

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

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Refer to: Ocean Science: os-2017-82, submitted on 03 Oct 2017 Manuscript Number: PAAG-D-14-00412R2; Title: Rising bubbles as mechanism for scavenging and aerosolization of diatoms; Authors: Roman Marks, Ewa Górecka, Kevin Mc Cartney, and Wojciech Borkowski Responses to the Anonymous Referee #2

First of all I would like to thank the Reviewer for many valuable comments. Base on them a new version of the manuscript was prepared. 1. Comment. The manuscript presents some interesting postulations on the mechanisms for the scavenging of diatomaceous material by rising bubbles and the aerosolization of scavenged material on bubble-bursting at the sea-air interface. I am reviewing this from the perspective of

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marine atmospheric science, where there is a substantial body of literature concerned with the bubble-mediated production of sea-spray 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.

2. Response. The literature concerned with bubble-mediated sea spray production is expanded in altered manuscript and include more recent references from marine atmospheric sciences. Our research on the bubble scavenge and aerosolization is based on experimental evidences that revealed ability of rising bubbles to develop bi-rotational and bi-ionic-electrical features, which are mechanistic and may be directly used to explain the principles of bubble mediated scavenge of bio-particles in the water column and aerosolization. The main evidences were gained by tracing photography records of rising bubble. The experiments require proper positioning of light source below the cylinder or at top, and more importantly the water has to be relatively clean (filtered) as well as relatively warm. The rotary tracers become foggy and not distinguishable when water contains too much of suspended materials. Since two distinct rotational systems were identified, we realized that also a charge polarity within the bubble upper and bottom half spheres is gathered during the bubble rise. The results are presented in (Marks, 2014; 2015). This is why we explain the diatoms/bacteria aerosolization base on both: the mechanistic and electrostriction principles. Considering the Reviewer comments, a new version of manuscript has been prepared and content of Literature expanded, see responses below.

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

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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.

2. The organic material in sea spray particles are aerosolized by both film and jet modes. The airborne dispersion of film mode is instant when bubble bursts, but particles accommodated to less abundant jet mode (although dominating in term of mass) contributes, since jet droplets may dry and undergo fragmentation. For example diatoms drying in droplets may even "explode" and break to fragments. These fragments are likely more dispersible and influence condensation of water vapour and "water uptake in the moist atmosphere". In addition list of literature is expanded.

3. [The importance of aerosolized organic materials as cloud condensation nuclei and

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their broader atmospheric functionality is reviewed by (O'Dowd et al., 2014). Apparently, also the contribution of diatomaceous is of more atmospheric significance than the other organic materials of a phytoplankton origin (Fuetnes et al., 2010). It has been also suggested that fragments of diatoms may provide ice nucleating particles (Wilson et al., 2015).]

[Bigg C., Leck E.K. (2005) Biogenic particles in the surface microlayer and overlaying atmosphere in the central Arctic Ocean during summer, *Tellus B*, 57,4, 305-316, DOI: 10.1111/j.1600-0889.2005.00148.x.

Lee C., Sultana C. M., Collins D. B., Santander M. V., Axson J. L., Malfatti F., Cornwell G. C., Grandquist J. R., Deane G. B., Stokes M. D., Azam F., Grassian V. H., Prather K. A. (2015) Advancing Model Systems for Fundamental Laboratory Studies of Sea Spray Aerosol Using the Microbial Loop. *J. Phys. Chem. A*, 119338860–7010.1021/acs.jpca.5b03488.

Lakhina G.S. (1993) Electrodynamic coupling between different regions of the atmosphere. *Current Science*, 64, 660-666.

O'Dowd C. D., Facchini M. C., Cavalli F., Ceburnis D., Mircea M., Decesari S., Fuzzi S., Yoon Y. J., and Putaud J.-P. (2004) Bio- genically driven organic contribution to marine aerosol, *Nature*, 431(7009), 676–680.

Patterson J. P., Collins D. B., Michaud J. M., Axson J. L., Sultana C. M., Moser T., ... Gianneschi N.C. (2016) Sea Spray Aerosol Structure and Composition Using Cryogenic Transmission Electron Microscopy. *ACS Central Science*, 2(1), 40–47. <http://doi.org/10.1021/acscentsci.5b00344>.

Wang X., Deane G.B, Moore K.A., Ryder O.S., Stokes M.D, Beall C.M., Collins D.B, Santander M.V., Burrows S.M., Sultana C.M. and Prather K.A. (2017) The role of jet and film drops in controlling the mixing state of submicron sea spray aerosol particles, *PNAS* 114 (27), 6978-6983, doi:10.1073/pnas.1702420114.

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Wilson T.W., Ladino L.A., Alpert P.A., Breckels M.N.B., Brooks I.M., Browse J., Burrows S.M., Carslaw K.S., Huffman J.A., Judd C., Kilhau W.P., Mason R.H., McFiggans G., Miller L.A., Nájera J.J., Polishchuk E., Rae S., Schiller C. L., Si M., Temprado J.V., Whale T.F., Wong J.P.S., Wurl O., Jakobi-Hancock J.D.Y., Abbatt J.P.D., Aller J.Y., Bertram A.K., Knopf D.A.K., and Murray B.J. (2015) A marine biogenic source of atmospheric ice-nucleating particles, *Nature*, 525, 234–238, <https://doi.org/10.1038/nature14986>.]

1. 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. 2. Accepting the Reviewer comment (Abstract) is shortened. Two first sentences are deleted and third altered to read:

3. [Abstract. Bubbles rising in relatively clean saline water cause steady displacement of ions at the bubble boundaries which creates electric polarity that draw biological particles dispersed in the water. Viable diatoms as well as bacteria develop negative charge on outer membranes that are attracted to the cationic bubble bottom half sphere. When bubble bursts at the air/water interface the diatoms and bacteria are ejected into the air with initial or secondary jet droplets that are projected upward with a small water column derived from a sub-bubble cationic vortex. Laboratory experiments conducted in brackish and oceanic saline water on *Nanofrustulum* and *Cyclotella* cells indicate that the average concentration of jet droplets compared to original water volume (here termed the enrichment factor) for aerosolized diatoms may range from 8 to 307.]

1. 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

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bio-molecules with special attention given to diatoms".

2. Introduction is shortened and reedited. In addition a new paragraph underlying contribution of atmospheric researchers to study the sea spray aerosol is inserted. Also the Reviewer comment related to section 1.3 is rephrased:

3. [Increasing interest in biological and chemical nature of sea salt aerosols has resulted in numerous field experiments and papers (e.g., Bigg and Leck 2005; Patterson et al., 2016). In particular the role of sea spray particles in determining electrical mobility of particles and related cloud properties was investigated by (Wang et al., 2017). In addition, evidences that aerosol samples contains phytoplankton and fragments of diatoms were reported (e.g., Bigg and Leck, 2005; Lee et al., 2015; Patterson et al., 2016). Furthermore, the importance of organic materials as cloud condensation nuclei and broader atmospheric functionality is reviewed by (O'Dowd et al., 2014). Apparently, the contribution of diatoms is of more atmospheric significance than other organic materials of a phytoplankton origin (Fuetnes et al., 2010). It has been also suggested that diatoms fragments may provide ice nucleating particles (Wilson et al., 2015). However, no particular investigations on enrichment of diatom cells in jet droplets have been reported.]

[The present research further explores the bubble mediated scavenge and aerosolization of biological particles with special attention given to diatoms.] 1. 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  $1.667 \times 10^{-5}$  L/s. Each 1.2 mm diameter bubble is  $0.905 \times 10^{-6}$  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  $3.5 \times 10^{-7}$  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

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(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.

2. The air flow rate was  $\sim 0.35$  mL/s; however the main attention was given to produce separated bubbles that were not forming foam and to collect separated droplets under highest possible air humidity, needed for diatoms counting. That was achieved by covering the glass breaker by microscopy slides during the aeration. Then slides were immediately closed in Petri Dish with small volume of water, in order to avoid the damage of diatoms. In closed Petri Dish air humidity was 100%. The sizes of bubbles were measured optically by sizing the photo-tracers of bubbles in the water or at the surface as well as tracers of jet droplets just after ejection into air. In order to collect set of samples during each run, much longer time was needed than 2-3 seconds. Considering the reviewer comment the expression (cation availability) was deleted and accordingly the first sentence in page 7 was altered. In order to provide more information on electric charge distribution around bubbles we inserted a following:

3. [In order to detect electric charge distribution around the rising bubble boundaries an oscilloscope, type HANTEK<sup>®</sup>-DS01201BV was used, allowing to trace the voltage polarity between a two probe heads in mV with 0.1 mV detection limit and 0.5% accuracy. One head was submerged to the depth of 2 cm in the bulk water, and the second probe tip was placed just at the water/air/bubble interface. The measurements were conducted in a small glass tank filled with 700 mL of saline (35 g/kg) water of 21.0°C temperature. Bubbles  $D = 1.2$  mm were generated by capillary aerator with a rate allowing ca. 20 mm distance between a successive bubbles. These rose about 6 cm in a free water column and then were slowed on a glass plate tilted to 30° that reduced a zig-zag motion of bubbles, allowing interactions with the tip of probe head.

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The measurements showed negative charge introduced by bubbles outer layer that typically adhered to the tip at water/air/bubble interface. However, from time to time also strong positive charge was measured, indicating interaction with cationic domain gathered within surfacing bubble vortex. The maximum negative and positive values were considered as revealing polarity of ions gathered around the bubble upper and bottom half spheres.] 1. 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."

2. Thank you for that comment the word (increased) was replaced by (decreased). The sentence was rewritten:

3. [However, after analyzing that set of data we noticed that the efficiency of diatom aerosolization decreased in time of aeration, indicating that rising bubbles reduced the concentrations of diatoms in the water penetrated by bubbles (solution was gradually cleaned). In addition, the development of microlayer and changes in physicochemical properties of the water/air interface gradually suppressed the bubble mediated diatoms aerosolization.] 1. 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 descriptions 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.

2. Following the Reviewer comment, the first paragraph and in particular (lines 219-222) are rephrased. Lines 230-232 are corrected. In addition, the context of Lines 240-247 with expanded explanation is inserted to that paragraph.

[Considering that a single burst of bubble  $D = 1.2$  mm may eject about 4-8 droplets from

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a clean water at temperature ( $T_w$ ) of 20°C (Blanchard and Syzdek, 1982), the number of jet droplets that contained diatoms were counted and compared with droplets that lacked diatom cargo. The comparison showed that only 20-25% of jet droplets were enriched by diatoms, which suggests that the diatom aerosolization process might be influenced by a combination of factors operative in the water column, or at air-water interface. To determine whether the diatoms concentration in the top jet droplet is always highest, as reported for bacteria (Blanchard and Syzdek, 1978), jet droplets were collected at different elevations above the water level with special attention given to initial droplet. The observations indicated that not exclusively top jet droplets were diatom enriched.] [Since diatom cells are relatively large, as compared with bacteria cells, these are more resistant to sudden displacement with cationic jet, thus contribute into sub-initial droplets.]

[In addition, a build-up of microlayer at the water/air interface might suppress the efficiency of water jet formation by bursting bubbles.] 1. 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 or 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). 2. Accepting the Reviewer comment the number of samples collected during each set of experiments was inserted to Table 1 and 2. 3. (Table 1) [(N) number of samples 20 8 ] (Table 2) [(N) number of samples 20 20 ]

1. 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

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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.

2. In order to investigate the distribution of diatoms with droplets high, the other set up (probably modification of that used by Blanchard and Syzdek, 1978, would be required). We designed our experiments to estimate the enrichment factor for aerosolized diatom cells of two shapes and considering two suspensions. However, I do agree that much more research is needed and in particular exploration by methods that are in hands of chemist might provide new evidences. I strongly feel that these will support our findings. Additional explanation and graphical description of droplets and diatoms distribution was included according to the Reviewer postulates.

3. [Figure 5] [An example of typical data distribution and related variability is show in Fig. 5, where number of droplets with and without diatom cells as well as number of diatom cells in each of 20 samples is plotted. Data refer to experiment with suspension of Nanofrustulum cells aerosolized by uniform bubbles of  $D = 1.2$  mm rising in water column containing 840000 cells/mL under 22.1oC temperature and 35 g/kg salinity.] 1. 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.

2. Both sections include more information and were reedited.

1. 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.

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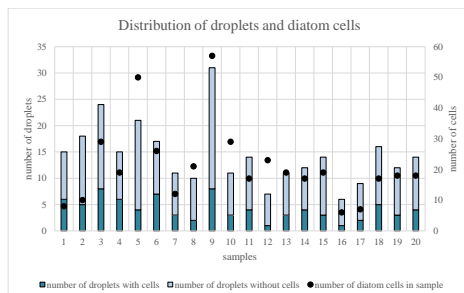
2. Thank you for that remarks. The sentence on line 48 was shortened and punctuation (while, during rain) inserted to read:

3. [A typical breaking wave generates a downward circulating rotor-like motion reaching a depth equal to the wave height (Thorpe, 2001) while, during rain bubbles may occur to about 10-20 cm depth (Blanchard, 1963; Katsaros and Buettner, 1969).] 1. Line 66 "Cl/Na atomic mass..." add "RATIO" 2. The "ratio" was added. 3. [Using a Cl/Na ratio of atomic mass equal to 1.542 (Kropman and Bakker, 2001). . .]

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**Figure 5.** Number of droplets containing *Nanofrustulum* diatom cells, without cells and number of cells in each sample aerosolized by bubbles of  $D = 1.2$  mm.

Fig. 1.

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