

This manuscript by Barhelmeß et al extends the classic research into marine gel particles with this report on hydrogels (TEP and CSP) from the Southwestern Pacific Ocean, as well as in nascent aerosols generated in a bubbling tank. Using colorimetric staining assays and microscopic, size-resolved quantification they compare data on hydrogels in the spray generated from samples collected in the Southwestern Pacific Ocean. The motivation for this study is to extend our understanding of the relationship between polysaccharidic and proteinaceous gels in the sea surface microlayer to aerosolized particles and atmospheric processes including the formation of cloud condensation nuclei (CCN) and ice nucleating particles (INP). As part of the justification, in LINE 15 why should it matter that “the few studies which exist were conducted in the Northern hemisphere” (and Hemisphere should be capitalized! Why would you expect a major difference? This should be part of the motivation for this study. Just collecting more samples by itself is not a justification.

In general, this study is relevant for improving our understanding of ocean-atmosphere interactions in the context of aerosols and although with very limited data, worth publishing. However, there is room for improvement. The limited number of samples is unfortunate, but still allows for some conclusions. The authors do oversell the study with the title. CONTROL is a very strong word. More accurately Biogeochemical regimes influence the chemical characteristics of aerosolized hydrogels. In general they show this, but not convincingly given the limited data set. The authors are encouraged to address not only the specific issues highlighted, but overall make the manuscript more rigorous in presentation and interpretation of the data.

For example, the introduction provides a good summary of the current literature, an overview of sources and chemical composition of gels/ hydrogels in the SML and relevance to sea spray aerosols (SSA). However, it is dense, often redundant, and sometimes ambiguous which detracts from the flow. Several sentences are overly long exceeding 30 words, making it difficult to follow the authors intentions. Frequent parenthetical citations interrupt flow too. Some terms are introduced without immediate definition (e.g., “hydrogels” appear before TEP/CSP are defined). And, there are often a repetition of ideas (e.g., hydrogels as CCN/INP is mentioned multiple times). It would be much more helpful to the reader if the authors would make sure there are clear transitions between themes (particle type and composition, atmospheric relevance, oceanic production, and knowledge gaps), break long sentences into two or more, move citations to the end of sentences to complete a thought, introduce TEP/CSP earlier or define hydrogels more explicitly at first mention, and consolidate repeated statements about the potential role of these gels in atmosphere processes. For example: The term “hydrogels” is used broadly. It is not clear if the authors are referring to polymer networks, colloidal gels, or operationally defined TEP/CSP.

The hypothesis (LINE 495) illustrated in Figure 7 that bacterial processing in the SML favors the accumulation of surface-active, proteinaceous rather than carbohydrate-rich material with TEP rapidly degrading leading to larger carbohydrate rich particles which are aerosolized and smaller protein-rich particles is consistent with finding in Aller et al., 2017 that CSP in the surface waters and the SML was concentrated in larger size particles while TEP was concentrated in smaller particles with both aerosolized.

In the abstract, LINE 22 the statement about carbohydrate-to-protein ratio talks about “marine samples”. Weren’t all the samples from marine waters?

While the methods section contains all essential methodological details, the narrative is dense, occasionally repetitive, and sometimes unclear in organization. Several sentences are overly long, and some information is not in logical order. The narrative jumps between topics (e.g., chamber design to aerosol sampling to filter storage to blanks to staining), which makes it more difficult to follow. The description of oceanic regimes is followed by aerosol chamber details but is disconnected from the SSA sampling workflow. The plunging-jet system description is split across multiple sentences and paragraphs. The transition from SSA sampling to filter staining is abrupt.

LINE 140. It is not clear why the SSA collection onto filters was conducted over a ~6-hour period with no replication. Regarding the dual staining. Why were the number of filters limited? Not clear why an apology is necessary. If Engels method is followed why the apology? Rather than use the same filter for dual staining was cutting the one filter in half and using one half for the TEP and the other for CSP considered? Did it matter whether you stained first with the CBB then Alcian Blue or vice versa? Would it make any difference? If they have been divided, the number of samples could have been increased from 1 or 2. The authors defer all discussion of deviation of methods from what one would expect to the supplemental section.

One of my main problems with this paper concerns the first method that Engel proposed to contrast carbohydrate and proteinaceous particles, but subsequent studies took a more quantitative analytical approach with xanthum gum and bovine albumin serum calibration. LINES 124 and 386-390. Some comparisons to literature are not fully parallel (e.g., comparing XG equivalents to colorimetric -area-based measurements). The use of the colorimetric technique vs XG and BSA equivalents are not explained and no direct comparison is made before adopting this technique. Methodological differences when comparing to other studies should be clarified in the main text and not buried in a supplemental section. While they state that their color technique relies on a classical approach (Engle, 2009) it only provides “approximate the contribution of CSP and TEP”. Nevertheless, their results are quantitative. Any discussion about comparison of a microscopic method with the widely accepted quantitative colorimetric technique (XG and BSA equivalents) needs to be made in the text.

Also, regarding methods, the chamber description is mixed with sampling volumes and residence times, then the description reverts back to chamber operation. The section would benefit from clearer structuring, more concise phrasing, and improved flow between procedural steps.

The results give a good introduction to the 4 oceanic regions and their biological characteristics; however, this section often jumps between regimes, soot events, size bins, and Na⁺ normalization without clear transitions which makes it difficult to follow the progression from raw observations to the patterns and then to the interpretation. For example, soot contamination is introduced abruptly in the middle of the results, interrupting the flow of hydrogel abundance patterns. The anomalous soot events and heavy rain events are identified and contextualize helping to justify the exclusion of some samples. That being said, listing daily values (e.g., “on the 23rd of

March...”) LINE 232 stops the flow of the narrative. Given that the date is important for the pattern of results perhaps the information could be reorganized by date followed by the results.

The authors need to clarify the implications of limited SSA replication. The quantitative detail provided obscures the fact that the results are VERY LIMITED. There is no explicit statistical framing, only the statement that “no statistical tests were performed” for SSA (LINEv229) due to limited sample size”. This is misleading. Presumably, the lack of replication was not intentional but, it still leaves the possibility that the degree of variability is much greater or much less.

So while there are definitive values for abundance, area, CSP/TEP ratios, and enrichment factors, stating that the SSA hydrogel concentrations “ranged from $0.9 \pm 0.7 \times 10^5 \text{ m}^{-3}$... to $0.2 \times 10^5 \text{ m}^{-3}$...” gives the impression that the number of samples (n) is greater than 2. In this case, just giving the 2 values is better given the VERY LIMITED data.

Interpretation creeps into the results section in many instances. For example, the authors should just present the fact that the CSP fraction decreased with particle size and not include interpretation that proteinaceous particles were the predominate type. The contrast between SSW and SSA size-bin patterns is well articulated. LINES 237-243) Results make no sense as written “CSP contributed especially to the smallest size bin reaching its minimum in the largest size bin”. Totally needs clarification.

Some key findings are buried and should be highlighted more explicitly. The highest concentration of hydrogels and highest relative enrichment of hydrogels in the Subtropical Front is a major finding and deserves a more prominent discussion, perhaps in the conclusion.

The figures and table are well aligned with the narrative. The use of pie diagrams to illustrate proportional abundance in Figure 5 is effective. Figure 5 and Table 1 directly support the text and clearly illustrating the size-resolved patterns. However, size-resolved abundance appears in the text, in Figure 5, and in Table 1. sometimes with overlapping phrasing. Consolidating these into a single, coherent subsection would improve readability.

This section would benefit from reorganization into subsections: (1) SSW patterns, (2) SSA patterns, (3) size-resolved patterns, (4) enrichment to help improve transitions and maintain the flow of the results. There is a certain amount of redundancy and repetition. For example, hydrogels as CCN/INP is mentioned in two separate places. Bubble bursting mechanisms are described in detail, but similar information appears earlier in the literature review. The authors are encouraged to consolidate discussion of the roles of hydrogels in atmospheric processes into a single, well-structured paragraph and to keep bubble-bursting details in the description of the study design.

Sections 3.3 Biogeochemical characteristics mixes descriptive statistics with previously published findings. and 3.4 Biogeochemical factors influencing hydrogel occurrence in seawater and aerosols contains detailed results and comparison with previous studies, but the narrative jumps between particle size classes, phytoplankton groups, CSP and TEP in the water and in SSA, to ecological interpretation, and then to mechanistic explanations. This loses the reader who can no longer follow the main points. The connection between phytoplankton community composition and

hydrogel types are interesting but the claim for example that *Synechococcus* driving CSP enrichment are speculative given that they present no data or referenced discussion of blooms of these cyanobacteria, or mucus or exudate production which would provide hydrogel material.

The fact that the result section extends beyond results and incorporates discussion leads to some repetition in the discussion section. In keeping with the journal's structure, the results should be kept in the results section and move all mechanistic interpretations to the discussion.

There are repeated references to Chl a patterns (i.e. LINES 302-305, 326, 333-335, 368, 435) repeated statements about nutrient regimes (STF vs MIX vs SAW vs STW), and multiple mentions of nanophytoplankton abundance patterns. Consolidate repeated descriptions of regime characteristics. Just present each biogeochemical variable once, then refer back to it when needed.

There are occasional grammatical errors ("22rd", "24st") in the Fig. 4 legend

Some phrases are vague (LINE 273) regimes were characterized by specific changes and the use of the future tense (LINE 272). Occasional awkward phrasing LINE 476 ("aggregation time of TEP are extended").

Some anthropomorphic phrasing LINE 431 ("bubbling scavenges, up concentrates and aggregates"). LINE 481 ("bacteria benefit from concentrated substrate").

Some claims are speculative without explicit qualifiers and should be changed. (e.g., soot interfering with bubble bursting). LINES 440. The authors have no evidence for soot particles on the aerosol filters which they presumably confirmed with microscopic examination, they speculate that soot may have interfered with bubble scavenging and bursting.

The occasional over-interpretation of patterns (e.g., attributing enrichment differences solely to polymer size) I attribute to the paucity of data. Without data to support the interpretation, implying causation should be avoided.