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Interactive comment on "Particle-associated dissolved elemental fluxes: revising the stochiometry of mixed layer export" *by* A. N. Antia

A. N. Antia

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Response to Reviewer 2

This reviewer brings up the important consideration of where the excess dissolved concentrations in cup supernatants originate from, and provides a framework for estimating the relative contributions of each. The issue is relevant for the avoidance of or compensation for solubilization in traps deployed for short periods, possibly with unpoisoned cups. I agree with this reviewer that the emphasis on aggregate pore water as the sole source of dissolved elements in supernatent, as originally put forward, presents an incomplete picture. This issue is brought up, indirectly, by the first review as well and other responses I have got to the paper. Several passages have been altered in the manuscript to acknowledge the possible different sources of dissolved particles, specifically the following:

The first line of the abstract has been altered to read: "Sinking particles, once caught in

sediment trap jars, release dissolved elements into the overlying water through leaching from their pore waters, chemical dissolution and the activity of free exoenzymes"

The citation of rapid discussion of pore waters on p. 284 that the reviewer points to has been deleted, and the studies cited now reads: " Nonetheless, where the rate of remineralisation by aggregate bacteria exceeds uptake of substrate and loss through outward dissolution, as found by Smith et al. (1992), pore waters of aggregate are replenished faster than depleted."

The Discussion now includes the following: "The presence of excess dissolved material in the trap supernatent water is the most direct evidence that particles settled within the cups are altered and solubilized during storage. No matter how these excess dissolved concentrations result: from continual particle degradation, passive leaching of aggregate pore fluids, through gradual physical breakdown of dead membranes and organic entities, or through chemical dissolution such as for silica and calcium carbonate, a correct estimation of the flux caught by the trap requires correcting for this artifact. Determining the contribution of each of these processes to net solubilization is however important in considering traps deployed in differing environments with different conditions of sample storage, treatment and analyses. The analyses here is restricted to mid-water traps with low swimmer abundances, where long-term deployments and storage of samples allows a straightforward determination of excess elements in supernatent. Shallow deployments for merely hours to days in high-swimmer environments would require a different approach, since swimmers are a major source of dissolved elements in trap samples."

"Aside from passive leaching out of large particles, other processes such as continued microbial degradation, the activity of free hydrolytic enzymes, physical destruction of dead organisms and damaged membranes and chemical dissolution could also contribute to the excess concentrations of dissolved elements measured. Particle breakdown by microbial activity is assumed to be stopped by the addition of poison to the cup solution, but this may not be instantaneous. Although at the concentration of mercuric

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chloride used in this study Lee et al. (1992) report cessation of microbial activity in a concentrated particle suspension, penetration of the poison into aggregates would be rate-limited by the diffusion coefficient applicable. There is little data on the diffusion constants applicable for aggregates, but these are likely to be considerably slower than for single cells due to the fractal, mucous/gel structure of organic aggregates. Brzezinski et al. (1987) estimate 2 orders of magnitude slower diffusivity rates for silica from diatom aggregates than those for elemental silica through a permeous membrane. This considerably slower transport for solutes into aggregates could result in some continuation of microbial degradation before poison concentrations in the aggregates are effective. Free enzymes such as phosphatases and proteases are also concentrated in aggregates and though their activity is strongly inhibited by both mercuric chloride and formaldehyde, at the concentrations used to poison trap samples (Christian Karl, 1995; Liu et al., 1997), the same time constraints on penetration of poison as discussed above would apply. In as far as phosphatases degrade larger dissolved organic entities to phosphate and low molecular weight organic material, they would contribute to alteration and not production of dissolved species."

I think this gives credit to the comments of the reviewer and acknowledges the important point brought up, that numerous processes contribute to excess dissolved elements in the sediment trap jars. Thanks for contributing to improving the focus of the paper!

Interactive comment on Biogeosciences Discussions, 2, 275, 2005.