Response Letter to Editor/Reviewers' Comments

We sincerely appreciate the time and effort you have invested in reviewing our manuscript. And thanks very much for the helpful comments and suggestions from editors and reviewers. Now we have revised the manuscript carefully according to these comments point by point. The changes can be seen in the modified version of the revised manuscript. The major changes are also cited in the response letter, and marked with red color.

Referee #1:

General comments:

Contamination of groundwater systems by high nitrogen emissions from agricultural land use are one of the most common and serious problems of water resources management and research at the global scale. Thus, the paper addresses a highly relevant problem. The manuscript combines hydrochemical analysis (PCA-APCS-MLR) and stable isotope mixing modeling (MixSIAR) to identify and quantify NO_3^--N pollution sources in groundwater for unconfined and confined aquifers. It compares the result of computing the apportionment, assuming the two aquifers as a single water body, or as two water bodies. The argument for such comparison is that previous studies on NO_3^--N source apportionment have largely neglected aquifer type. I tend to accept it after appropriate revisions.

Response:

Thank you very much for your thorough and constructive review of our manuscript, and we sincerely appreciate the time and effort you have invested in evaluating our work. We are truly grateful for your valuable and professional comments, which will be of great help in enhancing the quality of our work. We have refined our manuscript carefully to bring our work to the high standards expected by the journal.

Detailed comments:

1. The term "burial conditions" is not appropriate in this context. Please consider using more precise hydrogeological terminology, such as "groundwater occurrence conditions", to better reflect the intended meaning.

Response:

Thank you for this helpful comment. As suggested, we have replaced “burial conditions” with the more precise hydrogeological term “groundwater occurrence conditions” throughout the revised manuscript.

Page 1, Title:

Impact of ~~burial~~ occurrence conditions on NO_3^--N source apportionment in groundwater: Insights from PCA-APCS-MLR and MixSIAR methods

Please see L. 24, L. 25, L. 41, L. 58, L. 64, L. 87, L. 94, L. 108, L. 115, L. 117, L. 156, L. 161, etc. in the revised manuscript for other changes.

2. The manuscript uses several abbreviations without introducing their full forms upon first use. The full form of each abbreviation should be provided at its first occurrence (e.g., Nitrate-N (NO_3^--N)).

Response:

Thank you for highlighting this oversight. We have carefully reviewed the manuscript and have now introduced the full form for each abbreviation at its first occurrence (e.g., "Nitrate-N (NO_3^--N)").

Page 1, Abstract, Line 20:

Nitrate-N (NO_3^--N) contamination in groundwater poses a significant threat to drinking water safety and ecosystem health, with accurate source identification being crucial for effective pollution control.

Page 4, Introduction, Line 67:

Groundwater nitrate-N (NO_3^--N) contamination has persisted for nearly a century worldwide, emerging as a critical environmental challenge that threatens both human health and ecological security.

3. Sample-testing and correlation methods lack procedural detail and citations. Please add detailed information.

Response:

Thank you for this important comment. We acknowledge that the description of the sample-testing and correlation methods was insufficient. In the revised manuscript, we have added detailed procedural steps for both the experimental assays and the statistical correlation analysis. Furthermore, we have included appropriate citations to support the chosen methodologies.

Page 10, Materials and methods, Line 239:

~~The concentration of NO_3^--N was determined using ultraviolet spectrophotometry. The concentrations of K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, and SO_4^{2-} were measured using an ion~~

~~chromatograph (ICS-3000, Dionex, USA), the concentration of HCO_3^- was determined by acid-base titration.~~

The concentration of NO_3^--N was determined using the ultraviolet spectrophotometric method (at 220 nm and 275 nm) following filtration through a 0.45 μm membrane. The concentrations of major ions (K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, and SO_4^{2-}) were measured using an ion chromatograph (ICS-3000, Dionex, USA). The separation was achieved with an IonPac AS23 analytical column and an AG23 guard column, using a carbonate eluent. The concentration of HCO_3^- was determined by acid-base titration with a standardized HCl solution (0.02 M) to a bromocresol green-methyl red endpoint. All analyses of water quality indicators adhered to standard methods (Greenberg et al., 2005).

References

Greenberg, A.E., Trussell, R.R., Clesceri, L.S., Association, A.W.W., 2005. Standard methods for the examination of water and wastewater : supplement to the sixteenth edition. American Journal of Public Health & the Nations Health 56(3), 387.

Page 13, Materials and methods, Line 317:

The stable isotope mixing model used in this study was run in the R package MixSIAR (R version x64 4.3.2). ~~The Pearson correlation test was employed to evaluate the relationships between hydrochemical indices, with data analysis conducted using SPSS 20.~~ Statistical analysis was performed using SPSS 20 software. To evaluate the linear relationships between hydrochemical parameters, the Pearson correlation coefficient (r) was calculated. Correlations were considered statistically significant at a two-tailed p-value < 0.05.

Page 12, Materials and methods, Line 282:

Subsequently, on the basis of PCA, the absolute principal component scores (APCS) were determined. A multiple linear regression (MLR) was performed with the measured pollutant concentrations as the dependent variables and the absolute principal component scores as the independent variables (Thurston and Spengler, 1985).

References

Thurston, G.D., Spengler, J.D., 1985. A quantitative assessment of source contributions to inhalable particulate matter pollution in metropolitan Boston. Atmospheric

Environment (1967) 19(1), 9-25.

Page 13, Materials and methods, Line 303:

The principle of the MixSIAR method is to use the Dirichlet distribution as the prior distribution and to obtain the posterior distribution characteristics of the contributions, such as the mean, variance, and probability density, through the application of Bayes' theorem (Moore and Semmens, 2008).

References

Moore, J.W., Semmens, B.X., 2008. Incorporating uncertainty and prior information into stable isotope mixing models. Ecol. Lett. 11(5), 470-480.

4. L. 235: Add citation to the original work by Thurston and Spengler (1985) (https://doi.org/10.1016/0004-6981(85)90132-5) who proposed the APCS-MLR method.

Response:

Thank you for your valuable comment. We have added the suggested citation to the original work by Thurston and Spengler (1985). The reference has been included in the revised manuscript.

Page 12, Materials and methods, Line 282:

Subsequently, on the basis of PCA, the absolute principal component scores (APCS) were determined. A multiple linear regression (MLR) was performed with the measured pollutant concentrations as the dependent variables and the absolute principal component scores as the independent variables (Thurston and Spengler, 1985).

References

Thurston, G.D., Spengler, J.D., 1985. A quantitative assessment of source contributions to inhalable particulate matter pollution in metropolitan Boston. Atmospheric Environment (1967) 19(1), 9-25.

5. L. 256: Add citation to Moore and Semmens (2008) (DOI: 10.1111/j.1461-0248.2008.01163.x) and Stock and Semmens (2016) (doi:10.5281/zenodo.1209993.)for the MixSIAR method and R code.

Response:

As requested, we have added the following citations on line 256 (and in the reference list) of the revised manuscript.

Page 13, Materials and methods, Line 303:

The principle of the MixSIAR method is to use the Dirichlet distribution as the prior distribution and to obtain the posterior distribution characteristics of the contributions, such as the mean, variance, and probability density, through the application of Bayes' theorem (Moore and Semmens, 2008).

References

Moore, J.W., Semmens, B.X., 2008. Incorporating uncertainty and prior information into stable isotope mixing models. Ecol. Lett. 11(5), 470-480.

6. What do you mean by "aquifer burial conditions"? Does that term refer to the differentiation of "single-layer" and "double-layer" aquifers (but sometimes called "single-aquifer layer" and "double-aquifer layer" instead)?

Response:

Thank you for your question regarding the term "aquifer burial conditions". You are correct in your understanding – it does refer to the differentiation between "single-layer" and "double-layer" aquifers. We adopted this specific terminology because it is commonly used in relevant hydrological literature to describe aquifer architecture (Baudron et al., 2013; Qian et al., 2020). In addition, we have revised the manuscript to replace "burial conditions" with the more standard term "occurrence conditions" to avoid any potential ambiguity.

References

Baudron, P., Alonso-Sarría, F., García-Aróstegui, J.L., Cánovas-García, F., Martínez-Vicente, D., Moreno-Brotóns, J., 2013. Identifying the origin of groundwater samples in a multi-layer aquifer system with Random Forest classification. J. Hydrol. 499, 303-315.

Qian, H., Chen, J., Howard, K.W., 2020. Assessing groundwater pollution and potential remediation processes in a multi-layer aquifer system. Environ. Pollut. 263, 114669.

7. Add an explanation on why atmospheric deposition (4-5 %) was kept in the model even though its contribution is minor, this justifies its inclusion for completeness.

Response:

Thank you for this helpful suggestion. Although atmospheric deposition contributes only 4-5% to the total NO_3^--N in the studied aquifers, it was retained in the MixSIAR model for three main reasons:

(1) Completeness of source apportionment: Atmospheric deposition is a known natural input of nitrate in groundwater systems, and its inclusion ensures that the model accounts for all potential sources, thereby reducing structural uncertainty in the source apportionment.

(2) Isotopic distinction: Atmospheric nitrate exhibits distinct $\delta^{15}N$ and $\delta^{18}O$ signatures (typically low $\delta^{15}N$ and high $\delta^{18}O$), which help to better constrain the mixing space and improve the discrimination among the other major sources (e.g., chemical fertilizers, manure & sewage, and soil nitrogen).

(3) Model consistency and comparability: Retaining all recognized sources aligns with established practices in isotopic mixing modeling and facilitates comparison with other regional or global studies where atmospheric deposition may play a more significant role.

Omitting this minor source could artificially inflate the contributions of the remaining sources and affect the robustness of the proportional estimates, even if its absolute contribution is small.

8. The discussion section remains descriptive and does not yet place the new findings in the wider context of existing literature. Please systematically compare your findings with previous studies.

Response:

Thank you for your careful review. We fully agree with your suggestion. In response, we have significantly revised and expanded the Discussion section to enhance its scientific depth.

Page 24, Discussion, Line 501:

The PCA-APCS-MLR method has the advantages of being rapid and convenient, but it has the disadvantage of being unable to further identify soil nitrogen as a pollution source. Accurately apportioning soil-derived nitrate is particularly challenging for hydrochemical-based methods due to the overlapping ionic signatures between soil nitrogen and the leaching of synthetic fertilizers or organic wastes (Yu et al., 2022). To compensate for this limitation, the MixSIAR method was further employed to analyze the sources of pollution.

References

Yu, L., Zheng, T., Yuan, R., Zheng, X., 2022. APCS-MLR model: a convenient and fast method for quantitative identification of nitrate pollution sources in groundwater. J. Environ. Manage. 314, 115101.

Page 24, Discussion, Line 513:

Because NO_3^--N from different sources (atmospheric deposition, soil nitrogen, chemical fertilizers, and manure & sewage) has distinct isotopic signatures, and isotopic fractionation occurs during the transport and transformation processes of nitrogen in the groundwater system (such as ammonification and nitrification), leading to changes in the $\delta^{15}N$ and $\delta^{18}O$ values of NO_3^--N (Shu et al., 2024). Our MixSIAR model incorporated fractionation factors of specific nitrogen transformation processes. This approach, essential for reliable quantification, aligns with established practice in nitrate source apportionment studies, where such constraints are proven to substantially reduce uncertainty (Wang et al., 2023).

References

Wang, D., Li, P., Yang, N., Yang, C., Zhou, Y., Li, J., 2023. Distribution, sources and main controlling factors of nitrate in a typical intensive agricultural region, northwestern China: vertical profile perspectives. Environ. Res. 237, 116911.

Page 25, Discussion, Line 525:

The MixSIAR method further revealed that soil nitrogen is a dominant pollution source for unconfined groundwater, with a higher contribution rate than that of chemical fertilizers. The identification of soil nitrogen as a major contributor in the unconfined groundwater is significant. Legacy soil nitrogen constitutes a dominant source in the unconfined aquifer, a finding that shifts the pollution focus from direct fertilizer inputs to cumulative soil nitrogen pools. This finding is consistent with previous nitrate source studies conducted in other regions (Cui et al., 2023). For confined groundwater, MixSIAR also confirmed that manure & sewage are the major sources of NO_3^--N.

References

Cui, R., Zhang, D., Hu, W., Zhao, X., Yan, H., Liu, G., Chen, A., 2023. Nitrogen in soil, manure and sewage has become a major challenge in controlling nitrate pollution in groundwater around plateau lakes, Southwest China. J. Hydrol. 620, 129541.

Page 25, Discussion, Line 540:

While NH_4^+-N can be adsorbed and immobilized by the soil, NO_3^--N can leach into the deeper vadose zone and aquifer through atmospheric precipitation or agricultural irrigation, directly contaminating unconfined groundwater (Wan et al., 2024). This process underscores the phenomenon of the soil and vadose zone as a dynamic “nitrogen reactor and buffer”. Similar delayed release mechanisms from legacy nitrogen stored in thick unsaturated zones have been documented in arid regions (Li et al., 2025), indicating that the risk of groundwater contamination may persist long after surface inputs are reduced. Therefore, in assessing the sources of NO_3^--N pollution in regional groundwater, it is crucial not only to focus on the application rates of chemical fertilizers but also to pay attention to the storage of nitrogen in the soil and vadose zone.

References

Li, S., Chow, R., Su, H., Han, F., Li, Z., 2025. Multiple isotopes and GIS analyses reveal sources and drivers of nitrate in the Loess Plateau's groundwater. Environ. Pollut., 127022.

Page 26, Discussion, Line 562:

These pollutants can enter surface water bodies through surface runoff or infiltration and then gradually transport to deeper aquifers via the interflow recharge process between unconfined and confined aquifers, resulting in persistent contamination (McDonough et al., 2022). This may highlight a potential mechanism for sustained nitrate pollution in confined systems, which can be attributed to manure/sewage sources transported via aquifer exchange, providing a continuous input of recalcitrant nitrogen that gradually accumulates in this zone (Zhang et al., 2026). Therefore, for the prevention and control of NO_3^--N pollution in confined aquifers, it is crucial to focus on the source control of manure & sewage to block the migration pathways of pollutants and mitigate their long-term impacts on confined aquifers.

References

Zhang, T., Lei, Q., Liu, H., Ding, K., Luo, J., Qiu, W., Ma, Y., Xu, Q., Zhao, Y., Liu, X., 2026. Vertical migration of multi-source nitrates driven by multiple water inputs in groundwater base on combined nitrogen-oxygen and hydrogen-oxygen isotopes. J. Hazard. Mater., 141181.

Page 27, Discussion, Line 581:

First, the sources and recharge mechanisms of groundwater in unconfined and confined aquifers differ significantly (Liu et al., 2025), leading to distinct isotopic compositions and characteristic values. For example, the isotopic signature of a pollution source in an unconfined aquifer may resemble that of another source in a confined aquifer. Such overlapping isotopic signals can lead to ambiguous source identification. However, previous studies either do not explicitly distinguish between groundwater occurrence conditions or rely on land-use as a primary proxy for pollution source identification (Yu et al., 2020). When mixed calculations are performed without considering the actual occurrence conditions, the isotopic differences are obscured, resulting in confusion in pollution source identification, inaccurate contribution rate calculations, and incomplete analysis of pollution processes.

References

Liu, W., Du, Y., Qiu, W., Deng, Y., Wang, Y., 2025. Constraints on vertical variability of geogenic ammonium in multi-layered aquifer systems. Water Res. 268, 122639.

Yu, L., Zheng, T., Zheng, X., Hao, Y., Yuan, R., 2020. Nitrate source apportionment in groundwater using Bayesian isotope mixing model based on nitrogen isotope fractionation. Sci. Total Environ. 718, 137242.

Page 27, Discussion, Line 593:

Hydrogeological conditions can influence the intensity of biogeochemical processes such as ammonification, nitrification, denitrification (Dai et al., 2023), and adsorption (Huang et al., 2022; Li et al., 2023), which further alter NO_3^--N concentrations and isotopic signatures.

References

Dai, H., Zhang, Y., Fang, W., Liu, J., Hong, J., Zou, C., Zhang, J., 2023. Microbial community structural response to variations in physicochemical features of different aquifers. Front. Microbiol. 14, 1025964.

9. I suggest putting forward more specific policy recommendations and summarizing the limitations of the research and its future directions in the discussion.

Response:

We thank the reviewer for this constructive suggestion. In the revised manuscript, we have added the dedicated content that outlines more specific policy recommendations based on groundwater occurrence conditions, such as promoting optimized irrigation and slow-release fertilizers in unconfined aquifers and strengthening manure-and-sewage source control in confined aquifers, and also summarises the limitations of this work (e.g., spatiotemporal sampling constraints and uncertainties in isotopic signatures) together with future research directions, notably the integration of reactive-transport modelling with isotopic mixing models to better elucidate nitrogen transformation processes. These additions strengthen the policy relevance and scholarly rigour of the study.

Page 27, Discussion, Line 596:

Hydrogeological conditions can influence the intensity of biogeochemical processes such as ammonification, nitrification, denitrification, and adsorption (Huang et al., 2022; Li et al., 2023), which further alter NO_3^--N concentrations and isotopic signatures. This ultimately affects the accuracy and reliability of pollution source apportionment. Consequently, effective management of NO_3^--N in groundwater systems requires policy interventions tailored to specific groundwater occurrence conditions. In unconfined groundwater, which are highly vulnerable to surface-derived inputs, management should prioritize agricultural best practices such as optimized irrigation scheduling — reducing both frequency and volume of irrigation — coupled with the promotion of slow-release or stabilized nitrogen fertilizers. These measures can significantly decrease the rapid leaching of soil nitrogen pools, thereby mitigating short-term, large-scale pulses of NO_3^--N into groundwater. Besides, the confined groundwater, often affected by legacy pollution, requires long-term strategies focused on source control. This includes stricter regulation and monitoring of manure storage facilities, improved wastewater treatment infrastructure, and the implementation of containment systems to prevent leaching from historical contamination hotspots. Given the limited attenuation capacity and persistent nature of NO_3^--N in confined groundwater, remediation efforts may also need to consider engineered attenuation or pump-and-treat systems in severely affected zones. Future research should integrate reactive-transport modeling with isotopic mixing models to better capture the dynamic behavior of nitrogen in dual-layer aquifer systems and to further reduce uncertainty in source apportionment under varying hydrogeological conditions.

Referee #2:

This manuscript investigates the sources and quantification of nitrate in groundwater, with emphasis on the role of burial conditions. PCA-APCS-MLR and MixSIAR are combined to apportion nitrate origins in groundwater samples collected from different layers of aquifers. The results are interesting, highlighting innovative concepts and methodologies in source apportionment and groundwater pollution control strategies, which finally could merit publication.

Response:

Thank you very much for your thoughtful feedback on our manuscript. We sincerely appreciate your recognition of the novelty and significance of our work, as well as your positive assessment of the methodological approach and its potential contribution to the field of groundwater pollution control. We have carefully revised the manuscript accordingly to enhance its quality.

I have a number of comments:

1. The term "NO_3^--N" is used extensively in the introduction. While it is likely that readers familiar with groundwater contamination will understand this abbreviation, it would be helpful to define it explicitly at its first appearance in the text. For example, you could write "NO_3^--N (nitrate-N)" the first time it appears.

Response:

Thank you for pointing this out. We have revised the manuscript to explicitly define “NO_3^--N” as “nitrate-N” upon its first appearance in the text.

Page 1, Abstract, Line 20:

Nitrate-N (NO_3^--N) contamination in groundwater poses a significant threat to drinking water safety and ecosystem health, with accurate source identification being crucial for effective pollution control.

Page 4, Introduction, Line 67:

Groundwater nitrate-N (NO_3^--N) contamination has persisted for nearly a century worldwide, emerging as a critical environmental challenge that threatens both human health and ecological security.

2. The objectives of the study are listed clearly at the end of the introduction, but transition from the discussion of existing methods and challenges to the specific aims of your study is somewhat abrupt. I recommend adding a brief paragraph to explicitly link the identified gaps in current research to the specific goals of your study.

Response:

We agree that a smoother transition would improve the logical flow of the Introduction. As recommended, we have added a bridging paragraph before the statement of objectives to explicitly link the identified research gaps to the specific aims of our study.

Page 4, Introduction, Line 162:

To bridge the methodological gap associated with overlooking groundwater occurrence conditions in NO_3^--N source apportionment and to elucidate the genesis of "high-nitrogen groundwater" in the Old County groundwater source area, this study undertook an integrated field campaign and laboratory analysis. ~~The Old County groundwater source area is a vital water supply hub in the central region of Shandong Province. However, with the development of industry and agriculture and the increasing level of urbanization, the Old County source area is facing severe NO_3^--N contamination in groundwater. Identifying the sources of NO_3^--N in groundwater under different occurrence conditions in this region is crucial for elucidating the genesis of "high-nitrogen groundwater". In this study,~~Groundwater samples were collected from 64 wells, and soil, fertilizer, manure, and precipitation samples were also gathered within the study area. The water chemistry indicators and isotopic characteristics of these samples were analyzed. Subsequently, PCA-APCS-MLR and MixSIAR methods were employed for data analysis. The objectives of this study are (1) to quantify the concentration and distribution of NO_3^--N in groundwater within the study area; (2) to quantitatively identify the sources of NO_3^--N contamination in groundwater under different occurrence conditions using hydrochemical analysis and the MixSIAR method; and (3) to define the error in the analysis of groundwater NO_3^--N sources apportionment without considering occurrence conditions.

3. The introduction provides a comprehensive background on the problem of NO_3^--N contamination and the methods used for source apportionment. However, it lacks a clear statement of the hypothesis. Please clearly state the hypothesis in the introduction.

Response:

We appreciate your suggestion to strengthen the clarity and structure of the introduction. As recommended, we have revised the introduction to include a clear statement of the hypothesis.

Page 8, Introduction, Line 177:

The objectives of this study are (1) to quantify the concentration and distribution of NO_3^--N in groundwater within the study area; (2) to quantitatively identify the sources of NO_3^--N contamination in groundwater under different occurrence conditions using hydrochemical analysis and the MixSIAR method; and (3) to define the error in the analysis of groundwater NO_3^--N sources apportionment without considering occurrence conditions. We hypothesize that the primary sources of NO_3^--N pollution differ significantly between unconfined and confined aquifers, and neglecting groundwater occurrence conditions will introduce a discrepancy in the results of quantitative NO_3^--N source apportionment. The study aims to provide a more accurate basis for assessing the risk of NO_3^--N contamination in regional groundwater.

4. In the materials and methods section, the descriptions of sample testing methods are brief and there are no references provided to support these methods. Please provide detailed descriptions of the sample testing methods and cite relevant literature.

Response:

Thank you for your constructive comment. We have accordingly revised the manuscript to include more detailed methodological descriptions and relevant citations.

Page 10, Materials and methods, Line 239:

~~The concentration of NO_3^--N was determined using ultraviolet spectrophotometry. The concentrations of K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, and SO_4^{2-} were measured using an ion chromatograph (ICS-3000, Dionex, USA), the concentration of HCO_3^- was determined by acid-base titration.~~

The concentration of NO_3^--N was determined using the ultraviolet spectrophotometric method (at 220 nm and 275 nm) following filtration through a 0.45 μm membrane. The concentrations of major ions (K^+, Na^+, Ca^{2+}, Mg^{2+}, Cl^-, and SO_4^{2-}) were measured using an ion chromatograph (ICS-3000, Dionex, USA). The separation was achieved with an IonPac AS23 analytical column and an AG23 guard column, using a carbonate eluent. The concentration of HCO_3^- was determined by acid-base titration with a standardized HCl solution (0.02 M) to a bromocresol green-methyl red endpoint. All analyses of water quality indicators adhered to standard methods (Greenberg et al., 2005).

References

Greenberg, A.E., Trussell, R.R., Clesceri, L.S., Association, A.W.W., 2005. Standard methods for the examination of water and wastewater : supplement to the sixteenth edition. American Journal of Public Health & the Nations Health 56(3), 387.

5. The materials and methods section on data analysis briefly mentions the use of Pearson correlation tests but lacks details on the specific procedures used for these analyses.

Response:

Thank you for your thoughtful comment. We have revised the manuscript accordingly.

Page 13, Materials and methods, Line 317:

The stable isotope mixing model used in this study was run in the R package MixSIAR (R version x64 4.3.2). ~~The Pearson correlation test was employed to evaluate the relationships between hydrochemical indices, with data analysis conducted using SPSS 20.~~ Statistical analysis was performed using SPSS 20 software. To evaluate the linear relationships between hydrochemical parameters, the Pearson correlation coefficient (r) was calculated. Correlations were considered statistically significant at a two-tailed p-value < 0.05.

6. In the results section, the terms "generalized single-aquifer layer" and "actual double-aquifer layer" are used to describe different scenarios, but they are not clearly clarified. Define these terms explicitly at the beginning of the results section.

Response:

We agree that the terms "generalized single-aquifer layer" and "actual double-aquifer layer" should be explicitly defined to improve clarity. We have added a brief clarification at the beginning of the Results section.

Page 16, Results, Line 361:

The results of Pearson correlation analysis demonstrate that, in the generalized single-aquifer layer (refers to the simplified analytical scenario in which groundwater samples from both unconfined and confined aquifers are pooled and treated as a single homogeneous aquifer, without considering differences in occurrence conditions) ~~without consideration of groundwater occurrence conditions (hereinafter referred to as the generalized single-aquifer layer)~~ (Fig.4a), there is a strong correlation among the nine hydrochemical indicators.

Page 17, Results, Line 368:

In the actual double-layer aquifer (refers to the realistic scenario in which unconfined and confined aquifers are analyzed separately, respecting their distinct hydrogeological settings, recharge mechanisms, and pollution pathways)~~where groundwater occurrence conditions are taken into account (hereinafter referred to as the actual double-layer aquifer)~~ (Fig.4b and Fig.4c), the indicators also show strong correlations.

7. In the discussion section, please include a more detailed comparison with previous studies on NO_3^--N pollution and source apportionment. Discuss how your results align with or differ from those of other studies and provide possible explanations for these differences.

Response:

Thank you for your careful review . We fully agree with your suggestion. In response, we have significantly revised and expanded the Discussion section to enhance its scientific depth.

Page 24, Discussion, Line 501:

The PCA-APCS-MLR method has the advantages of being rapid and convenient, but it has the disadvantage of being unable to further identify soil nitrogen as a pollution source. Accurately apportioning soil-derived nitrate is particularly challenging for hydrochemical-based methods due to the overlapping ionic signatures between soil nitrogen and the leaching of synthetic fertilizers or organic wastes (Yu et al., 2022). To compensate for this limitation, the MixSIAR method was further employed to analyze the sources of pollution.

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Page 24, Discussion, Line 513:

Because NO_3^--N from different sources (atmospheric deposition, soil nitrogen, chemical fertilizers, and manure & sewage) has distinct isotopic signatures, and isotopic fractionation occurs during the transport and transformation processes of nitrogen in the groundwater system (such as ammonification and nitrification), leading to changes in the $\delta^{15}N$ and $\delta^{18}O$ values of NO_3^--N (Shu et al., 2024). Our MixSIAR model incorporated fractionation factors of specific nitrogen transformation processes. This approach, essential for reliable quantification, aligns with established practice in nitrate source apportionment studies, where such constraints are proven to substantially reduce uncertainty (Wang et al., 2023).

References

Wang, D., Li, P., Yang, N., Yang, C., Zhou, Y., Li, J., 2023. Distribution, sources and main controlling factors of nitrate in a typical intensive agricultural region, northwestern China: vertical profile perspectives. Environ. Res. 237, 116911.

Page 25, Discussion, Line 525:

The MixSIAR method further revealed that soil nitrogen is a dominant pollution source for unconfined groundwater, with a higher contribution rate than that of chemical fertilizers. The identification of soil nitrogen as a major contributor in the unconfined groundwater is significant. Legacy soil nitrogen constitutes a dominant source in the unconfined aquifer, a finding that shifts the pollution focus from direct fertilizer inputs to cumulative soil nitrogen pools. This finding is consistent with previous nitrate source studies conducted in other regions (Cui et al., 2023). For confined groundwater, MixSIAR also confirmed that manure & sewage are the major sources of NO_3^--N.

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Page 25, Discussion, Line 540:

While NH_4^+-N can be adsorbed and immobilized by the soil, NO_3^--N can leach into the deeper vadose zone and aquifer through atmospheric precipitation or agricultural irrigation, directly contaminating unconfined groundwater (Wan et al., 2024). This process underscores the phenomenon of the soil and vadose zone as a dynamic “nitrogen reactor and buffer”. Similar delayed release mechanisms from legacy nitrogen stored in thick unsaturated zones have been documented in arid regions (Li et al., 2025), indicating that the risk of groundwater contamination may persist long after surface inputs are reduced. Therefore, in assessing the sources of NO_3^--N pollution in regional groundwater, it is crucial not only to focus on the application rates of chemical fertilizers but also to pay attention to the storage of nitrogen in the soil and vadose zone.

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Page 26, Discussion, Line 562:

These pollutants can enter surface water bodies through surface runoff or infiltration and then gradually transport to deeper aquifers via the interflow recharge process between unconfined and confined aquifers, resulting in persistent contamination (McDonough et al., 2022). This may highlight a potential mechanism for sustained nitrate pollution in confined systems, which can be attributed to manure/sewage sources transported via aquifer exchange, providing a continuous input of recalcitrant nitrogen that gradually accumulates in this zone (Zhang et al., 2026). Therefore, for the prevention and control of NO_3^--N pollution in confined aquifers, it is crucial to focus on the source control of manure & sewage to block the migration pathways of pollutants and mitigate their long-term impacts on confined aquifers.

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Page 27, Discussion, Line 581:

First, the sources and recharge mechanisms of groundwater in unconfined and confined aquifers differ significantly (Liu et al., 2025), leading to distinct isotopic compositions and characteristic values. For example, the isotopic signature of a pollution source in an unconfined aquifer may resemble that of another source in a confined aquifer. Such overlapping isotopic signals can lead to ambiguous source identification. However, previous studies either do not explicitly distinguish between groundwater occurrence conditions or rely on land-use as a primary proxy for pollution source identification (Yu et al., 2020). When mixed calculations are performed without considering the actual occurrence conditions, the isotopic differences are obscured, resulting in confusion in pollution source identification, inaccurate contribution rate calculations, and incomplete analysis of pollution processes.

References

Liu, W., Du, Y., Qiu, W., Deng, Y., Wang, Y., 2025. Constraints on vertical variability of geogenic ammonium in multi-layered aquifer systems. Water Res. 268, 122639.

Yu, L., Zheng, T., Zheng, X., Hao, Y., Yuan, R., 2020. Nitrate source apportionment in groundwater using Bayesian isotope mixing model based on nitrogen isotope fractionation. Sci. Total Environ. 718, 137242.

Page 27, Discussion, Line 593:

Hydrogeological conditions can influence the intensity of biogeochemical processes such as ammonification, nitrification, denitrification (Dai et al., 2023), and adsorption

(Huang et al., 2022; Li et al., 2023), which further alter NO_3^--N concentrations and isotopic signatures.

References

Dai, H., Zhang, Y., Fang, W., Liu, J., Hong, J., Zou, C., Zhang, J., 2023. Microbial community structural response to variations in physicochemical features of different aquifers. Front. Microbiol. 14, 1025964.

Referee #3:

This study investigated nitrate sources in groundwater by collecting and analyzing samples from aquifers with different burial conditions. The authors employed an integrated approach combining PCA-APCS-MLR and MixSIAR for source identification and quantification. The results were compared between confined and unconfined aquifers, highlighting distinct dominant pollution sources for each setting, which enhanced the accuracy of pollution source identification and the effectiveness of protection strategies. It is an interesting work. The approach and results in this study seem acceptable, but some minor revisions are needed. My specific comments are as follows.

Response:

Thank you very much for taking time to review our manuscript. We sincerely appreciate your positive feedback and valuable comments, which have helped us improve the quality of our work. All revisions have been carefully considered and incorporated into the revised manuscript.

(1) Title: The term "burial conditions" is non-standard. Replace it with a precise hydrogeological expression such as "aquifer confinement conditions" or "aquifer occurrence conditions".

Response:

Thank you for your valuable and feedback. To enhance the clarity and precision of our manuscript, we have replaced "burial conditions" with the more precise hydrogeological term "occurrence conditions" throughout the text, as you recommended.

Page 1, Title:

Impact of ~~burial~~ occurrence conditions on NO_3^--N source apportionment in groundwater: Insights from PCA-APCS-MLR and MixSIAR methods

Please see L. 24, L. 25, L. 41, L. 58, L. 64, L. 87, L. 94, L. 108, L. 115, L. 117, L. 156, L. 161, etc. in the revised manuscript for other changes.

(2) L. 56: Add the WHO safe limit of 11.3 mg L^{-1} for NO_3^--N in drinking water for readers to see the exact threshold behind the health risk.

Response:

You rightly pointed out the need to explicitly state the WHO safe limit for NO_3^--N in drinking water, which provides readers with a clear health risk threshold. We have revised the manuscript accordingly.

Page 4, Introduction, Line 72:

~~As a highly toxic pollutant, NO_3^--N poses significant health risks including methemoglobinemia and cancer when ingested through drinking water (Picetti et al., 2022), while also causing severe ecological impacts such as aquatic eutrophication (Romanelli et al., 2020).~~

As a highly toxic pollutant, NO_3^--N poses significant health risks including methemoglobinemia and cancer through drinking water (Picetti et al., 2022), particularly when its concentration exceeds the WHO drinking water standard of 11.3 mg N L^{-1}. It also causes severe ecological impacts such as aquatic eutrophication, primarily through groundwater discharge into rivers, lakes, and coastal waters (Romanelli et al., 2020).

(3) L. 58: Clarify how groundwater NO_3^- reaches surface water; without this pathway the link to aquatic eutrophication appears as a logical leap.

Response:

We appreciate this valuable suggestion. NO_3^--N can migrate from aquifers to surface waters through several key pathways, which vary depending on aquifer type and local hydrogeology. In unconfined aquifers, NO_3^--N-enriched groundwater often discharges directly into streams, lakes, or coastal zones via baseflow, especially in gaining stream systems. In confined aquifers, NO_3^--N may reach surface water through upward leakage where aquitards are discontinuous, or via pumping and irrigation return flows. In response to your comment, we have revised the relevant section in the manuscript.

Page 4, Introduction, Line 74:

~~As a highly toxic pollutant, NO_3^--N poses significant health risks including methemoglobinemia and cancer when ingested through drinking water (Picetti et al., 2022), while also causing severe ecological impacts such as aquatic eutrophication (Romanelli et al., 2020).~~

As a highly toxic pollutant, NO_3^--N poses significant health risks including methemoglobinemia and cancer through drinking water (Picetti et al., 2022), particularly when its concentration exceeds the WHO drinking water standard of 11.3 mg L^{-1}. It also causes severe ecological impacts such as aquatic eutrophication, primarily through groundwater discharge into rivers, lakes, and coastal waters (Romanelli et al., 2020).

(4) L. 61: NO_3^--N instead of this contaminant.

Response:

Thank you for your careful reading and precise suggestion. In response to your comment, we have revised the sentence in the manuscript.

Page 4, Introduction, Line 80:

The environmental persistence of NO_3^--N is exacerbated by limited natural attenuation in groundwater systems due to weak denitrification processes, resulting in long-term accumulation of NO_3^--N ~~this contaminant~~ (Rivett et al., 2008).

(5) L. 77: Add citations.

Response:

Thank you for your comment. We have added the relevant citation(s) to L. 77. The revision has been made in the manuscript accordingly.

Page 5, Introduction, Line 98:

While this simplification facilitates analysis, it introduces substantial limitations due to fundamental differences between unconfined and confined aquifers in terms of recharge mechanisms, flow paths, hydraulic characteristics, and contaminant transport behavior (Liang et al., 2017).

References

Liang, X., Zhan, H., Zhang, Y.K., Schilling, K., 2017. Base flow recession from unsaturated-saturated porous media considering lateral unsaturated discharge and aquifer compressibility. Water Resour. Res. 53(9), 7832-7852.

(6) L.93-94: Delete.

Response:

We have removed the content in the revised manuscript as recommended.

Page 5, Introduction, Line 114:

~~Therefore, elucidating the sources of NO_3^--N pollution in groundwater with different occurrence conditions and revealing the discrepancies between these results and those obtained without considering occurrence conditions can provide a more accurate basis for groundwater NO_3^--N pollution risk assessment.~~

(7) L.132: Explain why both hydrochemical tracers and MixSIAR are needed together instead of using just one of them.

Response:

Thank you for raising this important point. The combined use of hydrochemical tracers (e.g., PCA-APCS-MLR) and the stable isotope mixing model (MixSIAR) is essential for NO_3^--N source apportionment, as each approach has unique strengths and limitations that, when integrated, provide a more accurate and reliable assessment than either method alone.

1) Complementary strengths and limitations

Hydrochemical tracers (PCA-APCS-MLR) are effective in identifying pollution sources based on ion correlations and concentration patterns. They can quantify contributions from sources with distinct chemical fingerprints (e.g., fertilizers, sewage) but may struggle to distinguish between sources with similar chemical profiles (e.g., soil nitrogen and atmospheric deposition) and cannot directly account for isotopic fractionation processes. MixSIAR, based on $\delta^{15}N$ and $\delta^{18}O$ isotopes, excels at differentiating between sources with overlapping chemical signals but distinct isotopic signatures (e.g., manure vs. soil nitrogen).

2) Enhanced accuracy and reliability

By using both methods, we cross-validate the results. For instance, PCA-APCS-MLR identified chemical fertilizers as a major source in the unconfined aquifer, while MixSIAR highlighted soil nitrogen as the dominant contributor. This discrepancy prompted a deeper investigation, revealing that soil nitrogen — a legacy source with a chemical signature similar to fertilizers — was being overlooked by the chemical method alone. The integration thus provided a more nuanced understanding: soil nitrogen is a critical, persistent source that must be managed alongside current fertilizer inputs.

In summary, the integration of hydrochemical and isotopic methods provides a more comprehensive and validated apportionment of NO_3^--N sources in groundwater, enhancing the scientific rigor and practical relevance of our findings.

(8) L. 144: Add research hypotheses.

Response:

Thank you for your constructive suggestion. We have added research hypotheses to the revised manuscript in the Introduction section.

Page 8, Introduction, Line 177:

The objectives of this study are (1) to quantify the concentration and distribution of NO_3^--N in groundwater within the study area; (2) to quantitatively identify the sources of NO_3^--N contamination in groundwater under different occurrence conditions using hydrochemical analysis and the MixSIAR method; and (3) to define the error in the analysis of groundwater NO_3^--N sources apportionment without considering occurrence conditions. We hypothesize that the primary sources of NO_3^--N pollution differ significantly between unconfined and confined aquifers, and neglecting groundwater occurrence conditions will introduce a discrepancy in the results of quantitative NO_3^--N source apportionment. The study aims to provide a more accurate basis for assessing the risk of NO_3^--N contamination in regional groundwater.

(9) L.228, L.257: Add citations.

Response:

We have added relevant citations in the revised manuscript accordingly.

Page 12, Materials and methods, Line 282:

Subsequently, on the basis of PCA, the absolute principal component scores (APCS) were determined. A multiple linear regression (MLR) was performed with the measured pollutant concentrations as the dependent variables and the absolute principal component scores as the independent variables (Thurston and Spengler, 1985).

References

Thurston, G.D., Spengler, J.D., 1985. A quantitative assessment of source contributions to inhalable particulate matter pollution in metropolitan Boston. Atmospheric Environment (1967) 19(1), 9-25.

Page 13, Materials and methods, Line 303:

The principle of the MixSIAR method is to use the Dirichlet distribution as the prior distribution and to obtain the posterior distribution characteristics of the contributions, such as the mean, variance, and probability density, through the application of Bayes' theorem (Moore and Semmens, 2008).

References

Moore, J.W., Semmens, B.X., 2008. Incorporating uncertainty and prior information into stable isotope mixing models. Ecol. Lett. 11(5), 470-480.

(10) L.275: Add detailed information of the Pearson test.

Response:

We acknowledge that the detailed information of the Pearson test was insufficient. In the revised manuscript, we have added detailed procedural steps for the statistical correlation analysis. Furthermore, we have included appropriate citations to support the chosen methodologies.

Page 13, Materials and methods, Line 317:

The stable isotope mixing model used in this study was run in the R package MixSIAR (R version x64 4.3.2). ~~The Pearson correlation test was employed to evaluate the relationships between hydrochemical indices, with data analysis conducted using SPSS~~

~~20.~~ Statistical analysis was performed using SPSS 20 software. To evaluate the linear relationships between hydrochemical parameters, the Pearson correlation coefficient (r) was calculated. Correlations were considered statistically significant at a two-tailed p-value < 0.05.

(11) L. 313: “Generalized single-aquifer layer” and “actual double-aquifer layer” are used without definition. Clarify at the beginning of the section.

Response:

We apologize for this oversight. We have added a brief clarification at the beginning of the Results section.

Page 16, Results, Line 361:

The results of Pearson correlation analysis demonstrate that, in the generalized single-aquifer layer (refers to the simplified analytical scenario in which groundwater samples from both unconfined and confined aquifers are pooled and treated as a single homogeneous aquifer, without considering differences in occurrence conditions) ~~without consideration of groundwater occurrence conditions (hereinafter referred to as the generalized single-aquifer layer)~~ (Fig.4a), there is a strong correlation among the nine hydrochemical indicators.

Page 17, Results, Line 368:

In the actual double-layer aquifer (refers to the realistic scenario in which unconfined and confined aquifers are analyzed separately, respecting their distinct hydrogeological settings, recharge mechanisms, and pollution pathways)~~where groundwater occurrence conditions are taken into account (hereinafter referred to as the actual double-layer aquifer)~~ (Fig.4b and Fig.4c), the indicators also show strong correlations.

(12) L.405: How did the authors derive their fractionation data? What consequences would disregarding fractionation have for the MixSIAR results?

Response:

We appreciate the opportunity to clarify this key methodological aspect.

1) Determination of fractionation data

●**Ammonification:** The isotopic effect during ammonification is generally very small (< 2‰) and difficult to quantify precisely in complex environmental matrices. Given its minor magnitude relative to other processes, and consistent with approaches in similar studies, the fractionation associated with ammonification was neglected in our calculations.

●**Nitrification:** Nitrification is a major process affecting the isotopic signature of nitrate derived from ammonium-based sources (e.g., fertilizer, mineralized soil organic nitrogen). The fractionation factor for this process is known to vary widely (-35‰ to -5‰) depending on soil type and environmental conditions. Following common practice to obtain a representative value, we used the average of the range: -20.0 ± 2.7‰.

●**Ammonia Volatilization:** Manure and sewage are characterized by high ammonium content, making them susceptible to ammonia volatilization. Based on the previous study (which investigated volatilization in open systems relevant to manure management), the fractionation coefficient range of ammonia volatilization is 25-45‰. We adopted a fractionation factor of 24.1 ± 2.8‰ for this process.

To determine the nitrogen isotope fractionation factors of different pollution sources, it is necessary to consider the nitrogen transformation processes and their isotope fractionation effects. For atmospheric deposition, this study directly tests the nitrogen and oxygen isotope characteristic values of nitrate in precipitation. Therefore, there is no need to consider the isotope fractionation effect of atmospheric deposition pollution sources. For soil nitrogen and chemical fertilizer pollution sources, the fractionation effect of nitrification needs to be considered. Therefore, the fractionation factors for soil nitrogen and chemical fertilizer sources are both -20.0 ± 2.7‰. For manure sources, both isotope fractionation during ammonia volatilization and nitrification processes need to be considered. Therefore, the calculated isotope fractionation factor for manure sources is 16.8 ± 2.8‰.

2) Consequences of disregarding fractionation in MixSIAR results

Disregarding isotopic fractionation would critically compromise the validity of MixSIAR results. Without accounting for process-driven isotopic shifts, the model would misinterpret measured groundwater $\delta^{15}N$ and $\delta^{18}O$ values, leading to systematic bias in source attribution. For example, nitrate derived from soil or fertilizer that has undergone nitrification (~20‰ depletion in $\delta^{15}N$) could be incorrectly assigned to atmospheric deposition or other light-isotope sources, thereby underestimating agricultural contributions. Similarly, neglecting the enrichment effect of ammonia volatilization in manure systems would cause the model to undervalue the role of manure and sewage. Ultimately, omitting fractionation would treat nitrate transport as a conservative mixing process, ignoring the transformative biogeochemical history of nitrogen in the aquifer. This would not only reduce the quantitative accuracy of source apportionment but also obscure key nitrogen cycling pathways, weakening the reliability of conclusions and the effectiveness of any resulting management recommendations.

(13) L. 470: Add citations.

Response:

Thank you for your comment. We have added the relevant citation(s) in the revised manuscript accordingly.

Page 25, Discussion, Line 534:

Nitrogen from these fertilizers can directly leach into the unconfined aquifer, causing NO_3^--N pollution (Xie et al., 2025).

References

Xie, L., Li, P., Fida, M., Elumalai, V., 2025. Characteristics and potential health risk of inorganic nitrogen in phreatic water in the central and southern parts of Yinchuan Plain (Northwest China). Expo. Health 17(2), 581-598.

(14) L. 498-499: Without a statistical analysis, reporting absolute errors is inappropriate. Replace these percentages with a qualitative statement.

Response:

We appreciate the reviewer’s suggestion. Accordingly, we have removed the quantitative error percentages and replaced them with a qualitative statement regarding the discrepancy in source apportionment results when occurrence conditions are not considered.

Page 2, Abstract, Line 39:

These findings suggest that unconfined groundwater in regions with high soil nitrogen reserves is at persistent risk of NO_3^--N contamination. ~~In addition, neglecting groundwater occurrence conditions would introduce absolute errors of 22%-24% in source apportionment results obtained from both PCA-APCS-MLR and MixSIAR approaches.~~ This study highlights that aquifer confinement must be rigorously considered as a critical factor in NO_3^--N source identification and pollution control strategies to enhance the accuracy of source apportionment and the effectiveness of management measures.

These findings suggest that unconfined groundwater in regions with high soil nitrogen reserves is at persistent risk of NO_3^--N contamination. In addition, ignoring the groundwater occurrence conditions leads to marked deviations in the source apportionment results derived from both the PCA-APCS-MLR and MixSIAR approaches. This study highlights that aquifer confinement must be rigorously considered as a critical factor in NO_3^--N source identification and pollution control strategies to enhance the accuracy of source apportionment and the effectiveness of management measures.

Page 27, Discussion, Line 575:

~~This study compared the errors in source apportionment of NO_3^--N in groundwater with and without consideration of occurrence conditions. The absolute errors for the PCA-APCS-MLR method were 4%-20% and 5%-24%, while those for the MixSIAR method were 1.1%-8.5% and 1.5%-22%. The causes of these errors can be attributed to two main factors.~~ First, the sources and recharge mechanisms of groundwater in unconfined and confined aquifers differ significantly, leading to distinct isotopic compositions and characteristic values.

This study assessed the discrepancy in source apportionment of NO_3^--N in groundwater obtained under different groundwater occurrence conditions. This discrepancy can be attributed to two main factors. First, the sources and recharge mechanisms of groundwater in unconfined and confined aquifers differ significantly, leading to distinct isotopic compositions and characteristic values.

(15) To translate research findings into actionable groundwater protection strategies, the authors should explicitly outline how their results can guide targeted management practices.

Response:

Thank you for your thoughtful comment. We agree that translating scientific findings into actionable management guidance is crucial. In response to your suggestion, we have revised the Discussion and Conclusion sections to more explicitly outline the management implications derived from our findings.

Page 27, Discussion, Line 596:

Hydrogeological conditions can influence the intensity of biogeochemical processes such as ammonification, nitrification, denitrification, and adsorption (Huang et al., 2022; Li et al., 2023), which further alter NO_3^--N concentrations and isotopic signatures. This ultimately affects the accuracy and reliability of pollution source apportionment. Consequently, effective nitrate management in groundwater systems requires policy interventions tailored to specific aquifer burial conditions. In unconfined aquifers, which are highly vulnerable to surface-derived inputs, management should prioritize agricultural best practices such as optimized irrigation scheduling — reducing both frequency and volume of irrigation — coupled with the promotion of slow-release or stabilized nitrogen fertilizers. These measures can significantly decrease the rapid leaching of soil nitrogen pools, thereby mitigating short-term nitrate spikes. Additionally, land-use zoning that limits intense agricultural or livestock activities near recharge areas could further reduce nitrate loading. In contrast, confined aquifers, often affected by legacy pollution, require long-term strategies focused on source control. This includes stricter regulation and monitoring of manure storage facilities, improved wastewater treatment infrastructure, and the implementation of containment systems to prevent leaching from historical contamination hotspots. Given the slow migration and persistent nature of nitrate in confined settings, remediation efforts may also need to consider engineered attenuation or pump-and-treat systems in severely affected zones. Future research should integrate reactive-transport modeling with isotopic mixing models to better capture the dynamic behavior of nitrogen in dual-layer aquifer systems and to further reduce uncertainty in source apportionment under varying hydrogeological conditions.

Page 29, Conclusion, Line 652:

The study quantitatively analyzed the pollution sources of NO_3^--N in groundwater under different occurrence conditions using PCA-APCS-MLR and MixSIAR methods. The results showed that the groundwater NO_3^--N concentration in the study area ranged from 0 to 68 mg N L^{-1}, with an exceedance rate of 75%. The NO_3^--N pollution in unconfined groundwater (average concentration 29.9 mg N L^{-1}) was more severe than that in confined groundwater (average concentration 20.1 mg N L^{-1}). NO_3^--N in unconfined groundwater predominantly originates from soil nitrogen (58%), with a non-negligible contribution from chemical fertilizers. Therefore, it is necessary to focus on the storage of nitrogen in the soil and improve agricultural irrigation practices to prevent rapid infiltration of NO_3^--N into unconfined groundwater, which could lead to persistent contamination. NO_3^--N enrichment in confined groundwater is primarily attributed to manure & sewage (37.9%). Regulations and infrastructure for the treatment and disposal of domestic sewage and livestock waste should be strengthened to prevent their extensive accumulation in confined groundwater. Ignoring the groundwater occurrence conditions leads to marked deviations in the source apportionment results, pollution source identification and control policies for groundwater must explicitly distinguish between unconfined and confined groundwater.

Additional Revisions：

In addition to the specific revisions requested by the reviewers, I have also taken this opportunity to carefully proofread the entire manuscript. Following your instructions, I have corrected typographical errors, standardized terminology, verified all author affiliations, and refined figures and textual expressions. All these general improvements have been integrated into the revised manuscript. The specific revisions are outlined below.

1. The author affiliations have been updated.

Page 1, Line 9:

3. Key Laboratory of Marine Environment and Ecology, Ministry of Education, Ocean University of China, Qingdao 266100, China

2. We have inserted the specific numerical data into this sentence.

Page 2, Abstract, Line 29:

The results demonstrate that NO_3^--N concentrations in 75% of the groundwater samples exceeded the standard for drinking water quality of China ($\leq$ 10 mg N L^{-1}).

3. The expression in this section has been refined.

Page 2, Abstract, Line 30:

NO_3^--N in unconfined groundwater predominantly originates from soil nitrogen (58%), with a non-negligible contribution from chemical fertilizers. NO_3^--N enrichment in confined groundwater is primarily attributed to manure & sewage (37.9%).

4. The wording of this sentence has been improved.

Page 2, Abstract, Line 44:

This study highlights that considering the occurrence conditions serves as a key indicator for distinguishing the primary sources of NO_3^--N in groundwater, which can enhance the accuracy of source apportionment and the effectiveness of management measures.

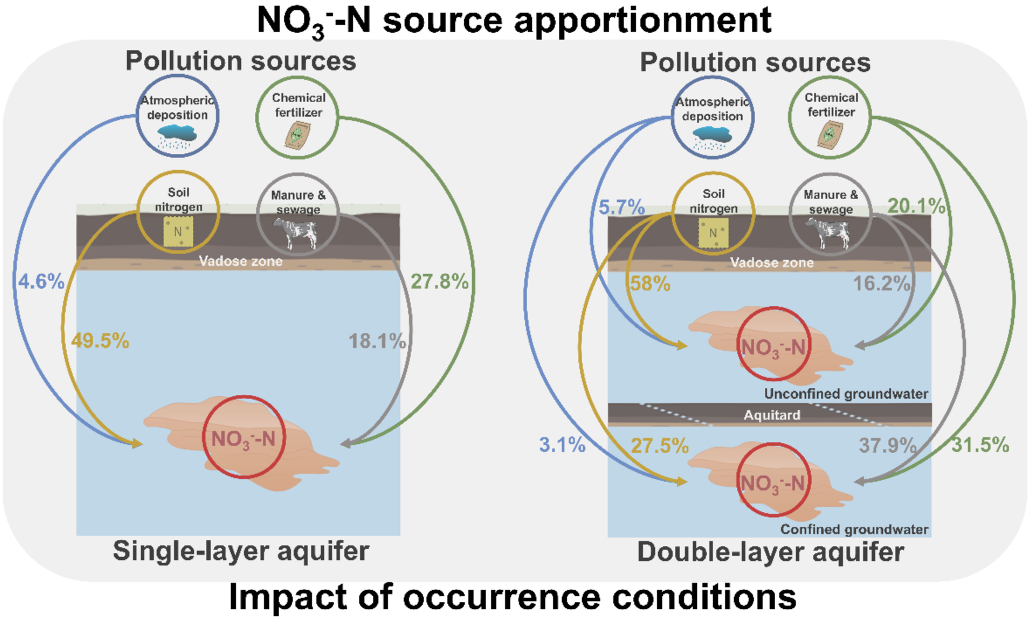
5. The keywords have been modified.

Page 2, Keywords, Line 49:

Keywords: Groundwater; occurrence conditions; NO_3^--N source apportionment; PCA-APCS-MLR; MixSIAR

6. We have updated the graphical abstract.

Page 3, Graphical Abstract:



7. The content of Highlights has been modified.

Page 4, Highlights, Line 62:

- Neglecting occurrence conditions leads to significant deviations in source apportionment.

8. Explanatory content on the prior information has been added.

Page 7, Materials and methods, Line 147:

The MixSIAR method, by integrating isotope data with prior information (the ranges of isotopic values and initial estimates of their contributions) on pollution sources, is capable of quantifying the relative contributions of different pollution sources and assessing the uncertainty of the results.

9. Fig. 2 has been updated.

Page 14, Results, Line 335:

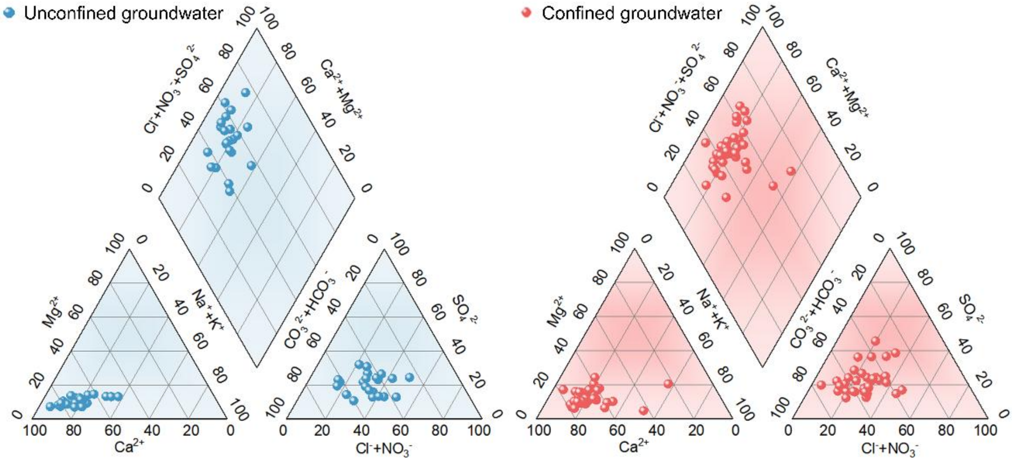


Fig.2. Piper graph illustrating hydrochemical types of groundwater.

10. The sentence has been rephrased.

Page 28, Conclusion, Line 621:

The study quantitatively analyzed the pollution sources of NO_3^--N in groundwater under different occurrence conditions using PCA-APCS-MLR and MixSIAR methods.

11. The sentence has been rewritten for clarity.

Page 29, Conclusion, Line 629:

NO_3^--N in unconfined groundwater predominantly originates from soil nitrogen (58%), with a non-negligible contribution from chemical fertilizers. NO_3^--N enrichment in confined groundwater is primarily attributed to manure & sewage (37.9%).

12. The Acknowledgments section has been updated.

Page 30, Acknowledgments, Line 659:

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China (No. 2024YFC3213900), the National Natural Science Foundation of China (No. 42422207 and No. 42507105), and the Postdoctoral Research Project (No. QDBSH20240202055).