

*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 #1**

Authors state that the particles associated with the bubbles, almost entirely originated from the sediments, rather than from the water. Will this statement hold true in case of turbid waters? Please clarify.

*We do not actually know whether the sediment particles have been scavenged from the plume of sediment in the water column, or the sediment directly. We have evidence to suggest that only a small portion (~10%) seem to originate in the water column from the column experiments, but the concentration of particles in the water column could have been different between experimental conditions in the column and field. As such, more turbid waters could result in larger concentrations originating from the water column as compared to the sediment, but further work is needed to understand this difference.*

*We were also not clear about the water column conditions when we conducted our tests for particle scavenging in the experimental column. Because these tests were done after tests where bubbles were emitted from the sediment bed, the water column was visibly turbid and contained many suspended particles. We have added two sentences to clarify this point:*

*(In Methods) "Scavenging tests were conducted after particle transport tests, so the water column above the sediment bed was turbid and contained a plume of sediment particles."*

*(In Results) "We conducted the scavenging tests when the water column was visibly turbid and contained a plume of suspended particles from previous tests."*

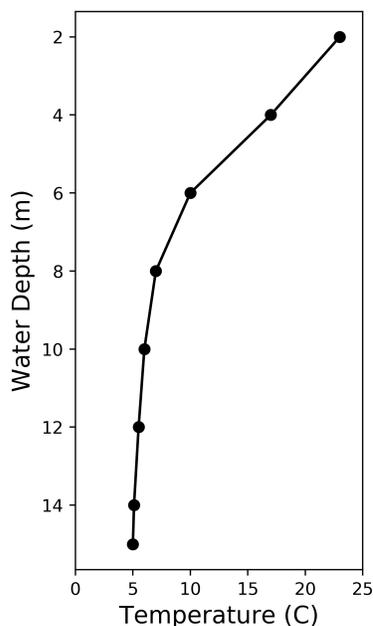
*We also add this as a possible mechanism in section 3.1:*

*"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."*

Add the details of dissolved oxygen concentration, temperature and total suspended matter in the water column at the lake sampling station.

*We have added a figure (Fig. S3) showing the temperature profile taken during the June 26, 2018 sampling event. Previous work on Upper Mystic Lake has shown that*

dissolved oxygen tracks closely with temperature (Delwiche and Hemond, 2017). We do not have a total suspended matter profile.



**Figure S3.** Water temperature profile taken during June 16, 2018 sampling event on Upper Mystic Lake.

Delwiche, K. B., and Hemond, H. F.: Methane Bubble Size Distributions, Flux, and Dissolution in a Freshwater Lake, *Environ Sci Technol*, 51, 13733-13739, <https://doi.org/10.1021/acs.est.7b04243>, 2017.

Did you observe any bubble breakup during the transport through the flexible tubing? If yes, does it affect the final bubble size count and volume transported?

*The bubble size sensor was placed below the sample cup set-up, which contained the flexible tubing, so any breakup within the tubing (which did occur) did not affect the measured size distribution. However, the size distribution could have been affected by rapid bubble flux, which can cause bubbles to coalesce within the funnel constriction leading to the bubble size sensor (as described in Delwiche et al, 2017). To address this fact, we have modified the text:*

*Anchor-triggered bubbles were significantly smaller (average diameter 5.6 mm) than those measured for natural bubbling events (average diameter 6.4 mm) during a 2016 field campaign [Fig. S7, (Delwiche and Hemond, 2017)], even though relatively high bubble flux events (such as those triggered by anchor dropping) can lead to some bubble coalescence within the funnel constriction in the bubble size sensor [ (as described previously (Delwiche and Hemond, 2017))].*

Line 114, please add the grade of HNO<sub>3</sub> used for rinsing.

*We used reagent grade HNO<sub>3</sub> for all acid washing, and have amended the text to reflect this:*

*“All sample cups were soaked in 5-10% **reagent grade** HNO<sub>3</sub> for 24 hours...”*

Authors dropped a cinderblock to trigger bubble release. Please state the difference in bubble volume during natural release and forced release.

*This information is presented in section 3.2 Triggered bubbles are smaller than natural bubbles, but both are larger than 1 mm where differences between sizes decrease, making it unlikely that their difference in size should substantially change transport.*

The impact of cinderblock on the lake floor would have re-suspended a significant amount of sediments. Does the forced release, thus suggest a much larger than natural bubble release mediated particle transport?

*We agree with the reviewer that triggering a bubble release with an anchor drop suspends a significant amount of sediments. We also wondered if this suspended sediment would artificially raise the measured rates of bubble particle transport. To address this question, we conducted the particle scavenging experiments in the bubble column, as described in section 2.3. The scavenging tests were done when the water column had significant amounts of suspended sediment from previous trials. Bubbles passing through this sediment cloud had only around 10% of the particle mass from bubbles emitted from the sediment, indicating that while particle scavenging does occur, it is relatively minor. However, we agree that anchor dropping could still influence bubble mediated particle transport, and future research is needed to assess the particle transport rates for naturally occurring bubbles.*

The collection of sediment by dredge and subsequent transport in bucket, would have resulted in the release of a significant amount of gas from the sediments. Can the authors provide the difference in the gas content of in-situ sediments and those collected by dredge and brought to the lab in a bucket?

*The gas content of the sediment was not measured, but would certainly be lower once removed from the environment by the dredge and placed into the bucket. However, the gas content of the sediment was not critical to the development of bubbles in the experimental bubble chamber. We used a syringe pump to inject gas into the sediment bed. For this reason, we did not find it critical to measure the gas content of the sediments collected in the environment.*

What was the percentage of bubbles breaking up, when striking the inverted funnel and releasing the cyanobacteria?

*As the reviewer points out, there are a number of potential experimental artifacts that could decrease the measured amount of sediment and cyanobacteria transport (including particles adhering to the sampling apparatus, as discussed earlier and now included in the manuscript). However, we have not found that bubbles break up when encountering an inverted funnel. Previous work looking at potential bubble break-up when bubbles reach the bubble sensor funnel found instead that bubble coalescence can occur when bubble flux is high enough. This coalescence relates to the reviewer's previous comment on how bubbles break up could affect size measurements, so we encourage the reviewer to see that response.*

Authors used air, instead of methane in the laboratory experiment. Will there be a difference in the particle transport by an air bubble as compared to methane bubble? Please discuss in the text.

*The composition of the air in the bubble was dramatically different between the experimental column and the field, given the origins of both gases. If the experiment was conducted at high pressure, such as in the deep ocean, this difference in gas composition in the bubble could reach a critical point where it could affect the bubbles and particle transport. However, at the pressures found within our system (both lake and column), the composition of gas is unlikely to influence bubble properties or particle transport.*

*In support of the conclusion above, using either air in the column or gas from the sediment resulted in a similar amount of particle transport per ml gas ("0.01 ± 0.006 mg/mL in the bubble column, compared to 0.01 ± 0.01 mg/mL on June 2018 in the field"). However, the differences between those amounts and the amounts measured in the field in October 2017 (0.09 ± 0.07 mg/mL) are substantial, so we do not fully understand all of the factors (potentially gas composition) that influence particle transport.*

We also added some general caveats to this approach, which would include gas composition (e.g.):

*" 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."*

*" While this variability in cell transport between column measurements and estimates of potential field transport highlights the need for continued research, it is useful to estimate the potential range of cyanobacterial transport."*

How did the authors decide the rate of injection of air into the sediments? What happened to the gases already present in the sediments when authors injected the air?

We have added the following text to the manuscript to clarify these points:

*“The bubbling rate was calibrated to achieve a relatively steady release of bubbles without substantial wait time in between. While we expect that much of the gas naturally existing within the sediment was released during sediment collection and as it was transferred to the sample bed (indeed we did not observe natural bubble release from the sediment bed prior to experimental trials), remaining gas could have been incorporated in to rising bubbles.”*

Line 266, authors did not estimate the gas reserve in the sediments. How can they infer that the lower gas volume did not indicate a smaller gas reserve?

*As you point out, we did not measure the gas reserve in the sediment, so we cannot speculate as to the cause of the lower gas volume in June 2018. We have re-framed the section to focus on the observations and avoid undue speculation:*

*“Both field and bubble column experiments demonstrate that bubbles **can** transport particles from **the sediment** to the lake surface. A positive correlation ( $p < 0.05$  level for October 2017 ( $r^2 = 0.76$ ),  $p = 0.15$  ( $r^2 = 0.38$ ) for June 2018) was found between total particle mass and gas volume in bubble traps for both field sampling campaigns (Fig. 1). The general magnitudes of particle loadings on bubbles in column experiments and on bubbles observed in triggered experiments in the field were of similar magnitude;  $0.01 \pm 0.006 \text{ mg mL}^{-1}$  in the column vs  $0.09 \pm 0.07 \text{ mg mL}^{-1}$  on October 2017 and  $0.01 \pm 0.01 \text{ mg mL}^{-1}$  on June 2018 in the field.”*

If the positioning of boat influenced the bubble release, then how can they quantify the bubble volume and associated particle transport?

*It was indeed a challenge to position the boat above the sample plume, particularly when winds blew us off course between anchor drop and bubbles reaching the surface. However, since we were interested in particle transport **per gas volume**, our results should not be affected by whether we captured all gas from a particular bubbling event.*

*We note that this sentence is now re-written in response to other comments, as mentioned above.*

Line 273, I do not agree with the comparison of experimental column release with that from the natural lake environment. As stated above the conditions in the lab were completely different than that in the lake, and thus any comparison between the two is superfluous.

*As any controlled environment will have many differences from the natural environment, we hope that you will agree that the experimental columns were within the range observed in the field, thus can be used to verify that cyanobacteria can move quickly on these bubbles. The bubble column work was necessary to test the importance of particle shedding and scavenging (something we could not test in the field), and the fact that bubble column particle transport was of similar magnitude to*

field results ( $0.01 \pm 0.006$  mg/mL in the bubble column versus  $0.09 \pm 0.07$  mg/mL and  $0.01 \pm 0.01$  mg/mL in the field) indicated that the bubble column results could inform field processes. However, to acknowledge the necessary differences between the controlled and natural environments, we have added the following text:

*"Although this is significantly higher than the measurements made in the bubble column, the conditions in the column are substantially different from the conditions in the field and the sediments used in column had been stored for 8 months, so the cyanobacteria cell concentration was 10 times less than fresh sediments. While this variability in cell transport between column measurements and estimates of potential field transport highlights the need for continued research, it is useful to estimate the potential range of cyanobacterial transport."*

Authors state a large difference in the size of natural and forced release of bubbles. Then what is the reliability of the volume and particle transport estimated by the authors?

*There is a large amount of uncertainty in amount of particle mass transported per ml of bubble volume in our measurements, which was not properly emphasized before in the manuscript. The differences in bubble size could be one aspect of this uncertainty. In response to this comment and other referee comments, we have emphasized the uncertainty in the text and removed amounts of cells or arsenic transported from the abstract. Even with these large uncertainties, we can still put our results into context by saying that we expect that this type of transport might be small compared to other inputs for arsenic, but that bubble-mediated cell transport could be a substantial part of the life cycle of cyanobacteria in this lake. This provides contexts for what should be pursued in future experiments while still emphasizing the uncertainty in our measurements. We hope that this provides better insight into the reliability of these measurements.*

Line 25, change 'Concentrations' to 'Concentration'

Line 27, change 'concentrations' to 'concentration'

*We have also changed the "A concentration of  $10^5$  cyanobacteria cells mL<sup>-1</sup> is considered to present a risk of both acute and chronic health effects (Backer, 2002), and many states, including Massachusetts, issue public health warnings for recreational water bodies when the cyanobacteria cell concentration exceeds this value."*

Line 40, modify 'et. al.' with 'et. al.'

*According to other referee comments, we have changed this sentence to "Previous research showed that recruitment..."*

Line 48, insert space after 2008;

*It seems that many of the references required spaces to separate them. This has been addressed here and in many other instances in the text.*

Line 71, change 'volumes' to 'volume'

*This has been changed.*

Line 74, change 'greatest' to 'a considerable'

*We agree that removing greatest is advisable, but tried to improve the sentence structure with the following "This potential transport pathway could be relatively **more important** for metal and cyanobacteria transport in eutrophic, deep, stratified lakes, such as UML."*

Line 79, change 'distribution' to 'distribution'

*This "s" has been removed from "distribution".*

Line 119, change 'mixing from of the' to 'mixing from the'

*This has been changed to "preventing mixing **of** sediment to the surface"*

Line 123, change 'an' to 'a'

*This has been changed.*

Line 148, change 'column is comprised' to 'column comprised'

*This has been changed to "The column **is composed of** four section..."*

Line 176, change 'um' to '\_m'

*This has been changed.*

Line 180, change 'metals analysis on bulk sediment' to 'metal analysis in bulk sediment'

*This has been changed.*

Line 185, change 'which use' to 'with use'

*This part of the sentence has been removed.*

Line 186, change 'analysis on' to 'analysis of'

*This was changed.*

Line 188, 5  $\mu$ mol filter? Is it correct?

*umol was not correct and we changed to 5  $\mu$ m.*