Response to reviewer 2:

This study examined the dynamics of major pollution sources contributing to eutrophication in the fifty large lakes of the Yangtze River Plain, a region with 340 million people. These dynamics were studied over a forty-year period (1979-2018) and correlated with satellite-tracked algal bloom events. Pollutant sources studied include leached nitrogen in agro-ecosystems, phosphorus sources, and industrial waste. Remarkable efforts have been made to characterize in space and time the leaching of nitrogen which is a very diffuse source of pollution. This characterization was made using a carbon-nitrogen coupled ecosystem model constrained with meteorological, soil and agronomic data. This study is exceptional by the size of the studied region, the diversity of ecosystems considered (crops, terrestrial and aquatic ecosystems) and used variables. Below I made some comments/criticisms with the aim of improving this manuscript that merit publication.

Response: We sincerely thanks for your valuable comments and questions. We have addressed each in detail below.

The title and conclusive sentences of the abstract only mention the impact of N leaching. However, your study also suggests an impact of P applications and industrial wastes. This must appear more clearly.

Response: Thanks for spotting these out. We have changed the title of this manuscript to "Long-term changes of nitrogen leaching and the contributions of terrestrial nutrient sources to lake eutrophication dynamics on the Yangtze Plain, China" and also the last sentences of the abstract to: "Our results revealed the importance of terrestrial nutrient sources for long-term changes in eutrophic status over the fifty lakes of the Yangtze Plain. This calls for region-specific sustainable nutrient management (i.e., nitrogen and phosphorus applications in agriculture and industry) to improve the water quality of lake ecosystems."

From my point of view, on the most important finding of your study is the suggestion that the origins of lake eutrophication change with space (and maybe with time). Indeed, this finding signifies that policies and technical changes that need to be implemented to decrease lake eutrophication must be adapted to local conditions.

Response: Thanks for this comment. Our study concluded that the primary nutrient causes of eutrophication vary with space, suggesting that specific policies and measures are required to be implemented on local scales to improve eutrophication issues. Now, we have added some discussion into lines 428-432: "Our study revealed that the primary nutrient causes of eutrophication changes varied with regions over the Yangtze Plain, where agricultural nutrient sources were strongly linked with eutrophication changes in western and central lakes, while industrial wastewater showed a significantly positive correlation with PEO trends in eastern lakes. Such spatial variations indicated that scientific policies and measures were required to be implemented at local scales to mitigate eutrophication issues in lake ecosystems."

In the discussion, you mention that the eutrophication induced by industrial wastes is now solved thanks to policies set by the government and changes made in industries. YOU CAN NOT CONCLUDE on that point because your data do not show that at all. You can only mention the fact that the government has become aware of the problem and set up incentives to decrease industrial pollutions.

Response: Thanks for pointing out this issue. Yes, we agree with you that our analysis cannot reach the conclusions as eutrophication induced by industrial wastewater has been solved currently. Now, we have revised the discussion about the linkage between industrial wastewater and eutrophication changes in eastern lakes as lines 449-456: "In such cases, various national strategies and policies have been gradually implemented to promote the green growth of industries on the Yangtze Plain. Considerable efforts were made to encourage the reclamation of wastewater, investment in the advances in wastewater treatment technology and installment of municipal wastewater treatment plants (Li et al., 2013; Lyu et al., 2016). Furthermore, industrial structures were also encouraged to transform from secondary to tertiary industries under the environment-friendly targets of economic development (Huang et al., 2015). All these measures potentially contribute to the decline in industrial sewage on the Yangtze Plain."

The manuscript discussion starts by saying that it is now possible to deplete lake eutrophication because we know one of its main sources: low NUE and high N leaching. One might argues that we know since decades that N leaching causes eutrophication. The challenge rather lies in the setup of good agricultural practices leading to a decrease in nutrient loss. You should mention/list some examples of these practices (agriculture precision, cover cropping, residues managements etc) and how they are implemented or not in this region. More generally, I would suggest to start the discussion with your finding that the causes of eutrophication can be multiple and change in space requiring incentive policies adapted to local conditions.

Response: Thanks for this great suggestion and we have changed the order of the text in the discussion. We start with discussing spatial variations of potential causes of eutrophication on the Yangtze Plain, and what relevant policies and measures should be implemented at local scales to mitigate eutrophication issues (see lines 433-437). Separately, sustainable agriculture development should be encouraged to improve nitrogen/phosphorus use efficiency and thus reduce agriculture nutrient sources available for western and central lakes to potentially control eutrophication issues. In recent years, several agriculture practices have been recommended and implemented, such as optimal fertilization schemes and residue removal, to pursue high-efficiency agriculture on the Yangtze Plain (Cui et al., 2018; Shi et al., 2020). In the revised discussion, these potential management practices have been discussed in terms of their contributions to improving agriculture sustainability (see lines 435-442).

L153 It seems that soil properties have been implemented in the model using the world database on soil types/soil pedology. How did you consider the impact of land use on these soil properties?

Response: Not only soil type but also soil properties including fractions of sand, clay and silt, organic carbon content, C:N, pH, and bulk density, were used as LPJ-GUESS inputs. In the model, soil properties can influence soil hydrological and thermal properties and are not varying throughout the simulation period. For cropland areas, the model does consider the impacts of for instant, nutrients and crop residues on soil organic matter and nutrient contents, but does not simulate other land use impacts, such as grazing and soil compactions etc.

L199 This approach using the linear relationship is not clear? Why did you not directly use the model to estimate the N leaching?

Response: Thanks for pointing this out. We have directly used the modelled N leaching and the linear regression is just to obtain the changing rate/slope over time. Please see the revised text on lines 237-240: "We assessed long-term changes in nitrogen use efficiency (NUE) and nitrogen leaching over the past four decades. For the LPJ-GUESS simulated NUE and leached nitrogen, a linear regression was conducted on the annual mean values for the whole Yangtze Plain to determine the associated change rates (i.e., the regression slopes), and the significance was tested by a t-test."

L212-213 I do not understand why it is a problem since the model can provide those results.

Response: Thanks for this comment. We have used the same levels of fertilizer and manure applications in 2014 for the simulation years of 2015 to 2018 on the Yangtze Plain, due to the un-availability of gridded fertilizer and manure application data. This model setup might not represent the trends in later years, as there are some studies reporting the decreasing trends in total anthropogenic nitrogen input over the Yangtze Plain (Deng et al., 2021; Zhao et al., 2022). Furthermore, although the sown area of each crop changed slightly from year to year, the unavailability of crop cultivation information for the period of 2015-2018 would certainly introduce uncertainties to estimated crop nitrogen uptake. For all these above-mentioned reasons, we only selected the period when we have both PEO data and also the good quality of simulated leaching data, to examine their linkage with terrestrial nutrient sources on the Yangtze Plain.

L217 P sources are not simulated right? This should be specified.

Response: No, P sources were not simulated and they are inventory data. Now, it has been specified as the lines 267-268.

L225 These choices are a bit strange. In general, N (especially mineral N) is more mobile than P. Thus, N leaching is likely to be more linked to N applications than P leaching to P applications. Despite this, you used a complex modelling to estimate N leaching whereas you simply used the P source for P leaching/pollution. You should better explain your choices, and maybe consider to also examine the correlation between N leaching and N applications to have something comparable for the different sources of pollutions.

Response: We agree with the reviewer that N is more mobile than P. Our main argument is that previous studies have identified the low phosphorus use efficiency (< 40%) for agriculture ecosystems on the Yangtze Plain (Zheng et al., 2018). With the reported phosphorus fertilizer application increasing from 6.5 kg P ha^{-1} in 1980 to 22.0 kg P ha^{-1} in 2014, the large proportion of these added fertilizer will end up in the lake systems, suggesting that agricultural phosphorus sources can be considered as potential causes of eutrophication changes. In addition, the leached nitrogen also showed strong dependence ($R^2 = 0.92$, $p < 0.001$ in Fig. S4) on nitrogen applications over the Yangtze Plain for the past four decades. Now, we have added these details into the discussion (lines 464-469) about why we used agricultural phosphorus sources to examine the linkage between terrestrial nutrient sources and eutrophication changes on the Yangtze Plain.

L256 What do you mean with climatological NUE?

Response: The climatological NUE here represents the average NUE for the period of 1979 to 2018, and was used here to demonstrate spatial variations in NUE over the Yangtze Plain. Now, it has been clarified at lines 302-303: "The average NUE for 1979 to 2018 was calculated to examine the spatial patterns of plant nitrogen uptake on the Yangtze Plain".

L270-273 There are considerable differences between crops, which is worth to discuss. The negative effect of double crops is surprising since maintaining a permanent plant cover generally promote N retention in agrosystems.

Response: In terms of large difference in NUEs between crops, we have discussed their difference on lines 306-309: "The NUE values also differed among different crop types for the past four decades. The largest NUE values were found for soybean (74.0 % ± 11.0 %, Fig 5), while the lowest values were found for late-season rice (15.9% ± 4.3%)".

Now, we have added more discussion about comparison with previous studies, the reasons of large NUE difference among crops and the lower NUE for crop rotations as lines 414-426: "Considerable differences in NUE were examined among different crops, with the largest NUE values in soybean for the past four decades (Fig. 5) as previously-documented NUE variations from 1961 to 2011 (Zhang et al., 2015). Generally, soybean has high NUEs mostly due to high protein contents (i.e., > 50%) in its grains (Fabre and Planchon, 2000). With the enhanced leaf nitrogen concentrations related to its biological fixation, soybean tends to achieve a higher photosynthesis rate and delay leaf senescence (Kaschuk et al., 2010; Ma et al., 2022), both of which potentially contributed to its generally high NUE. Furthermore, double-cropping rice showed an overall lower NUE than single-season rice (Fig. 5). It has been previously reported to occur in other double-cropping systems based on field experiments, such as rice-wheat cropping (Liu et al., 2016; Yi et al., 2015), rice-rapeseed cropping (Wang et al., 2021), and wheat-maize cropping (Xiao et al., 2021). In this type of system, fertilizer applications applied for the former crop could have accumulated nitrogen in soil that can be also taken by the latter cultivated crop for their growth and development (Shi et al., 2020). In this regard, chemical fertilizer applications for the latter crop can potentially lead to lower NUE."

Figure 6 -> N leaching under natural ecosystems also increase during the period, which merits discussions.

Response: Thanks for this comment. Yes, the leached nitrogen from natural ecosystems also significantly increased for the past four decades (marked by red stars in Fig. 6h), which were primarily associated with increasing atmospheric deposition (Chen et al., 2016; Xu et al., 2018). Specifically, it was reported that nitrogen emission induced by industry development (i.e., chemical plants and factories) and livestock contributed dominantly to the growth of atmospheric deposition from 1980 to 2012 (Chen et al., 2016). Such enhanced atmospheric nitrogen deposition provided substantial nitrogen available for natural ecosystems and thus resulted in significantly increasing leached nitrogen. Now, all these descriptions have been added into lines 340-343.

L292-293 -> put the treatments close to the numbers to facilitate the reading.

Response: Thanks for this valuable suggestion. Now, I have put the treatments close to the numbers in line 339.

L298 How did you define the drainage area? Watershed?

Response: The drainage area of each lake indicates its catchment area, which is provided by the HydroLAKES dataset (Messager et al., 2016). Now, it has been revised as the entire catchment of each studied lake provided by the HydroLAKES dataset (line 349).

L303 What about the role of atmospheric N depositions? Did you consider this aspect or not? This should be specified.

Response: Yes, atmospheric nitrogen deposition is an input for the model (see description on lines 187-190), so contributes to the modelled nitrogen cycle. In our simulations, the atmospheric deposition is the main nitrogen sources (75.8% ± 6.8%) for natural ecosystems, while it only accounted for 5.8% ± 0.9% of nitrogen sources for agriculture ecosystems. Now, we have added the role of atmospheric N deposition in leached nitrogen from natural ecosystems into lines 340-343.

L305 -> please recall us the acronyms of these variable to facilitate the reading.

Response: We have added the acronyms of these three nutrient variables into the revised manuscript (line 356) to facilitate the reading.

L377 Revise the sentence. The source of uncertainty is not "potential impacts of terrestrial nutrient losses" but the transport processes that mediate the quantity and quality of nutrients that will be transferred from sources to surface water (lakes?)

Response: Thanks for this comment. This sentence has been revised as lines 475-476:" Another source of uncertainty is associated with the lateral transport processes that mediate the quantity and quality of terrestrial nutrients discharged to surface water ecosystems, as well as the impacts of aquaculture-related nutrient sources".

Reference

Chen, F., Hou, L., Liu, M., Zheng, Y., Yin, G., Lin, X., Li, X., Zong, H., Deng, F., and Gao, J.: Net anthropogenic nitrogen inputs (NANI) into the Yangtze River basin and the relationship with riverine nitrogen export, Journal of Geophysical Research: Biogeosciences, 121, 451-465, 2016.
Cui, Z., Zhang, H., Chen, X., Zhang, C., Ma, W., Huang, C., Zhang, W., Mi, G., Miao, Y., and Li, X.: Pursuing sustainable productivity with millions of smallholder farmers, Nature, 555, 363-366, 2018.
Deng, C., Liu, L., Peng, D., Li, H., Zhao, Z., Lyu, C., and Zhang, Z.: Net anthropogenic nitrogen and phosphorus inputs in the Yangtze River economic belt: spatiotemporal dynamics, attribution analysis, and diversity management, Journal of Hydrology, 597, 126221, 2021.
Fabre, F. and Planchon, C.: Nitrogen nutrition, yield and protein content in soybean, Plant Science, 152, 51-58, 2000.

Huang, C., Zhang, M., Zou, J., Zhu, A.-x., Chen, X., Mi, Y., Wang, Y., Yang, H., and Li, Y.: Changes in land use, climate and the environment during a period of rapid economic development in Jiangsu Province, China, Science of the Total Environment, 536, 173-181, 2015.
Kaschuk, G., Hungria, M., Leffelaar, P., Giller, K., and Kuyper, T.: Differences in photosynthetic behaviour and leaf senescence of soybean (Glycine max [L.] Merrill) dependent on N2 fixation or nitrate supply, Plant Biology, 12, 60-69, 2010.
Li, Y., Luo, X., Huang, X., Wang, D., and Zhang, W.: Life cycle assessment of a municipal wastewater treatment plant: a case study in Suzhou, China, Journal of cleaner production, 57, 221-227, 2013.
Liu, X., Xu, S., Zhang, J., Ding, Y., Li, G., Wang, S., Liu, Z., Tang, S., Ding, C., and Chen, L.: Effect of continuous reduction of nitrogen application to a rice-wheat rotation system in the middle-lower Yangtze River region (2013–2015), Field Crops Research, 196, 348-356, 2016.
Lyu, S., Chen, W., Zhang, W., Fan, Y., and Jiao, W.: Wastewater reclamation and reuse in China: opportunities and challenges, Journal of Environmental Sciences, 39, 86-96, 2016.
Ma, J., Olin, S., Anthoni, P., Rabin, S. S., Bayer, A. D., Nyawira, S. S., and Arneth, A.: Modeling symbiotic biological nitrogen fixation in grain legumes globally with LPJ-GUESS (v4. 0, r10285), Geoscientific Model Development, 15, 815-839, 2022.
Messager, M. L., Lehner, B., Grill, G., Nedeva, I., and Schmitt, O.: Estimating the volume and age of water stored in global lakes using a geo-statistical approach, Nature communications, 7, 13603, 2016.
Shi, X., Hu, K., Batchelor, W. D., Liang, H., Wu, Y., Wang, Q., Fu, J., Cui, X., and Zhou, F.: Exploring optimal nitrogen management strategies to mitigate nitrogen losses from paddy soil in the middle reaches of the Yangtze River, Agricultural Water Management, 228, 105877, 2020.
Wang, C., Yan, Z., Wang, Z., Batool, M., El-Badri, A. M., Bai, F., Li, Z., Wang, B., Zhou, G., and Kuai, J.: Subsoil tillage promotes root and shoot growth of rapeseed in paddy fields and dryland in Yangtze River Basin soils, European Journal of Agronomy, 130, 126351, 2021.
Xiao, Q., Dong, Z., Han, Y., Hu, L., Hu, D., and Zhu, B.: Impact of soil thickness on productivity and nitrate leaching from sloping cropland in the upper Yangtze River Basin, Agriculture, Ecosystems & Environment, 311, 107266, 2021.
Xu, W., Zhao, Y., Liu, X., Dore, A. J., Zhang, L., Liu, L., and Cheng, M.: Atmospheric nitrogen deposition in the Yangtze River basin: Spatial pattern and source attribution, Environmental Pollution, 232, 546-555, 2018.
Yi, Q., He, P., Zhang, X., Yang, L., and Xiong, G.: Optimizing fertilizer nitrogen for winter wheat production in Yangtze River region in China, Journal of Plant Nutrition, 38, 1639-1655, 2015.
Zhang, X., Davidson, E. A., Mauzerall, D. L., Searchinger, T. D., Dumas, P., and Shen, Y.: Managing nitrogen for sustainable development, Nature, 528, 51-59, 2015.
Zhao, Z., Zhang, L., and Deng, C.: Changes in net anthropogenic nitrogen and phosphorus inputs in the Yangtze River Economic Belt, China (1999–2018), Ecological Indicators, 145, 109674, 2022.
Zheng, J., Wang, W., Cao, X., Feng, X., Xing, W., Ding, Y., Dong, Q., and Shao, Q.: Responses of phosphorus use efficiency to human interference and climate change in the middle and lower reaches of the Yangtze River: historical simulation and future projections, Journal of Cleaner Production, 201, 403-415, 2018.