



Measurement report: Assessing the ammonia characteristics over a high-altitude mountain site in Shanxi province, China: a comparison with the observations in the North China Plain

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Abstract:

Ammonia (NH₃) acts as the dominant alkaline gas and plays a crucial role in atmospheric chemistry, thereby influencing air quality and ecological systems. Previous NH₃ measurement studies have primarily focused on near-ground environments or relied on passive sampling methods; however, continuous, high-resolution NH₃ observations at high-altitude sites remain scarce. This study investigated NH₃ characteristics at a high-altitude mountain site (WTM) in northern China, using high-resolution and real-time measurement data spanning a full annual cycle. It further conducted comparative analyses with a regional background site (SDZ) and an urban site (BMS) to better understand the regional features of NH₃ in northern China. A multi-method approach was employed, integrating in situ NH₃ measurements, meteorological data analyses, Convergent Cross Mapping (CCM), Potential Source Contribution Function (PSCF), and WRF-Chem modeling, to identify NH₃ source regions and clarify underlying transport mechanisms. The results indicated that NH₃ emissions from the North China Plain (NCP) can reach WTM and SDZ through distinct circulation patterns: mountain-plain circulations for WTM and mountain-valley circulations for SDZ. Notably, despite significant differences in altitude, geography, and pollutant transport

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32 mechanisms between WTM and SDZ, regional agricultural emissions in the NCP were
33 the dominant factor driving the similarity in NH_3 levels at the two sites. This study
34 enhances the understanding of how surface emissions influence NH_3 concentrations at
35 high-altitude mountain site and highlights the critical role of NCP emissions in
36 influencing regional NH_3 levels, thereby providing insights for formulating strategies
37 to mitigate regional NH_3 pollution.

38 **Keywords:** ammonia, high-altitude mountain site, background station, Convergence
39 Cross Mapping (CCM), transport mechanism

41 1. Introduction

42 Ammonia (NH_3) mainly comes from fertilizers and animals, and other sources
43 include industry, fossil fuels, crops, soils, oceans, and biomass burning (Warner et al.,
44 2017; Warner et al., 2015). As the most important gas-phase alkaline species, NH_3
45 contributes considerably to the formation and development of fine particles ($\text{PM}_{2.5}$),
46 which have implications for human health, degrade regional air quality, and influence
47 the global radiation budgets. However, NH_3 emission control would mitigate nitrogen
48 deposition and haze pollution but worsen acid rain (Liu et al., 2019). The atmospheric
49 lifetime of NH_3 ranges from hours to days, depending on factors like deposition
50 processes and the presence of other reactive species (Baek et al., 2004); the global
51 average atmospheric lifetime is about 11 h (Xu et al., 2012).

52 Due to its short lifetime, NH_3 is highly concentrated in the planetary boundary
53 layer and near emission sources. Therefore, most of the current NH_3 measurements
54 were implemented at near-surface sites (Pu et al., 2023; Kuang et al., 2020; Zhang et
55 al., 2023; Elser et al., 2018). However, aircraft measurements revealed there were NH_3
56 pollution layers, where the concentrations were comparable to that of the surface,
57 within the lower free troposphere (FT) over the North China Plain (NCP) (Pu et al.,
58 2020a). Aircraft-borne campaign measurements over the US obtained concentration
59 profiles in the FT reaching altitudes of about 6 km (Nowak et al., 2007; Nowak et al.,
60 2010; Nowak et al., 2012; Leen et al., 2013; Schiferl et al., 2016). Although aircraft
61 could capture the vertical distribution of NH_3 in FT, only for the limited durations of



62 measurement campaigns. Satellite NH_3 observations complement current ground and
63 airborne measurements by providing unique insights on NH_3 distributions from
64 regional to global scales, such as the Tropospheric Emission Spectrometer (TES)
65 instrument on the NASA Aura satellite (Shephard et al., 2011), the Infrared
66 Atmospheric Sounding Interferometer (IASI) instrument on the MetOp-A and MetOp-
67 B satellites (Van Damme et al., 2014), and the Cross-track Infrared Sounder (CrIS)
68 instrument on the NASA satellite (Dammers et al., 2019). However, the vertical
69 sensitivity of these satellite retrievals is mainly limited to the lower troposphere up to
70 approximately 3 km, and no altitude resolution is achieved (Höpfner et al., 2016). These
71 findings underscore the significance of studying ammonia at high-altitude sites, where
72 its impact on large-scale pollution may be even more pronounced.

73 High-altitude mountain sites have long been recognized as suitable places for
74 characterizing the chemical composition of the lower troposphere. These sites allow for
75 measurements that are representative of continental to hemispheric scales by focusing
76 on air masses that have travelled far from emission sources and had sufficient time to
77 mix. Although NH_3 concentration observations have been conducted at mountain sites
78 in previous studies, the relevant data remain relatively limited. Among these,
79 continuous observation data with high-resolution is particularly scarce. Such
80 continuous measurements offer an opportunity to establish connections between NH_3
81 concentrations and the dynamic changes in source emissions as well as transport
82 patterns.

83 To improve our understanding of the characteristics and influence factors of NH_3
84 in the lower FT, a one-year measurement was conducted at a high-altitude mountain
85 site (Mountain Wutai, WTM) in northern China. Due to WTM is located in west of the
86 NCP, one of the NH_3 high-emission regions in the world (Van Damme et al., 2018), it
87 provides an opportunity to compare its patterns with those of other sites within the NCP.
88 Overall, the primary goals of this work were to characterize NH_3 levels at WTM and
89 other sites in NCP and to further interpret the similarities or different transport patterns
90 between these sites.



91

92 **2. Experimental and Methods**

93 **2.1. Experimental site and Instrument**

94 From June 2020 to May 2021, continuous in situ measurements were carried out
95 at a mountain site in Shanxi province, a background site and an urban site in Beijing.
96 The mountain site, Mountain Wutai (WTM; 38.95 °N, 113.52 °E; 2208 m a.s.l.) is
97 located in the northeast Shanxi province (Fig.1), which is connected to the continental
98 plateau extending to the west. Due to its high altitude and remote location, there are no
99 major pollution sources, making it an appropriate site for characterizing lower FT NH₃.
100 The fluctuations in the hourly and daily average for temperature (T), relative humidity
101 (RH), wind direction (WD), wind speed (WS), and precipitation, are depicted in Fig.S1.
102 The annual average T, RH, and total precipitation are 2.5 °C, 62.6 %, and 892.7 mm,
103 respectively. Prevailing winds are generally westerly, with an average speed of 5.8 m/s.

104 The background site, Shangdianzi (SDZ; 40.65°N, 117.11°E; 293.9 m a.s.l.), lies
105 in the transitional region between the NCP and the Mountain Yan area (Fig. 1). It acts
106 as both a regional background station in China and a regional site under the Global
107 Atmosphere Watch program. The mountainous areas around the SDZ station only
108 contains small villages, where populations are sparse and anthropogenic emission
109 sources are negligible. Therefore, the station's atmospheric pollution levels can
110 represent the background concentration of air pollutants in North China. Nevertheless,
111 orchards surrounding SDZ may have ammonium dibasic phosphate and urea applied as
112 soil fertilizers during spring.

113 The urban site is situated at the Beijing Meteorological Service (BMS; 39.93°N,
114 116.27°E; 90 m a.s.l.) (Fig. 1), which lies in the northwestern sector of the Beijing urban
115 area, surrounded by varied land use types (commercial areas, park, residential areas,
116 and traffic areas).

117 Real-time NH₃ concentrations at the above-mentioned stations were measured
118 using three analyzers (907, Los Gatos Research Inc., US). To ensure data accuracy and
119 reliability, rigorous quality assurance (QA) and quality control (QC) procedures,



including daily zero and span checks, monthly multi-point calibrations using certified
 NH₃ standard gas, were implemented throughout the campaign. Other than NH₃
 observations, methane (CH₄) was also measured at SDZ by a Picarro G2401
 CO₂/CO/CH₄ cavity-ringdown spectrometer during summer 2020 and spring 2021.
 Meteorological data of the surface in this study were achieved from the China
 Meteorological Data Service Center (<http://data.cma.cn/en>). The planetary boundary
 layer height (BLH) and U, V, and W winds are achieved from the ERA5
 (<https://cds.climate.copernicus.eu/datasets>), a fifth-generation European Centre for
 Medium-Range Weather Forecasts (ECMWF) reanalysis data with a spatial resolution
 of 0.25°×0.25°.

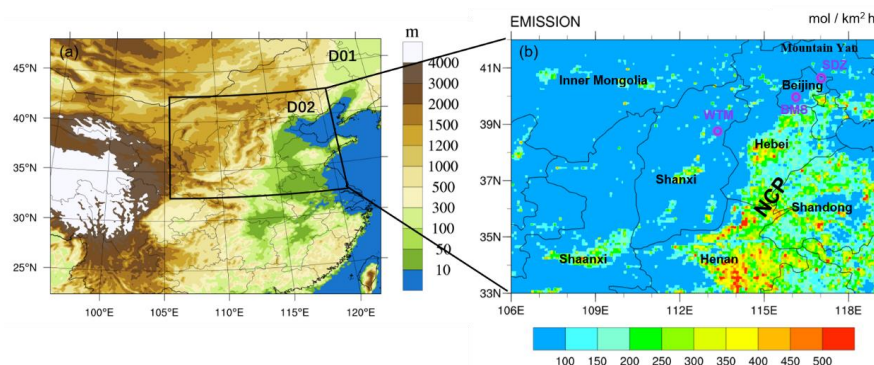


Fig. 1. Simulation domains and topographic distribution (a); spatial distribution of NH₃ emission intensity and locations of the measurement sites (indicated by purple open circles) (b).

2.2. Convergence cross mapping (CCM) method

Given the complex interactions in atmospheric environments and the strong
 nonlinearity of atmospheric systems, simple correlation analyses (e.g., Pearson
 correlation, which is only applicable to linear systems) cannot effectively quantify the
 causal effects of individual meteorological factors on NH₃ concentrations. To address
 this limitation, Sugihara et al. (2012) developed Convergent Cross Mapping (CCM) —
 a robust causal analysis approach—designed to extract the coupling relationships
 between individual variables in complex systems. This method serves as a suitable
 alternative for identifying nonlinear associations within the same system and evaluating
 weak-to-moderate coupling effects, while also enabling the determination of interaction



143 directionality. Therefore, it has been widely adopted in related studies.

144 In CCM, the predictive skill of variable A for variable B is calculated and denoted
 145 as ρ (ranging from 0 to 1). This metric provides a quantitative basis for comparing the
 146 influence magnitudes of different variables on a target variable. Detailed descriptions
 147 of the CCM algorithm have been published in previous literature (Chen et al., 2022;
 148 Ziyue et al., 2018; Rawat et al., 2024). The CCM analysis in this study was conducted
 149 using R software (version 4.3.1), with the support of the rEDM package (Sugihara et
 150 al., 2012) and the multispatialCCM package (Clark et al., 2015).

151 **2.3. Potential source contribution function (PSCF)**

152 To identify the locations of sources influencing different pollutant concentrations,
 153 PSCF analysis was applied, with its calculation relying on measured concentrations and
 154 associated trajectory data. A high PSCF value denotes that the corresponding region
 155 transports substantial amounts of the target atmospheric pollutant to the receptor site.
 156 In contrast, a low PSCF value may reflect either minimal emissions from the region or
 157 the absence of pollutant transport pathways from that region to the receptor site.

158 From June 2020 to May 2021, 48-hour backward trajectories were calculated 24
 159 times per day (00:00 to 23:00 UTC, with initiation at 10 m above ground surface) using
 160 the HYSPLIT model developed by the National Oceanic and Atmospheric
 161 Administration Air Resource Lab (NOAA;
 162 <http://www.arl.noaa.gov/ready/hysplit4.html>). Meteorological input data were derived
 163 from the FNL global analysis data—generated by the National Center for
 164 Environmental Prediction (NCEP) model and processed by the Global Data
 165 Assimilation System (GDAS) with a spatial resolution of $1^\circ \times 1^\circ$
 166 (<http://www.rl.noaa.gov/ss/transport/archives.hrml>). Detailed methodologies for the
 167 PSCF approach and weighted potential source contribution function (WPSCF) analysis
 168 have been reported in previous studies (Wang, 2014; Pu et al., 2019). The 70th percentile
 169 of NH_3 concentrations over the entire observation period was used as the criterion value.

170 **2.4. WRF-Chem**

171 The WRF-Chem model (version 4.2.1) was employed to simulate NH_3



172 concentrations and meteorological conditions in this study. A detailed description of
 173 WRF-Chem is provided by Grell et al. (2005). The model uses the CBM-Z gas phase
 174 chemical mechanism (Zaveri and Peters, 1999) to represent the transformation
 175 reactions of NH_3 in the atmosphere.

176 In this study, two nested domains with horizontal resolutions of 9 km and 3 km
 177 were established within the WRF-Chem model, centered at 116°E , 39°N (Fig. 1). The
 178 inner domain (D02, shown in Fig. 1a) covers the primary agricultural region of the
 179 North China Plain, which includes three major crop-producing provinces: Henan, Hebei,
 180 and Shandong. In the vertical direction, the model was configured with 30 sigma-
 181 pressure layers extending from the surface up to 50 hPa, 12 of which are located within
 182 the atmospheric boundary layer. Meteorological initial and boundary conditions were
 183 derived from the NCEP Final Operational Global Analysis (FNL) dataset, which has a
 184 spatial resolution of $1.0^\circ \times 1.0^\circ$ and a temporal resolution of 6 hours. Anthropogenic
 185 emissions were based on the inventory developed by Zhang et al. (2009), which was
 186 subsequently updated to a resolution of $0.1^\circ \times 0.1^\circ$ for the year 2019 (MEIC-2019,
 187 Zhang, personal communication). Biomass burning emissions were obtained from the
 188 Fire Inventory from NCAR (FINNv1.5, Wiedinmyer et al., 2011). Chemical initial and
 189 boundary conditions were initialized using model-default idealized profiles. The
 190 simulation was conducted from May 1 to 12, 2021, with the first 11 days considered as
 191 model spin-up and results from May 12, 2021, used for subsequent analysis.

192

193 **3. Results and Discussion**

194 **3.1. Overall features of NH_3 concentration**

195 The continuous variability of NH_3 concentrations at WTM during 2020 and 2021
 196 is shown in Fig. 2. The hourly NH_3 concentration ranged from 1 to 52.7 ppb, with a
 197 year-round average of 9.0 ± 6.7 ppb. The NH_3 concentrations in this study exhibited a
 198 seasonal variation (Table S1), with means of 8.9 ± 6.3 , 15.9 ± 5.4 , 8.8 ± 4.2 , and $2.1 \pm$
 199 1.2 ppb in spring, summer, autumn, and winter, respectively. Compared with other high-
 200 altitude sites (Table S2), the average concentration of NH_3 at WTM is obviously higher



201 than most of the sites, particularly at Rocky Mountain National Park, Beaver Meadows,
 202 Timber Creek, and Gore Pass in the USA and the Happono site in Japan. In contrast, NH_3
 203 levels at WTM were substantially lower than those recorded at rural and agricultural
 204 sites, such as Wuwei in China and Greeley and Kersey in the United States, highlighting
 205 the influence of anthropogenic emissions. Notably, within the same type of observation
 206 site, forest or grassland, the NH_3 concentrations measured at WTM are elevated. These
 207 discrepancies imply that local sources, atmospheric transport processes, or
 208 topographical factors may contribute to enhanced ammonia accumulation at this site
 209 compared to similar ecological settings.

210 Situated in a remote, high-altitude area in the western of the NCP, this unique
 211 location endows comparative studies between WTM and other sites located in the NCP.
 212 As can be seen from Figure 2(a), the NH_3 concentration and its variation pattern at the
 213 WTM site exhibit a high degree of similarity to those at the SDZ site, while showing
 214 significant differences from the BMS site, where the NH_3 concentration levels are
 215 notably higher. To further quantify the similarity of NH_3 variation tendency among the
 216 sites, this study employed a variety of analytical methods, including Dynamic Time
 217 Warping (DTW), Euclidean distance, Pearson correlation coefficient (r), Root Mean
 218 Square Deviation (RMSD), and Mean Absolute Bias (MAB) (Table S3). The DTW was
 219 processed in R version 4.3.1, along with the dtw R package (Giorgino, 2009). It is
 220 noteworthy that despite the greater spatial straight-line distance (350 km) between the
 221 SDZ and WTM sites compared to the BMS and WTM distance, as well as an altitude
 222 difference of 1.9 km, the values of DTW, Euclidean distance, RMSD, and MAB
 223 between WTM and SDZ are all lower than those between WTM and BMS. Meanwhile,
 224 the r between WTM and SDZ is significantly higher than that between WTM and other
 225 sites. These comprehensive quantitative analysis results fully demonstrate a high degree
 226 of consistency in the variation trends and numerical distributions of the NH_3
 227 concentration time series at the WTM and SDZ sites.

228 Since WTM is in a remote area, emission sources in its vicinity are relatively
 229 scarce. In contrast, the BMS site is located in an urban area, with obvious emission



sources such as vehicle exhaust emissions (Pu et al., 2023) and residential emissions. These complex local emission sources significantly influence the NH_3 concentration level and its variation pattern at this site, leading to distinct differences from the WTM. Interestingly, despite considerable altitude and spatial separation between WTM and SDZ, they exhibited similar NH_3 variation characteristics and close concentration. The subsequent sections will conduct analyses from meteorological effects, potential source regions, and transport patterns to illustrate the primary factors driving this similarity.

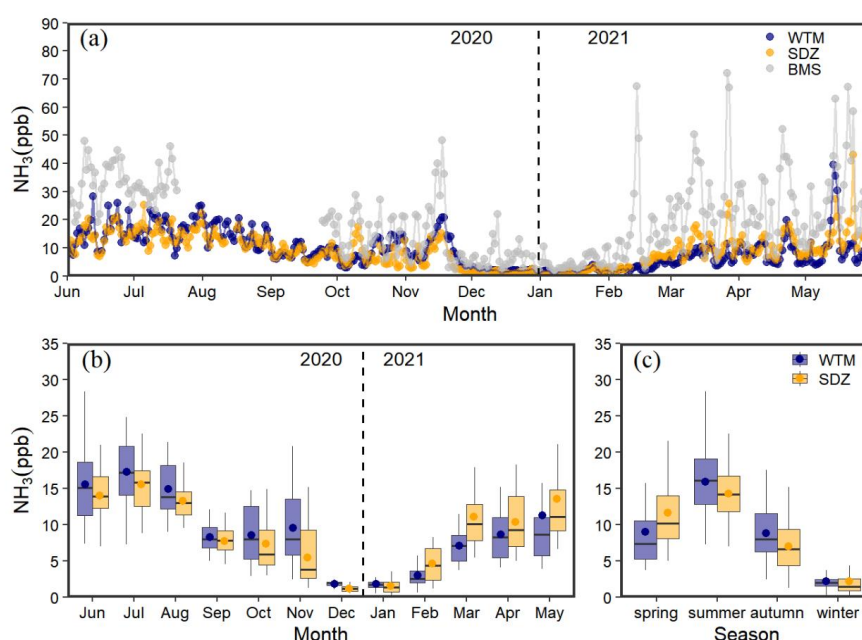


Fig. 2. Daily (a), monthly (b) and seasonal (c) variation of NH_3 concentration from Jun 1, 2020 to May 31, 2021. The solid dots represent the mean values, the horizontal lines in the box denote the median, the limits of the boxes correspond to the 25th and 75th percentiles, and the whiskers of the boxes are the 10th and 90th percentiles (b, c).

3.2. Effects of meteorological factors

Local meteorological conditions exert a significant impact on the NH_3 levels (Zhang et al., 2018). Given the complex interactions within atmospheric environments, it remains challenging to quantify how individual meteorological factors affect local NH_3 concentrations. In order to solve this problem, CCM was employed to examine the coupling relationships between these factors and NH_3 . The CCM analysis based on



248 hourly data is shown in Table 1, and the influence of local meteorological factors —
249 including WS, WD, T, RH, pressure (P), and BLH — on NH₃ in different seasons is
250 determined. The convergent cross maps were used to explain the quantitative coupling
251 between NH₃ concentration and each individual meteorological factor (Fig. S2). The ρ
252 -value from CCM methods is designed to understand the coupling between two
253 variables by excluding influences from other factors. Nevertheless, it lacks the ability
254 to directly identify the positive or negative nature of the causality between the two
255 variables.

256 CCM analysis reveals that RH and T are found to show strong influence on WTM
257 NH₃ during summer, while RH and T are also found to have strong effect on SDZ NH₃
258 during spring. However, high NH₃ levels are generally associated with high temperature
259 and humidity (Lan et al., 2021). P shows a strong impact on NH₃ concentrations of
260 WTM and SDZ NH₃ in autumn and spring, respectively. That could be attributed to P
261 mainly affecting the transport and accumulation of pollutants by indirectly influencing
262 other meteorological factors (e.g., wind and humidity) (Chen et al., 2020). Although
263 various meteorological parameters affect NH₃ concentrations differently across seasons
264 at different sites, WS, WD, and BLH significantly impact NH₃ concentrations at both
265 WTM and SDZ year-round (Table 1). This indicates that wind-driven processes (e.g.,
266 atmospheric transport or dispersion) along with the boundary layer variation play
267 crucial roles in governing NH₃ levels at these sites throughout the year.

268 A bivariate polar plot is presented in Fig. 3 to analyze the function of WS and WD
269 in each season. Despite the prevailing wind being westerly (Fig. S1), the higher NH₃
270 concentrations at WTM were observed in the southeast and southwest sectors in spring
271 and summer, while elevated levels mainly occurred in the easterly direction during
272 autumn and winter. In spring and summer, the higher concentrations were accompanied
273 by a wide range of WS between 0 and 15 m/s, suggesting that local emissions and air
274 masses transported from NH₃ intensity emission regions (Fig.1b) were important
275 factors affecting NH₃ concentrations at WTM during warm seasons. However, in
276 autumn and winter, higher NH₃ concentrations were detected with a WD of 5 – 10 m/s,

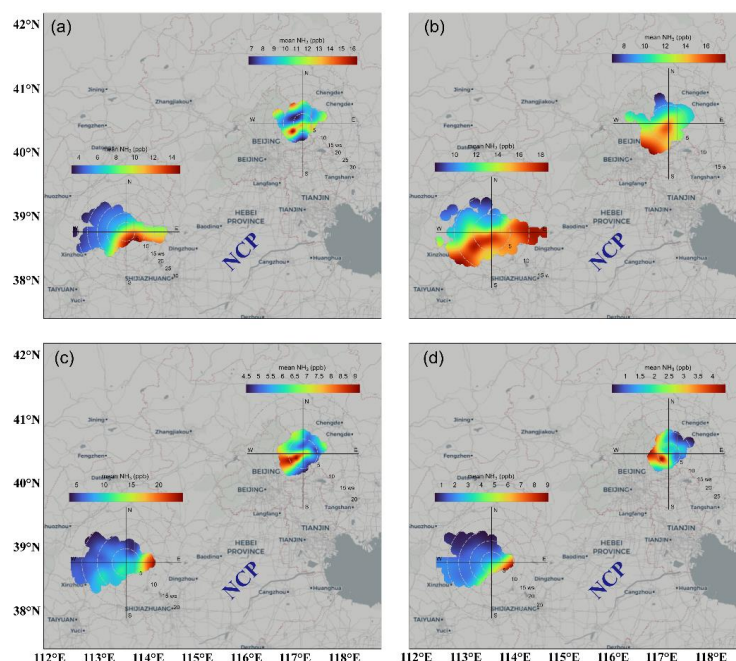


277 indicating that WTM was mainly under the influence of long-distance transport during
278 cold seasons. Unlike WTM, higher SDZ NH₃ concentrations were mainly concentrated
279 in the southwest direction. Based on NH₃ concentration variations and WS in each
280 season at SDZ, the site was affected by both local emissions and air mass transport.
281 Nevertheless, the impact of transport on NH₃ was more significant, as elevated
282 concentrations corresponded to WS exceeding 2 m/s. Notably, the NCP is located to the
283 southeast of WTM and to the southwest of SDZ – directions that directly correspond to
284 the WD associated with higher levels at both sites. This spatial alignment strongly
285 suggests that NCP might act as a potential emission hotspot for both WTM and SDZ.

286 Table 1 Seasonal correlations and causal relationships between individual meteorological
287 parameters and NH₃ concentration.

Site	Spring	Summer	Autumn	Winter
WTM	WS (0.42, 0.00**)	WS (0.15, 0.00**)	WS (0.46, 0.00**)	WS (0.46, 0.00**)
	WD (0.38, 0.01**)	WD (0.14, 0.00**)	WD (0.33, 0.00**)	WD (0.39, 0.00**)
	BLH (0.33, 0.00**)	T (0.22, 0.00**)	P (0.35, 0.00**)	RH (0.33, 0.00**)
		RH (0.63, 0.03*)	BLH (0.25, 0.01*)	BLH (0.20, 0.01*)
		BLH (0.23, 0.04*)		
SDZ	WS (0.31, 0.00**)	WS (0.24, 0.00**)	WS (0.36, 0.00**)	WS (0.16, 0.00**)
	WD (0.12, 0.00**)	WD (0.13, 0.02*)	WD (0.35, 0.00**)	WD (0.26, 0.00**)
	T (0.26, 0.01*)	T (0.24, 0.00**)	BLH (0.1, 0.01*)	RH (0.34, 0.00**)
	RH (0.62, 0.02*)	BLH (0.19, 0.02*)		BLH (0.21, 0.00**)
	P (0.28, 0.03*)			
	BLH (0.19, 0.00**)			

288 The causality of different meteorological factors on NH₃ is presented predictive skill (ρ) from the
289 CCM method with p-values (p). * and ** denote the p less than 0.01 and 0.05, respectively.



290
 291 Fig. 3. Bivariate polar plots of NH_3 concentrations at WTM and SDZ in four seasons (a: spring, b:
 292 summer, c: autumn, and d: winter).

293 3.3. Potential sources identified

294 To confirm whether NCP was a potential emission area for WTM and SDZ, PSCF
 295 analysis was performed. The PSCF map distributions of WTM and SDZ during the
 296 study period and each season are examined (Fig. 4 and Fig. S3). Grid cells characterized
 297 by high PSCF values (i.e., > 0.7) were identified as regions with a high likelihood of
 298 being NH_3 source areas for the receptor sites. Fig. 3 indicates that the potential sources
 299 for WTM and SDZ were primarily in the broader NCP, particularly in southern Hebei,
 300 western Shandong, and eastern Henan. Importantly, these locations corresponded with
 301 the NH_3 emission distribution in the NCP, as shown in Fig. 1b. This region is
 302 characterized by elevated anthropogenic emissions of NH_3 , largely from agricultural
 303 activities (Li et al., 2021). In addition to NCP, SDZ was also affected by emissions from
 304 the Bohai Sea. This might be attributed to the ocean-going vessels that installed the
 305 marine selective catalytic reduction technology (SCR) system to reduce NO_x emissions.
 306 As catalyst activity decreases over time, NH_3 slip increases (Zhang et al., 2021). In



spring and summer, the potential source ranges of both sites were more scattered in NCP; however, during autumn and winter, these ranges were more concentrated in the south or east of Hebei (Fig. S3). Under the impact of southerly wind, emissions originating from NCP can be transported to WTM and SDZ.

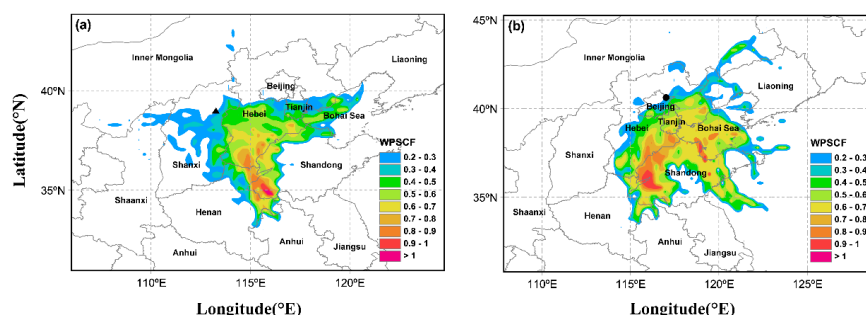
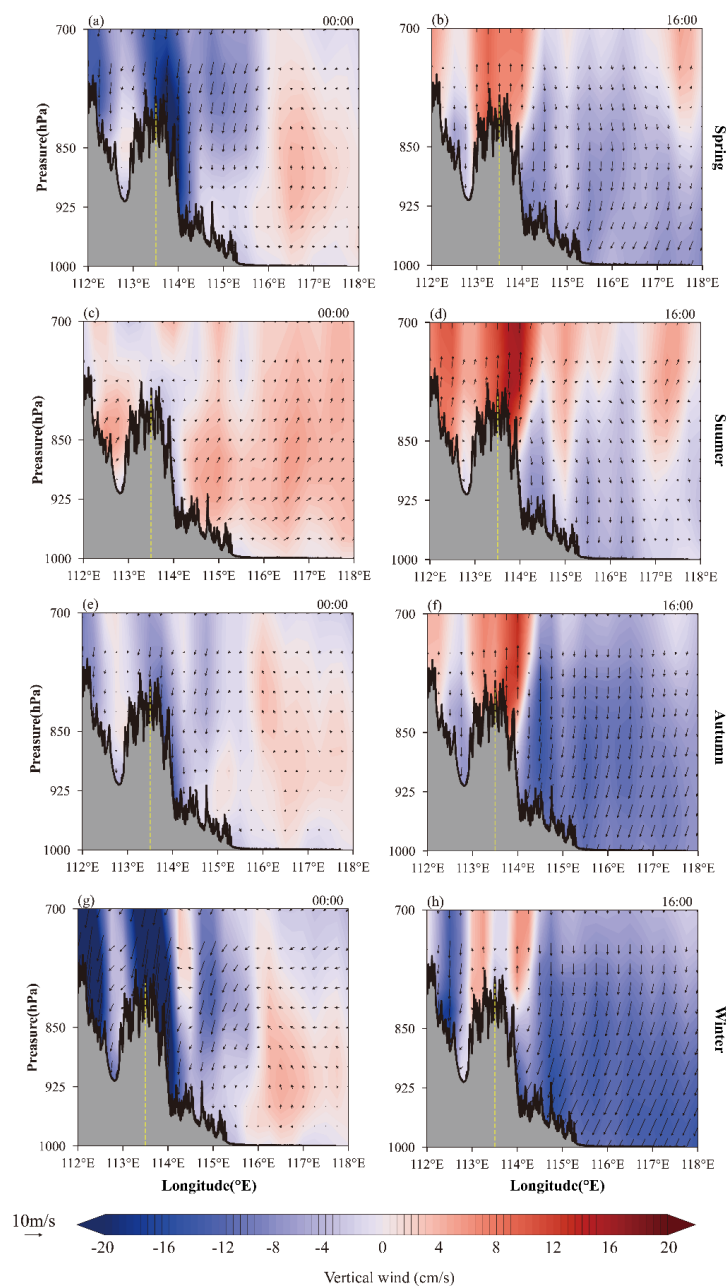


Fig. 4. Weighted potential sources contribution analysis (PSCF) of NH_3 at WTM (a) and SDZ (b) (triangle and circle denote WTM and SDZ, respectively).

3.4. Transport patterns of NH_3

3.4.1. Diurnal variation of the atmospheric circulation

Atmospheric circulation determines the migration and transport patterns of pollutants, so to study its impact on the distribution of NH_3 concentration, the east-west cross-sections of seasonal mean wind vectors and vertical velocity were first analyzed. As shown in Fig. 5, during the daytime, low-level winds close to mountain slopes warm early, resulting in upward movement from the plain toward the mountains. At night, this pattern switches to downslope motions due to the pressure difference between mountains and plains. Therefore, although WTM at a high altitude, local sources from NCP can still have an influence, depending largely on the vertical transport and associated convective mixing. Owing to ground surface heating, the magnitude of vertical velocity varies seasonally, peaking in summer (~ 16 cm/s) and reaching its lowest in winter (~ -20 cm/s). This mountain-plain circulation directly affects the day and night difference in NH_3 concentrations at WTM throughout the year, following those variations.



330
 331 Fig. 5. Meridional cross-section of vertical velocity (contour region) with superimposed wind
 332 vectors. The yellow dotted lines represent the location of WTM. The four rows are the different
 333 seasons, including spring (a-b), summer (c-d), autumn (e-f), and winter (g-h).

334



3.4.2. Relationship between the diurnal variations of NH_3 and BLH

As mentioned in section 3.3, NCP, especially in the south of Hebei, the west of Shandong, and the east of Henan provinces, represents the shared common source area influencing WTM and SDZ. To figure out how the emissions emitted from this region affected two sites, diurnal variation of NH_3 and boundary layer height were analyzed (Fig. 6). In order to better illustrate the diurnal variation of NH_3 at these two sites more clearly, the normalized concentrations were applied. As shown in Fig. 6, NH_3 concentrations at SDZ didn't rise as the BLH elevated, except in winter. This might be due to the good dispersion and dilution conditions as the BLH increases that reduce the NH_3 levels. Since SDZ lies in the transitional zone between the NCP and the Mountain Yan area, it is influenced by mountain-valley circulation (Lin et al., 2008). In the afternoon, under the impact of valley wind, lots of the pollutants from NCP were transported to SDZ by the southerly wind, leading to a peak concentration of NH_3 around 18:00 in seasons. However, apart from the late afternoon peak, morning spikes of NH_3 can also be found during summer and autumn. Based on previous studies, the morning NH_3 enhancement can be linked to dew evaporation since dew is a nighttime reservoir and morning source for NH_3 (Wentworth et al., 2016; Kuang et al., 2020).

However, different from the diurnal behavior of SDZ, NH_3 levels at WTM began to rise steadily around 8:00. During spring and autumn, peaks occurred around 18:00, while in winter and summer, peak durations were longer (12:00 – 18:00), followed by a rapid decline. It is particularly noteworthy that the diurnal variation of NH_3 concentration at WTM shows good consistency with the diurnal variation of BLH compared to SDZ, showing an increase in concentration as BLH rises and a decrease as BLH falls. This consistency might suggest that the mountain wind system transports the pollutants to the mountaintop during the day via upslope winds, along with the development of BLH through convective mixing. Conversely, nighttime downslope winds lead to the dispersion of these pollutants.

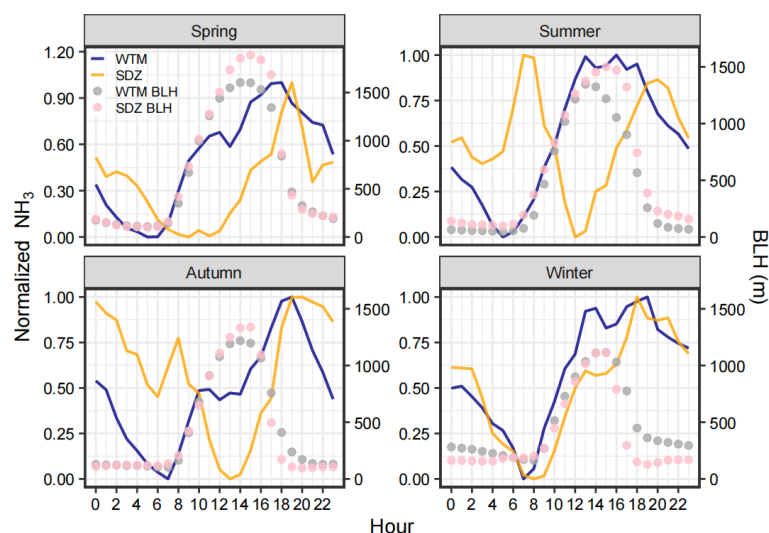


Fig. 6. Diurnal variation of normalized NH_3 concentration and BLH at WTM (blue line and grey dots) and SDZ (orange line and pink dots), respectively.

However, despite the fact that the distance between WTM and SDZ is more than 350 km and they are controlled by different transport patterns (i.e., mountain-plain and mountain-valley circulations in WTM and SDZ, respectively), their NH_3 levels still exhibit a high similarity. To find out the underlying factor driving this convergence, CH_4 data at SDZ was used to explain.

Fig. 7a presents the variations in the NH_3 concentration difference between SDZ and WTM, along with the average CH_4 concentration across different periods. During Stage I (summer to early autumn), the fluctuation of the NH_3 concentration difference is relatively small, ranging approximately from -5 to 5 ppb. In contrast, during Stage II (spring), the fluctuation amplitude of the difference increases significantly, and the values are mostly positive except for the period from May 12 to May 15, 2021 (Fig. 7b), indicating that the NH_3 concentration at SDZ is higher than that at WTM during this period. Corresponding to these patterns, the CH_4 concentration at SDZ is higher in stage I and lower in stage II.

As CH_4 is a key indicator of agricultural emissions, and the CH_4 concentration at SDZ can represent the background level of agricultural emissions in North China, the relationship between the NH_3 concentration difference and CH_4 levels in stages I and II



can be interpreted as follows: during stage I, when regional agricultural emissions are strong, the NH_3 concentrations at WTM and SDZ are more closely aligned, reflecting the dominant role of agricultural sources in homogenizing the concentrations at the two sites. Conversely, during stage II, when regional agricultural emissions are weak, the NH_3 concentration at SDZ is significantly affected by emissions from urban Beijing due to the transport of southerly winds. This leads to a larger concentration difference between the two sites, while the lower CH_4 concentration at SDZ further confirms the reduced influence of agricultural sources during this period.

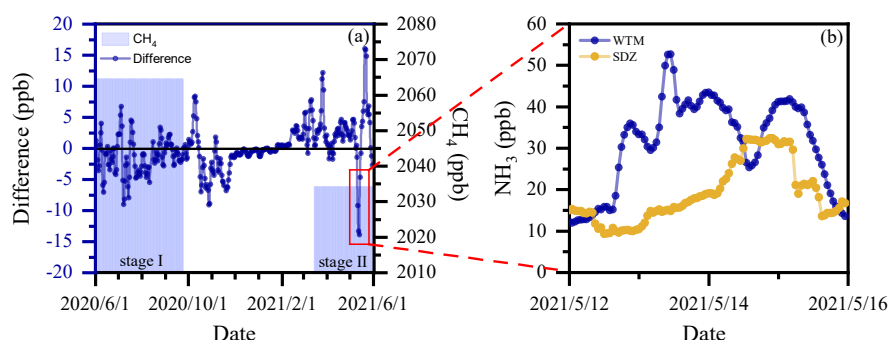


Fig. 7. Time series of average CH_4 concentration at SDZ and daily concentration difference between SDZ and WTM (a) and hourly concentration at each site (b).

3.4.3. A typical case simulation

To find out why NH_3 at WTM exhibited a higher level than that at SDZ in stage II, a typical case of May 12, 2021, was simulated to more clearly illustrate how NH_3 concentration varies with the evolution of the wind field and BLH at WTM. Fig. S4 shows the simulated and observed hourly mean NH_3 concentration, WD, and WS at the WTM and SDZ on 12 May, 2021. Despite the model has evident deviations in simulating the concentration values, it accurately captures the diurnal variations of NH_3 levels and surface wind fields at both stations. Consequently, the simulation results are deemed reliable for further analysis of transport characteristics.

As shown in Fig. S4, on May 12, 2021, the prevailing wind direction in the NCP region was southeast, with a strong east wind system traversing the mountainous areas west of the NCP. NH_3 concentration exhibited a transport pattern from the eastern plain

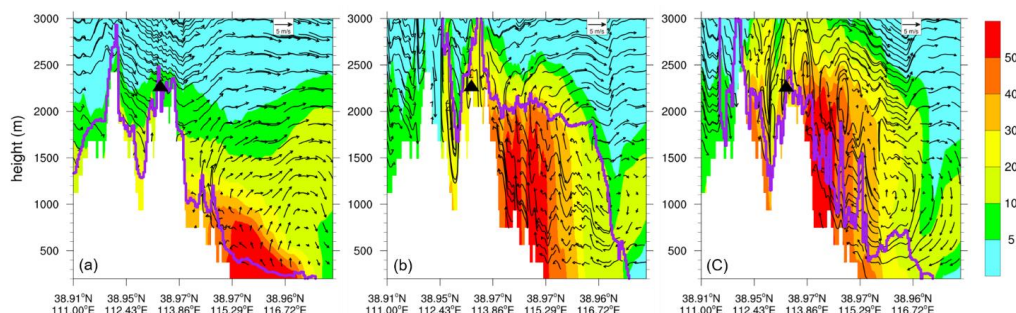


405 to the western mountains, significantly impacting the WTM. During this period, the air
406 mass affecting the SDZ station mainly originated from the Bohai Sea area, with no
407 significant pollutant transport from the NCP, thus maintaining consistently low NH_3
408 levels (Fig. S4a).

409 In addition to the transport effect of the southeasterly wind in the horizontal
410 direction, the lifting of the boundary layer height also plays a crucial role in the increase
411 in NH_3 concentration at the WTM station. To more finely illustrate the process of NH_3
412 concentration varying with the evolution of wind fields and boundary layer height, an
413 east-to-west cross section (shown in Fig. S4b) has been selected for detailed analysis.
414 As shown in Fig. 8, although similarly affected by the easterly wind, the NH_3 emitted
415 from the plain area on the morning of May 12 was trapped in the plain area due to the
416 lower BLH, resulting in low concentrations at the WTM (Fig. 8a). In the afternoon, air
417 masses with higher NH_3 levels from the eastern plain region rise upward with the
418 increase of BLH and are simultaneously pushed up along the slope by the prevailing
419 easterly winds within the boundary layer, gradually affecting the WTM area (Fig. 8b).
420 At the same time, both observations and simulations at WTM showed a rapid increase
421 in ground-level NH_3 concentrations (Fig. S4a). After nightfall, this high concentration
422 situation persisted at WTM in the absence of strong weather systems (Fig. 8c). Our
423 simulation result validates the aforementioned analysis that anthropogenic emissions
424 from NCP, influenced by the regional transport and boundary layer lifting, would lead
425 to an increase in NH_3 levels in the relatively distant mountain areas with high altitude.

426 It is noteworthy that the modeling study of the typical case on 12 May 2021 was
427 designed to corroborate the analysis based on observational data and to qualitatively
428 elucidate the horizontal and vertical transport pathways of NH_3 , as well as their
429 relationship with boundary layer dynamics. However, the simulation results have
430 limitations in accurately capturing NH_3 concentrations within a regional area and in the
431 quantitative assessment of transport impacts. These aspects will be the focus of further
432 investigation in future work.

433



440

441 Fig. 8. West-east vertical distributions of NH_3 mixing ratios (ppb) and vertical wind velocity (cm/s)
 442 at (a) 08:00 LST, (b) 16:00 LST, and (c) 19:00 LST on 12 May 2021. Note that vertical velocities
 443 are scaled by a factor of 100 in the wind vectors. The cross-section location is indicated by the solid
 444 black line in Fig. S4b. Wind vectors are overlaid on the NH_3 distributions. The thick purple curves
 445 denote the top of the atmospheric boundary layer. The approximate location of the WTM site is
 446 marked with a black triangle.

447

448 4. Conclusion

449 Continuous high-resolution and real-time NH_3 observations made from Jun 1,
 450 2020 to May 31, 2021 were simultaneously conducted at WTM, SDZ, and BMS. The
 451 average concentration of NH_3 at WTM was 9.0 ± 6.7 ppb during the study period.
 452 Compared to the same type of observation site in previous studies (e.g., forest or
 453 grassland mountain sites), the NH_3 concentration measured at WTM was much higher
 454 than those reported by Pan et al. (2018), Ban et al. (2016), and Benedict et al. (2013).
 455 On the other hand, although WTM is located in a remote area, its NH_3 levels were very
 456 close to those at the background station (SDZ) in the northern edge of NCP, while being
 457 much lower than those at the megacity station (BMS) in NCP. To identify the primary
 458 factors driving the similarity in NH_3 levels between WTM and SDZ, meteorological
 459 effects, potential source regions, and transport patterns were analyzed.

460 CCM analysis between NH_3 and meteorological variables revealed that WD, WS,
 461 and BLH significantly regulate NH_3 levels at both WTM and SDZ across all seasons.
 462 The relationships of wind and NH_3 concentration as well as PSCF results suggested that
 463 these sites were influenced by a shared common source area in the NCP, especially in
 464 the south of Hebei, the west of Shandong, and the east of Henan provinces.

465 The influence of pollutants emitted from NCP on WTM and SDZ was different.



466 NH₃ concentrations at SDZ were mainly controlled by the mountain-valley circulation,
467 which could transport the NH₃ from NCP to it by valley wind. For WTM, under the
468 effect of mountain-plain circulation and the diurnal variation of BLH, the pollutant from
469 NCP was carried up to the top of the mountain by the vertical transport and convective
470 mixing. This transport pattern was further confirmed by WRF-Chem simulation.

471 Furthermore, the correlation between CH₄ concentrations and the NH₃ level
472 difference between SDZ and WTM indicated that regional agricultural emissions in
473 NCP were the dominant factor contributing to the NH₃ similarity between these two
474 sites – even though the two sites differed significantly in altitude and geography.

475 Our study provides a full annual cycle of measurements at a location that is
476 underrepresented in the global ammonia monitoring network, and highlights that
477 emissions from the NCP can influence NH₃ concentrations at both high-altitude and
478 background areas in northern China by different transport mechanisms. These results
479 could help policymakers develop effective strategies for mitigating regional air
480 pollution. However, our results cannot distinguish specific NH₃ sources and their
481 quantitative contributions. Therefore, additional $\delta^{15}\text{N}$ isotope measurements are needed
482 in future studies.

483

484 **Data availability.**

485 The data are available at <https://zenodo.org/records/17089450> (Pu et al., 2025)

486

487 **Competing interests.**

488 The authors declare that none of the authors has any competing interests.

489

490 **Author contributions.**

491 WP analyzed the data and prepared the manuscript. JX revised the manuscript. LZ
492 and JD conducted the measurements. ZM supervised the project. All co-authors
493 discussed the results and commented on the manuscript.

494

495



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