Anonymous Referee #1

The authors have comprehensively addressed most of my previous comments through a revised title and an enhanced discussion of the relationship between NH_3 concentration, deposition, and their emission contributions. To further strengthen the manuscript, the following minor revisions are required:

We sincerely appreciate and thank the reviewer 1 for the second round of valuable suggestions and comments. We have revised this MS based all comments, more details are replied below.

1. Line 143 and 561: explain the terms "ABL" and "CAGR method" is

Done as suggested, the term “ABL” is atmospheric boundary layer which has been explained on line 99 “the atmospheric boundary layer (hereafter ABL)", where it first appeared; and the “CAGR method” is “compound annual growth rate”. To make clarification, we have added corresponding explanation on line 561 as “We used compound annual growth rate (CAGR) method”.

2. Line 266-268: Is there a reference for this linear regression approach?

Done as suggested, we added two references for the linear regression approach on line 264 as “(Hu et al., 2017; Liu et al., 2024)”.

Liu C, Huang J, Hu C. et al. Sensitivity of surface downward longwave radiation to aerosol optical depth over the Lake Taihu region, China, Atmospheric Research, 305 (2024) 107444

Hu, C, Wang Y, Wang W, et al. Trends in evaporation of a large subtropical lake. Theoretical and Applied Climatology, 2017, 129, 159–170, https://doi.org/10.1007/s00704-016-1768-z.

3. Line 348-350: add references

Done as suggested, we have added two references and citations as of “Crippa et al., 2019; Liu et al., 2024” on line 349.

Crippa, M., Janssens-Maenhout, G., Guizzardi, D., Van Dingenen, R., and Dentener, F.: Contribution and uncertainty of sectorial and regional emissions to regional and global $PM_{2.5}$ health impacts, Atmos. Chem. Phys., 19, 5165–5186, https://doi.org/10.5194/acp-19-5165-2019, 2019.

Liu, H., Hu, C., Xiao, Q., Zhang, J., Sun, F., Shi, X., Chen, X., Yang, Y., and Xiao, W.: Analysis of anthropogenic CO_2 emission uncertainty and influencing factors at city scale in Yangtze River Delta region: One of the world’s largest emission hotspots, Atmospheric Pollution Research, 15, 102281, https://doi.org/10.1016/j.apr.2024.102281, 2024

4. Line 614: The statement “y = 0.35 + 0.16 was applied to all scatter plots” is unclear. If you applied a multiplicative scaling (i.e., $y_i = a \times y\_pred$), both R^2 and RMSE would change, as the variance of residuals relative to y changes nonlinearly. If you instead applied an additive shift (bias correction), R^2 might remain almost the same (since R^2 is based on variance, not the mean) or slightly increase. Could you clarify which correction was applied? In addition, I believe what you are doing is a form of “K-theory” (gradient diffusion theory), based on the well-mixed assumption in the ABL. It assumes that transport flux can be represented analogously to molecular diffusion, where fluxes are

proportional to the mean gradient of the transported quantity.

Thanks so much for this suggestion, yes, the approach we used here is applying an additive shift (bias correction), where R^2 remain almost the same, as mentioned in this comment. It's also based on "K-theory" (gradient diffusion theory) with the well-mixed assumption in the ABL. It assumes that transport flux can be represented analogously to molecular diffusion, where fluxes are proportional to the mean gradient of the transported quantity.

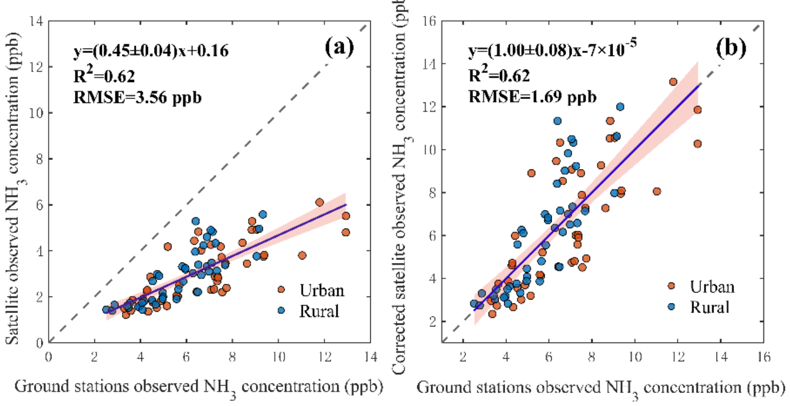
To make clarification, we also added more explanation on lines 611-616 as "The approach we used is applying an additive shift (bias correction), where R^2 remain almost the same (Figures 4a-b), it's also based on "K-theory" (gradient diffusion theory) with the well-mixed assumption in the ABL. This method assumes that transport flux can be represented analogously to molecular diffusion, where fluxes are proportional to the mean gradient of the transported quantity.".

5. Line 789: Should the contribution results correspond to fig. 10b?

Thanks so much and we have double checked the results on line 789, the contribution results correspond to figure 10a, and we feel sorry of lacking corresponding descriptions. To avoid misunderstanding, we have added more explanation of how the relative contributions were calculated on lines 817-830 as "The above relative contributions are calculated by the method that using the emissions and meteorological-hydrological factors from 2013 as the baseline (more details in Method Section 2.6.1), we first simulated the NH_3 concentration for 2022 using the emissions and meteorological-hydrological factors from that year. The simulated concentration was 3.08 ppb, which was consistent with the satellite-observed concentration for 2022, yielding a relative error of 0.1%. Subsequently, we replaced the emissions data with those from 2013 while keeping the 2022 meteorological-hydrological factors constant, resulting in a simulated concentration of 3.14 ppb. We then replaced the meteorological-hydrological factors with those from 2013 while keeping the 2022 emissions constant, leading to a simulated concentration of 3.10 ppb. By subtracting the two simulated concentrations from the 2022 NH_3 concentration simulation, we quantified the effects of changes in emissions and meteorological-hydrological factors on NH_3 concentration. Finally, the results were normalized, revealing that the relative contributions of emissions and meteorological-hydrological factors to the concentration changes were 77.4% and 22.6%, respectively. ".

6. Fig. 4: adjust the y-label or include it in the title of plot (a) and (b) to clarify their differences.

Done as suggested, we have revised the y-label of figure 4b with "Corrected satellite NH_3 concentration (ppb)".



7. Table S9: Specify whether the values represent annual emission reductions.

Yes, the values in Tables S9 represent annual emission reductions, to make clarification, we have revised the label of Table S9 as "Table S9. Average annual reduction (Tg yr^{-1}), annual reduction rates (% yr^{-1}), and relative rates between 2013 and 2023 (%) of SO_2, NO_x, and NH_3."

8. Figure S2: The figure does not display properly (appears black).

Thanks so much for pointing it out, it may be caused by transformation from word version to pdf version, we have replaced this figure in new version.

Referee #2

The manuscript attempts to explore decade variations atmospheric ammonia (NH_3) in terms of methodological integration, spatiotemporal resolution, and data-processing framework. It represents an important contribution to understanding NH3 pollution and dry deposition dynamics across China. Therefore, the manuscript can be accepted after a minor revision.

However, to further enhance the paper's impact and reliability, the authors should strengthen and deepen several aspects, particularly regarding uncertainty quantification and potential changes in NH3 sinks.

We sincerely appreciate and thank reviewer 2 for the second round of valuable suggestions and comments. We have revised this MS based all comments, especially added more explanation and discussions on the uncertainty quantification and potential changes in NH_3 dry deposition and sinks, more details are replied below.

1.Although the authors empirically converted CrIS "near-surface" concentrations (0–1 km) to "ground-level" concentrations (1–1.5 m), they did not sufficiently validate potential systematic biases under varying thermal contrast and boundary-layer conditions, especially in high-altitude or low-temperature regions. Based on reading of this review, vertical profiles of atmospheric NH3 likely changed little in the height below 300 m in today's China. This linear extrapolation neglects the strong vertical gradient of NH_3, which could lead to overestimation in high-concentration regions and underestimation in low-concentration regions. The authors should quantify how such bias may affect the overall uncertainty of their results.

Thanks so much for pointing it out, we agree that there may be spatial changes of relationship between "near-surface" concentrations (0–1 km) and "ground-level" concentrations (1–1.5 m), where the variations of scatter plots in Figure 4a-b also reflects such uncertainty. We further calculated the uncertainty extent of regression slope, which were 0.45±0.04, indicating the above mentioned potential systematic biases under varying thermal contrast and boundary-layer conditions can be 9%, calculated by dividing uncertainty extent of 0.04 with 0.45. These uncertainties can be decreased by conducting more ground site based NH_3 observations in different regions.

Furthermore, we have added more clarifications and discussions on lines 624-633 as "It is important to acknowledge that spatial-temporal uncertainties or potential systematic biases may exist in the relationship between ground-based and satellite-derived NH_3 observations across different regions and under varying thermal contrast and boundary-layer conditions. As demonstrated in the scatter plots of Figure 4a-b, which exhibit significant variability. Nevertheless, the regression slope and associated uncertainty were 0.45 ± 0.04, indicating that the potential systematic biases mentioned above could result in an error of approximately 9% when deriving ground-level NH_3 concentrations and dry deposition rates. This error was calculated by dividing the uncertainty extent (0.04) by the regression slope (0.45). These uncertainties can be mitigated by increasing the number of ground-based NH_3 observations in diverse regions in future studies."

2.The manuscript mentions the combined use of the GEOS-Chem and random forest (RF) models but does not clearly describe the RF model's input variables or the feature-importance ranking. These details should be added to ensure methodological transparency and reproducibility.

Done as suggested, we have added the figure of the feature-importance ranking below and discussions in Supplementary file, and more details have also been added as "The feature-importance ranking figure illustrates the relative importance of eight driving factors in predicting NH_3 concentrations using the Random Forest model (Figure S15, *SI*). Among the emission and meteorological-hydrological factors, the latter plays a more prominent role in explaining the spatial and temporal variability of NH_3 concentrations. Within the meteorological-hydrological factors, the 10-meter wind speed (20.3%), 2-meter temperature (14.9%), and boundary layer height (13.1%) are the most influential variables affecting the NH_3 concentration simulation. These variables collectively reflect the role of atmospheric diffusion capacity and volatilization conditions in regulating the distribution of NH_3 concentrations. Total precipitation (11.0%) and surface soil moisture content (13.6%) contribute to the removal of NH_3 from the atmosphere, though their relative importance is lower. Among the emission factors, NH_3 emissions (16.4%) are the most significant, followed by NOx (11.0%) and SO_2 (5.1%) emissions. This suggests that, in addition to direct emissions, precursor chemical processes also have an indirect influence on the distribution of NH_3 concentrations. ".

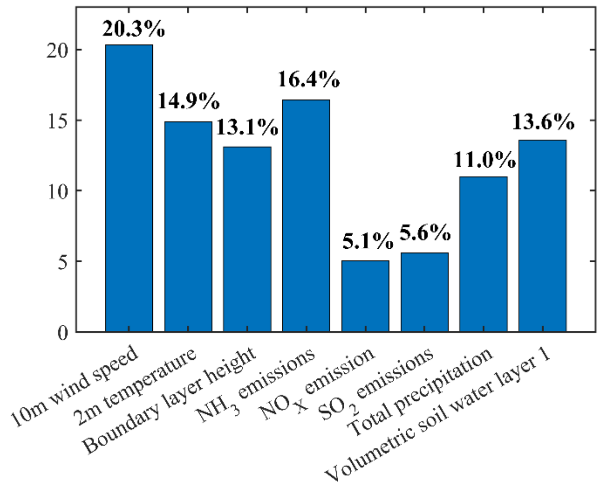


Figure S15. Feature-importance ranking figure illustrates the relative importance of eight driving factors in predicting NH_3 concentrations using the Random Forest model.

3.The conclusion states that "77.4% of the trend was explained by anthropogenic sources," yet the structure of variables, characteristics of the training set, and the attribution approach are not clearly explained. The change in NH3 sinks might not support the statement, based on a simple comparison between the decrease in particulate NH4+ and corresponding increase in atmospheric NH3 in US as well as the decrease in particulate NH4+in China.

Done as suggested, we have provided more details and discussions regarding how these values were calculated and potential uncertainty on lines 799-812 as "The above relative contributions are calculated by the method that using the emissions and meteorological-hydrological factors from 2013 as the baseline, we first simulated the NH_3 concentration for 2022 using the emissions and meteorological-hydrological factors from that year. The simulated concentration was 3.08 ppb, which was consistent with the satellite-observed concentration for 2022, yielding a relative error of 0.1%. Subsequently, we replaced the emissions data with those from 2013 while keeping the 2022 meteorological-hydrological factors constant, resulting in a simulated concentration of 3.14 ppb.

We then replaced the meteorological-hydrological factors with those from 2013 while keeping the 2022 emissions constant, leading to a simulated concentration of 3.10 ppb. By subtracting the two simulated concentrations from the 2022 NH_3 concentration simulation, we quantified the effects of changes in emissions and meteorological-hydrological factors on NH_3 concentration. Finally, the results were normalized, revealing that the relative contributions of emissions and meteorological-hydrological factors to the concentration changes were 77.4% and 22.6%, respectively." .

4.The language could benefit from additional professional editing or AI-assisted polishing to improve fluency and conciseness, particularly by simplifying lengthy or complex sentences.
Done as suggested, three co-authors are native speaker of English and also editor for many journals, they have provided professional editing during the draft written and revision process.

5. The effective number are total off through the manuscript, and should be corrected.
Done as suggested, we have double checked through this MS, and the relative changes of NH_3 concentration values are displayed with one decimal place, concentration values are displayed with two decimal places, and the trend analysis are displayed with three decimal places considering these values are relatively small.

6. Lines 485-487 "In China, summer fertilization for maize cultivation—often involving both mineral and organic fertilizers—contributes to the observed summer peak (Paulot et al., 2014)." This is not true in today's China, on basis of the reviewer's summer traveling experience therein.
Thanks so much for pointing it out, the main agricultural crops in China contains maize and rice paddy, corn and wheat, which is applied with fertilized in summer. We have revised this sentence as: "In China, summer fertilization is applied for the key agricultural crops as rice paddy, maize, corn and wheat—often involving both mineral and organic fertilizers—contributes to the observed summer peak (Paulot et al., 2014; Luo et al., 2025)"