



EGUsphere, referee comment RC2
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Comment on egusphere-2022-27

Anonymous Referee #2

Referee comment on "The ocean fine spray" by Alfonso M. Gañán-Calvo, EGU sphere,
<https://doi.org/10.5194/egusphere-2022-27-RC2>, 2022

The author proposes that a single mechanism is responsible for most sea spray aerosol, ranging from 5 nm to about 0.5 mm and due to bursting bubbles from 10um through the jet drop process. I consider that this paper is of interest to the community as it presents a new vision of the problem. However, I have several comments that I believe need to be addressed before the paper can be published. It is also worth mentioning that the paper is relatively hard to read and follow.

I think the author needs to be clearer on what has been proven by experimental data at this point and which part of its theory remains hypothetical. This is not necessarily a problem but it needs to be made very clear that several statements are not backed up by data at this point (as far as I understand). If I followed correctly, the discussion on rarefaction remains highly hypothetical since no experimental data can back the claims from the author. I remain particularly impressed by the 2017's paper from the author on jet drop, but that theory was backed by many data. I am also unclear why the author did not consider any of the dry particles measurements by bursting from many other authors (e.g. Forestieri et al, Salter et al, Prather et al; Frossard et al, Sellegri et al etc). The literature is huge and I understand it is hard to keep track but it seems that the choice of data set might be at least discussed, so that the reader can understand why certain data are used and not others.

As a general point of discussion, I am not convinced that a single mechanism can explain all of the sea spray aerosols for two main reasons: a) there is plenty of chemical evidence of the importance of film drops, as it is expected to have more organic material in the film drops than the jet drops; and b) can the author clarify how the bubble population down to 10um at the surface is justified? Again, this is not a show stopper but hypothesis and limitations should be discussed more openly.

Major comments:

1/ When discussing sizes, the author needs to specify if they mention the size at production, the size at 80% humidity or the dry aerosol size (all of which are being used in various parts of the literature). There are typically factor 2 to 4 variations for sea salt which make a big difference when arguing about "100nm" drops/particles. I do not recall jet drop data demonstrating 200nm drop size, as Brasz et al 2018 show a 1µm drop (which would be about 250 dry radius for a salt particle (so not a drop but a solid salt particle). This needs to be clarified throughout the text. Is the author talking about salt dry size? Liquid size? If considering salt, then field data suggest that particles below 10nm are essentially organic (see literature cited in the introduction).

2/ Several papers have analyzed the composition of atmospheric aerosols. I am not a chemist but it seems that organic aerosol mostly originate from film drops (see recent work by Quinn, Prather, etc). How would the author reconcile its claim with the atmospheric chemistry literature? (some of it being cited in the manuscript). This is also related to my previous point as the link between organic aerosol and initial drop size is not obvious (at least to me).

3/ Do we have observations of bursting bubbles of 10-1000µm at the surface of the ocean? Such small bubbles are observed down in the water column as they can be transported by turbulent processes. Simulations of bubble dynamics in upper ocean by JH. Liang and coauthors show significant transport of the small bubbles down in the mixed layer, leading to their full dissolution. There are discussions in papers on bubble gas exchange by Thorpe and Woolf which assume that bubbles below a certain size fully dissolve as their rise velocity would be too small to overcome turbulence fluctuations and those bubbles might never reach the surface. It seems that these questions deserve some discussion as they will be important to test the applicability of the proposed theory.

4/ I am not sure that the current literature "describes in great detail the chemical composition of ocean aerosols and their dependence on ambient (wind temperature)" There is a lot of uncertainties in the role of temperature, with some studies predicting an increase in ssa while other predict a decrease (see for example figure 1 in Forestieri et al 2018). Scatter in the role of wind spans two to three orders of magnitude (see review from deLeeuw et al 2011). This is a topic of intense scrutiny due to the difficulty of the field measurements.

5/ When the author mentions "*Two basic mechanisms are responsible of this droplet emission: bubble film breakup*" (p Lhuissier and Villermaux (2012); Jiang et al. (2022)), "I would encourage to cite previous work by Spiel, Blanchard, and others. The cited papers are not (by far) the first ones to describe film drops. The review of Lewis and Schwartz 2004 does an excellent job at summarizing the film drop evidence in the literature, in particular with respect to sub-micron aerosols. This part of the literature should not be overlooked as the Lhuissier and Villermaux (2012); Jiang et al. (2022) papers (while wonderful) present a specific vision of the problem (in the same way as the present paper presents the specific vision of the author). All of this is fine and part of the scientific process but the introduction should not focus on a specific subset of authors.

6/ The fitting and reanalysis done in figure 2 needs to be clarified. I would encourage the author to first show the dimensional data and then the rescaling as the rescaling is not obvious/common in the aerosol literature. I would also encourage to provide a plot of the mean diameter/radius of the drops as a function of the bubble, as it seems to emerge from that rescaling and is not presented in the original paper (I think). The measurements from Jiang et al 2022 are presented in dry size so the scaling factors used in the reanalysis need to be mentioned.

7/ The discussion on the rarefaction effect is rather unclear to me. I would encourage the author to explain this better. Figure 7 is used as proof of the rarefaction effect but if I understand correctly comes from simulations. Does the author have any experimental evidence of this effect? I see this as a major issue with the claims of the author. They propose a theory leading to very strong claims but have no data to back it up. I would be more clear on some of the hypothesis that remain to be tested.

Other comments:

8/ In 2.1.; "rd<0.5" is that micron?

9/ I do not understand the statement: "The super-micron size range is comprised by wet aerosols and its presence is fundamentally reduced to marine and coastal regions Boyce (1954)"

10/ I do not understand the statement "From this study and entirely attributing the fitted pdfs to micron- and submicron film droplets, one would conclude that (i) the droplet generation by film flapping will be distributed according to a lognormal for the nondimensional variable $\eta = \chi dLa^{-1/5}$, which reflects a reasonable dependency on the bubble size, while (ii) the more "classical" film rim fragmentation Lhuissier and Villermaux (2012) would yield droplets distributed according to a generalized inverse Gaussian as (1) independently of La."

Could the author be more explicit?

11/ The discussion on enrichment for very small bubbles seems interesting but I am not sure I am fully following. It also seems to contrast with some other papers. I would encourage the author to discuss this in more details and clarify its hypothesis compared to other work in the literature (see the work from W.C. Keene, P. Quinn, A. Frossard, K. Prather, etc).

12/ I am not sure that the first sentence of the abstract is accurate. It is true in remote

marine environment but probably wrong as a general statement.