**Responses to Reviewer:**

[Authors' response]

We would like to sincerely thank the reviewer for his/her supporting and for taking the time to review our manuscript. Your good suggestions have increased our papers quality. Thank you very much!

This manuscript presents a monthly analysis of TN and NH3-N concentrations in 3020 lakes across China and their spatial and temporal relationship to select influential factors over a 15-year period (2010-2024). By applying an ensemble machine learning framework, the authors assessed the relative strength of different factors in predicting the spatial and temporal patterns in TN and $NH_3$-N concentrations. The main findings are that, depending on the limnological region of the country, external nitrogen loading, hydro-climatic dynamics, soil N & land cover, and topography structure each appeared as dominant factors in predicting the spatial variability of TN, while only external nitrogen loading and soil N & land cover appeared as dominant factors in predicting the spatial variability of $NH_3$-N. A similar distribution of dominant factors was found with respect to trends in TN over the 15-year study period, while $NH_3$-N trends were related predominantly to external N loading. Differences between the relationships with factors were attributed to the influence of legacy nitrogen stored in the system on TN concentrations and the higher reactivity of $NH_3$-N concentrations in response to continued external N loading. The findings led the authors to recommend nitrogen-form-specific management strategies to account for the differing influences.

In reviewing this manuscript, I was not able to assess the ensemble machine learning framework applied, as it is outside my expertise. My review therefore focuses primarily on data used, the interpretation of the results and the recommendations.

Overall, I found the manuscript to present a thorough and comprehensive analysis of a remarkable dataset. The results are significant and of value, at least with respect to Total N, in addressing problems of N contamination across China and more broadly. It is also well written.

[Authors' response]

We sincerely thank the reviewer for the positive assessment of our study and for the constructive comments. We also greatly appreciate the reviewer's important concerns regarding the suitability of $NH_3$-N as a response variable in national- and catchment-scale analyses, the appropriate scope of interpretation of the results, and the use of causal language. In response to these concerns, we have systematically revised the manuscript and provide detailed point-by-point responses below.

My primary concern centres around the use of NH3-N as a response variable for such a national and catchment scale analysis and the influence this has on the final conclusions. The analysis infers cause and effect relationships between the predictor variables (e.g. external N loading) and the response variables (TN and $NH_3$-N concentrations). This leads to the following statement in the Conclusions section "Based on these findings, we postulate that both the spatial distribution and long-term trend of lake TN across a large geographical region are reflected by watershed-scale cumulation of nitrogen, and that the long-term trend of lake $NH_3$-N is primarily affected by external nitrogen loading". From this the authors go on to state "The different responses of lake nitrogen forms in spatial and temporal characteristic across China highlight the need for nitrogen-form-specific management strategies."

While I judge the analysis and eventual conclusions related to TN concentrations to be reasonable, I do not feel the same about those for $NH_3$-N. Ammonia ($NH_3$) is a highly reactive compound of nitrogen in natural waters generated during the ammonification of organic nitrogen and generally quickly converted to ammonium ($NH_4$+), oxidised to nitrate ($NO_3$-), or taken up and assimilated by plants. The predictor variables considered in this study are quite distal from the conditions in the lakes themselves. $NH_3$-N concentrations could be much better predicted using TN concentrations in the lake, pH and dissolved O2 concentrations. Nitrate would have been the better dissolved inorganic form of N to use for the analysis and may have even resulted in similar conclusions, but I understand that $NH_3$-N was the only option available in the CNEMC dataset. CNEMC monitoring of $NH_3$-N is understandable because it is a priority pollutant and serves as a rapid warning indicator of failures in water treatment systems or flushing events from nearby contamination sources (like feedlots). It is valuable for detecting local effects, but it is not a good indicator of catchment scale characteristics.

The poor utility of $NH_3$-N concentrations as an indicator of external N loading is evident in the concentrations reported in the manuscript. The observed mean concentrations reported in lines 336-339 (maximum 0.159 mg/L) are low and acceptable for water quality Classes I and II in China. By comparison the national multi-year mean TN concentration of 2.07 mg/L in the lakes is slightly above what is acceptable even for water quality Class V. So, comparatively, TN concentrations reflect highly enriched conditions while $NH_3$-N concentrations do not. In short, mean regional

TN concentrations in the lakes are a water quality problem, while mean $NH_3$-N concentrations are not.

TN concentrations may very well be influenced by external N loading in the catchment, reflecting as the authors state "the widespread inertia inherent in watershed- and lake-scale nitrogen pollution and recovery processes at the global scale". But $NH_3$-N concentrations are likely only influenced by the TN concentrations in the lake, which when combined with pH and dissolved oxygen conditions in the lake are not so high as to result in widespread $NH_3$-N concentrations exceeding national water quality standards.

I believe these considerations may be so significant as to remove $NH_3$-N from the analysis. If the authors choose to keep $NH_3$-N in the analysis, I recommend that the considerations raised above be reflected in the revision of the manuscript. TN and $NH_3$-N should not be assumed to respond in equal strength to the variations in the predictor variables distal to the river. The limitations of using $NH_3$-N for such a study should be acknowledged. Results and conclusions related to $NH_3$-N should also therefore be qualified.

[Authors' response]

We thank the reviewer for raising this important concern regarding the suitability of $NH_3$-N for national- and catchment-scale analyses. We carefully re-evaluated the scientific role of $NH_3$-N and the associated conclusions in the original manuscript, and we agree with the reviewer's central point that TN and $NH_3$-N have distinct biogeochemical characteristics and therefore should not be assumed to have the same responses to, or environmental significance with respect to, catchment-scale environmental predictors. TN integrates multiple nitrogen forms and can, to some extent, reflect nitrogen accumulation and legacy effects, making it more suitable for characterizing long-term nitrogen accumulation and enrichment at the catchment scale. In contrast, $NH_3$-N is a more reactive and rapidly transformed nitrogen form whose concentration is associated not only with external nitrogen inputs but also with in-lake processes such as ammonification, nitrification, and biological uptake, as well as local physicochemical conditions including pH, dissolved oxygen, and temperature. We therefore agree that statistical relationships between catchment-scale environmental variables and $NH_3$-N alone are insufficient to infer direct control of lake $NH_3$-N concentrations by external nitrogen loading, and that $NH_3$-N should not be regarded as a direct proxy for catchment-scale nitrogen loading.

After carefully considering the reviewer's suggestion to remove $NH_3$-N from the analysis, we decided to retain it but have substantially revised its scientific role and scope of interpretation throughout the manuscript. Our purpose in retaining $NH_3$-N is not to treat it as equivalent to TN as an indicator of catchment-scale nitrogen pollution, but rather to use it as a complementary reactive nitrogen form with distinct biogeochemical characteristics, allowing comparison with TN in terms of spatial patterns, long-term trends, and predictive relationships with environmental variables. Accordingly, the revised manuscript no longer emphasizes a "rapid and direct response" of $NH_3$-N to external nitrogen loading. Instead, relationships between $NH_3$-N and catchment-scale environmental variables are explicitly interpreted as statistical/predictive associations rather than direct causal effects.

Based on this revised framing, we systematically revised the statements and conclusions related to $NH_3$-N throughout the manuscript. First, we removed or revised causal wording such as "governed by," "driven by," and "affected by," and now consistently describe the relationships between external nitrogen loading and $NH_3$-N as statistical/predictive associations. Second, we explicitly acknowledge the scale-related limitations of $NH_3$-N in the Discussion, noting that its concentration is strongly modulated by rapid in-lake nitrogen transformations and local physicochemical conditions. Consequently, the observed long-term changes in $NH_3$-N cannot be interpreted simply as a direct response to changes in catchment-scale external nitrogen loading (Lines 538–544). Third, the revised Abstract now notes that the comparatively lower predictive performance for $NH_3$-N suggests that catchment-scale environmental predictors alone are insufficient to fully capture its variability, thereby limiting its applicability as a direct indicator of catchment-scale nitrogen loading (Lines 35–41). In the Conclusions, we similarly revised the original statement that "long-term $NH_3$-N trends were primarily affected by external nitrogen loading" to indicate instead that external nitrogen loading variables showed strong statistical associations with long-term $NH_3$-N trends, while again emphasizing the scale-related limitations arising from the sensitivity of $NH_3$-N to in-lake processes (Lines 648–655).

In addition, following the reviewer's comment, we reconsidered the environmental significance of the observed $NH_3$-N concentrations and accordingly moderated the associated pollution and management implications. The revised manuscript no longer interprets elevated $NH_3$-N concentrations as indicating the same degree of catchment-scale nitrogen pollution as TN, nor do we use these results to directly support catchment-scale source-control recommendations specifically for $NH_3$-N. Instead, we now adopt distinct interpretive frameworks for the two nitrogen forms: TN is treated as a more integrative indicator of long-term nitrogen accumulation and enrichment at the catchment scale, whereas $NH_3$-N is treated as a complementary, more reactive nitrogen form that is more sensitive to local in-lake physicochemical and biogeochemical processes. Its relationships with catchment-scale environmental variables are therefore interpreted primarily as broad-scale statistical/predictive associations.

We believe that this revised framing directly addresses the reviewer's concerns regarding the catchment-scale representativeness and environmental interpretation of $NH_3$-N, while preserving the scientific value of comparing the spatiotemporal patterns and environmental predictive relationships of different nitrogen forms.

Other points for consideration

Throughout the manuscript the authors describe TN of NH3-N spatial patterns or trends to be "attributed to", "governed by", "affected by", "influenced by", etc., the variables representing drivers. For example, lines 461-462 say "the long-term changes in $NH_3$-N concentrations were primarily governed by external nitrogen loading". This leads to the clear inference of the direct cause-and-effect relationships I referred to above. But the associations between N concentrations and variables used in the analysis are not causal relationships… they are predictive statistical relationships. This language should be revised throughout the manuscript, replacing words that imply causality with correct representations such as "associated with", "statistically related to", "predictive of", etc. This change in language will partially, but not fully, address the concerns raised above about the use of $NH_3$-N.

[Authors' response]

Thank you for pointing out this important issue. We fully agree that the ensemble machine-learning and SHAP analyses used in this study identify statistical associations between environmental predictors and TN and $NH_3$-N concentrations and their long-term trends, as well as the relative contributions of these predictors to model predictions, rather than establishing direct causal relationships. We acknowledge that the use of terms such as "attributed to," "governed by," "affected by," "influenced by," and "driven by" in the original manuscript could imply stronger causal interpretations than warranted by our analyses.

To address this concern, we systematically reviewed and revised the relevant language throughout the manuscript. Terms that could imply causality were replaced with expressions more consistent with the statistical nature of our analyses, such as "associated with," "showed statistical associations with," "showed stronger predictive contributions," and "was positively/negatively associated with." For example, the original statement regarding long-term changes in $NH_3$-N: "the long-term changes in $NH_3$-N concentrations were primarily governed by external nitrogen loading" has been revised to: "external nitrogen loading variables showed the strongest statistical associations with long-term $NH_3$-N trends across most sub-basins." (Lines 648–650)

Accordingly, the relevant Results section has also been revised to state, for example: "Long-term TN trends showed spatially varying statistical associations with external nitrogen loading and hydro-climatic variables, whereas long-term $NH_3$-N trends showed a more spatially consistent association with external nitrogen loading variables." (Lines 470 – 473)

In addition, we revised the description of SHAP analysis in the Methods. SHAP is no longer presented as a tool for "mechanistic attribution" or "identification of driving mechanisms," but rather as an approach for quantifying the relative predictive contributions of environmental variables to model outputs. In the Discussion, we similarly avoid inferring causal mechanisms directly from SHAP results and instead use more cautious expressions such as "consistent with," "may reflect," and "may be related to" when discussing plausible ecological processes. For $NH_3$-N specifically, its relationship with external nitrogen loading is now explicitly interpreted as a statistical/predictive association rather than a direct causal effect.

Through these revisions, we have more clearly distinguished model-derived predictive relationships from potential ecological causal mechanisms and ensured that the interpretation of our findings is consistent with the statistical nature of the analytical framework.

Lines 66-67: The text states “Previous studies have documented the differential responses of nitrogen forms to climatic, hydrological, and land-use changes at lake or watershed scales”. Careful in claiming that studies have already "documented" the responses of different N forms to climate change. Your citations here do not support that. Du et al. (2023) investigated organic matter with no analysis of N forms, and Jiang et al. (2026) report modeling results of future conditions. If you want to keep the statement, your references should document studies of N responses to climate change that have already occurred.

[Authors’ response]

Thank you for your careful review. To address the concern that the previous references did not adequately support our statement regarding historically observed responses of specific nitrogen forms, we have replaced them with three empirical studies based on long-term observational datasets that more directly examine how different nitrogen forms or nitrogen loads vary in relation to climatic, hydrological, and land-use factors.

Study 1: Using historical monitoring data from sub-watersheds of the Upper Mississippi River Basin during 2001–2020, Zhao et al. (2025) documented changes in oxidized nitrogen ($NO_3^-$ + $NO_2^-$-N) yields associated with hydrological variability and agricultural activities.

Study 2: He et al. (2026) explicitly examined the contrasting spatiotemporal dynamics of $NH_3$-N and TN in China's surface waters, including their seasonal coupling/decoupling and associations with thermal and hydrological conditions.

Study 3: Using several decades of observational records from the United States, Zhao et al. (2023) quantified the responses of riverine nitrogen loading to climatic variability, particularly changes in total and extreme precipitation and temperature.

To more accurately reflect the evidence provided by these studies, we have revised the corresponding sentence in the Introduction as follows:

"Previous observational studies have documented differential patterns and responses of aquatic nitrogen forms and loads in relation to climatic, hydrological, and land-use drivers at lake and watershed scales." (Lines 69-73)

He, Y., Nong, X., Chen, L., Wei, J., 2026. National-scale nitrogen stoichiometry in China's surface waters: spatiotemporal dynamics, proxy utility, and threshold-driven controls. npj Clean Water 9, 49. https://doi.org/10.1038/s41545-026-00592-5

Zhao, G., Merder, J., Ballard, T.C., Michalak, A.M., 2023. Warming may offset impact of precipitation changes on riverine nitrogen loading. Proc. Natl. Acad. Sci. 120, e2220616120. https://doi.org/10.1073/pnas.2220616120

Zhao, Q., Peng, B., Ma, Z., Jia, M., McIsaac, G.F., Robertson, D.M., Saad, D.A., Warner, R.E., Wu, X., Zhou, Q., Guan, K., 2025. How do hydrological variability and human activities control the spatiotemporal changes of riverine nitrogen export in the upper Mississippi river basin? Environ. Sci. Technol. 60, 1028 – 1039. https://doi.org/10.1021/acs.est.5c06476

Lines 107-110: The sentence “This highly compressed spatiotemporal heterogeneity makes it difficult for analyses based on single catchments to systematically reveal the divergent drivers of different nitrogen forms across spatial states and long-term evolutionary processes” can be deleted.

[Authors’ response]

Following your suggestion, we have removed this sentence from the revised manuscript.

Section 2.1 Study Area: The first sentence says the study analysed 3020 lakes, but then in line 131 the distribution of study lakes across the five limnological regions is presented as “483, 690, 153, 1, 206, and 488”. These numbers sum to only 2021 lakes, with only 1 indicated in the EPL. Why the difference?

[Authors’ response]

Thank you for pointing out this error. In the original manuscript, an incorrect space in the thousands separator (1,206) caused the number 1,206 to be mistakenly split into two separate numbers (1 and 206), resulting in six numbers being displayed instead of five. The correct numbers of lakes in the five limnological regions are: IMXL, 483; TPL, 690; YGPL, 153; EPL, 1,206; and NPML, 488, for a total of 3,020 lakes.

To eliminate any ambiguity caused by thousands separators in the list, we have restructured the sentence in the revised manuscript (Lines 131-133) so that each region is directly paired with its corresponding number of lakes in parentheses: “The study lakes are greater than 2 $km^2$ in surface area and are distributed across five limnological regions: IMXL (483), TPL (690), YGPL (153), EPL (1206), and NPML (488).” (Lines 131－133)”

Lines 428-429: Large seasonal variability is indicated in external nitrogen loading, but nitrogen loading data in Table 1 are annual. I was not able to find the description of how the annual data were distributed seasonally? Is this described somewhere? If not it needs to be added.

[Authors' response]

Thank you for pointing out this potential ambiguity. The external nitrogen loading data used in this study are annual data (Table 1) and were not seasonally disaggregated or redistributed. Our original statement was intended to describe seasonal variation in the model-derived predictive contributions of the external nitrogen loading features, rather than seasonal variation in the nitrogen loading data themselves. We apologize for the ambiguity caused by the original wording.

To avoid this confusion, we have revised the sentence in the Results section from: "With external nitrogen loading showing particularly large seasonal variability during summer." to: "Notably, the predictive contribution of the external nitrogen loading features varied substantially among seasons, with the largest contributions observed during summer." (Lines 430-432)

Thus, the seasonal differences reported in Fig. 7 reflect variation in the model-derived predictive contributions of these annual predictors across seasonal observations, rather than seasonal variation in the underlying nitrogen loading data.

Line 133: Fig 1b is cited, but I do not see a 1b in the figure.

[Authors' response]

Thank you for pointing this out. We apologize for this oversight. We have corrected the erroneous figure citation and carefully checked the manuscript to ensure that all figure references now correspond to the appropriate panels.

Line 166: “(ERA5)” should be (ERA5-Land).

[Authors’ response]

Thank you for pointing this out. We have corrected “(ERA5)” to “(ERA5-Land)” in the revised manuscript.

Lines 515-519: Citation needs to be added for decrease in N loads from 6.8 to 3.5 million tons. What is time unit… per yr?

[Authors’ response]

Thank you for bringing this to our attention. We have clarified the unit as million tons per year and added the appropriate reference in the revised manuscript to support this statement. The revised text now reads:

“This study found that the North China Plain exhibited the most significant decreasing trends in both TN and $NH_3$-N, consistent with the findings of Deng et al. (2024), who reported that improved wastewater management and reduced agricultural emissions during China’s urbanization process have substantially reduced nitrogen loads to inland waters and oceans, with the annual load projected to decline from 6.8 million tons per yr in 2017 to 3.5 million tons per yr by 2050 and the most pronounced reductions occurring in the North China Plain.” (Lines 523-529)

Deng, O., Wang, S., Ran, J., Huang, S., Zhang, X., Duan, J., Zhang, L., Xia, Y., Reis, S., Xu, Jiayu, Xu, Jianming, de Vries, W., Sutton, M.A., Gu, B., 2024. Managing

urban development could halve nitrogen pollution in China. Nat. Commun. 15, 401.
https://doi.org/10.1038/s41467-023-44685-y