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Interactive comment on "Rising bubbles as mechanism for scavenging and aerosolization of diatoms" *by* Roman Marks et al.

Anonymous Referee #2

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The manuscript presents some interesting postulations on the mechanisms for the scavenging of diatomaceous material by rising bubbles and the aerosolisation of scavenged material on bubble-bursting at the sea-air interface. I am reviewing this from the perspective of marine atmospheric science, where there is a substantial body of literature concerned with the bubble-mediated production of seaspray aerosol. Convergence of interpretations of evidence in this field will be extremely important in resolving outstanding mechanistic uncertainties and the current work aims to provide mechanistic insight into this debate. However, as it is written, I have some general problems in understanding the concepts and implications of this work and there are quite a few points that must be addressed before the manuscript is publishable.

Context: First, I have a couple of points about the context of the work. Much atmo-

spheric work has focussed on the enrichment of organic material in seaspray particles. Owing to the much greater number of particles in sizes from around one to a few hundred nm and the importance of these as potential cloud nuclei, much of the work has concerned characterisation of organic material in these particles in terms of its broad chemical functionality and influence on water uptake in the moist atmosphere (O'Dowd et al., 2004; Wang et al., 2017). As recognised at the top of section 1.3 in the current manuscript, primary marine particles in these size ranges are from the "film" mode, formed when the bubble cap shatters on bursting, not from the larger sized, but less abundant, "jet" mode of particles. It has been shown that enrichment of diatomaceous exudate in this film mode is of more atmospheric significance than that of organic material of other phytoplankton origin (Fuentes et al. 2010), but there has been less recent atmospheric focus on aerosolised intact diatoms since they will generally be too large to be found in the abundant film particle mode and will not thereby influence liquid cloud droplets. However, there has been recent interest in the potential roles of diatoms as the much scarcer ice nucleating particles (Wilson et al., 2015). For this reason (and for mass budgeting and biogeochemical cycling purposes, as recognised in the few recent studies in this area), mechanisms for their atmospheric aerosolisation will be important (particularly under low wind speed conditions relevant for the Arctic atmosphere). I believe these contextual considerations are important to set the scene for the current work. However, once the atmospheric context is appreciated, the relevance of enrichment of diatoms in droplets of 120 microns diameter becomes apparent and the likelihood of such enrichments playing a role on atmospherically important timescales must be seen as negligible.

Abstract: It is not clear to me that the abstract serves the usual purpose of an abstract. Almost all the text is background information that does not summarise the findings of the paper and refers to previous literature (mainly from the first author). Indeed, the only part of the abstract that relates specifically to the paper findings is the last sentence.

Introduction: I found this rather meandering and not all directly relevant to the current work. The last paragraph of section 1.3 outlines the findings of previous experiments on the dynamics of bacterial scavenging by rising bubbles and states that the novelty of the current study is "...the bubble mediated scavenge [sic] and aerosolization of bio-molecules with special attention given to diatoms".

Methodology: I have a number of queries about the experimental procedure. i) what is the air pump flow rate? This would be useful to understand the scavenging dynamics. 1 sccm would be 1.667x10ˆ-5 L/s. Each 1.2 mm diameter bubble is 0.905x10ˆ-6 L, so this would be 18.42 bubbles per second (@ 1 sccm), so 3 seconds of bubbles with 7 jet drops per bubble would be 387 droplets collected at this flow rate. At a droplet diameter of 0.12 mm, this is 3.5 x 10ˆ-7 L = 0.35 microlitre. ii) how was the bubble size measured (optically or acoustically)? iii) how was the droplet size measured? iv) was the RH actively controlled to ensure the water evaporation rate was constant for all experiments and hence the diatom concentration was being measured at the same water activity (and hence normalised to ionic strength)

Given the 150 ml volume, I do not understand how sampling roughly of the order of 1/3 microlitre in 3 seconds can substantially lead to "both diatoms concentrations in the water suspension and water salinity (or more precisely the availability of cations) decrease during aeration". Obviously the air pump flow rate could be very much higher, but then this would lead to many thousands of droplets on the slides.

Consequently, I am not sure of the statement on lines 194-196: "after analyzing that set of data we noticed that the efficiency of diatom aerosolization increased, indicating that rising bubbles reduced concentrations of diatoms (solution was gradually cleaned) during the experiments."

Results: My most significant concern is with the presentation of the results - or more precisely, the lack of systematic presentation of results. The results should be detailed along with errors and statistical distributions to support the hypotheses. The descrip-

tions of the results is hard to follow as it is currently written, since it is frequently mixed with discussion material (e.g. lines 219-222, 230-232, 240-247). When this discussion is removed, it become apparent that there are few results presented. Table 1 refers to very few droplets, from which I cannot understand the methodology (number of repeats, flow rates and sampling strategy etc...) Moreover, many of the the discussion points are either incorrect of logical non-sequiturs (e.g. line 239 - there is no reason for an inability of bubbles to scavenge diatoms to lead to a reduction in jet droplets and line 258 - there is no reason that an enrichment factor indicates that diatoms were from the bubble bottom layer, though they may have been).

I am not sure that I follow the attempt to distinguish the initial and secondary droplets through the ejection heights as outlined in lines 253-257 when it is clearly stated that there was "not exclusively top jet droplets were enriched by diatoms" on 219 and "Conducted screening showed that only 20-25% of jet droplets were enriched by diatoms, which suggests that the process of diatom aerosolization might be influenced by a combination of factors operative in the water column, or at air-water interface". I would have liked to have seen some statistical analysis of the distribution of enrichments with height (for example) to support the mechanistic contentions and postulations.

Discussion & Conclusions: The discussions section presents some interesting conjecture, but does little to draw on and add to the results of the paper. Furthermore, I do not believe that the authors have presented results that can be evaluated sufficiently rigorously to support the stated conclusions.

Minor The sentence starting on line 48 reads peculiarly. The statement "generated at wind velocity 8 m/s" makes the sentence appear to be a non-sequitur, since the previous sentence states simply that the lume depth is the same as the wave height (with no wind speed dependence). If this is simply additional information, the sentence can straightforwardly be rephrased. The second phrase needs further punctuation "while, during rain," line 66 "Cl/Na atomic mass..." add "RATIO"

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