Measurement report: Exploring the NH_3 behaviors at urban and suburban Beijing: Comparison and implications

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ABSTRACT

Ammonia (NH_3) plays an important role in particulate matter formation, and hence its atmospheric level is relevant to human health and climate change. Due to different relative distributions of NH_3 sources, the concentrations of atmospheric NH_3 may behave differently in urban and rural areas. However, few parallel long-term observations of NH_3 to reveal the different behaviors of the NH_3 concentrations at the urban and rural sites in a same region. In this study, online ammonia analyzers were used to continuously observe atmospheric NH_3 concentrations at an urban site and a suburban site in Beijing from January 13, 2018, to January 13, 2019. The observed mixing ratio of NH_3 averaged 21 ± 14 ppb (range: 1.6–133 ppb) at the urban site and 22 ± 15 ppb (range: 0.8–199 ppb) at the suburban site. The NH_3 mixing ratios at the urban and suburban sites exhibited similar seasonal variations, with high values in summer and spring and low values in autumn and winter. The hourly mean NH_3 mixing ratios at the urban site were highly correlated ($R = 0.849$, $P < 0.01$) with those at the suburban site. However, the average diurnal variations in the NH_3 mixing ratios at the urban and suburban sites differed significantly, which implies the different contributions of NH_3 sources and sinks at the urban and suburban sites. In addition to the emission sources, meteorological factors were closely related to the changes in the NH_3 concentrations. For the same temperature (relative humidity) at the urban and suburban sites, the NH_3 mixing ratios increased with relative humidity (temperature). Relative humidity was the factor with the strongest influence on the NH_3 mixing ratio in different seasons at the two sites. The relationships between the NH_3 concentrations and temperature (relative humidity) varied from season to season and showed differences between the urban and suburban sites. The reasons for the different relationships need to be investigated in future studies. Higher wind speed mainly from the northwest sector lowered the NH_3 mixing ratios at both sites. Similar with other primary pollutants in Beijing, the NH_3 mixing ratios were high under impacts of air masses from the south sector.

Keywords: NH_3; variations; simultaneous observation

1. Introduction

Ammonia (NH_3) is the most abundant alkaline trace gas in the atmosphere (Meng et al., 2017). An excessive NH_3 concentration directly harms the ecosystem; causes water eutrophication and soil acidification; and leads to forest soil erosion, biodiversity reduction, and carbon uptake variations (Pearson and Stewart, 1993; Reay et al., 2008; Van Breemen et al., 1983; Erisman et al., 2007). NH_3 can react with acidic gases to form ammonium salts, which might significantly influence the mass concentration and composition of particulate matter (Wu et al., 2009). As major components of fine particle, ammonium salts contribute largely to the scattering of solar radiation and hence influence climate change (Charlson et al., 1991). Therefore, atmospheric NH_3 is one of the key species relevant to human health, ecosystem and climate change.

After the implementation of policies such as the *12th Five-Year Plan for the Key Regional Air Pollution Prevention and Control in Key Regions* (Ministry of Ecology and Environment of the People's Republic of China, 2012) and the *Air Pollution Prevention and Control Action Plan* (General Office of the State Council, PRC, 2013), China, especially the capital city Beijing, has been effectively controlling the emissions of sulfur dioxide (SO_2) and nitrogen oxide (NO_x), which are key precursors of fine particles. However, the pollution caused by fine particles is still serious (Krotkov et al., 2016; UN Environment, 2019), particularly in winter in the North China Plain, where excess NH_3 promote the haze formation through heterogeneous reactions (Ge et al., 2019). Studies have indicated that when the SO_2 and NO_x concentrations are reduced to a certain extent, reducing NH_3 emissions is the most economical and effective method to decrease the $PM_{2.5}$ concentration (Pinder et al., 2008). In China, the main anthropogenic sources of NH_3 are livestock and poultry feces (54%) and fertilizer volatilization (33%) (Huang et al., 2012). Moreover, the atmospheric NH_3 concentration in China has increased with the expansion of agricultural activities, control of SO_2 and NO_x, and increase in temperature (Warner et al., 2017). This increase in the NH_3 concentration might weaken the effectiveness of SO_2 and NO_x emission control in reducing $PM_{2.5}$ pollution (Fu et al., 2017).

The North China Plain is a region with high NH_3 emission (Zhang et al., 2017), and Beijing has one of the highest NH_3 concentrations in the world (Chang et al., 2016b; Pan et al., 2018). Compared with studies on pollutants such as SO_2 and NO_x, considerably fewer studies have been conducted on the NH_3 concentration in Beijing. Chang et al. (2016a) collected gaseous NH_3 samples during the 2014 APEC summit (October 18 to November 29, 2014) in the Beijing urban area and concluded that the overall contributions of traffic, garbage, livestock, and fertilizers to the NH_3 concentration were 20.4%, 25.9%, 24.0%, and 29.7%, respectively. According the data from Huang et al (2012), the NH_3 emissions in Beijing were from livestock and poultry farming (34.55%), nitrogen-fixing plants (33.57%), fertilizer use (13.06%), household garbage treatment (8.29%), traffic emissions (5.20%), industrial emissions (0.14%), biomass combustion (0.42%), and agricultural soil (0.84%). Zhang (2016) measured the NH_3 concentrations in urban and rural areas of Beijing from January to July 2014 and found that NH_3 concentration in urban areas was approximately 65% higher than that in rural areas. Meng et al. (2011) reported that the highest NH_3 concentration in Beijing occurred in summer and the lowest one occurred in winter, and their results indicated traffic to be a significant source of NH_3 in urban areas. Zhang et al. (2018) reported the vertical variability of NH_3 in urban Beijing based on one-year passive sampling in 2016/2017 and concluded that local sources such as traffic emissions were important contributors to urban NH_3. Meng *et al*. (2020) investigated the significant increase in winter NH_3 and its contribution to the increasing nitrate in $PM_{2.5}$ from 2009 to 2016, and they also concluded that vehicles exhaust was an important contributor to NH_3 in urban Beijing in winter.

Currently, NH_3 is not included in the routine environmental monitoring operation in China. Research data on NH_3 monitoring, particularly on the synchronous observations of NH_3 concentrations with a high temporal resolution in urban and suburban areas, are relatively scarce. In this study, high-time-resolution observations of NH_3 were obtained simultaneously at an urban site and a suburban site in Beijing. The variation characteristics and influencing factors of the NH_3 concentration were analyzed with meteorological data to provide a scientific basis for NH_3 pollution control in Beijing.

2. Materials and methods

2.1. Measurement sites

From January 2018 to January 2019, continuous and simultaneous observations of atmospheric NH_3 were conducted at an urban site and a suburban site in Beijing. The urban site was located on the roof of the Science and Technology Building of Minzu University of China (39.95°N, 116.32°E, altitude: 102 m) and the suburban site was in the Changping Meteorological Station (40°13′N, 116°13′E, altitude: 77 m). The suburban site is in the NW direction relative to the urban site and the shortest distance between these two sites is approximately 32 km (Figure 1). More farm land and glass land are around the suburban site than the urban site.

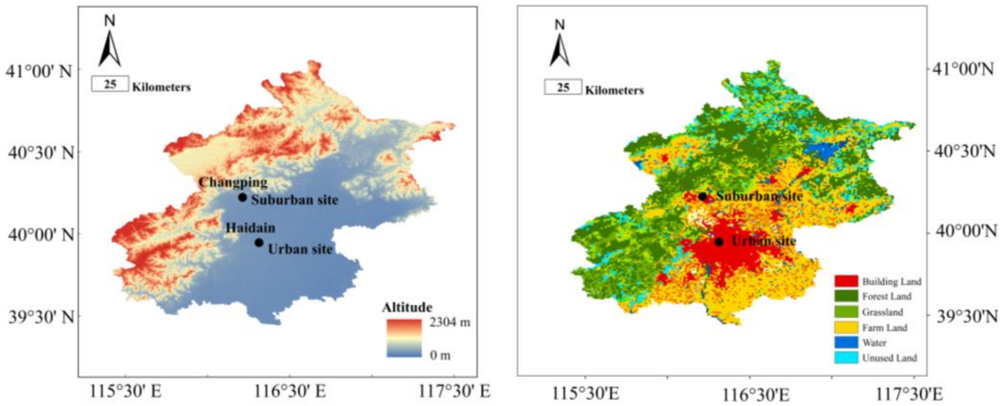

Fig. 1. Location of the observation sites, the topography, and land use of Beijing city.

2.2. Measurements and data acquisition

NH_3 concentrations were measured using two NH_3 analyzers (Ammonia Analyzer-Economical, Los Gatos Research Inc., USA), which have the minimum detection limit of <0.2 ppb and the maximum drift of 0.2 ppb/24hrs. The NH_3 analyzers were deployed in air-conditioning rooms. These analyzers use off-axis integrated cavity output spectroscopy (OA-ICOS) technology, which is a fourth-generation cavity-enhanced absorption technique, to simultaneously measure NH_3 and water vapor (H_2O) in the atmosphere. The incident laser beam of the OA-ICOS technology deviates from the optical axis, which differs from the traditional coaxial incidence mode. The axial incidence mode of the OA-ICOS technology can increase the optical path, stimulate additional high-order transverse modes, effectively suppress the noise of the cavity mode, reduce the cross interferences and errors due to contaminants existing in the cavity, and improve the detection sensitivity (Baer *et al*., 2002; Baer *et al*., 2012). The analyzer method is a quasi-absolute measurement, which theoretically does not require calibration. However, to ensure the comparability of the obtained data with other monitoring data, NH_3 standard gas (Beijing AP BAIF Gases Industry Co., Ltd.) was used for comparison measurement before the observation. The recorded concentrations were revised with respect to the reference NH_3 concentration in the standard gas mixture.

Ambient air was drained at >0.4 L/min through Teflon lines (1/4'OD) from a manifold. The lengths of the Teflon lines were designed as short as possible (less than 2 m from the manifold). Particulate matters were filtered by Teflon membranes with a pore size less than 5 μm. Since NH_3 easily "sticks" to surfaces (like inside walls of tubes), heated sample lines were suggested by many measurement studies. However, according our test (Fig. S1) in the lab, when heating (70℃) was on, there did have a peak lasting 5–6 min minutes and then deceasing to the normal levels in ambient air, which means a new balancing process has been established in less than 10 min. This suggests that heating is not necessarily a solution for NH_3 sticking. Keeping the relatively stable balance between adsorption and desorption of NH_3 in the sampling system is important. When tested using air of different humidity, only very sharply changes of humidity obviously influenced and changed the balance, and a new balance needed tens of minutes to reestablished (Fig. S2). Under the normal weather conditions, humidity changes in a relatively smoothing way unless a quickly changing weather system, like rain, is approaching. The minute-level data were converted into hourly averages in the data analysis process and the hourly resolution can smooth the effect to some extent caused by variations in humidity and temperature during the observation.

The balancing idea was also used to carry out multi-point calibrations on NH_3 analyzers (Fig. S3). A high mixing ratio (e.g. 400 ppb or higher) of NH_3 mixing gases were firstly produced by a dynamic diluter and measured by the NH_3 analyzers overnight. After the signals reached the stable level, other lower span values were switched in turn. At each span point, the measurement time was lasting at least 40 minutes or longer. Then a linear regression function was obtained with R^2 higher than 0.999. Nowadays, NH_3 in compressed gas cylinder is also trustworthy, as confirmed by the comparison with the NH_3 standard in a permeation tube (Fig. S4).

Totally, 7645 and 8342 valid hourly mean observations were obtained for the urban (Haidian) and suburban (Changping) sites, respectively. In addition, the urban and suburban meteorological data (temperature, relative humidity, wind direction, and wind speed) during the sampling period were obtained from the Haidian Meteorological Observation Station and Changping Meteorological Station, respectively.

3. Results and discussion

3.1. Overall variations in the NH_3 mixing ratios

Fig. 2 displays the time-series variations in the NH_3 mixing ratios, temperatures, and relative humidity at the urban and suburban sites in Beijing. At the urban site, the mean ± 1σ, median, maximum, and minimum values of the hourly average NH_3 mixing ratio during the observation period were 21 ± 14 ppb, 17 ppb, 133 ppb and 1.6 ppb, respectively. At the suburban site, the corresponding values were 22 ± 15 ppb, 18 ppb, 199 ppb, and 0.8 ppb, respectively. The annual average and range of the NH_3 mixing ratio at the suburban site were marginally higher than those at the urban site. The characteristics of the weekly smoothed data indicate that the NH_3 variations and temperature/humidity fluctuations at the two sites were practically consistent, which suggests that both sites were under the influence of similar weather systems. The hourly mean NH_3 concentrations at the urban site were significantly correlated ($R = 0.849$, $P < 0.01$) with those at the suburban site.

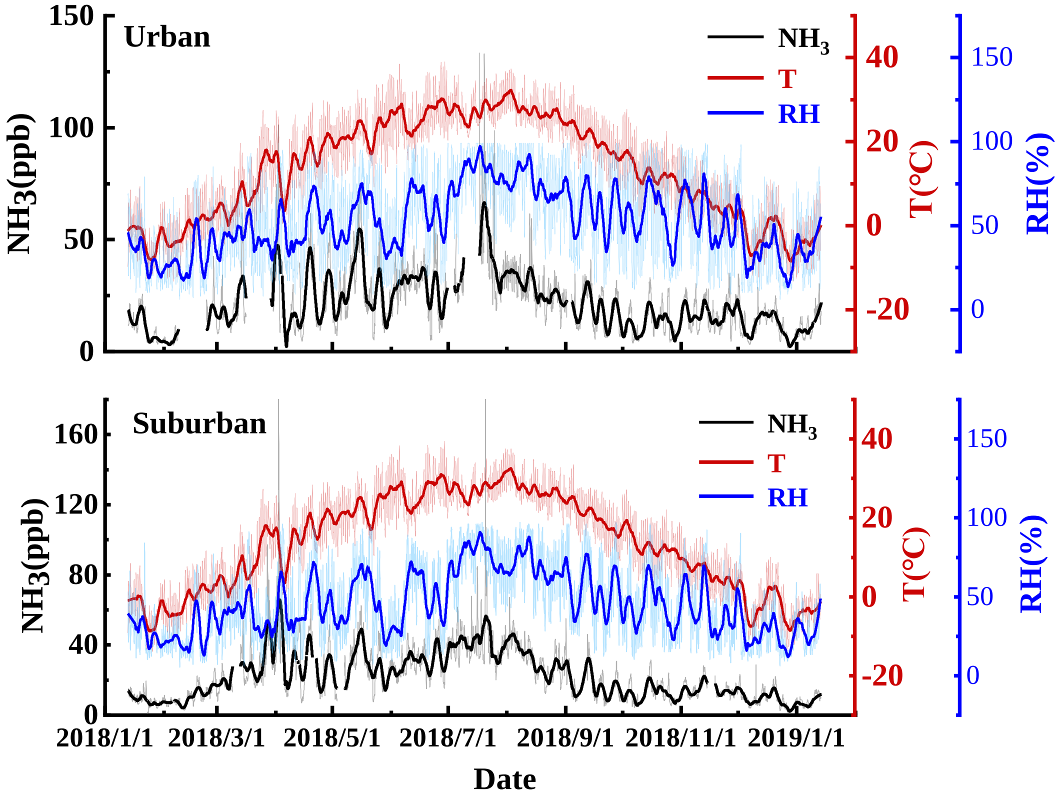

Fig. 2. Temporal variations in the hourly average NH_3 mixing ratios, temperatures (*T*) and relative humidity (*RH*) at the urban and suburban stations in Beijing. Continuous thick lines were smoothed with 168 points (7 days) by using the Savitzky–Golay method.

Table 1 shows the comparison of atmospheric NH_3 concentrations (ppb) observed in different areas.

Meng et al. (2011) obtained an average NH_3 mixing ratio of 22.8 ± 16.3 ppb for the period 2008-2010 in Beijing urban area, which is very close to our result (21 ± 14 ppb) for 2018-2019. Therefore, the annual average NH_3 mixing ratio in urban Beijing did not change significantly from 2008 to 2019. Moreover, results from this study and Meng et al. (2011) indicate that the NH_3 concentrations at the urban and suburban sites were higher than those in the background areas. The observed NH_3 concentrations in Beijing were higher than those in northwest China (Meng et al., 2010) and the Yangtze River Delta region (Chang et al., 2019). The average annual NH_3 concentration in the urban area of Shanghai, a mega city in the Southeast China (31° N), was approximately 50% lower than that in urban Beijing. This might be related to the fact that the North China Plain, in which Beijing is located, is one of the most intensive agricultural production regions in China. The differences in the soil properties of Beijing and Shanghai may be another reason because the loss of soil NH_3 can increase with an increase in the soil pH (Ju et al., 2009). Shanghai and its surrounding areas are dominated by acidic soil of paddy fields (Zhao et al., 2009), whereas Beijing is dominated by the alkaline soils of dry land (Wei et al., 2013). In addition, the climate in Beijing is much drier than in Shanghai so that less atmospheric NH_3 in Beijing can be removed than in Shanghai by wet deposition.

Table 1. Comparison of the atmospheric NH_3 concentrations (ppb) observed in different areas.

Period	Location	Methodology	Types	Concentration	Reference
2018.01-2019.01	Beijing, CN	Online monitor	Urban	20.8±13.7	This study
			Suburban	21.9±14.9	
2008.02-2010.07	Beijing, CN	Passive sampler	Urban	22.8±16.3	Meng et al., 2011
2007.01-2010.07			Background	10.2±10.8	
2014.5-2015.6	Shanghai, CN	Passive sampler	Urban	7.8	Chang et al. 2019
			Suburban	6.8	
2006.04-2007.04	Xi'an, CN	Passive sampler	Urban	18.6	Cao et al. 2009
			Suburban	20.3	
2017.12-2018.2	Hebei, CN	Online monitor	Rural	16.7±19.7	He et al. 2020
2008	Qinghai, CN	Passive sampler	Rural	4.1±2.2	Meng et al. 2010

2003.7-2011.9	Toronto, CA	Passive sampler	Urban	2.3-3.0	Hu et al. 2014
			Rural	0.1-4	
2016.4-2017.10	New York, US	Active and passive system	Urban	2.2-3.2	Zhou et al. 2019
			Rural	0.6-0.8	
2017.12	Tokyo, JP	semi-continuous microflow analytical system	Urban	4.1	Osada et al. 2019
2013.1-2015.12	Delhi, IN	Automatic analyzer	Urban	53.4±14.9	Saraswati et al., 2019
2012.10-2013.9	Jaunpur, IN	Glass flask sampling	Suburban	51.6±22.8	Singh and Kulshrestha, 2014
2008.1-2009.2	Bamako, MLI	Passive sampler	Urban	46.7	Adon et al., 2016
2006.3-2017.4	Edmonton, CA	Online monitor	Urban	2.4±0.6	Yao et al., 2016
2010.9-2011.8	Seoul, KR	Online monitor	Urban	10.9±4.25	Phan et al., 2013
2004.3-2004.7	Munster, DE	Wet denuder	Urban	5.2	Vogt et al., 2005

Table 1 also shows observational results of atmospheric NH_3 from some other countries. The NH_3 mixing ratios in the United States (Edgerton et al., 2007; Nowak et al., 2006; Zhou et al. 2019), Scotland (Burkhardt et al., 1998), Canada (Hu et al., 2014), Japan (Osada et al., 2019), and Germany (Vogt et al., 2005) were 0.23–13 ppb, 1.6–2.3 ppb, 0.1–4 ppb, 4.1 ppb, and 5.2 ppb, respectively. These values are considerably lower than those in Beijing. However, Delhi, India (Saraswati et al., 2019), exhibited higher NH_3 mixing ratio (53.4±14.9 ppb) than Beijing did. This result might be attributed to the well-developed livestock breeding activities in Delhi. This comparison indicates that in the decade before 2019, the NH_3 concentration in Beijing did not change considerably, but it is of the highest in big cities in China and much higher than those observed in developed countries in America, Europe and Asia.

3.2. Seasonal variations

Fig. 3 displays the monthly statistics for the NH_3 mixing ratios at the urban and suburban sites in Beijing. The seasonal variations in the NH_3 mixing ratios were very similar at the urban and suburban sites, with higher mixing ratios in the spring and summer and low ones in the autumn and winter. The daily mean concentrations fluctuated considerably in the spring, particularly in April. The highest mean

NH_3 concentrations at the urban and suburban sites were 42± 17 ppb and 42 ± 8.2 ppb, respectively. Both occurred in July, when the NH_3 concentrations fluctuated considerably as well. On average, the seasonal NH_3 mixing ratios at the urban and suburban sites can be arranged as follows: summer > spring > autumn > winter. The main grain crops in the rural area of Beijing are corn and wheat. Corn is categorized as spring corn and summer corn, which are sown in April and June, respectively. Usually, a large amount of base fertilizer is applied when planting corn and the topdressing after 2 months. Wheat is sown from September to October, and the topdressing is applied in the following spring. The volatilization of nitrogen fertilizers can cause an increase in atmospheric NH_3 mixing ratios and its fluctuations in fertilization seasons (Zhang et al., 2016). In addition, the high temperature in summer should also be responsible to the high NH_3 mixing ratios in this season. An increase in the temperature can increase the biological activity and thus enhance the NH_3 production and emission. High temperature is also conducive for the volatilization of the urea and diammonium phosphate applied to crops. Moreover, the equilibrium among ammonium nitrate particles, gaseous NH_3, and nitric acid is transferred to the gas phase at high temperature, which increases the NH_3 concentration (Behera et al., 2013). Sewage treatment, household garbage, golf courses, and human excreta are crucial NH_3 sources that are easily neglected (Pu et al., 2020). The relatively low NH_3 concentrations in the autumn and winter might be caused by the decrease in NH_3 emission in the soil and vegetation, the decrease in the NH_4NO_3 decomposition capacity at low temperatures, and the reduced human activities caused by a large floating population returning to their native locations outside Beijing during the Spring Festival (Liao et al., 2014). In the spring and summer, the NH_3 mixing ratios at the suburban site were higher than those at the urban site, which might be related to the higher agricultural activity around the suburban site. In the autumn and winter, the NH_3 mixing ratios at the urban site were marginally higher than those at the suburban site. In the autumn and winter seasons, the influences of agricultural activities on the NH_3 concentration were weakened, whereas the influences of other sources (such as traffic sources) were enhanced. According to Wang et al. (2019), the traffic NH_3 emission per unit area in Haidian (urban site) was three times higher than that in Changping (suburban site). This difference in traffic source emissions might have resulted in higher NH_3 concentrations at the urban site than at the suburban site in the autumn and winter.

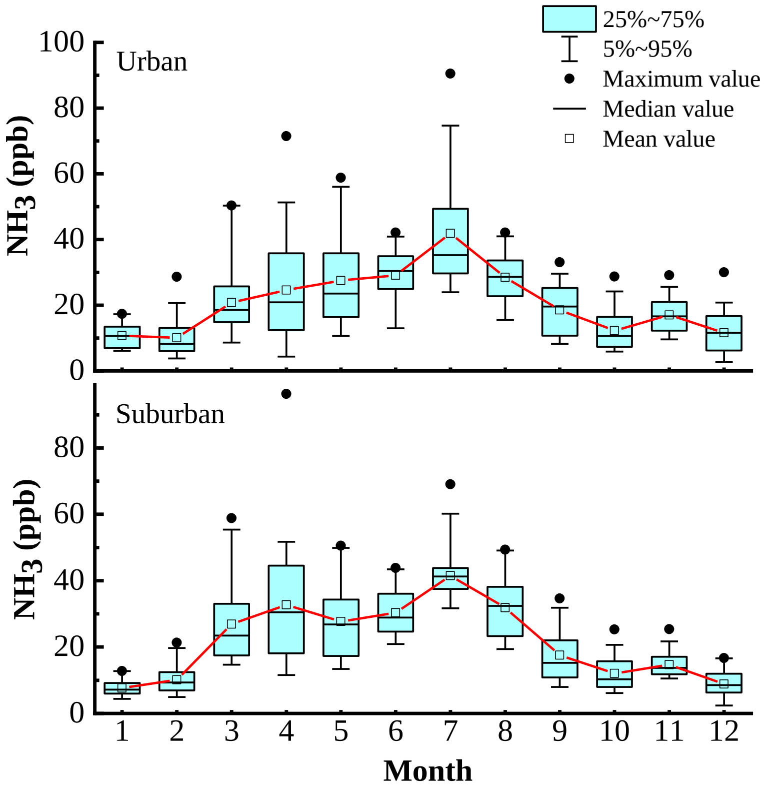

Fig. 3. Monthly statistical variation in the NH_3 mixing ratios at the urban and suburban sites in Beijing.

Table 2. NH_3 mixing ratios (ppb) measured at the urban and suburban sites in Beijing.

Site	Time period	Mean	Standard deviation	Minimum	Median	Maximum
Urban	Annual	21	14	1.6	17	133
	Spring	25	16	1.9	21	101
	Summer	32	12	5.0	30	133
	Autumn	16	7.5	3.8	15	41

	Winter	11	6.7	1.6	9.9	42
	Annual	22	15	0.8	18	198
	Spring	29	16	6.8	26	180
Suburban	Summer	35	12	12.1	33	199
	Autumn	15	6.8	4.1	13	55
	Winter	9.2	4.5	0.8	8.4	29

3.3. Diurnal variations

Figure 4 displays the average diurnal variations in the NH_3 and H_2O mixing ratios in different seasons at the urban and suburban sites in Beijing. Ambient NH_3 exhibited different diurnal behaviors in different seasons.

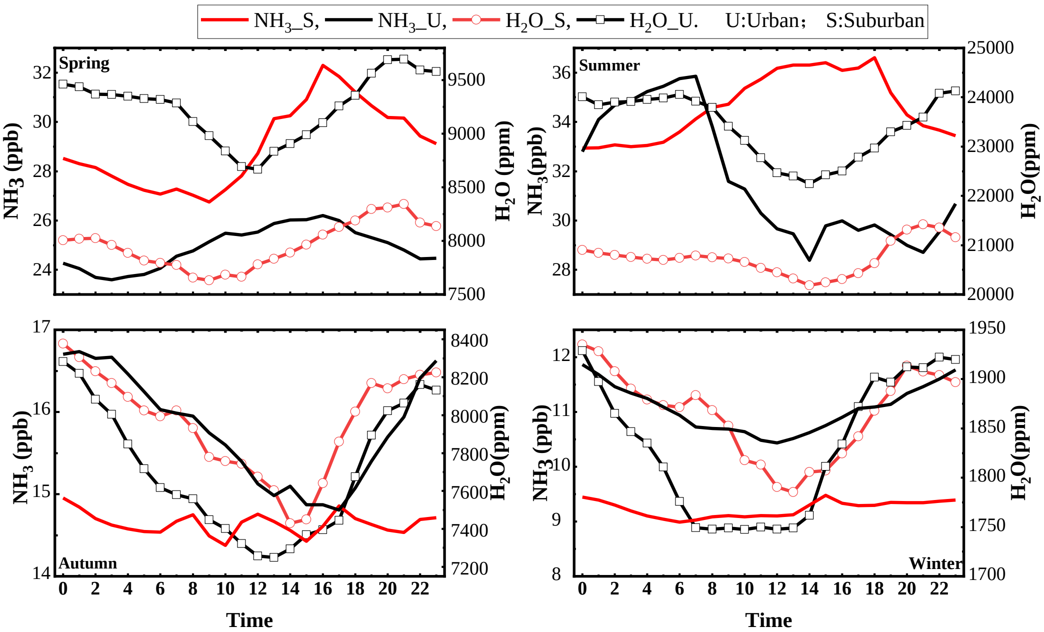

Fig. 4. Average diurnal variations in the NH_3 and H_2O mixing ratios in different seasons at the urban and suburban sites in Beijing.

In spring, the average diurnal variations in the NH_3 mixing ratio were similar at the urban and suburban sites. The diurnal variations exhibited a single-peak pattern with high values in the daytime and low values at night. The NH_3 mixing ratio began to increase in the morning, reached its maximum value at 16:00, and then decreased gradually. The lowest mixing ratios at the urban and suburban sites occurred at 03:00 and 09:00, respectively. The NH_3 mixing ratio began to increase earlier at the urban site than at the suburban site. A plausible explanation to the earlier increase in the NH_3 emission at the urban site is the traffic emission in the morning rush hours. In spring, the mixing ratio of NH_3 was higher at the suburban site than that at the urban site, with an average difference of 4.1 ppb and a maximum difference of 6.1 ppb. The average diurnal amplitude of the NH_3 mixing ratio at the suburban site was 5.3 ppb, which was higher than that (2.6 ppb) at the urban site. At the urban site, the average diurnal variations in the NH_3 and H_2O mixing ratios exhibited nearly opposite trends. The H_2O mixing ratio had high values in the night and low values in the day. At the suburban site, the variation characteristics of NH_3 and H_2O were very similar; however, the peak NH_3 concentration occurred 5 hours earlier than the peak H_2O concentration. In spring, in contrast to the NH_3 mixing ratio, the H_2O mixing ratio at the urban site was 1279 ppm higher than that at the suburban site.

The diurnal variation in the NH_3 mixing ratio at the suburban site in summer was similar to that in spring. This phenomenon was also observed in the rural areas of Shanghai by Chang et al. (2019). The diurnal variations of NH_3 at the suburban site were considerably affected by the temperature and the contribution from volatile NH_3 sources. However, the diurnal summer variation of NH_3 at the urban site was completely different from that at the suburban site. The summer level of NH_3 at the urban site was obviously lower during the daytime and evening than that at the suburban site, increased gradually from 21:00 to levels higher its suburban counterpart, dropped after reaching its peak value at 7:00, and then reached its lowest value at 14:00. The diurnal pattern (with a peak in early morning) has been observed in other areas, such as rural (Ellis et al., 2011), urban (Gong et al., 2011), and steppe areas located far away from human activity (Wentworth et al., 2016). Kuang et al. (2020) believed that such diurnal pattern of NH_3 was caused by the evaporation of dew in the morning, which resulted in the release of NH_3 originally stored in the droplets. A lag was observed between the changes in the NH_3 and H_2O concentrations in the early morning, which supported the hypothesis of Kuang et al (2020). In addition, the increase in the NH_3 concentration in the morning might have been caused by the breakup of the boundary layer formed at night. The downward mixing of air with a higher NH_3 concentration in the residual layer led to a morning increase in the NH_3 concentration on the ground (Bash et al., 2010). In summer, the NH_3 concentrations at the suburban site were significantly higher than those at the urban site during the daytime and first half of the night. The average diurnal amplitude of the NH_3 concentration was 7.5 ppb and 3.7 ppb at the urban and suburban sites, respectively. Similar to the situation in spring, the H_2O concentrations at the urban site were significantly higher than those at the suburban site in the summer.

In autumn, the NH_3 concentration at the suburban site was relatively stable and remained nearly all the time lower than that at the urban site, which showed low values during the day and high values during the night, with a peak at midnight and a minimum (about 2.0 ppb lower than the peak) at 17:00. The H_2O concentration was marginally lower (250 ppm) at the urban site than at the suburban site. The diurnal profiles of H_2O at both sites resemble that of NH_3 at the urban site, but the lowest values of H_2O occurred earlier than the lowest value of NH_3 at the urban site.

The diurnal patterns of NH_3 and H_2O in winter were similar to those in autumn though the mixing ratios of NH_3 and H_2O were lower than their autumn counterparts. There were two slight differences: (1) the mixing ratios of NH_3 at both sites exhibited lower fluctuations than those in autumn and (2) the mixing ratio of NH_3 at the urban site reached its minimum in winter earlier than that in autumn.

The above results indicate that although the two sites were under the influence of similar weather systems, the diurnal variations in the NH_3 mixing ratios at the two sites were different in different seasons. This finding suggests that different NH_3 sources and possibly sinks had different contributions to the NH_3 concentrations at the urban and suburban sites. Additional studies should be conducted to better understand the behaviors of atmospheric NH_3 and its influencing factors.

3.4. Effect of meteorological factors on the NH_3 levels

Table 3 presents the annual and seasonal correlation coefficients between the daily means of NH_3 mixing ratios and those of the temperature, relative humidity, and wind speed at the two sites. Annually, the NH_3 mixing ratios at both sites were positively correlated with temperature and relative humidity and negatively correlated with wind speed, and the correlations are all highly significant. However, the correlations deteriorated somewhat in warm seasons. In summer and autumn, no significant correlations were noted between ambient NH_3 and temperature at the two sites. The correlation between NH_3 and wind speed in summer was much weaker than in the other seasons. The relative humidity was stronger correlated with the NH_3 concentration at the two sites than temperature, which can be perceived in Fig 2. Also, the correlation between NH_3 and relative humidity did not vary much from season to season. This implies a possibility that relative humidity exerts a certain influence on the variation of the NH_3 level in the surface layer.

Table 3. Correlations between the daily mean values of NH_3 and meteorological elements (Spearman's rank correlation coefficient)

Site	Time Period	Temperature	Relative humidity	Wind speed
	Annual	0.680**	0.706**	-0.370**
	Spring	0.450**	0.645**	-0.540**
Urban	Summer	0.043	0.488**	-0.106**
	Autumn	0.101	0.759**	-0.413**
	Winter	0.596**	0.690**	-0.449**

	Annual	0.745**	0.730**	-0.325**
	Spring	0.256*	0.518**	-0.391**
Suburban	Summer	0.126	0.576**	-0.061**
	Autumn	0.135	0.792**	-0.618**
	Winter	0.676**	0.663**	-0.545**

*: at the 0.05 significant level; **: at the 0.01 significant level.

Fig. 5 displays the seasonal mean diurnal variations in the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites, with their correlation coefficients shown in Fig. S5. At the urban site, the seasonal-hourly means of the NH_3 mixing ratio were positively (negatively) correlated with those of temperature (relative humidity) in spring, but the correlations were reversed in the other seasons. At the suburban site, the seasonal-hourly means of the NH_3 mixing ratio were positively (negatively) correlated with those of temperature (relative humidity) in the spring and summer, but less correlated in autumn and winter. Similar correlation behaviors (diurnal variations) were found at both sites in spring, but in other seasons the correlations (diurnal variations) at the urban site behaved differently from those at the suburban site. The inconsistent behaviors in summer, autumn and winter caused urban-suburban differences in the annual-diurnal patterns of NH_3, temperature and relative humidity as well as the NH_3-temperature (relative humidity) correlations, as can be seen in Fig. S6. Figure 6 displays the contour maps of the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites. The annual contour maps are shown in Fig. S7. As shown in these contour maps, the NH_3 mixing ratios at both sites increased with relative humidity at same temperature and increased with temperature at same relative humidity. Although there are some scatterings in the contour maps, high NH_3 levels are generally associated with high temperature and humidity. In winter, when air temperature was low (< 0 °C), the NH_3 mixing ratios at both sites often had low values except in high humidity (>60%). An increase in temperature caused higher NH_3 mixing ratios at both sites; however, the NH_3 concentration at the suburban site was more significantly correlated with temperature than that at the urban site (Table 3), suggesting that volatile NH_3 sources might have a higher contribution to the NH_3 concentration in suburban than in urban area. A higher amount of NH_3 removal through chemical transformation is expected during the day at the urban site than at the suburban site because the urban area had higher relative humidity and amounts of particulate matters, and higher emissions of acid gases (particularly NO_x) than the suburban area. In 2018, the concentrations of $PM_{2.5}$, SO_2 and NO_2 were 50 μg/m^3, 5 μg/m^3, 43μg/m^3 in Haidian, and 46 μg/m^3, 6 μg/m^3, 35 μg/m^3 in Changping, respectively, as reported by Beijing Ecology and Environment Statement.

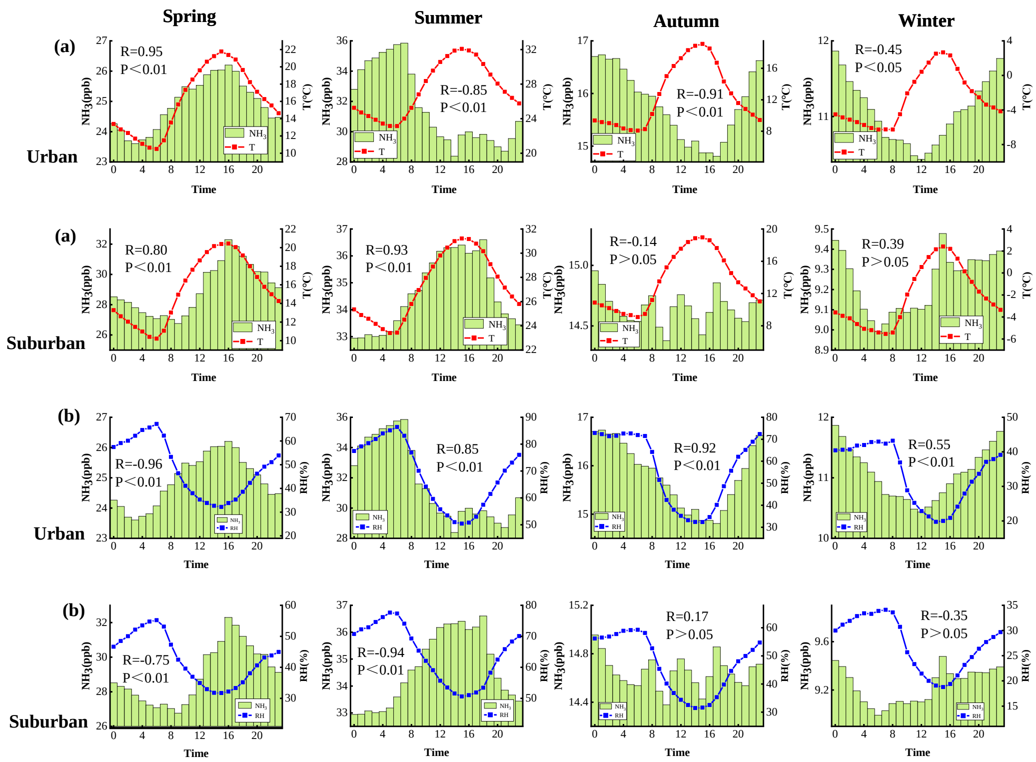

Fig. 5. Diurnal variations in and correlation coefficients between the NH_3 mixing ratios and temperature (a), relative humidity (b) in different seasons at the urban and suburban sites in Beijing.

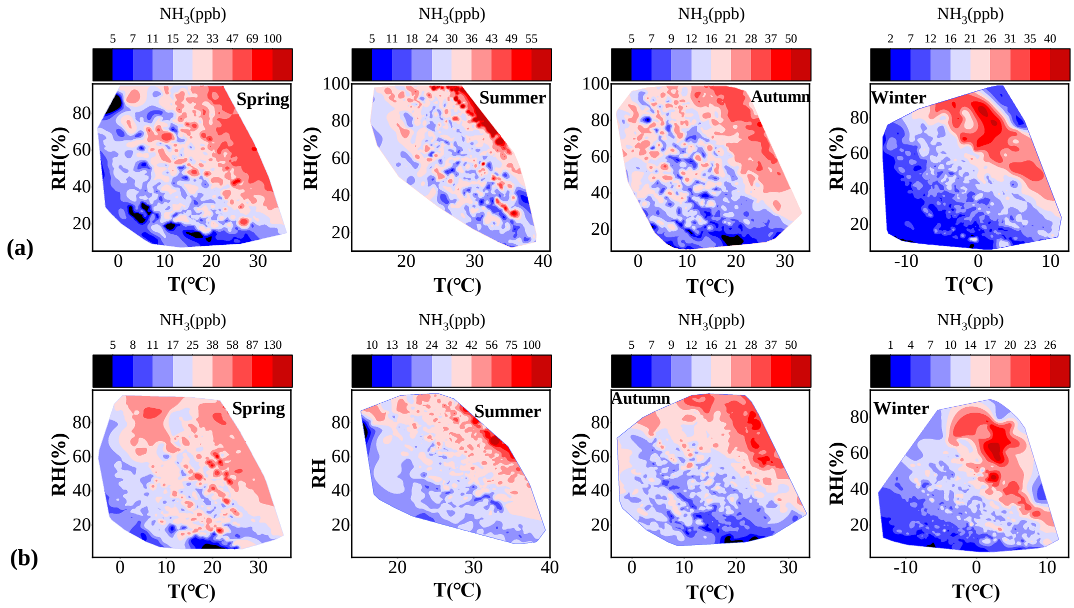

Fig. 6. Contour maps of the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites in Beijing (a: Urban, b: Suburban).

To explore the influence of wind on the NH_3 mixing ratios, rose charts were drawn for the hourly mean concentration of NH_3, wind direction frequency, and wind speed during the observation period (Fig. 7). The large-scale wind circulation in the North China Plain is often influenced by the mountain-plain topography; therefore, the dominant winds in this region are southerly (from noon to midnight) and northerly (from midnight to noon) (Lin et al., 2009; Lin et al., 2011). As displayed in Fig. 7, some differences existed in the distributions of the surface wind between the urban and suburban sites. The prevailing surface winds were northeasterly and southwesterly at the urban site and northwesterly and easterly at the suburban site. At the urban site, the NH_3 mixing ratios were relatively high when the winds originated from the southern sectors and relatively low when the winds originated from the northwest sectors. Therefore, under southwest wind, air masses from the south of Beijing carry not only air pollutants but also higher levels of NH_3 to the urban site. Meng et al. (2017) examined the effect of longrange air transport on the urban NH_3 levels in Beijing during the summer through trajectory analysis. They concluded that the air mass from the southeast has a cumulative effect on the NH_3 concentration. Although the dominant wind direction at the suburban site was different from that at the urban site, the NH_3 mixing ratios were also relatively high in the south sectors. Thus, winds from the southeast, south, and southwest can elevate levels of atmospheric NH_3 at both the urban and suburban sites. The NH_3 mixing ratios were relatively low when air masses originated from the northwest sector at urban site and from the west sector at the suburban site. The west and northwest winds were stronger and promoted the dilution and diffusion of NH_3 emitted into the boundary layer.

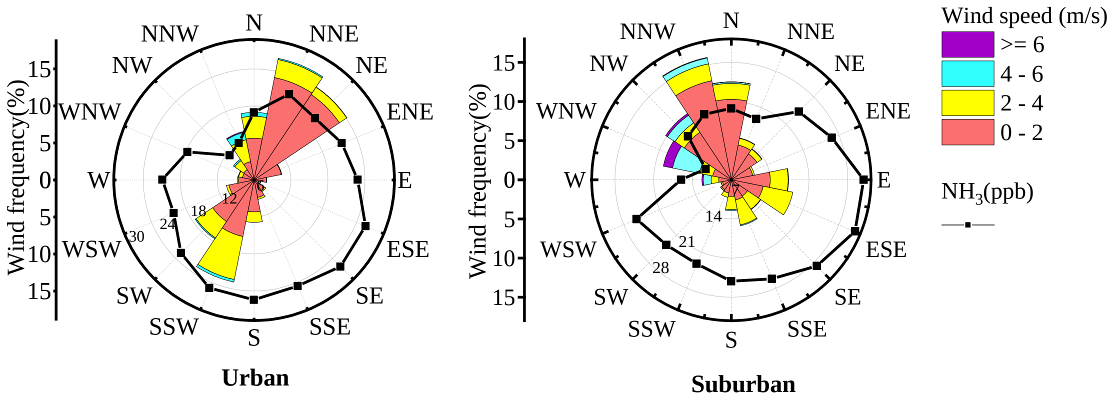

Fig. 7. Rose maps of the NH_3 mixing ratios, wind frequency, and wind speed in different wind direction sectors.

As a water-soluble gas, NH_3 can be impacted by precipitation. Heavy rainfall occurred on August 18, 2018 (Fig. 8). Before the rainfall, the NH_3 concentration at the urban site was higher than the average level in August. After the rainfall, the NH_3 concentration decreased rapidly, and it was significantly lower than the mean value in August. However, the diurnal pattern of NH_3 on that day did not differ considerably from the average diurnal pattern in August. On the same day, the NH_3 mixing ratio at the suburban site remained at a low level during the rainfall period, which was considerably lower than the August mean NH_3 concentration during the same time of day. However, the NH_3 mixing ratio increased rapidly after the precipitation and reached the mean level at 17:00. The rainfall might have an obvious clearing effect on NH_3 but more case studies are needed to reach a robust conclusion.

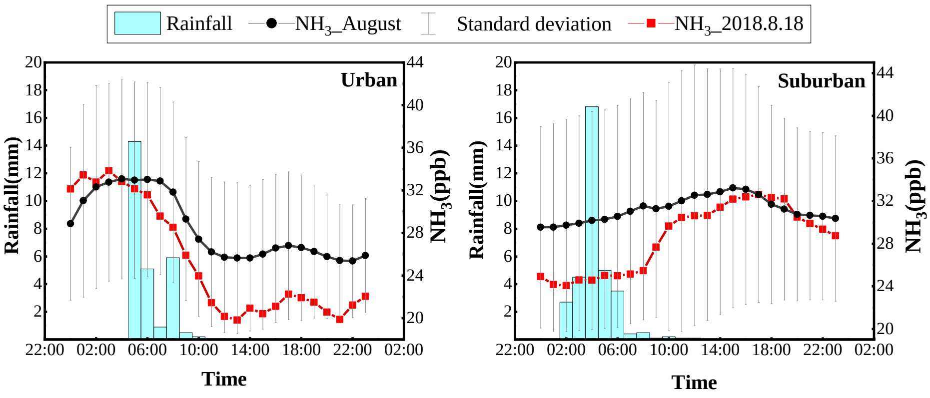

Fig. 8. Diurnal variations in the rainfall and NH_3 concentration on August 18, 2018.

4. Conclusions

In this study, the atmospheric NH_3 concentrations at an urban site and a suburban site in Beijing were continuously and simultaneously observed from January 2018 to January 2019. The mean NH_3 mixing ratios were 21 ± 14 ppb and 22 ± 15 ppb at the urban and suburban sites, respectively. These NH_3 levels are among the highest mean values found in China and much higher than those reported for some developed countries in America, Europe and Asia. In the summer and spring, the NH_3 mixing ratios at the suburban site were slightly higher than those at the urban site. In the autumn and winter, however, the situation was reversed. The highest NH_3 mixing ratios at the urban and suburban sites were all found in July. The lowest NH_3 mixing ratio occurred in February at the urban site and in January at the suburban site. A comparison with data from literature shows that the mean concentration of NH_3 in Beijing did not change considerably in the decade before 2019.

The hourly mean NH_3 mixing ratios at the urban site were highly correlated (R = 0.849, P < 0.01) with those at the suburban site. However, the mean diurnal variations in the NH_3 mixing ratios at the urban and suburban sites were different. At the urban site, lower NH_3 mixing ratios were observed in the daytime and higher ones at night. The opposite trend was observed at the suburban site. Although both sites were under the influence of similar weather systems, the seasonal-diurnal variations in the NH_3 mixing ratio were different at the urban and suburban sites, suggesting that NH_3 sources had different relative contributions to the NH_3 levels at the urban and suburban sites.

The relationship of meteorological factors with the NH_3 mixing ratio was complex. Overall, the NH_3 mixing ratios increased with relative humidity and temperate at both sites. Relative humidity was stronger correlated with the NH_3 mixing ratio at both sites. The situation in different seasons varied and was site-dependent, which warrants further studies. A high wind speed (mainly under northwesterly) suppressed the levels of NH_3 at both sites. The NH_3 mixing ratios were higher under southerly wind conditions. Rainfall had a certain scavenging effect on NH_3 but had little effect on the diurnal variations in the NH_3 concentration.

Data availability. The data of stationary measurements are available upon request to the contact author Weili Lin (linwl@muc.edu.cn).

Author contributions. ZL and WL developed the idea for this paper, formulated the research goals, and carried out the measurement at urban site. WP and ZM carried out the NH_3 field observations at the suburban site.

Competing interests. The authors declare that they have no conflict of interest.

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