

Interactive comment on “Does ocean acidification induce an upward flux of marine aggregates?” by X. Mari

Anonymous Referee #1

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General comments: This study investigates the effect of seawater acidification on aggregation and sinking of transparent exopolymer particles (TEP). Natural seawater filtered through 0.2 μm polycarbonate filters was acidified with sulfuric acid (ΔpH of 0.2, 0.4, and 0.8 units). After addition of 6 μm diameter microspheres (beads) the samples were exposed to small-scale turbulence of three different intensities generated by an oscillating grid. TEP size spectra and the adherence of beads were examined microscopically following Alcian Blue staining.

Seawater acidification caused an increase in the size and abundance of TEP. A 100-fold difference in shear stress did not have any detectable effect on TEP formation and bead adhesion. A decrease in sinking velocity and an increase in ascending velocity in response to the acidification treatment was deduced from changes in TEP and bead

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concentrations with time in the top and bottom layer of tissue flask. Implications of these results with respect to ocean acidification and the ocean's biological pump are discussed.

While this study addresses a potentially important and timely issue, the effect of ocean acidification on TEP formation, aggregation and sinking, the experimental approaches used to determine the pH/CO₂ sensitivities of these processes raise some questions:

1. Significant differences between TEP size and abundance were obtained between control and pH treatments, however, no difference was observed between the different pH levels, despite a three-fold difference in acid addition. This suggests that the observed effect may be in response to the acid treatment itself rather than the level of acid-induced changes in the carbonate system. Considering that seawater acidification was achieved through the addition of concentrated H₂SO₄, a transient drastic pH perturbations likely occurred in the sample (or parts of it) upon acid addition. The effect of this on the properties of TEP precursors may have been much stronger than that of the comparatively moderate pH change sustaining after equilibration of the seawater carbonate system. To ensure that the observed responses are not an artifact of the short-term severe pH perturbation, it would have been worthwhile to apply alternative modes of perturbing the carbonate system (e.g. diluting the concentrated acid with organic-free seawater before addition to the samples, applying acid addition of lower molarity or addition of CO₂ saturated seawater).

2. Sinking and ascending rates were measured in 200 ml tissue flasks through sampling of the top and bottom layers after 1, 2, 3, and 4, assuming an initially homogenous sample. According to the author's description, this approach was inappropriate to provide representative samples of the sinking and ascending TEP. As stated on p. 1640, lines 8-10: "...particles 'escape' the surface sampling ... by adhering to the surface microlayer (SML)". And further: "The sampling procedure does not seem to recover satisfactorily the TEP and beads that are likely accumulating in the SML" (p. 1640, lines 12-14). To circumvent the apparent methodological failure, the author modified

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the data analysis for calculating ascending rates (p. 1640, lines 24-28): "Since the surface sampling apparently failed to detect the surface accumulation of particles, the upward velocity in the pH treatments was calculated from changes in TEP and beads concentrations occurring in the bottom layer during the first hour." Obviously, sampling of the middle layer (missing in this analysis) would have helped to clarify the inconsistency between expected and measured surface layer concentrations. Without this measurement, it is not possible to verify the assumption that the missing material accumulated in the SML. Given the importance of this assumption to the data interpretation, I am surprised that no attempt was taken to analyze the surface microlayer for TEP and bead accumulations. In essence, because this approach relies on some critical but untested assumptions, I think it is not an appropriate method to estimate particle ascending rates.

Some questions also arise regarding the extrapolation of the experimental results to the natural system:

1. A change in seawater pH of 0.2 units is well in the range of pH variability naturally occurring for example during the seasonal cycle in temperate to high latitude regions or during upwelling events. Is there any field evidence that pH changes of this magnitude influence TEP production, aggregation or sinking (ascending) in areas where pH changes of this magnitude occur naturally?
2. Because the maximum effect on TEP formation and aggregation was observed already at a pH decrease of 0.2 units, shouldn't one expect the change in surface ocean pH of 0.12 units since pre-industrial times already have affected these processes? Is there any indication in long-term data sets (e.g. from the time series stations BATS, HOTS, ESTOC) to support this?
3. Given i) the methodological uncertainty (perturbation with concentrated sulfuric acid), ii) the untested assumption about accumulation of TEP and beads in the SML), iii) the surprising finding that a three-fold difference in acidification resulted in identical

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changes in TEP abundance, size and adherence of beads, is it justified to extrapolate these results to infer about possible future changes in the ocean's biological pump? Personally, I think the data interpretation and its extrapolation to the 'real world' requires a more critical evaluation of the results.

In summary, I cannot recommend this manuscript for publication in Biogeosciences in its present form.

Minor comments: 1. P. 1632, line 1 of the abstract: Change to "absorption" See also p. 1634, line 5. 2. P. 1633, lines 2-5, p. 1634, lines 7-9: At present there is no evidence that the biological pump is contributing to fossil fuel CO₂ sequestration in the ocean. 3. P. 1633, line 10: Projected anthropogenic ocean acidification is not expected to exceed a pH decrease of 0.8-1 pH unit over the next few hundred years. 4. P. 1636, line 1: Give normality of H₂SO₄. 5. P. 1641, lines 12-14: Doesn't this statement contradict some of the results reported in the previous section? 6. P. 1643, first paragraph: I don't see how the results of this study specifically relate to the SOIREE iron fertilization experiment. If TEP production indeed reduces particle sinking, shouldn't this be a typical phenomenon after any regular

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