

1 Measurement report: Exploring the NH₃ behaviors at urban and suburban Beijing:
2 Comparison and implications

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8 **ABSTRACT**

9 Ammonia (NH₃) plays an important role in particulate matter formation; however, few long-term
10 observations with a high temporal resolution have been conducted on the NH₃ concentrations in Beijing.
11 In this study, online ammonia analyzers were used to observe continuously the atmospheric NH₃
12 concentrations at an urban site and a suburban site in Beijing from January 13, 2018, to January 13, 2019.
13 The average mixing ratio of NH₃ at the urban site was 21 ± 14 ppb (range: 1.6–133 ppb) and that at the
14 suburban site was 22 ± 15 ppb (range: 0.8–199 ppb). The NH₃ mixing ratios at the urban and suburban
15 sites exhibited similar seasonal variations, with high values being observed in the summer and spring
16 and low values being observed in the autumn and winter. The hourly mean NH₃ mixing ratios at the urban
17 site were highly correlated ($R = 0.849$, $P < 0.01$) with those at the suburban site. However, the average
18 diurnal variations in the NH₃ mixing ratios at the urban and suburban sites differed significantly, which
19 indicated the different contributions of NH₃ sources and sinks at the urban and suburban sites. In addition
20 to the emission sources, meteorological factors were closely related to the changes in the NH₃
21 concentrations. For the same temperature (relative humidity) at the urban and suburban sites, the NH₃
22 mixing ratios increased with the relative humidity (temperature). The relative humidity was the factor
23 with the strongest influence on the NH₃ mixing ratio in different seasons at the two sites. In general, a

24 high wind speed promoted a reduction in the NH₃ mixing ratio. Similar with other primary pollutants in
25 Beijing, the NH₃ mixing ratios were high when winds originated from the south and low when winds
26 originated from the north and northwest.

27 **Keywords:** NH₃; variations; simultaneous observation

28

29 **1. Introduction**

30 Ammonia (NH₃) is the most abundant alkaline trace gas in the atmosphere (Meng et al., 2017). An
31 excessive NH₃ concentration directly harms the ecosystem; causes water eutrophication and soil
32 acidification; and leads to forest soil erosion, biodiversity reduction, and carbon uptake variations
33 (Pearson and Stewart, 1993; Reay et al., 2008; van Breemen et al., 1983). Thus, the NH₃ concentration
34 influences climate change (Charlson et al., 1991; Erisman et al., 2007). NH₃ can react with acidic gases
35 to form ammonium salts, which might significantly influence the mass concentration and composition
36 of particulate matter (Wu et al., 2009). After the implementation of policies such as the *12th Five-Year*
37 *Plan for the Key Regional Air Pollution Prevention and Control in Key Regions* (Ministry of Ecology
38 and Environment of the People's Republic of China, 2012) and the *Air Pollution Prevention and Control*
39 *Action Plan* (General Office of the State Council, PRC, 2013), China, especially for Beijing, has been
40 effectively controlling the emission of primary pollutants, such as sulfur dioxide (SO₂) and nitrogen oxide
41 (NO_x); however, the pollution caused by fine particles is still serious (Krotkov et al., 2016; UN
42 Environment, 2019). Studies have indicated that when the SO₂ and NO_x concentrations are reduced to a
43 certain extent, reducing NH₃ emissions is the most economical and effective method to decrease the
44 PM_{2.5} concentration (Pinder et al., 2008). In China, the main anthropogenic sources of NH₃ are livestock
45 and poultry feces (54%) and fertilizer volatilization (33%) (Huang et al., 2012). Moreover, the
46 atmospheric NH₃ concentration in China has increased with the expansion of agricultural activities,
47 control of SO₂ and NO_x, and increase in temperature (Warner et al., 2017). This increase in the NH₃
48 concentration might weaken the effectiveness of SO₂ and NO_x emission control in reducing PM_{2.5}
49 pollution (Fu et al., 2017).

50 The North China Plain is a region with high NH₃ emission (Zhang et al., 2017), and Beijing has one

51 of the highest NH₃ concentrations in the world (Chang et al., 2016b; Pan et al., 2018). Compared with
52 studies on pollutants such as SO₂ and NO_x, considerably fewer studies have been conducted on the NH₃
53 concentration in Beijing. Chang et al. (2016a) collected gaseous NH₃ samples during the 2014 APEC
54 summit (October 18 to November 29, 2014) in the Beijing urban area and concluded that the overall
55 contributions of traffic, garbage, livestock, and fertilizers to the NH₃ concentration were 20.4%, 25.9%,
56 24.0%, and 29.7%, respectively. According the data from Huang et al (2012), the NH₃ emissions in
57 Beijing were from livestock and poultry farming (34.55%), nitrogen-fixing plants (33.57%), fertilizer
58 use (13.06%), household garbage treatment (8.29%), traffic emissions (5.20%), industrial emissions
59 (0.14%), biomass combustion (0.42%), and agricultural soil (0.84%). Zhang (2016) measured the NH₃
60 concentrations in urban and rural areas of Beijing from January to July 2014 and found that NH₃
61 concentration in urban areas was approximately 65% higher than that in rural areas. Meng et al. (2011)
62 reported that the highest NH₃ concentration in Beijing occurred in summer and the lowest one occurred
63 in winter, and their results indicated traffic to be a significant source of NH₃ in urban areas. Zhang et al.
64 (2018) reported the vertical variability of NH₃ in urban Beijing based on one-year passive samples in
65 2016/2017 and concluded that local sources such as traffic emissions were important contributors to
66 urban NH₃. Meng *et al.* (2020) investigated the significant increase in winter NH₃ and its contribution to
67 the increasing nitrate in PM_{2.5} from 2009 to 2016, and they also concluded that vehicles exhaust was an
68 important contributor to NH₃ in urban Beijing in winter.

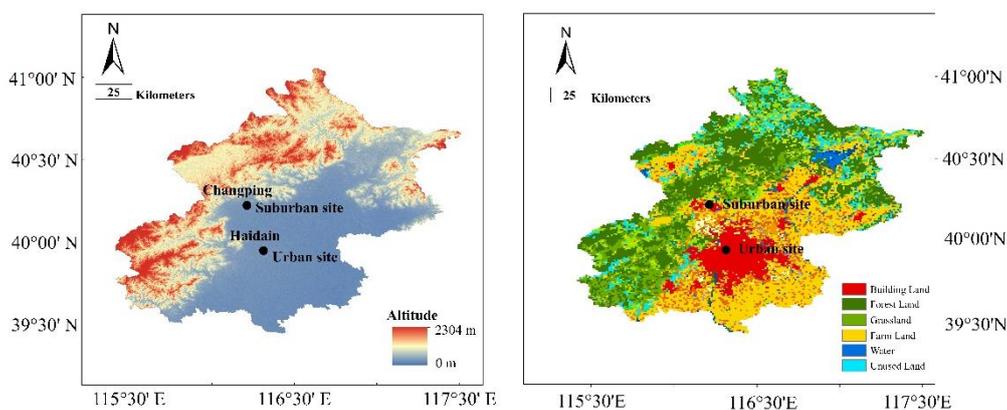
69 Currently, NH₃ is not included in the routine environmental monitoring operation in China. Research
70 data on NH₃ monitoring, particularly on the synchronous observation of the NH₃ concentrations with a
71 high temporal resolution in urban and suburban areas, are relatively scarce. In this study, high-time-
72 resolution observations of NH₃ were obtained simultaneously at an urban site and a suburban site in

73 Beijing. The variation characteristics and influencing factors of the NH_3 concentration were analyzed
74 with meteorological data to provide a scientific basis for NH_3 pollution control in Beijing.

75 2. Materials and methods

76 2.1. Measurement sites

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



85

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

87

88 2.2. Measurements and data acquisition

89 NH_3 concentration measurements were performed by using two NH_3 analyzers (Ammonia
90 Analyzer-Economical, Los Gatos Research Inc., USA), and the minimum detection limit was <0.2 ppb

91 and the maximum drift of 0.2 ppb/24hrs. The analyzers were deployed in air-conditioning rooms. These
92 analyzers use off-axis integrated cavity output spectroscopy (OA-ICOS) technology, which is a fourth-
93 generation cavity-enhanced absorption technique, to measure the NH₃ and water vapor (H₂O)
94 concentrations in the atmosphere. The incident laser beam of the OA-ICOS technology deviates from the
95 optical axis, which differs from the traditional coaxial incidence mode. The axial incidence mode of the
96 OA-ICOS technology can increase the optical path, stimulate additional high-order transverse modes,
97 effectively suppress the noise of the cavity mode, reduce the cross interferences and errors due to
98 contaminants existing in the cavity, and improve the detection sensitivity (Baer *et al.*, 2002; Baer *et al.*,
99 2012). The analyzer method is a quasi-absolute measurement, which theoretically does not require
100 calibration. However, to ensure the comparability of the obtained data with other monitoring data, NH₃
101 standard gas (Beijing AP BAIF Gases Industry Co., Ltd.) was used for comparison measurement before
102 the observation. The obtained concentration was revised with respect to a reference concentration.

103 Sample gases were drained by >0.4 L/min through Teflon lines (1/4"OD) from a manifold, which
104 lengths were designed as short as possible (less than 2 m from the manifold). Particulate matters were
105 filtered by Teflon membranes with a pore size less than 5 μm. Since NH₃ easily “sticks” to surfaces (like
106 inside walls of tubes), heated sample lines were suggested by many measurements. However, according
107 our test (Fig. S1) in the lab, when heating (70°C) was on, there did have a peak lasting several minutes
108 (5–6 min) and then decreasing to the normal levels in ambient air, which means a new balancing process
109 has been established in less than 10 min. This tells us that heating is not a solution for NH₃ sticking.
110 Keeping the relatively stable balance between adsorption and desorption of NH₃ in the sampling system
111 are the most important. When tested by different humidity air, only very sharply change of humidity
112 obviously influenced and changed the balance, and a new balance needed tens of minutes to reestablished

113 (Fig. S2). In the routine weather conditions, humidity changed in a relatively smoothing way except in a
114 quickly changing weather system, like rainy days. The minute-level data were converted into hourly
115 averages in the data analysis process and the hourly resolution can smoothing the effect to some extent
116 caused by variations in humidity and temperature during the sampling time.

117 The balancing idea was also used to carry out multiple calibrations on NH₃ analyzers (Fig. S3). A
118 high mixing ratio (e.g. 400 ppb or higher) of NH₃ mixing gases were firstly produced by a dynamic
119 diluter and measured by the NH₃ analyzer overnight. After the signals were keeping in stable level, other
120 lower span values were switched in turn. At each span point, the measurement time was lasting at least
121 40 minutes or more. Then a linear regression function was obtained with R² higher than 0.999. Nowadays,
122 NH₃ in compressed gas cylinder is also trustworthy, which result is concluded by the comparison with
123 NH₃ permeation tube (Fig. S4).

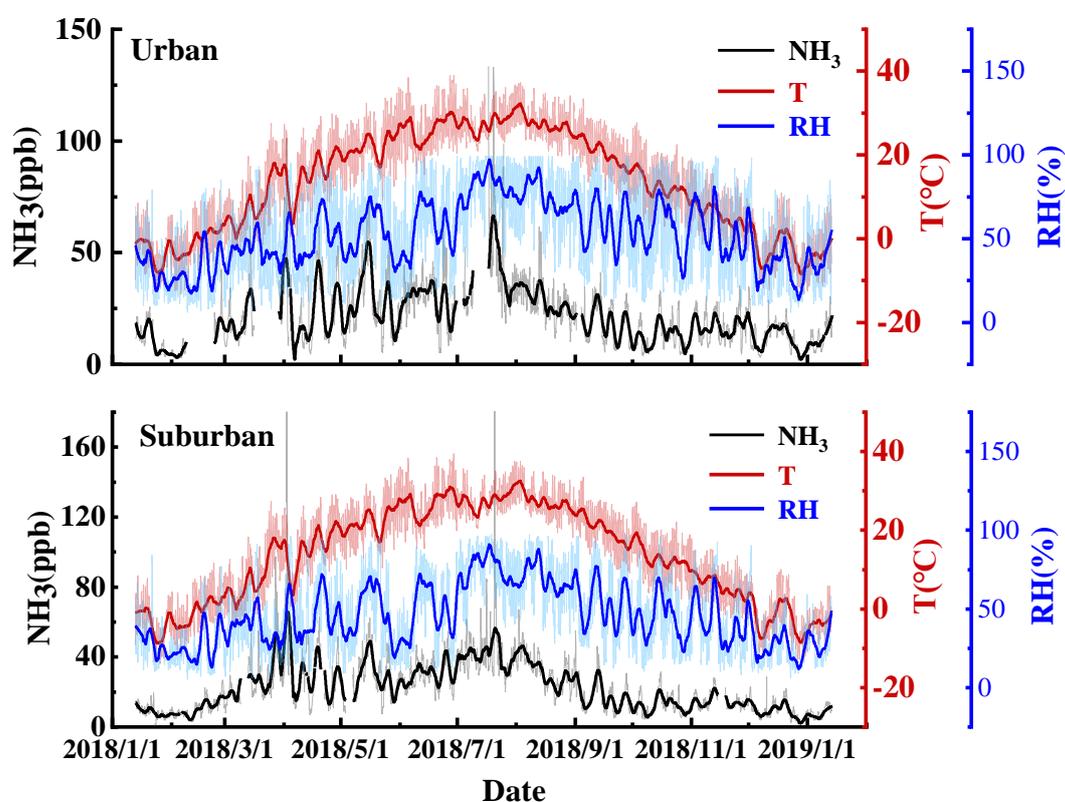
124 Finally, 7645 and 8342 valid hourly mean observations were obtained for the urban (Haidian) and
125 suburban (Changping) sites, respectively. In addition, the urban and suburban meteorological data
126 (temperature, relative humidity, wind direction, and wind speed) during the sampling period were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

129 3. Results and discussion

130 3.1. Overall variations in the NH₃ mixing ratios

131 Fig. 2 displays the time-series variations in the NH₃ mixing ratios, temperatures, and relative
132 humidity at the urban and suburban sites in Beijing. At the urban site, the mean $\pm 1\sigma$, median, maximum,
133 and minimum values of the hourly average NH₃ mixing ratio during the observation period were 21 ± 14 ,
134 17, 133 and 1.6 ppb, respectively. At the suburban site, the corresponding values were 22 ± 15 , 18, 199,

135 and 0.8 ppb, respectively. The annual average NH_3 mixing ratio and range of the NH_3 mixing ratio at the
 136 suburban site were marginally higher than those at the urban site. The variation characteristics of the
 137 weekly smoothing data indicated that the NH_3 variations and temperature/humidity fluctuations at the
 138 two sites were practically consistent, which suggested that both sites were under the influence of similar
 139 weather systems. The hourly mean NH_3 concentrations at the urban site were significantly correlated (R
 140 = 0.849, $P < 0.01$) with those at the suburban site.



141
 142 **Fig. 2.** Temporal variations in the hourly average NH_3 mixing ratios, temperatures (T) and relative humidity(RH) at the urban and suburban
 143 stations in Beijing. Continuous thick lines were smoothed with 168 points (7 days) by using the Savitzky–Golay method.

144
 145 Table 1 showed the comparison of the atmospheric NH_3 concentrations (ppb) in different areas.
 146 Meng et al. (2011) obtained an average NH_3 mixing ratio of 22.8 ± 16.3 ppb for the period 2008-2020 in
 147 Beijing urban area, which means there was no significant change in the annual average NH_3 mixing ratio
 148 from 2018 to 2019 compared with the change in the average NH_3 mixing ratio over the past decade.

149 Moreover, the NH₃ concentrations at the urban and suburban sites were higher than those in the
 150 background areas. The observed NH₃ concentrations in Beijing were higher than those in northwest
 151 China (Meng et al. 2010) and the Yangtze River Delta region (Chang et al. 2019). For example, the
 152 average annual NH₃ concentration in the urban area of Shanghai, a mega city in the south of China (31°
 153 N), was approximately 50% lower than that in Beijing. This result might be related to the fact that the
 154 North China Plain, in which Beijing is located, is one of the most intensive agricultural production
 155 regions in China. The differences in the soil properties of Beijing and Shanghai may be another reason
 156 because the loss of soil NH₃ can increase with an increase in the soil pH (Ju et al., 2009). Shanghai and
 157 its surrounding areas are dominated by acidic soil of paddy fields (Zhao et al., 2009), whereas Beijing is
 158 dominated by the alkaline soils of dry land (Wei et al., 2013).

159 **Table 1.** Comparison of the atmospheric NH₃ 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 |
| 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 |

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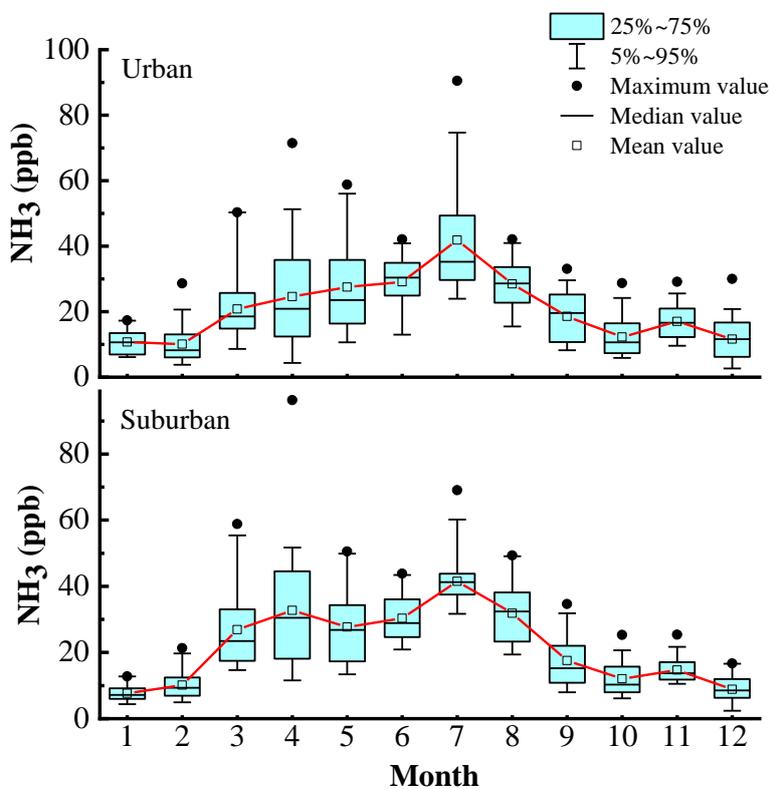
161 The NH₃ mixing ratios in the United States (Edgerton et al., 2007; Nowak et al., 2006; Zhou et al.
 162 2019), Great Britain (Burkhardt et al., 1998), Canada (Hu et al., 2014), and Japan (Osada et al., 2019)
 163 were 0.23–13, 1.6–2.3, 0.1–4, and 4.1 ppb, respectively. These NH₃ mixing ratios are considerably lower
 164 than that in Beijing. However, Delhi, India (Saraswati et al., 2019), exhibited a higher NH₃ mixing ratio
 165 (53.4±14.9 ppb) than Beijing did. This result might be attributed to the well-developed livestock breeding
 166 activities in Delhi. The comparisons indicate that in the past decade, NH₃ concentration in Beijing has
 167 not changed considerably, but that it is higher than in large cities in other developed countries.

168 *3.2. Seasonal variations*

169 Fig. 2 displays the monthly statistical results for the NH₃ mixing ratios at the urban and suburban
 170 sites in Beijing. According to the seasonal division standard of China Meteorological Administration, in
 171 China, March to May is spring, June to August is summer, September to November is autumn, and
 172 December to February is winter. As presented in Fig. 2, the seasonal variations in the NH₃ mixing ratios
 173 were very similar at the urban and suburban sites. The NH₃ mixing ratios showed high values in the
 174 spring and summer and low values in the autumn and winter. The daily mean concentrations fluctuated
 175 considerably in the spring, and the highest variations occurred in April. The highest mean NH₃
 176 concentrations occurred in July. The highest mean NH₃ concentrations at the urban and suburban sites
 177 were 42± 17 ppb and 42 ± 8.2 ppb, respectively. The NH₃ concentrations fluctuated considerably in July.
 178 On average, the NH₃ mixing ratios at the urban and suburban sites can be arranged according to season
 179 as follows: summer > spring > autumn > winter. The main grain crops in Beijing are corn and wheat.

180 Corn is categorized as spring corn and summer corn, which are sown in April and June, respectively. A
181 large amount of base fertilizer is applied when planting corn, and the topdressing is applied after 2 months.
182 Wheat is sown from September to October, and the topdressing is applied in the following spring. The
183 volatilization of nitrogen fertilizers can cause an increase in the NH_3 mixing ratios and fluctuations in
184 fertilization seasons (Zhang et al., 2016). In addition, the NH_3 mixing ratios are relatively high in the
185 summer season due to the relatively high temperature in this season. An increase in the temperature can
186 increase the biological activity and thus enhance the NH_3 emission. A high temperature is also conducive
187 for the volatilization of the urea and diammonium phosphate applied to crops. Moreover, the equilibrium
188 among ammonium nitrate particles, gaseous NH_3 , and nitric acid is transferred to the gas phase at high
189 temperature, which increases NH_3 concentration (Behera et al., 2013). Sewage treatment, household
190 garbage, golf courses, and human excreta are crucial NH_3 sources that are easily neglected (Pu et al.,
191 2020). The relatively low NH_3 concentrations in the autumn and winter might be caused by the decrease
192 in NH_3 emission in the soil and vegetation, the decrease in the NH_4NO_3 decomposition capacity at low
193 temperatures, and the reduced human activities caused by a large floating population returning to their
194 native locations outside Beijing during the Spring Festival (Liao et al., 2014). In the spring and summer,
195 the NH_3 mixing ratios at the suburban site were higher than those at the urban site, which might be related
196 to the higher agricultural activity around the suburban site. In the autumn and winter, the NH_3 mixing
197 ratios at the urban site were marginally higher than those at the suburban site. This result was obtained
198 possibly because in the autumn and winter seasons, the influence of agricultural activities on the NH_3
199 concentration weakened, whereas the influences of other sources (such as traffic sources) on the NH_3
200 concentration were enhanced. According to Wang et al. (2019), the traffic NH_3 emission per unit area in
201 Haidian was three times higher than that in Changping. This difference in traffic source emissions might

202 have resulted in higher NH₃ concentrations at the urban site than at the suburban site in the autumn and
 203 winter.



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Fig. 3. Monthly statistical variation in the NH₃ mixing ratios at the urban and suburban sites in Beijing.

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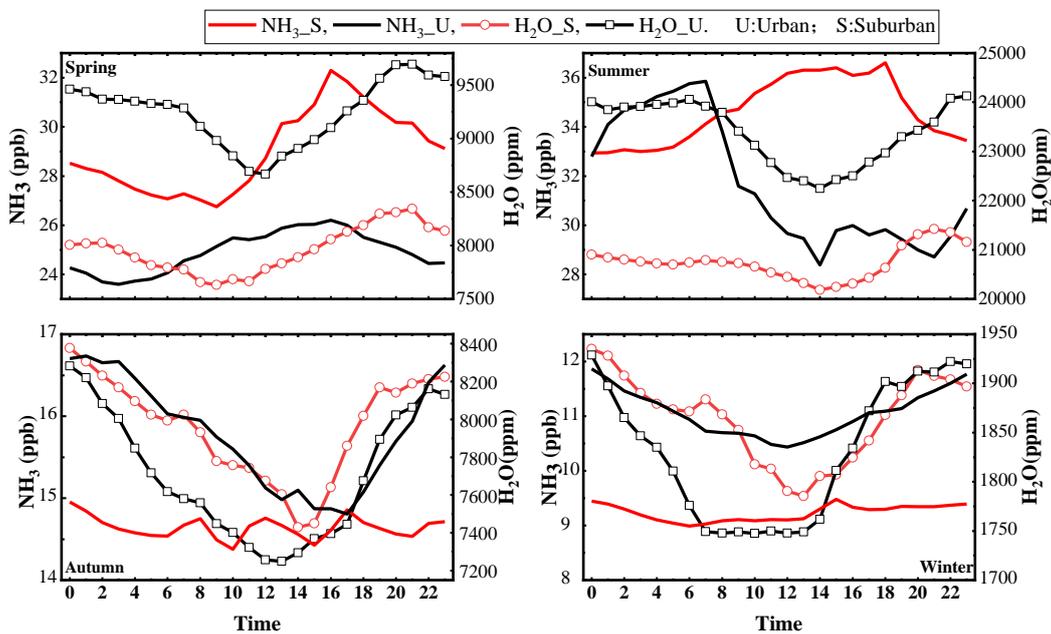
Table 2. NH₃ mixing ratios (ppb) measured at the urban and suburban sites in Beijing.

| 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 |

208

209 3.3. Diurnal variations

210 Figure 4 displays the average diurnal variations in the NH₃ and H₂O mixing ratios in different
 211 seasons at the urban and suburban sites in Beijing. NH₃ exhibited different diurnal behaviors in different
 212 seasons.



213

214 Fig. 4. Average diurnal variations in the NH₃ and H₂O mixing ratios in different seasons at the urban and suburban sites in Beijing.

215

216 In the spring, the average diurnal variations in the NH₃ mixing ratio were similar at the urban and
 217 suburban sites. The diurnal variations exhibited a single-peak pattern with high values in the daytime and
 218 low values at night. The NH₃ mixing ratio began to increase in the morning, reached its maximum value
 219 at 16:00, and then decreased gradually. The lowest mixing ratios at the urban and suburban sites occurred
 220 at 03:00 and 09:00, respectively. The NH₃ mixing ratio began to increase earlier at the urban site than at
 221 the suburban site. This result was obtained possibly due to the increased NH₃ emission at the urban site

222 due to traffic in the morning rush hours. On average, the mixing ratio of NH_3 was considerably higher at
223 the suburban site than that at the urban site, with an average difference of 4.1 ppb and a maximum
224 difference of 6.1 ppb in the NH_3 mixing ratios of the sites. The average fluctuation in the NH_3 mixing
225 ratio at the suburban site was 5.3 ppb, which was higher than that (2.6 ppb) at the urban site. At the urban
226 site, the average diurnal variations in the NH_3 and H_2O mixing ratios exhibited opposite trends. The H_2O
227 mixing ratio had high values in the night and low values in the day. At the suburban site, the variation
228 characteristics of NH_3 and H_2O were very similar; however, the peak NH_3 concentration occurred 5 hours
229 earlier than the peak H_2O concentration. In the spring, in contrast to the NH_3 mixing ratio, the H_2O mixing
230 ratio at the urban site was 1279 ppm higher than that at the suburban site.

231 The diurnal variations in the NH_3 mixing ratio at the suburban site in the summer were similar with
232 those in the spring. This phenomenon was also observed in the rural areas of Shanghai by Chang et al.
233 (2019). The diurnal variations were considerably affected by the temperature and the contribution from
234 volatile NH_3 sources. However, at the urban site, the diurnal variations in the NH_3 mixing ratio were
235 different. In the summer, the NH_3 mixing ratio increased gradually from 21:00, decreased after reaching
236 its peak value at 7:00, and then reached its lowest value at 14:00. The diurnal variability pattern (with a
237 peak value in the morning) has been observed in other areas, such as rural (Ellis et al., 2011), urban
238 (Gong et al., 2011), and steppe areas located far away from human activity (Wentworth et al., 2016).
239 Kuang et al. (2020) believed that the diurnal variability pattern was caused by the evaporation of dew in
240 the morning, which results in the release of NH_3 that was originally stored in the droplets. A lag was
241 observed between the changes in the NH_3 with H_2O concentrations in the early morning, which supported
242 the hypothesis of Kuang et al (2020). In addition, the increase in the NH_3 concentration in the morning
243 might have been caused by the breakup of the boundary layer formed at night. The downward mixing of

244 air with a high NH_3 concentration in the residual layer led to an increase in the NH_3 concentration on the
245 ground in the morning (Bash et al., 2010). The NH_3 and H_2O concentrations at the urban and suburban
246 sites exhibited opposite diurnal variations patterns in the spring. In the summer, the NH_3 concentrations
247 at the suburban site were significantly higher than those at the urban site during the daytime and first half
248 of the night. However, the NH_3 concentrations at the urban site were significantly higher than those at
249 the suburban site during the second half of the night. The average fluctuation in the NH_3 concentration
250 was 7.5 and 37 ppb at the urban and suburban site, respectively. Similar with the situation in the spring,
251 the H_2O concentrations at the urban site were significantly higher than those at the suburban site in the
252 summer.

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

262 In the winter, the NH_3 mixing ratios at the urban and suburban sites exhibited similar diurnal
263 variation patterns. The mixing ratios exhibited high values in the night and low values in the day.
264 However, the NH_3 mixing ratio at the urban site was higher than that at the suburban site. This result was
265 related to the decrease in the boundary layer height and temperature as well as the slow conversion and

266 easy accumulation of pollutants in the night. In the daytime, increases in the temperature and boundary
267 layer height enhanced the diffusion of pollutants, and the NH₃ mixing ratio decreased. The H₂O mixing
268 ratio at the suburban site was close to that at the urban site, but higher in the morning. In the winter, the
269 average diurnal variation in the NH₃ concentration was well correlated with that in the H₂O concentration
270 at the urban site ($R = 0.89$). The correlation was close to that ($R = 0.93$) obtained by Teng et al. (2017)
271 between the NH₃ and H₂O concentrations in the winter. However, at the suburban site, the mean diurnal
272 variation in the NH₃ mixing ratios had a poor correlation with that in the H₂O mixing ratios.

273 The results indicated that although the two sites were under the influence of similar weather systems,
274 the diurnal variations in the NH₃ mixing ratios at the two sites were different in different seasons. This
275 finding suggested that different NH₃ sources and possibly sinks had different contributions to the NH₃
276 concentrations at the urban and suburban sites. Additional studies should be conducted on the behaviors
277 of NH₃.

278 3.4. Effect of meteorological factors on the NH₃ levels

279 Table 3 presents the correlations between the daily mean NH₃ mixing ratios and the diurnal mean
280 values of the temperature, relative humidity, and wind speed at the two sites. Annually, the correlations
281 were highly significant as the NH₃ mixing ratios at both sites were significantly and positively correlated
282 with the temperature and relative humidity and negatively correlated with the wind speed. However, in
283 the summer and autumn, no significant correlations were noted between the NH₃ and temperature at the
284 two sites. The relative humidity had the stronger influence on the NH₃ concentration at the two sites than
285 temperature, which phenomenon also can be found in Fig 2. The fluctuations between NH₃ and relative
286 humidity were much more consistent.

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

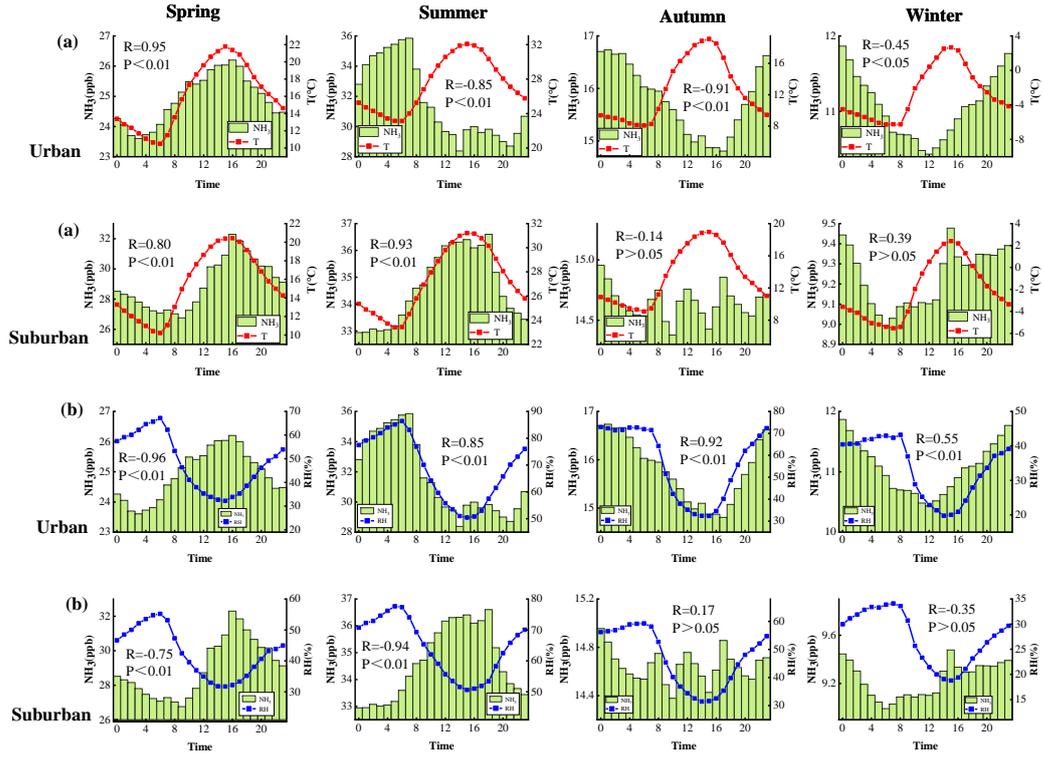
| Site | Time Period | Temperature | Relative humidity | Wind speed |
|----------|-------------|-------------|-------------------|------------|
| Urban | Annual | 0.680** | 0.706** | -0.370** |
| | Spring | 0.450** | 0.645** | -0.540** |
| | Summer | 0.043 | 0.488** | -0.106** |
| | Autumn | 0.101 | 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** |

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

293

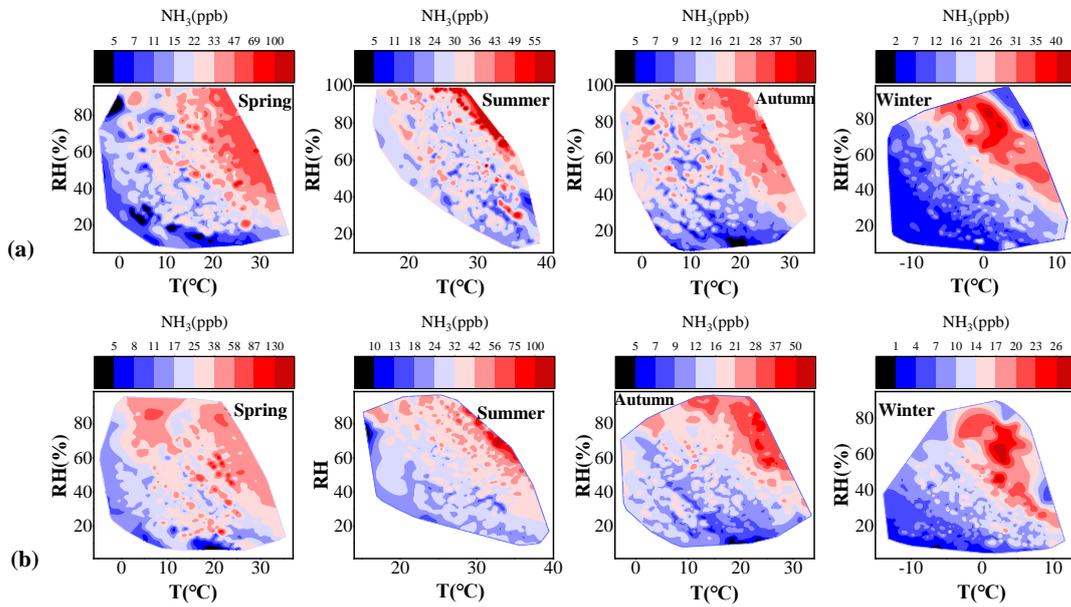
294 Figs. 5 display the seasonal mean diurnal variations in the NH₃ mixing ratio, temperature, and
295 relative humidity with their correlation coefficients (also see in Fig. S5) in different seasons at the urban
296 and suburban sites. At urban site, the diurnal variations in the NH₃ mixing ratio at the urban site were
297 positively (negatively) correlated with the temperature (relative humidity) in spring, while in contrast,
298 the diurnal variation of NH₃ mixing ratio were negatively (positively) correlated with temperature
299 (relative humidity) in summer and autumn, and less correlated in winter. At suburban site, the diurnal
300 variations in the NH₃ mixing ratio were positively (negatively) correlated with the temperature (relative
301 humidity) in the spring and summer, but less correlated in the fall and winter. Similar behaviors were
302 found in spring, but different in other seasons. In general, the annual diurnal behaviors of NH₃ with
303 temperature and relative humidity were different at the urban and suburban sites (see Fig. S6). Figure 6
304 displays the contour maps of the NH₃ mixing ratio, temperature, and relative humidity in different seasons

305 at the urban and suburban sites. As displayed in Fig. 6 and Fig. S7, the NH₃ mixing ratios at both sites
306 increased with the relative humidity at the same temperature and increased with the temperature at the
307 same relative humidity. In winter, when the temperature was low (< 0 °C), the NH₃ mixing ratios at both
308 sites often had low values except in high humidity (>60%). An increase in the temperature increased the
309 NH₃ mixing ratios; however, the NH₃ concentration at the suburban site was more significantly affected
310 by the temperature than that at the urban site (Table 3), indicating that volatile NH₃ sources might have
311 a higher contribution to the NH₃ concentration at the suburban site than at the urban site. A higher amount
312 of NH₃ removal through chemical transformation was expected during the day at the urban site than at
313 the suburban site because the urban site had a higher relative humidity and higher amounts of primary
314 particulate matter, NO_x, and SO₂ acid gas emissions than the suburban site did. In 2018, the
315 concentrations of PM_{2.5}, SO₂ and NO₂ were 50μg/m³, 5μg/m³, 43μg/m³ in Haidian, and 46μg/m³, 6μg/m³,
316 35μg/m³ in Changping, respectively, which were reported by Beijing Ecology and Environment
317 Statement, 2018.
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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.

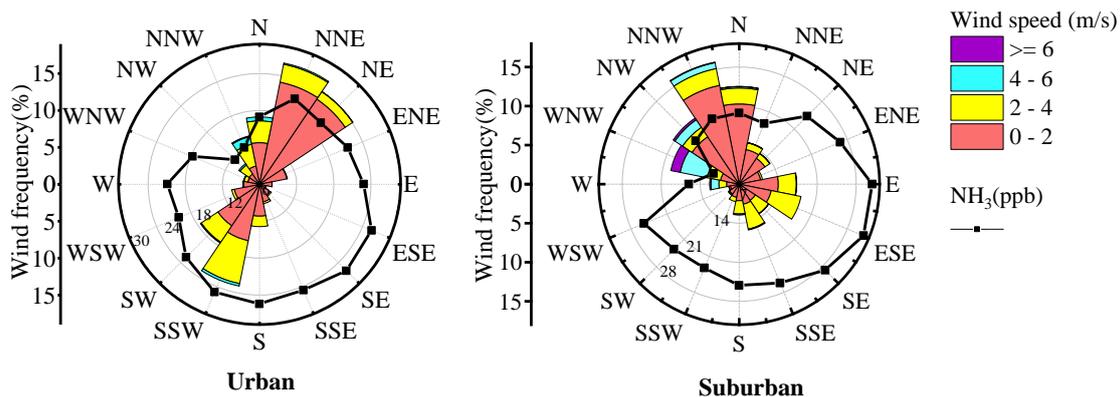


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Fig. 6. Contour maps of the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites in Beijing (a: Urban, b: Suburban).

To explore the influence of wind on the NH_3 mixing ratios, rose charts were drawn for the hourly

327 mean concentration of NH_3 , wind direction frequency, and wind speed during the observation period (Fig.
328 7). The large-scale wind circulation in the North China Plain is often influenced by the mountain-plain
329 topography; therefore, the dominant winds in this area originate from the south (often in the day) and
330 north (often at night). As displayed in Fig. 6, some differences existed in the distributions of the surface
331 wind between the urban and suburban sites. The dominant surface winds originated from the northeast
332 and southwest at the urban site and from the northwest and east at the suburban site. At the urban site,
333 the NH_3 mixing ratios were relatively high when the winds originated from the southern sectors and
334 relatively low when the winds originated from the northwest sectors. Therefore, under the action of the
335 southwest wind, a polluted air mass from the south of Beijing can be easily transported to the urban site.
336 Meng et al. (2017) examined the effect of long-range air transport on the urban NH_3 levels in Beijing
337 during the summer through trajectory analysis. The authors concluded that the air mass from the southeast
338 has a cumulative effect on the NH_3 concentration. Although the dominant wind direction at the suburban
339 site was different from that at the urban site, the NH_3 mixing ratios were relatively high in the south
340 sectors for both sites. Thus, winds from the southeast, south, and southwest had a cumulative effect on
341 the NH_3 mixing ratios at both the urban and suburban sites. The NH_3 mixing ratios were relatively low
342 when the wind originated from the northwest sector at urban site and from the west sector at the suburban
343 site, in which the wind speed was strong, which indicated that the northwest/west wind could promote
344 NH_3 dilution and diffusion.



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

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Heavy rainfall occurred on a few days in Beijing in 2018. Heavy precipitation occurred for a long

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duration on August 18, 2018 (Fig. 8). Before the rainfall, the NH_3 concentration at the urban site was

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higher than the average level in August. After the rainfall occurred, the NH_3 concentration decreased

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rapidly, and it was significantly lower than the mean value in August. However, the diurnal variation of

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NH_3 on the rain day did not differ considerably from the average diurnal variation in August. On August

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18, 2018, the NH_3 mixing ratio at the suburban site remained at a low level during the rainfall period and

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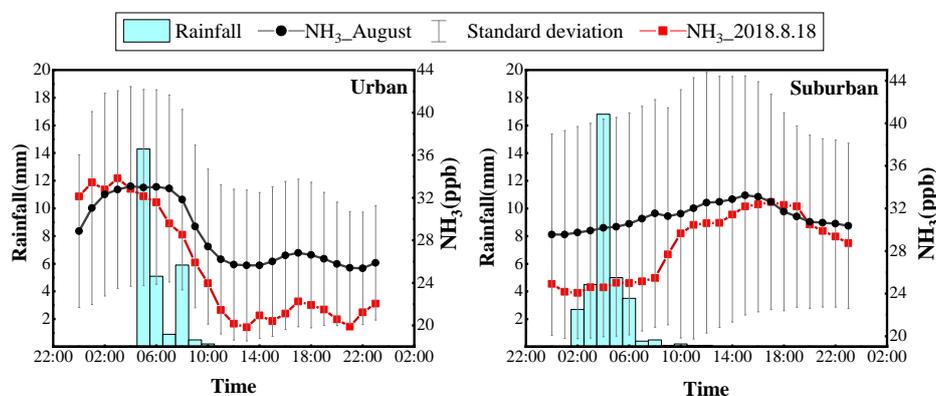
was considerably lower than the mean NH_3 concentration in August. However, the NH_3 mixing ratio

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increased rapidly after the precipitation and reached the mean level at 17:00. The rainfall might have an

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obvious clearing effect on NH_3 but needed more cases to support.



357

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

359

360 4. Conclusion

361 In this study, the atmospheric NH₃ concentrations at an urban site and a suburban site in Beijing
362 were continuously and simultaneously observed from January 2018 to January 2019. The mean NH₃
363 mixing ratios were 21 ± 14 and 22 ± 15 ppb at the urban and suburban sites, respectively. The annual
364 average NH₃ mixing ratio at the suburban site was higher than that at the urban site. Moreover, the
365 variation range of the NH₃ mixing ratio was larger at the suburban site than at the urban site. In the
366 summer and spring, the NH₃ mixing ratio at the suburban site was higher than that at the urban site. In
367 the autumn and winter, the NH₃ mixing ratio at the suburban site was lower than that at the urban site.
368 The highest NH₃ mixing ratios at the urban and suburban sites were observed in July. The lowest NH₃
369 mixing ratio at the urban site occurred in February, and the lowest NH₃ mixing ratio at the suburban site
370 occurred in January. In the past decade, the concentration of NH₃ in Beijing did not change considerably,
371 and the NH₃ levels in Beijing were higher than those in other large cities.

372 The hourly mean NH₃ mixing ratios at the urban site were highly correlated ($R = 0.849$, $P < 0.01$)
373 with those at the suburban site. However, the mean diurnal variations in the NH₃ mixing ratios at the
374 urban and suburban sites were different. At the urban site, low NH₃ mixing ratios were observed in the
375 day and high NH₃ mixing ratios were observed in the night. The opposite trend was observed at the
376 suburban site. Although both sites were under the influence of similar weather systems, the seasonal
377 diurnal variations in the NH₃ mixing ratio were different at the urban and suburban sites. This result
378 indicated that NH₃ sources had different contributions to the NH₃ levels at the urban and suburban sites.

379 The influence of meteorological factors on the NH₃ mixing ratio was complex. At the same
380 temperature, the NH₃ mixing ratios increased with the relative humidity at the urban and suburban sites.

381 At the same relative humidity, the NH₃ mixing ratios increased with the temperature at both sites. The
382 relative humidity had the strongest influence on the NH₃ mixing ratio in different seasons at the two sites.
383 No strong correlation was observed between the NH₃ concentration and the temperature in the summer
384 and autumn at the two sites. A high wind speed promoted a decrease in the NH₃ concentration. The NH₃
385 mixing ratios were higher when the winds originated from the south than when the winds originated from
386 the north and northwest. Rainfall had a certain scavenging effect on NH₃; however, it had little effect on
387 the diurnal variations in the NH₃ concentration.

388

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

391

392 **Author contributions.** ZL and WL developed the idea for this paper, formulated the research goals, and
393 carried out the measurement at urban site. WP and ZM carried out the NH₃ field observations at the
394 suburban site.

395

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

397

398 **Acknowledgments.** This study was funded by the National Natural Science Foundation of China
399 (Grant No. 91744206) and the Beijing Municipal Science and Technology (Z181100005418016).

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