

Dear Editor,

We are truly grateful to yours and other reviewer's critical comments and thoughtful suggestions on our manuscript "Measurement report Nitrogen Isotope ($\delta^{15}\text{N}$) Signatures of Ammonia Emissions from Livestock Farming Implications for Source Apportionment of Haze Pollution". Those comments and suggestions are helpful not only for improving the present manuscript but also our future research. The paper has been carefully revised based on the comments and suggestions. And we hope our revision has met with your approval now. The changes in the revised manuscript are marked in blue.

The following is the point-point change list.

Modification to the comments of Reviewer

Reviewer #1: L51: Delete "for."

Thank you for your suggestion. We have already removed the error in the original text.

2: L84: Clarify the name, principle, and technical details of the MixSAIR model.

Thank you for your suggestion. The Bayesian stable isotope mixing model MixSAIR is primarily used to allocate contributions of atmospheric emission sources through isotope analysis. MixSIAR is a stable isotope mixing model based on the Bayesian statistical framework, designed to quantitatively analyze the relative contributions of multiple potential sources to the isotopic composition of observed mixtures. Its fundamental assumption posits that the isotopic signature of a mixture can be expressed as a linear combination of the isotopic characteristics of each source weighted by their proportional contributions, while explicitly accounting for source variability, measurement errors, and isotopic fractionation.

3: L108: Specify the principle, detection limit, and potential interferences of the method.

Thank you for your suggestion. The sampling principle is based on active air

sampling combined with aqueous absorption. Ambient air was continuously drawn through an impinger containing deionized water, in which gaseous NH_3 was absorbed and converted to dissolved NH_4^+ . After sampling, the absorption solution was quantitatively recovered for subsequent laboratory analysis. Under the applied sampling flow rate and duration, the method detection limit for atmospheric NH_3 was on the order of 3 ppb, which is adequate for resolving ambient concentration variations during the observation period. Potential interferences include the co-collection of particulate NH_4^+ and the absorption of other water-soluble alkaline gases. These effects were minimized by controlled sampling duration, appropriate flow rates, and blank correction procedures, and are considered to have a negligible influence on the measured NH_3 concentrations.

4: L109: Flow rate may affect calculated NH_3 concentrations. Please explain how results obtained with different flow rates were compared.

Thank you for your suggestion. All NH_3 samples were collected at a constant and identical flow rate throughout the study period, with all flow rates complying with the National Standard GB 3095-2012.

5: L116: Remove “T.”

Thank you for your suggestion, we have already removed the error in the original text.

6: Figure 1 is cluttered and confusing. Please distinguish which data are from your experiments and which are from the literature. Were sampling periods during haze and clean conditions conducted at the same locations? Are the cattle, layer, and fattening pig farms all located in the same city? If so, please include a detailed map showing the sampling points.

Thank you for your feedback. We have made changes to Figure 1.

7: L117–118: Specify the absorption solution used.

Thank you for your suggestion. According to the sample requirements for mass spectrometry pretreatment, we used deionized water as the absorption solution.

8: L122: Which building was selected? Please justify its representativeness for intensive laying-hen farms.

Thank you for your suggestion. Sampling was conducted in a typical commercial intensive laying-hen house with a conventional cage-based rearing system, representative of large-scale laying-hen farms in northern China. This question was not answered. We collected gases from the exhaust vents of chicken houses and pig houses. These types of animal housing have centralized air inlets and outlets, so collecting from the exhaust vents can represent the ammonia emissions from these two types of housing into the atmosphere. For cattle farms, since the barns are open, we selected cattle sheds located in the middle of the farm to more effectively collect ammonia gas.

9: L144: Should this be “mg L⁻¹”?

Thank you for your suggestion, We have checked and revised it though out the manuscript.

10: L149: Specify what analytical method is being referred to.

Thank you for your suggestion. The analytical method for N isotope determination employs the hypobromite-hydroxylamine hydrochloride chemical method(Song et al., 2024).

11: The statistical methods should be clearly described.

Thank you for your suggestion. NH₃ concentrations and $\delta^{15}\text{N}$ values are presented as mean \pm standard error (SE). Differences in $\delta^{15}\text{N}$ values among livestock categories were evaluated using one-way analysis of variance (ANOVA). When data did not meet the assumptions of normality or homogeneity of variance, non-parametric tests were applied. Statistical significance was defined at $p < 0.05$. All statistical analyses were conducted using standard statistical software.

12: L185: Should this be “1.7 to 2.5”?

Thank you for your suggestion. We have checked and revised it though out the manuscript.

13: L205: Add a space after “(a)” and “(b).”

Thank you for your suggestion. We have checked and revised it though out the manuscript.

14: L188: Replace the comma before “For” with a period.

Thank you for your suggestion. We have checked and revised it though out the manuscript.

15: Discuss the relationship between temperature and NH₃ emissions, and cite relevant references to support the explanation.

Thank you for your suggestion. Temperature is the primary environmental driver of ammonia volatilization from agricultural sources (including livestock manure and manure management systems). As air and surface temperatures rise, the thermodynamic equilibrium shifts toward gaseous ammonia while the solubility of NH₃ in water decreases, thereby increasing the volatilization rate. Higher temperatures also elevate the vapor pressure and diffusion rate of NH₃ at the manure surface, and accelerate microbial processes such as ammonification. These factors collectively contribute to increased NH₃ emissions under high-temperature conditions.

16: L190: Add a space before “ $\delta^{15}\text{N}$.”

Thank you for your suggestion. We have checked and revised it though out the manuscript.

17: L192: Delete “From May to June”; change to “ $\delta^{15}\text{N-NH}_4^+$.”

Thank you for your suggestion. We have already corrected this error in the manuscript.

18: L194–195: The variations appear too large to support significant differences.

Thank you for your suggestion. Although relatively large variability was observed within each livestock category, the differences in mean $\delta^{15}\text{N}$ values among groups were statistically significant (one-way ANOVA, $p < 0.05$). The large within-group variability reflects realistic operational and environmental heterogeneity and does not negate the statistically significant differences observed among livestock categories.

19: L230: Change to “Martine et al.”

Thank you for your suggestion. We have already corrected this error in the original text.

20: L234: Should this be “David et al. (Felix et al., 2014)”?

Thank you for your suggestion. This citation was marked as an error and has been corrected in the original text.

21: L241–243: Add citations to support the argument.

Thank you for your suggestion. We have added relevant literature to support the argument.

22: L251–254: Include references and source apportionment calculations to substantiate the conclusions.

Thank you for your suggestion. We conducted source apportionment for haze and clean weather using the MixSIAR model. The results showed that combustion and traffic were the main contributing sources, with combustion accounting for 29.0%, traffic for 38.0%, agriculture for 15.1%, and livestock for 17.8%.

23: L257: Add a space after “(a)” and “(b).”

Thank you for your suggestion. We have already corrected this error in the original text.

24: Figure 4: Clarify the meaning of the boxes or violin plots.

Thank you for your suggestion. Comparison of $\delta^{15}\text{N-NH}_4^+$ values among different livestock systems and with previously reported source signatures. Boxes represent the interquartile range, the horizontal line within each box denotes the median value, and whiskers indicate the minimum and maximum values excluding outliers. Individual points outside the whiskers represent statistical outliers.

25: Figure 5: Provide P-values. The R^2 values are below 0.1 in both sub-figures, indicating very weak correlation between GDP/year and $\delta^{15}\text{N-NH}_4^+$.

Thank you for your suggestions. We have provided *p-values* for GDP and $\delta^{15}\text{N-NH}_4^+$, as well as for year and $\delta^{15}\text{N-NH}_4^+$, with both sets of *p-values* being $p < 0.001$.

26: L268–271: Figure 5 does not present data on contributions from combustion and vehicular sources. Please provide supporting data.

Thank you for your suggestion. The original text has been revised accordingly.

As shown in Figure 5b, the $\delta^{15}\text{N}$ values exhibited an increasing trend from 2018 to 2020.

27: L272: Add a space after “signatures.”

Thank you for your suggestion. We have already corrected this error in the original text.

28: L274: Add a space after “emissions.”

Thank you for your suggestion. We have already corrected this error in the original text.

29: L282–283: Figure 5b does not show the proportion of $\delta^{15}\text{N-NH}_4^+$ attributed to combustion and vehicular sources. Please address this.

Thank you for your suggestion. The original text has been revised accordingly.

As shown in Figure 5b, the $\delta^{15}\text{N}$ values exhibited an increasing trend from 2018 to 2020.

References.

Song, L., Wang, A., Li, Z., Kang, R., Walters, W. W., Pan, Y., Quan, Z., Huang, S., and Fang, Y.: Large seasonal variation in nitrogen isotopic abundances of ammonia volatilized from a cropland ecosystem and implications for regional NH_3 source partitioning, *Environ. Sci. Technol.*, 58, 1177–1186, <https://doi.org/10.1021/acs.est.3c08800>, 2024.