

Modifications in trace gases flux by Very Severe Cyclonic Storm '*Hudhud*' in the coastal Bay of Bengal

V.R. Kumari, V.V.S.S. Sarma, G.D. Rao, R. Viswanadham, B. Navita, T.N.R. Srinivas, M.S. Krishna and N.P.C. Reddy

CSIR-National Institute of Oceanography,
Regional Centre, 176 Lawsons Bay Colony,
Visakhapatnam – 530 017
Email: sarmav@nio.org

Abstract

A very severe cyclonic storm, '*Hudhud*', made landfall at Visakhapatnam city, central east coast of India, on 12th October 2014 and it is the most destructive cyclone that ever hit Indian subcontinent since past two decades. In order to examine its impact on the flux of trace gases to the atmosphere, a study was made in the coastal Bay of Bengal, off Visakhapatnam, after the cyclone and compared with the pre-cyclone conditions. '*Hudhud*' suppressed vertical mixing of water column due to occurrence of strong salinity stratification associated with torrential rainfall. The land runoff and precipitation brought significant amount of ammonium to the coastal waters resulting in increased pH. The increased pH shifted inorganic carbon equilibrium towards formation of bicarbonate resulting in decreased partial pressure of carbon dioxide ($p\text{CO}_2$) after cyclone '*Hudhud*'. Undersaturation of CO_2 and nitrous oxide (N_2O) with respect to atmospheric equilibrium was observed during post- compared to pre-cyclone period. About 80% of the post-cyclone decrease in N_2O (>2 nM) and $p\text{CO}_2$ (150 to 200 μatm) was contributed by dilution of coastal waters with the precipitated waters. In contrast, methane (CH_4) concentrations were increased by 0.5 to 2.8 nM during post cyclone than pre-cyclone period, and were attributed to the input of domestic sewage through land runoff. Dimethyl sulphide (DMS) and the total Dimethyl sulphonio-propionate (DMSP_i) concentrations decreased by 0.4 to 3.9 nM and 0.2 to 6.0 nM respectively during post cyclone in comparison to pre-cyclone period and it was consistent with lower phytoplankton biomass during the former than the latter. The sea-to air flux of CO_2 , N_2O and DMS were 1.3 ± 0.5 $\text{mmol C m}^{-2} \text{d}^{-1}$, 0.9 ± 0.3 $\mu\text{mol m}^{-2} \text{d}^{-1}$ and 5.8 ± 3 $\mu\text{mol m}^{-2} \text{d}^{-1}$ during pre-cyclone period respectively. The corresponding values during post-cyclone were lower at -2.0 ± 1 $\text{mmol C m}^{-2} \text{d}^{-1}$, -0.4 ± 0.1 $\mu\text{mol m}^{-2} \text{d}^{-1}$ and 2.8 ± 2 $\mu\text{mol m}^{-2} \text{d}^{-1}$ respectively. In contrast, the sea-to-air flux of CH_4 increased from 0.6 to 1.5 $\mu\text{mol C m}^{-2} \text{d}^{-1}$ from pre- to post-cyclone period. This study suggested that the cyclone '*Hudhud*' modified the magnitude of the biogenic gases fluxes to the atmosphere from the coastal Bay of Bengal, than hitherto hypothesized.

Key words: Trace gases, Carbon dioxide, tropical cyclone, fluxes, Bay of Bengal

1. Introduction

Tropical cyclones are extreme atmospheric events which strongly affect the physical structure of the upper ocean along their track (D'Asaro, 2003; Cione and Uhlhorn, 2003; Levy et al., 2012). As a response to tropical cyclones, sea surface temperature significantly cools by several degrees (2-10° C; Chiang et al., 2011; Maneesha et al., 2011), associated with increase in phytoplankton biomass (Di Tullio and Laws, 1991). This is mainly caused from the wind-driven entrainment of subsurface cold and nutrients enriched waters (Price, 1981; Vincent et al., 2013).

Influence of cyclones on trace gas fluxes have been studied at several regions (Perrie, 2004; Nemoto et al., 2009). It was noticed that hurricanes in the North Atlantic Ocean removed significant amount of CO₂ from the ocean surface resulting in its undersaturation (Bates et al., 1998). It was reported that cyclones cause enormous efflux of CO₂ from the ocean to the atmosphere to the extent of affecting the annual local efflux (Perrie, 2004; Nemoto et al., 2009). Though cyclones are short lived and travel at considerable speed, they affect significant areas (several 100 km) (Willoughby et al., 2006). It was hypothesized that they exert a significant influence on the global air-sea flux of CO₂ (Bates et al., 1998); however, measurements under cyclone conditions are rather sparse to examine its influence. Crosswell et al. (2014) observed extensive CO₂ emissions from the shallow coastal waters during passage of Hurricane Irene over the Mid-Atlantic Coast of the USA. Huang and Imberger (2010) modelled impact of Hurricane Frances on CO₂ fluxes using a coupled three dimensional hydrodynamic and carbon model. They noticed that the passage of Hurricane Frances was estimated to have caused a CO₂ efflux of about 3.5 to 10.3 TgC (1 Tg=10¹² g) from ocean to the atmosphere. Levy et al. (2012) noticed that tropical cyclones influence CO₂ fluxes significantly at the regional levels; however, their impact on global level is rather insignificant at longer time scales due to consequential removal of CO₂ through biological processes promoted by the entrained nutrients.

Naik et al. (2008) observed that tropical cyclone in the Arabian Sea cooled surface water temperature by ~4° C and increased nutrients and primary production. The new production supported by cyclone was estimated to be equivalent to ~5% of the annual organic carbon export to the deep sea (beyond continental margin) for the entire Arabian Sea. They further noticed that entrainment of N₂O from the thermocline led to more than doubling of its concentration in the mixed layer. Unlike the Arabian Sea, which is weakly stratified due to less fresh water discharge from the rivers, the Bay of Bengal receives significant amounts of freshwater from various rivers such as Ganges, Brahmaputra, Mahanadi, Godavari etc, resulting in low surface water salinity by 3-5 units compared to the Arabian Sea (UNESCO, 1979; Varkey et al., 1996).

The Bay of Bengal is considered to be low productive zone due to inhibition of vertical mixing by strong vertical salinity gradient (Prasanna Kumar et al., 2002). Faster removal of organic matter through mineral ballast further lowers the decomposition of organic matter in the water column (Ittekkot et al., 1991). As a result, Kumar et al. (1996) suggested that Bay of Bengal is a net sink for atmospheric CO₂. Based on the seasonal observations in the surface waters during pre and post monsoon period, it was observed that Bay of Bengal is a mild source of N₂O to the atmosphere (Naqvi et al., 1994; Hashimoto et al., 1998). Recent observations along east coast of India suggested that river discharge from the peninsular region of India brought CO₂ and N₂O rich waters to the coastal region and acted as a mild source to the atmosphere (Sarma et al., 2012; Rao and Sarma, 2014). More recently, Sarma et al. (2015a) found enhanced acidification in the northern than southern Bay of Bengal due to increased air pollution associated with the recent increase in industrial activities and concluded that the northern Bay of Bengal is acting as a perennial source of CO₂ to the atmosphere. Very few observations have been carried out so far on CH₄ in the water and over atmosphere in the Bay of Bengal. Berner et al. (2003) measured CH₄ concentrations in the Northern Bay of Bengal during northeast monsoon and reported that the strong sources of CH₄ to the atmosphere at the mouth of Ganges and Brahmaputra rivers, and on annual scale, the Bay of Bengal is acting as a source ($6.65 \pm 7.36 \mu\text{mol m}^{-2} \text{d}^{-1}$) to the atmosphere. No studies have been carried so far in the coastal Bay of Bengal to examine its contribution to the atmospheric CH₄. Shenoy et al. (2006) and Rao et al. (2015) noticed that coastal Bay of Bengal is a strong source for atmospheric Dimethyl sulphide (DMS) during monsoon. Therefore, in all earlier studies, the coastal Bay of Bengal has been observed as a strong to mild source of several trace gases (CO₂, N₂O, CH₄ and DMS) to the atmosphere.

The flux of trace gases depends on the piston velocity and the difference of gas concentrations between atmosphere and ocean surface. The piston velocity which is a function of wind speed strongly increased during the cyclone conditions (Liss and Merlivat, 1986; Wanninkhof, 1992, D'Asaro and McNeil, 2007; McNeil and D'Asaro, 2007). In addition to this, high surface winds churn up the upper ocean that causes injection of cool, nutrients and trace gas rich (mainly for CO₂, CH₄ and N₂O) waters to the surface (Malone et al., 1993; Shiah et al., 2000; Naik et al., 2008; Levy et al., 2012). The enhanced nutrients increase primary production that would modify production/consumption of some of the trace gases. In addition to this, heavy precipitation associated with cyclone, and landfall brings land-derived nutrients that may have significant impact on trace gas concentrations in the coastal regions.

The fluxes of trace gases are expected to be modified under extreme atmospheric events such as cyclones. The objective of the present study is to examine the variability in trace gases concentrations and their fluxes in the coastal Bay of Bengal after cyclone 'Hudhud' compared to the pre-cyclone period.

2. Material and methods

2.1. Description of cyclone 'Hudhud'

A Very Severe Cyclonic Storm 'Hudhud' hit the east coast of India on 12 October 2014 and it was the strongest tropical cyclone over the North Indian Ocean in past two decades. Cyclone 'Hudhud' originated from a low pressure system that formed under the influence of an upper-air cyclonic circulation in the Andaman Sea on 6 October 2014. 'Hudhud' intensified into a cyclonic storm on 8 October and as a Severe Cyclonic Storm on 9 October 2014. 'Hudhud' underwent rapid strengthening in the following days and was classified as a Very Severe Cyclonic Storm by the Indian Meteorological Department (IMD). Shortly before landfall near Visakhapatnam city, Andhra Pradesh, on 12 October, 'Hudhud' reached its peak strength with three minute wind speeds of 217 km h⁻¹ and a minimum central pressure of 960 mbar. After the landfall, the system drifted northwards towards Chathisgarh, Uttar Pradesh and Nepal, causing widespread rains in those areas and heavy snowfall in Nepal. 'Hudhud' caused extensive damage to the city of Visakhapatnam and the neighbouring districts of Vizianagaram and Srikakulam of Andhra Pradesh. Damages are estimated to be between US\$ 9.7 and 11 billion. At least 96 deaths have been confirmed, a majority of them from Andhra Pradesh state.

2.2. Sampling and methods

As a part of Ocean Finder program, *in situ* measurements were carried out in the coastal Bay of Bengal, off Visakhapatnam (Transect A) and off Achutapuram (Transect B) during 14th and 15th October 2014, from close to coast to about 22.5 and 30 kms offshore respectively where water depth was up to 75m (Fig. 1). The transect A is our time-series observation region since 2007 (Sarma et al., 2013) and transect B is sampled during this period as cyclone crossed coast at this location. Five stations were occupied along each transect and water samples were collected at standard depths from surface to bottom of the water column using 10 L Niskin sampler. Water column temperature and salinity was measured using portable conductivity-temperature-depth profiler (CTD; Sea Bird Electronics, USA). Nutrients (nitrate, nitrite, ammonium, phosphate and silicate) were measured following spectrophotometric method (Grashoff et al., 1983). About 3 litres of water was filtered

through glass fibre filter (GF/F; 0.7 μm ; Whatman) and pigment retained on the filter was extracted to dimethyl formamide (DMF) and the fluorescence of the extract was measured using spectrofluorophotometer (Cary Eclipse, Varian Instruments, USA) following Suzuki and Ishimaru (1990).

The pH and total alkalinity (TA) were measured by potentiometric (Metrohm, Zofingen, Switzerland) Gran-titration method following standard operating procedures suggested by the Department of Energy (DOE, 1998). Dissolved inorganic carbon (DIC) was measured using a Coulometer (model CM 5014; UIC Inc., Joliet, Illinois, USA) attached to an automated subsampling system (Sarma, 1998). The precisions for pH, TA and DIC were ± 0.002 , ± 2.0 and $\pm 1.8 \mu\text{mol l}^{-1}$, respectively. Based on the certified reference material (CRM) supplied by Dr. A.G. Dickson, Scripps Institute of Oceanography, USA, the accuracy of the measurements of DIC, TA and PH was estimated to be 0.03, 0.05 and 0.02% respectively.

Dissolved N_2O and CH_4 in the water were determined by a multiphase head space equilibration technique (McAuliffe, 1971) coupled with Gas Chromatographic (GC) analysis. Predetermined volume (25 ml) was equilibrated with equal volume of ultrapure helium in a gas tight syringe by vigorously shaking the syringe at room temperature for 5 minutes using wrist action shaker. After equilibrium, the head space was dried over drierite and removed CO_2 using ascarite scrubber and then injected through a sampling loop into a gas chromatograph (Agilent - 6820, USA) and separated over a chromosorb column (80/100 mesh) at 35°C . N_2O and CH_4 peaks were detected with an Electron Capture and Flame Ionization Detectors respectively followed by Jayakumar et al. (2001) and Patra et al. (1998). The calibrated N_2O and CH_4 gases (Spectra gases, USA) were used for calibration of GC. The precision of N_2O and CH_4 measurements was $<3\%$.

Dimethyl Sulphide (DMS) and total Dimethyl sulphoniopropionate (DMSP_t) samples were collected into 40 ml amber colour glass bottles to prevent their photochemical decomposition and were analysed using the Agilent 7890 Series GC. The samples were stored in the dark at 4°C until analysis. The storage time never exceeded 4 to 6 hours. Calibration of the instrument has been carried out using standard DMSP (Research Plus, Netherlands) solutions. DMS and DMSP_t in the samples were analysed using purge and trap method (Shenoy, 2002). A known volume (sample volume varied from 5 to 20 ml depending on the concentration of DMS) of a seawater sample was purged with ultrapure nitrogen gas (99.9995%) for 15 minutes. The released gases were trapped cryogenically in a Teflon column in line with 6 port valco valve. The trapped gases were injected by inserting the Teflon column into boiling water (temperature $>80^\circ\text{C}$) and switching the valco valve to

inject position. The trapped and released gases were then separated over a Chromosil 330 column (Shenoy, 2002; Turner et al., 1990) and detected by Flame Photometric Detector (FPD). The DMS was measured first followed by DMSP_t. About 2ml of 10 M NaOH was added to the same samples, after DMS has been analysed, and purged immediately for 20 min to hydrolyze DMSP_t to DMS. This results in cleavage of DMSP_t to DMS and acrylic acid (Turner et al., 1990) and the DMS produced by cleavage was measured as detailed above. The precision of the DMS measurements was <8% (RSD).

2.3. Computations

The pCO₂ was computed using measured salinity, temperature, nutrients (phosphate and silicate), pH and DIC using CO₂ SYS program (Lewis and Wallace, 1998). The CO₂ dissociation constants given by Millero et al. (2006) in the 0-40 salinity range were used. The estimated pCO₂ carries an error of 8 μatm. The solubility of N₂O and CH₄ were computed using *in situ* temperature and salinity using coefficients given by Weiss and Price (1980) and Wiesenburg and Guinasso (1979) respectively. Wind speed data from the coastal Visakhapatnam city were obtained from the Indian Meteorological Department (IMD) at Visakhapatnam. The air-water flux of trace gases was estimated using formulations given by Wanninkhof (1992) based on concentration of gas at the surface, solubility of gas in the surface waters at *in situ* temperature and salinity and wind speeds measured at 10 m above the sea level.

3. Results and Discussion

3.1. Variations in hydrographic properties in the coastal Bay of Bengal after cyclone Hudhud

Sarma et al. (2018b) discussed the impact of cyclone ‘Hudhud’ on upper ocean physical processes, phytoplankton biomass and primary production in the coastal Bay of Bengal using field measurements, satellite data and models. However, we will briefly discuss the changes in the water column properties before and after cyclone and mean variation in temperature, salinity, Chl-a and nutrients during before and after ‘Hudhud’ are given in Table 1. For comparison of the after cyclone conditions (henceforth called ‘post cyclone period’) with pre-cyclone condition (henceforth called ‘pre-cyclone period’), the data collected during September 2014, two weeks prior to cyclone were used. The satellite SST images evidenced decrease in surface water by 1-3° C along the cyclone track in the offshore whereas the decrease was <1° C close to the coast (Sarma et al., 2018b). The weaker cooling close to the coast was attributed to strong vertical salinity stratification driven by heavy precipitation associated with cyclone (Fig. 2). The sea surface height (SSH) was shallower along the

cyclone track in the offshore compared to outside suggesting that divergence may be possible (Sarma et al., 2015). The existence of cold core eddy was observed at the study regions during pre-cyclone period and it was absent during post cyclone period due to divergence of waters close to coast. Waters were relatively cooler and saltier during pre-cyclone compared to post cyclone period due to influence of cold core eddy during former period (Fig. 2). The thermal structure along the transect showed upsloping of isotherms towards the coast indicating signatures of upwelling from a depth of 20-30 m during pre-cyclone period. In contrast, the water column in the upper 75 m was relatively fresher (~ 10) and warmer ($>1^{\circ}\text{C}$) during post-cyclone than pre-cyclone period due to convergence at the coast associated with deeper SSH. The upsloping of isolines during pre-cyclone period changed to a thick lens of low saline water in the upper surface along both transects (Fig. 2). The thickness of the fresh water lens decreased towards offshore.

Relatively higher concentrations of ammonium were observed during post-cyclone (1.5 to 3 μM) compared to pre-cyclone period (0.2-0.5 μM) (Fig. 3). Such high concentrations of ammonium in the entire coastal water column were attributed to the input through land-runoff associated with heavy precipitation. In addition to this ammonium was also added to the coastal water through rainwater and their concentration was as high as 300 to 500 μM (Sarma et al., 2018b). Relatively higher concentrations of nitrite (0.05 to 0.5 μM) and nitrate (3.4-6 μM) were observed during post-cyclone than pre-cyclone period (<0.15 μM , and <2 μM respectively; Fig. 3). Despite higher nutrients in the study region, the concentration of Chl-a (Fig. 2) and the column integrated primary production was low during post-cyclone than pre-cyclone period due to increase in turbidity which led to decrease in light penetration (Sarma et al., 2018b).

3.2. Variations in inorganic carbon components and fluxes of CO_2

The distribution of dissolved inorganic carbon (DIC) followed salinity in the entire water column. Higher DIC concentrations (2035-2138 μM) were observed with upsloping of high DIC contours to the surface during pre-cyclone period. In contrast, concentrations of DIC in the upper 20 m along transects A and B decreased to ~ 1650 and 1750 μM respectively compared to pre-cyclone period and it is consistent with the salinity distribution (Fig. 4). Relatively basic waters (higher pH by 0.07 - 0.15) were observed in the entire water column during post-cyclone compared to pre-cyclone period (Fig. 4). The magnitude of difference in pH between pre- and post-cyclone was higher in the upper 20m and decreased with depth. Relatively basic pH waters during post-cyclone are consistent with higher concentrations of ammonium suggesting that land-derived sewage might be the potential reason. Upsloping of isolines of total alkalinity was noticed during pre-cyclone with higher levels

(~2250 μM) than during post-cyclone period (1900 to 2000 μM) (Fig. 4). Land runoff brings significant amount of organic matter and terrestrial bacteria to the coastal Bay, however, decomposition rates may not be increased due to salinity shock on bacteria (both marine and terrestrial) resulted from sharp decrease in salinity. Hence, biological influence on inorganic carbon system may be rather minimal. Sewage also bring significant amount of dissolved inorganic carbon and total alkalinity to the coastal Bay as their concentrations were higher in sewage (~9350 μM and ~9800 μM respectively), however, its impact was masked by dilution by the precipitated water which is 4000 times less diluted than sea water. Hence, salinity (S) displayed strong linear relation with DIC ($40.41 \times S + 804.2$; $r^2=0.97$; $p<0.001$) and TA ($34.39 \times S + 1143$; $r^2=0.96$; $p<0.001$) and inverse relation with pH ($-0.022 \times S + 8.882$; $r^2=0.86$; $p<0.001$) suggesting that freshwater inputs have significant impact on inorganic carbon system in the study region. This study indicates that heavy precipitation associated with cyclone '*Hudhud*' brought significant changes in the inorganic carbon components and decreased concentrations of DIC, TA and increased pH in the coastal Bay compared to the pre-cyclone conditions.

The CO_2 in the surface water was oversaturated with respect to atmospheric equilibrium in the entire study region during pre-cyclone period (373-510 μatm), except at the offshore region (371 μatm) associated with low salinity (Fig. 4). These observations are consistent with earlier reports (Kumar et al., 1996; Sarma et al., 2012; 2015a). Sharp increase in pCO_2 was observed with depth following salinity. In contrast, the CO_2 concentrations were significantly undersaturated in the upper 30m of water column during post cyclone period along both transects A and B where pCO_2 levels were 150 to 200 μatm lower than that of atmospheric levels (Fig. 4). It was further noticed that pCO_2 levels at deeper depths were also lower during post-cyclone compared to pre-cyclone period due to convergence in the coastal region. Such low pCO_2 in the upper ocean could be due to increase in pH in response to increase in ammonium concentrations resulting in shifting of the inorganic carbon system towards formation of bicarbonate leading to decrease in pCO_2 . On the other hand, decrease in pCO_2 in the upper 30m could also be due to increased flux of CO_2 to the atmosphere due to heavy winds associated with cyclone '*Hudhud*'. The measured wind speed ranged from 50 to 100 km h^{-1} during cyclone. Such heavy winds increase piston velocity and thus, flux of CO_2 to the atmosphere. Bates et al. (1998) observed under-saturation of CO_2 at the Bermuda Atlantic Time-Series (BATS) location in the North Atlantic associated with hurricane and attributed the enhanced flux to high wind speeds. Enhanced CO_2 fluxes due to Hurricane Irene and Frances was observed in the Atlantic coast (Crosswell et al., 2014; Huang and Imberger, 2010). In addition to this, enhanced biological

production due to increased nutrient levels may contribute to understaturation of CO₂. Sarma et al. (2015b) noticed that the depth integrated phytoplankton biomass and primary production to the photic zone did not increase in the study region after cyclone compared to those before the cyclone and attributed it to inhibition of light due to increased turbidity. Therefore, decrease in pCO₂ may not be due to biological production.

In order to examine the dominant controlling process on pCO₂ variability during post-cyclone period, the contribution of pCO₂ change due to flux and salinity dilution was computed. Salinity showed linear relation with pCO₂ in the study region (Fig.5). The regression analysis suggested that decrease in salinity by one unit reduced the pCO₂ levels by 22.4 µatm. Compared to pre-cyclone conditions, salinity decreased by 8 units after cyclone, therefore, it contributes to decrease in pCO₂ by 179 µatm due to dilution compared to pre-cyclone period. Based on pCO₂ level during pre-cyclone period of ~471 µatm (average pCO₂ during September 2014), mean mixed layer depth during September 2014 (~12 m) and mean wind speed of 8.3 km h⁻¹, the flux of CO₂ to the atmosphere between 25 September 2014 and 11th October 2014 (precyclone period observation to one day before cyclone ‘Hudhud’) was computed to be 40 µatm. Therefore, the resultant pCO₂, after removing the effects of dilution and removal to the atmosphere, would be expected to 252 µatm (471-(179+40) µatm), which is slightly lower than that of the measured pCO₂ of 274 µatm. At the same time, this study suggests that decrease in pCO₂ after cyclone was contributed by dilution with precipitated water (80%) and that the impact of sea-to-air flux was relatively small due to strong stratification that inhibits mixing of pCO₂ rich subsurface water with surface. The sea-to-air flux of CO₂ was 1.3±0.5 mmolC m⁻² d⁻¹ during pre-cyclone period and it turned negative (-1.8±0.9 and -2.2±1 mmolC m⁻² d⁻¹ along transect A and B respectively) after cyclone. This study suggested that freshwater input to the coastal Bay of Bengal due to heavy rainfall derived by cyclone and land runoff enhanced concentrations of ammonium and increased pH between 0.07 and 0.15 units that led to undersaturation of CO₂. As a result, this region, which was previously a significant source of CO₂ to the atmosphere, turned into a sink after cyclone ‘Hudhud’.

3.3. Variations in concentrations of nitrous oxide (N₂O) and its fluxes

The concentrations of N₂O ranged between 6.0 and 7.7 nM during pre-cyclone period and its saturation varied from 113 to 138% at surface (Fig. 6). The concentrations of N₂O increased with depth and its saturation reached to >300% at a depth of 75m. Like pCO₂, the concentrations of N₂O decreased by >2 nM in the upper 30 m of water column during post- cyclone period along both transects compared to pre-cyclone period. Relatively lower concentrations of N₂O (by about 1 nM)

were observed along transect B than transect A associating with higher ammonium concentrations along the former than latter transect (Fig. 3). The concentration of N₂O below 40 m was relatively higher during post-cyclone than pre-cyclone period and it was associated with higher salinity. Salinity showed linear relation with N₂O concentrations ($r^2=0.76$; $p<0.001$; Fig. 5) suggesting that salinity controlled their concentrations in the coastal Bay. There are two possible mechanisms responsible for production of N₂O in the marine waters and they are nitrification and denitrification. Since the coastal waters are well ventilated and oxygen concentrations are high (>200 μ M at surface and ~50 μ M at 75 m; Sarma et al., 2015b), denitrification may not be triggered (Rao et al., 1994; Sarma et al., 2013). Hence nitrification is solely responsible for N₂O production in the study region. In our study region, sharp decrease in salinity in the coastal waters associated with heavy precipitation, decreases activity of bacteria due to salinity shock. Fernandes et al (2008) noticed that freshwater bacteria received by various rivers were inactive in the Bay of Bengal due to salinity shock. del Giorgio and Bouvier (2002) observed decrease in growth and possible loss of activity of bacteria in the fresh to salt water transition. This would result in decrease in nitrification rates and production of N₂O and it is consistent with the decrease in nitrification at low salinity (Rysgaard et al., 1999; DeBie et al., 2002; Megalhaes et al., 2005; Bange et al., 2006; Aslan and Simsek, 2012; Rao and Sarma, 2013). In addition to this, lower N₂O concentrations were observed in the sewage waters (1 to 2.5 nM) due to possible occurrence of denitrification as near anoxic conditions exists in the sewage. Therefore, decrease in salinity due to heavy precipitation and land runoff associated with cyclone might have decreased the nitrification rates and diluted with low N₂O waters that resulted in lower concentrations of N₂O in the coastal Bay of Bengal.

The entire water column was supersaturated with N₂O (>120%) during pre-cyclone period in the upper 50 m suggesting that the study region acted as a strong source of N₂O to the atmosphere (Fig. 6). In contrast, undersaturation of N₂O was noticed during post-cyclone period in the upper 30 m suggesting that this region became a sink for the atmospheric N₂O. On the other hand, waters below 40 m were relatively more saturated after the cyclone than pre-cyclone period suggesting that a greater production of N₂O may be linked to higher ammonium concentrations observed in the study region during post cyclone period. Though the concentration of ammonium increased in the entire water column due to land runoff associated with the cyclone, low salinity in the upper ocean did not favour intense nitrification due to decrease in bacterial activity. In addition to this, the concentrations of N₂O in the sewage was under-saturated (75-96%) due to denitrification resulted from formation of anoxic conditions. Hence, sewage did not contribute significant amount of N₂O to

the coastal Bay. Therefore, the variations in N₂O concentrations and saturation between pre- and post-cyclone periods were contributed by decrease in salinity and also rapid removal of N₂O to the atmosphere associated with heavy winds.

In order to examine the influence of salinity and winds, the contribution of these two processes is evaluated. Based on the relationship between salinity and N₂O concentration, it was observed that 0.18 nM of N₂O decreased with decrease in per unit of salinity (Fig. 5). About 8 units of salinity decreased after cyclone compared pre-cyclone conditions and would lead to decrease in N₂O concentrations by ~1.4 nM due to lowering of salinity. The flux of N₂O to the atmosphere was computed using pre-cyclone N₂O concentrations and mean wind speed of 8.3 km h⁻¹ during before cyclone. It was observed that sea-to-air flux removed 0.9±0.4 nM of N₂O to the atmosphere from the surface waters. The mean concentration of N₂O in the surface water during pre-cyclone and post-cyclone was 7.1±0.6 nM and 5.3±0.7 nM respectively suggesting that ~2.0 nM of N₂O was decreased during latter period. It is suggested that about 80% of decrease in N₂O after cyclone was contributed by freshwater dilution and associated with decrease in nitrification. The flux of N₂O at air-water interface suggests that the study region acted as a significant source of N₂O to the atmosphere during pre-cyclone period (0.9±0.3 μmol m⁻² d⁻¹), but as a sink (-0.2±0.1 and -0.6±0.2 μmol m⁻² d⁻¹ along transect A and B respectively) after cyclone due to decrease in salinity and nitrification rates.

3.4. Variations in concentrations of methane (CH₄) and its fluxes

The concentration of CH₄ was ranged between 1.8 and 6.1 nM during pre-cyclone period and its saturation varied from 114 to 532% in the water column (Fig. 6). The concentration of CH₄ increased with depth with maximum at the mid waters (~30 m) during both pre- and post cyclone periods associated with sharp salinity gradient. This subsurface maximum is more prominent during post cyclone than pre cyclone period. The concentration of CH₄ was significantly higher during post cyclone associated with relatively low salinity (Fig. 2). Unlike pCO₂ and N₂O, the concentration of CH₄ increased in the water column after cyclone (by 0.5 to 2.8 nM) compared to the pre-cyclone period (Fig. 6). Relatively higher concentration (by about 2 nM) was observed along transect B than transect A associated with higher ammonium concentrations. The concentration below 40 m was relatively higher during post-cyclone than pre-cyclone period and was associated with lower salinity. The concentration of CH₄ in sewage was an order of magnitude higher than in the coastal waters (~500 nM). The salinity to CH₄ relationship (Fig. 5) shows higher variability in concentrations of CH₄ at lower salinity (<25) suggesting that the land drainage could be a potential source of CH₄ to

the coastal Bay of Bengal. The higher concentration of CH₄ at deeper depths could be due to diffusion of CH₄ from the benthic boundary layer. This gas, however, did not diffuse to the surface due to strong vertical stratification leading to accumulation in the subsurface layer resulting in higher concentrations (Fig. 2).

The water column was above saturation in CH₄ with reference to that at surface in equilibrium with the atmospheric CH₄ during pre-cyclone period (114-130%) and it was increased to 140-200% after cyclone (Fig. 6). The super-saturation of CH₄ increased with depth (up to 700%) suggesting that sediments may be a strong source of CH₄ as production of CH₄ in the sediments is reported (Araujo et al., 2018) and its further diffusion to the surface waters is possibly hindered by strong stratification. The saturation of CH₄ in the water column was relatively higher along transect A than B as the former transect is close to the Urban region of Visakhapatnam city while the latter is close to rural region. Hence, more sewage associated with the land-fall is expected at transect A than B resulting in higher concentration of CH₄ in the former than latter. It was further confirmed by higher concentrations of ammonium as observed along the transect A than B (Fig. 3). It would be difficult to understand the source of CH₄ (*in situ* production or sewage) due to lack of isotopic data in this study. The flux of CH₄ at air-water interface suggests that the study region acts as a mild source of CH₄ to the atmosphere during pre-cyclone period ($0.6 \pm 0.3 \mu\text{mol m}^{-2} \text{ d}^{-1}$) that turned into a significant source (1.9 ± 1 and $1.3 \pm 0.8 \mu\text{mol m}^{-2} \text{ d}^{-1}$ along transect A and B respectively) after the cyclone 'Hudhud' (Table 1).

3.5. Variations in concentrations of DMS and DMSP_t and fluxes

During pre-cyclone period the concentrations of DMS and DMSP_t were in the ranges of 8.8 to 33.5 nM and 2.0 to 28.3 nM respectively (Fig. 7) and decreased significantly after cyclone 0.4 to 3.9 nM (DMS) and 0.2 to 6.0 nM (DMSP_t) respectively. Both DMS and DMSP are produced through biological processes like microbial activity such as bacterial conversion of DMSP to DMS, and microzooplankton grazing of particulate DMSP. The lower concentrations of DMS and DMSP after cyclone is mainly caused by decrease in phytoplankton biomass as evidenced from chl-a (Fig. 2). Furthermore decrease in DMS and DMSP_t during post cyclone period could also due to dilution of coastal water with fresh water from the land-drainage which contain no DMS/DMSP. The strong linear relationships of DMS and DMSP_t with salinity suggest the role of dilution of freshwater in their distribution in coastal waters of the Bay of Bengal (Figure. 8). The concentrations of DMS and DMSP_t were relatively lower along transect A compared to transect B and it is consistent with the concentrations of Chl-a (Fig. 2) suggesting that higher phytoplankton biomass is responsible for

increased production of DMS along the latter transect. Though Chl-a concentrations were not significantly higher during pre-cyclone compared to post cyclone period, lower DMS and DMSP during post cyclone may be by dilution with land drainage (Fig. 2). The production of DMSP within the plankton's cell and subsequent release of DMS would depend on a variety of factors such as phytoplankton species composition (Liss et al., 1997), physico-chemical parameters (salinity and temperature), microbial community (Taylor and Gilchrist, 1991) and DMSP lyase activity (Stienke et al., 2002). Phytoplankton composition was dominated by diatoms during pre-cyclone period whereas picoplankton (cyanobacteria) was abundant during post-cyclone period. The production of DMS by phytoplankton is species dependent (Turner et al., 1988). For instance, the high DMS concentrations were associated with high abundance of DMSP producing phytoplankton groups (prymesiophytes and dinoflagellates) over diatoms and cyanobacteria as the latter have relatively low intracellular DMSP concentrations compared to the former. Shifts in plankton speciation were also noticed in association with modifications in the concentrations and ratios of nutrients (Lancelot et al., 1987; Mee, 1992; VandenBerg et al., 1996). Significant modification in concentrations and ratio of nutrients were observed between pre- and post cyclone period (Table 1; Sarma et al., 2018b). In addition to this, microzooplankton grazing of particulate DMSP and bacterial conversion to DMS is also responsible for variations in concentrations of DMS and $DMSP_t$ (Jones et al., 1998; Stefels et al., 2007). We have not measured zooplankton grazing during this study; hence are not able to confirm the possible mechanism responsible for lower DMS and $DMSP_t$ after the cyclone '*Hudhud*'. In addition to this, lower concentration of DMS and $DMSP_t$ after cyclone *Hudhud* could also due to higher suspended particulate matter and low Chl-a concentrations as the former decreased the light availability resulting in low production of DMS and $DMSP_t$. Therefore, dilution of low DMS/DMSP waters from the precipitation, decrease in production due to increased suspended load and also variations in composition of phytoplankton might have contributed lower DMS/DMSP concentrations during post cyclone compared to pre-cyclone conditions.

The flux of DMS at air-water interface suggests that the study region which acts as a strong source of DMS to the atmosphere during pre-cyclone period ($5.8 \pm 3 \mu\text{mol m}^{-2} \text{d}^{-1}$), turned in to being a mild source (1.2 ± 0.5 and $3.8 \pm 2 \mu\text{mol m}^{-2} \text{d}^{-1}$ along transect A and B respectively) after the cyclone '*Hudhud*'.

4. Summary and Conclusions

In order to examine the influence of the very severe cyclonic storm '*Hudhud*' on the concentrations and fluxes of trace gases such as CO_2 , N_2O , CH_4 and DMS to the atmosphere, a study was conducted

in the coastal Bay of Bengal, and compared them with the pre-cyclone period measurements. The study region received nutrients through upwelling during pre-cyclone period and the same was suppressed due to cyclone 'Hudhud'. High precipitation associated with cyclone brought land-derived nutrients such as ammonium that increased pH of coastal waters. Contrarily, the inorganic carbon components (DIC, TA and pCO₂) were significantly low during post-cyclone compared to their measurements during pre-cyclone period. The concentrations of N₂O also decreased during post-cyclone compared to pre-cyclone period and this was attributed to the decrease in nitrification due to low salinity and lower concentrations of N₂O in the sewage waters. The relation of pCO₂ and N₂O with salinity suggested that about 80% of the decrease is attributable to dilution with rain water and the remaining to enhanced gas fluxes to atmosphere by cyclonic winds. In contrast, CH₄ concentrations were significantly higher during post-cyclone than pre-cyclone due to input of anthropogenic CH₄ through land runoff. The DMS and DMSP_t concentrations decreased during post-cyclone in comparison to pre-cyclone period and it is consistent with low phytoplankton biomass during former than latter. Due to lack of isotopic data, the process responsible for higher concentrations of CH₄ could not be unequivocally explained in this study. This study suggested that very severe cyclonic storm 'Hudhud' changed the magnitude of trace gases emissions from significant source to sink, except CH₄, in the coastal Bay of Bengal contrary to the existing belief.

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Table 1: The mean physico-chemical characteristics of coastal waters before and after cyclone Hudhud.

Parameter	September 2014	October 2014	
	Off Visakhapatnam	Off Visakhapatnam	Off Atchutapuram
SST (°C)	28.85±0.6	30.02±0.1	30.06±0.1
SSS	31.77±2.0	21.22±1.6	23.29±1.4
Chl-a (mg m ⁻³)	0.52±0.1	0.39±0.2	0.18±0.1
Turbidity (mg l ⁻¹)	6.2±1	10.2±4	18.4±4
NO ₃ (μM)	1.8±1	0.79±0.3	4.3±2
NH ₄ (μM)	0.2±0.1	1.7±0.7	0.9±1
PO ₄ (μM)	2.0±0.5	2.3±0.6	2.2±0.4
SiO ₄ (μM)	8.1±2	20.9±2	18.0±2
N:P	3.9±1.0	1.2±0.3	1.4±0.2
Si:N	2.4±0.9	9.3±1.0	4.1±0.8

Legend to figures

1. Locations of the stations where sampling was conducted in the coastal Bay of Bengal. The letter A and B denotes transect A (Red fill circle) and B (blue star). The track of cyclone Hudhud was shown in the inset.
2. Water column temperature ($^{\circ}\text{C}$), salinity, and Chlorophyll-a (mg m^{-3}) in the coastal Bay of Bengal during pre-cyclone (September 2014 in the upper panel), along transect A (middle panel) and transect B (lower panel) during post-cyclone period.
3. Concentrations of ammonium (μM), nitrite (μM), and nitrate (μM) in the coastal Bay of Bengal during pre-cyclone (upper panel), along transect A (middle panel) and transect B (lower panel) during post-cyclone period.
4. Distribution of dissolved inorganic carbon (DIC; μM), pH, total alkalinity (TA; μM), and pCO_2 (μatm) during pre-cyclone (upper panel), along transect A (middle panel) and transect B (lower panel) during post-cyclone period.
5. Relation of pCO_2 , N_2O and CH_4 with salinity during both pre (blue) and post cyclone period (Transect A- Red closed circles and Transect B- open circles). The horizontal line (pink) denotes atmospheric value.
6. Distribution of concentration of N_2O (nM), its saturation (%), CH_4 (nM) and its saturation (%) during pre-cyclone (upper panel), along transect A (middle panel) and transect B (lower panel) during post-cyclone period.
7. Distribution of DMS (nM), and DMSP (nM) during pre-cyclone (upper panel), along transect A (middle panel) and transect B (lower panel) during post-cyclone period.
8. Relationships of DMS and DMSP with salinity during pre (black) and post-cyclone periods (Transect A- Red Triangle and Transect B-Blue circles).
9. " Relationships of DMS and DMSPt with salinity during pre- (black) and post-cyclone periods (Transect A- Red and Transect B-Blue)."

Figure 1

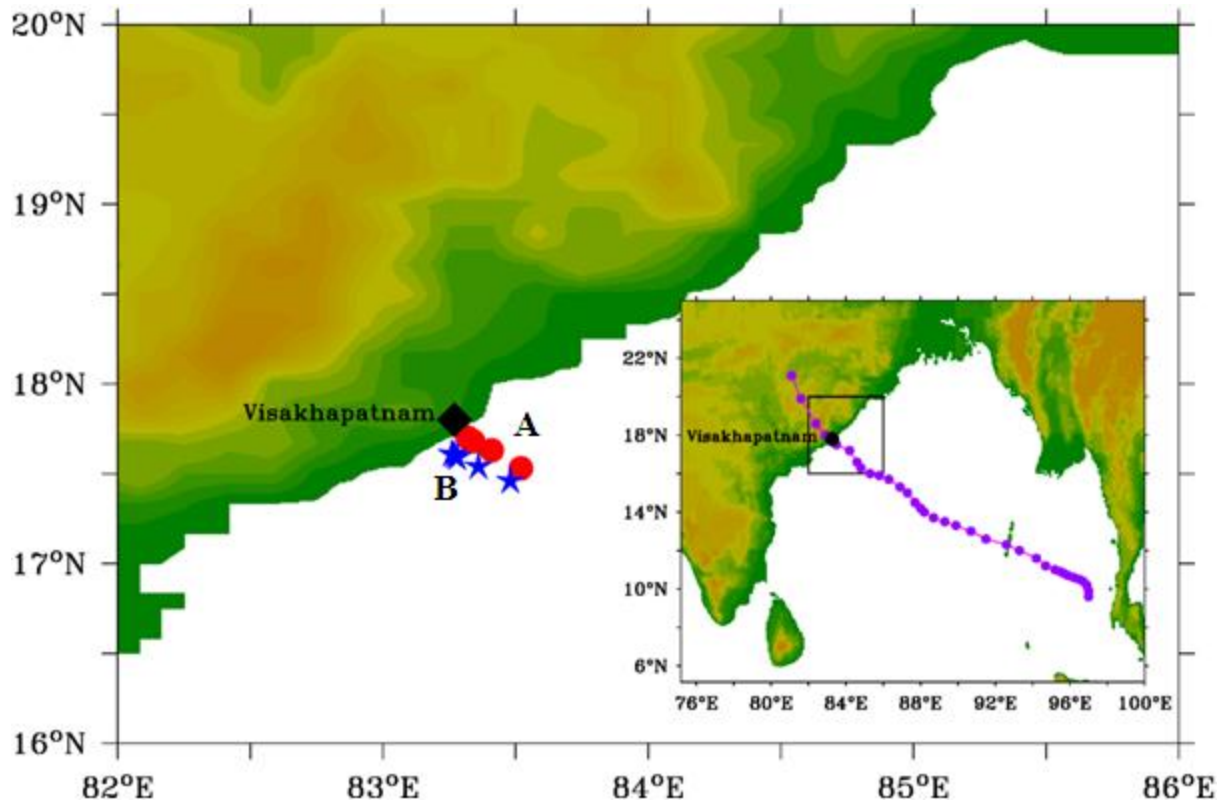


Figure 2

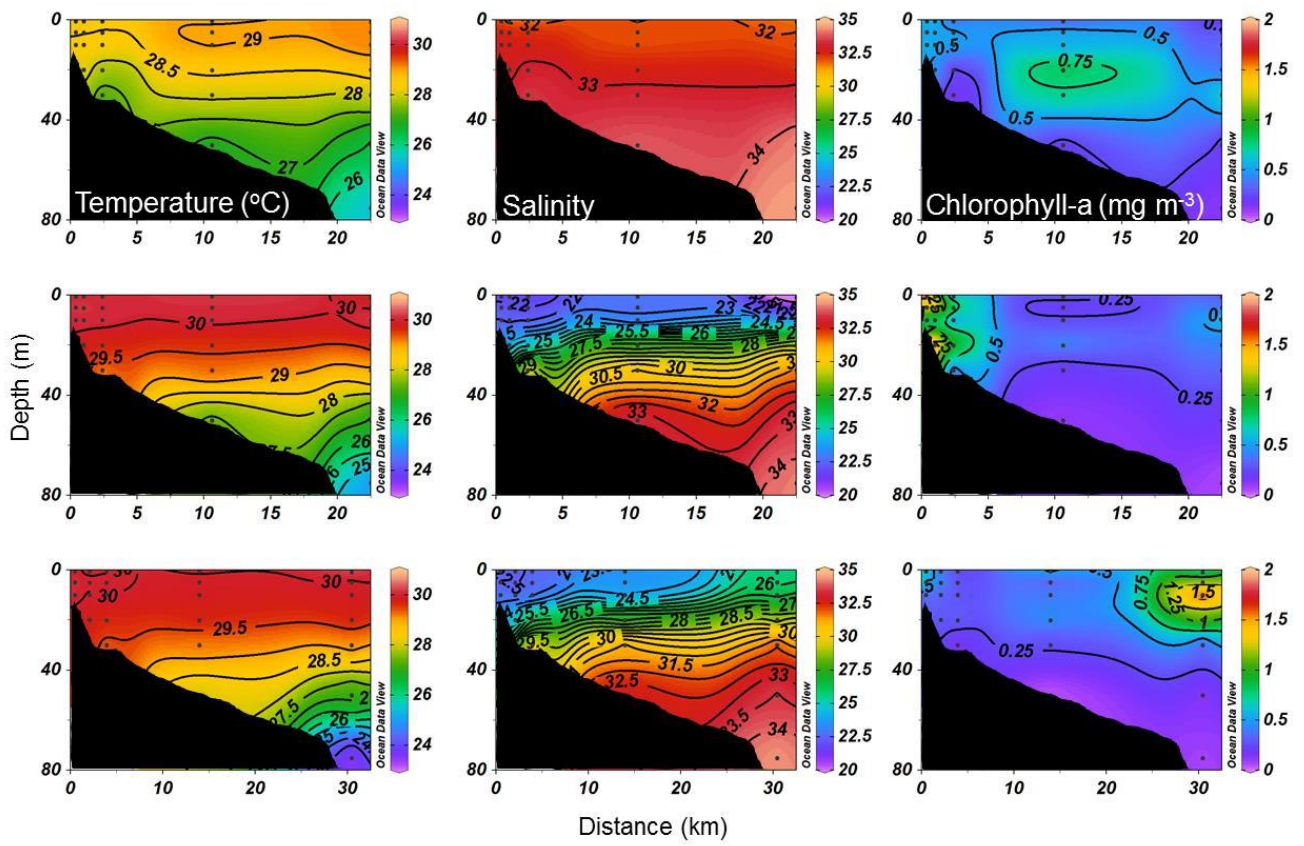


Figure 3

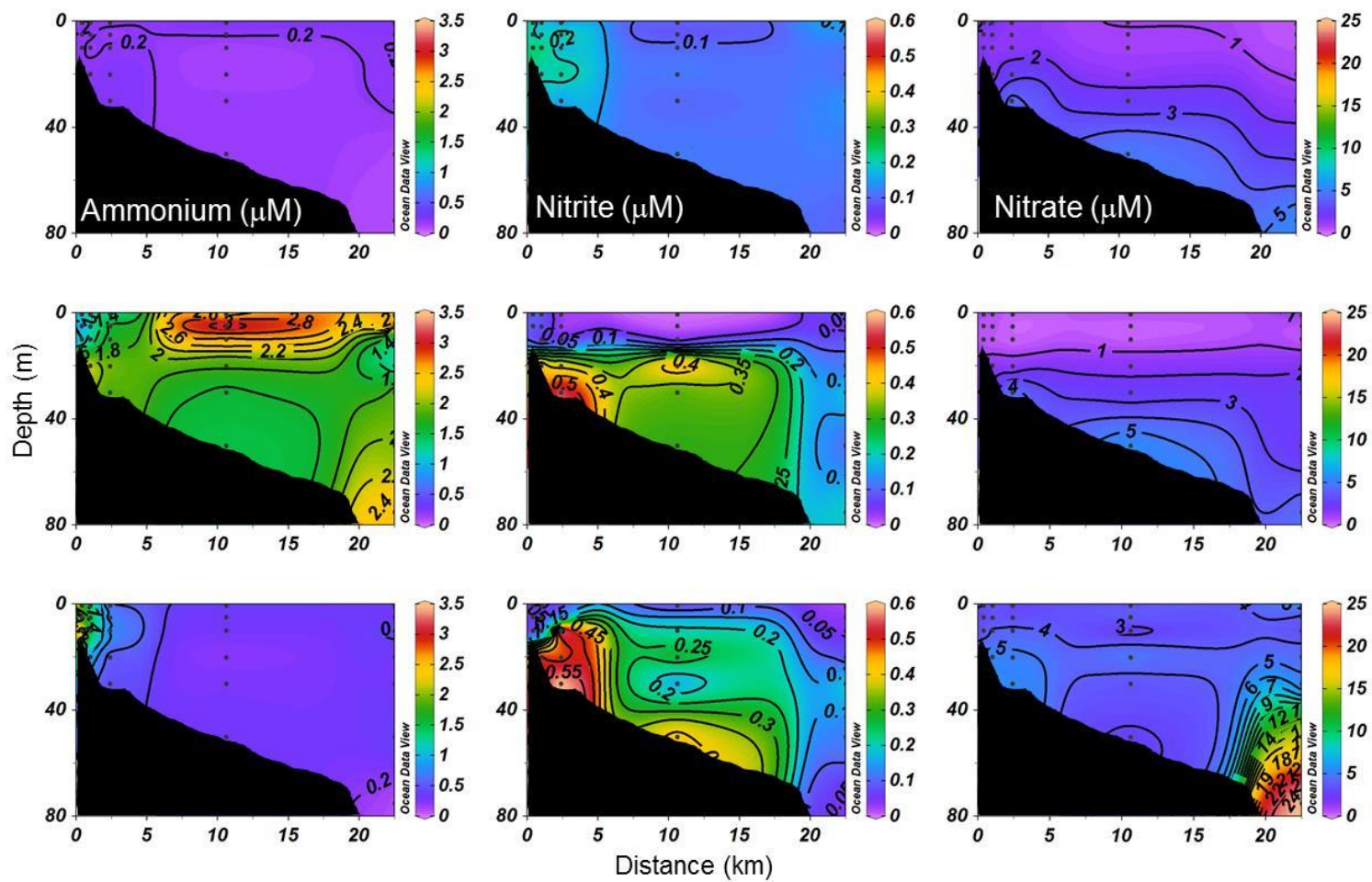


Figure 4

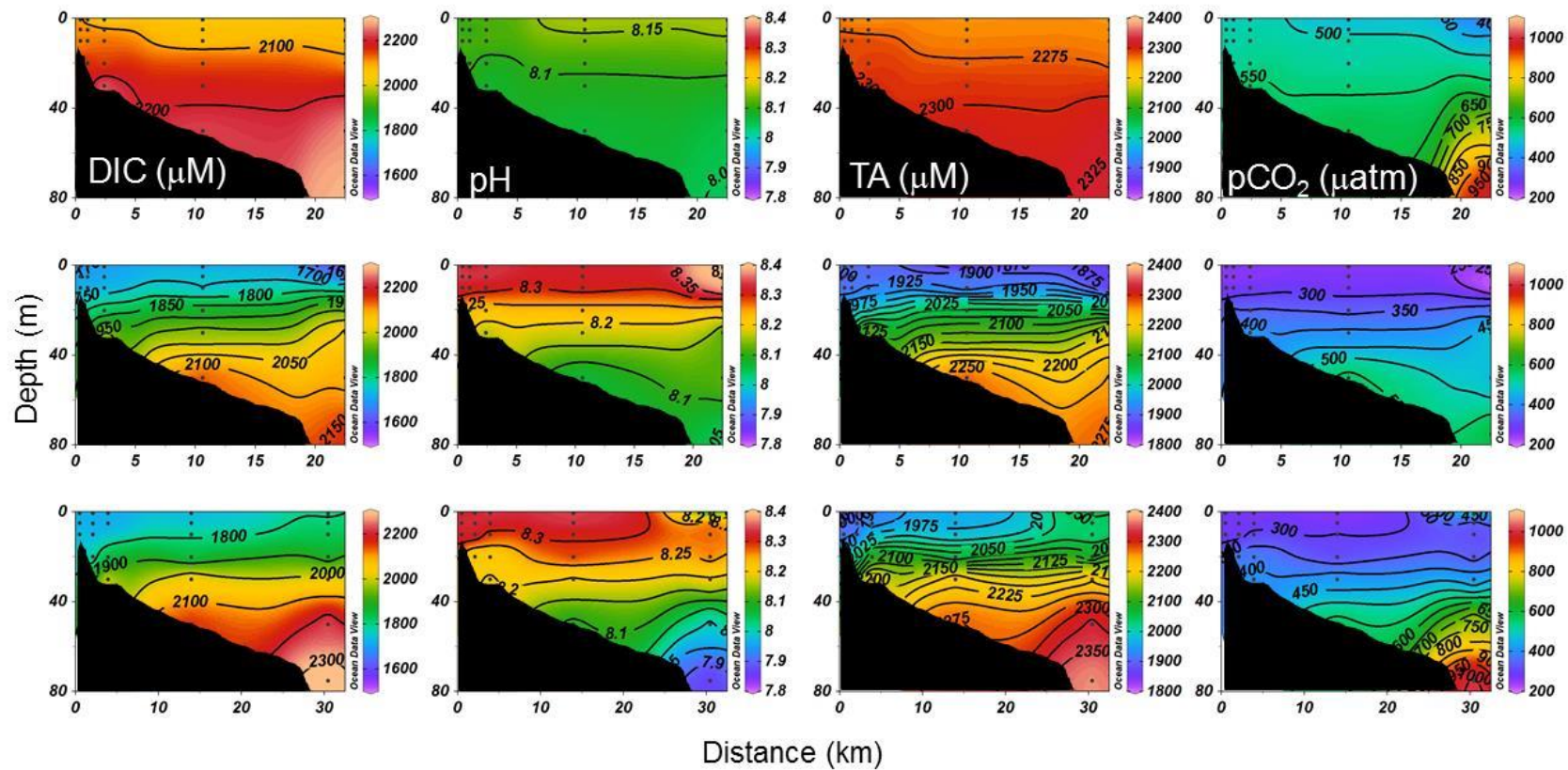


Figure 5

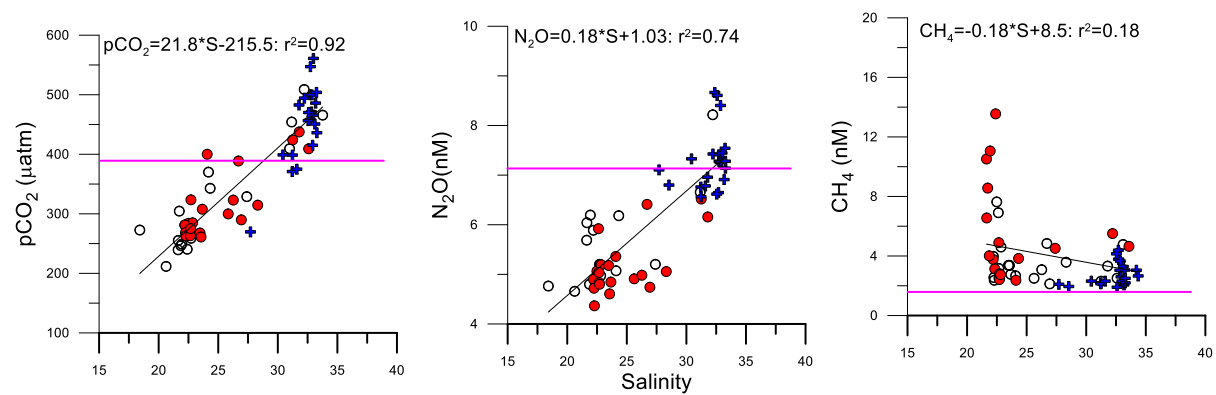


Figure 6.

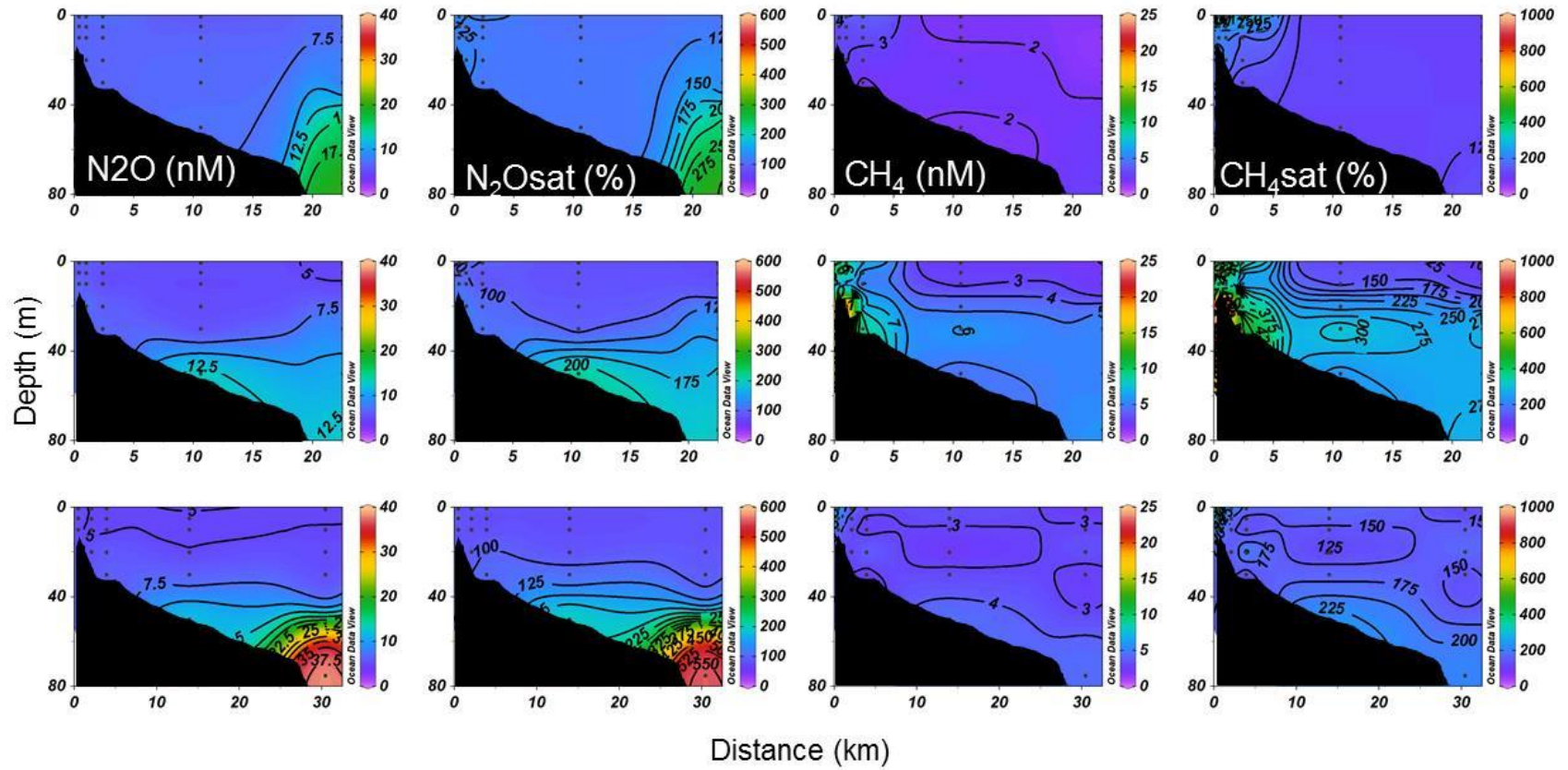


Figure 7:

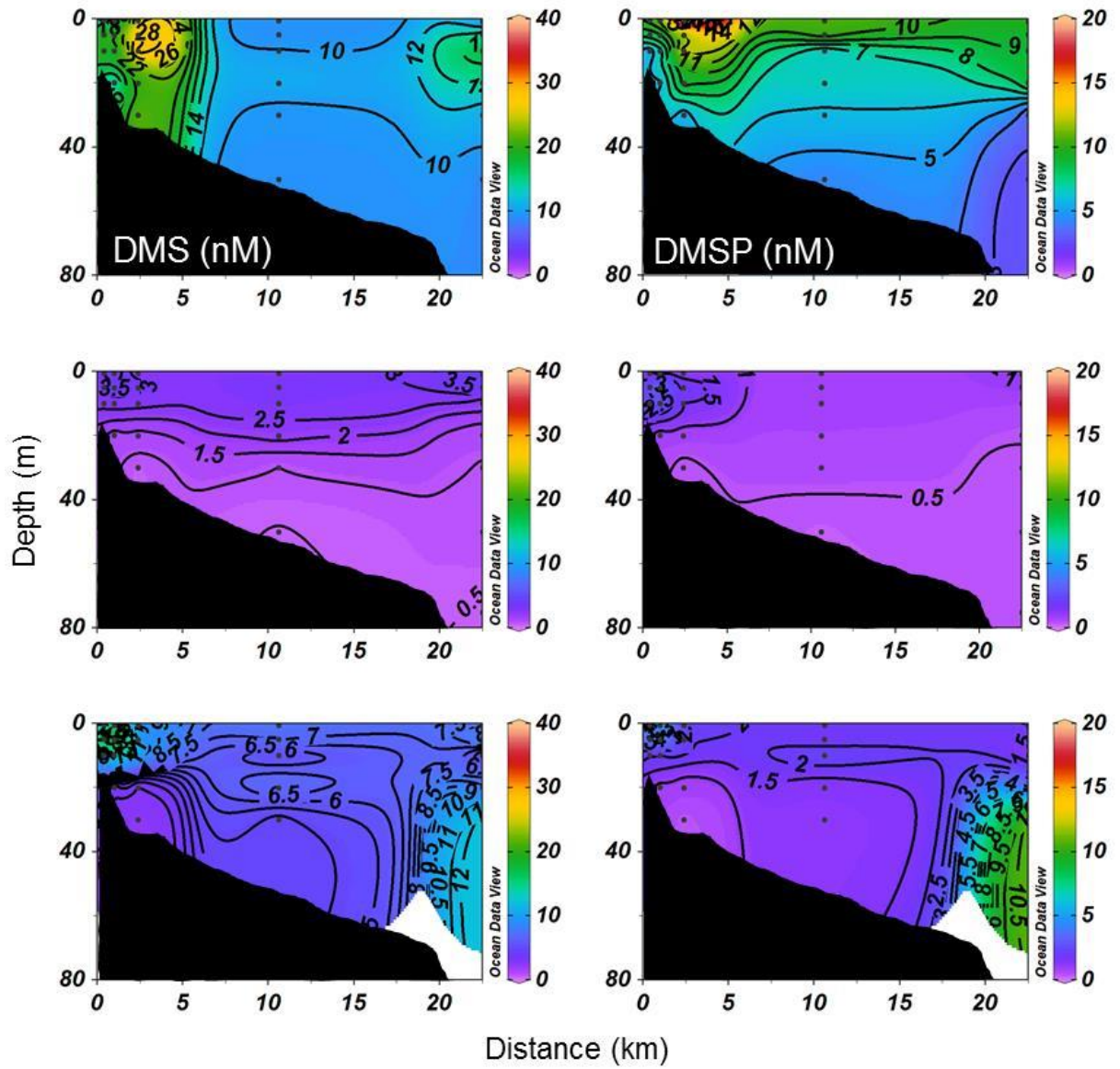


Figure 8:

