



Measurement of ambient NH₃ over Bay of Bengal during W_ICARB Campaign

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Abstract. Concentrations of ambient NH₃, NO, NO₂ and SO₂ were measured over Bay of Bengal (BoB) during 28 December 2008 to 25 January 2009 to study their diurnal variation and relationship of NH₃ with other trace gases over BoB. The measurements were done under the winter phase of Integrated Campaign on Aerosols and Radiation Budget (W_ICARB). For the first time, ambient NH₃ was monitored precisely over BoB based on chemiluminescence method, having estimation efficiency more precise than the chemical trap method. The average concentration of ambient NH₃, NO, NO₂ and SO₂ were recorded as 4.78 ± 1.68 , 1.89 ± 1.26 , 0.31 ± 0.14 and $0.80 \pm 0.30 \mu\text{g m}^{-3}$, respectively, over BoB. The prominent latitudinal and longitudinal variations of the trace gases were observed over BoB, whereas NH₃ and NO showed the non-significant diurnal variation. Results reveal that the concentration of ambient NH₃ negatively correlated with ambient NO₂ ($r^2 = -0.56$), SO₂ ($r^2 = -0.58$) and ambient temperature ($r^2 = -0.27$) during the study.

Keywords. Atmospheric composition and structure (General or miscellaneous)

1 Introduction

In the atmosphere and ocean, NH₃ and its ionized form NH₄⁺ are ubiquitous. Naturally and anthropogenically produced NH_x (NH₃ + NH₄⁺) are transported through the atmosphere and generally their concentrations in air decrease as the distance from land increases. It has been suggested that in pre-industrial times, the oceans were probably a net source of NH_x of the continents (Duce et al., 1991), but this is not the case today (Sutton et al., 1995, 2000). NH_x is produced in surface water by the biological reduction of nitrate (ei-

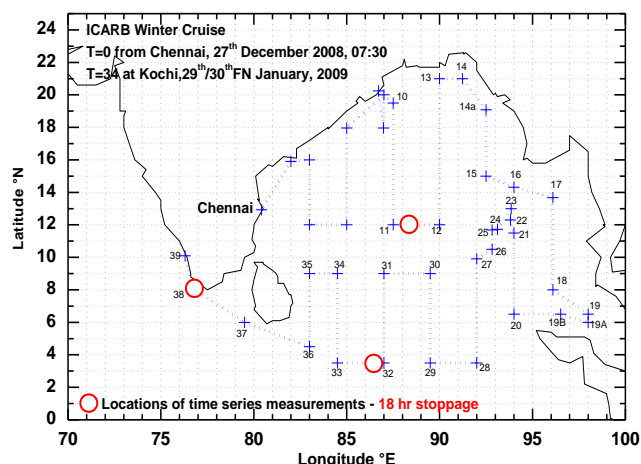
ther directly or via the degradation of biologically synthesized organic nitrogenous material/agricultural run-off). In a solution, NH_x is partitioned between NH₄⁺ and NH₃ according to equilibrium thermodynamics: the proportion of NH_x that occurs as NH₃ (depending on pH, temperature and ionic strength of the medium) is available for emission to the atmosphere (Aneja et al., 2001). NH₃ is also emitted to the atmosphere by plants, animals and its environments, by soil micro-organisms and by various industrial and agricultural processes, including the direct volatilization of solid NH₄NO₃ salts and fertilizers (Sutton et al., 2000; Li et al., 2006; Sharma et al., 2010a, b). There is also evidence of volcanic source of NH_x to the atmosphere (Uematsu et al., 2004) and of substantial NH₃ emissions from seabird colonies (Blackall et al., 2007; Theobald et al., 2006)

Norman and Leck have reported NH₃ of the order of 0.05–0.2 nmol m⁻³ (0.0085–0.0034 μg m⁻³), whereas, Gibb and Mantoura (1999) and Gibb et al. (1999) have reported 10–20 nmol m⁻³ (0.17–0.34 μg m⁻³) in the central Indian Ocean and 2.5–5.6 nmol m⁻³ (0.043–0.095 μg m⁻³) over coastal Arabian Sea and 0.4–1.8 nmol m⁻³ (0.007–0.031 μg m⁻³) over remote Arabian Sea. Schafer et al. (1993) have reported the concentration of NH₃ gas over BoB of the order of 14.29–29.29 nmol m⁻³ (0.243–0.500 μg m⁻³). Johnson et al. (2008a, b) has reviewed the measurement of ambient NH₃ gases along with sea water NH_x (NH₃ + NH₄⁺) over Pacific, Atlantic and Indian Ocean. Table 1 summarizes the results on concentrations of ambient NH₃ over various oceans reported by different researchers.

Ambient NH₃ plays an important role not only in the formation of secondary aerosols while combining with atmospheric acid gases (sulfuric acid, nitric acid and hydrochloric acid) but also contributes to adverse health effects, mainly respiratory diseases (Warneck, 1988) and climate change.

Table 1. Comparison of ambient NH₃ (nmol m⁻³) over various locations.

Location	Maximum	Minimum	Average	Reference
Southern Ocean	6.0	0.6	3.5	Ayers and Gras (1980)
Central Atlantic Ocean	30.0	1.0	–	Zhuang and Huebert (1996)
Central Atlantic Ocean	18.4	3.7	9.7	Norman and Leck (2005)
Central Pacific Ocean	3.4	0.01	0.67	Quinn et al. (1990)
Southern Atlantic Ocean	7.7	0.1	2.1	Norman and Leck (2005)
Southern Indian Ocean	4.4	2.2	–	Ayers and Gras (1980)
Southern Indian Ocean	2.1	0.3	1.1	Norman and Leck (2005)
Central Indian Ocean	0.2	0.05	0.1	Norman and Leck (2005)
Central Indian Ocean (coastal)	5.6	2.5	3.8	Gibb and Mantoura (1999)
Central Indian Ocean (remote)	1.8	0.4	1.0	Gibb et al. (1999)
Bermuda	20			Lebel et al. (1985)
Atlantic (transect S-N)	7.6	1.3	4.3	Johnson et al. (2008b)
Atlantic (transect N-S)	1.0	0.02	0.2	Johnson et al. (2008a)
Atlantic transect	3.2	0.07	1.9	McKee (2001)
Berhampur (coastal)	–	–	329.4	Carmichael et al. (2003)
Bhunbaneswar (coastal)	–	–	288.2	Carmichael et al. (2003)
Bay of Bengal (coastal)	211.8	117.6	158.8	Khemani et al. (1987)
Bay of Bengal (coastal)	675.0	105.2	265.2	Biswas et al. (2005)
Bay of Bengal	441.2	11.7	281.2	Present study

**Fig. 1.** Scheduled track of *Sagar Kanya* SK-254 cruise for measurement of trace gases over BoB.

Sulfuric acid (H₂SO₄) and nitric acid (HNO₃) are the major acid gases in the atmosphere that occur from oxidation of SO₂ and NO_x, respectively. These acid gases are neutralized by NH₃ in the atmosphere, thereby forming NH₄HSO₄ and (NH₄)₂SO₄ and NH₄NO₃ aerosols, respectively.

This study is the first time ambient NH₃ has been monitored precisely over BoB based on chemiluminescence method along with the other trace gases NO, NO₂ and SO₂ onboard *Sagar Kanya* (a research vessel). The meteorological parameters (temperature, sea surface temperature (SST), relative humidity (RH), wind direction and wind speed) were

also recorded over BoB during the campaign to correlate with trace gases. The main objective of this campaign was to study the diurnal variation of ambient NH₃ and other trace gases over BoB and its interaction.

2 Experimental setup

2.1 Cruise track of W_ICARB

Measurement of trace gases, i.e. NH₃, NO, NO₂, and SO₂, were made over BoB under the W_ICARB campaign from 28 December 2008 to 25 January 2009 onboard *Sagar Kanya* (SK-254) cruise (a research vessel) as per its scheduled track given in Fig. 1. The cruise tracks covered the region ranging from 21° N to 3.5° N latitude and 76.3° E to 98° E longitudes over BoB. The cruise embarked on 27 December 2008 from Chennai and ended at Kochi (9.96° N, 76.3° E), India, on 31 January 2009. The ship halt times are also indicated in the figure by open circles. The cruise stationed at 3 places for 18 h to take time series/diurnal variation measurements on 5, 8 and 23 January 2009. Sampling inlets of all analyzers were connected to the sampling system, which was placed in the opposite direction of the cruise ship plume to reduce the self-contamination; the portable weather station was also mounted at same height (about 11 m from sea surface) and same location on the deck of the ship. These instruments operated continuously for the entire period of the campaign (28 December 2008 to 25 January 2009).

Table 2. Average concentration of NH₃, NO, NO₂ and SO₂ (μg m⁻³) over BoB.

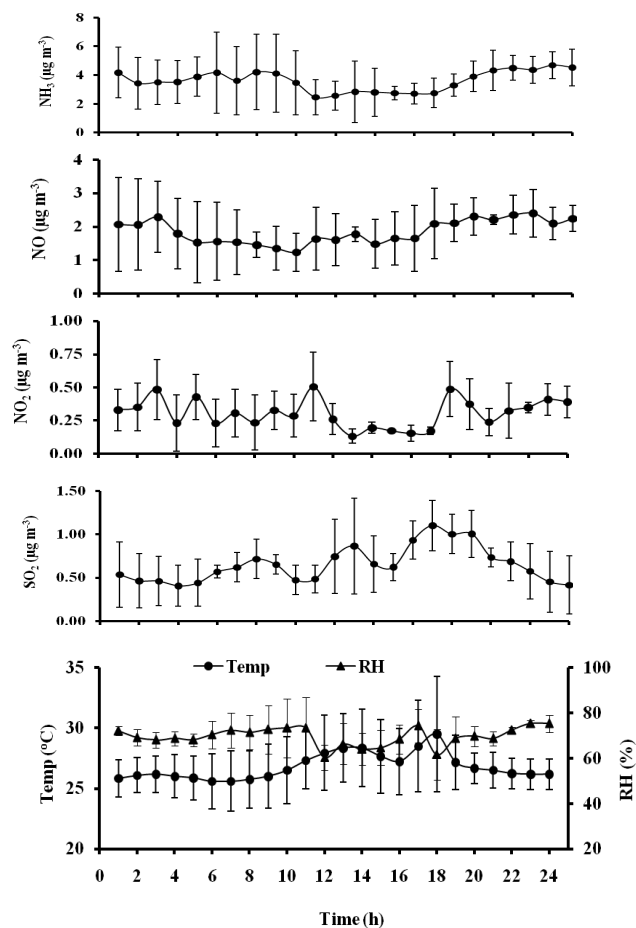
	Concentration			
	Average	Day	Night	D/N
NH ₃	4.78 ± 1.68	4.85 ± 1.91	4.70 ± 1.44	1.03
NO	1.89 ± 1.26	1.87 ± 1.24	1.90 ± 1.27	0.98
NO ₂	0.31 ± 0.14	0.27 ± 0.12	0.34 ± 0.16	0.79
SO ₂	0.80 ± 0.30	0.86 ± 0.33	0.75 ± 0.27	1.15

± Standard deviation

2.2 Measurement of trace gases

Concentration of ambient NH₃ was measured continuously using an NH₃-analyzer (model: CLD88CYp, M/s. ECO Physics AG, Switzerland) operating on chemiluminescence method (having estimation efficiency >90 % compared to chemical trap method having reproducibility of 4.7 %). In this analyzer, two catalytic converters of different characteristics allow sequential detection of NO_x and NO_x-amines by converting them into NO at 375 °C and 650 °C, respectively. Concentration of ambient NH₃ was calculated from the difference between NO_x and NO_x-amine (NH₃ = NO_x-amine - NO_x). The measurement range of NH₃ analyzer varied from 0–5 ppb to 0–5000 ppb (accuracy ±0.050 ppb of all the ranges). NO and NO₂ were measured continuously using an NO_x-analyzer (model: CLD88p, M/s. ECO Physics AG, Switzerland) with photo catalytic converter (model: PLC860, M/s. ECO Physics AG, Switzerland) also based on chemiluminescence method. The estimation ranges of NO_x-analyzer varied from 0–5 ppb to 0–5000 ppb (accuracy ±0.050 ppb of all the ranges). The response time of these analyzers are <1 s with a signal noise of 1 % of measured value and the estimation efficiency is >90 %. The zero air calibrations of these analyzers were done using a pure air generator (model: PAG-003, M/s. ECO Physics AG, Switzerland) having air pollutant elimination capacity <0.010 ppb. The analyzers were calibrated and validated using NIST-USA traceable certified NO gas (250 ppb ± 2.5 %, M/s Spectra Gases Inc., USA). The analyzers showed less than ±1 % error rate during calibration. Zero and span calibrations (before and after the measurement) of these analyzers were performed for a week to obtain reproducible values. Ambient SO₂ was measured continuously using a calibrated SO₂-analyzer (model: APSA 360A, M/s. Horiba Ltd, Japan) at the same location. The instrument was calibrated daily using in-built calibrator for zero and span.

Meteorological parameters such as temperature (accuracy: ±1 °C), relative humidity (accuracy: ±2 %), wind direction (accuracy: ±3 °) and wind speed (accuracy: ±2 % of full scale) were recorded at hourly intervals using calibrated portable weather station.

**Fig. 2.** Average diurnal variation of NH₃, NO, NO₂ and SO₂ over BoB.

The gas analysers were used to record the ambient NH₃, NO, NO₂ and SO₂ concentration at 1 min intervals throughout the study period. The concentrations of all the above trace gases were converted into μg m⁻³ from ppb by respective factors for uniform representation of the results. Statistical analysis of all the data sets collected during the study period was performed using standard recommended methods.

3 Results and discussion

The concentration of trace gases, i.e. NH₃, NO, NO₂ and SO₂, were measured over BoB during W ICARB campaign. The average values of trace gases with day and night average values are summarized in Table 2. The average concentration of ambient NH₃ was recorded as 4.78 ± 1.68 μg m⁻³ with a day/night ratio of 1.03, whereas average concentration of NO was recorded 0.31 ± 0.14 μg m⁻³ with a day/night ratio of 0.79.

Figure 2 shows the average diurnal variation of NH₃, NO, NO₂ and SO₂ along with ambient temperature and RH over

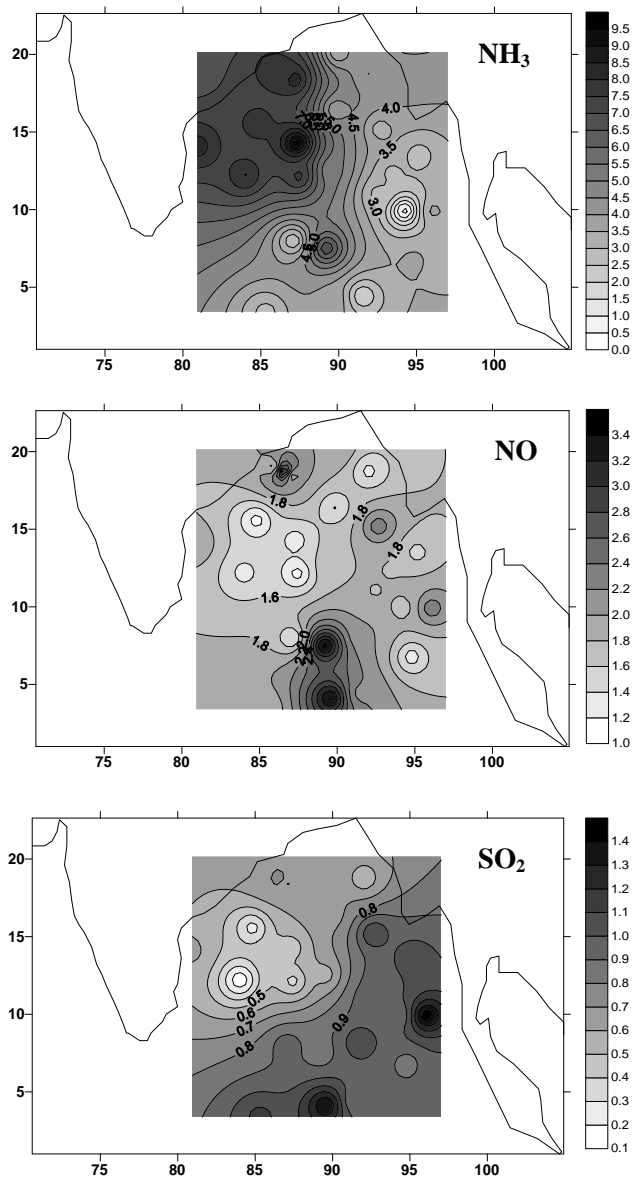


Fig. 3. Latitudinal and longitudinal variations of trace gases (NH₃, NO, SO₂) over BoB.

BoB. In the present observations, non-significant diurnal variations of ambient NH₃ and NO (Fig. 2) were observed, whereas NO₂ and SO₂ showed significant diurnal variations. The average daytime concentration of NO₂ was recorded as $0.27 \pm 0.12 \mu\text{g m}^{-3}$, whereas average nighttime NO₂ concentration recorded was as $0.34 \pm 0.16 \mu\text{g m}^{-3}$. Nighttime increase in NO₂ concentration may be attributed to conversion of NO to NO₂ with the reaction of O₃ as well as lowering of the boundary layer during winter. Figure 3 represents the spatial distribution of trace gases (NH₃, NO, SO₂) over BoB region. The average concentration of ambient NH₃ during campaign recorded as $4.78 \pm 1.68 \mu\text{g m}^{-3}$ with a range of $0.23\text{--}14.34 \mu\text{g m}^{-3}$. Concentration of ambient NH₃ gas in

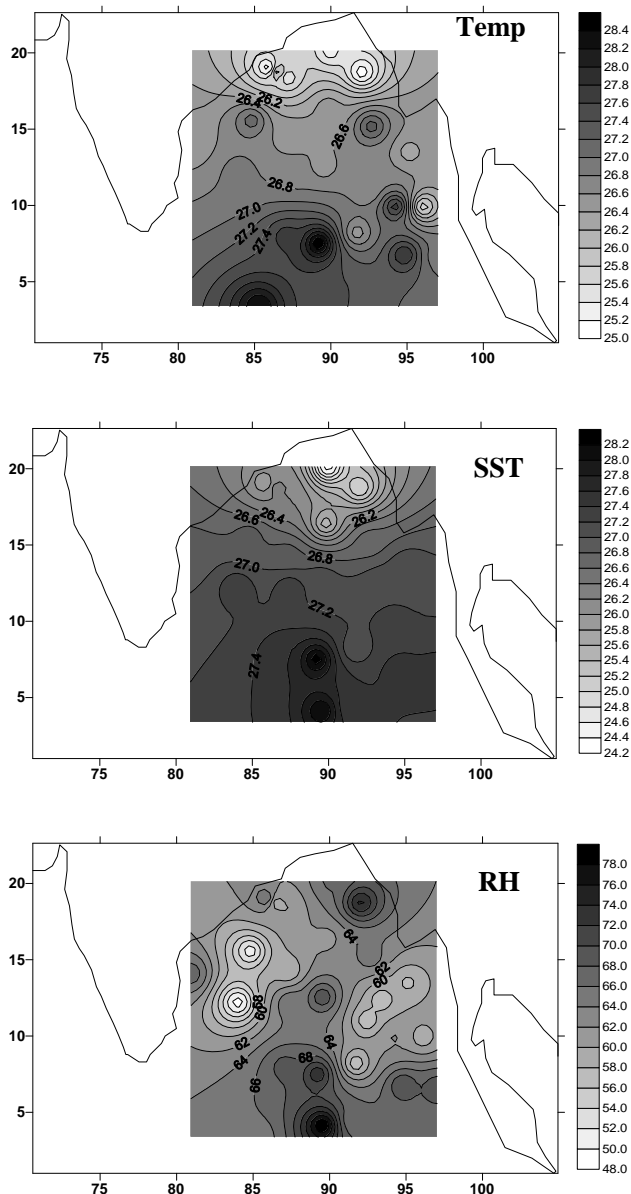


Fig. 4. Latitudinal and longitudinal variations of meteorological parameters over BoB.

the present study is almost one order higher than the earlier reported values over the central Indian Ocean as well as BoB (Norman and Leck, 2005; Schafer et al., 1993) (Table 1).

A strong positive west-east (zonal) gradient (0.184) was observed in ambient NH₃ concentration ($6.0\text{--}9.0 \mu\text{g m}^{-3}$) over the area close to the west coast of BoB as compared to other parts of the BoB (Fig. 3). It is to be noted that agricultural activities (Sharma et al., 2010a, b), livestock, biomass burning and transport may contribute to the emission of large amounts of NH₃ (Sutton et al., 1995, 2000). Khemani et al. (1987) reported the concentration of NH₃ in coastal region of BoB of the order of $1.4\text{--}2.9 \mu\text{g m}^{-3}$. Carmichael et al. (2003) also reported high NH₃

concentrations (6.4–7.1 $\mu\text{g m}^{-3}$) at two sites (Bhubneswar and Berhampur) of the western coast of BoB. West-east positive gradient observed in the western coast of BoB could be due to transport of NH₃ locally, since we do not have any record of dissolved NH₃ in seawater and NH₄⁺ in the sea water. It is difficult to comment on comparative quantification of biogenic oceanic source of NH₃ due to phytoplankton and sea birds as compared to anthropogenic activities. Norman and Leck (2005) reported distribution of NH₃ with the range of 1.1–3.2 nmol m^{-3} (0.019–0.054 $\mu\text{g m}^{-3}$) in the marine boundary layer over Atlantic and South Indian Ocean during Cruise99. They also connected peak value of NH₃ to biomass combustion and dust sources on the African continent. Compared to these data, it may be concluded that NH₃ concentrations over BoB are quite high. Since the lifetime of ambient NH₃ gas is short (1–5 d), particularly in the humid oceanic atmosphere, conversion to particulate NH₄⁺ is supposed to be very fast (30 % h⁻¹). In the middle region of BoB and southern part of BoB, ambient NH₃ is of the order of 1–2 $\mu\text{g m}^{-3}$.

Observation of nitric oxide (NO) over BoB is not reported so far except over the Indian Ocean (Naja et al., 1999; Rhoads et al., 1997). The reported value over the Arabian Sea and North Indian Ocean varies in the range of 0.06–0.19 $\mu\text{g m}^{-3}$. In the present study, average concentration of NO was of the order of 1.89 $\mu\text{g m}^{-3}$ with day/night ratio 0.98, one order higher than the reported value over the Arabian Sea. Spatial distribution of NO shows two peaks in the southern part of BoB with low values in the coastal region and middle of BoB. The peak value of NO resembles the peak of SST and RH. Due to its short lifetime, source of NO in the southern BoB is obviously marine rather than continental transport. The conversion from gaseous NO to particulate NO₃⁻ is unlikely in the southern BoB.

Observation of in-situ NO₂ over the Indian Ocean, particularly BoB, is very limited and only supported by satellite observations (Kunhikrishnan et al., 2004; Franke et al., 2009). They showed that the central Indian Ocean in the Southern Hemisphere is not always as pristine as observed earlier during the winter monsoon period, but is polluted during the monsoon transition periods by pollution plumes from Africa and Southeast Asia. Generally, the most polluted region is the BoB, which is influenced by Indian and south-east Asian outflow during most of the year and China during part of the year. In the present study, average concentration of NO₂ was recorded as 0.31 $\mu\text{g m}^{-3}$ with a day/night ratio 0.79. Concentration of NO₂ is very low and very often it is observed to be below the lower detection limit (LDL \pm 0.04 $\mu\text{g m}^{-3}$) of the instrument (NO_x-analyzer; model: CLD88p; M/s. ECO Physics AG) at few locations over BoB. Correlations of these trace gases with meteorological parameters are summarized in Table 3.

Similarly, measurement of SO₂ over the Indian Ocean is also very limited (Reiner et al., 2001; Shon et al., 2001;

Table 3. Correlation matrix of ambient NH₃, NO, NO₂ and SO₂ with meteorological parameters over BoB.

	NH ₃	NO	NO ₂	SO ₂	Temp	SSTemp	RH
NH ₃	1.00						
NO	0.04	1.00					
NO ₂	-0.56*	0.28	1.00				
SO ₂	-0.58*	0.51	0.25	1.00			
Temp	-0.27	0.13	0.26	0.26	1.00		
SSTemp	-0.09	0.17	0.21	0.34	0.76*	1.00	
RH	-0.09	0.21	-0.50*	0.29	0.20	0.09	1.00

* significant at 5 % level ($n = 600$)

Putaud et al., 1992) and mostly done over the southern Indian Ocean. Using aircraft data, Reiner et al. (2001) have reported SO₂ profiles with surface concentration of the order of 0.63 $\mu\text{g m}^{-3}$ within the area of 8.1–8.3° N and 69.7–70.1° E. They have reported an elevated layer with concentration > 1.9 $\mu\text{g m}^{-3}$ just above 2000 m altitude. In the present study, average concentration of SO₂ was recorded as 0.80 $\mu\text{g m}^{-3}$ with a day/night ratio of 1.15. Since its lifetime is a few days, transport of continental SO₂ to BoB, which is surrounded by densely polluted areas, changes the concentration, depending on the wind pattern. In the eastern coastal region, gaseous SO₂ shows low value and it seems that the gas-phase of SO₂ has been immediately converted to SO₄²⁻ in the region.

Spatial distribution of sea surface temperature (SST), ambient temperature and relative humidity (RH) are plotted over BoB (Fig. 4). North-to-south positive gradient in ambient temperature and SST is noticed with peak value in the southern of BoB, whereas spatial distribution of RH shows zonal gradient. Influences of ambient temperature and RH on aerosol could be identified in nitrate formation (Seinfeld and Pandis, 2006). During daytime with increasing UV radiation, NO rapidly converts into NO₂ in the presence of O¹D and atmospheric NO₂ reacts with hydroxyl radical (OH) to form the nitric acid (HNO₃). However, SO₂ also reacts with the hydroxyl radical (OH) to form sulphuric acid (H₂SO₄). During nighttime, NO₃⁻ is the source of the HNO₃ in the atmosphere. NO₃⁻ reacts either with the NO₂ to form N₂O₅, which reacts with OH to form HNO₃, or NO₃⁻ directly reacts with water vapor to form HNO₃ during nighttime. The reaction of HNO₃ or H₂SO₄ with NH₃ is reversible and forms NH₄NO₃ and (NH₄)₂SO₄, respectively.

4 Conclusion

In the present study, the concentration of ambient NH₃, NO, NO₂ and SO₂ were measured over BoB. The prominent latitudinal and longitudinal variations of the trace gases were observed over BoB, whereas NH₃ and NO showed the non-significant diurnal variation. The average concentration of

ambient NH₃, NO, NO₂ and SO₂ were recorded as 4.78 ± 1.78 , 1.89 ± 1.26 , 0.31 ± 0.14 and $0.80 \pm 0.30 \mu\text{g m}^{-3}$, respectively, with a range of 0.23–14.34, 0.21–9.80, 0.10–1.00 and 0.35–3.53 $\mu\text{g m}^{-3}$, respectively. Coastal region of BoB reported large concentrations of NH₃, NO, NO₂ and SO₂, suggesting the role of continental influence in addition to biogenic sources, particularly near Kolkata and Chennai regions. Presence of higher concentrations of SO₂ than expected in the south of BoB suggest biogenic sources, particularly the role of DMS (dimethyl sulfide). Since we do not have measurement of DMS or its precursor MSA (methanesulfonic acid), it is difficult to conclude for certain.

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