Response to comments on "Atmospheric NH₃ in urban Beijing: long-term variations and implications for secondary inorganic aerosol control"

Ziru Lan¹, Xiaoyi Zhang², Weili Lin¹, Xiaobin Xu², Zhiqiang Ma³, Jun Jin¹, Lingyan Wu², Yangmei Zhang²

¹Key Laboratory of Ecology and Environment in Minority Areas, Minzu University of China, National Ethnic Affairs Commission, Beijing 100081, China

²Institute of Atmospheric Composition, Chinese Academy of Meteorological Science, Beijing 100081, China

³Institute of Urban Meteorology, China Meteorological Administration, Beijing 100089, China *Correspondence to*: Weili Lin(<u>linwl@muc.edu.cn</u>)

General comments:

This paper describes the implications of standard/state-of-the-art NH₃ measurements taken in Beijing between 2009 and 2020. NH₃ is an air pollutant that is principally emitted from human activities, affecting ecosystems, air quality and climate. The manuscript is a good fit for the ACP audience. I found the methods used to be generally sound relative to the conclusions drawn, and the paper is well written and clear. I would recommend this article for publication after the following comments are addressed (some major, some minor).

----- We greatly appreciate your time spent reviewing our manuscript and providing constructive comments. We have revised the manuscript according to your suggestions.

Specific comments:

Major:

I feel that there should be better acknowledgement of the uncertainty inherent in relocating the measurement in 2017. While there have been references showing that the NH3 vertical profile is relatively constant up to 300 m in Beijing, a measurement on the 14 story could be on the cusp of surpassing 300 m. There has also been recorded greater variability in the vertical profile in other locations. The manuscript raises that there is spatial variability in NH3, so it's not clear why this couldn't also extend to measurements of the vertical profile (in other words, could the vertical profile of NH3 differ in

different parts of Beijing?). In my opinion, it would be better to acknowledge the uncertainty associated with this move, raise plausible hypotheses, and indicate that there is too much uncertainty to draw confident conclusions about the drop in 2017. Could emissions or changes in the urban topography have contributed? These aren't discussed here but might also be relevant. The authors may also consider splitting the dataset in 2017, but it is probably worth maintaining the dataset as continuous because the horizontal distance between the two locations is so small.

----- Thank you for your suggestion. We acknowledge the necessity of addressing the uncertainties associated with our observations and have made the following modifications accordingly: ①We have added detailed descriptions regarding the heights of the observation locations: The ground-floor elevations of both buildings are 56 m, and the observation heights above the ground are 10 m on the 3rd floor and 56 m on the 14th floor. Studies of vertical observations of atmospheric NH₃ in Beijing have been conducted at the Beijing Meteorological Tower, which is situated in an urban area. Both locations are approximately 6.7 km apart and have the same elevation of 56 m. Given that the 14th floor of our observation site is 56 m above the ground, which is considerably lower than 300 m, we believe that these vertical profile results are applicable as a reference for our study. ②We have added a discussion on splitting the dataset into three segments, comparing the segmented data results with those from the continuous dataset. Overall, the revised Section 3.1 now reads as follows:

From June 2009 to July 2020, the hourly average mixing ratio of atmospheric NH₃ in Beijing was 26.9 \pm 19.3 ppb (median, 23.5 ppb). Table S1 summarizes results from various NH₃ monitoring studies conducted in urban areas. The results of the present study are basically consistent with the annual NH₃ mixing ratio averages that were observed in urban Beijing by other researchers through optical instrument. The primary reasons for this phenomenon are the frequent agricultural activities and the presence of highly alkaline soils in the North China Plain, where Beijing is located. As a densely populated country with intensive agriculture activities, China contains several areas that are major global hotspots for the atmospheric NH₃ concentration. The monitoring results of the present study indicate that the overall NH₃ mixing ratio in Beijing is lower than that in Delhi but considerably higher than those in other developed cities such as New York, Toronto, and Rome. Even within China, the NH₃ mixing ratio in Beijing is higher relative to that in Shanghai, which is also a megacity (i.e., the NH₃ mixing ratio in Shanghai is less than one-third of that in Beijing), and only a few cities in North China have mixing ratios

comparable to that in Beijing. The primary reasons for this phenomenon are the frequent agricultural activities and the presence of highly alkaline soils in the North China Plain, where Beijing is located.

Due to significant data gaps from January 2013 to June 2013 and from May 2017 to August 2017, the period from 2009 to 2020 was divided into three segments for linear regression analysis (Figure S7). From June 2009 to January 2013, the observed hourly average atmospheric NH₃ mixing ratio showed a decreasing trend (R = -0.23, p < 0.05, slope = -0.01); from June 2013 to May 2017, the NH₃ mixing ratio increased (R = 0.04, p < 0.05, slope $= 0.22 \times 10$ -2); and from September 2017 to July 2020, the NH₃ concentration exhibited a decreasing trend again (R = -0.03, p < 0.05, slope $= -1.42 \times 10^{-3}$). Similar to in situ observations, the satellite observations of NH₃ concentration showed a decreasing trend from June 2009 to January 2013 (R = -0.19, p < 0.05, slope $= -3.88 \times 10^{-4}$) and an increasing trend from June 2013 to May 2017 (R = 0.12, p < 0.05, slope $= 3.65 \times 10^{-4}$), but differed from in situ atmospheric NH₃ trends as it continued to rise from September 2017 to July 2020 (R = 0.23, p < 0.05, slope $= 1.17 \times 10^{-3}$).



Figure S7. Trends in atmospheric NH₃ concentrations observed in situ and by satellite from June 2009 to January 2013, from June 2013 to May 2017 and from September 2017 to July 2020.

To further analyze the long-term trends of the atmospheric NH₃ concentration, the present study referred to the findings of Vu et al. (2019) and used meteorological factors to construct a random forest model for imputing missing values. The computed time series for the atmospheric NH₃ concentration is presented in Figure S8. The complete dataset obtained through EEMD was used to characterize the changes in atmospheric NH₃ concentrations in Beijing (Figure 1). The NH₃ mixing ratio initially exhibited a slight decrease but started to increase in 2012 and peaked in 2017, subsequently declining. From 2009 to 2017, the NH₃ mixing ratio increased by 50%, but by 2020, the NH₃ mixing ratio had decreased by 49% from its peak in 2017. A comparison of monthly average NH₃ concentrations obtained from satellite observations revealed that prior to 2018, the trend for the surface NH₃ mixing ratio was similar to that observed by satellites, exhibiting a decline followed by an increase in atmospheric NH₃ concentrations in 2018, these two trends diverged, with satellite observations indicating a continued increase in NH₃ concentrations, while the surface NH₃ mixing ratio exhibited a decreasing trend.

The monitoring results of this study were compared with NH₃ monthly concentrations observed by NNDMN in Beijing from April 2011 to December 201 and from January 2005 to August 2020 (Figure S9). From April 2011 to December 2015, both the NH_3 mixing ratio observed in this study (R = 0.27, p < 0.05) and the satellite-observed concentrations (R = 0.28, p < 0.05) exhibited increasing trends, while the NNDMN station did not show a significant trend (R = 0.16, p > 0.05). The NNDMN station observations from January 2009 to August 2020 were significantly correlated with this study's observations ($R_{Aug} = 0.66$, p < 0.05; $R_{Jan} = 0.65$, p < 0.05), but neither the present study's observations nor the NNDMN observations were significantly correlated with satellite-observed NH₃ concentrations. Satellite observations showed a strong correlation between NH₃ concentrations in the Beijing urban area and the Beijing-Tianjin-Hebei region (Figure S3). However, measurements by Zhang et al. (2020) at five stations in Beijing indicated that four stations had lower NH₃ concentrations in 2017 (winter) than in 2020 (winter + spring), while one station had higher concentrations in 2017 than in 2020, indicating variability in observation results even within the same city. Due to the short atmospheric lifetime, low transport altitude, high dry deposition rate, limited transport distance, and abundance of atmospheric NH₃, its complex temporal and spatial characteristics contribute to the complexity of NH₃ variations. Satellite observations are limited by the observation height and spatial resolution, which may mask

variations in local surface NH₃ concentrations. Additionally, differences between the present study's observations and satellite observations may also be due to changes in the monitoring location and observation height in September 2017. However, tower observations conducted by the Institute of Atmospheric Physics, Chinese Academy of Sciences (6.7 km from the present study's site) in the urban area showed only slight variations within a 300 m altitude range. Therefore, the change in observation altitude may have had a limited impact on the change in NH₃ mixing ratio trends after 2017.



Figure S9. (a) Atmospheric NH₃ monthly average concentrations observed in this study, NNDMN Beijing station, and by satellite in the Beijing urban area and their trends from April 2011 to December 2015. (b) Correlations between NH₃ concentrations observed in this study, NNDMN, and by satellite from January 2009 to January 2020. (c) Correlations between NH₃ concentrations observed in this study, NNDMN, and by satellite from January 2009 to August 2020.

I found it strange that there is an anticorrelation between NH3 and temperature (on an average daily basis) and feel that the authors should include more interpretation of this finding. Are there any other papers showing an NH3-T anticorrelation?

----- Several studies have examined the diurnal variations of NH₃, showing similar patterns during spring, summer, and autumn as observed in our study, but without discussing their relationship with temperature (Buijsman et al., 1998; Sharma et al., 2014; Gu et al., 2022). In our previous research, the urban site exhibited a negative correlation between NH₃ concentrations and temperature's diurnal characteristics during summer, autumn, and winter, whereas the rural site showed a positive correlation (Lan et al., 2021). Buijsman et al. (1998) suggested that lower daytime NH₃ concentrations in high emission areas might be due to higher wind speeds and more favorable mixing conditions, with NH₃ accumulating during the night within a shallower boundary layer. Our current manuscript delves deeper into these issues, with the revised paragraph as follows:

Several studies have suggested that temperature plays a pivotal role in driving diurnal variations in atmospheric NH₃ concentrations (Clarisse et al., 2021; Langford et al., 1992). However, the present study shows that NH₃ concentrations are significantly influenced by temperature across seasonal changes (Figure 3); in terms of diurnal patterns, the days showing positive and negative correlations between NH_3 concentrations and temperature each constitute nearly half of the effective observation days (Figure S11). The mean diurnal variations in different seasons typically exhibit lower concentrations during the day and higher at night in spring, summer, and autumn (Figure S12). This difference suggests that correlations observed on a seasonal (climatic scale) tend to obscure lower-frequency data relationships, with daily variations in NH₃ concentrations being more influenced by the transition from day to night (meteorological or weather scale). This highlights the complex factors affecting NH₃ concentrations in the urban areas of Beijing and underscores the importance of high temporal resolution in observations. Several studies have noted a reduction in NH₃ concentrations during daylight hours (Buijsman et al., 1998; Sharma et al., 2014; Gu et al., 2022; Lan et al., 2021). Increased temperatures during the day promote the volatilization of NH₃, but lower daytime concentrations may result from higher wind speeds and more favorable mixing conditions, whereas at night, NH₃ tends to accumulate within a shallower boundary layer (Buijsman et al., 1998). The diurnal variation of the boundary layer height in Beijing exhibits a single-peak pattern, rising rapidly from 6:00 to 8:00, reaching its peak between 14:00 and 15:00, then declining sharply, and stabilizing after 18:00 to 20:00 (Figure S12). During the daytime, NH₃ concentrations are influenced by a combination of temperature (which promotes emissions) and changes in the boundary layer height (which causes dilution), with the valley value of NH₃ concentrations lagging behind the peak times of boundary layer height and temperature. Moreover, during spring, summer, and autumn, the continuous decline in NH₃ concentrations during the daytime indicates that vertical mixing transport has a limited impact on atmospheric NH₃ in the urban areas of Beijing. In winter, the evaporation of dew or frost in the early morning leads to a rapid increase in H₂O and NH₃ concentrations (Wentworth et al., 2016), while the effects of afternoon temperature and vertical mixing dilution are comparable, keeping NH₃ concentrations relatively stable.



Figure S12. Average diurnal variations in NH₃, temperature, absolute humidity and boundary layer height in different seasons in Beijing urban area. Boundary layer height data are from the ERA5 global atmospheric reanalysis (Hersbach et al., 2023)

Figure 3 - To my eye there generally appears to be a positive correlation between air T and NH3. The text starting on L241 also suggests there is a pretty even balance of days with positive or negative correlation between NH3 and air T (my understanding is that this is correlating hours within a day). It seems to me that there could plausibly be a weak negative correlation on a daily average basis, but I don't understand how there are strong negative correlations between air T and NH3 for most seasons in L246. Is this correct? Otherwise, could the interpretation among these relationships between air T and NH3 be expanded to clarify?

----- It is widely accepted that NH₃ concentration positively correlates with temperature, and our study's

findings on the seasonal variations of NH₃ concentrations also support this consensus. During the observation period, the temperature in Beijing followed the seasonal sequence of summer (being the warmest), spring, autumn, and winter. The rankings of NH₃ concentrations across the seasons were consistent with this trend. In Figure 3, we calculated average values of NH₃ concentrations and temperatures in each season annually. We found a significant correlation only in winter (R = 0.38, p < 0.05), with no significant correlations in other seasons. Line 241 discusses the results from Figure S11, where we analyzed the hourly averages of NH₃ concentration and their correlation with temperature on diurnal variations. Line 246 presents the correlation of average daily variations between NH₃ concentrations and temperature over an eleven-year observation period (Figure S12). The strong negative correlation reported on L246 might seem counterintuitive. However, such diurnal variation patterns are more prevalent during the seasons with higher concentrations (summer, spring, and autumn), and tend to obscure individual daily patterns after averaging. This seemingly contradictory result reveals the complexity and difference between climatic scale and weather scale factors. We have added the following content in the original text for further explanation: However, the present study shows that NH₃ concentrations are significantly influenced by temperature across seasonal changes (Figure 3); in terms of diurnal patterns, the days showing positive and negative correlations between NH₃ concentrations and temperature each constitute nearly half of the effective observation days (Figure S11). The mean diurnal variations in different seasons typically exhibit lower concentrations during the day and higher at night in spring, summer, and autumn (Figure S12). This difference suggests that correlations observed on a seasonal (climatic scale) tend to obscure lower-frequency data relationships, with daily variations in NH₃ concentrations being more influenced by the transition from day to night (meteorological or weather scale). This highlights the complex factors affecting NH₃ concentrations in the urban areas of Beijing and underscores the importance of high temporal resolution in observations.

Minor:

Generally, I found the abstract to be a well written and succinct summary that draws out keen points of interest for this manuscript. My only suggestion is to strengthen the motivation for controlling NH3 concentrations in the last sentence. The preceding sentence expresses that SIA concentrations are not very sensitive to NH3, so it could be helpful to provide additional rationale (for example, that reducing

NH3 is cheaper? It's more effective once NH3 has been reduced to some extent?—both discussed in the paper).

----- Thank you for your suggestion. We have revised the last sentence of the abstract as follows: "Although reducing NH₃ concentrations can improve air quality during winter, controlling acid gas concentrations has a greater effect than controlling NH₃ concentrations on reducing SIA concentrations, until NH₃ and acidic gas concentrations are reduced below 80% of their current levels. Nevertheless, the increase in the proportion of ammonium salts in SIAs during the observation period indicates that future control measures for NH₃ concentrations may need to be prioritized in Beijing."

L38 – suggest clarifying that "particulate matter 2.5" is "particulate matter with a diameter less than 2.5 μ m in size".

----- Done.

Sentence starting on L36 ("However, long-term...") – I think that this paragraph motivates the utility of long-term trends in ground-based atmospheric NH3 well, but it would benefit from expanding on the urban aspect of this study. For example, are NNMDN sites generally in rural areas?

----- We have adjusted this section to make it more relevant to the content of the study: In China, according to the monitoring results from the Nationwide Nitrogen Deposition Monitoring Network (NNDMN), NH₃ concentrations at 12 urban sites and 43 rural sites increased by approximately 80% from 2011 to 201. Satellite data analysis by Dong et al. (2023) indicated a significant increase (~32%) in NH₃ vertical column densities in China from 2008 to 2019. In the North China Plain, a hotspot for global NH₃ emissions, Luo et al. (2020) found a rapid increase in urban NH₃ concentration from 2011 to 2018. Wen et al. (2024) found a 26% decrease in Beijing NH₃ concentrations from August 2005 to August 2020, and a 50% increase from January 2005 to January 2020. Currently, long-term ground-based observations of atmospheric NH₃ at high temporal resolution are relatively rare in China, and the contrasting trends between NH₃ emissions, satellite and in-situ measured concentrations in urban areas have not been fully explore.

L78 – Describe source of emission data in Figure S1 either in the main text or figure caption.

----- We have added a description of the sources of the emission data: Figure S1. NH₃ emission in and around Beijing (a) and topographic map of Beijing (b). The NH₃ emissions data represent the total for 2017, sourced from emission inventories (Huang et al., 2012; Kang et al., 2016).

L79 – I assume that the inlet is outside of the building—could you clarify?

----- The inlet is located outside the building. In our setup, the inlet lines are positioned approximately 1.5 m above the floor level of the building. It is installed through a hole in the window using a polytetrafluoroethylene (PTFE) tube that connects to the instrument, with the hole fitting snugly around the sampling tube. The conduit extends outward by about 15 cm, and to prevent water ingress, there is a downward-sloping shield above the conduit. To clarify further, I have added details about the sampling setup in the second paragraph of Section 2.1: At CMA site, the air had been drained into an air-conditioned room with a 4.5 m long Teflon line and the inlet height is 1.8 m above the rooftop (about 12 m above ground level). At MUC site, air is introduced from outside the sealed window through a borehole, with the air inlet extending 20-30 cm outside the window. Since it is on the 14th floor, the air outside the building flows smoothly.

L80 – Later in the manuscript it is noted that there is not a strong vertical profile of NH3 in Beijing (although this has been observed in other locations). It may be worth mentioning that here as well.

----- Thank you for your suggestion. Presenting the vertical profile characteristics of NH_3 in Beijing here would be beneficial for understanding the reliability of our data. However, to maintain the focus and flow of this section, we have opted to discuss this in the data analysis section later in the manuscript.

L100 – Do you have any thoughts on why the slope is higher in this study than in Zhang et al. 2021?
----- The discrepancies in the observation results can be attributed to differences in the timing of

instrument comparisons between Zhang et al. and our study, as well as variations in instrument conditions over time. Furthermore, the longer duration of observations in our study exposes our instruments to a broader range of environmental conditions, which may also contribute to the observed differences in slopes.

L112 - Could you please clarify why you switched the source of the met data?

----- Due to the unavailability of meteorological data from the Haidian weather station, which is closest to our observation site before 2012, we utilized meteorological data from the Capital Airport for continuity in data analysis. By comparing the correlation of meteorological data (Figure S5), we believe that it is feasible to use temperature and relative humidity data from the Capital Airport to supplement the analysis in this study.

L122 – Could you please clarify where is the Chinese Academy of Meteorological Sciences relative to the other measurement locations of interest?

----- We apologize for the ambiguity in our expression that may have caused a misunderstanding. The original sentence: "PM_{2.5} samples collected on filters were analyzed for ion components (Na⁺, SO₄²⁻, NH₄⁺, NO₃⁻, Cl⁻, Ca²⁺, K⁺, and Mg²⁺) at the Chinese Academy of Meteorological Sciences, resulting in the acquisition of 184 data sets." Here, the Chinese Academy of Meteorological Sciences was the location where the samples were analyzed, not where the monitoring occurred. We have revised this sentence to: "The collected PM_{2.5} samples on filters were subsequently sent to the Chinese Academy of Meteorological Sciences for chemical analysis of ion components (Na⁺, SO₄²⁻, NH₄⁺, NO₃⁻, Cl⁻, Ca²⁺, K⁺, and Mg²⁺), from which 184 data sets were obtained."

Section 2.2.1 – It would be helpful to briefly state why this particular method is being used (to gap fill?).

----- Thank you for your comment. The use of EEMD is not intended for filling data gaps, but rather for a clearer analysis of long-term trends. Traditional trend analysis tools have limitations, especially when dealing with weak and non-linear trends. For instance, the linear regression method does not represent the true trends of non-linear data accurately. The combined use of the Mann-Kendall test and Theil-Sen (TS) methods for long-term trend analysis is affected by seasonal fluctuations in the data series. In contrast, Ensemble Empirical Mode Decomposition (EEMD) is an advanced time-frequency analysis technique that is well-suited for extracting trends from time series that are non-linear and exhibit irregular cycles. The temporal characteristics of atmospheric NH3 are non-linear and non-stationary, hence using EEMD allows for a more comprehensive understanding of underlying trends and cycles often masked in traditional analysis methods. We have included additional descriptions of EEMD in our manuscript: Compared to traditional long-term trend analysis tools, EEMD shows greater stability in decomposing non-linear and non-stationary data series. It is unaffected by the seasonal variations in data series and can resolve non-linear changing trends, making it more suitable for environmental data trend analysis. Therefore, using EEMD enables more accurate extraction of genuine signal variations.

Section 2.2.2 – Could you please describe the model configuration? For example, are the PM5 ion measurements used to run the model? It might be helpful to make that connection and remind the reader of that available data.

----- We reorganized this section: The ISORROPIA-II model is mainly used to simulate the physical state and concentration of inorganic components of the aerosol system at thermodynamic equilibrium. A distinct advantage of the ISORROPIA-II model over other thermodynamic models is the inclusion of the K^+ , Ca^{2+} , and Mg^{2+} ions in the calculations, and taking these components into account significantly improves the accuracy of the model simulations (Allen et al., 2015). Additionally, the high precision and computational efficiency of the ISORROPIA II mode have been widely demonstrated (Fountoukis and Nenes, 2007). To assess the sensitivity of sulfate, nitrate, and ammonium (SNA) to changes in precursor concentrations, the present study employed the ISORROPIA II thermodynamic equilibrium, version 2.3 (http://isorropia.epfl.ch). The model was run in "forward + metastable" mode, taking inputs such as temperature (unit is k), relative humidity (up to 1), and concentrations of particulate components (SO4²⁻, $Cl^- + HCl_NO3^- + HNO3_NH4^+ + NH3_NNa^+_NK^+_NCa^{2+}$ and Mg^{2+}) expressed in $\mu g m^{-3}$ for calculations.

Table S1 – It's not totally clear what's gained from comparing this study's results to other studies located

in different parts of the world. I am also unclear on what the numbers in the "This study" column represent – are those the averages from the same timeframe as the study that they are being compared with? Please expand/clarify.

----- Thank you for your comment. This column was intended to compare NH₃ concentrations during the same period with other studies. However, we agree that this column does not significantly contribute to the analysis, so we have decided to remove it.

L227 – I don't understand how the variation is being calculated. It is described as "average annual temperature" but then specified by season. Please clarify?

----- Thank you for your comment highlighting the ambiguity in our expression. In our manuscript, the term "average annual temperature" refers to the calculation of average temperatures for each season within a year. We calculate the average temperature for each season and then discuss the variation coefficients of these seasonal averages for each year. Accordingly, we have revised the original text from "The interannual trends pertaining to temperature and NH₃ mixing ratios across multiple seasons (Figure 3) revealed that temperature remained stable in summer and autumn over the years; when calculated in Kelvin, the average annual temperature exhibited variation coefficients of 0.42% in spring, 0.15% in summer, 0.17% in autumn, and 0.51% in winter." to "The interannual trends for temperature and NH₃ mixing ratios across multiple seasons (Figure 3) revealed that temperature and 0.51% in winter." to "The interannual trends for temperature and NH₃ mixing ratios across multiple seasons (Figure 3) revealed that temperature and NH₃ in winter." to "The interannual trends for temperature and NH₃ mixing ratios across multiple seasons (Figure 3) revealed that temperature exhibited variation coefficients of 0.42% in spring, 0.15% in summer, 0.17% in autumn, and 0.51% in winter." to "The interannual trends for temperature and NH₃ mixing ratios across multiple seasons (Figure 3) revealed that temperature remained stable in summer and autumn over the years; when calculated in Kelvin, the average seasonal temperatures exhibited interannual variation coefficients of 0.42% in spring, 0.15% in summer, 0.17% in autumn, and 0.51% in winter."

Generally, I felt that the authors could be clearer about the specificity of their results for Beijing (as appropriate), in particular in the conclusions and when discussing the policy prioritization in the abstract (e.g. "measures to control NH3 concentrations should be prioritized [in Beijing].").

----- Thank you for your suggestion; we have made the necessary modifications.

L367 – reiterate that this is for Beijing——"Therefore, reducing acidic gas emissions is still a primary focus for controlling fine particulate matter pollution [in Beijing].

----Done.

L376 – Could you recall the evidence supporting "And in the future, more attention will be needed to focus on controlling NH3"?

----- Since this sentence is speculative and seems inappropriate for the conclusion section, we have decided to remove it.

Technical comments

There is often a missing space between a word and the opening parenthesis for the citations

----- We were really sorry for our careless mistakes. Thank you for your reminder.

Figure S3 caption – It's a little confusing that the subpart labels first follow and then precede the description of the content. It would be more intuitive to keep it consistent (e.g. "Monthly (a) and annual (b) variations and correlations between satellite observations... 116.5E) (c) and the average observations... \sim 118.5E) (d)."

----- Thank you for your suggestions. We have revised the text as follows: Monthly (a) and annual (b) variations and correlations between satellite observations during the observation period at the grid points around the monitoring stations (39.5°N, 116.5°E and 40.5°N, 116.5°E) (c) and the average observations in the region selected for the present study (36.5°N~42.5°N, 113.5°E~118.5°E) (d).

Consider referencing Figure 1 earlier, where the results are first mentioned (I believe in L172?)

----- Done.

L374 I found this wording awkward: "the current reduction of SIA remains less significant in response to NH3 than acid gases." Consider: "SIA formation is more sensitive to acid gases than NH3.

----- This has been done as suggested.

References

Hersbach, H., Bell, B., Berrisford, P., Biavati, G., Horányi, A., Muñoz Sabater, J., Nicolas, J., Peubey, C., Radu, R., Rozum, I., Schepers, D., Simmons, A., Soci, C., Dee, D., Thépaut, J-N.: ERA5 hourly data on single levels from 1940 to present. Copernicus Climate Change Service (C3S) Climate Data Store (CDS), https://doi.org/10.24381/cds.adbb2d47, 2023.

Buijsman, E., Aben, J. M. M., Van Elzakker, B. G., and Mennen, M. G.: An automatic atmospheric ammonia network in the Netherlands set-up and results, Atmos. Environ., 32, 317–324, https://doi.org/10.1016/S1352-2310(97)00233-1, 1998.

Gu, M., Pan, Y., Walters, W. W., Sun, Q., Song, L., Wang, Y., Xue, Y., and Fang, Y.: Vehicular Emissions Enhanced Ammonia Concentrations in Winter Mornings: Insights from Diurnal Nitrogen Isotopic Signatures, Environ. Sci. Technol., 56, 1578–1585, https://doi.org/10.1021/acs.est.1c05884, 2022.

Lan, Z., Lin, W., Pu, W., and Ma, Z.: Measurement report: Exploring NH3 behavior in urban and suburban Beijing: comparison and implications, Atmospheric Chem. Phys., 21, 4561–4573, https://doi.org/10.5194/acp-21-4561-2021, 2021.

Sharma, S. K., Mandal, T. K., Sharma, C., Kuniyal, J. C., Joshi, R., Dhyani, P. P., Rohtash, Sen, A., Ghayas, H., Gupta, N. C., Sharma, P., Saxena, M., Sharma, A., Arya, B. C., and Kumar, A.: Measurements of Particulate (PM2.5), BC and Trace Gases Over the Northwestern Himalayan Region of India, MAPAN, 29, 243–253, https://doi.org/10.1007/s12647-014-0104-2, 2014.

Wentworth, G. R., Murphy, J. G., Benedict, K. B., Bangs, E. J., and Collett Jr., J. L.: The role of dew as a night-time reservoir and morning source for atmospheric ammonia, Atmospheric Chem. Phys., 16, 7435–7449, https://doi.org/10.5194/acp-16-7435-2016, 2016.