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

Anonymous Referee #1

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The manuscript entitled "Rising bubbles as mechanism for scavenging and aerosolization of diatoms" discusses experimental studies of the enrichment of diatoms in jet droplets produced by bursting bubbles in laboratory experiments. The basic finding of the study indicates that diatoms are highly enriched in bubble bursting aerosol (also called sea spray aerosol). This simple finding is reasonable and aligns with prior studies of similar marine microbiota, although this reviewer finds various problems with the mechanistic explanations, including various possible simpler explanations that are equally valid based on what is known about bursting bubbles in the literature, and are noted in the comments below.

Experimental data are not presented in the manuscript, except for certain broad statistics in basic tables – some of which are highly qualitative. The figures entirely consist

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of cartoons or exemplary images. The findings of this study should be provided in such a way that the reader (and reviewer) can evaluate the data, assess its quality, and use the manuscript as an aid when attempting to reproduce the findings at some time in the future. Based on the current form of the manuscript, this is not possible, and the manuscript should certainly not be accepted for publication until this is corrected.

It is recommended that this manuscript should be considered for publication in Ocean Science only after very major revisions are made to nearly all aspects of the paper.

The introduction could be substantially simplified. It seems to ramble about various aspects of bubble physics, not all of which are related to the focused finding of this paper. It is the recommendation of this reviewer that substantial revisions be made to this section overall.

Line 102: "...no bubble related research has focused on diatom scavenging [sic]..." Atmospheric chemists have been increasingly interested in this topic over the past 5-10 years. Images of phytoplankton and their fragments in sea spray aerosol samples can be found in the literature (e.g., Bigg and Leck, Tellus B 2005; Patterson et al., ACS Cent. Sci. 2016; Lee et al., J. Phys. Chem A 2015).

The initial/sub-initial droplets generated in this study are very large and will sediment quickly, rendering their atmospheric relevance to be minimal. Table 1 shows that the secondary drops are also d = 0.12 mm. Were these measured or deduced based on bubble diameter? In a recent study (Wang et al., Proc. Nat. Acad. Sci. 2017), secondary droplets were proposed to be smaller than the initial/sub-initial jet droplets. The aforementioned study by Wang et al. also provides a detailed discussion of sea spray production and its charge distribution from an aerosol perspective, and may be of general utility in the discussion of the results of the present study.

Line 191: "Availability of cations decreases during aeration" Please provide a citation for this assertion.

Line 198: Please expand on the manner in which the electric charge distribution around rising bubbles were collected. The explanation is inadequate to allow a reader to reproduce the measurement.

Line 237: "Bubbles rising in more concentrated diatom suspensions produced fewer jet droplets" The conclusion given is that the rising bubble had a limited ability to interact with the diatoms. The problem with this argument is that the observation in question is a change in total jet droplet production, not diatom-included droplets. A reduction in diatom inclusion may not drive a reduction in droplet production – but the converse may be true! A simpler, and equally valid, explanation is that jet droplet production was suppressed by a change in the composition of the water (and likely its physicochemical properties, like surface tension and viscosity), which also changed the mechanics of diatom inclusion in the initial/sub-initial droplet.

Line 240: It is difficult to conceptualize the relationship between bubble rise speed and the kinetic energy of the aerosol resulting from the bursting event. These are more likely separate processes, where the kinetic energy in the bubble rise is dissipated into the interfacial tension of the fluid at the air sea interface, and/or translational/rotational degrees of freedom of the bubble itself that are not related to the fluid fragmentation process of the bubble bursting event. Bubbles may reside at the water surface for a time before bursting, for lengths of time that depend on various parameters. The authors' explanation is difficult to verify based on the given information. In addition, the ability of a bubble to eject a cell is likely not limited by the energy required for ejection. The density of a cell is very close to that of the surrounding seawater (while it is acknowledged that cells are known to adjust their density to control buoyancy). The energy required to eject a diatom-free droplet is therefore likely very similar to the energy required to eject a diatom-laden droplet, although physical quantities to describe this difference are not known to this reviewer.

Suppression of jet droplet production has been shown in seawater enriched in diatoms and their associated dissolved organic matter when foams form on the water surface (e.g., King et al., Environ. Sci. Technol. 2013; Collins et al., Atmos Meas Tech 2014). It is important to consider the diatoms while including the dissolved organic carbon that they produce routinely. This comment also relates to the discussion on lines 328-338.

Figure 4: The conceptual framework of this figure is confusing. Consideration is not given to the hydrodynamic features of the diatoms themselves, yet such consideration is given to the much smaller ionic species. An explanation of diatoms existing at the base of a droplet may be associated with hydrodynamic drag during bubble ascent, where the larger particles migrate to the base of the bubble. More importantly, Phase C seems to suggest that the bubble bursting event is driven by the production of a cationic jet prior to film cap rupture. Such a concept is inconsistent with the known mechanism for bubble bursting. The detailed treatment by Lhuisseur and Villermaux (J. Fluid. Mech. 2012) [along with others prior and since] shows that the formation of the jet is related to the collapse of the bubble cavity following film cap rupture. This figure and associated conceptual model is therefore extremely confusing at best and may include fundamental flaws.

Table 1: If the initial/sub-initial droplets and secondary droplets are distinguished by ejection height, how was a lack of enrichment factor derived for secondary droplets. Any collections at low ejection heights would necessarily require the collection of initial/sub-initial droplets as well by consequence. Please clarify the method used to derive these quantities and provide data in plots and/or in entirety as supplemental information.

Table 2 shows that the range of the number of diatoms existing in enriched droplets widens by a factor of 2 for the more concentrated sample. A listing of the statistical parameters of the distribution of observations from these experiments would substantially improve the clarity of these results in the context of the discussion within the text. Providing a histogram of observations (e.g., a frequency distribution of observing 'N diatoms per droplet') would help mightily.

Line 357-363: This conceptual framework (paired with Figure 4) may be inconsistent with the current paradigm based on detailed observations by multiple investigators – or the explanation may be unclear. A greater defense of the proposed mechanism should be provided and/or the authors' argument should be presented in light of the current evidence-based understanding of the bubble bursting mechanism in which the basal jet is formed during bubble cavity collapse, and not associated with the bubble stopping as it approaches the (bulk) air-water interface.

Line 365: In order to support this argument, please cite evidence to suggest that diatom cells are more dense than seawater and bacteria cells.

Line 367: Diatoms are indeed larger than bacteria, and therefore are more subjected to hydrodynamic drag as bubbles ascend through the water column. This simple argument (as discussed above) would provide an equally valid case for inclusion of diatoms in the initial/sub-initial droplets that are derived from the jet. It is important not to forget that the concentration of bacteria in the ocean also dwarfs the number concentration of diatoms by multiple orders of magnitude, so the statistical likelihood of finding a bacterium in a droplet is simply greater.

Line 374-376: Can the data that support this statement be shown in the manuscript or the supplement?

Line 403-410: This discussion is off topic and confusing. Define a 'spiraling chain of cations'. What sort of cations are involved? This is particularly hard to envision with inorganic ions. How is this related to the topic of the manuscript? Where are the DNA and RNA bases coming from? This paragraph could be easily omitted without a loss of context or substantive discussion of the results. In general, the discussion of the cationic vortex and related items are largely irrelevant to the stated motivation of the study. The discussion of this topic is also confusing and not well buttressed by evidence. It is interesting to think about the charge separation that reportedly occurs in the rising bubble, but the deeper mechanistic discussion strays from the important

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point.

Line 446-453: Much greater detail on the methods and findings (including visualized data!) that reflect the description in this paragraph should be included in the manuscript.

Line 507: The conclusions relating to differences in concentration should be tempered, as the mechanism may not be well explained or explored by this study, and only two concentrations were attempted.

Line 513: The claim in the last sentence is almost entirely unrelated to the study at hand, and at the same time, is a significant stretch in logic. Prebiotic chemistry is, indeed, an exciting field, but the simple fact that a vortex exists beneath a rising bubble does not necessarily suggest that this is related to proto-nucleic acid formation – although in making this argument, it is clear to see that this subject of the detailed structure of cationic vortexes is quite off topic for the current manuscript.

Other comments: Line 132: “bubble mediated scavenge [sic] and aerosolization of biomolecules with special attention given to diatoms” The authors perhaps mean to say ‘aerosolization of biomaterials’ as diatoms are not cells and not molecules.

The proper terminology for diatom morphology should be used: Cyclotella are ‘centric’ diatoms, while Nanofrustulum are ‘pennate’ diatoms.

Diatoms have been referred to in this manuscript in general terms using “bio-cells” or “bio-matter”. These are not accepted terms and the authors should simply refer to them as “cells”, “biological material”, or even “biological particles”, as are often used in the oceanography literature when referring to filter-retained material in the ocean.

Concentrations should be provided using standard unit notation (cells/mL) rather than noted as “cells / 1 mL.” In tables, units of volume (mL) are consistently given instead of units of concentration. It is suggested that the authors seek further editing of English usage and grammar for this manuscript.

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