

Reviewer #1:

General comment:

This paper evaluated the effects of human land uses on sediment denitrification and N₂O production in Yangtze lakes. A broad-wide monitoring and spatial analysis in 20 lake's watersheds around Yangtze River are invaluable. The evaluation of the indirect effects of human-dominated land uses (HDL) in watersheds on sediment denitrification and N₂O production in the lakes by structural equation modeling is a new approach and can be also appreciated in this study area. However, it is bit difficult to understand for me whether this MS provides new insight relative to current knowledge. The discussion on the results of the study was insufficient and did not support conclusions adequately.

1) There is little discussion on the results of SEM analysis (Table 4 and Figure 4) that are the main results of this study. More discussion on the results of SEM analysis is needed to support conclusion of this study.

Response: We have added more discussion on the results of SEM analysis (Pages 19-20 Lines 438-459). Thanks for your comments.

2) In regard to SEM analysis, I think the authors should not delete NH₄⁺ from the analysis (P. 7824 line 2 -4) because NO₃⁻ and NH₄⁺ concentration in water column is regulated by different mechanisms and NH₄⁺ can be influenced on sediment denitrification directly and indirectly.

Response: This is an important suggestion. We have revised our SEM model and the NH₄⁺ has been included in the final model (Pages 33 and 37).

3) I did not understand the causal relation between DO and NO₃⁻ (DO → NO₃⁻; standardised path coefficients -0.34) in Figure 4. How do the authors explain the causal relation (also the causal relation between ORP and NO₃⁻). The authors should explain the results in Figure 4 and Table 4 more carefully.

Response: We have explained the causal relation between DO and NO₃⁻ in the revised manuscript (Page 11 Lines 247-249). The ORP has been deleted from the SEM model because it is positively related to DO. Some studies also suggest that DO can be a proxy for ORP (e.g., Heduit and Thevenot 1989).

Heduit A, Thevenot D. 1989. Relation between redox potential and oxygen levels in activated-sludge reactors. Water science and technology, 21: 947-956.

4) The authors should also discuss on the result in Figure 2 that showed the relationship between Ln-Background denitrification rate and Sqrt-N₂O production rate. What does it mean?

Response: The figure 2 shows that N₂O production rates significantly increased with increasing background denitrification rates. We have added more discussion on this result (Pages 17-18 Lines 386-396).

Specific comments:

5) P. 7819 line 25-P. 7820 line 2: The authors should state the summary of 20 lakes based on Table S1.

Response: Revised (Page 6 Lines 125-127).

6) P. 7824 line 2 -4: The authors should not delete NH_4^+ from the SEM analysis because NO_3^- and NH_4^+ concentration in water column is regulated by different mechanisms and the correlation between the two variables was not so strong.

Response: Please see the response to the comment 2 and Page 37.

7) P. 7824 line 6 -9: Please explain how to estimate the values of indirect effects in Table 4 with an example using standardised path coefficients in Figure 4.

Response: For instance, the indirect effect of HDL on background denitrification rate via NO_3^- is 0.58 (Page 33 Table 4). We can calculate it through the following steps. Firstly, the total effect of HDL on NO_3^- (0.72) is calculated by adding direct effect (0.52) and indirect effect via DO and NH_4^+ ($[-0.44 \times -0.46] + [-0.44 \times -0.15 \times -0.02] + [0.53 \times -0.02] = 0.20$; please see Fig 4B in Page 37). Secondly, the total effect of NO_3^- on background denitrification rate (0.81) is calculated by adding direct effect (0.81) and indirect effect (0.00; Fig 4B). Therefore, the indirect effect of HDL on background denitrification via NO_3^- is 0.58 ($0.72 \times 0.81=0.58$).

8) P. 7825 line 8 -10: Please show r, and P values.

Response: Revised (Page 13 Lines 281-283).

9) P. 7825 line 22 -23: Please show r, and P values.

Response: Revised (Page 14 Line 300).

10) P. 7828 line 1 -: The authors should pay attention to the significant digits of the values used in this paragraph.

Response: Revised and 4 significant digits were retained (Page 16 Lines 360-366).

11) P. 7828 line 1 -8: Is it possible to compare the annual values calculated from the one-time monitoring data to the literature annual values? The authors should mention potential limitations of the data.

Response: Revised (Pages 16-17 Lines 367-371).

12) P. 7828 line 13-22: I did not understand the significance of this paragraph. The authors only entered the residence time in the equation of Seitzinger et al. (2006), not in discussion the validity of the results in this study.

Response: This paragraph has been deleted. Thank you for this comment.

13) P. 7828 line 16: Please show the unit of WRT.

Response: As the paragraph has been deleted, the unit of WRT is not presented in the revised manuscript.

14) P. 7829 line 4-5: I understood the incubation in this study was conducted under anoxic condition (P. 7821, line 11-12).

Response: Yes, our study was conducted under anoxic but not anaerobic condition. We have deleted this unsuitable sentence in discussion.

15) P. 7829 line 18-P.7830 line18: This paragraph did not discuss on the results of SEM analysis (Figure 4 and Table 4).

Response: Please see the response to the comment 1 and page 19 and 20.

16) Table 1: The authors should pay attention to the significant digits of the values.

Response: The values were revised and two or three significant digits were retained (Table 1 Page 30).

17) Figure 3: Please explain the figs (A) and (B) in the caption.

Response: Revised (Please see the page 36).

18) Figure 4: Please explain the figs (A) - (D) in the caption.

Response: Revised (Please see the page 37).

Reviewer #2:

General Comment:

This is a timely and important analysis of N₂O production and water quality related to human land use in lake catchments. It fills a gap in our understanding of the factors that contribute to N₂O production in lacustrine systems. As a general recommendation, however, I would advise the authors to emphasize more clearly those findings which are new and add unique insight. It is generally known that land use affects water quality and N₂O production in terrestrial environments. However this work is unique because the clear documentation of these connections in lacustrine environments, especially the effect on STN, was lacking until this publication.

Specific Comments:

1) In the Abstract on lines 10-11, the following sentence seems ambiguous: "Increased background denitrification rate would result in increased N₂O production rate." Is this meant to indicate that increased background denitrification would hypothetically or theoretically result in increased N₂O production rates? Or that it in fact *did* result in increased N₂O production rates? Later it is stated that "N₂O production rates increased with increasing background denitrification rates."

Response: Thanks for your valuable suggestion. We have revised this sentence according to your suggestion (Page 2 Line 22).

2) In the introduction it is stated that "the percentage of eutrophic lakes...increased from 41% in 1980 to 85% in 2005." It seems more appropriate to say that there was an increase in the percentage of lakes *classified* as 'eutrophic' (presumably according to a classification method outlined in Liu et al., 2010). It's not necessary to spell out what exactly is defined as 'eutrophic', but keep in mind this word is often used to describe relative as well as absolute nutrient levels. This also applies to lines 18-20 in the "Study sites" section which also reference eutrophication. In addition, there is a reference to 'built-up' lands. It might be good to say "the percentage *classified as* built-up lands" to make it clear you are talking about a land classification scheme. I notice later in the paper there is an explanation of the method used to classify land use.

Response: Revised (Page 3 Line 42; Page 6 Line 114 and 118).

3) Later in the introduction it is stated that "the relative N₂O production for heavily polluted river and estuary sediments is approximately 0.03." I'm assuming you mean the N₂O : N₂ ratio is 0.03? Many readers may be familiar with the work of Seitzinger and Kroeze and will know what is meant, but it would be good to make this more clear.

Response: Thanks for your suggestion. We have revised this sentence to make it more clear (Page 4 Line 72).

4) In the section called "Watershed land use calculation," the categories 'cropland' and 'built-up land' are combined to create a variable called 'human-dominated land uses.' However it seems likely that crop production might have a distinct effect on water quality when compared to other human land uses (for example sewage treatment and urban runoff and groundwater effects). Is there reason to believe that the different land uses will affect the lake in similar ways, with respect to the variables measured? For example, is there reason to believe that both 'built-up' land and 'cropland' add the same type of inorganic nitrogen to the water column in these systems? Could it be the case that the N additions from these two sources have a distinct effect on denitrification? It might be a good idea to add a sentence or two justifying the choice to combine these variables.

Response: Thanks for your friendly reminder. We know that the cropland may have a distinct effect on water quality when compared to built-up land. It has been also reported that land use types in the catchments have a significant effect on the ratio of NH₄⁺ to NO₃⁻ in exported runoff (e.g., Coulter et al. 2004. Water quality in agricultural, urban, and mixed land use watersheds. Journal of The American Resources Association, 40: 1593-1601). In our study, higher nitrogen concentration was found in both some urban lakes (e.g., Lake Linghu) and agricultural lakes (e.g., Lake Wuchanghu), which lead to nonsignificant correlations between lake nitrogen concentration and percentage of cropland or percentage of built-up land. Therefore, we used a combined variable (i.e., the percentage of human-dominated

land uses) in statistical analyses. In the text, we have added a sentence to explain why we have combined the two variables (Please see the Page 10 Lines 219-221).

5) As far as adding NO₃ to the model and excluding NH₄⁺ idea to add some insight into how much the model would change if NH₄⁺, for example, were included or substituted for NO₃⁻. If the conclusions hold even when NH₄⁺ is added or substituted into the model, then this could bolster the assumption that NO₃⁻ can stand in for NH₄⁺ and TN in the model. Or maybe there is some other reason to think NH₄⁺ shouldn't have its own unique effect in these systems, apart from the strong correlation with NO₃⁻?

Response: We have revised our SEM model and found that our conclusions hold even when NH₄⁺ is added into the model (Pages 33 and 37).

6) Related to point 5, it is stated that "100% of the positive indirect effect of HDL on background denitrification (0.55) was mediated through water quality (principally via NO₃⁻). This is where NH₄⁺ could make a difference. If HDL affects NH₄⁺, which in turn is nitrified to NO₃⁻, then this could be an additional indirect effect of HDL on denitrification and N₂O production, although it would require one extra step. It may be that the experiment described cannot fully evaluate this possibility, since the nitrification rate was not measured. However later in the paper it is stated that "nitrification processes [are] the major source of N₂O." This raises the possibility that future research on lake N₂O and land use will have to measure nitrification directly, or risk missing the most important process of N₂O production.

Response: Thanks for your important suggestion. We have revised our SEM model and the NH₄⁺ was included in the final model (Page 37). We agree with your views that nitrification may be an important process of N₂O production in shallow lakes and future studies are needed to measure nitrification directly.

7) It is stated the "The relative N₂O production >1 implies that the production of N₂O through nitrification must have occurred..." But it is also stated that the relative N₂O production was 0.17 - it would be good to clarify what this means.

Response: In the present study, the relative N₂O production ranged between 0.02 and 0.62, and averaged 0.17 (Page 30, Table 1). However, some studies have found that the relative N₂O production can be larger than 1 (e.g., Xu et al., 2008). Thanks for your suggestion. We have revised this sentence to make it more clear (Page 18 Lines 405-407).