

*We would like to thank Dr. Dileep Kumar 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.*

#### Abstract

Line 9: 'Bubbles adsorb and transport particulate matter both in industrial and marine systems' – include lakes systems here.

*We agree that this opening sentence falls short of encompassing the full scope of the impact of bubbles. Industrial and marine systems have been the primary focus of the research, but this phenomenon applies more broadly, and certainly applies to lakes as we show in the manuscript. We adjusted the opening sentence to provide a broader significance to the work.*

*"Bubbles adsorb and transport particulate matter **in a variety of natural and engineered settings, including industrial, freshwater, and marine systems.**"*

Line 9-12: 'methane-containing bubbles emitted from anoxic sediments are found extensively in aquatic ecosystems' – the word "extensively" is inappropriate for marine systems since methane-containing bubbles can only be found in a few select coastal ecosystems. However, this issue assumes greater and global significance in vertical transportation of dissolved and particulate materials scavenged across a few meters below to sea surface by the rising wind-induced bubbles, particularly in shallow marginal systems.

*Thank you for this comment. We changed the sentence to reflect that methane containing bubbles would be a particular concern in freshwater systems such as lakes, reservoirs, and wetlands ("are found **widely in freshwater ecosystems**"), and also broadened the first sentence to bring in more of the global significance of bubble-mediated transport (above).*

#### Introduction

Lines 34-35: 'Metals can be mobilized from sediments via solubilization by oxidation reduction reactions, and by sediment resuspension or bioturbation' – mobilization can also occur through acidification of lakes.

*Thank you for bringing this omission to our attention. We have added acidification to the list of mechanisms.*

*"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**). "*

Line 35-36: 'transport to surface waters of contaminants mobilized from the sediment is affected by lake hydrodynamic conditions, notably stratification' – an interesting question to the current investigation be how does stratification influence

methane bubble rising to surface during minimal wind induced turbulent conditions? A strongly stratified upper water column will inhibit (slow down speed of rising) or even prevent particularly small sized but proportionately with large surface areas from rising across the strong pycnocline. This is possible if vertical profiling is done with close intervals of sampling to find density gradients across the pycnocline and assessing the bubble rise rates in hypo- and epilimnion layers.

*The impact of stratification on bubble rise is an interesting question to both methane-emission from lakes and bacterial or chemical transport that should be addressed in future work. If stratification does prevent small bubbles from penetrating, bubbles may be an additional mechanism concentrating organisms or chemicals at these interfaces, resulting in the thin layers of organisms that can often be seen within the water column. However, the changes in density across naturally occurring pycnoclines might be gradual enough and the bubbles buoyant enough pass through it without bursting or accumulating. The proposed vertical profiling experiment would make a very good follow-up study.*

*We have also added a discussion of these questions*

*"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."*

Lines 40-41: 'Verspagen et. al. (2005) showed that recruitment from sediments of the potentially toxic cyanobacterium *Microcystis* was a major driver of the summer bloom Verspagen et al., 2005)' – referenced twice in the same sentence!

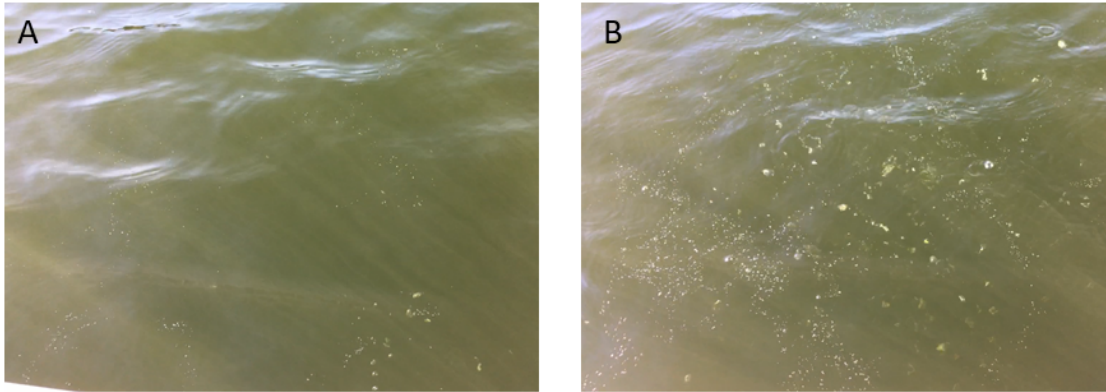
*Thank you for pointing out this redundancy. It has been changed to "Previous research showed .."*

Lines 66-67: 'the full extent of bubble particle flotation in aquatic systems remains unknown.' – even the present manuscript cannot make it 'full', which requires many attempts by many investigators!

*We agree we cannot hope to determine the full extent of bubble particle flotation with this study, and have removed full from this sentence.*

Line 70: Fig. S1 should show pictures before and after the bubble event to highlighting the emergence of particles following the bubbling.

*We have changed Figure S1 to show the water surface near the beginning of a bubble triggering event, as well as at the end. This highlights the visible accumulation of particulate matter on the water surface.*



**Figure S1.** Picture of the lake surface near the beginning (A) and end (B) of a triggered bubble event at 15 m depth showing an accumulation of particulate matter (visible as light specks on the water surface).

Lines 78-80: 'Given the expected importance of both bubble size and total bubble volume, we used a bubble size sensor (Delwiche et al., 2015; Delwiche and Hemond, 2017) to measure bubble diameter distributions both in the lake and in the laboratory.' – adsorption or scavenging of particles by bubbles is expected to be proportional to the surface area of the bubble (similar to metal adsorption on to a particle) and therefore representing bubble characteristic in terms of 'surface area' than its 'size' or 'volume' would be preferable.

*The surface area is an important bubble characteristic for transport along with other key characteristics, but typically the bubble diameter is provided as a key metric of bubble characteristics. We have rephrased this as. "Given the expected importance of bubble size on key characteristics (e.g. surface area, buoyancy, diffusion of gas), we used a bubble size sensor (Delwiche et al., 2015; Delwiche and Hemond, 2017) to measure bubble diameter distribution both in the lake and in the laboratory."*

Methods

Lines 101 and 250: 'another lake' – please name the lakes.

*The lakes referenced as "other lakes" in the text are **Lake Scharmützelsee and Lake Limmaren**, which have been explicitly stated in the text.*

Lines 111-112: 'All bubbles rising through the bubble size sensor or collection funnel entered the flexible tubing and rose into the sample cup.' – as the particles and the associated substances are adsorptive in nature it is likely that some of the rising bubble attached particles are adsorbed in the flexible tubing etc. before they reached sample cup. Authors may include a statement on this possible loss of particles during sample processing.

*The reviewer makes a valid point, transported particles were indeed adsorptive and some stuck to the sample tubing. We have added a statement to introduce this possible sample processing artifact. "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"*

Line 117: Word 'approx.' may not be necessary as the coordinates are specified to third decimal.

*This was removed.*

Lines 119-120: 'preventing mixing from of the sediment to the surface.' – requires rephrasing.

*We have rephrased as "preventing mixing of sediment to the surface"*

Lines 121-124: Good strategy.

*We found sediment contamination within the collection cups that were deployed for much longer periods capturing natural bubble events, but the possibility of contamination and subsequent growth, death or decay of transported cells made it impossible to have reliable estimates from this method. While there are some issues with this approach, it made the measurements of particle transport feasible.*

Line 250: Please correct the flux units 'cells m<sup>-2</sup>' to cells m<sup>-2</sup> d<sup>-1</sup>.

*This error was corrected.*

## Results and Discussion

Lines 262-263: 'demonstrate that bubbles transport particles from depths of at least 15 m to the lake surface.' – It may be revised as "demonstrate that bubbles transport particles from depths to the lake surface" since bubbles if formed even in deeper waters can transport materials to surface.

*This statement is confusing, and was revised to " Both field and bubble column experiments demonstrate that bubbles can transport particles from the sediment to the lake surface. "*

Line 307: Lines 134-135 mention 'On 26 June 2018 we sampled for cyanobacteria bubble transport using similar procedures, except we used a simple inverted funnel instead of a custom bubble size sensor to intercept rising bubbles' whereas Fig. S7 caption shows "Frequency distribution numbers are approximate because the bubble size sensor is unable to measure fast bubble flux or very small bubbles" – It is important to check the compatibility between these statements, particularly for data of 26 June 2018 if used.

The data from Fig. S8 is only from the column experiments, not collected during the 26 June 2018 sampling. Fig S7 shows the frequency of bubble diameter naturally occurring (from previous work) and triggered (this work, not for the 26 June 2018 date) only collected during the October 2017 sampling event, where cyanobacteria were not measured.

To clarify the difference between Fig. S7 and Fig. S8, and to further clarify the figures themselves, we have updated the figure captions to read:

**Figure S7.** Frequency of occurrence of bubble diameter during triggered (purple) and natural (gray) events *in Upper Mystic Lake*. Mean (black lines) and standard deviation (shaded regions) for each event type. Bubbles were triggered by dropping an anchor multiple times during the October 2017 sampling event, while natural bubble size distribution are based on continuous measurements from the summer of 2015 and 2016 (Delwiche and Hemond, 2017). *Frequency distribution numbers for the triggered bubbles are approximate because the bubble size sensor is unable to measure the rapid bubble flux that sometimes occurred with anchor-triggered bubble events.*

**Figure S8.** Frequency of bubble *diameter* observed *across multiple trials (a-k)* during the cyanobacteria experiment *in the laboratory bubble column*. Some *trials* had a bimodal *diameter* distribution. Panels *f* and *k* represent trials where air was bubbled directly *above the sediment*, *and remaining panels represent trials where air was bubbled into the sediment*. Note the different y-axis scales.

Line 317: replace ug with \_g.  
*This mistake was corrected.*

Lines 342-343: Besides ‘a significant fraction of the arsenic input to epilimnetic waters can be attributed to inflow from the Aberjona River (Hemond, 1995)’ aerial transport of dust associated arsenic/metals should be invoked here to be among the unknown inputs.

*We have not properly accounted for all other forms of input of metals to the lake by restricting the next sentence to just surface water input, so we have adjusted that to include atmospheric deposition with "However, bubble-mediated fluxes of arsenic or other sediment-borne metals may represent a larger fraction of epilimnetic input in other lakes having lower influx rates from surface water inflow **or other external sources, such as atmospheric deposition (Csavina et al., 2012).**"*

Lines 361-362: ‘Bubble-transported particulate matter contained cells at a rate of approximately 30 cells mL<sup>-1</sup> gas, indicating that bubbles are capable of transporting cyanobacteria through’ – May be revised as “Bubble-transported particulate matter contained cells at approximately 30 cells mL<sup>-1</sup> gas, indicating that bubbles are capable of transporting cyanobacteria through”. A ‘rate’ is expected to be material transferred during a specific duration (time). \*\*\*

*Thank you for identifying this error. It has been changed as suggested.*