Biogeosciences Discuss.,
https://doi.org/10.5194/bg-2019-243-AC3, 2020

Biogeosciences Discussions
Open Access
EGU

Interactive comment on "Vertical transport of sediment-associated metals and cyanobacteria by ebullition in a stratified lake" *by* Kyle Delwiche et al.

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Received and published: 18 February 2020

We would like to thank the referee for looking over this work and providing valuable critiques to our paper. The comments are thoughtful and bring up many important points, which we addressed individually below.

Anonymous Referee 4

The issues with sample collection make me call into question the quantitative results and budget. Please see my specific comments below for further details. Ultimately, the data need to be published, but the manuscript needs major revisions to remove

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the budgets which are likely inaccurate, given the sample collection procedure. Please refocus the manuscript to state the observations and cast your results in light of how the samples were collected.

We agree that the quantitative results and budget analysis are highly speculative, so the suggestion of removing the budget analysis would certainly be one way of addressing this issue. However, we propose keeping the budget calculations in the text, but making sure to emphasize the proper uncertainty associated with these budget estimates and to replace any specific estimates highlighted in the abstract or conclusions with a statement that more work is needed to calculate a proper budget for this mechanism. We hope that this approach would provide some context for the observations while remaining realistic about the fact that the information isn't at the level it needs to be for estimating a proper budget. We hope that our revisions have captured the spirit of this comment, while still providing some context to interpret our observations and to inspire future research.

Specific Comments: L 23-24: Define "problematic". What does this mean for cyanobacteria? Be more specific.

This statement was clarified as "In a 2012 national assessment, 15.2% of surveyed lakes in the U.S. were ***categorized as Most Disturbed due to the concentration of cyanobacteria, a significant increase in lakes with this categorization (8.3%, 95% confidence intervals 4.0-12.5%)*** *over the 2007 assessment (U.S. Environmental Protection Agency, 2016)."*

L 29-30: What about the "improved understanding"? What type of understanding? Be specific.

*We have changed this to be more specific as "****Identifying*** *the sources and mechanisms of transport of these substances within lake ecosystems can help predict the fate of contaminants and aid remediation efforts."*

L 110-111: How do you know the bubble transported biology and chemistry is no adhered to the inner walls of sampling equipment? Do your measurements represent an underestimate?

This is a point that was also brought up by a previous referee, so we have added a comment about this potential sampling artifact, which would underestimate transport:

"The interaction of bubbles with the flexible tubing resulted in visible particle attachment to the tubing, making our estimates of particle mass transport a lower bound."

L 172-173: Are these filter measurements meant to be volumetric? If so, do you know how much water passed through each filter before clogging?

For these filter measurements, we recorded the total volume filtered and the total mass accumulated, whether or not this was distributed over more than one filter because of clogging. Thus, we do not know the volumes passed through individual filters, only the total volume of water associated with a total particle mass.

We have amended our text to read: "Due to filter clogging, we typically used multiple filters for each ***sample, and total particulate transport per sample was calculated by summing the particle mass on each filter and dividing by the total gas volume associated with the sample.*** *"*

L181: I don't know how this relates to the accuracy and precision of your measurements? How do counts per second relate to concentration?

The relative standard deviation of the ICP-MS counts relates to the uncertainty in the measurements. The uncertainty for the sediment digests is quite low, and while it is higher in the less concentrated bubble transported particle samples, this uncertainty is still low relative to the experimental uncertainty. We have added the following line to the text: ***"These relatively low RSD values indicate that analytical uncertainty is low, especially compared experimental uncertainty."***

L 266: This is an excellent study and I think your experiments and testing shows bubbles play a role in lakes that has not been considering from a biological perspective. This study needs to be published, but I can't get over the anchor drop issue. I have thrown many anchors overboard in lakes and the plume of sediment is always significant. I have a hard time decoupling this disturbance with your results. There needs to be a paragraph describing how the laboratory results follow the lake results and the anchor had minimal impact on the lake results. Although, your laboratory results show sediment disturbance impact the bubble transported particles. How can you decouple these methodological problems with your results? What if you shift the focus of your manuscript to documenting that bubbles DO transport chemistry and biology, but stop short of the full budgets, as I think those are biased due to the methodological problems.

We agree with the reviewer that triggering bubbles with an anchor drop leads to substantially different conditions than naturally ebullition. We wish we could have collected samples from natural ebullition alone, but this would have resulted in long wait times and probable changes in the cyanobacteria population prior to sample analysis. We attempted to alleviate some of this concern by using the laboratory bubble column experiments to demonstrate that particle scavenging when bubbles rise through a plume of sediment is still a relatively minor contribution to total particle transport. However, we agree that this experiment alone cannot account for all potential effects of the anchor drop. We feel this is an excellent area for future research, either in systems with much higher ebullition rates such that natural bubbles could be used, or potentially with updated experimental apparatus that can utilize natural bubbles.

To address these concerns, we have re-worded the text in numerous areas to highlight the uncertainty while still providing context for whether these observations could substantially impact chemical cycling or cyanobacterial life cycle. Some examples include:

Abstract- " ***Although more work is needed to reduce uncertainty in budget estimates****, bubble-facilitated cyanobacterial transport* ***has the potential to contribute***

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substantially to the cyanobacteria cell recruitment to the surface of *this lake and may thus be of particular importance in large, deep, stratified lakes."*

Results- ***"These particle loadings on bubbles, and any ecosystem-wide flux estimates derived from them, must be qualified by the fact that neither triggered bubbles nor bubbles in the bubble column fully replicate natural bubbling. In particular, the triggering of bubbles with an anchor may have raised plumes of suspended sediment through which some fraction of produced bubbles had to rise, and within which the possibility of scavenging should be considered."***

" However, many questions remain regarding bubble-mediated transport in natural systems, including how the change in water density at the thermocline affects bubble rise and associated chemical and biological material."

"There remains the possibility that our measured bubble particle transport rates differ significantly from those from naturally emitted bubbles, and this remains an important area for future research. However, despite this uncertainty, broad-scale estimates of arsenic and cyanobacteria cycling can provide important context as to whether these processes may be significant in UML."

" These calculations demonstrate that bubble transported cyanobacteria could negatively impact water quality, though more research is warranted to improve these estimates."

" Using the maximum observed recruitment rate of 2.3 x 105 cells m-2 day-1 (Brunberg and Blomqvist, 2003) from sediments for the area of the lake above 12 meters, we estimate that bubbling could contribute 14 % of cyanobacterial recruitment in the lake, ***but 95% confidence intervals range from less than 0 to 46% of overall recruitment. While we cannot rule out the possibility that this is an insignificant source of cells given the large uncertainty in these measurements, the potential for bubble-mediated transport to contribute substantially to the source of cyanobacteria cells at the lake surface warrants further investigation."***

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*Conclusions- "Bubble mediated transport of cyanobacteria cells may contribute **substantially** to cellular recruitment from the sediment, **but the uncertainties in our measurements make these estimates speculative."***

L 268-270: This observation is baseless since you caused the ebullition.

The reviewer makes a good point that natural variation in ebullition has nothing to do with the variation in mass transport observed in our triggered bubbling events. We have removed this sentence.

L 277-280: This is analogous to dropping an anchor on the lake sediments. How do you reconcile these laboratory experiments with what you did in the field? Again, this is evidence the focus of the manuscript should be focused to an observation that bubbles do transport chemistry and biology, but do not calculate budgets because the evidence shows they are not accurate.

Two observations from the columns with recently disturbed sediments (similar to the anchor drop, as mentioned in the comment) are similar to those with "normal" sediment, so the impact of these disturbances creates a complicated relationship with particle transport that we can not fully understand. The combination of both measurements ("normal" and "recently disturbed") resulted in transport that were similar to one field collection date, so it is at least in a similar range to what is occurring in the field.

This comment again highlights the uncertainty in our measurements. We agree with this comment and address it by making the uncertainty in our calculations more prominent, downplaying numbers in the abstract and conclusions, but keeping the budgets for context. We have re-written the text in numerous locations to highlight sources of uncertainty (mentioned above). However, we do still see value in budget calculations, however uncertain they may be. For example, the rough budget calculations for arsenic show a several order of magnitude gap between potential bubble arsenic transport rates and other transport rates within UML, indicating that even if our estimates are biased low, they are unlikely to be high enough to matter in UML. Conversely, the

upper threshold for cyanobacteria transport in UML does fall within the realm of an important flux, which is a justification for further research in this area. We therefore think these estimates give a useful perspective, but we emphasize the large uncertainty that exists in these measurements and that the budgets are a best guess.

L 283-285: Were there particles to scavenge? This was tap water, right?

This reviewer and one other have helpfully pointed out that we were not clear about the water column conditions when we conducted our tests for particle scavenging. As discussed previously in this response, scavenging tests were done after tests where bubbles were emitted from the sediment bed, so the water column was visibly turbid and contained many suspended particles.

We have added a sentence to clarify this point: ***"We conducted the scavenging tests when the water column was visibly turbid and contained a plume of suspended particles from previous tests."***

Section 3.3 header: Again, I have a hard time reconcile the topic of this section that particles originated in the sediment after traveling through a plume of sediment. Maybe scavenging is a more active process and makes up a larger percentage of the particles when not passed through a plume of sediment.

We agree that bubble scavenging of particles within the water column could contribute to the particle burden, and thus not all particles originate in the sediment. Indeed, our scavenging tests shows that approximately 10% of the particles transported to the surface could be picked up within the relatively turbid water column. This indicates that within our experiments, a substantial fraction of the particles appear to come from the sediment bed itself. However, as pointed out previously, the artificial conditions for bubble release in both our laboratory and field experiment could influence our results. To acknowledge this uncertainty, we have changed the section title to:

"3.3 ***Bubble-transported particles have chemical and biological characteristics***

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similar to sediment *The data on bubble particle mass transport clearly shows that bubbles are capable of transporting particles from relatively deep depths, and minimal rates of particle shedding and scavenging in the water column* ***suggests*** *that these particles originate* ***primarily*** *in the sediment. "*

L 325-326: Observations like this are the reason this manuscript needs to be published.

We appreciate your support for the publication of this work. To highlight the finding of potential ephippia in the particles, we have added a reference to the specific panel in Figure S10 that may show ephippia (Fig. S10-B).

L 353-354: This is a major finding of this study and should be a highlight.

We appreciate the reviewer's enthusiasm for the content. The referenced sentence in L 353-354 speculates that since cyanobacteria overwinter in the lake sediments, bubble-mediated transport could be a mechanism of inoculating the upper water column with these cells. We believe we have highlighted this possibility with the mass budget calculations that compare potential bubble cell transport to other methods of cell recruitment. However, as discussed previously in responses to this reviewer, there remains a high degree of uncertainty around our estimated cell flux.

L 374: What does it mean to have a negative rate of transport? Are bubbles actually sequestering cells from the surface waters? This is another reason why I think the budgets need to be removed and the focus placed on the observations and laboratory experiments.

A negative transport rate is not meaningful, but is another aspect of the variability of our measurements that add uncertainty to the budgets. As discussed earlier, we agree with the reviewer that more attention should be given to the uncertain nature of our budget calculations, and have re-written portions of our text accordingly. Furthermore, we now conclude with the statement that: "Using the maximum observed recruitment rate of 2.3 x 105 cells m-2 day-1 (Brunberg and Blomqvist, 2003) from sediments for

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the area of the lake above 12 meters, we estimate that bubbling could contribute 14 % of cyanobacterial recruitment in the lake, ***but 95% confidence intervals range from less than 0 to 46% of overall recruitment. While we cannot rule out the possibility that this is an insignificant source of cells given the large uncertainty in these measurements, the potential for bubble-mediated transport to contribute substantially to the source of cyanobacteria cells at the lake surface warrants further investigation."***

L 400: Given the large errors in your bubble transport of cells, I have a hard time following how the error now is so small. The error propagation is not well explained.

This is an error, and the range of values reported comes from using both 9 meters and 12 meters as the cut-off for where cyanobacteria would be able to recruit to the surface without bubbles. We agree that this does suggest a smaller uncertainty in the final budget than is warranted from the data.

To address this and the comment from above, we propose to still include the budgets in the presentation of the data for perspective, but to better emphasize the speculative nature of these budget results and the uncertainty associated with it. This provides context for the results and motivates additional research in the future on this topic, while still being realistic about whether these transport rates are well constrained. Even with the large uncertainty in particle transport values, the arsenic transport is unlikely to be a substantial part of arsenic found in the lake surface, but bubbles could still be an important part of cyanobacteria transport.

Since there are a number of uncertainties associated with cyanobacteria transport, we can emphasize that bubble-mediated transport has the potential to be a significant source of cell recruitment, especially in deep, eutrophic lakes. However, more work is needed to better constrain these values to determine the actual contribution.

Technical Comments: L 22: Delete “are”. *Thank you for finding this glaring error in our first sentence, we have deleted the “are”.*

L 22-23: First sentence needs a citation.

We have added two references that provide an overview of how water quality is a wide-spread phenomenon that will be likely exacerbated with increases in urbanization and climate change. "Deterioration of water quality is wide-spread and expected to become more acute with increased urbanization and climate-change ***(Zhang, 2016; Paerl et al., 2011)."***

L 32-34: First sentence of the paragraph, poor sentence structure, please rewrite.

We have clarified this sentence to read:

"Because sediments are typically major repositories of contaminants (Nriagu et al., 1996; Pan and Wang, 2012; Taylor and Owens, 2009), ***it is important to understand the processes leading to contaminant mobilization."***

L 35-37: "However, transport to surface: : :" Poor sentence structure, please rewrite.

We agree that this sentence was poorly worded. We have restructured the whole paragraph to improve readability:

"Because sediments are typically major repositories of contaminants (Nriagu et al., 1996; Pan and Wang, 2012; Taylor and Owens, 2009), ***it is important to understand the processes leading to sediment mobilization****. Metals can be mobilized from sediments via solubilization by oxidation-reduction reactions, and by sediment resuspension,* ***acidification*** *or bioturbation (Calmano et al., 1993; Eggleton and Thomas, 2004; Schaller, 2014; Schindler et al., 1980).* ***Likewise, over-wintering cyanobacteria and algae concentrated in the sediments are mobilized through germination, wind-induced resuspension, or bioturbation (Ramm et al., 2017; Verspagen et al., 2004; Stahl-Delbanco and Hansson, 2002)****. In some cases, the number of resting cells in sediment can be predictive of the severity of subsequent bloom events (Anderson et al., 2005).* ***Previous research*** *showed that recruitment from sediments of the potentially toxic cyanobacterium Microcystis was a major driver of the summer bloom*

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(Verspagen et al., 2005). Cyanobacterial recruitment to surface waters from deep sediments is expected to be inhibited by stratification, low oxygen concentration, and low light levels (Ramm et al., 2017). ***Metals mobilized from sediment under stratified water columns will also be inhibited from reaching surface waters due to stratification (Wetzel, 2001).”***

45-46: “Bubbling from anoxic sediment: : :” Sentence missing numerous citations.

Thank you for bringing this to our attention, we have added the following two citations showing substantial contribution of methane bubbling to total freshwater emissions: Bastviken, D.; Tranvik, L. J.; Downing, J. A.; Crill, P. M.; EnrichPrast, A. Enrich- prast, A. Freshwater methane emissions offset the continental carbon sink. Science 2011, 331, 50−50.

Deemer, B.; Harrison, J.;Li, S.; Beaulieu, J.;DelSontro, T.; Barros, N.; Bezerra-Neto, J.; Powers, S.; Dos Santos, M.; Vonk, J. Greenhouse gas emissions from reservoir water surfaces: A new global synthesis. BioScience 2016, 66 (11), 949−964.

Citations for the ability of bubbles to transport particles are already provided in subsequent sentences detailing this process in industry and marine systems.

L 50- 53: “Bubble-mediated particle: : :” Poor sentence structure, confusing, please rewrite.

We agree this sentence was quite poorly written, and have changed it to:

“Bubble-mediated particle transport also occurs in the open ocean where bubbles are injected into the water by breaking waves, scavenge surface-active particles as they rise, and then deposit these particles on the ocean surface (Aller et al., 2005; Blanchard, 1975; Wallace et al., 1972; Liss, 1975).”

L184-185: “We filtered bubble: : :” I did not understand this sentence.

We agree this sentence is confusing, and have shortened it to say:

"We filtered bubble column samples using pre-weighed 5.0 μm and 0.2 μm Whatman Nuclepore membrane filters (47mm diameter)."

L 187: How much lower are the blanks? Actual numbers would be better. Two orders of magnitude can range from 110-fold lower to 900-fold lower. These are very different blanks.

To clarify the blank question, we have calculated that the Whatman filters contained less than a nanogram of arsenic contamination, far below the sample concentrations. For the Nucleopore membranes, the 5 μm filters had arsenic levels below the ICP-MS detection limit, and the 0.2 μm filters had 0.003 $\pm$ 0.002 μg per filter for the 0.2 μm filter (less than 1% of the arsenic found in the least concentrated sample). We have added the following text:

"Duplicate analysis of clean Nuclepore membranes (blank) was used to determine arsenic contamination of the filters and was below the detection limit for the 5 μm filters and 0.003 $\pm$ 0.002 μg per filter for the 0.2 μm filter (less than 1% of the arsenic found in the least concentrated sample). "

L 249: mL-1 gas volume or mL gas volume-1?

was changed to "mL gas volume-1"

L 250: Estimate – estimated (past tense).

Thank you, we have made this change.

L 258: Bring eq. 1 up so that the reader knows the equation before getting the variables.

We have Equation 1 to the top of the paragraph, along with a summary description of each variable to aid in readability.

Rewrite the part about the depth interval for germination. I was lost.

*We have improved the readability of this section as: "**We conservatively assumed that germination could occur to a depth of 12 meters based on typical** light, temperature, and oxygen **levels observed in** UML (Varadharajan, 2009). The fraction (Fg) of the surface area (SA = 580,000 m2) of lake above 12 meters that could support cyanobacterial recruitment through germination is**approximately** 0.50 (Varadharajan, 2009)."*

L 362: This is a concentration, not a rate.

Thank you, we have eliminated "a rate of".

L 365: Keep units consistent. Use slash or exponent throughout.

Thank you for noticing this inconsistency. We have used exponents throughout.

Please also note the supplement to this comment:
https://www.biogeosciences-discuss.net/bg-2019-243/bg-2019-243-AC3-supplement.pdf

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Interactive comment on Biogeosciences Discuss., https://doi.org/10.5194/bg-2019-243, 2019.

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