https://doi.org/10.5194/acp-2020-1047
Preprint. Discussion started: 2 November 2020

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

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Abstract. Ammonia (NH_3) plays an important role in particulate matter formation; however, few long-term observations with a high temporal resolution have been conducted on the NH_3 concentrations in Beijing. In this study, online ammonia analyzers were used to observe continuously the atmospheric NH_3 concentrations at an urban site and a suburban site in Beijing from January 13, 2018, to January 13, 2019. The average mixing ratio of NH_3 at the urban site was 21 ± 14 ppb (range: 1.6–133 ppb) and that at the suburban site was 22 ± 15 ppb (range: 0.8–199 ppb). The NH_3 mixing ratios at the urban and suburban sites exhibited similar seasonal variations, with high values being observed in the summer and spring and low values being observed in the 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 indicated 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 the relative humidity (temperature). The relative humidity was the factor with the strongest influence on the NH_3 mixing ratio in different seasons at the two sites. In general, a high wind speed promoted a reduction in the NH_3 mixing ratio. Similar with other primary pollutants in Beijing, the NH_3 mixing ratios were high when winds originated from the south and low when winds originated from the north and northwest.

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). Thus, the NH_3 concentration influences climate change (Charlson et al., 1991; 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). 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

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the *Air Pollution Prevention and Control Action Plan* (General Office of the State Council, PRC, 2013), China, especially for Beijing, has been effectively controlling the emission of primary pollutants, such as sulfur dioxide (SO_2) and nitrogen oxide (NO_x); however, the pollution caused by fine particles is still serious (Krotkov et al., 2016; UN Environment, 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. 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 samples 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 observation of the 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 the atmospheric NH_3 concentration were conducted in an urban area and a suburban in Beijing. The urban site was located on the roof of the Science and Technology Building of Minzu University of China (referred to as the urban site, 39°95′N, 116°32′E, altitude: 102 m) and the suburban

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site was in the Changping Meteorological Station (referred to as the suburban site, 40°13′N, 116°13′E, altitude: 77 m), respectively. 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).

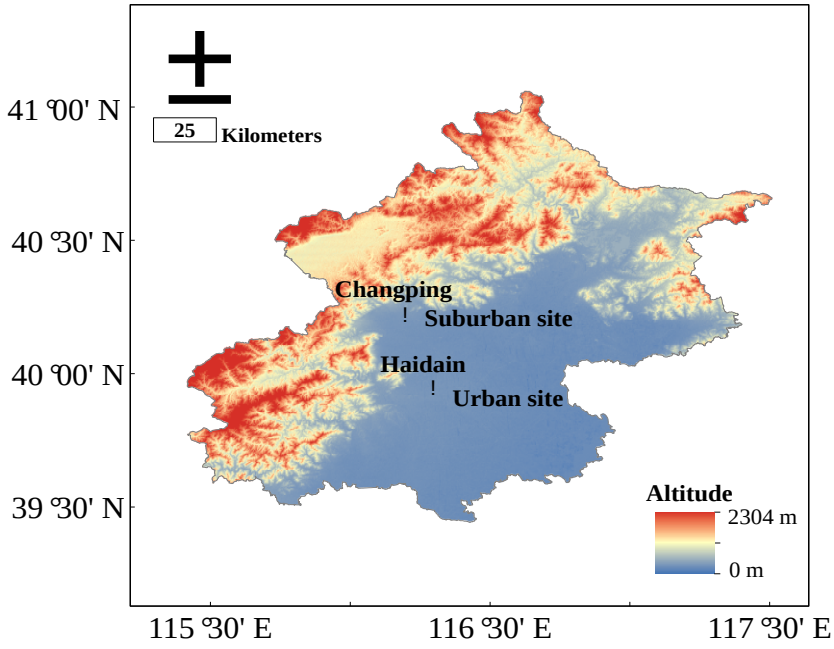

Figure 1. Location of the observation sites and the topography of Beijing city.

2.2. Measurements and data acquisition

NH_3 concentration measurements were performed by using two NH_3 analyzers (Ammonia Analyzer-Economical, Los Gatos Research Inc., USA), and the minimum detection limit was <0.2 ppb. These analyzers use off-axis integrated cavity output spectroscopy (OA-ICOS) technology, which is a fourth-generation cavity-enhanced absorption technique, to measure the NH_3 and water vapor (H_2O) concentrations 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 obtained concentration was normalized with respect to a reference concentration.

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Sample gases were drained through Teflon lines (1/4'OD), which lengths were designed as short as possible (less than 2 m). 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 measurements. However, according our test using certain concentrations of NH_3 in the lab, when heating (70℃) was on, there did have a peak lasting several minutes and then deceasing to the normal level in when heating was off. This tells us that heating is not a solution for NH_3 sticking. Keeping the relatively stable balance between adsorption and desorption of NH_3 in the sampling system are the most important. When tested by different humidity air, only very sharply change of humidity obviously influenced and changed the balance, and a new balance needed tens of minutes to reestablished. In the routine weather conditions, humidity changed in a relatively smoothing way except in a quickly changing weather system, like rainy days. The minute-level data were converted into hourly averages in the data analysis process and the hourly resolution can smoothing the effect to some extent caused by variations in humidity and temperature during the sampling time.

The balancing idea was also used to carry out multiple calibrations on NH_3 analyzers. A high mixing ratio (e.g. 400 ppb) of NH_3 mixing gases were firstly produced by a dynamic diluter and measured by the NH_3 analyzer overnight. After the signals were keeping in stable level, other lower span values were switched in turn. At each span point, the measurement time was lasting at least two hours. Then a linear regression function was obtained with R^2 higher than 0.999. Nowadays, NH_3 in compressed gas cylinder is also trustworthy, which result is concluded by the comparison with NH_3 permeation tube.

Finally, 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 NH3 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, 17, 133 and 1.6 ppb, respectively. At the suburban site, the corresponding values were 22 ± 15, 18, 199, and 0.8 ppb, respectively. The annual average NH_3 mixing ratio and range of the NH_3 mixing ratio at the suburban site were marginally higher than those at the urban site. The variation characteristics of the weekly smoothing data indicated that the NH_3 variations and temperature/humidity fluctuations at the two sites were practically consistent, which suggested 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.

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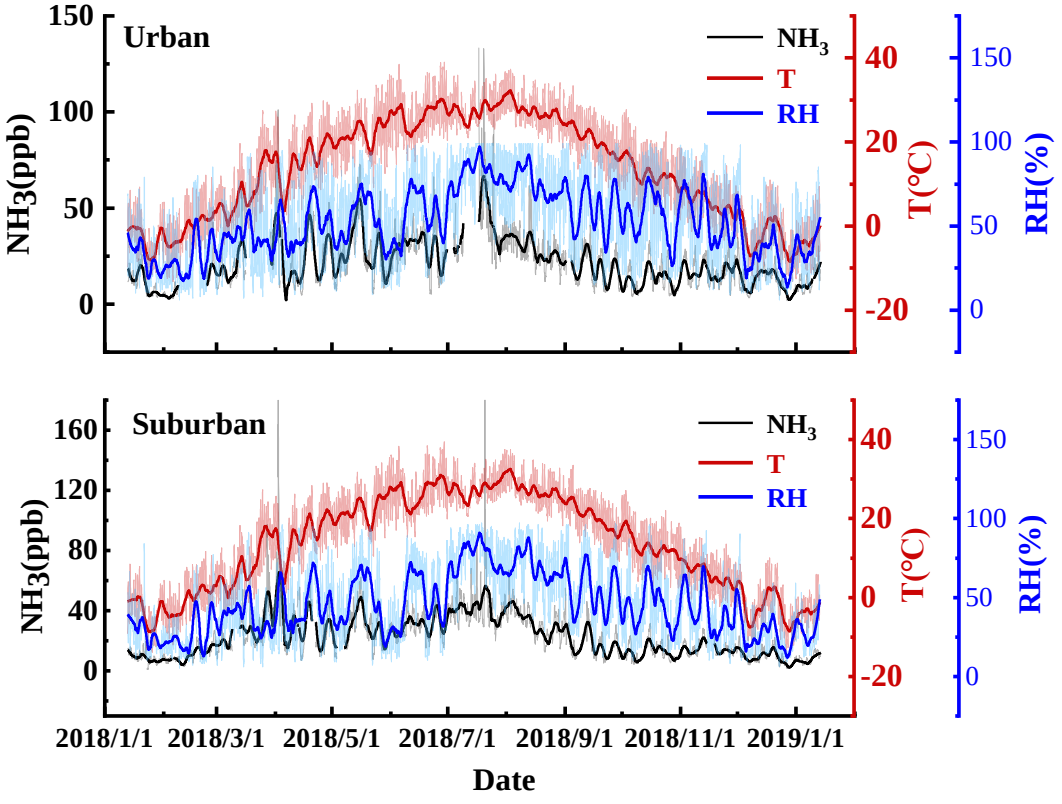

Figure 2. Temporal variations in the hourly average NH_3 mixing ratios, temperatures (T) and relative humidity(RH) at the urban and suburban stations in Beijing. Continuous thick lines were smoothed with 168 points (7 days) by using the Savitzky–Golay method.

Table 1 showed the comparison of the atmospheric NH_3 concentrations (ppb) in different areas. Meng et al. (2011) obtained an average NH_3 mixing ratio of 22.8 ± 16.3 ppb for the period 2008-2020 in Beijing urban area, which means there was no significant change in the annual average NH_3 mixing ratio from 2018 to 2019 compared with the change in the average NH_3 mixing ratio over the past decade. Moreover, 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). For example, the average annual NH_3 concentration in the urban area of Shanghai, a mega city in the south of China (31 °N), was approximately 50% lower than that in Beijing. This result 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).

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Table 1. Comparison of the atmospheric NH_3 concentrations (ppb) 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

The NH_3 mixing ratios in the United States (Edgerton et al., 2007; Nowak et al., 2006; Zhou et al. 2019), Great Britain (Burkhardt et al., 1998), Canada (Hu et al., 2014), and Japan (Osada et al., 2019) were 0.23–13, 1.6–2.3, 0.1–4, and 4.1 ppb, respectively. These NH_3 mixing ratios are considerably lower than that in Beijing. However, Delhi, India (Saraswati et al., 2019), exhibited a 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. The comparisons indicate that in the past decade, NH_3 concentration in Beijing has not changed considerably, but that it is higher than in large cities in other developed countries.

3.2. Seasonal variations

Fig. 2 displays the monthly statistical results for the NH_3 mixing ratios at the urban and suburban sites in Beijing. According to the seasonal division standard of China Meteorological Administration, in China, March to May is spring, June to August is summer, September to November is autumn, and December to February is winter. As presented in Fig. 2, the seasonal

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variations in the NH_3 mixing ratios were very similar at the urban and suburban sites. The NH_3 mixing ratios showed high values in the spring and summer and low values in the autumn and winter. The daily mean concentrations fluctuated considerably in the spring, and the highest variations occurred in April. The highest mean NH_3 concentrations occurred in July. The highest mean NH_3 concentrations at the urban and suburban sites were 42 ± 17 ppb and 42 ± 8.2 ppb, respectively. The NH_3 concentrations fluctuated considerably in July. On average, the NH_3 mixing ratios at the urban and suburban sites can be arranged according to season as follows: summer > spring > autumn > winter. The main grain crops in Beijing are corn and wheat. Corn is categorized as spring corn and summer corn, which are sown in April and June, respectively. A large amount of base fertilizer is applied when planting corn, and the topdressing is applied 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 the NH_3 mixing ratios and fluctuations in fertilization seasons (Zhang et al., 2016). In addition, the NH_3 mixing ratios are relatively high in the summer season due to the relatively high temperature in this season. An increase in the temperature can increase the biological activity and thus enhance the NH_3 emission. A 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 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. This result was obtained possibly because in the autumn and winter seasons, the influence of agricultural activities on the NH_3 concentration weakened, whereas the influences of other sources (such as traffic sources) on the NH_3 concentration were enhanced. According to Wang et al. (2019), the traffic NH_3 emission per unit area in Haidian was three times higher than that in Changping. 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.

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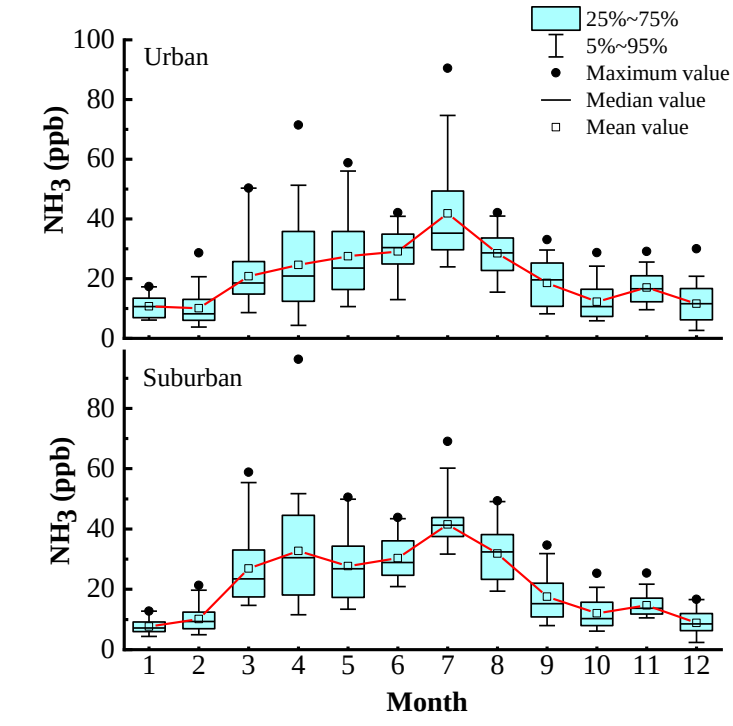

Figure 3. Monthly statistical variation in the NH_3 mixing ratios at the urban and suburban sites in Beijing.

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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
Suburban	Annual	22	15	0.8	18	198
	Spring	29	16	6.8	26	180
	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

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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. NH_3 exhibited different diurnal behaviors in different seasons.

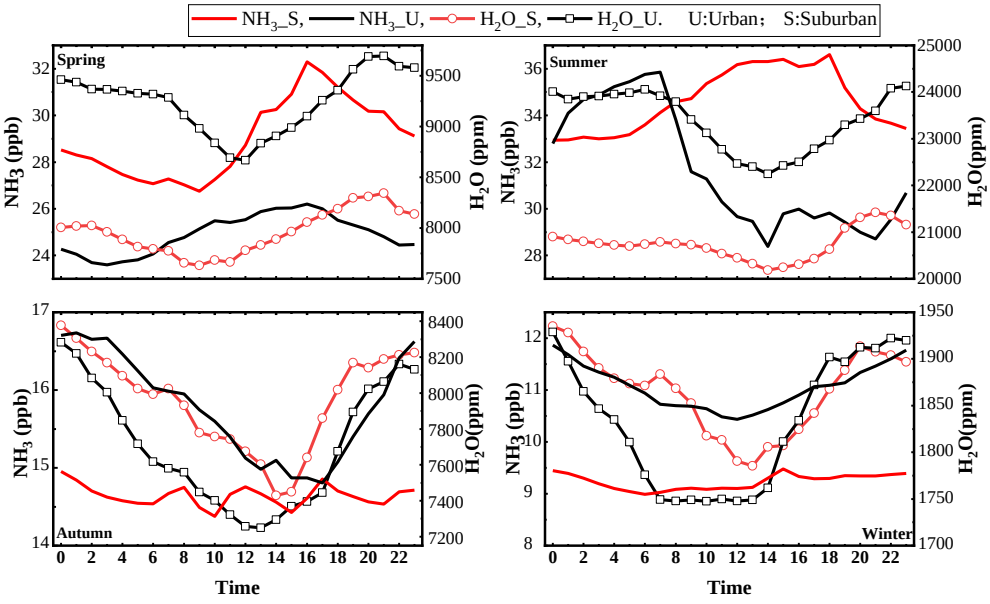

Figure 4. Average diurnal variations in the NH_3 and H_2O mixing ratios in different seasons at the urban and suburban sites in Beijing.

In the 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. This result was obtained possibly due to the increased NH_3 emission at the urban site due to traffic in the morning rush hours. On average, the mixing ratio of NH_3 was considerably 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 in the NH_3 mixing ratios of the sites. The average fluctuation in 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 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 the 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.

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The diurnal variations in the NH_3 mixing ratio at the suburban site in the summer were similar with those in the spring. This phenomenon was also observed in the rural areas of Shanghai by Chang et al. (2019). The diurnal variations were considerably affected by the temperature and the contribution from volatile NH_3 sources. However, at the urban site, the diurnal variations in the NH_3 mixing ratio were different. In the summer, the NH_3 mixing ratio increased gradually from 21:00, decreased after reaching its peak value at 7:00, and then reached its lowest value at 14:00. The diurnal variability pattern (with a peak value in the 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 the diurnal variability pattern was caused by the evaporation of dew in the morning, which results in the release of NH_3 that was originally stored in the droplets. A lag was observed between the changes in the NH_3 with 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 high NH_3 concentration in the residual layer led to an increase in the NH_3 concentration on the ground in the morning (Bash et al., 2010). The NH_3 and H_2O concentrations at the urban and suburban sites exhibited opposite diurnal variations patterns in the spring. In the 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. However, the NH_3 concentrations at the urban site were significantly higher than those at the suburban site during the second half of the night. The average fluctuation in the NH_3 concentration was 7.5 and 37 ppb at the urban and suburban site, respectively. Similar with the situation in the spring, the H_2O concentrations at the urban site were significantly higher than those at the suburban site in the summer.

In the autumn, the NH_3 concentration at the urban site had low values during the day and high values during the night. The peak NH_3 concentration occurred at midnight, and the lowest NH_3 concentration occurred at 17:00. There was essentially no diurnal variation in the NH_3 concentration at the suburban site, but obvious at the urban site with a fluctuation of 2.0 ppb. The concentration of NH_3 at the urban site was 1.2 ppb higher than that at the suburban site. The H_2O concentration was marginally lower (250 ppm) at the urban site than at the suburban site. The correlation between the diurnal variations in the NH_3 and H_2O concentrations was strong; however, the lowest value of NH_3 occurred later than the lowest value of H_2O at the urban site. The correlation between the diurnal variations in the NH_3 and H_2O concentrations was poor at the suburban site.

In the winter, the NH_3 mixing ratios at the urban and suburban sites exhibited similar diurnal variation patterns. The mixing ratios exhibited high values in the night and low values in the day. However, the NH_3 mixing ratio at the urban site was higher than that at the suburban site. This result was related to the decrease in the boundary layer height and temperature as well as the slow conversion and easy accumulation of pollutants in the night. In the daytime, increases in the temperature and boundary layer height enhanced the diffusion of pollutants, and the NH_3 mixing ratio decreased. The H_2O mixing ratio at the suburban site was close to that at the urban site, but higher in the morning. In the winter, the average diurnal variation in the NH_3 concentration was well correlated with that in the H_2O concentration at the urban site (R = 0.89). The correlation was close to that (R = 0.93) obtained by Teng et al. (2017) between the NH_3 and H_2O concentrations in the winter. However, at the suburban site, the mean diurnal variation in the NH_3 mixing ratios had a poor correlation with that in the H_2O mixing ratios.

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The results indicated 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 suggested 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 on the behaviors of NH_3.

3.4. Effect of meteorological factors on the NH_3 levels

Table 3 presents the correlations between the daily mean NH_3 mixing ratios and the diurnal mean values of the temperature, relative humidity, and wind speed at the two sites. Annually, the correlations were highly significant as the NH_3 mixing ratios at both sites were significantly and positively correlated with the temperature and relative humidity and negatively correlated with the wind speed. However, in the summer and autumn, no significant correlations were noted between the NH_3 and temperature at the two sites. The relative humidity had the stronger influence on the NH_3 concentration at the two sites than temperature, which phenomenon also can be found in Fig 2. The fluctuations between NH_3 and relative humidity were much more consistent.

Table 3. Correlations between the daily mean values of NH3 and meteorological elements (Spearman's rank correlation coefficient)

Site	Time Period	Temperature	Relative humidity	Wind speed
Urban	Annual	0.680**	0.706**	-0.370**
	Spring	0.450**	0.645**	-0.540**
	Summer	0.043	0.488**	-0.106**
	Autumn	0.101	0.759**	-0.413**
	Winter	0.596**	0.690**	-0.449**
Suburban	Annual	0.745**	0.730**	-0.325**
	Spring	0.256*	0.518**	-0.391**
	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.

Figure 5 display the seasonal mean diurnal variations in the NH_3 mixing ratio, temperature, and relative humidity with their correlation coefficients (also see in Fig. S3) in different seasons at the urban and suburban sites. At urban site, the diurnal variations in the NH_3 mixing ratio at the urban site were positively (negatively) correlated with the temperature (relative humidity) in spring, while in contrast, the diurnal variation of NH_3 mixing ratio were negatively (positively) correlated with temperature (relative humidity) in summer and autumn, and less correlated in winter. At suburban site, the diurnal variations in the NH_3 mixing ratio were positively (negatively) correlated with the temperature (relative humidity) in the spring and

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summer, but less correlated in the fall and winter. Similar behaviors were found in spring, but different in other seasons. In general, the annual diurnal behaviors of NH_3 with temperature and relative humidity were different at the urban and suburban sites (see Figure S1). Fig. 6 displays the contour maps of the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites. As displayed in Fig. 6 and Fig. S2, the NH_3 mixing ratios at both sites increased with the relative humidity at the same temperature and increased with the temperature at the same relative humidity. In winter, when the temperature was low (< 0 ℃), the NH_3 mixing ratios at both sites often had low values except in high humidity (>60%). An increase in the temperature increased the NH_3 mixing ratios; however, the NH_3 concentration at the suburban site was more significantly affected by the temperature than that at the urban site (Table 3), indicating that volatile NH_3 sources might have a higher contribution to the NH_3 concentration at the suburban site than at the urban site. A higher amount of NH_3 removal through chemical transformation was expected during the day at the urban site than at the suburban site because the urban site had a higher relative humidity and higher amounts of primary particulate matter, NO_x, and SO_2 acid gas emissions than the suburban site did. In 2018, the concentrations of $PM_{2.5}$, SO_2 and NO_2 were 50μg/m^3, 5μg/m^3, 43μg/m^3 in Haidian, and 46μg/m^3, 6μg/m^3, 35μg/m^3 in Changping, respectively, which were reported by Beijing Ecology and Environment Statement, 2018.

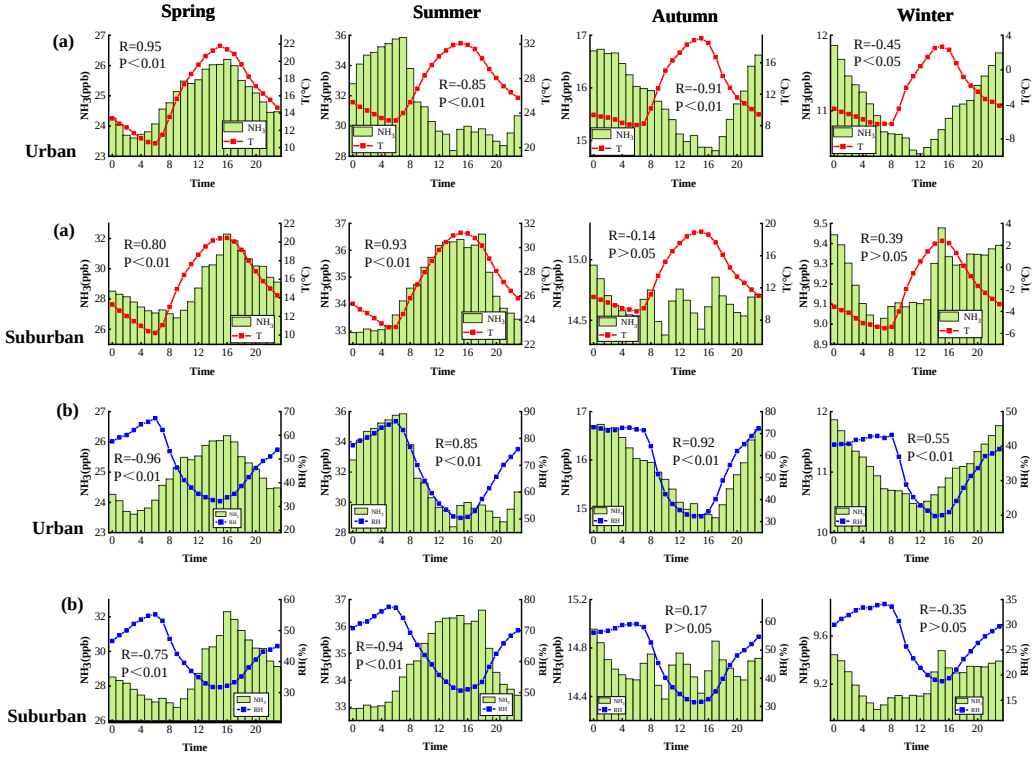

Figure 5. Diurnal variations in and correlation coefficients between the NH3 mixing ratios and temperature (a), relative humidity (b) in different seasons at the urban and suburban sites in Beijing.

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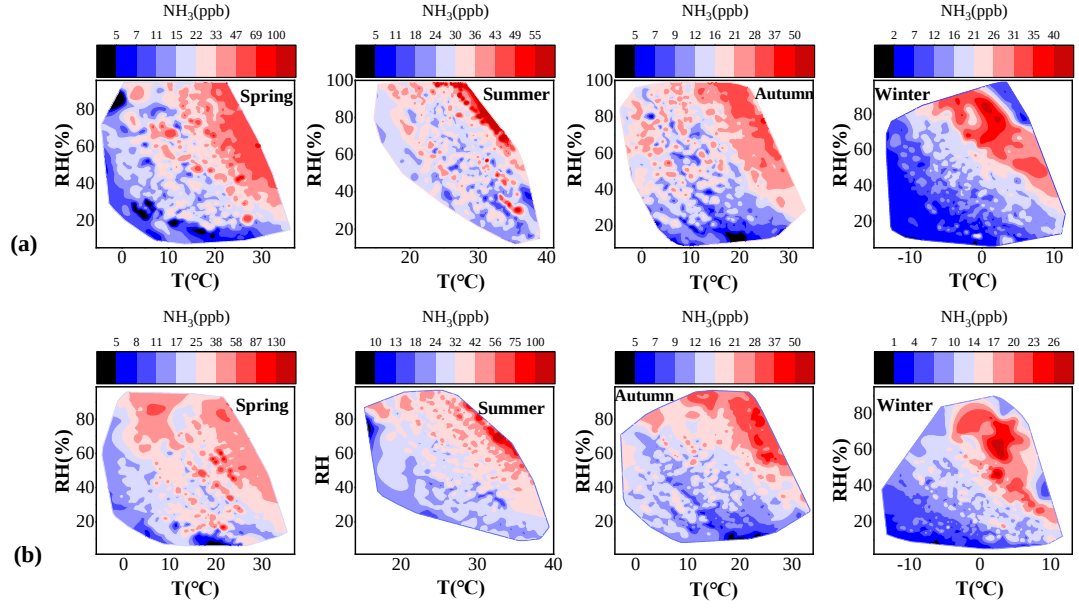

Figure 6. Contour maps of the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites in Beijing (a: Urban, b: Suburban).

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 area originate from the south (often in the day) and north (often at night). As displayed in Fig. 6, some differences existed in the distributions of the surface wind between the urban and suburban sites. The dominant surface winds originated from the northeast and southwest at the urban site and from the northwest and east 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 the action of the southwest wind, a polluted air mass from the south of Beijing can be easily transported to the urban site. Meng et al. (2017) examined the effect of long-range air transport on the urban NH_3 levels in Beijing during the summer through trajectory analysis. The authors 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 relatively high in the south sectors for both sites. Thus, winds from the southeast, south, and southwest had a cumulative effect on the NH_3 mixing ratios at both the urban and suburban sites. The NH_3 mixing ratios were relatively low when the wind originated from the northwest sector at urban site and from the west sector at the suburban site, in which the wind speed was strong, which indicated that the northwest/west wind could promote NH_3 dilution and diffusion.

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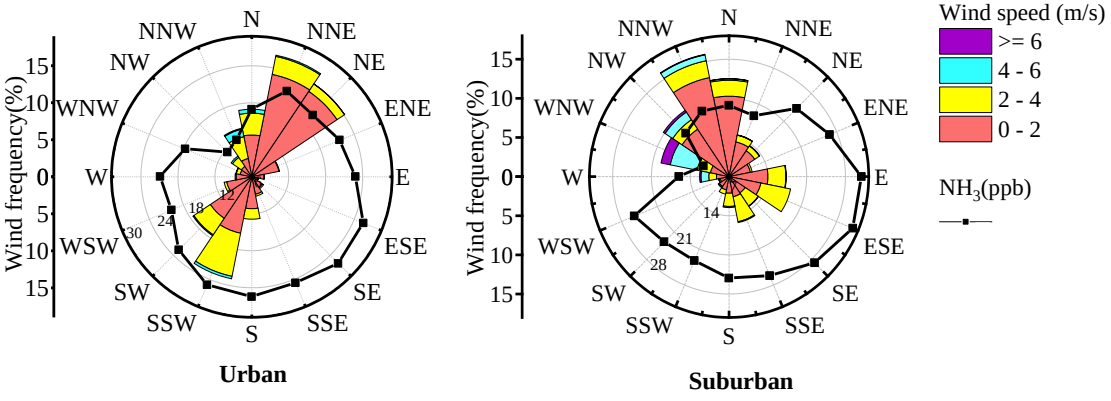

Figure 7. Rose maps of the NH_3 mixing ratios, wind frequency, and wind speed in different wind direction sectors.

Heavy rainfall occurred on a few days in Beijing in 2018. Heavy precipitation occurred for a long duration 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 occurred, the NH_3 concentration decreased rapidly, and it was significantly lower than the mean value in August. However, the diurnal variation of NH_3 on the rain day did not differ considerably from the average diurnal variation in August. On August 18, 2018, the NH_3 mixing ratio at the suburban site remained at a low level during the rainfall period and was considerably lower than the mean NH_3 concentration in August. 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 needed more cases to support.

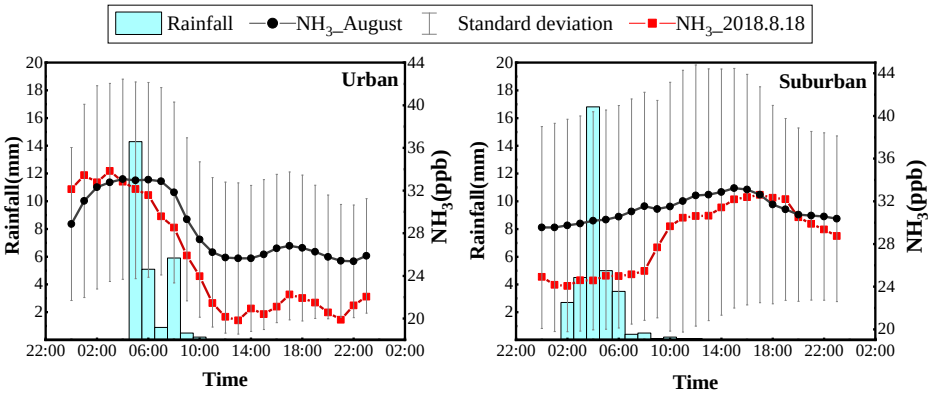

Figure 8. Diurnal variations in the rainfall and NH_3 concentration on August 18, 2018.

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4. Conclusion

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 and 22 ± 15 ppb at the urban and suburban sites, respectively. The annual average NH_3 mixing ratio at the suburban site was higher than that at the urban site. Moreover, the variation range of the NH_3 mixing ratio was larger at the suburban site than at the urban site. In the summer and spring, the NH_3 mixing ratio at the suburban site was higher than that at the urban site. In the autumn and winter, the NH_3 mixing ratio at the suburban site was lower than that at the urban site. The highest NH_3 mixing ratios at the urban and suburban sites were observed in July. The lowest NH_3 mixing ratio at the urban site occurred in February, and the lowest NH_3 mixing ratio at the suburban site occurred in January. In the past decade, the concentration of NH_3 in Beijing did not change considerably, and the NH_3 levels in Beijing were higher than those in other large cities.

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, low NH_3 mixing ratios were observed in the day and high NH_3 mixing ratios were observed in the 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. This result indicated that NH_3 sources had different contributions to the NH_3 levels at the urban and suburban sites.

The influence of meteorological factors on the NH_3 mixing ratio was complex. At the same temperature, the NH_3 mixing ratios increased with the relative humidity at the urban and suburban sites. At the same relative humidity, the NH_3 mixing ratios increased with the temperature at both sites. The relative humidity had the strongest influence on the NH_3 mixing ratio in different seasons at the two sites. No strong correlation was observed between the NH_3 concentration and the temperature in the summer and autumn at the two sites. A high wind speed promoted a decrease in the NH_3 concentration. The NH_3 mixing ratios were higher when the winds originated from the south than when the winds originated from the north and northwest. Rainfall had a certain scavenging effect on NH_3; however, it 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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Acknowledgments. This study was funded by the National Natural Science Foundation of China (Grant No. 91744206) and the Beijing Municipal Science and Technology (Z181100005418016).

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