



# Influence of Vehicular Emissions (NO, NO<sub>2</sub>, CO and NMHCs) on the Mixing Ratio of Atmospheric Ammonia (NH<sub>3</sub>) in Delhi, India

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## Abstract

Mixing ratios of atmospheric ammonia (NH<sub>3</sub>), nitric oxide (NO), carbon monoxide (CO), nonmethane hydrocarbons (NMHCs), and methane (CH<sub>4</sub>) were measured to investigate the vehicular emissions, which are a dominant source of atmospheric NH<sub>3</sub> in urban sites of Delhi, India from January 2013 to December 2014. The annual average mixing ratios of NH<sub>3</sub>, NO, CO, NMHCs, and CH<sub>4</sub> were 21.2 ± 2.1 ppb, 21.2 ± 6.1 ppb, 1.89 ± 0.18 ppm, 0.67 ± 0.21 ppm and 3.11 ± 0.53 ppm, respectively. Considering NO as a tracer of vehicular plume, ambient NH<sub>3</sub> was correlated with NO during peak traffic hour in the morning (7:00–10:00 h) and evening (17:00–19:00 h) and observed significant positive correlation between them. Result reveals that the mixing ratio of atmospheric NH<sub>3</sub> significantly positive correlated with traffic related pollutants (NO, CO, and NMHCs) during all the seasons (winter, summer, and monsoon). During winter, the average mixing ratio of atmospheric NH<sub>3</sub> was increased by 1.2–3.5 ppb in the morning peak hour, whereas increased by 0.3–1.6 ppb in the evening peak hour. Similarly, an increase in NH<sub>3</sub> mixing ratio was observed during summer (morning: 1.2–2.7 ppb and evening: 1.5–1.6 ppb) and monsoon (morning: 0.4–3.6 ppb and evening: 0.9–1.4 ppb) seasons. The results emphasized that the traffic could be one of the dominant source of ambient NH<sub>3</sub> at the urban site of Delhi, as illustrated by positive relationships of NH<sub>3</sub> with traffic related co-pollutants (NO, CO and NMHCs).

Ammonia (NH<sub>3</sub>) is the third most abundant nitrogen containing gas in the atmosphere after N<sub>2</sub> and N<sub>2</sub>O (Seinfeld and Pandis 1998; Aneja et al. 2000) and principal reduced nitrogen components in the atmosphere (Chang et al. 2016). It plays a strong role in local as well as regional tropospheric chemistry and air quality by serving as a precursor to particulate formation (Seinfeld and Pandis 2006; Sharma et al. 2018). As a primary alkaline gas in ambient air, NH<sub>3</sub> can neutralize nitric acid and sulphuric acid gases to form ammonium nitrate and sulphate (Pinder et al. 2007; Sharma et al. 2014; Jain et al. 2019), which are important constituents of airborne fine particles or PM<sub>2.5</sub> (Chow et al. 1994;

Aneja et al. 2001; Huang et al. 2011a, b; Saraswati et al. 2019). The most recent consideration for NH<sub>3</sub> emissions on the global scale is linked to climate change based on its ability to form PM<sub>2.5</sub>, specifically ammonium particulates (Aneja et al. 2001). The need to better understand the role of this particular air pollutant has been underscored in recent years as its atmospheric concentration increases day by day. Effective management strategies are essential to mitigate the pollutants over public health concern and to enhance visibility (Brunekreef and Holgate 2002; Wang et al. 2012).

Major anthropogenic sources of ambient NH<sub>3</sub> are agricultural practices, livestock, transport, and industrial activities (Sutton et al. 2000; Li et al. 2006; Sharma et al. 2010), along with natural sources, such as forest fires and emissions from soil (Olivier et al. 1998; Lee et al. 2005). Although agriculture is the dominant source of atmospheric NH<sub>3</sub> at continental to global scales (Sutton et al. 2013), in urban areas a significant NH<sub>3</sub> source is gasoline vehicles equipped with three-way catalysts (TWC). The reduction of NO compounds in TWC converters of automobile exhaust and industrial power stations emissions are significant contributors to atmospheric NH<sub>3</sub> in urban environments ( $2\text{NO} + 2\text{CO} + 3\text{H}_2 \rightarrow 2\text{NH}_3 + 2\text{CO}_2$

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or  $2\text{NO} + 5\text{H}_2 \rightarrow 2\text{NH}_3 + 2\text{H}_2\text{O}$  (Gandhi and Shelef 1991). Because of the growing efficiency of TWC to reduce  $\text{NO}_x$  emissions and recent introduction of selective catalytic reduction (SCR) systems in diesel vehicles,  $\text{NH}_3$  is now a dominant-reactive nitrogen species emitted by vehicles (Stritzke et al. 2015). Correlation matrix of  $\text{NH}_3$  with CO also suggested the dependency of  $\text{NH}_3$  on the traffic intensity and air temperature (Perrino et al. 2002; Meng et al. 2011). Li et al. (2006) observed spikes in the  $\text{NH}_3$  time-series that was correlated with the spikes in the CO and inferred that the  $\text{NH}_3$  came from nearby traffic. Limited efforts had been done to explore the vehicle-emitted  $\text{NH}_3$  to the urban atmosphere of India (Sharma et al. 2017). Therefore, more efforts are needed to elucidate the contribution of vehicle-emitted  $\text{NH}_3$  to the urban atmosphere.

In the previous study, we reported the diurnal, seasonal, temporal, spatial, and annual variations in mixing ratio of ambient  $\text{NH}_3$  and its interaction with other trace gases ( $\text{NH}_3$ , NO,  $\text{NO}_2$ , CO,  $\text{SO}_2$ , and  $\text{HNO}_3$ ) under the prevailing meteorological conditions of the observational site of Delhi and reference therein (Sharma et al. 2010, 2014, 2017; Saraswati et al. 2018; 2019). The purpose of this study was to determine whether pollutants (NO,  $\text{NO}_2$ , CO, NMHCs, and  $\text{CH}_4$ ) emitted from roadside traffic are the source of atmospheric  $\text{NH}_3$  over the urban site of megacity Delhi, India? Hence, in this paper, we present the influence of vehicular emissions on the mixing ratio of  $\text{NH}_3$  in the urban atmosphere of megacity Delhi, India.

## Experimental Methods

Delhi, the capital of India, is considered one of the most polluted megacities of the world due to its geographical origin, rapid urbanization, overcrowding, and vehicular emissions (Nagpure et al. 2013; Sharma et al. 2018). The mixing ratios of ambient  $\text{NH}_3$ , NO, CO, NMHCs, and  $\text{CH}_4$  were measured at the CSIR-National Physical Laboratory, New Delhi ( $28^\circ 38' \text{ N}$ ,  $77^\circ 10' \text{ E}$ ; 218 m amsl) from January 2013 to December 2014 (Fig. 1). The observational site represents a typical urban atmosphere, amenable to free wind flow from all the directions and is surrounded by huge roadside traffic. Detailed descriptions of the observational site, including general meteorology (temperature, RH, wind speed, and wind directions) during the observational period are available in Table S1 (in Supplementary Information) and Saraswati et al. (2018).

Mixing ratios of ambient  $\text{NH}_3$  and NO were measured using a  $\text{NH}_3$ -Analyzer (AC32M&CNH<sub>3</sub>, Environment SA, Paris, FR), operating on a chemiluminescence method (accuracy:  $\pm 1.0$  ppb) from January 2013 to December 2014.  $\text{NH}_3$  analyzer was calibrated periodically using Pure Air Generator (Model: PAG-003, M/s. ECO Physics AG,

Switzerland, accuracy  $\pm 0.01$  ppb) and NIST certified NO span gas ( $500$  ppb  $\pm 5\%$ ). Mixing ratio of CO was recorded using a non-dispersive infrared (NDIR) gas filter correlation analyzer (Model 48C; Thermo Fisher Scientific, Waltham, MA). CO-Analyzer was calibrated using NIST traceable certified CO gas ( $8.1$  ppm  $\pm 5\%$ ) and Pure Air Generator. NMHCs and  $\text{CH}_4$  were measured using flame ionization detection (FID) based Hydrocarbon analyzer (APHA-360; Horiba, Japan) from April to December 2014. Zero and span gas of hydrocarbon analyzer were calibrated using Pure Air Generator and NIST certified Propane and Methane gases (Sharma et al. 2017). Statistical analysis of these trace gases ( $\text{NH}_3$ , NO, CO, NMHCs, and  $\text{CH}_4$ ) were performed using standard recommended methods.

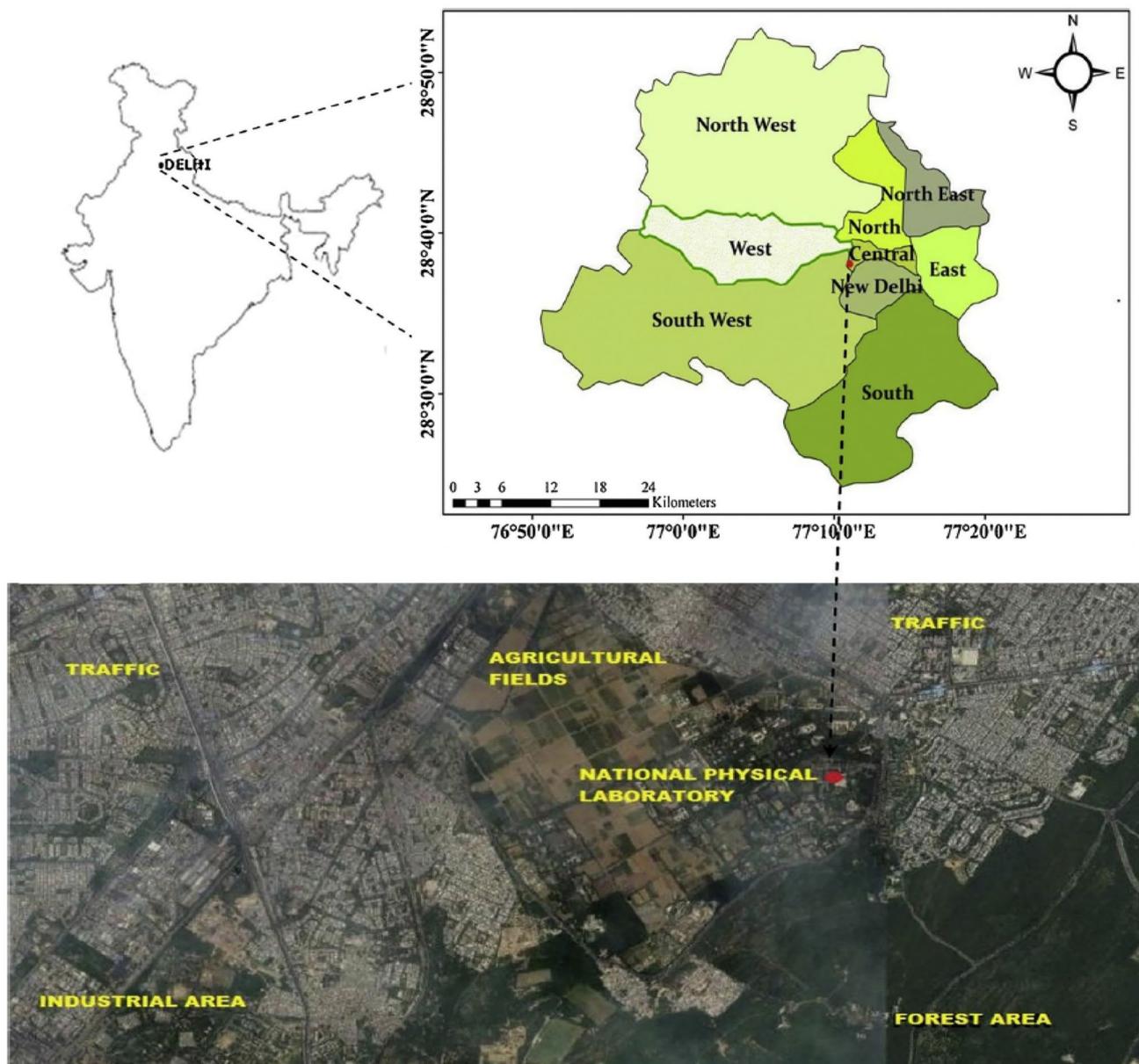
## Results and Discussion

### Mixing Ratios of $\text{NH}_3$ , NO, CO, NMHCs and $\text{CH}_4$

The annual average mixing ratios of ambient  $\text{NH}_3$ , NO, CO, NMHCs, and  $\text{CH}_4$  were  $21.2 \pm 2.1$  ppb;  $21.2 \pm 6.1$  ppb,  $1.89 \pm 0.18$  ppm,  $0.67 \pm 0.21$  ppm, and  $3.11 \pm 0.53$  ppm, respectively from January 2013 to December 2014 (Table 1). Seasonal average of ambient  $\text{NH}_3$ , NO, CO, NMHCs, and  $\text{CH}_4$  are tabulated in Table 1 with maxima during winter season. Figure 2 showed the average diurnal variations in mixing ratios of  $\text{NH}_3$ , NO, CO, and NMHCs during winter (NDJF), summer (MAMJ), and monsoon (JASO) seasons. Kapoor et al. (1992) reported the average  $\text{NH}_3$  mixing ratio of  $\text{NH}_3$  as  $47.3 \pm 13.6$  ppb in Delhi, whereas Sharma et al. (2010) reported the average mixing ratio of ambient  $\text{NH}_3$  as  $20.23 \pm 2.71$  ppb. Zutshi et al. (1970) reported the  $\text{NH}_3$  mixing ratio as 50.7 ppb at Mumbai, whereas Sharma et al. (2016) reported the  $\text{NH}_3$  mixing ratio as  $43.4 \pm 7.0$  ppb at Kolkata, India. The diurnal, seasonal, annual, and spatial variations in mixing ratio of ambient  $\text{NH}_3$  and its interaction with other trace gases ( $\text{O}_3$ , NO,  $\text{NO}_2$ , CO,  $\text{SO}_2$ , and  $\text{HNO}_3$ ) under the prevailing meteorological conditions at Delhi (same site) and other locations of India are already discussed in our previous studied (Sharma et al. 2010, 2014, 2017; Saraswati et al. 2018, 2019) and compared with other studies (Sharma et al. 2014).

### Diurnal Profiles of $\text{NH}_3$ and Insight Into Sources

Hourly measurements over long-term periods offer a unique opportunity to provide the robust diurnal profiles for each season. Figure 2 showed the average diurnal profiles of  $\text{NH}_3$ , NO, CO, and NMHCs mixing ratios during winter, summer, and monsoon seasons. On road vehicular exhaust contributes 36% of  $\text{NO}_x$ , whereas 83% of CO to the ambient air of Delhi (Sharma and Dikshit 2016). In Fig. 2, CO, NO, and



**Fig. 1** Map of observational site. *Source:* Google map

NMHCs shows a bimodal diurnal profile, with maxima in the morning (starting at 06:00 IST; Indian Standard Time) and the evening (starting at 17:00 IST), consistent with the traffic flow in Delhi (peak hours and non-peak hours). Therefore, variation in NO and CO mixing ratios can be used as a potential indicator of vehicular emissions.  $\text{NH}_3$  mixing ratio also shows a clear bimodal diurnal profile during all the seasons, similar to the NO and CO diurnal profile, suggesting a significant influence of on-road traffic emissions (NO and CO) on the ambient mixing ratio of  $\text{NH}_3$  in the urban area of Delhi. During winter, the average mixing ratio of ambient  $\text{NH}_3$  was increased by 1.2–3.5 ppb in morning peak hour

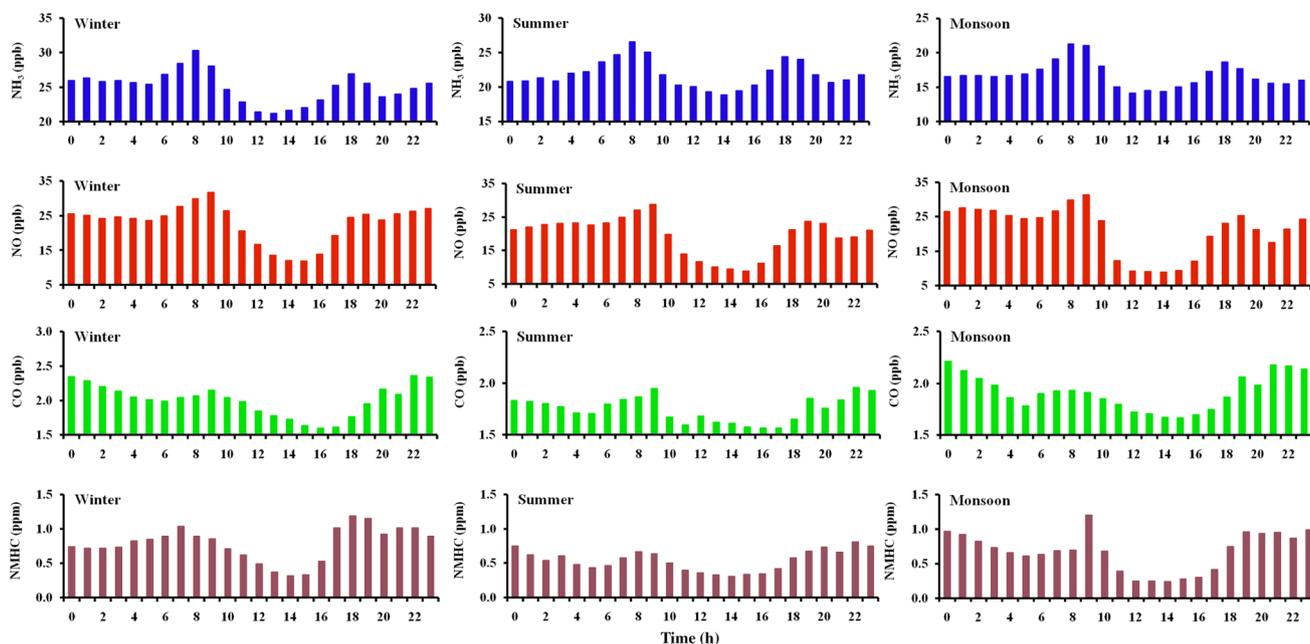
(starting at 06:00 IST), whereas increased by 0.3–1.6 ppb in evening peak hour (starting at 17:00 IST). Similarly, increase in  $\text{NH}_3$  mixing ratio was also observed during summer (morning: 1.2–2.7 ppb and evening: 1.5–1.6 ppb) and monsoon (morning: 0.4–3.6 ppb and evening: 0.9–1.4 ppb) seasons (Table 2). NO and CO are also showing the same trend during winter, summer and monsoon seasons (Fig. 2), indicating the influence in mixing ratio of ambient  $\text{NH}_3$ .

The hypothesis that traffic exhausts (NO and CO) could be the dominant source of ambient  $\text{NH}_3$  at urban site is supported by the good correlation between  $\text{NH}_3$  versus CO and NO (Meng et al. 2008; Chan and Yao 2008). The correlation

**Table 1** Average mixing ratios of NH<sub>3</sub>, NO, CO, NMHC, and CH<sub>4</sub> during winter, summer, and monsoon seasons at Delhi

Seasons	NH <sub>3</sub> (ppb)	NO (ppb)	CO (ppm)	NMHC (ppm)	CH <sub>4</sub> (ppm)
Winter	25.1 ± 2.3	22.9 ± 5.5	2.01 ± 0.23	0.79 ± 0.24	3.13 ± 0.53
Summer	21.8 ± 2.0	19.5 ± 5.7	1.75 ± 0.12	0.54 ± 0.15	3.07 ± 0.56
Monsoon	16.8 ± 1.9	21.2 ± 7.2	1.92 ± 0.17	0.68 ± 0.28	3.11 ± 0.50
Average	21.2 ± 2.1	21.2 ± 6.1	1.89 ± 0.18	0.67 ± 0.21	3.11 ± 0.53

± Standard deviation

**Fig. 2** Average diurnal variations of NH<sub>3</sub>, NO, CO, and NMHCs during winter, summer, and monsoon seasons

matrix between NH<sub>3</sub> & NO, NH<sub>3</sub> & CO, and NH<sub>3</sub> & NMHCs are summarized in Table 3 along with CH<sub>4</sub>. The correlation matrix of NH<sub>3</sub> with NO shows good correlation during winter ( $r^2=0.84$ ), summer ( $r^2=0.80$ ) and monsoon ( $r^2=0.78$ ) seasons, respectively. The positive linear correlation between NH<sub>3</sub> and NO during winter season at Beijing was also reported by Meng et al. (2011) and considered that traffic was one of major sources of ambient NH<sub>3</sub> over Beijing, China. The strong links was also observed between NH<sub>3</sub> emissions and traffic in roadside measurements in the United Kingdom (Capr et al. 2004) and Europe (Kirchner et al. 2012; Perrino et al. 2002). Ianniello et al. (2010) also reported good correlation of NH<sub>3</sub> with NO<sub>x</sub> ( $r^2=0.65$ ) and CO ( $r^2=0.67$ ) in winter season and attributed traffic as one of the major source at the observational site. However, the insignificant correlation of NH<sub>3</sub> with CO ( $r^2=0.29$ ) during monsoon season suggests that other non-traffic sources may become more important (other biological sources like human, sewage treatment, and agriculture) (Whiteland et al. 2007; Fu et al. 2013).

In order to identify the possible local sources of ambient NH<sub>3</sub> and trace gases (O<sub>3</sub>, NO, NO<sub>2</sub>, CO, SO<sub>2</sub>, and HNO<sub>3</sub>) using surface wind speed and wind direction alongwith trace gases had already been discussed in our previous papers and references therein (Sharma et al. 2014; Saraswati et al. 2018, 2019). The highest NH<sub>3</sub> mixing ratio was observed during the lowest wind speed ( $1-2 \text{ m s}^{-1}$ ) from downwind direction indicates the possible nearby sources (roadside traffic in SW, NE, and SE directions) (Saraswati et al. 2018). The higher NH<sub>3</sub> mixing ratio was recorded during winter season may also be due to the lowest wind speed which associated with local sources from SE and SW direction (Fig. S1; in supplementary information) may be due to major influence of on-road traffic.

**Table 2** Hourly changes in mixing ratios of NH<sub>3</sub>, NO, CO, NMHC, and CH<sub>4</sub> when compared with average mixing ratios at 06:00h and 17:00h

Time (in h)	NH <sub>3</sub> (ppb)	NO (ppb)	CO (ppm)	NMHCs (ppm)	CH <sub>4</sub> (ppm)
<i>Winter</i>					
06:00	26.9	24.9	2.00	0.90	3.50
07:00	1.6	2.7	0.05	0.14	0.06
08:00	3.5	5.0	0.07	0.02	0.07
09:00	1.2	6.8	0.16	0.06	-0.33
10:00	-2.2	1.6	0.05	-0.19	-0.62
17:00	25.3	19.3	1.61	1.02	2.49
18:00	1.6	5.3	0.15	0.17	0.30
19:00	0.3	6.1	0.34	0.13	0.70
<i>Summer</i>					
06:00	23.6	23.2	1.80	0.47	3.45
07:00	1.1	1.7	0.05	0.11	0.11
08:00	2.9	4.0	0.07	0.20	-0.18
09:00	1.4	5.7	0.15	0.17	-0.70
10:00	-1.8	-3.3	-0.13	0.04	-1.07
17:00	22.5	16.4	1.57	0.42	2.47
18:00	1.5	4.8	0.09	0.15	0.12
19:00	1.6	7.3	0.29	0.25	0.40
<i>Monsoon</i>					
06:00	17.6	24.8	1.90	0.63	3.55
07:00	1.5	1.9	0.02	0.06	-0.02
08:00	3.6	5.1	0.03	0.07	-0.25
09:00	3.4	6.5	0.01	0.58	-1.41
10:00	0.4	-1.0	-0.05	0.04	-1.07
17:00	15.0	19.3	1.75	0.42	2.54
18:00	1.4	3.8	0.12	0.34	0.32
19:00	0.9	6.0	0.31	0.55	0.51

**Table 3** The correlation matrix of NH<sub>3</sub> and other trace gases during winter, summer, and monsoon seasons

Seasons		NH <sub>3</sub>	NO	CO	NMHC	CH <sub>4</sub>
Winter	NH <sub>3</sub>	1				
	NO	0.84*	1			
	CO	0.44*	0.75*	1		
	NMHC	0.67*	0.75*	0.36	1	
	CH <sub>4</sub>	0.64*	0.83*	0.89*	0.62*	1
Summer	NH <sub>3</sub>	1				
	NO	0.80*	1			
	CO	0.48*	0.74*	1		
	NMHC	0.42	0.70*	0.85*	1	
	CH <sub>4</sub>	0.21	0.65*	0.71*	0.66*	1
Monsoon	NH <sub>3</sub>	1				
	NO	0.78*	1			
	CO	0.29	0.63*	1		
	NMHC	0.52*	0.81*	0.85*	1	
	CH <sub>4</sub>	0.02	0.52*	0.69*	0.46*	1

\*Significant at  $p < 0.05$

## Conclusions

Mixing ratios of ambient  $\text{NH}_3$ ,  $\text{NO}$ ,  $\text{CO}$ , NMHCs, and  $\text{CH}_4$  were measured to investigate the vehicular emissions are dominant source of atmospheric  $\text{NH}_3$  in urban site of Delhi, India, from January 2013 to December 2014. The annual average mixing ratios of  $\text{NH}_3$ ,  $\text{NO}$ ,  $\text{CO}$ , NMHCs, and  $\text{CH}_4$  were  $21.2 \pm 2.1$  ppb,  $21.2 \pm 6.1$  ppb,  $1.89 \pm 0.18$  ppm,  $0.67 \pm 0.21$  ppm, and  $3.11 \pm 0.53$  ppm, respectively. Result reveals that the mixing ratio of ambient  $\text{NH}_3$  significantly positively correlated with traffic-related pollutants ( $\text{NO}$ ,  $\text{CO}$ , and NMHCs) during all the seasons. The results emphasized that the traffic could be one of the dominant source of ambient  $\text{NH}_3$  at the urban site of Delhi, as illustrated by positive correlations of  $\text{NH}_3$  with traffic related co-pollutants. However, it is still unknown whether small fraction of  $\text{NH}_3$  emissions from vehicles in Delhi is just the result of its local traffic composition or they are generally true worldwide.

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