1   Measurement report: Exploring the NH$_3$ behaviors at urban and suburban Beijing:

2   Comparison and implications

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8 **ABSTRACT**

9 Ammonia (NH$_3$) plays an important role in particulate matter formation, and hence its atmospheric level

10 is relevant to human health and climate change. Due to different relative distributions of NH$_3$ sources,

11 the concentrations of atmospheric NH$_3$ may behave differently in urban and rural areas. However, few

12 parallel long-term observations of NH$_3$ to reveal the different behaviors of the NH$_3$ concentrations at the

13 urban and rural sites in a same region. In this study, online ammonia analyzers were used to continuously

14 observe atmospheric NH$_3$ concentrations at an urban site and a suburban site in Beijing from January 13,

15 2018, to January 13, 2019. The observed mixing ratio of NH$_3$ averaged 21 ± 14 ppb (range: 1.6–133 ppb)

16 at the urban site and 22 ± 15 ppb (range: 0.8–199 ppb) at the suburban site. The NH$_3$ mixing ratios at the

17 urban and suburban sites exhibited similar seasonal variations, with high values in summer and spring

18 and low values in autumn and winter. The hourly mean NH$_3$ mixing ratios at the urban site were highly

19 correlated ($R = 0.849$, $P < 0.01$) with those at the suburban site. However, the average diurnal variations

20 in the NH$_3$ mixing ratios at the urban and suburban sites differed significantly, which implies the different

21 contributions of NH$_3$ sources and sinks at the urban and suburban sites. In addition to the emission

22 sources, meteorological factors were closely related to the changes in the NH$_3$ concentrations. For the

23 same temperature (relative humidity) at the urban and suburban sites, the NH$_3$ mixing ratios increased

24 with relative humidity (temperature). Relative humidity was the factor with the strongest influence on

25 the NH_3 mixing ratio in different seasons at the two sites. The relationships between the NH_3

26 concentrations and temperature (relative humidity) varied from season to season and showed differences

27 between the urban and suburban sites. The reasons for the different relationships need to be investigated

28 in future studies. Higher wind speed mainly from the northwest sector lowered the NH_3 mixing ratios at

29 both sites. Similar with other primary pollutants in Beijing, the NH_3 mixing ratios were high under

30 impacts of air masses from the south sector.

31 **Keywords:** NH_3; variations; simultaneous observation

32

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

55   expansion of agricultural activities, control of SO_2 and NO_x, and increase in temperature (Warner et al.,

56   2017). This increase in the NH_3 concentration might weaken the effectiveness of SO_2 and NO_x emission

57   control in reducing $PM_{2.5}$ pollution (Fu et al., 2017).

58   The North China Plain is a region with high NH_3 emission (Zhang et al., 2017), and Beijing has one

59   of the highest NH_3 concentrations in the world (Chang et al., 2016b; Pan et al., 2018). Compared with

60   studies on pollutants such as SO_2 and NO_x, considerably fewer studies have been conducted on the NH_3

61   concentration in Beijing. Chang et al. (2016a) collected gaseous NH_3 samples during the 2014 APEC

62   summit (October 18 to November 29, 2014) in the Beijing urban area and concluded that the overall

63   contributions of traffic, garbage, livestock, and fertilizers to the NH_3 concentration were 20.4%, 25.9%,

64   24.0%, and 29.7%, respectively. According the data from Huang et al (2012), the NH_3 emissions in

65   Beijing were from livestock and poultry farming (34.55%), nitrogen-fixing plants (33.57%), fertilizer

66   use (13.06%), household garbage treatment (8.29%), traffic emissions (5.20%), industrial emissions

67   (0.14%), biomass combustion (0.42%), and agricultural soil (0.84%). Zhang (2016) measured the NH_3

68   concentrations in urban and rural areas of Beijing from January to July 2014 and found that NH_3

69   concentration in urban areas was approximately 65% higher than that in rural areas. Meng et al. (2011)

70   reported that the highest NH_3 concentration in Beijing occurred in summer and the lowest one occurred

71   in winter, and their results indicated traffic to be a significant source of NH_3 in urban areas. Zhang et al.

72   (2018) reported the vertical variability of NH_3 in urban Beijing based on one-year passive sampling in

73   2016/2017 and concluded that local sources such as traffic emissions were important contributors to

74   urban NH_3. Meng *et al.* (2020) investigated the significant increase in winter NH_3 and its contribution to

75   the increasing nitrate in $PM_{2.5}$ from 2009 to 2016, and they also concluded that vehicles exhaust was an

76   important contributor to NH_3 in urban Beijing in winter.

77   Currently, NH₃ is not included in the routine environmental monitoring operation in China. Research

78   data on NH₃ monitoring, particularly on the synchronous observations of NH₃ concentrations with a high

79   temporal resolution in urban and suburban areas, are relatively scarce. In this study, high-time-resolution

80   observations of NH₃ were obtained simultaneously at an urban site and a suburban site in Beijing. The

81   variation characteristics and influencing factors of the NH₃ concentration were analyzed with

82   meteorological data to provide a scientific basis for NH₃ pollution control in Beijing.

83   **2. Materials and methods**

84   *2.1. Measurement sites*

85   From January 2018 to January 2019, continuous and simultaneous observations of atmospheric NH₃

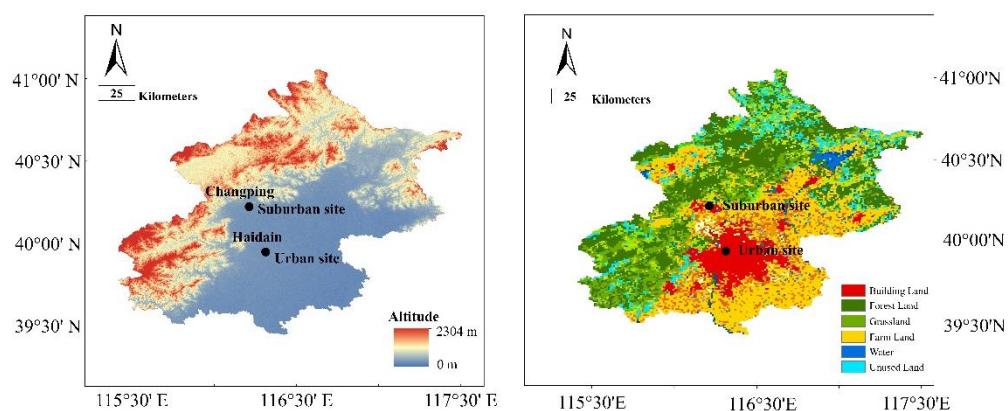
86   were conducted at an urban site and a suburban site in Beijing. The urban site was located on the roof of

87   the Science and Technology Building of Minzu University of China (39.95°N, 116.32°E, altitude: 102

88   m) and the suburban site was in the Changping Meteorological Station (40°13′N, 116°13′E, altitude: 77

89   m). The suburban site is in the NW direction relative to the urban site and the shortest distance between

90   these two sites is approximately 32 km (Figure 1). More farm land and glass land are around the suburban

91   site than the urban site.

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

94

95   *2.2. Measurements and data acquisition*

96   NH$_3$ concentrations were measured using two NH$_3$ analyzers (Ammonia Analyzer-Economical, Los

97   Gatos Research Inc., USA), which have the minimum detection limit of <0.2 ppb and the maximum drift

98   of 0.2 ppb/24hrs. The NH$_3$ analyzers were deployed in air-conditioning rooms. These analyzers use off-

99   axis integrated cavity output spectroscopy (OA-ICOS) technology, which is a fourth-generation cavity-

100 enhanced absorption technique, to simultaneously measure NH$_3$ and water vapor (H$_2$O) in the atmosphere.

101 The incident laser beam of the OA-ICOS technology deviates from the optical axis, which differs from

102 the traditional coaxial incidence mode. The axial incidence mode of the OA-ICOS technology can

103 increase the optical path, stimulate additional high-order transverse modes, effectively suppress the noise

104 of the cavity mode, reduce the cross interferences and errors due to contaminants existing in the cavity,

105 and improve the detection sensitivity (Baer *et al*., 2002; Baer *et al*., 2012). The analyzer method is a

106 quasi-absolute measurement, which theoretically does not require calibration. However, to ensure the

107 comparability of the obtained data with other monitoring data, NH$_3$ standard gas (Beijing AP BAIF Gases

108 Industry Co., Ltd.) was used for comparison measurement before the observation. The recorded

109 concentrations were revised with respect to the reference NH$_3$ concentration in the standard gas mixture.

110   Ambient air was drained at >0.4 L/min through Teflon lines (1/4'OD) from a manifold. The lengths

111 of the Teflon lines were designed as short as possible (less than 2 m from the manifold). Particulate

112 matters were filtered by Teflon membranes with a pore size less than 5 μm. Since NH$_3$ easily "sticks" to

113 surfaces (like inside walls of tubes), heated sample lines were suggested by many measurement studies.

114 However, according our test (Fig. S1) in the lab, when heating (70℃) was on, there did have a peak

115 lasting 5–6 min minutes and then deceasing to the normal levels in ambient air, which means a new

116 balancing process has been established in less than 10 min. This suggests that heating is not necessarily

117   a solution for NH$_3$ sticking. Keeping the relatively stable balance between adsorption and desorption of

118   NH$_3$ in the sampling system is important. When tested using air of different humidity, only very sharply

119   changes of humidity obviously influenced and changed the balance, and a new balance needed tens of

120   minutes to reestablished (Fig. S2). Under the normal weather conditions, humidity changes in a relatively

121   smoothing way unless a quickly changing weather system, like rain, is approaching. The minute-level

122   data were converted into hourly averages in the data analysis process and the hourly resolution can

123   smooth the effect to some extent caused by variations in humidity and temperature during the observation.

124   The balancing idea was also used to carry out multi-point calibrations on NH$_3$ analyzers (Fig. S3).

125   A high mixing ratio (e.g. 400 ppb or higher) of NH$_3$ mixing gases were firstly produced by a dynamic

126   diluter and measured by the NH$_3$ analyzers overnight. After the signals reached the stable level, other

127   lower span values were switched in turn. At each span point, the measurement time was lasting at least

128   40 minutes or longer. Then a linear regression function was obtained with R^2 higher than 0.999.

129   Nowadays, NH$_3$ in compressed gas cylinder is also trustworthy, as confirmed by the comparison with the

130   NH$_3$ standard in a permeation tube (Fig. S4).

131   Totally, 7645 and 8342 valid hourly mean observations were obtained for the urban (Haidian) and

132   suburban (Changping) sites, respectively. In addition, the urban and suburban meteorological data

133   (temperature, relative humidity, wind direction, and wind speed) during the sampling period were

134   obtained from the Haidian Meteorological Observation Station and Changping Meteorological Station,

135   respectively.

136   **3. Results and discussion**

137   *3.1. Overall variations in the NH$_3$ mixing ratios*

138   Fig. 2 displays the time-series variations in the NH$_3$ mixing ratios, temperatures, and relative

139   humidity at the urban and suburban sites in Beijing. At the urban site, the mean ± 1σ, median, maximum,

140   and minimum values of the hourly average NH₃ mixing ratio during the observation period were 21 ± 14

141   ppb, 17 ppb, 133 ppb and 1.6 ppb, respectively. At the suburban site, the corresponding values were 22

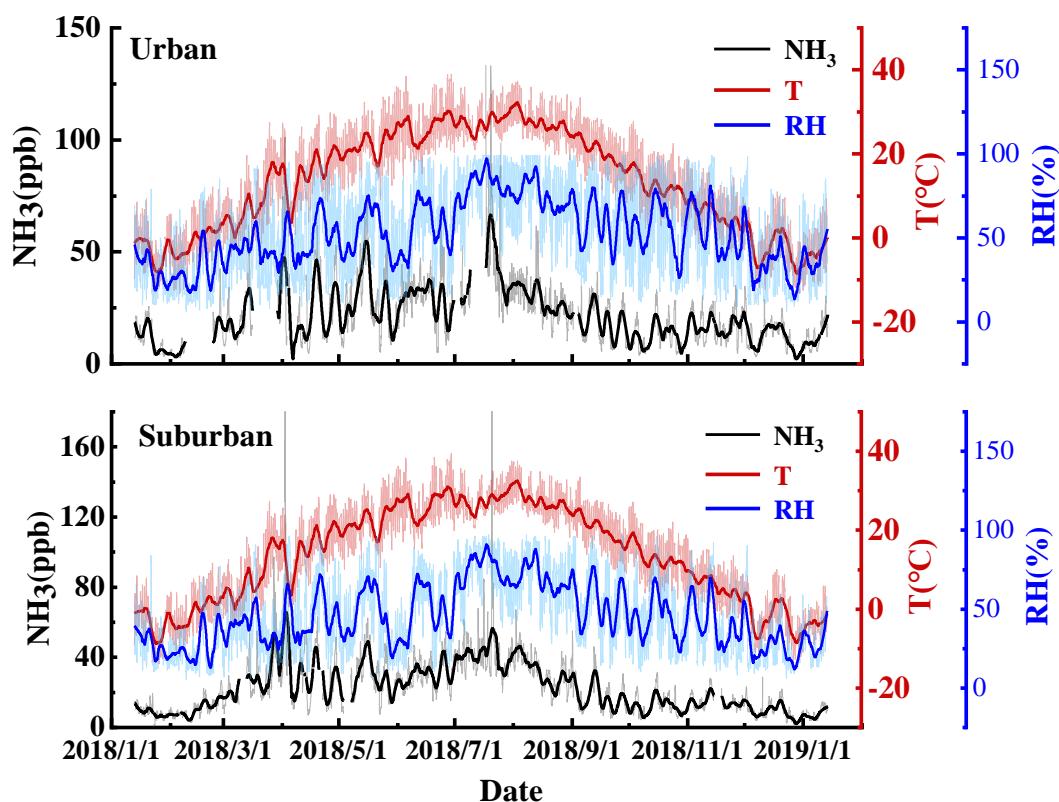
142   ± 15 ppb, 18 ppb, 199 ppb, and 0.8 ppb, respectively. The annual average and range of the NH₃ mixing

143   ratio at the suburban site were marginally higher than those at the urban site. The characteristics of the

144   weekly smoothed data indicate that the NH₃ variations and temperature/humidity fluctuations at the two

145   sites were practically consistent, which suggests that both sites were under the influence of similar

146   weather systems. The hourly mean NH₃ concentrations at the urban site were significantly correlated (R

147    $= 0.849$, $P < 0.01$) with those at the suburban site.

149   **Fig. 2.** Temporal variations in the hourly average NH₃ mixing ratios, temperatures (T) and relative humidity (RH) at the urban and suburban
150   stations in Beijing. Continuous thick lines were smoothed with 168 points (7 days) by using the Savitzky–Golay method.

151

152   Table 1 shows the comparison of atmospheric NH₃ concentrations (ppb) observed in different areas.

153   Meng et al. (2011) obtained an average NH_3 mixing ratio of 22.8 ± 16.3 ppb for the period 2008-2010 in

154   Beijing urban area, which is very close to our result (21 ± 14 ppb) for 2018-2019. Therefore, the annual

155   average NH_3 mixing ratio in urban Beijing did not change significantly from 2008 to 2019. Moreover,

156   results from this study and Meng et al. (2011) indicate that the NH_3 concentrations at the urban and

157   suburban sites were higher than those in the background areas. The observed NH_3 concentrations in

158   Beijing were higher than those in northwest China (Meng et al., 2010) and the Yangtze River Delta region

159   (Chang et al., 2019). The average annual NH_3 concentration in the urban area of Shanghai, a mega city

160   in the Southeast China (31° N), was approximately 50% lower than that in urban Beijing. This might be

161   related to the fact that the North China Plain, in which Beijing is located, is one of the most intensive

162   agricultural production regions in China. The differences in the soil properties of Beijing and Shanghai

163   may be another reason because the loss of soil NH_3 can increase with an increase in the soil pH (Ju et al.,

164   2009). Shanghai and its surrounding areas are dominated by acidic soil of paddy fields (Zhao et al., 2009),

165   whereas Beijing is dominated by the alkaline soils of dry land (Wei et al., 2013). In addition, the climate

166   in Beijing is much drier than in Shanghai so that less atmospheric NH_3 in Beijing can be removed than

167   in Shanghai by wet deposition.

168   **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

169

170   Table 1 also shows observational results of atmospheric NH_3 from some other countries. The NH_3

171   mixing ratios in the United States (Edgerton et al., 2007; Nowak et al., 2006; Zhou et al. 2019), Scotland

172   (Burkhardt et al., 1998), Canada (Hu et al., 2014), Japan (Osada et al., 2019), and Germany (Vogt et al.,

173   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

174   considerably lower than those in Beijing. However, Delhi, India (Saraswati et al., 2019), exhibited higher

175   NH_3 mixing ratio (53.4±14.9 ppb) than Beijing did. This result might be attributed to the well-developed

176   livestock breeding activities in Delhi. This comparison indicates that in the decade before 2019, the NH_3

177   concentration in Beijing did not change considerably, but it is of the highest in big cities in China and

178   much higher than those observed in developed countries in America, Europe and Asia.

179   *3.2. Seasonal variations*

180   Fig. 3 displays the monthly statistics for the NH_3 mixing ratios at the urban and suburban sites in

181   Beijing. The seasonal variations in the NH_3 mixing ratios were very similar at the urban and suburban

182   sites, with higher mixing ratios in the spring and summer and low ones in the autumn and winter. The

183   daily mean concentrations fluctuated considerably in the spring, particularly in April. The highest mean

184   NH_3 concentrations at the urban and suburban sites were 42 ± 17 ppb and 42 ± 8.2 ppb, respectively. Both

185   occurred in July, when the NH_3 concentrations fluctuated considerably as well. On average, the seasonal

186   NH_3 mixing ratios at the urban and suburban sites can be arranged as follows: summer > spring > autumn >

187   winter. The main grain crops in the rural area of Beijing are corn and wheat. Corn is categorized as spring

188   corn and summer corn, which are sown in April and June, respectively. Usually, a large amount of base

189   fertilizer is applied when planting corn and the topdressing after 2 months. Wheat is sown from

190   September to October, and the topdressing is applied in the following spring. The volatilization of

191   nitrogen fertilizers can cause an increase in atmospheric NH_3 mixing ratios and its fluctuations in

192   fertilization seasons (Zhang et al., 2016). In addition, the high temperature in summer should also be

193   responsible to the high NH_3 mixing ratios in this season. An increase in the temperature can increase the

194   biological activity and thus enhance the NH_3 production and emission. High temperature is also

195   conducive for the volatilization of the urea and diammonium phosphate applied to crops. Moreover, the

196   equilibrium among ammonium nitrate particles, gaseous NH_3, and nitric acid is transferred to the gas

197   phase at high temperature, which increases the NH_3 concentration (Behera et al., 2013). Sewage treatment,

198   household garbage, golf courses, and human excreta are crucial NH_3 sources that are easily neglected

199   (Pu et al., 2020). The relatively low NH_3 concentrations in the autumn and winter might be caused by the

200   decrease in NH_3 emission in the soil and vegetation, the decrease in the NH_4NO_3 decomposition capacity

201   at low temperatures, and the reduced human activities caused by a large floating population returning to

202   their native locations outside Beijing during the Spring Festival (Liao et al., 2014). In the spring and

203   summer, the NH_3 mixing ratios at the suburban site were higher than those at the urban site, which might

204   be related to the higher agricultural activity around the suburban site. In the autumn and winter, the NH_3

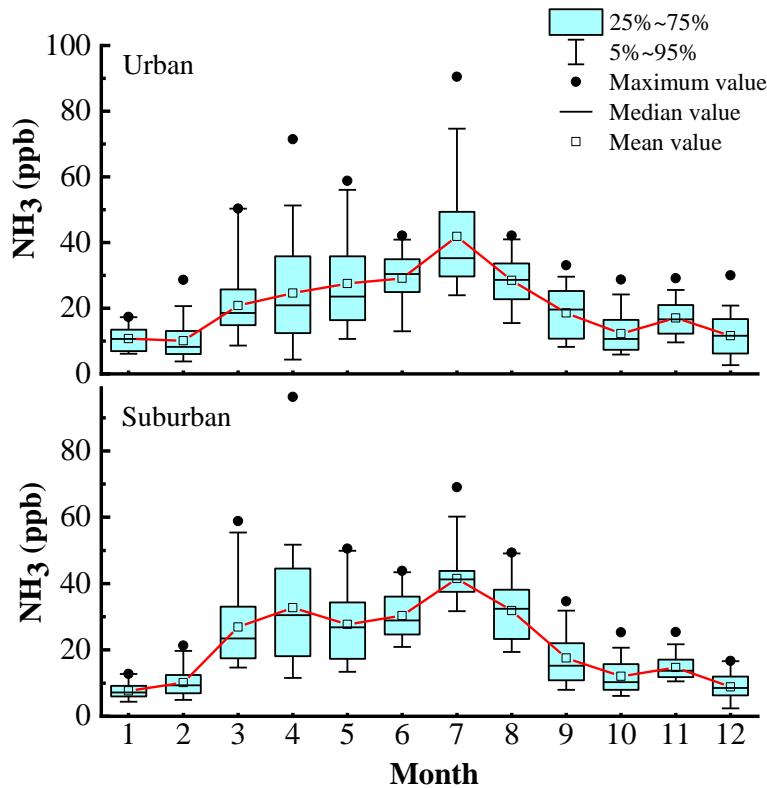
205   mixing ratios at the urban site were marginally higher than those at the suburban site. In the autumn and

206   winter seasons, the influences of agricultural activities on the NH$_3$ concentration were weakened,

207   whereas the influences of other sources (such as traffic sources) were enhanced. According to Wang et

208   al. (2019), the traffic NH$_3$ emission per unit area in Haidian (urban site) was three times higher than that

209   in Changping (suburban site). This difference in traffic source emissions might have resulted in higher

210   NH$_3$ concentrations at the urban site than at the suburban site in the autumn and winter.

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212   **Fig. 3.** Monthly statistical variation in the NH$_3$ mixing ratios at the urban and suburban sites in Beijing.

213

214   **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
	Annual	21	14	1.6	17	133
	Spring	25	16	1.9	21	101
Urban						
	Summer	32	12	5.0	30	133
	Autumn	16	7.5	3.8	15	41

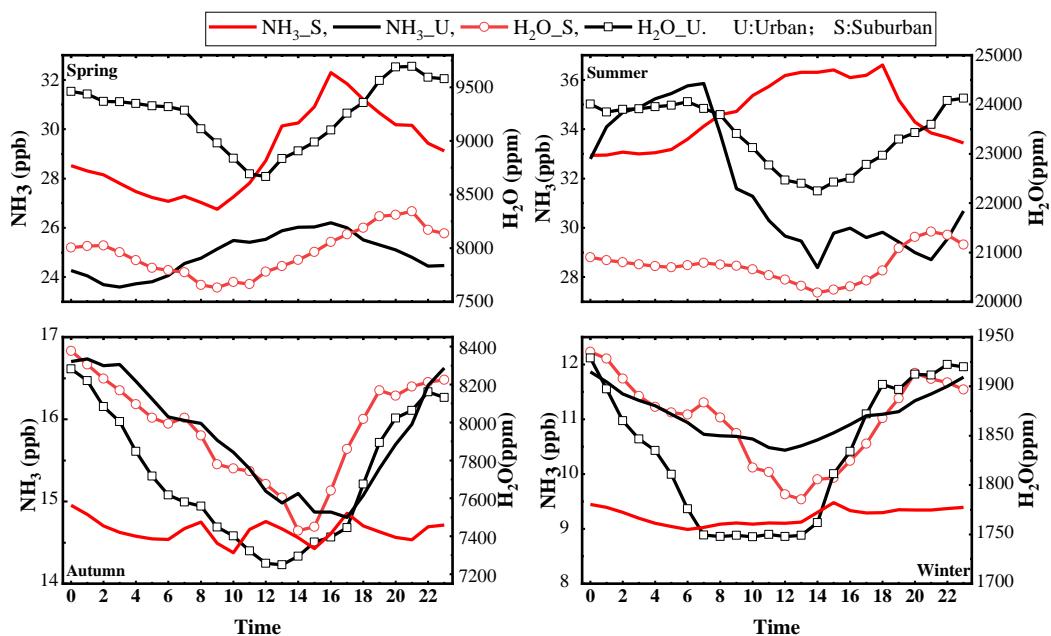
		Winter	11	6.7	1.6	9.9	42
		Annual	22	15	0.8	18	198
		Spring	29	16	6.8	26	180
Suburban		Summer	35	12	12.1	33	199
		Autumn	15	6.8	4.1	13	55
		Winter	9.2	4.5	0.8	8.4	29

215

216   *3.3. Diurnal variations*

217   Figure 4 displays the average diurnal variations in the NH_3 and H_2O mixing ratios in different

218   seasons at the urban and suburban sites in Beijing. Ambient NH_3 exhibited different diurnal behaviors in

219   different seasons.

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221   **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.

222

223   In spring, the average diurnal variations in the NH_3 mixing ratio were similar at the urban and

224   suburban sites. The diurnal variations exhibited a single-peak pattern with high values in the daytime and

225   low values at night. The NH_3 mixing ratio began to increase in the morning, reached its maximum value

226   at 16:00, and then decreased gradually. The lowest mixing ratios at the urban and suburban sites occurred

227   at 03:00 and 09:00, respectively. The NH_3 mixing ratio began to increase earlier at the urban site than at

228   the suburban site. A plausible explanation to the earlier increase in the NH_3 emission at the urban site is

229   the traffic emission in the morning rush hours. In spring, the mixing ratio of NH_3 was higher at the

230   suburban site than that at the urban site, with an average difference of 4.1 ppb and a maximum difference

231   of 6.1 ppb. The average diurnal amplitude of the NH_3 mixing ratio at the suburban site was 5.3 ppb,

232   which was higher than that (2.6 ppb) at the urban site. At the urban site, the average diurnal variations in

233   the NH_3 and H_2O mixing ratios exhibited nearly opposite trends. The H_2O mixing ratio had high values

234   in the night and low values in the day. At the suburban site, the variation characteristics of NH_3 and H_2O

235   were very similar; however, the peak NH_3 concentration occurred 5 hours earlier than the peak H_2O

236   concentration. In spring, in contrast to the NH_3 mixing ratio, the H_2O mixing ratio at the urban site was

237   1279 ppm higher than that at the suburban site.

238   The diurnal variation in the NH_3 mixing ratio at the suburban site in summer was similar to that in

239   spring. This phenomenon was also observed in the rural areas of Shanghai by Chang et al. (2019). The

240   diurnal variations of NH_3 at the suburban site were considerably affected by the temperature and the

241   contribution from volatile NH_3 sources. However, the diurnal summer variation of NH_3 at the urban site

242   was completely different from that at the suburban site. The summer level of NH_3 at the urban site was

243   obviously lower during the daytime and evening than that at the suburban site, increased gradually from

244   21:00 to levels higher its suburban counterpart, dropped after reaching its peak value at 7:00, and then

245   reached its lowest value at 14:00. The diurnal pattern (with a peak in early morning) has been observed

246   in other areas, such as rural (Ellis et al., 2011), urban (Gong et al., 2011), and steppe areas located far

247   away from human activity (Wentworth et al., 2016). Kuang et al. (2020) believed that such diurnal pattern

248   of NH_3 was caused by the evaporation of dew in the morning, which resulted in the release of NH_3

249   originally stored in the droplets. A lag was observed between the changes in the NH_3 and H_2O

250   concentrations in the early morning, which supported the hypothesis of Kuang et al (2020). In addition,

251   the increase in the NH_3 concentration in the morning might have been caused by the breakup of the

252   boundary layer formed at night. The downward mixing of air with a higher NH_3 concentration in the

253   residual layer led to a morning increase in the NH_3 concentration on the ground (Bash et al., 2010). In

254   summer, the NH_3 concentrations at the suburban site were significantly higher than those at the urban

255   site during the daytime and first half of the night. The average diurnal amplitude of the NH_3 concentration

256   was 7.5 ppb and 3.7 ppb at the urban and suburban sites, respectively. Similar to the situation in spring,

257   the H_2O concentrations at the urban site were significantly higher than those at the suburban site in the

258   summer.

259   In autumn, the NH_3 concentration at the suburban site was relatively stable and remained nearly all

260   the time lower than that at the urban site, which showed low values during the day and high values during

261   the night, with a peak at midnight and a minimum (about 2.0 ppb lower than the peak) at 17:00. The H_2O

262   concentration was marginally lower (250 ppm) at the urban site than at the suburban site. The diurnal

263   profiles of H_2O at both sites resemble that of NH_3 at the urban site, but the lowest values of H_2O occurred

264   earlier than the lowest value of NH_3 at the urban site.

265   The diurnal patterns of NH_3 and H_2O in winter were similar to those in autumn though the mixing

266   ratios of NH_3 and H_2O were lower than their autumn counterparts. There were two slight differences: (1)

267   the mixing ratios of NH_3 at both sites exhibited lower fluctuations than those in autumn and (2) the

268   mixing ratio of NH_3 at the urban site reached its minimum in winter earlier than that in autumn.

269   The above results indicate that although the two sites were under the influence of similar weather

270 systems, the diurnal variations in the NH_3 mixing ratios at the two sites were different in different seasons.

271 This finding suggests that different NH_3 sources and possibly sinks had different contributions to the NH_3

272 concentrations at the urban and suburban sites. Additional studies should be conducted to better

273 understand the behaviors of atmospheric NH_3 and its influencing factors.

274 *3.4. Effect of meteorological factors on the NH_3 levels*

275 Table 3 presents the annual and seasonal correlation coefficients between the daily means of NH_3

276 mixing ratios and those of the temperature, relative humidity, and wind speed at the two sites. Annually,

277 the NH_3 mixing ratios at both sites were positively correlated with temperature and relative humidity and

278 negatively correlated with wind speed, and the correlations are all highly significant. However, the

279 correlations deteriorated somewhat in warm seasons. In summer and autumn, no significant correlations

280 were noted between ambient NH_3 and temperature at the two sites. The correlation between NH_3 and

281 wind speed in summer was much weaker than in the other seasons. The relative humidity was stronger

282 correlated with the NH_3 concentration at the two sites than temperature, which can be perceived in Fig 2.

283 Also, the correlation between NH_3 and relative humidity did not vary much from season to season. This

284 implies a possibility that relative humidity exerts a certain influence on the variation of the NH_3 level in

285 the surface layer.

286

287 Table 3. Correlations between the daily mean values of NH_3 and meteorological elements (Spearman's

288 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**

		0.745**	0.730**	-0.325**
	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.

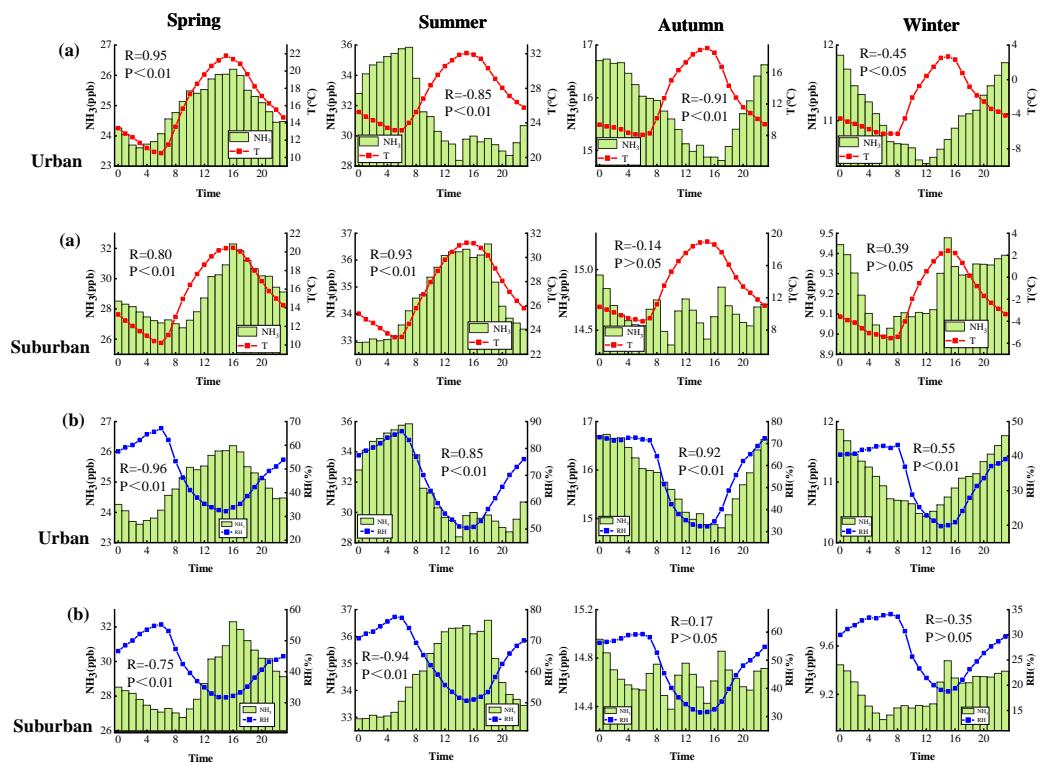



Fig. 5. Diurnal variations in and correlation coefficients between the NH$_3$ mixing ratios and temperature (a), relative humidity (b) in

different seasons at the urban and suburban sites in Beijing.

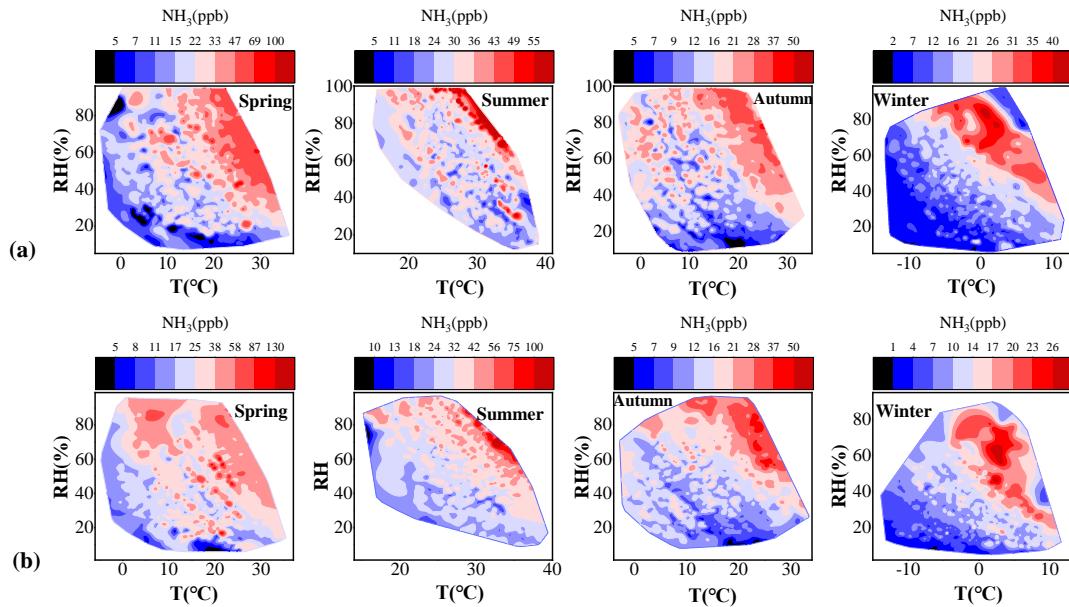



Fig. 6. Contour maps of the NH_3 mixing ratio, temperature, and relative humidity in different seasons at the urban and suburban sites in Beijing (a: Urban, b: Suburban).

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 long-

336 range air transport on the urban NH$_3$ levels in Beijing during the summer through trajectory analysis.

337 They concluded that the air mass from the southeast has a cumulative effect on the NH$_3$ concentration.

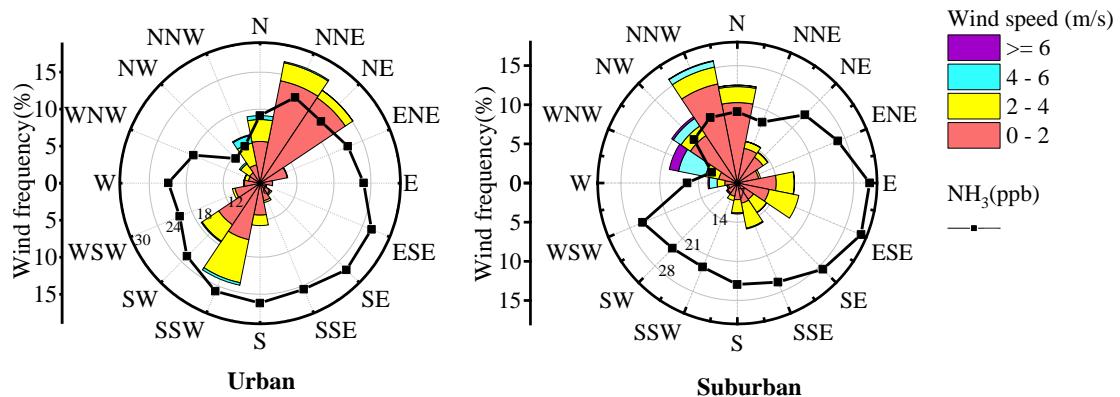
338 Although the dominant wind direction at the suburban site was different from that at the urban site, the

339 NH$_3$ mixing ratios were also relatively high in the south sectors. Thus, winds from the southeast, south,

340 and southwest can elevate levels of atmospheric NH$_3$ at both the urban and suburban sites. The NH$_3$

341 mixing ratios were relatively low when air masses originated from the northwest sector at urban site and

342 from the west sector at the suburban site. The west and northwest winds were stronger and promoted the

343 dilution and diffusion of NH$_3$ emitted into the boundary layer.

344

345 **Fig. 7.** Rose maps of the NH$_3$ mixing ratios, wind frequency, and wind speed in different wind direction sectors.

346

347 As a water-soluble gas, NH$_3$ can be impacted by precipitation. Heavy rainfall occurred on August

348 18, 2018 (Fig. 8). Before the rainfall, the NH$_3$ concentration at the urban site was higher than the average

349 level in August. After the rainfall, the NH$_3$ concentration decreased rapidly, and it was significantly lower

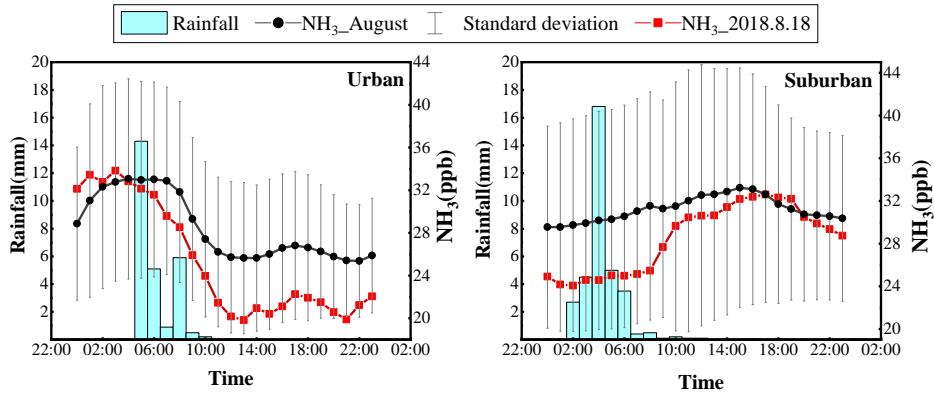
350 than the mean value in August. However, the diurnal pattern of NH$_3$ on that day did not differ

351 considerably from the average diurnal pattern in August. On the same day, the NH$_3$ mixing ratio at the

352 suburban site remained at a low level during the rainfall period, which was considerably lower than the

353 August mean NH$_3$ concentration during the same time of day. However, the NH$_3$ mixing ratio increased

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

355 clearing effect on NH_3 but more case studies are needed to reach a robust conclusion.

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

358

359 **4. Conclusions**

360   In this study, the atmospheric NH_3 concentrations at an urban site and a suburban site in Beijing

361 were continuously and simultaneously observed from January 2018 to January 2019. The mean NH_3

362 mixing ratios were 21 ± 14 ppb and 22 ± 15 ppb at the urban and suburban sites, respectively. These NH_3

363 levels are among the highest mean values found in China and much higher than those reported for some

364 developed countries in America, Europe and Asia. In the summer and spring, the NH_3 mixing ratios at

365 the suburban site were slightly higher than those at the urban site. In the autumn and winter, however,

366 the situation was reversed. The highest NH_3 mixing ratios at the urban and suburban sites were all found

367 in July. The lowest NH_3 mixing ratio occurred in February at the urban site and in January at the suburban

368 site. A comparison with data from literature shows that the mean concentration of NH_3 in Beijing did not

369 change considerably in the decade before 2019.

370   The hourly mean NH_3 mixing ratios at the urban site were highly correlated ($R = 0.849$, $P < 0.01$)

371 with those at the suburban site. However, the mean diurnal variations in the NH_3 mixing ratios at the

372 urban and suburban sites were different. At the urban site, lower NH_3 mixing ratios were observed in the

373 daytime and higher ones at night. The opposite trend was observed at the suburban site. Although both

374 sites were under the influence of similar weather systems, the seasonal-diurnal variations in the NH_3

375 mixing ratio were different at the urban and suburban sites, suggesting that NH_3 sources had different

376 relative contributions to the NH_3 levels at the urban and suburban sites.

377   The relationship of meteorological factors with the NH_3 mixing ratio was complex. Overall, the NH_3

378 mixing ratios increased with relative humidity and temperate at both sites. Relative humidity was stronger

379 correlated with the NH_3 mixing ratio at both sites. The situation in different seasons varied and was site-

380 dependent, which warrants further studies. A high wind speed (mainly under northwesterly) suppressed

381 the levels of NH_3 at both sites. The NH_3 mixing ratios were higher under southerly wind conditions.

382 Rainfall had a certain scavenging effect on NH_3 but had little effect on the diurnal variations in the NH_3

383 concentration.

384

385 *Data availability.* The data of stationary measurements are available upon request to the contact author

386 Weili Lin (linwl@muc.edu.cn).

387

388 *Author contributions.* ZL and WL developed the idea for this paper, formulated the research goals, and

389 carried out the measurement at urban site. WP and ZM carried out the NH_3 field observations at the

390 suburban site.

391

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

393

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