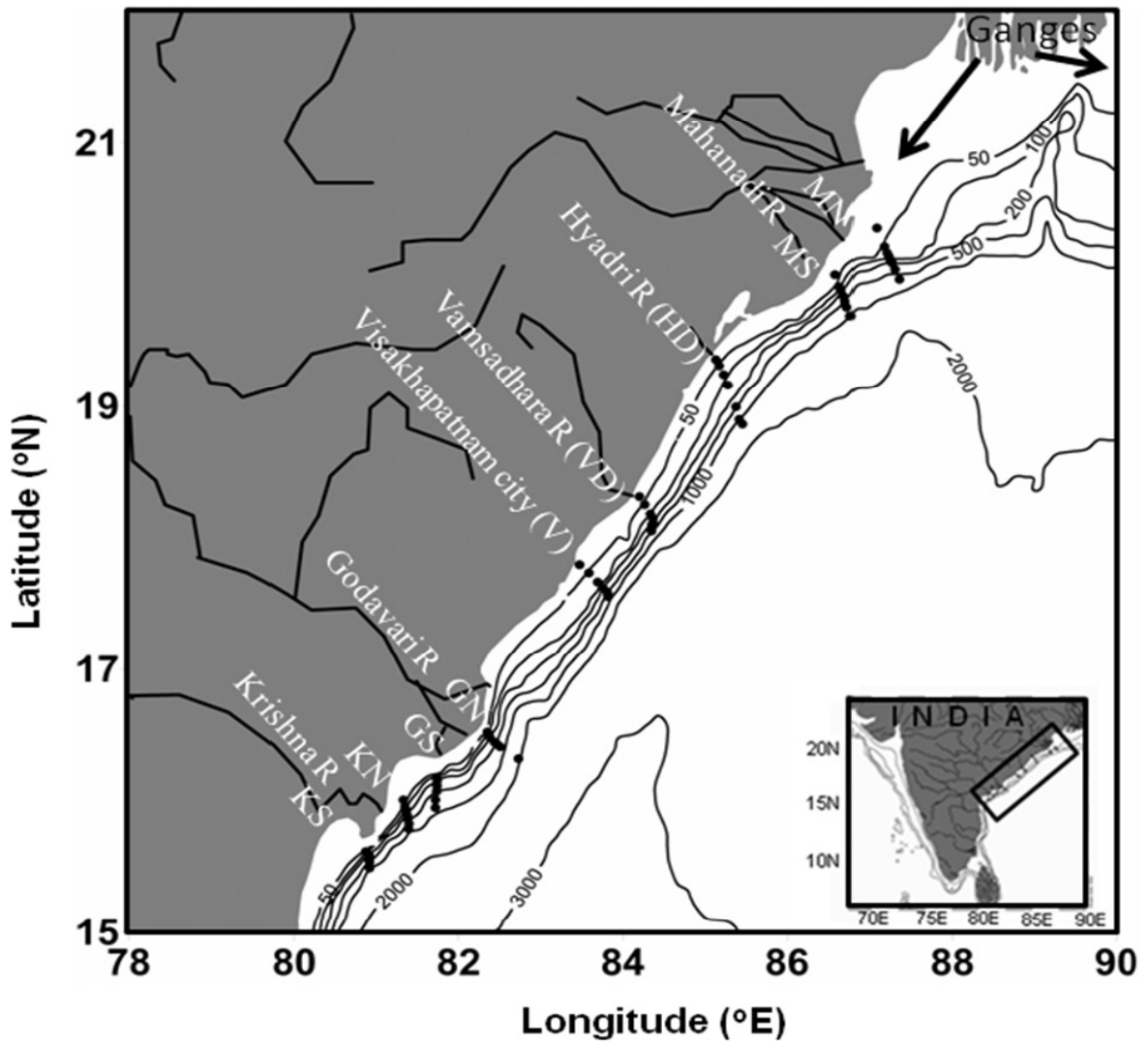


Figure 2:

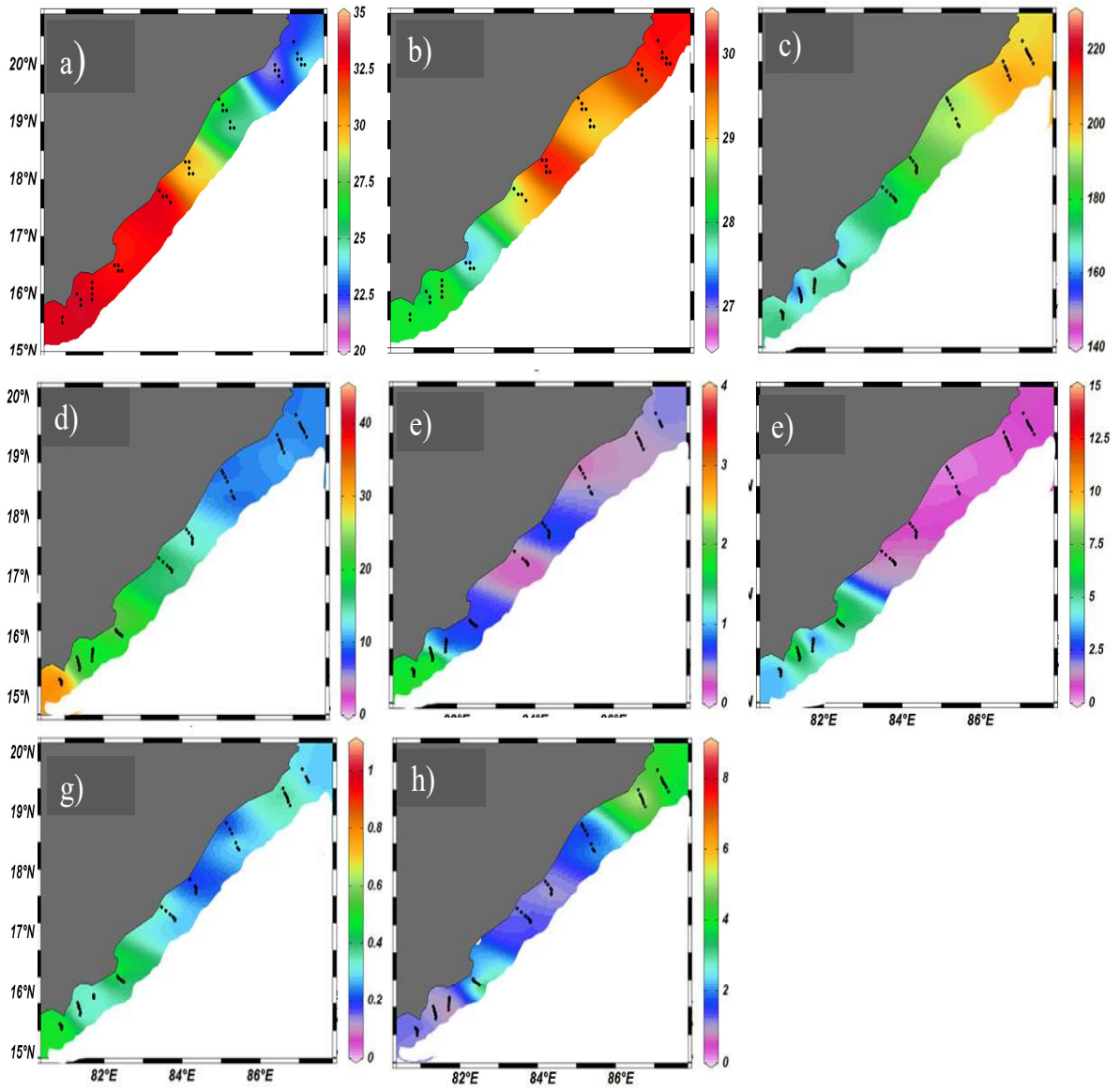


Figure 3:

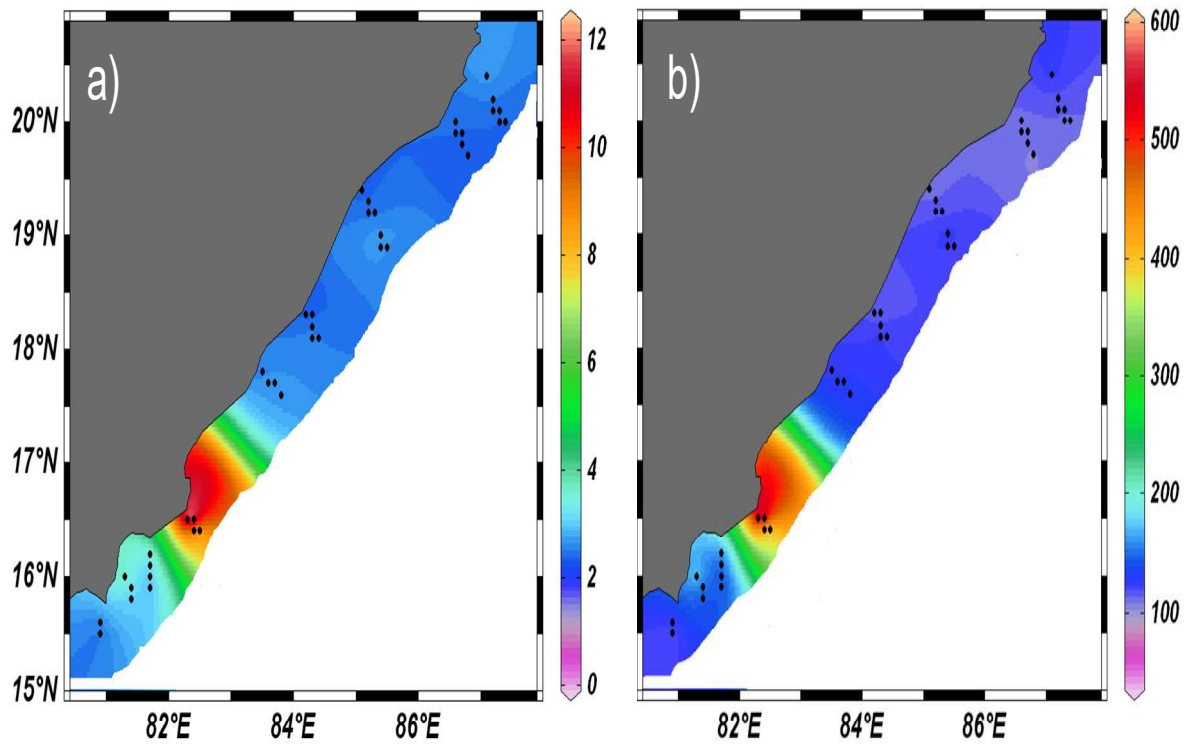


Figure 4 a):

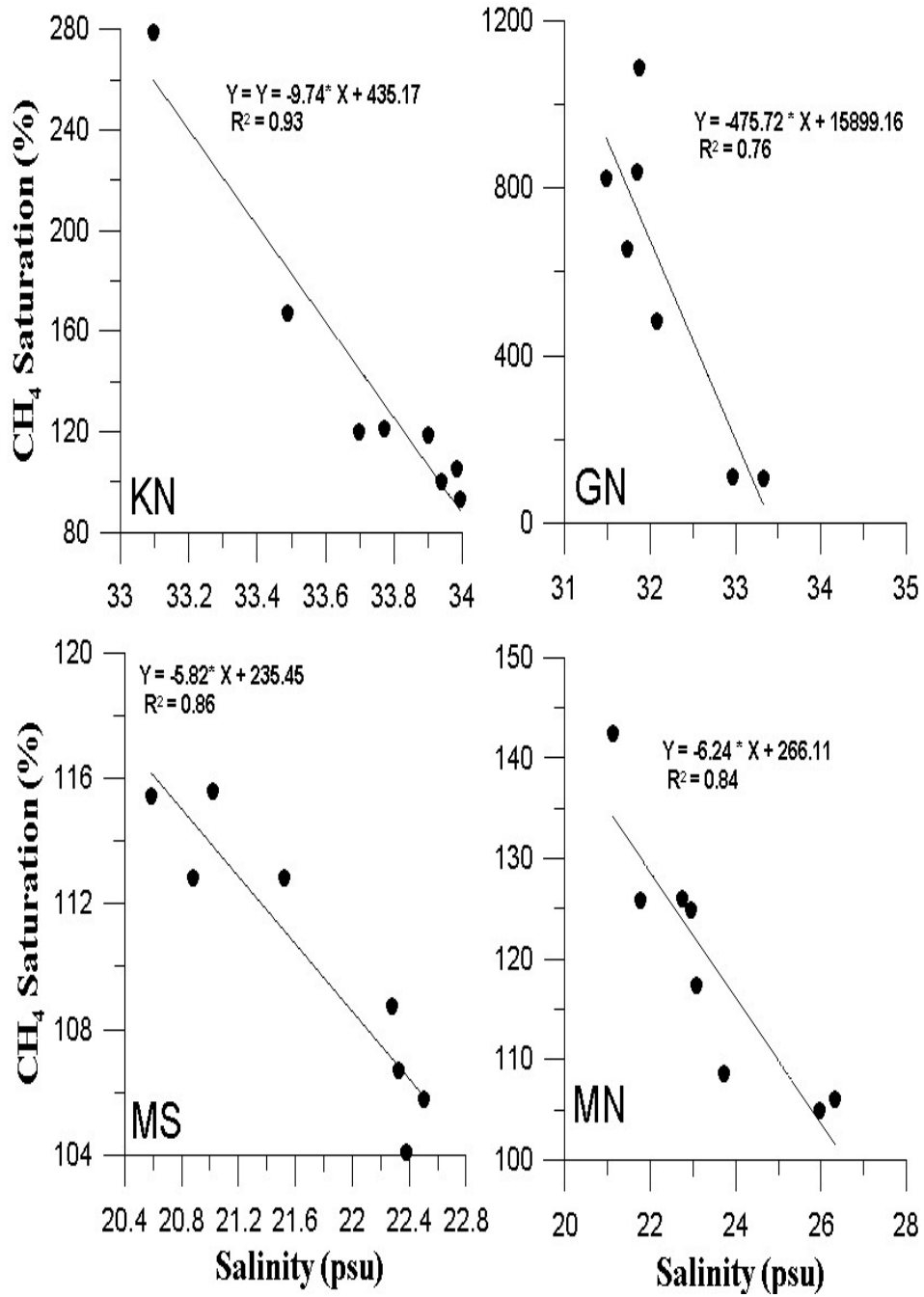


Figure 4 b)

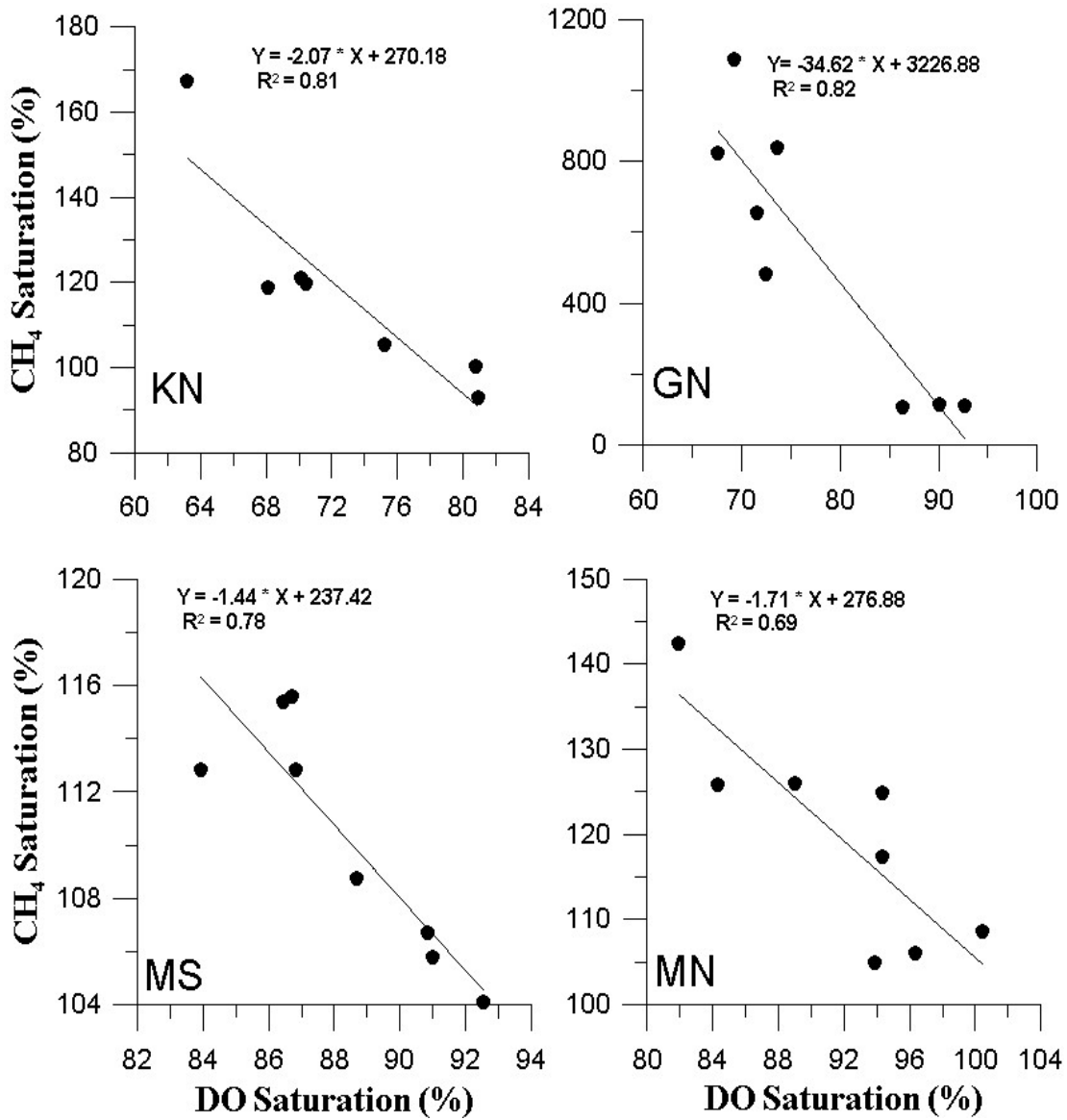


Figure 5:

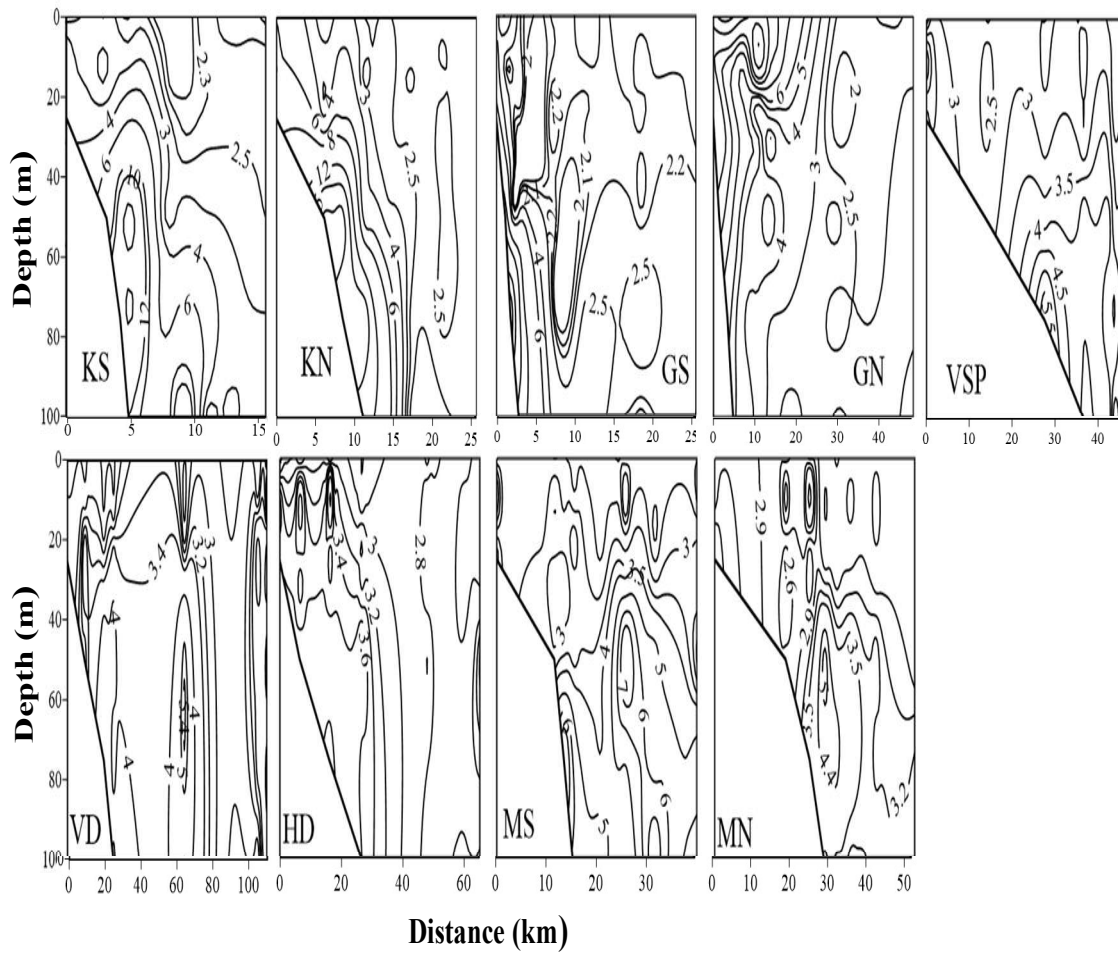


Table 1: Mean (\pm SE) of sea surface salinity, temperature ($^{\circ}$ C), DO (μ M), NO₃ (μ M), PO₄ (μ M), SiO₄ (μ M), Chl-a (mg m^{-3}), CH₄ (nM), CH₄ saturation (%), wind speed (m/s) and CH₄ flux ($\mu\text{mole m}^{-2} \text{d}^{-1}$) at water-air interface along the different cruise transects in the coastal Bay of Bengal.

Transect	Salinity	Temp ($^{\circ}$ C)	DO (μ M)	TSM (mg l^{-1})	NO ₃ (μ M)	PO ₄ (μ M)	SiO ₄ (μ M)	Chl-a (mg m^{-3})	CH ₄ (nM)	CH ₄ Saturation (%)	Wind (m/s)	Flux ($\mu\text{mole m}^{-2} \text{d}^{-1}$)	
SW Region	KS	32.9 \pm 0.5	28.3 \pm 0.2	171.9 \pm 3	33.7 \pm 2.7	3 \pm 0.3	0.5 \pm 0.2	7.3 \pm 1.4	1.3 \pm 0.2	2.4 \pm 0.1	111.4 \pm 4.8	13.1	15.3 \pm 0.2
	KN	34 \pm 0.1	27.8 \pm 0.1	152.7 \pm 6.5	29.4 \pm 9.3	7.7 \pm 2	0.3 \pm 0.1	3.1 \pm 0.3	1.1 \pm 0.2	3 \pm 0.5	138.2 \pm 21.7	18.1	35.8 \pm 1.2
	GS	33.9 \pm 0.1	28.2 \pm 0.2	171.3 \pm 2.3	19.9 \pm 2	2.4 \pm 0.4	0.2 \pm 0.1	1.9 \pm 0.4	0.9 \pm 0.2	3.1 \pm 0.7	144.7 \pm 29.4	16.8	32.3 \pm 1.4
	GN	32.2 \pm 0.3	27.4 \pm 0.3	166.2 \pm 7.1	26.9 \pm 7.1	5.4 \pm 0.8	0.7 \pm 0.3	5.9 \pm 0.7	3.1 \pm 1	11.5 \pm 3	528.5 \pm 136.1	16.7	119.9 \pm 7.5
	VSP	33.4 \pm 0.1	29 \pm 0.2	172.5 \pm 4.6	16.8 \pm 1.1	2.2 \pm 1.1	0.4 \pm 0.1	6.5 \pm 0.5	1.4 \pm 0.2	2.5 \pm 0.2	119.5 \pm 6.3	20.2	36.9 \pm 0.8
NW Region	VD	29.2 \pm 0.5	29.9 \pm 0.1	187.1 \pm 2.5	12.5 \pm 1.3	0.8 \pm 0.4	0.2 \pm 0.1	6.9 \pm 0.3	1.1 \pm 0.1	2.5 \pm 0.2	118.1 \pm 7.7	17.2	29.5 \pm 0.9
	HD	25.7 \pm 0.7	29.1 \pm 0.2	190.8 \pm 1.8	9.2 \pm 1.4	0.3 \pm 0.2	0.3 \pm 0.1	4.2 \pm 0.4	1.7 \pm 0.4	2.6 \pm 0.2	117.5 \pm 6	17.9	28.2 \pm 0.6
	MS	21.7 \pm 0.3	29.8 \pm 0.1	196.1 \pm 2.1	9.9 \pm 1	0.5 \pm 0.3	0.3 \pm 0.1	6.6 \pm 0.6	3.6 \pm 0.6	2.4 \pm 0.1	109.2 \pm 2	24.2	57.8 \pm 0.6
	MN	23.5 \pm 0.7	29.9 \pm 0.1	201.8 \pm 5	14.8 \pm 2.9	0.8 \pm 0.3	1.3 \pm 0.8	21.5 \pm 2.4	5.4 \pm 0.6	2.6 \pm 0.2	119.6 \pm 4.6	25.5	56.6 \pm 0.5

Table 2: Ranges and mean concentrations of River discharge, (\pm SE) salinity, concentration, saturation and fluxes of CH₄ in the estuaries.

Estuary	Mean discharge (m ³ s ⁻¹)	Salinity	CH ₄ conc (nM)	CH ₄ Saturation (%)	Flux (μmole m ⁻² d ⁻¹)
Krishna	2213 ^b	4.75-6.03 (5.27±0.6)	11.8-27.5 (19.3 ±4.5)	466-1085 (760 ± 179)	265-716 (480 ± 130)
Godavari	3505 ^b	0.08-0.3 (0.16±0.1)	10.4-36.1 (23 ± 4.2)	410-1407 (882 ± 161)	225-799 (485 ± 93)
Mahanadi	2121 ^b	0.06-0.07 (0.07±0.1)	23.7-29.4 (25.7 ±1.8)	980-1216 (1095 ± 68)	1161-1472 (1312 ± 90)
Ganges	15000 ^a	0.56-0.81 (0.71±0.1)	4.7-5.2 (5± 0.1)	196-220.6 (208.7 ±7)	10.6-13.4 (12 ± 0.8)

^a Data taken from Sarma et al.,2012^a

^bData taken from Sarma et al.,2012^b

Table 3: Pearson correlation between CH₄ and its saturation with physicochemical parameters.

	CH ₄	Saturation	Salinity	Temp	DIN	DO	Chl- <i>a</i>	AOU	TSM	pCO ₂	
CH ₄	1										
Saturation	1.0**	1									
Salinity	-0.57**	-0.56**	1								
Temp	-0.55**	-0.55**	-0.69**	1							
DIN	0.33**	0.33**	0.54**	-0.69**	1						
DO	-0.42**	-0.42**	-0.69**	0.79**	-0.68**	1					
Chl- <i>a</i>	-	-	-0.56**	0.37**	-0.33**	0.53**	1				
AOU	0.46**	0.47**	0.52**	-0.77**	0.69**	-0.95**	-0.41**	1			
TSM	0.31*	0.31**	0.41**	-0.48**	0.42**	-0.38**	-	0.36**	1		
pCO ₂	0.49**	0.49**	0.82**	-0.85**	0.67**	-0.84**	-0.42**	0.81**	0.49**	1	

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

Table: 4 Compilation of concentrations, saturation, and fluxes of CH₄ from the various estuaries and coastal and shelf waters. The fluxes for different continental coasts are given as ranges and were obtained from the surveys given in the table.

Study area	CH ₄ (nM)	CH ₄ Saturation (%)	CH ₄ Flux (μmol m ⁻² d ⁻¹)	Reference
NW Black Sea (Europe)	13.1±10.6	173–10500	53	Amouroux et al., 2002
Aegean Sea (Europe)	4.8±0.3	231±32	1.6	Bange et al., 1996
Yellow Sea (China)	3.4±0.2	121±5.4	0.8±0.5	Zhang et al., 2004
Jiaozhou Bay (China)	34.8 ± 75.5	-	36.9 ± 87.3	Zhang et al. 2007
Dalian Bay (China)	56.1 ± 69.3	-	113.4 ± 146.8	Wang et al., 2011
East China Sea (China)	6.2 ± 4.9	317±236	11.5 ± 111.9	Ye et al., 2016
Bohai Sea (China)	5.87 ± 2.02	288 ± 99	8.1 ± 4.2	Li et al., 2010
North Yellow Sea (China)	12.2 ± 7.5	515.2±231	14.6 ± 22.3	Yang et al., 2010
NS China Sea (China)	2.4–5.9	134–297	15.6±8.0	Zhou et al., 2009
East China Seas (China)	8.2 ± 6.2	-	16.9 ± 21.4	Sun et al., 2015
Northwest Africa (Africa)	-	-	0.4–1.7	Kock et al., 2008
Eastern North Pacific (US)	-	-	0.5–5.9	Sansone et al., 2004
Eastern South Pacific (US)	-	-	0.9–18.7	Farias et al., 2009
Northern Gulf of Mexico (US)	1.7-23.5	46-1196	-4.9-86.5	Hu et al., 2012
Gulf of Cadiz (Europe)	1.6-51.1	70-1820	4.7±4.6	Ferron et al., 2010
Eckernforde Bay (Europe)	6.6±0.2	554±317	6.3±14.7	Bange et al., 2010
Arabian Sea (India)	2.6-20.3	140-1091	11.3±3.7	Jayakumar, 2000
Bay of Bengal (India)	6.4±8.2	-	6.7±7.4	Barner et al., 2003
Bay of Bengal (India)	1.9-23.6	117-1088	26.5±31.9	Present study
Fluxes from coastal regions (μmol m⁻² d⁻¹)				
African coastal waters			0.1-1.7	
Chinese coastal waters			0.8-113.4	
European coastal waters			1.56-53	
American coastal waters			-4.9-86.5	
Indian coastal waters			1.7-46.3	

Table:5 CH₄ fluxes from the different components of the Indian coastal waters

Region	Area (km²)	CH₄ Flux ($\mu\text{mole m}^{-2} \text{d}^{-1}$)	CH₄ Emission ($\times 10^9 \text{g y}^{-1}$)	Reference
Estuaries	27450	13.4	3.90	Rao and Sarma,2016
Mangroves	570	5.9	0.1	Chauhan et al., 2008
Arabian Sea (wet)	50000	11.25	1.6	Jaikumar,2000
Arabian Sea (dry)	50000	1.37	0.2	Jaikumar,2000
Bay of Bengal (wet)	114000	38.9	15.7	Present Study
Bay of Bengal (dry)	114000	1.1	0.3	Rao, 2013
Total	252711	-	21.9	Present Study