Peer Review File

Quantification of Ammonium Release from an Aging Free Water Surface Constructed Wetland To Improve Treatment Performance

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Andrea Ludwig, University of Tennessee

First Review

Associate Editor's Summary for Manuscript Review

Manuscript # 1003

Title: "Quantification of ammonium release from an aging free water surface constructed wetland to improve treatment performance"

Reviewers (3):
Marc Beutel, PhD –University of California, Merced
Alex Horne, PhD – University of California, Berkeley (Emeritus)
Andrea Ludwig, PhD – University of Tennessee

Overall Recommendation: Accept After Moderate Revision

AE Summary

The three reviewers all agree that the manuscript is well-written and presents work that is relevant to JEED readership. There does not appear to be any conflicting review comments, and each review comment warrants a response. On the technical merit, there is consensus that the study was executed using sound methodology. No major issues were identified. There are multiple comments related to providing more context for the system studied and comparing their system more directly to other work in treatment wetland nitrogen budgets. One reviewer states that it is imperative that the overall long-term nitrogen mass balance of the field site (Walnut Cove WWTP) be addressed. One of the more significant comments includes revising the section describing the modeling approach to focus the emphasis on the appropriate model. On the format, there is a consensus that the paper would benefit from a combined results and discussion section (instead of the two sections as it currently reads), which will likely take care of some of the redundancy. Several comments were made about increasing font sizes in figures and improving overall readability.

Beutel First Review

Review of JEED manuscript: Quantification of ammonia release from an aging free water surface constructed wetland to improve treatment performance

Feb 1, 2023

Dr. Marc Beutel, UC Merced

The manuscript presents an interesting experimental assessment of internal release of ammonia in treatment wetlands. The authors do a good job in detailing the knowledge gap that aim to fill and the manuscript is well written and well presented. See detailed comments below. I would classify this review as recommending a "moderate" rewrite, somewhere between a minor and major rewrite. I recommend that you have the reviewers give the revised manuscript a second review to confirm changes made are on target. I would be happy to do that.

1. The authors need to make a more explicit link to how their study results inform design, a key aim of JEED. The current emphasis is on informing management, which is good. But a bit more thought and discussion on how results could inform design is needed. Perhaps authors could argue that by having a better constraint on the internal release term for N, wetland designers should consider upsizing low-loaded systems to allow for future increases in internal loading. Or perhaps model metrics can be used to look at different scenarios of potential internal release and how that could affect treatment performance, thereby informing sizing/design. Where "design" was mentioned on line 583, I did not really understand the point that was being made. That text should be revisited accordingly.
2. In general, the figures need larger font size. Please revisit figures with an eye to enhance readability. Make font size much larger.
3. More detail is needed regarding the study site and how the treatment wetland fits into the wastewater treatment plant. Is the wetland polishing secondary effluent, or is it taking fairly untreated wastewater and processing BOD? This will help readers understand the treatment wetland environment better. What is meant by "pre-

treated wastewater" on line 439? We need to know to understand how hard this wetland is working.

4. I suspect you would have presented this dataset if you had it, but do you have both inlet and outlet ammonia for the wetland. Figure 2, I think, is presenting outlet ammonia (make it more clear what is being presented in fig 2). It would be nice to show both in and out ammonia data, or % concentration removal for the long-term data set. How do we know patterns of outlet ammonia concentration are not simply following patterns of inlet ammonia concentration?

5. Regarding ammonia and nitrate analytical methods, please report method detection limits.

6. One factor that needs to be better addressed, and that can have a big impact on N cycling, is temperature. Please report some temperature values/ranges where it makes sense as you present study results. Were microcosm experiments performed at room temperature (~20 deg C)? Can rates be considered to represent ~20 deg C expected rates?

7. You note around line 225 and line 538 that DO declines in early day of experiment did not correspond with increases in nitrate, indicating limited nitrification. One reason it may have taken a number of days for nitrate to appeared is the time it took for nitrifying bacteria to get a foothold in your microcosms. These are finick and slow growing bacteria. Think of the good old BOD5 tests, keep it to 5 days partly so we assess DO demand of organic waste and not that of ammonia, since nitrifiers take longer than 5 days to get active. You might consider adding this possible mechanism to your discussion of this observation. Note too that some of the wording in these two sections was repetitive. Edit to alleviate that problem.

8. In Section 2.3 I suggest you drop the text regarding the k-C* model and focus your attention on the sequential N removal modeling approach. My thinking here is the k-C* model does take into account internal release – that is what the C* represents, the background concentration you can't get below due to internal production. Typically, C* is assumed to be 0 mg/L since ammonia can disappear completely via microbial processes. But as you note, in the sequential mass balance internal loading is not explicitly included, and that opens a window for your study. Focus readers attention on that model. Drop C* text, or if not you'll need to discuss what C* is in context of this study, which I think will just confuse things.

9. One of the more significant weaknesses of the paper is the discussion, rather than just presentation, of results in the results section, see 3.2 for example (discussion of inverse relation between concentration and release) and p 20 and top of 21 (discussion of difference in parameter values). This paper could benefit from formatting it as a results and discussion section. While the JEED paper template says this is not allowed, we are changing that. Either reformat as results and discussion (and rework subheads accordingly), or critically review and remove discussion from the results section.

10. Line 371 – ammonia a byproduct of respiration? Do you mean mineralization or decay, which is not necessarily linked to respiration?

11. While I consider it bad form to suggest in reviews that folks cite your own papers, in this case I think I can be of some help in putting your rates (~100-300 mg-N/m2-d) into context, by comparing your results to some of my own experimental anoxic flux rates of ammonia from sediment in eutrophic lakes. Not sure what temperature you were at, but I recently measured similar rates of ammonia release in incubations at 25 deg C. See report below p 28-29.

    https://www.vidwater.org/lake-henshaw-and-lake-wohlford-harmful-algal-blooms-management-and-mitigation-plan

    Also, rates measured in eutrophic lakes in cooler hypolimnion (~15 deg C) are on the order if 40 mg-N/m2-d – see https://www.sciencedirect.com/science/article/pii/S0925857406000929#fig3 (Inhibition of ammonia release from anoxic profundal sediments in lakes using hypolimnetic oxygenation, MW Beutel, Ecological Engineering 28 (3), 271-279).

Incorporate this info into your paper as you see fit. I think the takeaway here is enriched eutrophic aquatic sediment have capacity to release a lot of ammonia!

Horne First Review
Review of JEES Manuscript “Quantification of ammonium release from an aging free water surface constructed wetland to improve treatment performance” by Brock J.W. Kamrath et al.

Review by: Alex Horne, Prof Emeritus, Dept. CEE University of California Berkeley. anywaters@comcast.net

Summary: This MS is of good quality with a nice blend of field and lab work as well as the theoretical/modeling approach we were aiming for with (some articles) in the new journal. It also tackles a real-world performance concern, namely the loss of performance of a FSW taking real world treated effluent. The authors should be congratulated on their diligence and results. The replication and results seem to be a good estimate of the flux of NH4 from the sediments of a static (lab work) and quiescent (?) conditions in the WWTP effluent wetland treatment cells. Replication was adequate in the lab studies and the analysis and modelling are good. The fact that the flux rates declined as the base added ammonia was increased in the microcosms indicates proof of a commonsense result. However, more context is needed to give our readers a better understanding. *In particular, the implications of more N coming out than going in is a mass balance no-no. This must be cleared up before publication*.

My recommendations: MS is acceptable for JEES with the revisions suggested. I would be willing to review the MS after modification.

My concerns:

1. **High flux rate of NH4 in sediments (from organic detritus?) to the overlying water.**

   The flux rates measured (140 to 210 mg/m^2/d) are high but are not much higher than some other studies, for example, the large, well-studied Waco Lake Marsh, Texas (Scott et al. 2008) who report a high of 210 mg/m^2/d (assuming their hourly rate can be extrapolated to 24 hours), an average 64 mg/m^2/d. The also have some low flux rates and the odd negative result. These wetland fluxes are much higher than many others in the literature measuring sediment flux from eutrophic lake/estuarine sediments (5-84 mg/m^2/d and 15 mg/m^2/day is probably a good typical number). So, I do not question the author’s work here since it looks like their sediments are very N-rich. But they do need to be put in context. I suggest that context is that their Walnut Cove wetland is (may be?) unusual in that it receives high ammonia all the time and no nitrate to speak of. Others (included in their references) made the same mistake of putting full strength 2y effluent with 25 mg/L NH4 into a wetland then then wondering why the wetland does not change anything. For a wetland to work with nitrogen removal the ammonia needs to be low since NH4 is a highly reduced molecule and wetlands have, by definition, anoxic sediments. Up front aeration is a good answer and aeration systems are very cheap and easy to work with so that would be a solution to the Walnut Cove ammonia releases.

2. **Unstable long-term mass balance.**

   The authors comment that a reason for their work was the “poor performance of the NC treatment wetlands” and that is a good reason. However, it seems that there is always more ammonia coming out than goes into the wetland and their data shows this. If this were true in the long-term, we would not need the energy-intensive Haber-Bosh ammonia plants that dot the world today to make N-fertilizers and explosives. There cannot be more N going out than coming in on the long term, but I think the authors ignore this problem (maybe I missed something here). It seems like the inflow is mostly NH4 with almost no nitrate and an unknown amount of organic-N. The authors need to explain the problem of a positive mass balance. My own guess is that there must be times of year when the balance goes the other way (winter maybe, calm period when sediment flux is substrate inhibited?). Are there odd occasions when the inflow reaches more normal NH4 levels like 25 mg/L? I have found this for some specialized small WWTPs.

   My help here is twofold, some of my own work on wetlands and a comment of the performance of the Walnut Cove WWTP. Below I show inflow and outflow averaged over 24 years for mostly treated municipal effluent entering a 119-ha constructed treatment wetland (28 cells) at Prado in Orange County CA. The ammonia entering is low (0.12 mg/L), as expected from a modern POTW, nitrate is quite high (6.1 mg/L), and organic-N low (0.39 mg/L). This compares with the author's inflow of ammonia of 2.6 mg/L (spring) and 8 mg/L (winter). So, their wetland is not only heavily loaded with NH4 (as they comment) but is somehow storing ammonia then releasing it. I agree with the authors that the long-term increase in ammonia over 20+ years from their treatment wetlands is well-documented. Such high levels of NH4 are not good when discharged to surface waters, and puzzling if outflow total-N (mostly NH4 here it seems) is higher than the long term inflow. Long-term inflow needs to be on this figure 2 (if available) to sort out the true mass balance.

Table 1. Nitrogen dynamics (mg/L) over 24 years for Prado Wetland, Orange County, CA. The source is mostly treated POTW effluent, HRT about 5 days, area 119 ha, flow ~ 1m^3/s & n is about 300 for each N-species (*source Horne et al. J. Ecol, in review*).

	Inflow	Outflow	Difference	% Removal
NH4-N	0.12	0.07	0.04	38
NO3-N	6.1	2.6	3.5	57.4
Organic-N	0.39	0.64	-0.25	-65

3. **Role of detritus is not clear (to me).** The authors, correctly point out that "detritus" can cause the wetland to lose volume and they quote 1-3 cm/y, which is a good proportion of their originally 30 cm deep wetland. While this is true for the first year, the organic matter gets substantially compressed otherwise their wetland would have filled in long ago. I assume by detritus they mean dead *Typha* stems which collapse into the sediments each winter.These dead stems and leaves have a very low N-content so seem unlikely to be a (large) source of NH4 later. Are the authors assuming some sequestration by the living plants then release? But if so, there would be no mean annual effect due to recycling. One explanation is inflowing NH4 from other sources. This would expand the inflow so it is somehow higher than the authors suggest and fluxes in and out of the anoxic sediments over time depending on the concentration gradient. However, no such large external NH4 sources come to my mind. N_2-fixation is unlikely here since nitrogenase is strongly repressed by ammonia at these levels, so something must be coming in. Aerial deposition will occur but unless the POTW is close to a cattle farm (feeding lot) that seems also an unlikely source of NH4 (coming in as NH3 vapor probably).

4. **Figures.** These are hard to read. The authors should realize that the figures will be pretty small in a journal though they can be expanded online. The captions usually come out worse than the lines and data points. My suggestion is that the captions in particular need to be larger and bold and black (I suggest 14 point for the caption wording and 12 for everything else (years can be 11 point) – if you are using Times Roman. My eyes are not what they were 75 years ago and so will some of our readers be too. The color figure 1 may not show up well as shown but I just suggest they think about that. Who is going to pay for the color figure?

Ludwig First Review

Review of "Quantification of ammonium release from an aging free water surface constructed wetland to improve treatment performance"

Submitted to the Journal of Ecological Engineering Design

Conducted by A. Ludwig, University of Tennessee

March 20, 2023

This work aims to fill the gap in understanding the magnitude and implications of the detritus layer as an internal nitrogen source in free water surface constructed wetlands nitrogen budgets. The manuscript presents findings from a study that incorporates field data, benchtop microcosms, and resultant modeling effort that has implications for similar sites and perhaps beyond. The paper is well written, easy to follow, and has implications for ecological engineering design and practice, and therefore, is appropriate for consideration in this journal.

Recommendation: Acceptable after light revisions.

General Comments:

1. The stated conclusions from the experiment match the scale and scope of the study. The authors recognized the limitations of the study and addressed them as well as tied them to future research trajectories.

2. More detail would be beneficial to discuss the detritus substrate collection methods in the field. What was the range of mixed depths (line 169)? How was active vegetation removed (I assume, since it's not explicitly stated)? Was there any other preparatory filtering? How were they homogenized?

3. Throughout the text, it was hard to follow the experimental design in terms of treatments, replicates, number of microcosms, number of runs. I suggest clarifying it in the methods section (page 8) perhaps with a grid figure of the microcosms and treatments. Line 300 in the methods should be clarified.

4. The target treatment of 0 mg/L NH4-N ended up with detectable levels, which I assume was part of the tap water. Some mention of this would help explain in the methods as well as including detection limits of the water chemistry analyses.

5. There is quite a bit of overlap between results/discussion/conclusions. Consider shortening results and condensing discussion.

6. Related to the technical aspects of the study, the experimental design (treatments, replicates, and relation to real-world facts) is strong. Indeed more data points would be good, but the findings are presented in a way that discusses shortcomings and in an appropriate manner. Bringing field sampled sediments into a controlled laboratory is a good way to start the investigations into this subject. The selected methods (analytical, statistical) appear sound and on par with the current science.

Author response to first review

Journal Editor Summary

Overall Recommendation: Accept After Moderate Revision

The three reviewers all agree that the manuscript is well-written and presents work that is relevant to JEED readership. There does not appear to be any conflicting review comments, and each review comment warrants a response. On the technical merit, there is consensus that the study was executed using sound methodology. No major issues were identified. There are multiple comments related to providing more context for the system studied and comparing their system more directly to other work in treatment wetland nitrogen budgets. One reviewer states that it is imperative that the overall long-term nitrogen mass balance of the field site (Walnut Cove WWTP) be addressed. One of the more significant comments includes revising the section describing the modeling approach to focus the emphasis on the appropriate model. On the format, there is a consensus that the paper would benefit from a combined results and discussion section (instead of the two sections as it currently reads), which will likely take care of some of the redundancy. Several comments were made about increasing font sizes in figures and improving overall readability.

Response:

Thank you for compiling the comments. We have addressed each reviewer's comments below, and made a few other modifications within the text and figures to improve readability.

Reviewer #1: Marc Beutel
Reviewer Summary

The manuscript presents an interesting experimental assessment of internal release of ammonia in treatment wetlands. The authors do a good job in detailing the knowledge gap that aim to fill and the manuscript is well written and well presented. See detailed comments below. I would classify this review as recommending a "moderate" rewrite, somewhere between a minor and major rewrite. I recommend that you have the reviewers give the revised manuscript a second review to confirm changes made are on target. I would be happy to do that.

Response:

Thank you for your comments Dr. Beutel – they have helped us strengthen the article. We welcome a re-review if you deem necessary.

Specific Comments

1. The authors need to make a more explicit link to how their study results inform design, a key aim of JEED. The current emphasis is on informing management, which is good. But a bit more thought and discussion on how results could inform design is needed. Perhaps authors could argue that by having a better constraint on the internal release term for N, wetland designers should consider upsizing low-loaded systems to allow for future increases in internal loading. Or perhaps model metrics can be used to look at different scenarios of potential internal release and how that could affect treatment performance, thereby informing sizing/design. Where "design" was mentioned on line 583, I did not really understand the point that was being made. That text should be revisited accordingly.

   Response: We attempted to clarify this in a number of locations, specifically within the Introduction (Lines 135-137), discussion in Section 3.5 (for example lines 501-503, 540-550), Section 3.6, and most significantly Section 3.8 (Application of results)

2. In general, the figures need larger font size. Please revisit figures with an eye to enhance readability. Make font size much larger.

   Response: Figures have been updated and enlarged

3. More detail is needed regarding the study site and how the treatment wetland fits into the wastewater treatment plant. Is the wetland polishing secondary effluent, or is it taking fairly untreated wastewater and processing BOD? This will help readers understand the treatment wetland environment better. What is meant by "pre-treated wastewater" on line 439? We need to know to understand how hard this wetland is working.

   Response: We expanded the site description (Section 2.1 on Materials and Methods). In short, the wetlands at the site receive wastewater that has passed through two aerated lagoons and a duckweed raceway. The typical influent NH_4-N concentration to the wetlands is 1-12 mg/L and 1 mg/L NO_3-N.

4. I suspect you would have presented this dataset if you had it, but do you have both inlet and outlet ammonia for the wetland. Figure 2, I think, is presenting outlet ammonia (make it more clear what is being presented in fig 2). It would be nice to show both in and out ammonia data, or % concentration removal for the long-term data set. How do we know patterns of outlet ammonia concentration are not simply following patterns of inlet ammonia concentration?

   Response: You are correct - we would have presented long-term inlet data if we had it. Unfortunately, the only long-term data available is the WWTP outlet data (Figure 2), which is essentially wetland outlet data - as only a short stay in the chlorinator separates wetland outlet water from the water sampler. This lack of long-term data was the motivating factor for the Kamrath dissertation (from which this manuscript is derived). We did add our average inlet and outlet concentrations we did measure in two short term studies in the Introduction, lines 113-117 to communicate the only inlet/outlet data we do have (and will be the subject of a subsequent article). Based on our short-term observations, we do not believe that the observed outlet concentrations were a function of changes of wastewater entering the WWTP.

5. Regarding ammonia and nitrate analytical methods, please report method detection limits.

   Response: Method detection limits have been added. The method detection limits were 0.015 and 0.2 mg-N L-1 for ammonium and nitrate, respectively (Lines 254-255).

6. One factor that needs to be better addressed, and that can have a big impact on N cycling, is temperature. Please report some temperature values/ranges where it makes sense as you present study results. Were microcosm experiments performed at room temperature (~20 deg C)? Can rates be considered to represent ~20 deg C expected rates?

   Response: Temperature values (lines 280-282), along with additional water quality parameters, have been added to section 2.4.1 in the Methods. The experiments were conducted at room temperature.

7. You note around line 225 and line 538 that DO declines in early day of experiment did not correspond with increases in nitrate, indicating limited nitrification. One reason it may have taken a number of days for nitrate to appeared is the time it took for nitrifying bacteria to get a foothold in your microcosms. These are finick and slow growing bacteria. Think of the good old BOD5 tests, keep it to 5 days partly so we assess DO demand of organic waste and not that of ammonia, since nitrifiers take longer than 5 days to get active. You might consider adding this possible mechanism to your discussion of this observation. Note too that some of the wording in these two sections was repetitive. Edit to alleviate that problem.

   Response: We adjusted some of the verbiage in the paragraph (lines 256-272) to be more clear on this issue and reduced the text on this topic later in the manuscript

8. In Section 2.3 I suggest you drop the text regarding the k-C* model and focus your attention on the sequential N removal modeling approach. My thinking here is the k-C* model does take into account internal release – that is what the C* represents, the background concentration you can't get below due to internal production. Typically, C* is assumed to be 0 mg/L since ammonia can disappear completely via microbial processes. But as you note, in the sequential mass balance internal loading is not explicitly included, and that opens a window for your study. Focus readers attention on that model. Drop C* text, or if not you'll need to discuss what C* is in context of this study, which I think will just confuse things.

   Response: The co-authors took this comment very seriously and spent a significant amount of time discussing it. We feel strongly that we want to make the point that the P-k-C* model certainly represents the background concentrations early in the development of the wetland, but seems to fall short when the internal source increases significantly with time. Certainly you can make mathematical guesses as how to increase C* values for NH_4-N with time to achieve whatever outlet concentration you are seeing.

   So we choose to keep it in the manuscript starting in Section 2.5 (and we enhanced the description somewhat) to show how background concentrations are generally handled with the most popular design model, P-k-C*, and how our model demonstrates the limitation of the outlet concentration predictive power in older systems. In the Results and Discussion (section 3.5 lines 544-550) we circle back around and compare ours and the P-k-C* model estimated NH_4-N return rates at the Walnut Cove system. Essentially, our results indicate that if you design a wetland using the current P-k-C* model with recommended C* levels for initial design, it will fail to predict N removal as the wetland ages.

9. One of the more significant weaknesses of the paper is the discussion, rather than just presentation, of results in the results section, see 3.2 for example (discussion of inverse relation between concentration and release) and p 20 and top of 21 (discussion of difference in parameter values). This paper could benefit from formatting it as a results and discussion section. While the JEED paper template says this is not allowed, we are changing that. Either reformat as results and discussion (and rework subheads accordingly), or critically review and remove discussion from the results section.

   Response: We felt that way too, but followed the journal's initial guidance on this. The results and discussion sections have been combined which made the manuscript much better.

10. Line 371 – ammonia a byproduct of respiration? Do you mean mineralization or decay, which is not necessarily linked to respiration?

    Response: In organic matter, nitrogen is assimilated and stored in stored in reduced amine and amidine functional groups. Whether abiotic or in most cases biotic, decay results in the production of inorganic nitrogen that is in the form of ammonium. Because of anaerobic conditions in wetland organic sediment, ammonium is the stable form and accumulates in the porewater.

11. While I consider it bad form to suggest in reviews that folks cite your own papers, in this case I think I can be of some help in putting your rates (~100-300 mg-N/m2-d) into context, by comparing your results to some of my own experimental anoxic flux rates of ammonia from sediment in eutrophic lakes. Not sure what temperature you were at, but I recently measured similar rates of ammonia release in incubations at 25 deg C. See report below p 28-29. https://www.vidwater.org/lake-henshaw-and-lake-wohlford-harmful-algal-blooms-management-and-mitigation-plan Also, rates measured in eutrophic lakes in cooler hypolimnion (~15 deg C) are on the order if 40 mg-N/m2-d – see https://www.sciencedirect.com/science/article/pii/S0925857406000929#fig3 (Inhibition of ammonia release from anoxic profundal sediments in lakes using hypolimnetic oxygenation, MW Beutel, Ecological Engineering 28 (3), 271-279).

    Response: We are glad you shared these articles with us because we wanted to compare our results with other aquatic ecosystems to helpfully show our results were reasonable. An additional section has been added to section 3.5, lines 534 - 539

Reviewer #2: Alex Horne
Reviewer Summary:

This MS is of good quality with a nice blend of field and lab work as well as the theoretical/modeling approach we were aiming for with (some articles) in the new journal. It also tackles a real-world performance concern, namely the loss of performance of a FSW taking real world treated effluent. The authors should be congratulated on their diligence and results. The replication and results seem to be a good estimate of the flux of NH4 from the sediments of a static (lab work) and quiescent (?) conditions in the WWTP effluent wetland treatment cells. Replication was adequate in the lab studies and the analysis and modelling are good. The fact that the flux rates declined as the base added ammonia was increased in the microcosms indicates proof of a commonsense result. However, more context is needed to give our readers a better understanding. *In particular, the implications of more N coming out than going in is a mass balance no-no. This must be cleared up before publication*.

Response: Thank you for the review Dr. Horne. In response to your largest concern, we never meant to imply that the wetland system was a source of N over its entire life, only that while it had been mostly a sink in the first 10-15 years, then more recently there had become times during the year when the wetland became a source since the N it had accumulated over the previous years had started to release. We clarified that in multiple locations in the manuscript using the terms source and sink (e.g. line 122.)

Specific Concerns:

1. **High flux rate of NH4 in sediments (from organic detritus?) to the overlying water.**

    The flux rates measured (140 to 210 mg/m^2/d) are high but are not much higher than some other studies, for example, the large, well-studied Waco Lake Marsh, Texas (Scott et al. 2008) who report a high of 210 mg/m^2/d (assuming their hourly rate can be extrapolated to 24 hours), an average 64 mg/m^2/d. The also have some low flux rates and the odd negative result. These wetland fluxes are much higher than many others in the literature measuring sediment flux from eutrophic lake/estuarine sediments (5-84 mg/m^2/d and 15 mg/m^2/day is probably a good typical number). So, I do not question the author's work here since it looks like their sediments are very N-rich. But they do need to be put in context. I suggest that context is that their Walnut Cove wetland is (may be?) unusual in that it receives high ammonia all the time and no nitrate to speak of. Others (included in their references) made the same mistake of putting full strength 2y effluent with 25 mg/L NH4 into a wetland then then wondering why the wetland does not change anything. For a wetland to work with nitrogen removal the ammonia needs to be low since NH4 is a highly reduced molecule and wetlands have, by definition, anoxic sediments. Up front aeration is a good answer and aeration systems are very cheap and easy to work with so that would be a solution to the Walnut Cove ammonia releases.

    Response: Thank you for the comment. We have added context to the flux rates by adding values from eutrophic lakes (work by Buetel - section 3.5, lines 534 - 539).

2. Unstable long-term mass balance.

The authors comment that a reason for their work was the “poor performance of the NC treatment wetlands” and that is a good reason. However, it seems that there is always more ammonia coming out than goes into the wetland and their data shows this. If this were true in the long-term, we would not need the energy-intensive Haber-Bosh ammonia plants that dot the world today to make N-fertilizers and explosives. There cannot be more N going out than coming in on the long term, but I think the authors ignore this problem (maybe I missed something here). It seems like the inflow is mostly NH4 with almost no nitrate and an unknown amount of organic-N. The authors need to explain the problem of a positive mass balance. My own guess is that there must be times of year when the balance goes the other way (winter maybe, calm period when sediment flux is substrate inhibited?). Are there odd occasions when the inflow reaches more normal NH4 levels like 25 mg/L? I have found this for some specialized small WWTPs.

My help here is twofold, some of my own work on wetlands and a comment of the performance of the Walnut Cove WWTP. Below I show inflow and outflow averaged over 24 years for mostly treated municipal effluent entering a 119-ha constructed treatment wetland (28 cells) at Prado in Orange County CA. The ammonia entering is low (0.12 mg/L), as expected from a modern POTW, nitrate is quite high (6.1 mg/L), and organic-N low (0.39 mg/L). This compares with the author’s inflow of ammonia of 2.6 mg/L (spring) and 8 mg/L (winter). So, their wetland is not only heavily loaded with NH4 (as they comment) but is somehow storing ammonia then releasing it. I agree with the authors that the long-term increase in ammonia over 20+ years from their treatment wetlands is well-documented. Such high levels of NH4 are not good when discharged to surface waters, and puzzling if outflow total-N (mostly NH4 here it seems) is higher than the long term inflow. Long-term inflow needs to be on this figure 2 (if available) to sort out the true mass balance.

Table 1. Nitrogen dynamics (mg/L) over 24 years for Prado Wetland, Orange County, CA. The source is mostly treated POTW effluent, HRT about 5 days, area 119 ha, flow ~ $1 m^3/s$ & n is about 300 for each N-species (*source Horne et al. J. Ecol, in review*).

	Inflow	Outflow	Difference	% Removal
NH4-N	0.12	0.07	0.04	38
NO3-N	6.1	2.6	3.5	57.4
Organic-N	0.39	0.64	-0.25	-65

Response: Now that Prada dataset is impressive – if we only had that period of record AND the NO_3-N concentrations at our inlet. For some perspective, we do have a few years of measurements. We added our average inlet and outlet concentrations we did measure in two short term studies in the Introduction, lines 113-117 to communicate the only inlet/outlet data we do have (and will be the subject of a subsequent article). We have rewritten the paragraph preceding figure 2 (lines 111-122) to improve the clarity of our background site observations and study objectives.

From the historic data we have we are unable to determine if the historic trend in increasing ammonium at the outlet was a function of increasing ammonium at the inlet or increasingly poor N removal performance in the wetlands, but we strongly suspect the latter based on our period of sampling and the knowledge that the source of wastewater has not been influenced by new industry or population growth. We do suspect if they operated their aerators more frequently, that we would see more NO_3-N making its way to the wetland.

The mechanics for this internal nitrogen source would be that the first 10-15 years the wetland vegetation was a net sink for ammonium, then after years of decaying vegetation building up within the wetland and continuous inputs of ammonium to the wetland under reduced conditions, this former sink switched to a source over most of the year. We make this more clear in the manuscript.

3. Role of detritus is not clear (to me).

The authors, correctly point out that “detritus” can cause the wetland to lose volume and they quote 1-3 cm/y, which is a good proportion of their originally 30 cm deep wetland. While this is true for the first year, the organic matter gets substantially compressed otherwise their wetland would have filled in long ago. I assume by detritus they mean dead *Typha* stems which collapse into the sediments each winter. These dead stems and leaves have a very low N-content so seem unlikely to be a (large) source of NH4 later. Are the authors assuming some sequestration by the living plants then release? But if so, there would be no mean annual effect due to recycling. One explanation is inflowing NH4 from other sources. This would expand the inflow so it is somehow higher than the authors suggest and fluxes in and out of the anoxic sediments over time depending on the concentration gradient. However, no such large external NH4 sources come to my mind. N_2-fixation is unlikely here since nitrogenase is strongly repressed by ammonia at these levels, so something must be coming in. Aerial deposition will occur but unless the POTW is close to a cattle farm (feeding lot) that seems also an unlikely source of NH4 (coming in as NH3 vapor probably).

Response: The detritus in this wetland consists of dead *Typha* along with other types of decaying organic material (algae, duckweed, biofilms, etc.) and was found to contain a significant amount of nitrogen (see the added supplemental materials). We reported the N content of the detritus in the Materials and Methods section (lines 209-210). The wetland had been in operation for approximately 20 years and had accumulated approximately 30 cm of detritus across a majority of the cells. We think our wetland (sure the detritus compresses some with time) averaged about a bit over 1 cm per year to give us the 30 cm depth we observed. This is in line with the 1-3 cm/yr, obtained from the Kadlec and Wallace textbook and cited accordingly, for this wetland and likely other similar wetlands. Attached is a photo of the detritus and the depth of detritus determined during a cleanout of the cell lead by the authors. The authors are pretty sure that the main internal N source is from microbial decomposition of detritus. We can’t identify any other significant NH_4-N sources. Atm sources will be negligible, and N_2 fixation is likely small. This is the major point of this article that accumulated detritus can have a major impact on the N balance.

4. Figures.

These are hard to read. The authors should realize that the figures will be pretty small in a journal though they can be expanded online. The captions usually come out worse than the lines and data points. My suggestion is that the captions in particular need to be larger and bold and black (I suggest 14 point for the caption wording and 12 for everything else (years can be 11 point) – if you are using Times Roman. My eyes are not what they were 75 years ago and so will some of our readers be too. The color figure 1 may not show up well as shown but I just suggest they think about that. Who is going to pay for the color figure?

Response: Understood, the figure fonts have been increased.

Reviewer #3: Andrea Ludwig

Reviewer Summary:

This work aims to fill the gap in understanding the magnitude and implications of the detritus layer as an internal nitrogen source in free water surface constructed wetlands nitrogen budgets. The manuscript presents findings from a study that incorporates field data, benchtop microcosms, and resultant modeling effort that has implications for similar sites and perhaps beyond. The paper is well written, easy to follow, and has implications for ecological engineering design and practice, and therefore, is appropriate for consideration in this journal.

Response: Thank you Dr. Ludwig for your comments

General Comments:

1. The stated conclusions from the experiment match the scale and scope of the study. The authors recognized the limitations of the study and addressed them as well as tied them to future research trajectories.

   Response: Thank you for your comments

2. More detail would be beneficial to discuss the detritus substrate collection methods in the field. What was the range of mixed depths (line 169)? How was active vegetation removed (I assume, since it's not explicitly stated)? Was there any other preparatory filtering? How were they homogenized?

   Response: Methods of detritus sampling was updated in lines 204-211.

3. Throughout the text, it was hard to follow the experimental design in terms of treatments, replicates, number of microcosms, number of runs. I suggest clarifying it in the methods section (page 8) perhaps with a grid figure of the microcosms and treatments. Line 300 in the methods should be clarified.

   Response: We updated the methods and created a Figure 4 that now shows a schematic of the experimental setup

4. The target treatment of 0 mg/L NH4-N ended up with detectable levels, which I assume was part of the tap water. Some mention of this would help explain in the methods as well as including detection limits of the water chemistry analyses.

   Response: Method detection limits have been added. The method detection limits were 0.015 and 0.2 mg-N L-1 for ammonium and nitrate, respectively (Lines 254-255). We also added some reasoning in table 2 to why the NH4-N target concentrations were mostly higher (because of some mixing with the substrate during loading).

5. There is quite a bit of overlap between results/discussion/conclusions. Consider shortening results and condensing discussion.

   Response: We felt that way too, but followed the journal's initial guidance on this. The results and discussion sections have been combined which made the manuscript much better.

6. Related to the technical aspects of the study, the experimental design (treatments, replicates, and relation to real-world facts) is strong. Indeed more data points would be good, but the findings are presented in a way that discusses shortcomings and in an appropriate manner. Bringing field sampled sediments into a controlled laboratory is a good way to start the investigations into this subject. The selected methods (analytical, statistical) appear sound and on par with the current science.

   Response: Thank you for your comments

Specific Comments:

1. Line 38: Should this be 27 experiments? I believe there are 9 microcosms.

   Response: Changed to 27 experimental units

2. Line 91 (Figure 1): I don't understand the size scale here (30 cm) and associated reference in the figure title.

   Response: Figure 1 has been updated.

3. Line 112 (Figure 2): Where in the wetland is this dataset collected? Inlet, outlet, middle? I believe this is outlet.

   Response: Figure 2 title has been updated to show this is wetland outlet data.

4. Line 183: Tap water?

   Response: Yes tap water was used as source water for the experiment for mixing. This figure was moved to supplementary materials

5. Line 204 (Table 1): This is a tricky table. It is not totally clear what the numbers represent without units. The treatment titles on each column give the impression, but the overall "Treatment" title needs to be clear and include units associated with the numbers in the data rows.

   Response: Yes we missed that. Table one has been updated to include units and a better description

6. Line 299-300: This is confusing. Are there 9 replicates of each treatment? Or is it 9 mesocosms with three treatments in triplicate and thee runs?

   Response: Yes, this line has been updated for more clarity.

7. Line 346: Mineralization?

   Response: We think respiration is the appropriate term to use in this instance

8. Line 353 (Table 2): Need units for concentrations

   Response: Table 2 was updated

9. Line 456 (Table 4): To differentiate this from the target run concentrations or the measured concentrations, perhaps label this "Expected Water Column NH4-N Concentrations"

   Response: Table 4 updated

Second Review

EIC McMillan Review Comments (via email on 2023-08-28)

Thank you for submitting your revised manuscript, “Quantification of ammonium release from an aging free water surface constructed wetland to improve treatment performance”, to the Journal of Ecological Engineering Design (JEED). I am pleased to inform you that, following careful assessment of your resubmission, our editorial team would like to move forward with publishing your manuscript. Our production editor will follow up with you shortly.

Minor comments for your consideration include:

- The main revision requested was to add the theme of design recommendations. This is nicely laid out in section 3.8 and paragraph starting Line 96 about the design models. A couple additions would help frame this and elevate the design aspects, specifically:
  - Abstract: suggest that you expand the third sentence (L28-31) and the last sentence (L45-47) to include design in addition to or in lieu of management concerns,
  - End of the introduction (lines 135-137), same general comment as above for abstract, add theme of design in addition to or in lieu of management).

- Line 34 in the abstract - add "equal" to the loading of detritus to clarify that the OM was consistent across the nutrient treatments.
- Note that highlight should be, as noted in template, "Highlight: One-sentence summary clearly and concisely highlighting the key finding(s) of the study"
- Line 192: replace "mud" with "sediment".
- Lines 209 & 232: delete "muck"
- In Figure 6, change "muck" to "sediment" in Y axes labels.
- Add citation to all analytical methods (Line 197; Lines 249-255); also add detection limits.
- Define what error terms mean in section 2.4.1; several values are presented with plus/minus. What is the sample size, is this standard deviation, over what period of time? Also remove this subsection and combine with 2.4.
- Line 369: Can you clarify what "these values" refers to?
- Line 410: replace "were" with "was" (we did not thoroughly note all minor grammatical errors like this so please do a final proofread).
- Consider a more descriptive name for Section 3.4. These are statistical results but more informative for the reader to know the topic / context (i.e., what is different?). Something like "Drivers of variability among experiments".
- In lines 542-546 add in mass estimate for C* not only in kg/m2/yr but also in kg/yr (15 g/m2/yr x 7200 m2 = 108 kg/yr?) so reader can make complete comparison to study values in same units (370-550 kg/yr).
- Similar thought about title of Section 3.8 – this is a really nice addition to the influence on design and would be helpful to have the section heading reflect that.
- Perform a final proofread to catch a handful of minor typos.
- Ensure that all figures have text of readable size and that they are of high resolution.
- Reformat all in-text citations and the end references list using Council of Science Editors (CSE) Name-Year citation style. Include a DOI for all references consulted online. If a DOI is not available, provide a URL that does not require a password to access the material.
- The author is responsible for ensuring that both in-text citations and the reference list are complete and accurate, and that all DOIs/URLs are functional. Reference lists generated by citation managers are prone to error: Proofread the reference list and test all links prior to submission.

Beutel Review Comments (via email on 2023-07-29)

Hi Andrea, and thank you for the opportunity to give this manuscript a second review. It is now adequate for publication. Please pass the following recommendations on to the author. I will leave it to you as AE and EIC McMillan to confirm these changes are part of final submittal. I do not need to see the manuscript again. You might want to confirm with Aimee that references are in the right format, as now would be a good chance to ask for those edits as well.

- The main revision requested is to add the theme of design recommendations, nicely laid out in section 3.8, into the abstract (e.g., expand second sentence and last sentence to include design in addition to or in lieu of management concerns) and into the end of the introduction (lines 135-137, same general comment as above for abstract, add theme of design in addition to or in lieu of management).
- Note that highlight should be, as noted in template, "Highlight: One-sentence summary clearly and concisely highlighting the key finding(s) of the study"

- Add “JEED Original Research Paper” at top of paper as in template.
- Perform a final proofread to catch a handful of minor typos.
- Define what values mean in section 2.4.1 where plus/minus is presented. N = ? Over multiple years?, Is this standard deviation?
- In lines 542-546 add in mass estimate for C* not only in kg/m2/yr but also in kg/yr (15 g/m2/yr x 7200 m2 = 108 kg/yr?) so reader can make complete comparison to study values in same units (370-550 kg/yr).

Author response to second review

A formal response to individual comments was not requested because suggested changes were minor and editorial in nature. The authors incorporated the requested changes in the final submission.