

Article

## Study on mixing ratio of atmospheric ammonia and other nitrogen components

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### Abstract

Mixing ratios of atmospheric NH<sub>3</sub>, NO and NO<sub>2</sub> were measured during September-October 2008 and September-October 2009 over Delhi to study their variation and possible sources in respect of meteorological condition. Mixing ratio of atmospheric NH<sub>3</sub> ranges from 1.05 – 39.26 ppbv with an average value of 17.47 ± 3.25 ppbv during 2008. The average mixing ratio of atmospheric NH<sub>3</sub> was increased by 7.2% during 2009 over that of 2008. The average mixing ratio of NO<sub>2</sub> during 2008 recorded as 4.46 ± 1.26 ppbv with ranges between 1.05 – 16.06 ppbv. Significantly higher (P<0.05) NO<sub>2</sub> mixing ratio was recorded during 2008. Early morning increase in NO<sub>2</sub> mixing ratio may be attributed to conversion of NO to NO<sub>2</sub> with the interaction of O<sub>3</sub>. Diurnal and day-to-day variations in mixing ratio of atmospheric NH<sub>3</sub>, NO and NO<sub>2</sub> were observed during the study. Result reveals that the mixing ratio of atmospheric NH<sub>3</sub> is correlated with NO (r<sup>2</sup> = -0.86), NO<sub>2</sub> (r<sup>2</sup> = 0.35), relative humidity (r<sup>2</sup> = -0.87) and ambient temperature (r<sup>2</sup> = 0.88).

**Keywords** atmospheric NH<sub>3</sub>; NO; NO<sub>2</sub>; chemiluminescence method; diurnal variation.

### 1 Introduction

Ammonia is the only gaseous base in the troposphere that arises from both natural and anthropogenic sources. Anthropogenic sources of atmospheric NH<sub>3</sub> are agricultural practices, livestock establishment, roadside vehicles and industrial activities (Sutton et al., 2000; Li et al., 2006; Sharma et al., 2010a) along with natural sources like forest fire and losses from soil under native vegetation (Olivier et al., 1996; Lee et al., 2005). Total global NH<sub>3</sub> emission has been estimated to be 50 Tg N yr<sup>-1</sup> (Bouwman et al., 1997) of which 39.9 Tg is from anthropogenic activities. It is an important component of the nitrogen cycle and plays an important role in the acid-base chemistry of the troposphere. NH<sub>3</sub> although not a very toxic substance, contributes to eutrophication of oligotrophic ecosystems, acid rain (Erisman et al., 2001), photochemical smog, mentioned in connection of forest decline and global warming (Wang et al., 1976). NH<sub>3</sub> cannot be rained out from the troposphere without the atmospheric acids (Dickerson, 2003) unlike NO and NO<sub>2</sub>. Ammonia is the primary neutralizing agent for atmospheric acids (H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub> and HCl) and forms inorganic aerosols like (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, NH<sub>4</sub>NO<sub>3</sub> and NH<sub>4</sub>Cl (Aneja et al., 2001) which affects global radiative forcing (Walker et al., 2004). The mixing ratio of atmospheric NH<sub>3</sub> also depends on modification of the transport and deposition pattern of SO<sub>2</sub> and NO<sub>x</sub> (Sutton et al., 2000). Depending on the meteorological conditions NH<sub>3</sub> may be transported <10 to 100 km; however;

$\text{NH}_4^+$  may be transported much longer distance (100 to >1000 km) under certain environmental conditions (Asman et al., 1998). Thus contributes to international transboundary air pollutant issues addressed by UNECE Convention on Long Range Transboundary Pollution (Fowler et al., 1998).

One of the major problems originated by the air pollution in urban areas is the pollution caused by photochemical oxidants. Among these,  $\text{O}_3$  and  $\text{NO}_x$  are capable to cause adverse impacts on human health and environment (Lee et al., 1996; WHO 2000). These compounds lead to several health problems like lung tissue damage, emphysema and bronchitis. Sources of atmospheric  $\text{NO}_x$  vary from rural areas to cities. The oxides of nitrogen ( $\text{NO}_x$ ) are emitted from several sources (*viz.* thermal power plants, transport, biomass burning, industries) to the atmosphere as NO, which is simultaneously converted to  $\text{NO}_2$  and other nitrogenous species leading to formation of several photo oxidants (*viz.*  $\text{O}_3$ ,  $\text{N}_2\text{O}_5$ , PAN) (Gupta et al., 2003). The efficiency of emission control depends on the relationship between primary and secondary pollutants. Hydrogen produced in the water-gas shift reaction, can reacts with NO in presence of CO ( $\text{NO} + \text{CO} + \text{H}_2 \rightarrow \text{NH}_3 + \text{CO}_2 + \text{H}_2\text{O}$ ) in the atmosphere to generate  $\text{NH}_3$  as a secondary pollutant (Gandhi and Shelef, 1991). However, atmospheric  $\text{NH}_3$  can get oxidized to NO ( $\text{NH}_3 + \text{O}_2 \rightarrow \text{NO} + \text{H}_2\text{O}$ ;  $G_0 = -57.34 \text{Kcal mole}^{-1}$ .) and can oxidized with  $\text{NO}_2$  to  $\text{N}_2\text{O}$  ( $\text{NH}_3 + \text{NO}_2 \rightarrow \text{N}_2\text{O} + \text{H}_2\text{O}$ ) which is a potent greenhouse gas.

In India, distribution and sources of atmospheric  $\text{NH}_3$  has been reported in limited areas and time (Khemani et al., 1989; Kulshrestha et al., 1996; Parashar et al., 1996) using chemical trap method integrated over 3-4 hrs of sampling. The sampling efficiency of this method is of the order of 91% and the reproducibility of  $\text{NH}_3$  is 4.7% (Parmar et al., 2001). In spite of the important role of ambient  $\text{NH}_3$  in the atmospheric acid-base chemistry, its diurnal variations, sources and interactions with  $\text{NO}_x$  during different seasons has not been well characterized in India. However, Sharma et al. (2010b) has reported the temporal variations of ambient  $\text{NH}_3$  over Delhi using chemiluminescence method.

The mixing ratio of atmospheric  $\text{NH}_3$  has been measured during Sept.-Oct. 2008 and Sept.-Oct. 2009 to find out the contribution of sources other than agricultural practices. Inventories on  $\text{NH}_3$  emission have established that the agricultural sector (including livestock) contributes about 80-90% of the total atmospheric  $\text{NH}_3$  and most of the  $\text{NH}_3$  emission from agricultural field is after the application of inorganic nitrogen fertilizers. September and October is mainly the harvest month in north India and there is no fertilizer application in the agricultural fields. Hence, it is expected that agricultural contribution to the mixing ratio of atmospheric  $\text{NH}_3$  is minimum during this period. The study represent diurnal and temporal variations of the mixing ratio of atmospheric  $\text{NH}_3$ , NO and  $\text{NO}_2$  during Sept. – Oct. 2008 and Sept. – Oct. 2009 and analyze possible sources during the season based on meteorological conditions.

## 2 Materials and Methods

### 2.1 Description of the study site

The study was carried out at National Physical Laboratory, New Delhi ( $28^{\circ}38'N$ ,  $77^{\circ}10'E$ ; 220m m.s.l.), India (Fig. 1). New Delhi, the capital city of India, located in the northern part of India. It is one of the ten most polluted cities of the Asia and third most polluted city of the country having numerous industries in and around the city. The total number of registered vehicles in the city reached to about 5.67 million during the year 2009 (Delhi Statistical Handbook, 2009). Large agricultural fields of Indian Agricultural Research Institute (IARI) located at the NW to SW parts around 500 m away from the study site. Central ridge forest area is located in the SE direction. One industrial area is located about 6 km away in west and SW direction. NE direction is mainly dominated with the traffic during the rush hours (8 am-11am and 4 pm-8 pm) of the day. Wind flow and temperature ( $12^{\circ}\text{C}$  to  $35^{\circ}\text{C}$ ) remains moderate over Delhi during September – October. Most of the time wind blows from WNW, SW and NW directions over Delhi (Goyal and Sidhartha, 2002).

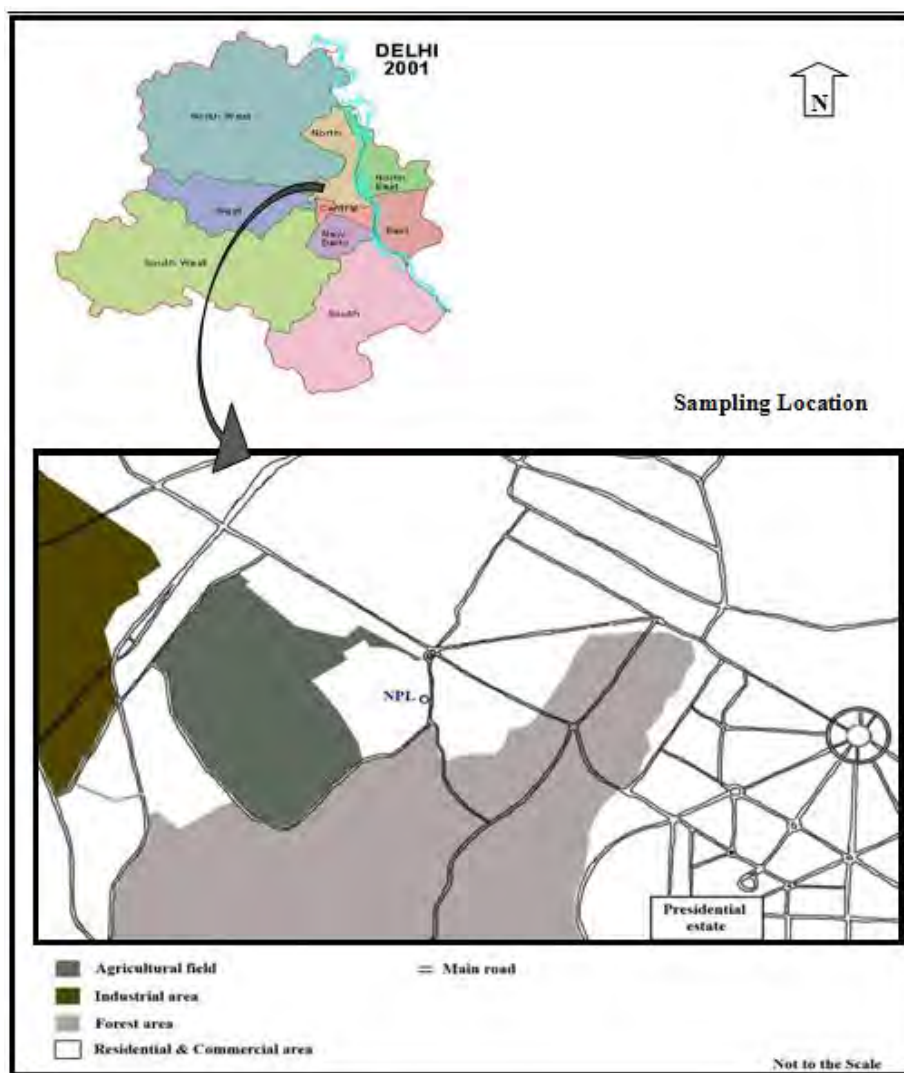


Fig. 1 Location map of the observational site.

## 2.2 Sampling and analysis

Atmospheric  $\text{NH}_3$  mixing ratio was measured using  $\text{NH}_3$ -analyzer (Model: CLD88CYp, M/s. ECO Physics, Switzerland) operating based on chemiluminescence method. In this analyzer two catalytic converters of different characteristics allow sequential detection of  $\text{NO}_x$  and  $\text{NO}_x$ -amines by converting them into  $\text{NO}$ .  $\text{NH}_3$  mixing ratio was calculated from the difference between  $\text{NO}_x$  and  $\text{NO}_x$ -amine ( $\text{NH}_3 = \text{NO}_x\text{-amine} - \text{NO}_x$ ). The measurement range of  $\text{NH}_3$  analyzer varies between 0–5000 ppbv with an accuracy of  $\pm 0.050$  ppbv.  $\text{NO}$  and  $\text{NO}_2$  were also measured using chemiluminescence method with  $\text{NO}_x$ -analyzer (Model: CLD88p, M/s. ECO Physics, Switzerland) equipped with photocatalytic converter (Model: PLC860, M/s. ECO Physics, Switzerland). The measurement range of  $\text{NO}_x$ -analyzer varies from 0–5000 ppbv with an accuracy of  $\pm 0.050$  ppbv. Sampling efficiency of the  $\text{NH}_3$  analyzer is more than 90% with a reproducibility of 91–96% which is much higher than the conventional chemical trapping method (reproducibility 4.7%). Both analyzers were calibrated and validated using NIST certified  $\text{NO}$  gas (500 ppbv  $\pm 5\%$ , M/s Spectra Gases Inc., USA) with

$\pm 1\%$  error during calibration. Analyzers were recorded the mixing ratio of atmospheric  $\text{NH}_3$ , NO and  $\text{NO}_2$  at 1 minute interval during the study period.

Meteorological parameters like temperature (accuracy:  $\pm 1^\circ\text{C}$ ), relative humidity (accuracy:  $\pm 2\%$ ), wind direction (accuracy:  $\pm 3^\circ$ ) and wind speed (accuracy:  $\pm 2\%$  of full scale) were recorded at every 1 minute interval using calibrated Portable Weather Logger (M/s. RainWise Inc., Bar Harbor, Maine) throughout the study period. All meteorological parameters were reported as 1h average.

Sampling inlets of all analyzers and the Portable Weather Logger were stationed at same height (10 m above ground) and place at the National Physical Laboratory, New Delhi. These instruments were operated continuously during 7th – 22nd September and 12th – 29th October- 2008) and 6th – 20th September and 10th – 26th October-2009. Statistical analysis of all parameters was completed following standard statistical method.

### 3 Results and Discussion

$\text{NH}_3$ , NO and  $\text{NO}_2$  were measured continuously alongwith different weather parameters during Sept.– Oct., 2008 and Sept.– Oct., 2009 over Delhi. The average mixing ratio of  $\text{NH}_3$ , NO and  $\text{NO}_2$  are summarized in Table 1. All trace gases were measured at 1 minute interval during the study period, hourly average of the mixing ratios of all three gases during 2008 and 2009 are represented as diurnal variations (Fig. 1). Similarly, average hourly wind direction from each  $10^\circ$  directions was calculated to compare with mixing ratios of all three trace gases (Fig. 2) to establish possible local source of these pollutants.

#### 3.1 Variation of atmospheric $\text{NH}_3$ mixing ratio

Average mixing ratio of atmospheric  $\text{NH}_3$  during 2008 was recorded as  $17.47 \pm 3.25$  ppbv with a range of 1.05 – 39.26 ppbv (Table 1), whereas it was  $18.75 \pm 4.85$  ppbv with a maximum value of 40.62 ppbv during 2009. However, there is no significant difference ( $P > 0.01$ ) in average mixing ratio of  $\text{NH}_3$  between the years. Fig. 2 shows the diurnal variation of atmospheric  $\text{NH}_3$ , NO and  $\text{NO}_2$  during 2008 and 2009. Maximum atmospheric  $\text{NH}_3$  mixing ratio was recorded between 10:00 to 15:00h. Average day and night time mixing ratio of atmospheric  $\text{NH}_3$  recorded as  $17.56 \pm 3.05$  ppbv and  $17.37 \pm 3.25$  ppbv respectively during 2008, whereas; average day and night time mixing ratio of atmospheric  $\text{NH}_3$  recorded as  $19.47 \pm 4.80$  ppbv and  $18.03 \pm 4.82$  ppbv respectively during 2009 (Table 1). However, there is no significant difference in the mixing ratio of  $\text{NH}_3$  between years ( $P > 0.05$ ). Results suggest that the day time average mixing ratio of atmospheric  $\text{NH}_3$  during both years are marginally higher than the night time. The study reveals that the atmospheric  $\text{NH}_3$  mixing ratio is significantly correlated with ambient temperature ( $r^2 = 0.88$ ) as reported by Parmar et al. (2001). Day time increase in ambient temperature attributes to increase in soil temperature which leads to increase soil ammonification and release of  $\text{NH}_3$  from soil (Sharma et al. 2010b). However, apart from the increase in ambient temperature, increase of vehicular density during day time may also contribute to increase in the ambient mixing ratio of  $\text{NH}_3$ . Similar results were also reported by Whitehead et al. (2007) over urban area of Manchester, a city of NW England.

The average mixing ratio of atmospheric  $\text{NH}_3$  during the study period was recorded lower than the winter season in Delhi ( $47.3 \pm 13.6$  ppbv) (Kapoor et al., 1992) during 2008. However, Sharma et al. (2010a) has reported a significant day and night time difference in the  $\text{NH}_3$  mixing ratio at the same observational site during winter. They had reported the higher night time  $\text{NH}_3$  mixing ratio related with significant decrease of temperature and boundary layer height. However, there is no significant difference among day and night time mixing ratio has been observed during the present study period as day and night time temperature variation during the study period ranged between  $20^\circ\text{C} - 25^\circ\text{C}$  which do not lead to significant decrease in the boundary layer height. However, the lower  $\text{NH}_3$  mixing ratio during the study period might be due to reduction in agricultural activities and no nitrogen fertilizer application at nearby agricultural fields as is generally the

harvest season in north India. The emission of NH<sub>3</sub> from the cultivated land with applied inorganic fertilizers is more compared to barren soil and nitrogen fertilizer application is regarded as the prime source of ambient NH<sub>3</sub> (Sakuri et al., 2003; Whitehead et al., 2007).

**Table 1** Variations in mixing ratio of atmospheric NH<sub>3</sub>, NO and NO<sub>2</sub>.

Parameter	Mixing ratio (ppbv)					
	Min	Max	Day	Night	Difference	Average
<b>NH<sub>3</sub></b>						
2008	1.05	39.26	17.56 ± 3.05	17.37 ± 3.45	ns	17.47 ± 3.25
2009	1.09	40.62	19.87 ± 4.80	17.63 ± 4.82	*	18.75 ± 4.81
<b>NO</b>						
2008	1.79	62.15	31.47 ± 10.66	34.61 ± 6.97	*	33.04 ± 8.82
2009	2.48	75.46	30.44 ± 8.69	34.22 ± 5.17	*	32.33 ± 6.93
<b>NO<sub>2</sub></b>						
2008	1.05	16.06	4.88 ± 1.46	4.02 ± 1.07	ns	4.46 ± 1.26
2009	1.65	12.30	5.60 ± 0.80	4.83 ± 0.86	*	5.23 ± 0.83

(± Value is standard deviation)

ns = Not Significant

\* Significantly difference at P < 0.05

n = 360

### 3.2 Variation of atmospheric NO mixing ratio

The average mixing ratio of NO was recorded as 33.04 ± 8.82 ppbv with a range of 1.79 to 62.19 ppbv during 2008 and 32.33 ± 6.93 with a range of 2.48 to 75.46 ppbv during 2009 (Table 1). There is no significant difference in the mixing ratio of NO between years (P > 0.05). The increase in the atmospheric NO mixing ratio observed between 03:00 to 05:00 h and 08:00 to 10:00 h; thereafter decreases to minimum throughout the day time (Fig. 2). Sharma et al. (2010a) has also reported an early morning increase in the NO mixing ratio. Early morning increase in the NO mixing ratio may be attributed to photochemical breakdown of stable nitrogenous compounds like peroxy-acetyl-nitrite (PAN) with the onset of the sunlight. Increase of NO mixing ratio between 08:00 to 10:00 h may attribute to increase in traffic density, production of NO from PAN and other atmospheric nitrogen compounds (e.g. N<sub>2</sub>O<sub>5</sub>) in presence of sunlight and higher atmospheric O<sub>3</sub> mixing ratio (Yin et al. 2008). Atmospheric NO mixing ratio was found significantly negative correlated (r<sup>2</sup> = -0.89) with ambient temperature.

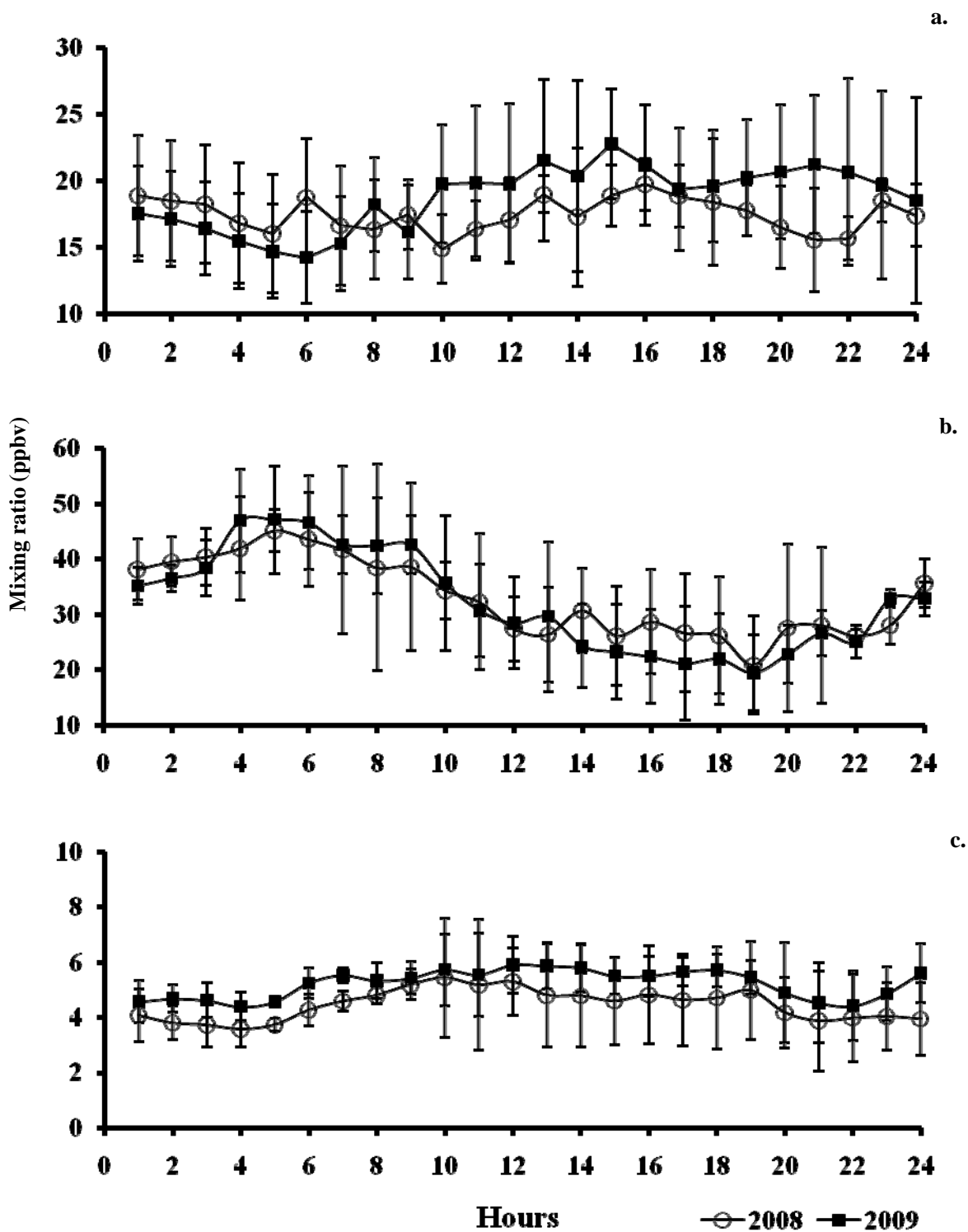


Fig. 2 Average diurnal variations of mixing ratio of atmospheric (a.)  $\text{NH}_3$ , (b.) NO and (c.)  $\text{NO}_2$  during study period

### 3.3 Variation of atmospheric $\text{NO}_2$ mixing ratio

The average mixing ratio of  $\text{NO}_2$  recorded as  $4.46 \pm 1.26$  ppbv and  $5.23 \pm 0.83$  ppbv during 2008 and 2009 respectively. However, the difference of average  $\text{NO}_2$  mixing ratio between the years is significant ( $P < 0.05$ ).

Increase in NO<sub>2</sub> mixing ratio (Fig. 2) between 9:00 to 15:00 h attributed to the photochemical formation of NO<sub>2</sub> from the reaction of NO with O<sub>3</sub>. Just after sunrise, PAN breaks into NO which rapidly converts into NO<sub>2</sub> in interaction with O<sub>3</sub> or O<sup>1</sup>D. Ambient mixing ratio of NO<sub>2</sub> found significantly correlated with NO ( $r^2 = -0.86$ ) and NH<sub>3</sub> ( $r^2 = 0.35$ ) during the study period.

### 3.4 Possible sources of atmospheric NH<sub>3</sub>, NO and NO<sub>2</sub>

Average surface wind pattern at the observational site was analyzed to demonstrate the possible local sources of the atmospheric NH<sub>3</sub>, NO and NO<sub>2</sub> during the study period. Fig. 3a shows wind speed from different directions during the study period. Higher wind speed (2.8 m s<sup>-1</sup>) was recorded from the NW direction; however there is occasional wind from NE, east and SE directions ranging from 1.2 – 2.3 m s<sup>-1</sup>. Fig. 3b represents the distribution of atmospheric NH<sub>3</sub> with respect to the wind directions. Result suggests that the higher mixing ratio of atmospheric NH<sub>3</sub> was recorded with the wind flows from NW, east and SW directions. However, Fig. 3a suggests higher wind speed and maximum wind events from these directions during the study period. The large experimental crop field is located in these directions of the observational site, however; fertilizer application to the field during the study period was minimum/nil. Higher wind speed (2.8 m s<sup>-1</sup>) from these directions may bring the NH<sub>3</sub> from far source, apart from the nearby crop land. Soil ammonification under higher ambient temperature in the crop lands located in these directions may contribute to the NH<sub>3</sub> from these directions, however; significant contribution may be from road traffic and the industrial area located within 5-10 km toward these directions, as NH<sub>3</sub> cannot be transported from very long distance source due to its rapid conversion to NH<sub>4</sub><sup>+</sup> (Asman and Jaarsveld 1992). Similar observations are also reported by other researchers from different parts of the world (Sakuri et al. 2003; Whitehead et al. 2007). Sporadic NH<sub>3</sub> mixing ratio was also recorded with wind flow from NE and east directions may attribute to the road traffic as low wind speed (1.2 – 2.3 m s<sup>-1</sup>) from these directions can transport the NH<sub>3</sub> from very near source like road traffic (Gandhi and Shelef, 1991) which is located within 100 m of the observation site and from the forest area (Anderson et al., 1993) which is located within 500 m of the observation site. NH<sub>3</sub> is also generated as a secondary product following the equations mentioned above (Pierson and Brachaczek, 1983; Gandhi and Shelef, 1991).

The NO and NO<sub>2</sub> mixing ratio was recorded higher (42.2 and 6.9 ppbv) with the wind flow from the NE and SE directions (Fig. 3c and 3d). Higher NO mixing ratio from NE direction alongwith low wind speed indicates that the major source of NO is the road traffic, which is about 100 m away from the observational site in the NE direction. NO mixing ratio from SE direction may also attribute to the road traffic. NO mixing ratio (range 26.8 – 29.8 ppbv) was also recorded with the wind flow from SW direction. This may be from the nitrification process in the dry soil of the agricultural fields as NO may not be transported from very long distance. NO<sub>2</sub> mixing ratio was recorded from NW, west and SW directions which may attributed to transport of NO<sub>2</sub> from the industrial area, located 5–6 km away from the observational site, however; other sources in the SW and west directions may also contribute to the NO<sub>2</sub> mixing ratio from these sites as wind speed from these directions was recorded high (1.9 to 2.8 m s<sup>-1</sup>) during the study period. NO<sub>2</sub> mixing ratio from NE and SE directions may attribute to local traffic as wind speed from these directions were comparatively low (1.2 to 2.3 m s<sup>-1</sup>).

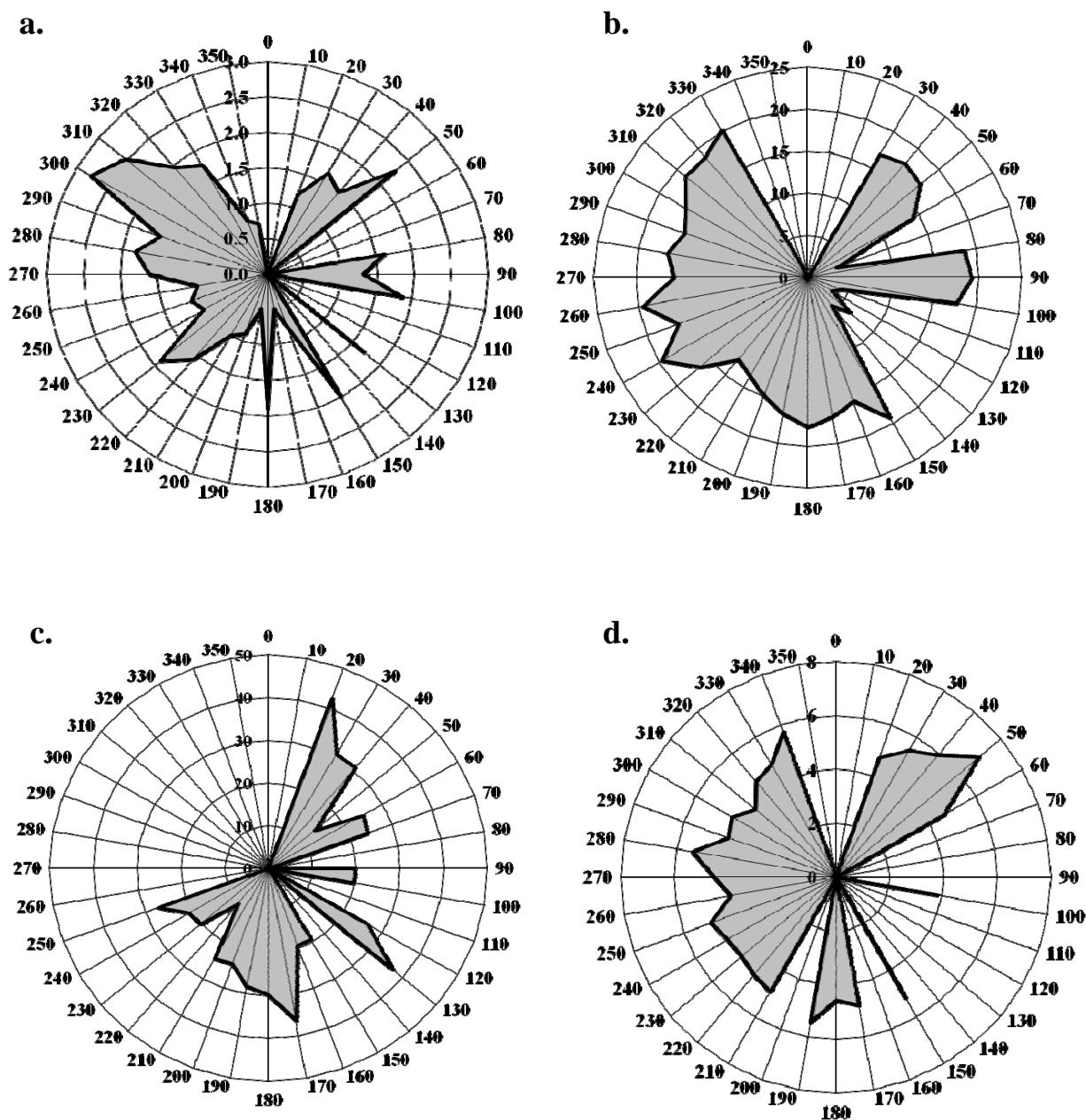


Fig. 3 Distribution of (a.) wind speed ( $\text{m s}^{-1}$ ) and atmospheric mixing ratio (ppbv) of (b.)  $\text{NH}_3$ , (c.)  $\text{NO}$ , (d.)  $\text{NO}_2$  with respect to wind direction during the study period.

#### 4 Conclusion

Average mixing ratio of atmospheric  $\text{NH}_3$  recorded as  $17.47 \pm 3.25$  and  $18.75 \pm 4.81$  ppbv during 2008 and 2009 respectively. The surface wind direction and wind speed analysis indicates that major possible local sources of atmospheric  $\text{NH}_3$  are road traffic and industries, however; soil ammonification may also contribute to atmospheric  $\text{NH}_3$ .  $\text{NO}_2$  can be transported from long distance sources with the high speed wind from NW and west directions. However,  $\text{NO}$  is completely from local sources, either from road traffic near the observational site or from the nitrification in soil in nearby crop land. There is significant difference in the



atmospheric mixing ratio of NO<sub>2</sub> between the year 2008 and 2009, however; there is no significant difference in average mixing ratio of NH<sub>3</sub> and NO<sub>2</sub> between years. Significant positive correlation of atmospheric NH<sub>3</sub> with ambient temperature established the day time higher mixing ratio of NH<sub>3</sub>. Negative correlation of atmospheric NH<sub>3</sub> with NO indicates the possibility of formation of NH<sub>3</sub> with the reaction with NO.

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