

On behalf of my co-authors, I greatly appreciate the reviewer's constructive comments and suggestions on our manuscript entitled "Measurement report: Assessing ammonia characteristics over a high-altitude mountain site in Shanxi Province, China: a comparison with observations in the North China Plain". We have carefully studied the comments and made corresponding revisions that we hope will meet with your approval. The revisions are marked in the manuscript, and our responses to the comments are as follows.

### **Reviewer 1**

Pu et al. reports valuable year-long measurements of NH<sub>3</sub> simultaneously captured at 3 distinct site types at different locations in the North China Plain, respectively representing high altitude background in the western North China Plain, regional background of the northern part of the Plain and urban pollution within the Plain area. This data was used to reveal similarities and differences within NH<sub>3</sub> measurements at distinct locations as well as to uncover the influences of emissions, local circulation and transport. Overall, the manuscript presented valuable insights into the homogeneity of NH<sub>3</sub> pollution in the North China Plain, and the impact of geographical circulations. However, the manuscript still needs to be further improved before it can be accepted for publication. Below are some detailed comments:

1. P1 L22-23: Grammar issue: "It further conducted comparative analyses"

The grammatical errors were corrected in line 22-24 of the new version.

P1 L30: Basically mountain-plain and mountain-valley circulations are caused by similar physical processes; they only differ in spatial scale. Is it accurate to say that the SDZ station is only influenced by mountain-valley circulation? Although the altitude of SDZ is not that high, it is still located within the Yan mountains, therefore must be also subjected to mountain-plain circulations.

Thank you for your suggestion. Mountain-plain and mountain-valley circulations are caused by similar physical processes, both driven by thermal contrasts over different terrains. Under certain conditions, the two circulation systems can coexist and interact. Their most significant distinction lies in the underlying topographic context and spatial scale. Mountain-plain circulation, as a mesoscale system, requires a substantial elevation difference (hundreds to thousands of meters) to establish pronounced thermal contrast and pressure gradients, typically occurring between a sizable mountain range and an adjacent broad plain. In contrast, mountain-valley circulation operates at a smaller, local scale, sustained mainly by thermal differences between slopes and valley bottoms, which usually arise from elevation differences of tens to hundreds of meters (Whiteman, C. D. ,2000; Weissmann, M., et al., 2005).

The SDZ is situated on the slopes of the Mountain Yan foothills and is thus influenced by both mountain-plain and mountain-valley circulations. However, considering its elevation (293.9 m a.s.l.), especially in comparison with the elevation of WTM station (2208 m a.s.l.), therefore, the revision stating that "mountain-valley circulation is the primary circulation system influencing NH<sub>3</sub> transport from emission source regions in the North China Plain to the SDZ station" is more accurate. Revisions have therefore been made in the abstract and relevant sections accordingly.

#### References:

Whiteman, C. D. (2000). *Mountain meteorology: fundamentals and applications*. Oxford University Press.

Weissmann, M., Braun, F. J., Gantner, L., et al. (2005). The Alpine Mountain-Plain Circulation: Airborne Doppler Lidar Measurements and Numerical Simulations. *Monthly Weather Review*, 133(11), 3095-3109. DOI: 10.1175/MWR3012.1

2. P3 L73-82: What scientific findings did previous measurements at high altitude sites present? How did these high-altitude measurements compare to those in nearby urban sites? What was still unexplained by these studies and needs to be further studied in this one?

Pan et al. (2018) and Xu et al. (2019) conducted measurements across China, including at urban and mountainous sites, using passive sampling methods with low temporal resolution. The results indicated that the observed NH<sub>3</sub> concentrations in urban areas were 2–3 times higher than those in mountainous sites. Despite providing insights into spatial distribution and seasonal variations at the national scale, these studies were limited by low temporal resolution, which impeded the investigation of NH<sub>3</sub> variability across multiple temporal scales, as well as the impacts of various meteorological variables and potential mountain-induced circulations on NH<sub>3</sub> levels. Our study utilized high-resolution, real-time NH<sub>3</sub> monitoring equipment and analytical techniques to elucidate the transport patterns governing NH<sub>3</sub> concentrations at the high-altitude mountain site.

#### Reference:

Pan, Y., Tian, S., Zhao, Y., Zhang, L., Zhu, X., Gao, J., Huang, W., Zhou, Y., Song, Y., Zhang, Q., and Wang, Y.: Identifying Ammonia Hotspots in China Using a National Observation Network, *Environmental Science & Technology*, 52, 3926-3934, 10.1021/acs.est.7b05235, 2018.

Xu, W., Zhang, L., and Liu, X.: A database of atmospheric nitrogen concentration and deposition from the nationwide monitoring network in China, *Scientific Data*, 6, 51,

10.1038/s41597-019-0061-2, 2019.

3. P4 L115: Should be "by various land use types" instead of "varied".

The sentence has been revised in accordance with your suggestion in line 114-115.

5. P8 L210-227: Regional air pollution occurs typically throughout the entire North China Plain under favorable meteorological conditions. Typically, there is an approximate cycle of 7 days in synoptic pattern variations, which might be mainly responsible for the observed "similarities" between observations at the two background sites.

I agree. The synoptic pattern is one of the main factors that influence observed "similarities" between two background stations. It can be found that two background stations exhibit almost same variation trends and fluctuation characteristics, while the BMS station shows higher concentration of  $\text{NH}_3$  in fig.2a. Therefore, we focus on the details of the similarities and provide some statistical parameters to assess the similarities among three stations (table s3). The quantitative results indicate that although synoptic patterns play crucial roles in the variations of  $\text{NH}_3$  in the NCP,  $\text{NH}_3$  showed different variation characteristics, especially at urban station, which are likely to be caused by emissions and transport conditions as discussed below.

6. P8 L228-229: What are the possible influences of microbial soil activities on  $\text{NH}_3$  at WTM and SDZ?

Microbial activities could affect nitrogen cycles in soil, which induce  $\text{NH}_3$  emission to the atmosphere by ammonification. However, the extent of ammonification depends dramatically on the amount of fertilizer. Both WTM and SDZ are located at background areas and are rarely affected by agricultural activities. Thus, microbial soil activities could account for a comparatively low percentage of  $\text{NH}_3$  concentration at WTM and SDZ.

7. What kind of data were used in the CCM analysis? Daily averages? How are intercorrelations among meteorological factors considered within this methodology (e.g.: intercorrelation between RH, T and P)

As described in Section 3.2, CCM calculations were based on hourly data. The correlations among meteorological factors varied across distinct seasons. For example, RH exerts a strong impact on P during summer, whereas it has a negligible effect in autumn. Therefore, the relationships among these factors are dependent on seasonal conditions. In this study, we mainly focus on the effect of meteorological parameters on  $\text{NH}_3$ .

8. L274: "NH<sub>3</sub> intensity emission regions" should be revised as "intense NH<sub>3</sub> emission regions"

Thanks. The sentence has been revised in the new version in line 274.

9. P11, L285: NCP is a region and cannot act like a potential emission "hotspot"

The sentence has been revised as "This spatial alignment strongly suggests that the NCP might act as a potential emission area for both WTM and SDZ" in line 284-285.

10. P15, L342: Might this be because cold and shallow wintertime BLH might trap pollutants over the plain region, preventing it from reaching SDZ? While during the other seasons the BLH was high enough for pollutants to mix up to the height of SDZ, under such context higher BLH would only result in more dilution and lower pollution levels.

According to the previous reports, the height of the fully developed planetary boundary layer over the NCP in winter typically ranges from several hundred meters (> 500 m) to around two kilometers (Su et al., 2018). The altitude of the SDZ site is 293 m, which is significantly lower than the typical daytime BLH over the adjacent plain areas in winter. Thus, the BLH is sufficiently high to enable the long-range transport of pollutants to the SDZ.

In winter, NH<sub>3</sub> concentrations remain at a low level of approximately 2.2 ppb under the influence of frequent northerly winds. Nevertheless, NH<sub>3</sub> emissions still occur from soil, sewage, and waste sources as temperatures rise after sunrise, even though winter temperatures are considerably lower than those in other seasons. Therefore, the diurnal variation in NH<sub>3</sub> exhibits the same characteristics as the BLH.

Reference:

Su, T., Li, Z., and Kahn, R.: Relationships between the planetary boundary layer height and surface pollutants derived from lidar observations over China: regional pattern and influencing factors, *Atmos. Chem. Phys.*, 18, 15921–15935, <https://doi.org/10.5194/acp-18-15921-2018>, 2018.

11. P15 L353: There seems to be two sources influencing NH<sub>3</sub> at the WTM site, one that is closer to the site, that influences the WTM earlier during the day and another later peak seems to be similar to that of SDZ, possibly more linked to mountain-plain circulation induced transport from the plain region. How do wind speed and wind directions vary diurnally at WTM and SDZ during distinct seasons?

In the manuscript, fig.6 illustrates the diurnal variation in normalized NH<sub>3</sub> concentrations, which would amplify the difference between the two peaks. However,

the actual difference between these two peaks is less than 0.3 ppb.

The diurnal variations in wind speed and direction are presented below (fig. R1). The WTM site is dominated by westerly winds year-round. As shown in Fig. 6, both the earlier and later peaks occur approximately between 10:00 and 18:00. During this period, in contrast to the SDZ site, the average wind direction at WTM exhibits no obvious shift. Therefore, it is difficult to conclude that a local source exists near WTM that contributes to the formation of the earlier peak. Further measurements will be conducted in future work to investigate the presence of local sources.

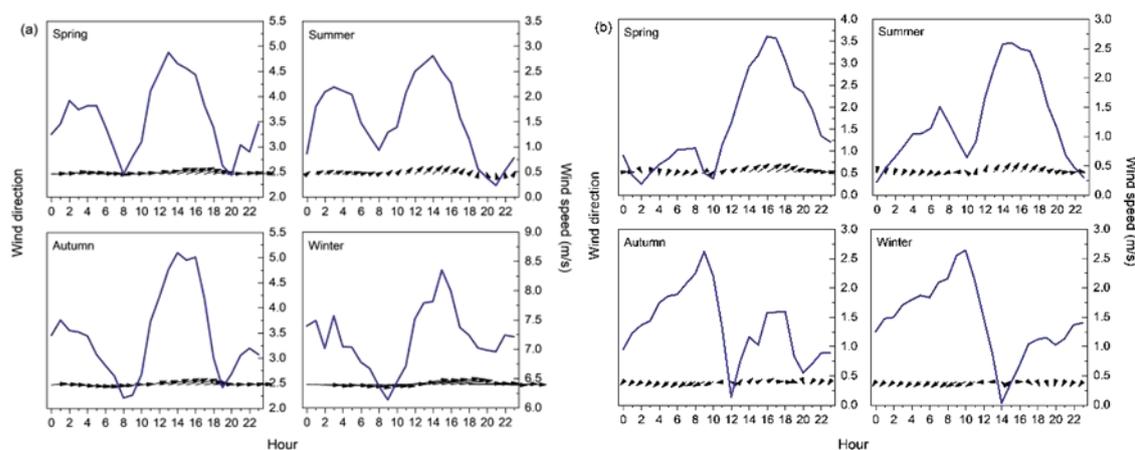


Fig. R1 Diurnal variation of wind direction (arrows) and wind speed (blue lines) at WTM (a) and SDZ (b)

12. P17 L 398-399 grammar issue.

The grammatical errors were corrected in line 399-401 of the new version.

13. While the case studies help understanding the role of topography and transport, the authors should fully exploit their long-term data and answer some critical questions such as: how does the regional background concentration vary with season, how does urban emissions add to the NH<sub>3</sub> burden over the NCP, how much influence do agricultural emissions contribute during distinct seasons? How frequent are morning peaks and how often are they linked to dew emissions?

Thank you for your suggestions. Our study aims to characterize the WTM site using measured data and elucidate why concentration variations at WTM and SDZ are highly similar. Case simulations are only used to verify the pollutant transport pattern.

Analyzing seasonal variations in background concentrations, quantifying urban and agricultural emission contributions to both sites, and measuring dew emissions are worthwhile, but these are not the main aims of our study. Further simulations and measurements will be conducted in future work to address these aspects.