

## ***Interactive comment on “Effect of wind speed on the size distribution of biogenic gel particles in the sea surface microlayer: Insights from a wind wave channel experiment” by Cui-Ci Sun et al.***

**Cui-Ci Sun et al.**

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We much appreciate the referee’s constructive and thoughtful comments. Below we have pasted in the entire review, and inserted our responses to the suggestions (indicated by two stars).

Interactive comment on “Effect of wind speed on the size distribution of biogenic gel particles in the sea surface microlayer: Insights from a wind wave channel experiment” by Cui-Ci Sun et al.. Anonymous Referee #2 This paper presents results from wind-wave channel experiments on how wind-driven water mixing affects dynamics of marine gels (TEP and CSP) in the sea surface microlayer (SML). The authors conducted

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detailed analysis of TEP and CSP concentration and size distribution. They concluded that wind speed controlled gel accumulation and size distribution in the SML under their experimental conditions. It is very difficult for me to evaluate the results and the conclusions in the present version of the manuscript because the description of the experiments as well as the presentation of some of the results are lacking important information (see details below). The manuscript would also benefit from shortening some of descriptive text in the Intro and in the Discussion. See below for some suggestions on that as well. I strongly recommend that the authors edit the text so that it is more focused and less wordy. \*\* We agree and will shorten the intro and discussion section.

1. Abstract: -In L. 9 use SML instead of surface microlayer. \*\* Will be done.

- Starting at L. 11: be more specific about the results on TEP and CSP; does this description refer to PSD of gels in the SML or bulk water? I suggest the following abbreviations for TEP and CSP in the SML (TEP-SML – CSP-SML) and in bulk water (TEP-bulk – CSP-bulk). Otherwise it is hard to distinguish between the two phases. \*\* This description refers to the PSD of gel particles in the SML. We think your suggestion is valuable. The abbreviations for TEP and CSP in the SML (TEP-SML – CSP-SML) and in bulk water (TEP-bulk – CSP-bulk) will be used across the whole MS to distinguish between the two phases.

- L. 17-18: You talk about the effects of TEP on aggregation and export. Since the focus of this paper is on TEP and CSP in the SML and the potential effects of gas exchange etc. you should focus/discuss potential effects on processes between the water and the atmosphere. In other words: if TEP settles out of the SML what that could mean for gas exchange processes between the water and the atmosphere. \*\*With respect to the potential effects of the accumulation and size distribution of gels particles on the sea-air exchange process, the more detailed analysis on the fraction of submicron gel particles (0.4-1 $\mu$ m) will be addressed in the whole manuscript: Below information will be added in the abstract: The contribution of submicron gels particle in the smallest

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size class, size 0.4-1  $\mu\text{m}$ , became larger at higher wind  $>6\text{ms}^{-1}$  after the addition of *E. huxleyi*, potentially impacting the emission of gels with sea spray aerosol.

The description below will be inserted in results section: ‘The abundance fractions of submicron particles (0.4-1  $\mu\text{m}$ ) in the SML were analyzed at low wind (LW) and high wind (HW) (Fig. S1). The results showed that the fraction of submicron gel particles became larger at high speed than at lower wind speed ( $<6.1\text{ms}^{-1}$ ) during the period after addition of *E. huxleyi* followed by a biogenic SML from a previous experiment ( $p=0.003$  for TEP-SML,  $p=0.02$  for CSP-SML, two sample-Kolmogorov-Smirnov test). The median of fraction of submicron gel increased from 33.7% at low wind to 43.0% at high wind speeds for submicron TEP-SML and from 38.5% to 46.0% for submicron CSP-SML, respectively. However, there were no enhancement found in submicron fraction at high wind speed before the addition of *E. huxleyi*, with the exception on day11 for TEP when the fraction of submicron TEP-SML increased from 37.7% at  $3.925\text{ms}^{-1}$  to 51.4% at  $18.208\text{ms}^{-1}$ ’.

The discussion below will be added: ‘In this study, we found that the fraction of submicron gels (0.4-1  $\mu\text{m}$ ) in the SML increased at high wind speeds after the addition of *E. huxleyi* and on day 11 with the peak concentration of bacterial abundance in SML. Due to the TEP’s quasi-particulate nature, a considerable number of small gels can pass through a filter with size of 0.4  $\mu\text{m}$  (Passow and Alldredge, 1995). It is therefore likely that the fraction of submicron gels was even higher at high wind speeds than observed. The changes of PSD in SML indicated that large gels were fragmented into smaller gels at high wind speed, or that submicron gels were generated. A strong enrichment of TEP in submicron SSA under field conditions has been observed before (Aller, et al 2017). Production of SSA in the field is driven by wind speed, and SSA in the size range 0.4-1  $\mu\text{m}$  in particular were observed to be higher at high wind speed (Lehahn et al., 2014).

References: Aller, J. Y., Radway, J. C., Kilthau, W. P., Bothe, D. W., Wilson, T. W., Vaillancourt, R. D., Quinn, P. K., Coffman, D. J., Murray, B. J., and Knopf, D. A.: Size-

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resolved characterization of the polysaccharidic and proteinaceous components of sea spray aerosol, *Atmos Environ*, 154, 331-347, 2017. Lehahn, Y., Koren, I., Rudich, Y., Bidle, K. D., Trainic, M., Flores, J. M., Sharoni, S., and Vardi, A.: Decoupling atmospheric and oceanic factors affecting aerosol loading over a cluster of mesoscale North Atlantic eddies, *Geophys Res Lett*, 41, 4075-4081, 2014. Passow, U., and A. L. Alldredge, Aggregation of a diatom bloom in a mesocosm: The role of transparent exopolymer particles (TEP), *Deep Sea Res., Part II*, 42(1), 99–109, 1995.

2. Introduction: Page 3 - L. 6: I don't think you need the abbreviation ULW. \*\* We agree and will delete the abbreviation ULW.

- L. 9: do you have a reference for this statement? \*\*References for this statement will be added:

Azetsu-Scott, K., and Niven, S. E. H.: The role of transparent exopolymer particles (TEP) in the transport of Th-234 in coastal water during a spring bloom, *Cont Shelf Res*, 25, 1133-1141, 10.1016/j.csr.2004.12.013, 2005. Ebling, A. M., and Landing, W. M.: Sampling and analysis of the sea surface microlayer for dissolved and particulate trace elements, *Mar Chem*, 177, 134-142, 10.1016/j.marchem.2015.03.012, 2015. Guasco, T. L., Cuadra-Rodriguez, L. A., Pedler, B. E., Ault, A. P., Collins, D. B., Zhao, D. F., Kim, M. J., Ruppel, M. J., Wilson, S. C., Pomeroy, R. S., Grassian, V. H., Azam, F., Bertram, T. H., and Prather, K. A.: Transition Metal Associations with Primary Biological Particles in Sea Spray Aerosol Generated in a Wave Channel, *Environ Sci Technol*, 48, 1324-1333, 10.1021/es403203d, 2014. Mari, X., Passow, U., Migon, C., Burd, A. B., and Legendre, L.: Transparent exopolymer particles: Effects on carbon cycling in the ocean, *Prog Oceanogr*, 151, 13-37, <http://dx.doi.org/10.1016/j.pcean.2016.11.002>, 2017.

- L. 14 -l. 2 on page 3 : In general, this text can be shortened as the focus is on SML sea-air exchange and not aggregation and particle export. Page 4 - L. 3: to me, your intro starts here. \*\* We agree and will shorten the Intro part.

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- L. 25 – l. 4 on page 5: In the first sentence you are saying that “TEP enrichment : : : is inversely related to wind speed : : :”. You don’t have to repeat this statement in the following sentence; the first part of that sentence can be shortened: “One explanation for this is that : : :”. \*\* We agree and will delete the repeated statement according to your suggestion.

- L. what are the “other mechanisms” \*\*It is proposed that gel particles formation within the SML is supported by bubble scavenging of DOM in the upper water column (Wurl et al., 2011), because more TEP precursors are lifted up the water-column. Moreover, compression and dilatation of the SML due to capillary waves may increase the rate of polymer collision, subsequently facilitating gel aggregation (Carlson, 1987).

Reference: Wurl, O., Wurl, E., Miller, L., Johnson, K., and Vagle, S.: Formation and global distribution of sea-surface microlayers, Biogeosciences, 8, 121-135, 10.5194/bg-8-121-2011, 2011. Carlson, D. J.: Viscosity of Sea-Surface Slicks, Nature, 329, 823-825, Doi 10.1038/329823a0, 1987.

3.Methods: Page 7 - L. 4: change to “November 3-24, 2014.” \*\*It will be done.

- L. 5: I am confused about the total volume of water collected for this study: Is it 20000 L with 14000 L of high sal water (what does high sal water mean??) + 8000 L at 5 m near Sylt? That does not add up, so remove “In total” in line 4, because your total is 42000 L. - L. 5: change to “were collected onboard FS Poseidon”. How did you collect the water? Pumping or niskins? \*\*20000 L is typo. It should be 22000L. The detail of sampling and collection will be presented in the method section:

‘Effects of different wind speeds on the size distribution of organic gel particles in the SML were studied during the Aeolotron experiment from November 3-28, 2014. 22,000 L of North Atlantic seawater were collected by the research vessel POSEIDON, including ~14000L collected at 55 m at 64° 4,90’ N, 8° 2,03’ E and ~8000 L collected at 5 m depth near the Island of Sylt in the German Bight, North Sea. The water was pumped into a clean (“food save”) road tanker and unloaded at the wind wave facility Aeolotron

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in Heidelberg the following day and stored in the dark and cool ( $\sim 10^{\circ}\text{C}$ ) until the start of the experiment.'

L. 11-12: Info about something that you haven't used in your study like Uref is not important, so delete this sentence. \*\* We agree and will delete this part.

- L. 16: This is the part where I am getting confused about the experiments: 7 experiments were conducted, and you refer to fig.1 and table 1 for explanation. Figure 1 shows the step wise increase of U which lets me believe that the 7 experiments were conducted under the same conditions of U. Table 1 leads me with a different impression as the values of U were quite different throughout the experiments (the table is lacking the unit for U; you also need to describe what 'NaN' means. This needs to be explained in the methods. - L. 22-24: does this apply to all the 7 experiments? \*\* We agree that some description on the wind speed setting were confusing. More details about experiment will be presented in the revised version. The unit for U10 is m s<sup>-1</sup> and it will be added in the table 1. NaN means no wind speed data on this condition. Information on the wind speed setting will be added:

Two strategies of experimental wind speed setting were conducted in the experiment. For the first strategy, the wind speed setting was shown in the Figure1. 7 experiments were conducted on days 2, 4, 9, 11, 15, 22 and 24, respectively, with stepwise increase in wind speeds (equivalent to U10,) ranging from 1.371 to over 18.652 m s<sup>-1</sup> as shown in Table 1. During some of the high wind speed conditions (Table 1), bubbles were generated in addition with a profiO2 oxygen diffuser hose to simulate strong breaking waves with bubble entrainment and spray formation. The second strategy was conducted on days 5, 12 and 23. Only one wind speed was arranged at about 18ms<sup>-1</sup> with and without bubbling for about 2hour, respectively. The aim was to evaluate the difference effects between bubbling and no bubbling condition.

Why are there no values for U at some days during experiment 7. \*\*U10 was determined by the method of Bopp and Jähne (2014). In this method, water velocity was

one of the important parameters to calculate the U10. Since data of water velocity at some conditions were absent, there no values for U10 could be obtained.

Page 8 - L.1-4: why was the light switched on in these two periods? Does that mean it was dark (0  $\mu\text{mol m}^{-2} \text{s}^{-1}$ ) throughout the rest of the incubation time? Why is this important? - L. 6: I could not find the Engel et al. 2017 reference in the list? Do you mean the Engel et al. (subm) reference? There is no way that we can get any information from this paper at this point. So you need delete this reference and give as many information of the methods as needed for this manuscript. - L. 9-11: why was E. hux added to the water? I suggest adding some explanation in the intro. Also, what do you mean by “adding a biogenic SML from a previous experiment”? That is too vague, I have no idea what a biogenic SML could be/look like, and how can this be added without disruption etc. - L. 19: It would help to show the collection volumes or give a range because it is hard to imagine how much water you collected from the SML. \*\* We will add details on the manipulations in the supplementary materials: During the experiment, a series of manipulations were conducted. To stimulate phytoplankton growth, lights were switched on from day 9 to day 16 and from day 20 to day 26, with a 12 Light:12 Dark regime, respectively,. On 14 November (day12), nutrients were added to final concentrations of 14.7  $\mu\text{mol L}^{-1}$  nitrate ( $\text{NO}_3$ ), 9.5.  $\mu\text{mol L}^{-1}$  silicate ( $\text{SiO}_4$ ) and of 0.48  $\mu\text{mol L}^{-1}$  phosphate ( $\text{PO}_4$ ). In order to induce phytoplankton growth and exudation,  $\sim 1\text{L}$  of an algal culture (*Emiliania huxleyi*,  $4.6 \times 10^5 \text{ cells ml}^{-1}$ ) was added to the tank on day 20. In addition, 6L of water enriched with organic matter, sampled from surface microlayer during a previous phytoplankton mesocosm experiment, was added to the tank on day 21. This water had been stored frozen at  $-20^\circ$  for about 6 month until the addition.

- L. 7-8: This statement is too general, and I don't see why this would be important to know at this point. \*\*We agree with you and it will be deleted.

Page 10: - L.2: what are the wind conditions 1 and 2? \*\*Wind conditions 1 and 2 were the first wind speed (1.66  $\text{ms}^{-1}$ ) and the second wind speed (2.89  $\text{ms}^{-1}$ ) condition on

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day11.

4. Results: As mentioned above, I cannot evaluate the quality of the results before the authors improve the description of the experimental set-up. For example, I really cannot tell if the TEP and CSP results described on page 11 and shown in figure 2 are average values of all 7 experiments. Figure 2 also lacks error bars. You also need to add more detail to the figure legends (e.g. figs 4 and 5 show error bars, this needs to be mentioned in the legends). L. 16: this is the first time that chl a is mentioned. This needs to be described in the methods section. \*\*Figure 2 showed average values of the different wind speed condition on each experimental day; the SD bars will be added. The error bars on figs 4 and 5 will be mentioned in the legends. The description on Chl a will be added in the method: 'Primary productivity was low during the whole experiment. Chlorophyll a (Chl a) concentrations were not detectable until days 20/21, after addition of the *E. huxleyi* culture and the SML water from a previous phytoplankton bloom experiment. Chl a concentration clearly increased after day 23'.

L. 20: what do you mean by "at the start of each wind experiment"?? Does that mean that you varied the wind speed over a course of a day from 0 - 20 or so (see also figures 4 and 5). \*\*It means "at the start of experiment on each day of days 2, 4, 9, 11, 15, 22 and 24". On these experimental days, wind started at about 8:00 in the morning and ended at about 20:30 in the evening. The wind speeds over the seven experiment days varied a little, but all followed the same strategy of setting shown in the figure 1.

At last, we are thankful for your time and valuable suggestions to improve this manuscript.

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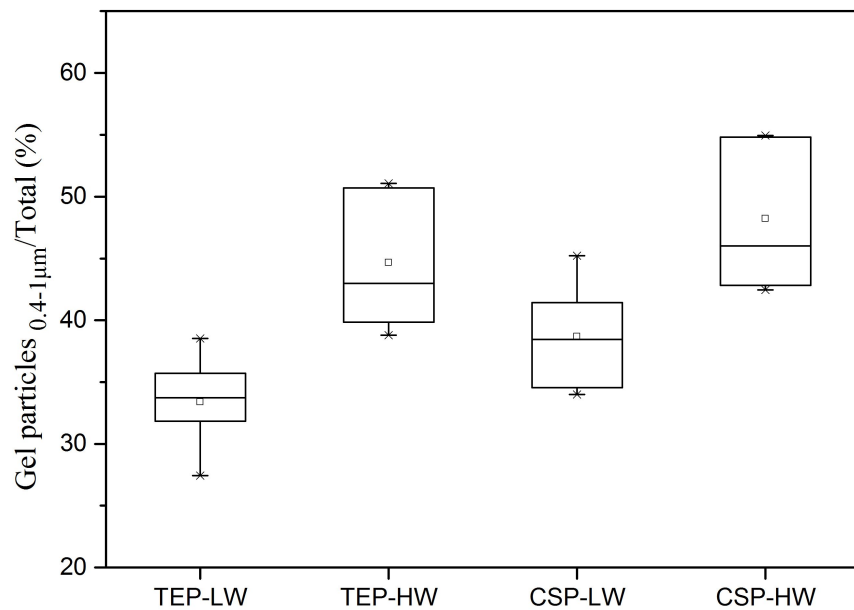
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**Fig. 1.** Figure S1 Changes in the submicron gel particles fraction with wind speed

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